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**ENHANCEMENT AND ANGULAR DEPENDENCE OF  
TRANSPORT CRITICAL CURRENT DENSITY IN PULSED  
LASER DEPOSITED  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x} + \text{BaSnO}_3$  FILMS IN  
APPLIED MAGNETIC FIELDS (POSTPRINT)**

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# Enhancement and angular dependence of transport critical current density in pulsed laser deposited $\text{YBa}_2\text{Cu}_3\text{O}_{7-x} + \text{BaSnO}_3$ films in applied magnetic fields

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$\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  (YBCO) films with nanoparticles of  $\text{BaSnO}_3$  (BSO) were processed using pulsed laser ablation of a special target made with dual phase sectors of YBCO and BSO. Transport critical current density ( $J_{ct}$ ) of these YBCO+BSO films in applied magnetic fields and angular dependence of  $J_{ct}$  on the applied field orientation was measured. It was observed that in the YBCO+BSO films, the  $J_{ct}$  ( $H\parallel c$  orientation) increased considerably as compared to regular YBCO films and was 1.3 times higher than  $J_{ct}$  in  $H\parallel ab$  orientation. Cross-sectional transmission electron microscopy images on YBCO+BSO films showed the presence of high density ( $3.5 \times 10^{11} \text{ cm}^{-2}$ ) of nanoparticles ( $\sim 10 \text{ nm}$  size) and nanocolumns that extended throughout the thickness of the films with high density of dislocations and stacking faults ( $1000 \mu\text{m}^{-2}$ ). The observed results of enhancements in  $J_{ct}$  in  $H\parallel c$  and  $J_{ct}$  in  $H\parallel ab$  orientations were discussed in the light of the observed microstructural details. © 2007 American Institute of Physics. [DOI: [10.1063/1.2783783](https://doi.org/10.1063/1.2783783)]

## I. INTRODUCTION

Improvements in the critical current density ( $J_c$ ) in applied magnetic fields are of great importance for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  (YBCO) coated conductors to be used in applications such as high temperature superconducting (HTS) generators and motors.<sup>1</sup> Flux pinning enhancement in YBCO films is one of the principal means to improve  $J_c$  in applied magnetic fields. The introduction of nanoparticles into YBCO films has been demonstrated as a viable method to introduce the required flux pinning centers for the YBCO coated conductors.<sup>2-5</sup> In addition to acting as flux pinning centers by themselves, the nanoparticles can also create defects, such as strain fields surrounding the particles, stacking faults, and dislocations, that can potentially enhance the flux pinning. Since the coils in HTS rotating machinery experience various magnetic field vectors, it is important to obtain pinning enhancements in all possible orientations of the applied magnetic field.

In undoped YBCO films, the  $J_{ct}$  is higher in the  $H\parallel ab$  orientation than the  $H\parallel c$  orientation due to the intrinsic pinning offered by the layered structure of YBCO.<sup>6</sup> Ideally, the pinning should maximize the transport current in the coated conductors while minimizing the angular dependence of the conductor to avoid a preferred orientation. This makes coil fabrication for HTS machines much simpler by avoiding field dependent designs. The addition of flux pinning centers

could change this anisotropic behavior of YBCO behavior by introducing  $c$ -axis correlated defects.<sup>7</sup> Since the addition of flux pinning centers could affect the angular dependence of  $J_{ct}$ , it is of interest to correlate the structure-property relationships in YBCO films processed with nanoparticles.

Several pinning materials including  $\text{Y}_2\text{BaCuO}_5$ ,  $\text{BaZrO}_3$ ,  $\text{BaIrO}_3$ ,  $\text{Nd}_2\text{O}_3$ , YSZ,  $\text{Y}_2\text{O}_3$ , and  $\text{BaSnO}_3$  (Refs. 2 and 8–13) have previously been investigated to improve the flux pinning properties of YBCO films. Depending upon the processing method, the composition, pinning material content, size, and distribution all determine the effectiveness of the nanoparticles for pinning enhancement. These differences in nanoparticle materials and processing methods result in YBCO films with pinning centers that can pin differently for particular magnetic field ranges. Hence, there is a need to identify a suitable material and processing method that will incorporate the nanoparticles in a manner that will obtain the desired effect.

