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Peer Review File

Ultra-high critical current density and pinning force in nanostructured, superconducting REBCO-based, coated conductor

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The authors report the fabrication of $(Y_{0.5}Gd_{0.5})Ba_2Cu_3O_X$ and $(Y_{0.5}Gd_{0.5})Ba_2Cu_3O_X$ + 2vol% BaZrO (RE)BCO thin films that incorporate self-assembled BZO nanocolumns fabricated by pulsed laser ablation with record values in the latter of critical current density, Jc (H, T), and pinning force, Fp (H, T). The results have been achieved by substituting a rare-earth (RE) element (Gd) on the Y in combination with the incorporation of self-assembled nanorods of BZO formed via a simultaneous phase separation and strain-driven ordering process. Significantly, these record values extend over a wide range of temperature and applied field, which is particularly relevant to a broad range of potential technologically important applications in a number of engineering sectors, such as healthcare, power generation and progress towards an environmentally sustainable global economy. The potential impact of the work will be considerable if the deposition technology can be extended from localised thin films on technical substrates to longer length coated conductors. Overall, this work makes a significant and important contribution to the field.

General comments
The authors, quite rightly, describe the need for long lengths of coated conductor that support high current in high fields on a commercial scale, and the background to this work and the introduction to the paper focuses on this aspect. Although both heteroepitaxial films were grown on a flexible, technical substrate (IBAD-based Hastelloy C276 flexible tape with a buffer configuration of Al_2O_3/Y_2O_3 /IBAD MgO/$LaMnO_3/CeO_2$), however, it is misleading to claim the resulting structure is anything other than a thin film on a discrete substate. The extrapolation of the potential of this technique for the fabrication of long length of high current carrying HTS wire, albeit very important, is a considerable way off and is presented in a misleading way by the authors. Achieving these record values of key parameters is an important development in its own right and should not be diluted by unreasonably extending the potential of the technology to long length coated conductors, at least at this stage. The very impressive results should be described clearly as record-breaking values for thin film structures deposited on technical substrates, and not yet on long length, coated conductors.

Specific comments
Change throughout the paper:
REBCO → (RE)BCO
$REBa_2Cu_3O_X$ → $(RE)Ba_2Cu_3O_X$
Paragraph 2, page 2.
The statement regarding mile-long single crystal HTS wires should be changed to mile-long single crystal-like HTS wires.

Reviewer #2 (Remarks to the Author):

This paper by Goyal et al. is very interesting, as it focuses on record values of critical current density, Jc, and pinning force, Fp, in REBCO (Gd doped YBCO) coated conductors with self-assembled BZO nanocolumns fabricated using pulsed laser deposition. The paper can be improved further by addressing the following points:

(1) What is the nature of epitaxy and the interface (relaxed versus unrelaxed) between BZO and REBCO along a- and c-axes?
(2) How much pinning is caused by the nonsuperconducting BZO phase, interface dislocations (relaxed strain) and unrelaxed elastic strain? This can be explained using domain matching epitaxy paradigm.
(3) The authors should be aware of higher Jc reported in B-doped Q-carbon, where pinning is derived by interface atomic structure and phase characteristics (Nanoscale, 2019,11, 9141-9154, and reference therein). However, it is not in the form of long wires yet!
(4) The strain-contrast and HAADF imaging may shed more light, compared to HRTEM phase contrast imaging, on the nature of interfacial strain between BZO and REBCO.

Reviewer #3 (Remarks to the Author):

