$Ba_2Y(Nb/Ta)O_6$ doped YBCO films on biaxially textured Ni-5at.% W substrates

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$Ba_2Y(Nb/Ta)O_6$ doped YBCO films on biaxially textured Ni-5at.% W substrates

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***Abstract*— The incorporation of nanoscaled pinning centers in superconducting films is one of the core topics for the optimization of coated conductors for applications. Therefore, a 1.6 µm thick film of pure $YBa_2Cu_3O_{7-\delta}$ (YBCO) and a similar film with additional 5 mol% of the mixed double-perovskite $Ba_2Y(Nb/Ta)O_6$ (BYNTO) were grown by pulsed laser deposition on buffered biaxially textured Ni-5at.% W tape with a growth rate of 1.6 nm/s. The doped samples nanostructure shows BYNTO rods and plates, that are incorporated with a defined crystallographic relationship towards the YBCO matrix as confirmed by X-Ray diffraction and transmission electron microscopy. Inductive measurements show only a slight decrease in the critical temperature T_c for the doped film. An improved homogeneity of the critical current density J_c over the sample was evaluated from trapped field profiles measured using a scanning Hall probe microscope. The mean J_c in rolling direction of the tape is 1.8 MA/cm² (77 K) and doubles the value of the undoped sample. Angular dependent measurements of the critical current density, $J_c(\Theta)$, show a decreased anisotropy of the doped film for various magnetic fields at 77 K as well as 64 K.**

***Index Terms*—BYNTO, pinning, pulsed laser deposition, RABiTS, YBCO**

I. Introduction

The use of high-temperature superconductors such as $YBa_2Cu_3O_{7-\delta}$ (YBCO) in applications as fault current limiters, motors, generators and high-field magnets, e.g. for fusion reactors, is limited by the current carrying capability in external magnetic fields. The critical current I_c can be increased by the immobilization of magnetic flux lines in the superconductor [1], [2]. These so-called pinning centers can be created in several ways, e.g. defect generation by irradiation [3], rare-earth mixing [4], substrate decoration [5], multi-layering approaches [6] or the incorporation of nanoparticles [7], [8], such as the mixed double-perovskite $Ba_2Y(Nb/Ta)O_6$ (BYNTO) [9], [10] where size, shape, density and distribution of the dopant can be adjusted to reach the favored performance in certain magnetic field and temperature regimes. A lower anisotropy is desirable for high-field coils based on superconducting tapes, where the lowest value of $I_c(B,\Theta)$ is the limiting criterion for applications. BYNTO-doped YBCO films on single-crystalline $SrTiO_3$ show pinning forces amongst the highest values ever reported, very high critical current density, J_c, values in magnetic fields up to 9 T, and strong matching effects in the $J_c(B,\Theta)$ behavior correlated to columnar grown BYNTO nanorods in the YBCO matrix [10]. Our goal is to transfer these structures to technical rolling-assisted biaxially textured substrates (RABiTS), which are produced in an industrial scale. Here, we present the structural and electrical properties of a 5 mol% BYNTO doped YBCO (BYNTO:YBCO) film, deposited on biaxially textured Ni-5 at.% W (Ni5W) [11] tapes by means of X-Ray diffraction, scanning and transmission electron microscopy, scanning Hall probe microscopy and transport current measurements.

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II. Experimental Details

A. Sample preparation

YBCO films with a thickness of 1.6 µm were grown on biaxially textured Ni5W tapes of 80 µm thickness with a chemically deposited buffer layer system of ~270 nm thick $La_2Zr_2O_7$ and ~30 nm thin CeO_2 by pulsed laser deposition using a Lambda Physics LPX305Pro KrF excimer laser (λ = 248 nm) with an energy density of 1.6 J/cm² at the target surface and a pulse frequency of 10 Hz (mean growth rate of 1.6 nm/s). All targets were prepared from precursor powder mixtures according to the stoichiometry of undoped YBCO and YBCO with additional 2.5 mol% Ba_2YNbO_6 + 2.5 mol% Ba_2YTaO_6 by pressing and sintering at 950°C in flowing oxygen for 24 h. Deposition took place in an atmosphere of 0.4 mbar at 830°C substrate temperature, which proved highest values of inductive critical temperature, T_c, and J_c in preliminary tests. Oxygen loading of the films was carried out at 765°C for 1 h in 400 mbar O_2 before cooling to room temperature. A silver cap layer of several 10 nm thickness was deposited to lower the contact resistance and protect the films.