Recently, YBCO+BSO (BSO= $\text{BaSnO}_3$ ) films were reported to have high  $J_c$  in applied magnetic fields showing more than an order of magnitude increase in  $J_c$ , determined by magnetization measurements in  $H\parallel c$  orientation at high fields  $> 2 \text{ T}$ .<sup>13</sup> These films were deposited using a pulsed laser ablation method, where a special target made with dual phase sectors was used. Nonlayered nanoparticles of  $\text{Y}_2\text{BaCuO}_5$  and BSO were introduced<sup>4,13</sup> using this method with considerable  $J_c$  improvements in YBCO. Melle *et al.*<sup>11</sup> also recently showed improvements in YBCO+YSZ (YSZ denotes yttria-stabilized zirconia) films processed by using

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an YBCO target with a YSZ sector. In the present paper, the critical transport current ( $J_{ct}$ ) measurements in applied magnetic fields and angular dependence of YBCO+BSO films processed by dual phase sector method were investigated. Cross-sectional transmission electron microscopy (TEM) results are presented, and the observed  $J_{ct}$  results are discussed in the light of the observed microstructures.

## II. EXPERIMENT

All the samples studied in the present work were processed by employing a pulsed laser ablation method using a dual sector pulsed laser deposition (PLD) target method as discussed in detail elsewhere.<sup>13</sup> Briefly, in this method, a thin sector (30° angle) cut from a disk of a sintered BSO target was attached to a top surface of an YBCO target. As the target was rotated, the BSO surface was ablated periodically, allowing the formation of BSO nanoparticles in the growing YBCO film. Due to the lattice mismatch between BSO and YBCO, nanoparticles were formed as opposed to continuous layers. All the depositions were done in a Neocera PLD chamber. A Lambda Physik KrF excimer laser (wavelength  $\lambda=248$  nm) was used to deposit films at an energy density of 2–4 J/cm<sup>2</sup>. The films were grown at a 4 Hz repetition rate. The substrate temperature was maintained at 780 °C during the deposition, while the substrate to target distance was set at 6 cm. The target was rotated at a speed of 15–20 rpm during the deposition of YBCO+BSO nanocomposite film. Films with  $\sim 300$  nm thickness were deposited on (100) LaAlO<sub>3</sub> single crystal substrates as well as on buffered biaxially textured Ni–W metallic substrates.

The pulsed laser ablation method discussed above with the dual phase sector target uses a single target to make the films in a scalable, continuous process. A particular advantage of this approach over a premixed target approach is that any possible chemical reactions between the pinning material and the YBCO in the target preparation stage are prevented as the second phase is physically separated from YBCO. The reaction time between the pinning material and YBCO coatings is reduced as the growth of the nanoparticles occurs simultaneously with the growth of YBCO film. Also, nanoparticles are introduced in a nonlayered fashion in contrast to an alternating target approach. The dual sector deposition approach provides a quick evaluation of different compositions by changing the second phase sector while maintaining the same YBCO target, thus avoiding differences that might be partially ascribed to differences in the batch of YBCO powder used.

The  $J_{ct}$  of the samples were measured by using a four probe transport current measurement method. Samples were prepared with 1–2 mm wide bridges using either laser ablation or chemical etch methods. The  $J_{ct}$  angular dependence was measured by rotating the sample through a range of  $-20^\circ$  to  $120^\circ$  about the sample normal in an applied magnetic field of 1–5 T at 77 K. The  $J_{ct}$  data were collected every  $2^\circ$  near  $H\parallel c$  and  $H\parallel ab$  orientations and every  $4^\circ$  elsewhere. A voltage criterion of 1  $\mu\text{V}/\text{cm}$  was used in the  $I$ - $V$  curves obtained at various angles to determine the  $I_c$  and calculate  $J_{ct}$ .

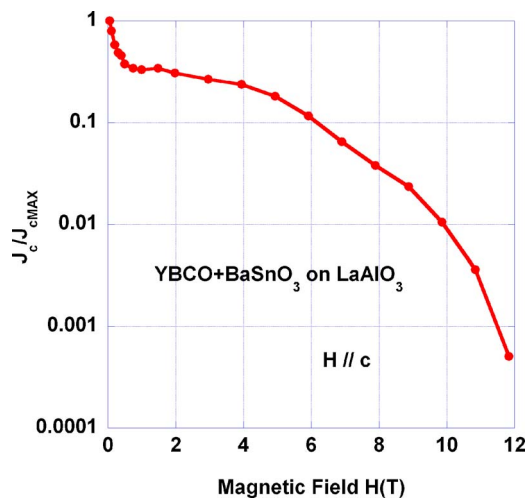


FIG. 1. Normalized plot of transport critical current density ( $J_{ct}$ ) in applied magnetic field of an YBCO+BSO sample on LaAlO<sub>3</sub> substrate at 77 K.