The manuscript presents very interesting and promising results on the critical current density and pinning force for superconducting cuprate REBCO thin films grown by pulsed laser deposition (PLD). In particular, it shows that the 2%vol of $BaZrO_3$ in $Y_{0.5}Gd_{0.5}Ba_2Cu_3O_x$ is able to reach 180 MA/cm^2 at 5 K and self-field and 80 MA/cm^2 at 5 K and 7 T, with pinning forces of 5.5 TN/m^3 at 5 K. The most relevant contribution is that this is achieved without process complication or cost increase, which makes it promising for the high temperature superconducting (HTS) materials industry, nowadays especially active thanks to the initiatives in magnetic fusion, as discussed in the manuscript. The results presented in this work give indeed hope for a further increase in material performance and, consequently, a reduction in the cost/performance ratio figure of merit, provided they could demonstrate them in films a factor 10 higher in thickness and in industrial competitive growth rates, since the industrially required property is the transport of high currents without losses.
However, while these results are interesting in themselves and deserve to be published, the content of the manuscript does not add sufficient knowledge in the area, bringing new ideas or innovations to the field to deserve its publication in Nature Communication. The reason for the good performance achieved is ascribed to a better control of the PLD system, without major details and studies. What was the critical parameter? The material, the nanostructuration, the methodology, the characterization used are well-known in the area, with one of the co-authors largely contributing in the past. Additional, some claims (as mentioned below) are overstretched and possibly wrong and should first be demonstrated experimentally.
-The remarkable results are achieved in the 2%vol BZO sample. What is the innovation that allows for these results? Is the interface between the BZO nanorods and YGdBCO different? How do the properties evolve with the volume content of BZO? How critical is that the matrix composition contains 50% Y and 50% Gd?
-Authors have used the correlation between the c-axis value, charge carrier density and doping level of $YBa_2Cu_3O_x$ to extrapolate the charge carrier density and doping level of $Y_{0.5}Gd_{0.5}Ba_2Cu_3O_x$ from its c-axis value and conclude that their sample is in the underdoped state. This assumes that the c-axis values for both superconducting materials are the same, which is not true. In fact, there is quite a difference specially if one wants to correlated to the charge carrier density and doping level due to the strong dependence with c-axis. In fact, the c-axis of GdBCO (11.7590 A0 for optimally doped) is much larger than that of YBCO (11.6767 A0), and therefore the claim that the film of this manuscript ($Gd_{0.5}Y_{0.5}BCO$) is in the underdoped state (larger c-axis than in the optimally doped) is not conclusive. For a matter of comparison, the article of Miura et al. NPG Asia Materials (2022) 14:85, shows how different are the values obtained for $Gd_{0.23}Y_{0.77}Ba_2Cu_3O_x$ or EuBCO (not cited). Therefore, it is not possible to extrapolate the values of charge carrier density for different REBCO compositions and instead this should be measured.
-c-axis value and Tc are affected by strain. Therefore, the c-axis value for YBCO films is different from that of nanocomposites due to the strain induced by the BZO/YBCO interfaces. This has been largely studied by the group of Prof. Wu (which recently has also demonstrated how to benefit from it. All these works have not either been properly cited either). The c-axis again increases in BZO-YBCO as compared to YBCO and therefore it is not possible to determine the charge carrier density from the published values on YBCO. Once more, the charge carrier density and doping level should be measured, by Hall effect or Thermopower measurements as has been previously reported by also Stangl et al, Sci Rep (2021) 11: 8176 or by E.F. Talantsev et al, APL 104, 242601 (2014).
-For both above reasons, it is very unlikely that the film of this manuscript is in the underdoped state given the values of Jc achieved. It is most probable that at least they are in the optimal state. Once again, experimental evidence should be given for the underdoped statements.
-Therefore, discussion on the value of the departing current and how far from the theoretical limit the nanocomposite sample is, should require a clear evidence of the doping level of the sample as well.
-Much emphasis is put in the interest of these results for the industry. However, to that end high thickness in the range of 1-2 micron should be demonstrated and specially for the High Rate PLD systems used in the industry. What is the growth rate used to achieve the results? How critical is the deposition rate to keep these results? What happens with the nanorod structure at the deposition rate of interest for the industry?
-The references used to categorize the achievements of today, are quite old (more than 10 years)

and they miss the results achieved by the community since then in several of the growth techniques (PLD, MOCVD, MOD, Reactive Evaporation, RCE-DR), also for industrial achievements.
-The TEM images from Figure 5 are not matching the quality of the images required for this investigation and published by many authors. Nowadays, there are good microscopes available for better analysis of the microstructure. The nanorods are difficult to be seen, their interfaces are not appreciated, their size and distance between each other is impossible to be determined by one of that images and statistics are required. Also the density of nanorods cannot be determined by the TEM images supplied.
-Additionally, the TEM technique is not the adequate technique to determine the thickness of a film, given the very small area under observation, though it can by an additional prove. Instead, perfilometry should be more adequate given the relevance of the thickness value in determining the value of the critical current density (thickness inversely proportion to Jc).
-Figure 6 compares the results from the manuscript with quite old results from many other authors. These should be now updated with present results from the community. Additionally, a consideration on the thickness should be done. It is easier to optimize thin films than thick films.
-The authors discuss the results of Jc(T,H) by the different types of defects present in the films. Many authors have explored this subject before, some have done a strong effort on developing models to identify the different types of defects. There is no reference on those, neither what model authors employ to propose what they claim. The fact that oxygen vacancies can be point defects is clear for the community nowadays, also that they can be localized inside other defects. All this discussion is incomplete in the manuscript.
-The authors calculate the microstrain of the layers, but they are not extracting additional information a part from commenting that they are of similar values as other PLD films. Angular dependent transport measurements to determine the anisotropy of the critical current densities at different magnetic fields and temperatures could help in the understanding of vortex pinning of these films.
-Finally, the results obtained by the authors are not so far from those published by Miura et al. NPG Asia Materials (2022) 14:85, in this later case for MOD growth technique. They demonstrated 130 MA/cm2 and pinning forces of 3 TN/m3 at 4.2 K. The later demonstrated that those samples were in the overdoped state by directly measuring the charge carrier density. Given the fact that the authors want to claim on a general breakthrough for the community, other growth techniques should have been checked and properly referenced.
For all these reasons, I confirm that this manuscript should not be published in Nature Communications.