B. Structural characterization

Transmission electron microscopy (TEM) was carried out on a FEI Osiris microscope operated at 200 kV as well as on a

FEI Titan³ electron microscope operated at 200 kV and 300 kV for high angle annual dark field scanning transmission electron microscopy (HAADF STEM) and energy dispersive X-ray spectroscopy (EDX).

The crystal structure was analyzed with a Bruker D8 Advance diffractometer (Co anode) in modified parallel-beam geometry (Θ-2Θ scans). The peak width is given as the full width at half maximum (FWHM) value from a Gaussian fit to the YBCO (005) peak in the Θ-2Θ plot. The *c*-axis parameter of YBCO was calculated from peak positions of the (00ℓ) peaks (ℓ > 6) by the Nelson-Riley formalism [12].

Pole figures of the YBCO (103) ($2\Theta = 32.5°$) and BYNTO (220) ($2\Theta = 29.9°$) [13] planes were taken at a Philip X`Pert PW3040 (Cu anode).

C. *Electrical properties*

The critical temperature $T_{c,50}$ and the transition width ΔT_c ($= T_{c,90} - T_{c,10}$) were measured inductively. Trapped field profile measurements were carried out at TU Wien to determine the local critical current, for detailed procedure see Ref. [14].

Angle resolved transport current measurements in magnetic fields, $J_c(B,\Theta)$, were conducted in maximum Lorentz force configuration in a four-point measurement assembly on laser-cut bridges [15] of 1 mm length and 300 µm width. Here J_c is defined by an electrical field criterion of 1 µV/cm.

III. Results and Discussion

A. *Structural properties*

The orientation of the grown films was studied with X-ray diffraction. The appearance of sharp YBCO (00ℓ) peaks (0.24° for pure YBCO, 0.32° for BYNTO:YBCO) in the Θ-2Θ scans proves the growth of highly *c*-axis oriented films on the chemically buffered Ni5W tapes, Fig. 1. An additional peak of the dopant is observed for BYNTO:YBCO, which is significantly broader and asymmetric. This might arise from the overlapping of several peaks of particle species with varying stoichiometry. Similar effects for Y-rich nanoparticles were reported by *Reich et al.* [8]. This is supported by the variation of the nanoparticle stoichiometry shown in the TEM-EDX data, Fig. 3.

A detailed analysis of the diffraction patterns indicated a change of the YBCO *c*-axis length from 11.69 Å (for the undoped YBCO, the increase of 0.10 Å compared to the bulk value is explained in Ref. [16]) to 11.72 Å (doped), respectively. A reduced oxygen content in the YBCO matrix is unlikely to account for the peak shift, as no reasonable drop in T_c is measured (Fig. 4). The shift of the YBCO peaks to lower 2Θ values for the doped sample in comparison to the undoped film might be explained by stretching the YBCO lattice in *c*-direction in order to match three unit cells of the cubic dopant ($a_{BYNTO} = 8.33$ Å) to two unit cells of YBCO ($c_{YBCO} = 11.68$ Å) by incorporating misfit dislocations similar to BHO-doped YBCO [16]. However, this needs to be confirmed with high resolution TEM.

Texture measurements were performed to study the in-plane alignment of the films. High intensities at $\varphi = 0°$ and 90° along $\psi = 45°$ in the BYNTO (220) pole figure (Fig. 2) indicate a biaxially oriented growth of the dopant inside the YBCO matrix. The poles are elongated along the rolling direction of the tape. Similar results are observed for the YBCO texture on such tapes [17]. The texture width, i.e. the texture quality, of all samples is in the typical range for YBCO on NiW tapes and only marginally influenced by the dopant.