The film microstructure was studied by using a SIRION high resolution scanning electron microscope (SEM). The cross-section TEM samples were prepared through a conventional TEM sample preparation technique involving gluing, grinding, polishing, and ion milling. The detailed microstructure of these samples was characterized by TEM and high resolution TEM using a JEOL-2010 analytical electron microscope with a point-to-point resolution of 0.21 nm.

## III. RESULTS AND DISCUSSION

Figure 1 shows the normalized  $J_{ct}$  data of an YBCO+BSO sample on a LaAlO<sub>3</sub> substrate in an applied magnetic field of 0–12 T. Even though the  $J_{ct}$  at self-field was found to be around 1 MA/cm<sup>2</sup>, the  $J_{ct}$  in the applied magnetic fields was found to be enhanced considerably. The increased  $J_{ct}$  in these films is believed to be due to the presence of 10 nm sized BSO nanoparticles that were formed in the YBCO films (to be discussed later).

It can be seen from Fig. 1 that the rate of fall in  $J_{ct}$  with the applied magnetic field in YBCO+BSO samples was very low as compared to regular YBCO samples. To illustrate this more clearly, alpha ( $\alpha$ ) measurements were taken using a vibrating sample magnetometer on YBCO+BSO and regular YBCO samples in the region where  $J_c$  is proportional to  $B^{-\alpha}$ , where  $B$  is the magnetic field. The temperature dependence of  $\alpha$  over a temperature range of 20–77 K was measured and is presented in Fig. 2 for an YBCO control sample and two of the YBCO+BSO samples. It can be seen that the typical value for  $\alpha$  for the YBCO control sample is 0.5 and is temperature independent. However, for the YBCO+BSO samples,  $\alpha$  was found to be around 0.1 at low temperatures and it rises slowly to 0.3 at higher temperatures giving an average value of 0.2 in the temperature range of 20–77 K for YBCO+BSO samples. A low  $\alpha$  value indicates that  $J_c$  drops with the magnetic field at a much lower rate. Such low values of  $\alpha$  at 77 K were also reported in other pinned YBCO samples with BaZrO<sub>3</sub> (BZO) nanoparticles,<sup>8</sup> corroborating the data of low  $\alpha$  value for these highly pinned

2 YBCO+BSO samples.

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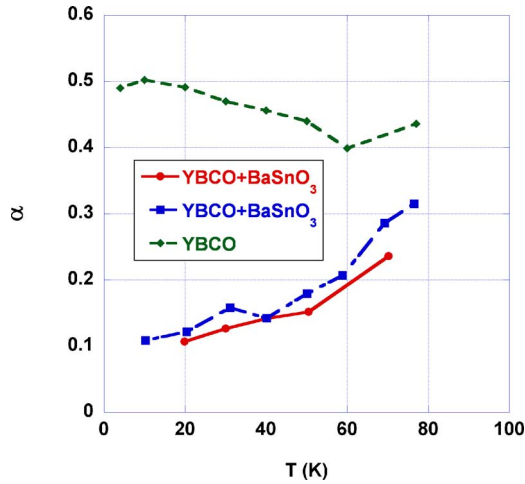


FIG. 2. Temperature dependence of  $\alpha$  values for a control YBCO and YBCO+BSO samples on  $\text{LaAlO}_3$  substrates.

Variations in  $J_{ct}$  with the applied magnetic field orientation to the sample normal at 77 K of an YBCO+BSO sample on a  $\text{LaAlO}_3$  substrate are shown in Fig. 3. The angular dependence at various applied fields of 1, 3, and 5 T was compared. It can be seen that the  $J_{ct}$  in  $H\parallel c$  orientation is higher than  $J_{ct}$  in  $H\parallel ab$  orientation in all of these applied fields. The presence of peaks in the  $J_{ct}$  in  $H\parallel c$  or  $H\parallel ab$  orientation is commonly attributed to the presence of correlated defects.<sup>6,7,14–16</sup> Evidence of the  $c$ -axis correlated defects in YBCO+BSO is provided in more detail later during the TEM analyses.