REVIEWER COMMENTS

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The authors report the fabrication of (Y0.5Gd0.5)Ba2Cu3OX and (Y0.5Gd0.5)Ba2Cu3OX + 2vol% BaZrO (RE)BCO thin films that incorporate self-assembled BZO nanocolumns fabricated by pulsed laser ablation with record values in the latter of critical current density, Jc (H, T), and pinning force, Fp (H, T). The results have been achieved by substituting a rare-earth (RE) element (Gd) on the Y in combination with the incorporation of self-assembled nanorods of BZO formed via a simultaneous phase separation and strain-driven ordering process. Significantly, these record values extend over a wide range of temperature and applied field, which is particularly relevant to a broad range of potential technologically important applications in a number of engineering sectors, such as healthcare, power generation and progress towards an environmentally sustainable global economy. The potential impact of the work will be considerable if the deposition technology can be extended from localised thin films on technical substrates to longer length coated conductors. Overall, this work makes a significant and important contribution to the field.

We thank the reviewer for commenting that this work is a "***significant and important contribution to the field***" and that "***Overall, this work makes a significant and important contribution to the field"***.

General comments
The authors, quite rightly, describe the need for long lengths of coated conductor that support high current in high fields on a commercial scale, and the background to this work and the introduction to the paper focuses on this aspect. Although both heteroepitaxial films were grown on a flexible, technical substrate (IBAD-based Hastelloy C276 flexible tape with a buffer configuration of Al2O3/Y2O3 /IBAD MgO/LaMnO3/CeO2), however, it is misleading to claim the resulting structure is anything other than a thin film on a discrete substate. The extrapolation of the potential of this technique for the fabrication of long length of high current carrying HTS wire, albeit very important, is a considerable way off and is presented in a misleading way by the authors. Achieving these record values of key parameters is an important development in its own right and should not be diluted by unreasonably extending the potential of the technology to long length coated conductors, at least at this stage. The very impressive results should be described clearly as record-breaking values for thin film structures deposited on technical substrates, and not yet on long length, coated conductors.

We fully agree with the reviewer and have now clearly specified that these results are on laboratory samples for REBCO films deposited on a coated conductor substrate (see Abstract and Summary sections). These results nevertheless provide a motivation for wire manufacturers to further optimize their deposition processes to obtain higher-J_c values approaching those reported in this paper.

Specific comments
Change throughout the paper:
REBCO → (RE)BCO

This change has been made in the paper.

REBa2Cu3OX → (RE)Ba2Cu3OX

This change has been made in the paper.

Editorial Note: Figures on p6 & p8 reprinted with permission from Cantoni et al., "Strain-driven oxygen deficiency in self-assembled, nanostructured, composit oxide films" *ACS Nano*, **5**, 4783–4789 (2011). https://doi.org/10.1021/nn2007628, © 2011 American Chemical Society.