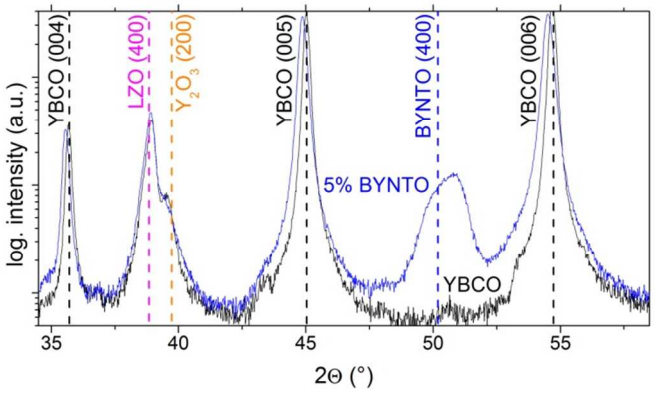

Fig. 1. X-ray diffraction pattern of pure YBCO and 5 mol% BYNTO:YBCO on Ni5W, dashed lines give theoretical peak positions.

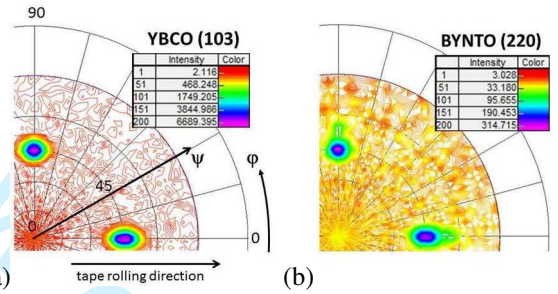

Fig. 2. Pole figures of the (a) YBCO (103) and (b) BYNTO (220) plane for 5 mol% BYNTO:YBCO on Ni5W tape.

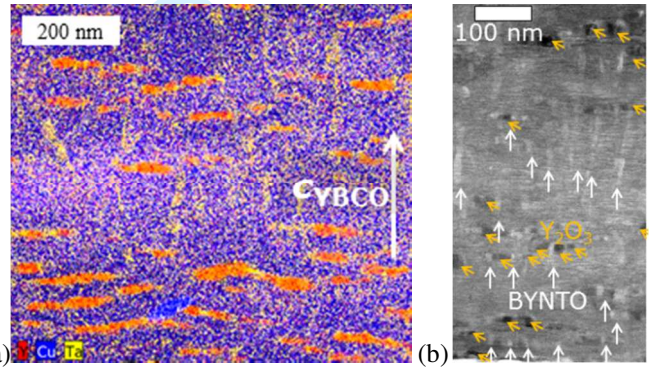

Fig. 3. (a) TEM-EDX analysis for Y (red), Copper (blue) and Tantalum (yellow) and (b) FIB cross section image (not the same area) of 5 mol% BYNTO:YBCO on Ni5W tape showing Y_2O_3 precipitates (orange arrows) and nanosized rods of BYNTO (white arrows) with a diameter of ~10 nm

Transmission electron microscopy reveals the formation of BYNTO nanostructures inside the YBCO matrix (Fig. 3). A few nanosized columns with a diameter of ~10 nm grow with a certain splay around the YBCO *c*-axis direction (Fig. 3(b))

and are decorated by Y_2O_3 particles. Furthermore we find large BYNTO plates parallel to tape surface (Fig. 3(a)), which have a slightly alternating stoichiometry that might explain the broad peak in the XRD pattern (Fig. 1). The microstructure with few nanocolumns and large plates is different from the results of BYNTO-doped YBCO on $SrTiO_3$ (STO) substrates [9], [10]. We think this discrepancy is a result of the chosen deposition parameters. Optimized deposition temperatures and/or growth rates are needed to produce more columnar or meandering structures.

B. Electrical properties

The BYNTO:YBCO sample has a T_c very close to the value of undoped YBCO with a sharp transition width (< 1 K) (Fig. 4). The critical current density maps, obtained from remanent field profiles on the complete sample surface, Fig. 5, show a doubling of the average self-field J_c in rolling direction of the tape at 77 K for the doped sample (1.8 MA/cm²) compared to the pure YBCO (0.9 MA/cm²) with a significantly improved uniformity of J_c along the complete tape length (Table I). The difference of J_c along the x and y axis has two reasons. First, the grains in RABiTS tapes such as Ni5W are usually slightly elongated in rolling direction leading to a better current percolation in this direction due to larger grain boundary areas ("brick wall model") and a larger number of grains per width (percolation threshold) [18]. Second, possible macroscopic defects such as grooves or scratches related to the rolling or winding processes may hinder current flow in transverse direction. This topic needs a further detailed analysis.