To compare the relative increase in  $J_{ct}$  in  $H\parallel c$  as compared to  $J_{ct}$  in  $H\parallel ab$  in YBCO+BSO films, angular dependence data of normalized  $J_{ct}$  with respect to  $J_{ct}$  in  $H\parallel ab$  are presented in Fig. 4. For comparison, data taken from a regular YBCO film on a buffered biaxially textured metallic Ni–W substrate (denoted as MS-6 in Fig. 4) are presented along with YBCO+BSO films on  $\text{LaAlO}_3$  (denoted as LAO in the Fig. 4) and Ni–W substrates. It can be seen that the  $J_{ct}$  in  $H\parallel c$  orientation is lower than  $J_{ct}$  in  $H\parallel ab$  in regular YBCO samples. However, in YBCO+BSO samples, the  $J_{ct}$  in  $H\parallel c$  is increased considerably. In this particular YBCO

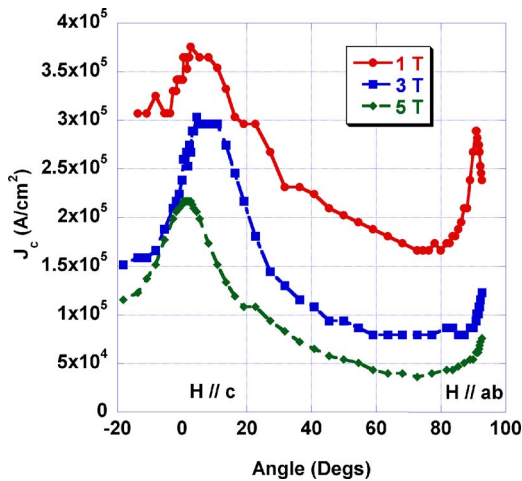


FIG. 3. Angular dependence of  $J_{ct}$  values of YBCO+BSO sample on a  $\text{LaAlO}_3$  substrate at various applied fields of 1, 3, and 5 T at 77 K.

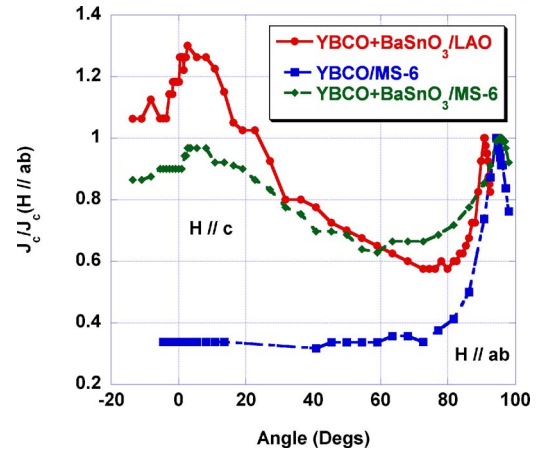


FIG. 4. Normalized  $J_{ct}$  with respect to  $J_{ct}$  in  $H\parallel ab$  of YBCO+BSO samples as compared to regular YBCO at 77 K (LAO= $\text{LaAlO}_3$  substrate and MS-6=buffered metallic substrate).

+BSO sample on  $\text{LaAlO}_3$ , the ratio of  $(J_{ct} \text{ in } H\parallel c)/(J_{ct} \text{ in } H\parallel ab)$  is about 1.3, whereas it is around 0.3 for regular YBCO.

Angular dependence of  $J_{ct}$  values of YBCO+BSO samples deposited on a buffered Ni–W metallic substrate was also measured to see if the improvements seen on  $\text{LaAlO}_3$  can also be observed on the more practical buffered metallic substrates. Figure 5 shows the actual  $J_{ct}$  values measured at 77 K in an applied field of 1 T, whereas the normalized values with respect to  $J_{ct}$  in  $H\parallel ab$  were shown earlier in Fig. 4. It can be seen that the  $J_{ct}$  in the  $H\parallel c$  orientation is increased compared to the  $J_{ct}$  in the  $H\parallel ab$  orientation similar to the behavior observed in the YBCO+BSO samples on  $\text{LaAlO}_3$  substrates. This illustrates that the angular dependence with YBCO+BSO is independent of the substrate used and is directly caused by the nanoparticles and/or the defects created by them. It can be seen that the  $J_{ct}$  of YBCO+BSO samples processed on buffered metallic substrates (shown in Fig. 5) is slightly higher than the  $J_{ct}$  of YBCO+BSO samples processed on  $\text{LaAlO}_3$  substrates (see  $J_{ct}$  at 1 T data in Fig. 3). The  $T_c$  of YBCO+BSO samples (90 K) processed on the buffered metallic substrates was noted to be slightly higher than the  $T_c$  of YBCO+BSO samples (88 K) pro-