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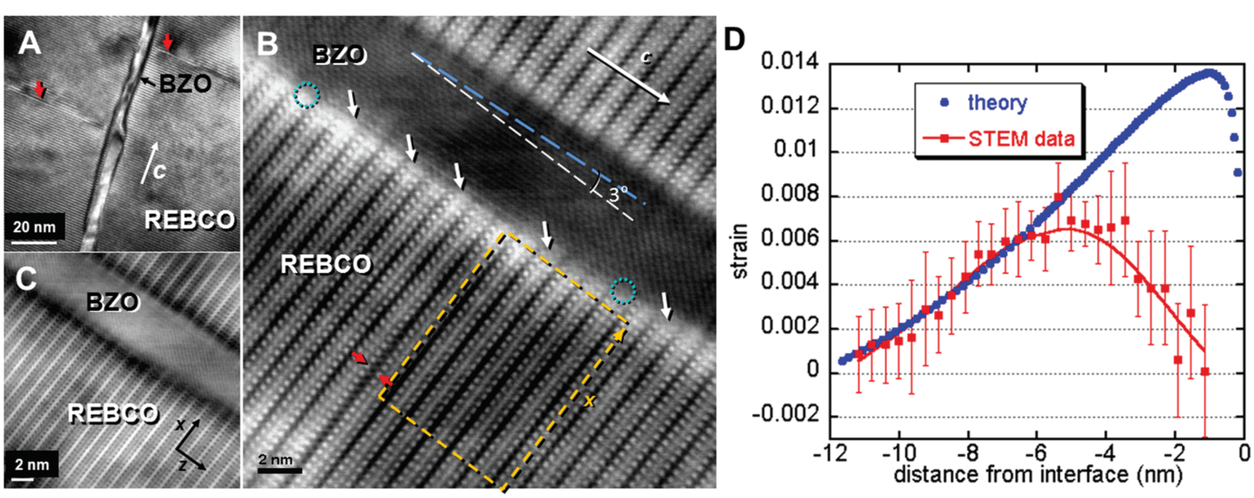
We thank the reviewer for commenting that this work is **very interesting**, *as it* ***focuses on record values of critical current density, J_c, and pinning force, F_p, in REBCO (Gd doped YBCO) coated conductors with self-assembled BZO nanocolumns fabricated using pulsed laser deposition***.

The paper can be improved further by addressing the following points:

(1) What is the nature of epitaxy and the interface (relaxed versus unrelaxed) between BZO and REBCO along a- and c-axes?

The film has relaxed partially. Details of the interface between BZO and REBCO was reported by us previously in the paper RS1 - "*Strain-Driven Oxygen Deficiency in Self-Assembled, Nanostructured, Composite Oxide Films*," Claudia Cantoni, Yanfei Gao, Sung Hun Wee, Eliot D. Specht, Jaume Gazquez, Jianyong Meng, Stephen J. Pennycook, and Amit Goyal, **ACS Nano**, 5 (2011) 4783-4789.
and also in RS2 - "Irradiation-free, columnar defects comprised of self-assembled nanodots and nanorods resulting in strongly enhanced flux-pinning in YBa2Cu3O7−δ films", AGoyal, SKang, K J Leonard, PMMartin, A A Gapud, MVarela, M Paranthaman, AOIjaduola, E D Specht, J R Thompson, D K Christen, S J Pennycook and F A List," **Supercond. Sci. Technol**. 18 (2005) 1533–1538.
There are misfit dislocations at the interface between BZO and REBCO film. In reference RS1, these are shown in Fig. 3 for a cross-section HRTEM/HAADF sample. This image is presented here for ease of viewing:



Editorial Note: Figure below reprinted with permission from Goyal et al. "Irradiation-free, columnar defects comprised of self-assembled nanodots and nanorods resulting in strongly enhanced flux-pinning in $YBa_2Cu_3O_{7-\delta}$ films," *Superconductor Science and Technology*, **18**, 1533 (2005), DOI 10.1088/0953-2048/18/11/021,

Figure 3. Local lattice deformation by high-resolution STEM imaging. (A) TEM micrograph of a sectioned BZO nanocolumn. (B) High-resolution Z-STEM image of the same nanocolumn in A. Dashed blue and white lines indicate the direction of the c axes of BZO and REBCO, respectively, showing a 3! tilt between the two lattices. Light blue circles indicate pure tilt dislocations; white arrows indicate misfit dislocations. (C) BF image acquired simultaneously to image B. (D) Comparison between analytically calculated strain (blue dots) and experimentally derived strain (red squares). The experimental strain is expressed using a c parameter obtained from interatomic distances within the yellow box in B. Red arrows in A and B indicate intergrowths having the Y124 structure.

In reference RS2, we show plan-view z-contrast, atomic-imaging of a BZO nanocolumn with misfit dislocations shown in Fig 2c of the paper. Again, for ease of viewing, this is shown below here:

(c)