BYNTO doping enhances the critical current density over a wide angular range of the magnetic field with respect to the tape normal (Fig. 6). Whereas J_c for magnetic fields parallel to the tape surface (±10° around $B \| ab$) is slightly decreased, J_c is enhanced for all other directions. Most probably the randomly distributed Y_2O_3 nanoparticles are responsible for this matter. A pronounced c-axis peak ($B \| c_{YBCO}$) is not visible at 77 K (Fig. 6(a)) but is clearly observed at lower temperatures (65 K, Fig. 6(b)) for both, the doped and the undoped films. The small amount of c-axis-oriented nanorods is most likely causing the peak for the BYNTO-doped sample, in the case of pure YBCO it might be due to dislocations. The decrease in J_c for $B \| ab$ is surprising considering the large amount of ab-oriented platelets. Either these platelets act as random particles rather than ab-correlated ones, or a secondary species of ab-defects not visible in these TEM images, such as stacking faults [19] is reduced in density by the addition of BYNTO. However, BYNTO:YBCO shows a reduced anisotropy at different temperature and magnetic field regimes (Table II), e.g. the minimum J_c is 50% higher for the doped film at 77 K, 1 T.

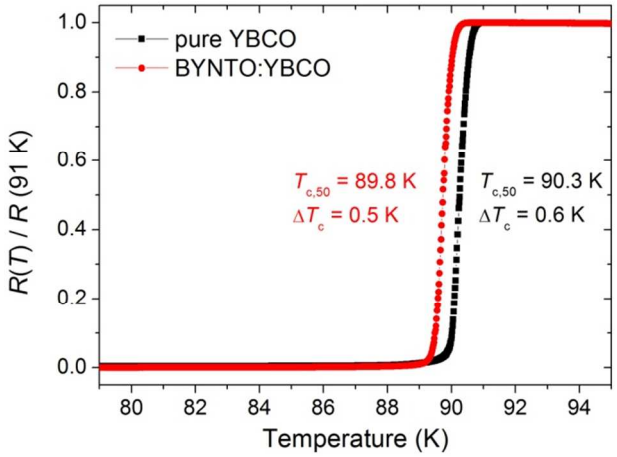

Fig. 4. Temperature-resistance curves of pure YBCO and 5 mol% BYNTO:YBCO on Ni5W tape.

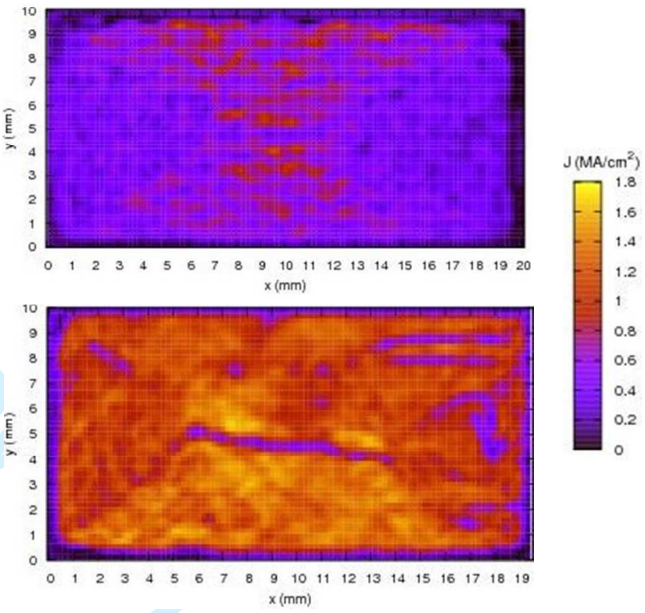

Fig. 5. Critical current density maps evaluated from trapped field profiles at 77 K for (a) pure YBCO and (b) 5 mol% BYNTO:YBCO on Ni5W tape.