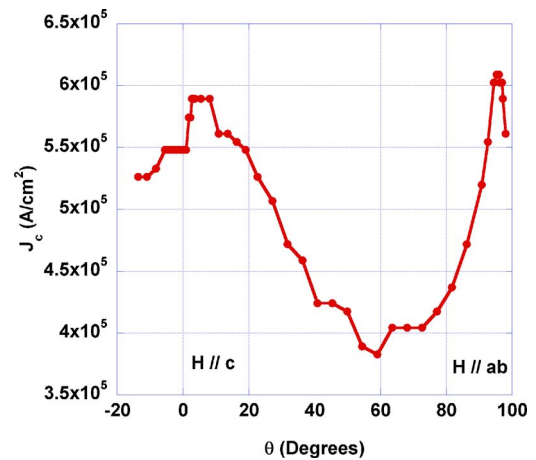


FIG. 5. Angular dependence of  $J_{ct}$  of an YBCO+BSO film on a buffered metallic Ni–W substrate in an applied magnetic field of 1 T at 77 K.

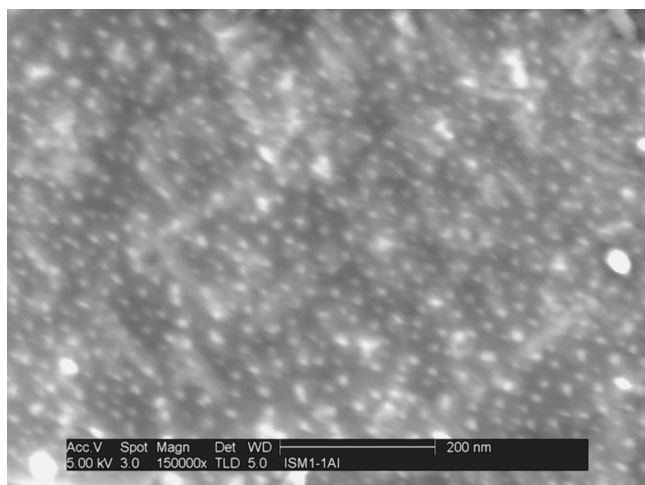


FIG. 6. Scanning electron micrograph of an YBCO+BSO sample on a buffered Ni-W metallic substrate.

cessed on  $\text{LaAlO}_3$  substrates. The slight increase in  $J_c$  could probably be due to the  $T_c$  differences. However, the  $J_{ct}$  behavior in the applied magnetic field in YBCO+BSO films, such as the increase in the  $H\parallel c$  orientation over  $H\parallel ab$  orientation, is similar irrespective of the substrates used.

Microstructural characterizations were carried out to understand the observed  $J_{ct}$  angular dependence. Figure 6 shows a scanning electron micrograph of an YBCO+BSO film on a buffered metallic substrate. The micrograph shows the presence of high density of nanoparticles ( $\sim 10$  nm bright particles) in an YBCO+BSO sample. It is known that as the applied magnetic field is increased, the magnetic flux line density increases in YBCO, and so accordingly the number density of pinning centers required to interact with the magnetic flux lines also needs to be high for high field applications. The number density of pinning centers in the present sample was estimated to be  $\sim 3.5 \times 10^{11} \text{ cm}^{-2}$  from the SEM pictures, which corresponds to a matching field of  $\sim 7$  T. This implies that at high magnetic field  $> 5$  T, the interaction between the flux lines and the particles was expected to be optimal.

In cross-sectional TEM images of the YBCO+BSO samples on  $\text{LaAlO}_3$  substrates, high quality epitaxial growth of YBCO/BSO nanocomposites was observed. Figure 7 shows the cross-sectional bright field TEM image obtained from one of the samples. The presence of nanocolumns extending the entire thickness of the films can be clearly seen. The average spacing between the nanocolumns is around  $\sim 20$ – $30$  nm. In the literature, nanorod structures were observed in YBCO+BZO or YBCO+YSZ films made using PLD of premixed targets.<sup>8,11</sup> In this study, without using a premixed target but using only a dual sector target, similar microstructural features were obtained. Also noteworthy, the columns seem to be formed as continuous rods as opposed to a structure of self-assembled nanodots as indicated for other YBCO+BZO samples.<sup>8</sup> The corresponding selected area diffraction pattern (also shown as an insert in Fig. 7) clearly shows the high quality epitaxial growth of YBCO/BSO on LAO substrate. The diffraction dots from BSO can be clearly seen and indexed to be BSO (002) and (020), correspond-