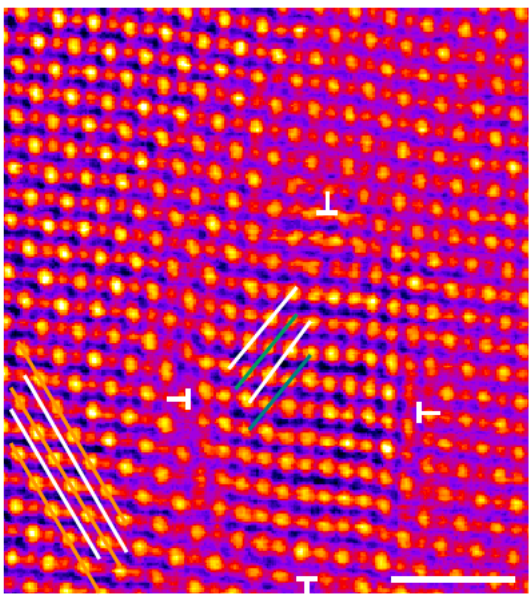


Fig. 2(c) Z-contrast STEM image of a single BZO nanodot. The extra semiplane in the edge dislocation cores is marked in white. Four misfit dislocations can be seen around the BZO nanodot. Since the BZO nanodots are aligned along the c-direction, the strain is also aligned, forming an extended defect. In the YBCO, the BaO planes have been marked with white lines, while CuO lines are marked with yellow lines. In the BZO nanoparticle, the ZrO planes have been marked with green lines, and the BaO planes again with white lines. The scale bar is 1.5 nm.

(2) How much pinning is caused by the nonsuperconducting BZO phase, interface dislocations (relaxed strain) and unrelaxed elastic strain? This can be explained using domain matching epitaxy paradigm.

This is not a simple analysis. We have pinning for H//c type configurations by extended BZO nanocolumns, collective pinning at lower temperatures by frozen-in oxygen point defects, pinning for H//ab configurations by stacking-faults such as the 124-type stacking fault with extra CuO plane, etc.

We have previously reported on detailed macro and micro strain as well as local strain and also theoretically calculated expected stratin using finite element methods in paper RS1 - "*Strain-Driven Oxygen Deficiency in Self-Assembled, Nanostructured, Composite Oxide Films*," Claudia Cantoni, Yanfei Gao, Sung Hun Wee, Eliot D. Specht, Jaume Gazquez, Jianyong Meng, Stephen J. Pennycook, and Amit Goyal, **ACS Nano**, 5 (2011) 4783-4789. Shown below is Fig. 1 of that paper:

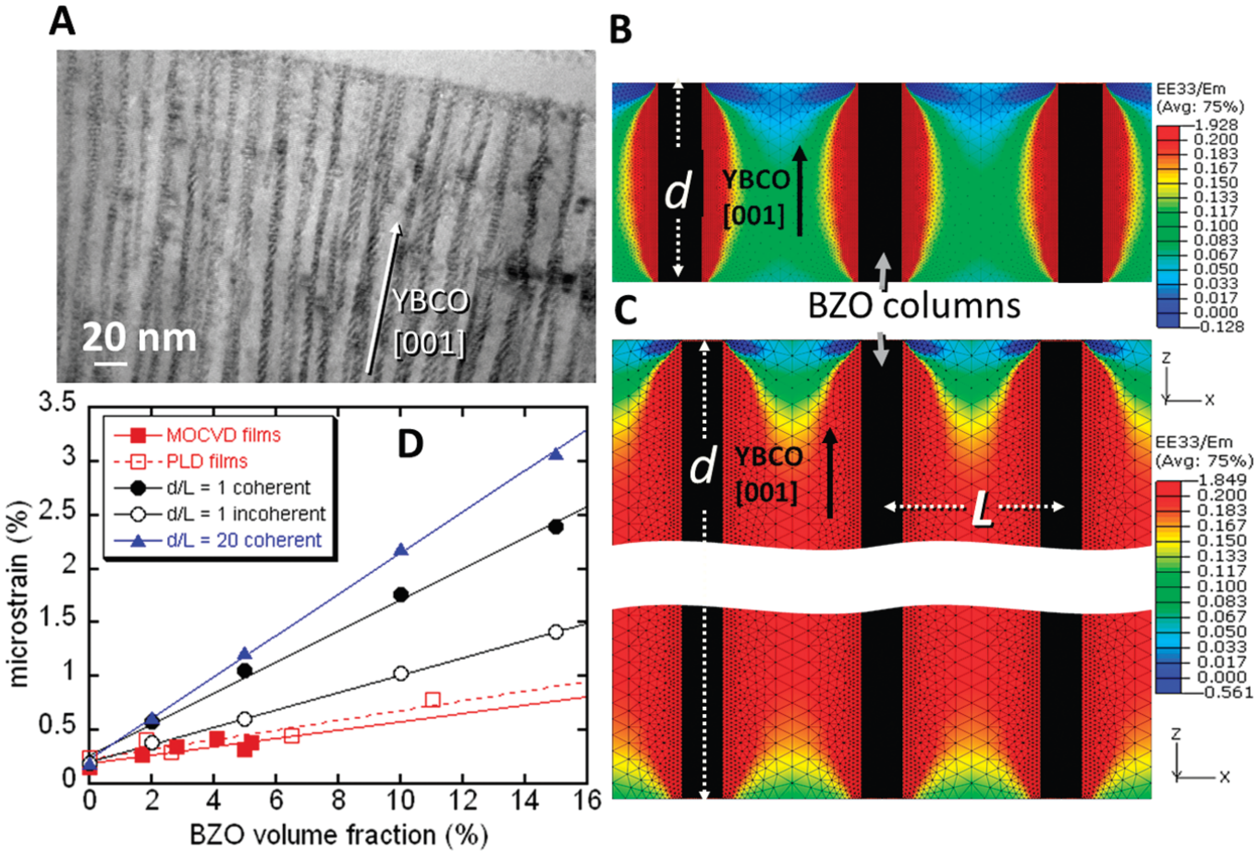


Figure 1. Evaluation of strain in BZO-doped REBCO composite films. (A) TEM micrograph illustrating the morphology of a 4 vol % BZO nanocolumn array embedded in the YBCO matrix. (B,C) Volumetric average of the out-of-plane strain (ε_{33}/f_o; f_o= lattice misfit) calculated through finite element method for the cases d/L = 1 (B) and d/L = 20 (C). (D) Strain versus BZO concentration as calculated by the finite element method for the cases d/L = 20 coherent interface (blue triangles), d/L = 1 coherent interface (black dots), d/L = 1 incoherent interface (black circles), and measured by X-ray diffraction for PLD fims (red, open squares) and MOCVD films (red, filled squares).

Fig. 3c in the same paper shown above shows how the local micro-strain varies as a function of distance from the REBCO/BZO interface.

Domain matching epitaxy paradigm developed by Narayan is indeed what explains the lattice-mismatched growth at all heteroepitaxial layers in the coated conductor substrate as well as for heteroepitaxial growth of the superconductor layer on the top oxide buffer layer.

Lastly, in Fig. 8 of the paper, we include strain maps obtained from geometrical phase analysis (GPA) on atomic resolution, HAADF-STEM images. The HAADF image beam current was 75 pA and probe size was 75 pm. The strain field was calculated with reference to the average spacing of atomic planes in the REBCO lattice. For strain mapping, the x, y direction and z direction are along the a-axis, b-axis and c-axis of REBCO primary lattice. The strain maps are color-coded with positive or tensile strain light green to red (20% strain) and negative or compressive strain from light green to blue. Fig. 8a shows a cross-section HAADF image of film showing vertical BZO nanorods. Fig. 8b and 8c show strain maps corresponding to e_{xx} and e_{zz} from GPA analysis of image shown in Fig. 8a. Fig. 8d shows a plan-view HAADF image of film matrix showing BZO nanorods. Fig. 8e and 8f show strain maps corresponding to e_{xx} and e_{yy} from GPA analysis of image shown in Fig. 8d. A typical pattern of tensile and compressive strain is seen around the BZO nanoparticles as expected and which is shown nicely in the strain maps. These maps are consistent with microstrain on a nanoscale we published previously.

(3) The authors should be aware of higher Jc reported in B-doped Q-carbon, where pinning is derived by interface atomic structure and phase characteristics (Nanoscale, 2019,11, 9141-9154, and reference therein). However, it is not in the form of long wires yet!

We thank the reviewer for pointing this excellent work. The work by Prof. J. Narayan's group on the discovery of superconductivity in B-doped Q-carbon is indeed very exciting and could be transformative. We look forward to reports from Prof. Narayan's group on wire fabrication using Q-carbon.

(4) The strain-contrast and HAADF imaging may shed more light, compared to HRTEM phase contrast imaging, on the nature of interfacial strain between BZO and REBCO.

Fully agree. Please refer to the four additional microscopy figures added to the paper and in particular, Fig.8 for the strain.

Reviewer #3 (Remarks to the Author):

The manuscript presents very interesting and promising results on the critical current density and pinning force for superconducting cuprate REBCO thin films grown by pulsed laser deposition (PLD).

We thank the reviewer for commenting that this manuscript presents **very interesting and promising results** *on the critical current density and pinning force for superconducting cuprate REBCO thin films grown by pulsed laser deposition (PLD).*

In particular, it shows that the 2%vol of BaZrO3 in Y0.5Gd0.5Ba2Cu3Ox is able to reach 180 MA/cm2 at 5 K and self-field and 80 MA/cm2 at 5 K and 7 T, with pinning forces of 5.5 TN/m3 at 5 K. The most relevant contribution is that this is achieved without process complication or cost increase, which makes it promising for the high temperature superconducting (HTS) materials industry, nowadays especially active thanks to the initiatives in magnetic fusion, as discussed in the manuscript. The results presented in this work give indeed hope for a further increase in material performance and, consequently, a reduction in the cost/performance ratio figure of merit, provided they could demonstrate them in films a factor 10 higher in thickness and in industrial competitive growth rates, since the industrially required property is the transport of high currents without losses.