TABLE I
AVERAGE $J_{C,X}$ (ROLLING DIRECTION) AND $J_{C,Y}$ (TRANSVERSE DIRECTION) OF PURE YBCO AND 5 MOL% BYNTO:YBCO ON NI5W TAPE

BYNTO CONTENT (MOL%)	$J_{C,X}$ (MA/CM²)	$J_{C,Y}$ (MA/CM²)	$J_{C,Y}/J_{C,X}$
0	0.9±0.2	0.5±0.1	0.6
5	1.8±0.2	1.5±0.2	0.8

TABLE II
J_C^{MAX}/J_C^{MIN} OF PURE YBCO AND 5 MOL% BYNTO:YBCO ON NI5W TAPE FOR SEVERAL TEMPERATURES AND MAGNETIC FIELDS

BYNTO CONTENT (MOL%)	ANISOTROPY (J_C^{MAX}/J_C^{MIN})			
	77 K 1 T	77 K 3 T	77 K 5 T	64 K 5 T
0	5.1	6.1	13.9	4.2
5	2.5	2.7	6.9	2.2

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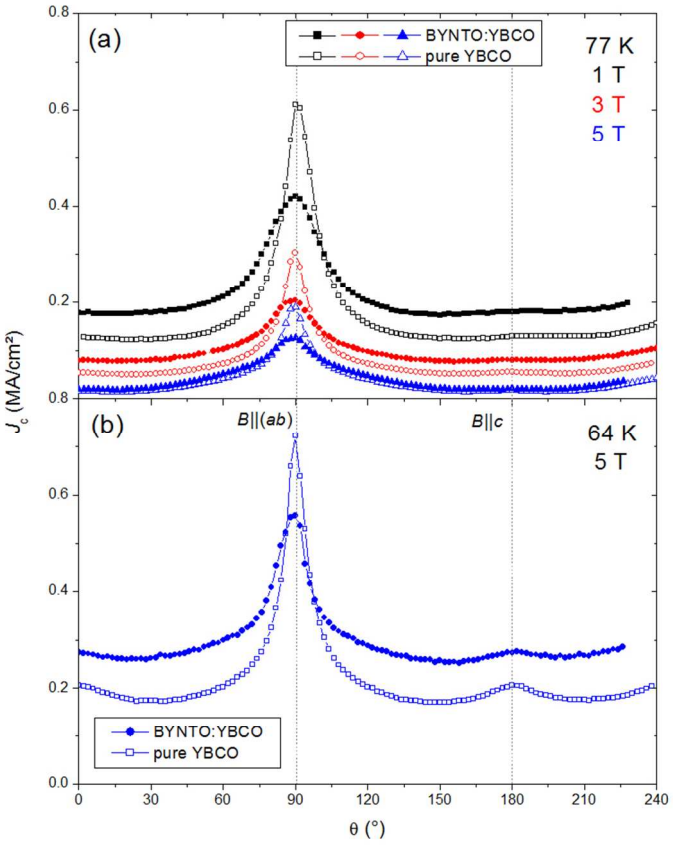

Fig. 6. Anisotropy of pure YBCO and 5 mol% BYNTO:YBCO on Ni5W tape at (a) 77 K and (b) 64 K for different magnetic fields.

CONCLUSION

In this work, we demonstrated the capability of producing 1.6 µm thick 5 mol% BYNTO:YBCO films on fully CSD-buffered Ni5W substrates with a reasonable growth rate. Nanosized BYNTO rods parallel to the YBCO *c*-axis and BYNTO plates parallel to the YBCO *ab*-plane are incorporated with biaxial orientation into the YBCO matrix. Inductive measurements show a high critical temperature and a very uniform average critical current density of 1.8 MA/cm² along the tapes rolling direction. BYNTO doping enhances the critical current in a wide range of magnetic field orientations. At lower temperatures (65 K), a *c*-axis peak becomes visible. Adjustment of the deposition parameters temperature and growth rate might further enhance the $J_C(B,\Theta)$ characteristics of BYNTO-doped YBCO films on biaxially textured Ni-W tapes.

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