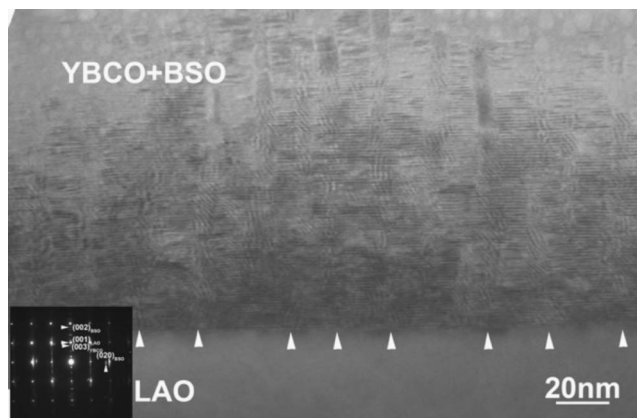


FIG. 7. Cross-sectional TEM image of an YBCO+BSO sample on a  $\text{LaAlO}_3$  substrate. The particles were identified to be  $\text{BaSnO}_3$  in the selected area diffraction pattern. Nanocolumns extending through the thickness, can be seen clearly, are marked by arrows.

ingly (marked in Fig. 7). The epitaxial relations are determined to be  $\text{YBCO}(003)\parallel\text{LAO}(001)\parallel(\text{BSO}(002))$  and  $\text{YBCO}(020)\parallel\text{LAO}(020)\parallel\text{BSO}(020)$ .

The fringe or moiré patterns in Fig. 7 around the rods are created due to the overlap of the YBCO and BSO lattices and their lattice mismatch. In order to reveal the detailed defect information at the interface of the YBCO/BSO, we conducted a detailed high resolution TEM (HRTEM) study on these cross-section samples. A high quality epitaxial growth of the YBCO/BSO nanocomposite was observed as shown in the HRTEM image given in Fig. 8. High density misfit dislocations and stacking faults generated at the interfaces of YBCO and BSO are shown in the corresponding fast Fourier transform (FFT) filtered image. The high density of these defects is strongly related to the high density of these BSO nanocolumns (spacing of  $\sim 20$  nm), the large lattice mismatch between YBCO and BSO ( $\sim 7\%$ ), and their vertical aligned orientation. From several cross-sectional images, the density of the misfit dislocations is estimated to be on the order of  $1000 \mu\text{m}^{-2}$ . In the FFT image, it is clearly shown that the formation of the stacking faults is closely related to the generation of misfit dislocations. Therefore, the density of the stacking faults is on the same order of magnitude as that of the misfit dislocations. The presence of these high density columnar defects (nanocolumns) and the  $c$ -axis cor-

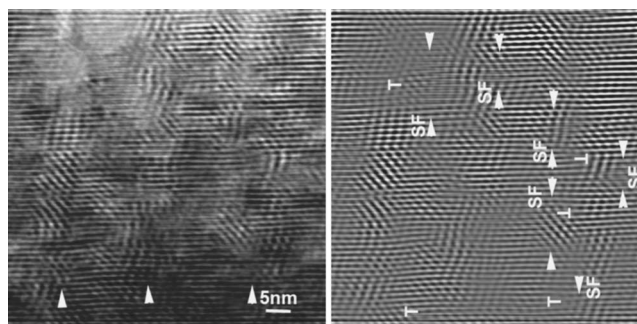


FIG. 8. High resolution TEM image of an YBCO+BSO film showing high density of defects. The FFT filtered image (shown on the right) shows the existence of high density of misfit dislocations and stacking faults at the YBCO/BSO interfaces.

related defects (misfit dislocations and stacking faults) could all contribute to enhance the  $J_{ct}$  in the  $H\parallel c$  orientation.

#### IV. CONCLUSIONS

It is shown that high quality YBCO+BSO films can be formed using a dual sector PLD with a BSO sector method on both  $\text{LaAlO}_3$  single crystal substrates as well as buffered Ni–W metallic substrates. These films were formed with high number density of nanoparticles ( $3.5 \times 10^{11} \text{ cm}^{-2}$ ) and nanocolumns with high density of dislocations and stacking faults. These defects and nanoparticles improved the transport current density ( $J_{ct}$ ) in both  $H\parallel c$  and  $H\parallel ab$  orientations.

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