We agree with the reviewer and thank the reviewer for commenting that "***this work give indeed hope for a further increase in material performance and, consequently, a reduction in the cost/performance ratio figure of merit"***, *provided HTS wire manufacturers could demonstrate them in films a factor 10 higher in thickness and in industrial competitive growth rates, since the industrially required property is the transport of high currents without losses.*

We have also included a reference for in-situ MOCVD films from low thicknesses to ~ 4.5 μm thick films with no degradation of J_c. In fact, stabilization of REBCO film growth and prevention of microstructural breakdown is a key attribute of REBCO films with self-assembled BZO nanorods.

The reason for the good performance achieved is ascribed to a better control of the PLD system, without major details and studies. What was the critical parameter? The material, the nanostructuration, the methodology, the characterization used are well-known in the area, with one of the co-authors largely contributing in the past.

We agree with the reviewer that this is an important point. So, we have added a lot more information on the film fabrication as well as included significant additional microstructural characterization and added 4-5 new figures that address this concern. This additional data and figures provide a very detailed characterization of the films.

Additional, some claims (as mentioned below) are overstretched and possibly wrong and should first be demonstrated experimentally.
-The remarkable results are achieved in the 2%vol BZO sample. What is the innovation that allows for these results? Is the interface between the BZO nanorods and YGdBCO different? How do the properties evolve with the volume content of BZO? How critical is that the matrix composition contains 50% Y and 50% Gd?

The answers to the questions above are shown below and all this information is included in the modified manuscript:
- The key difference here is the combined pinning by dual rare-earth point pins, BZO nanocolumns, oxygen point defects and CuO stacking faults.
- Carefully controlling the growth parameters in pulsed laser deposition is crucial to get good properties. We now include details of the growth procedure and show that we get these nice results only when the plume configuration is as shown and intersecting with the substrate surface. In such as case, the actual temperature of the substrate surface is likely also significantly higher than a configuration wherein the laser plume is not intersecting the sample surface.
- HAADF imaging plan-view has revealed that the BZO nanocolumns, much like in MOD samples have facets, implying that during the growth of the film some liquids similar to that in MOD growth may have been present. It appears that this higher temperature is important to obtain such remarkable performance.
- The areal density of BZO nanorods achieved was over an order of magnitude higher than what we observed previously in PLD YBCO films with nominal 2vol% BZO additions [48].
- The average spacing of BZO nanorods was also less at 15nm as opposed to 20nm.
- Another key finding was that under the deposition conditions used, BZO nanorods contain Y and Gd, which we did not observe in YBCO films with 2 vol% BZO additions [48]. The presence of Y and Gd in BZO nanorods may also be a result of the higher effective deposition temperatures.
- No atomic ordering of Y and Gd atoms in the REBCO lattice was observed.
Please refer to the modified paper for additional details.

We will explore variation of BZO volume % in future work. However, in our previous work with YBCO+BZO, the properties improve until 4vol% BZO additions and then increasing BZO content is detrimental. See reference 48 of this paper where we varied BZO from 0-10vol% [Wee et al., "*Engineering nanocolumnar defect configurations for optimized vortex pinning in high temperature superconducting nanocomposite wires*," Sci Rep 3, 2310 (2013). https://doi.org/10.1038/srep02310].

Lastly, w.r.t. Y and Gd composition, past work using MOCVD has shown that superior properties are obtained with higher total RE compositions, since it results in additional, randomly-oriented RE_2O_3 nanoparticles which can provide additional pinning (see reference 32 of paper). This provides an additional avenue to further increase performance in future work that we will probe. Hence, the composition we have used is not the limit or the only composition where excellent properties can be obtained.

-Authors have used the correlation between the c-axis value, charge carrier density and doping level of YBa2Cu3Ox to extrapolate the charge carrier density and doping level of Y0.5Gd0.5Ba2Cu3Ox from its c-axis value and conclude that their sample is in the underdoped state. This assumes that the c-axis values for both superconducting materials are the same, which is not true. In fact, there is quite a difference specially if one wants to correlated to the charge carrier density and doping level due to the strong dependence with c-axis. In fact, the c-axis of GdBCO (11.7590 A0 for optimally doped) is much larger than that of YBCO (11.6767 A0), and therefore the claim that the film of this manuscript (Gd0.5Y0.5BCO) is in the underdoped state (larger c-axis than in the optimally doped) is not conclusive. For a matter of comparison, the article of Miura et al. NPG Asia Materials (2022) 14:85, shows how different are the values obtained for Gd0.23Y0.77Ba2Cu3Ox or EuBCO (not cited). Therefore, it is not possible to extrapolate the values of charge carrier density for different REBCO compositions and instead this should be measured.

First, we thank the reviewer for pointing us to the nice paper by Miura. This is excellent work and is now extensively referenced in this manuscript. Second, we fully agree with the reviewer on all points. These figures have been removed from the manuscript.

-c-axis value and Tc are affected by strain. Therefore, the c-axis value for YBCO films is different from that of nanocomposites due to the strain induced by the BZO/YBCO interfaces. This has been largely studied by the group of Prof. Wu (which recently has also demonstrated how to benefit from it. All these works have not either been properly cited either). The c-axis again increases in BZO-YBCO as compared to YBCO and therefore it is not possible to determine the charge carrier density from the published values on YBCO. Once more, the charge carrier density and doping level should be measured, by Hall effect or Thermopower measurements as has been previously reported by also Stangl et al, Sci Rep (2021) 11: 8176 or by E.F. Talantsev et al, APL 104, 242601 (2014).

Once again, we fully agree with the reviewer comments and thank the reviewer for the comments. As suggested by the reviewer, we have conducted Hall measurements at a field of 9T at 300K. The measured carrier density is now reported in the paper.

Our group was also the first to report on the growth mechanism of self-assembled nanorods via a spontaneous phase separation and strain-driven self-assembly via a combined experimental, theoretical, and computational study, as reported in paper RS3 - "*Self-Assembly of Nanostructured, Complex, Multication Films via Spontaneous Phase Separation and Strain-Driven Ordering*," Sung Hun Wee, Yanfei Gao, Yuri L Zuev, Karren L More, Jianyong Meng, Jianxin Zhong, George M Stocks, Amit Goyal, **Advanced Functional Materials**, 23 (2013) 1912-1918; DOI: 10.1002/adfm.201202101. We have now included this paper in the references.

We have also included the key paper from WU's group related to this aspect (microstrain related to self-assembly) in the references.

-For both above reasons, it is very unlikely that the film of this manuscript is in the underdoped state given the values of Jc achieved. It is most probable that at least they are in the optimal state. Once again, experimental evidence should be given for the underdoped statements.

Experimental evidence for the state of the film is now included. The measured carrier density shows that the film is in a slightly underdoped state in light of Miura's results shown in Fig. 1 of Miura. The T_c (zero) of our film is also 86.5K and the R vs T shape is also consistent with it being underdoped.

-Therefore, discussion on the value of the departing current and how far from the theoretical limit the nanocomposite sample is, should require a clear evidence of the doping level of the sample as well.

The doping level is now addressed via measurement of the carrier density.

-Much emphasis is put in the interest of these results for the industry. However, to that end high thickness in the range of 1-2 micron should be demonstrated and specially for the High Rate PLD systems used in the industry. What is the growth rate used to achieve the results? How critical is the deposition rate to keep these results? What happens with the nanorod structure at the deposition rate of interest for the industry?

Please note that when the first report of strain-driven self-assembly in YBCO films was reported by us in RS2 - "Irradiation-free, columnar defects comprised of self-assembled nanodots and nanorods resulting in strongly enhanced flux-pinning in $YBa_2Cu_3O_{7-\delta}$ films", AGoyal, SKang, K J Leonard, PMMartin, A A Gapud, MVarela, M Paranthaman, AOIjaduola, E D Specht, J R Thompson, D K Christen, S J Pennycook and F A List," **Supercond. Sci. Technol**. 18 (2005) 1533–1538, the films were also just 0.2 μm thick. Following that report, industrial wires now have kilometer-long films which are a factor of 10 thicker with the similar performance to what was reported in RS2.

it is important. to note that incorporation of self-assembled BZO nanorods stabilizes the growth of REBCO films (thicknesses 0.2 μm to 4.5μm) allowing fabrication of very thick films with little or no microstructural degradation **[16, 48, 53]**. It has been reported that in MOCVD $(Y,Gd)Ba_2Cu_3O_x$ + BZO films, there is linear increase in I_c with a constant J_c in going from thin-films to ~ 4.5 μm thick films using roll-to-roll deposition at reasonable deposition rates **[53]**. Given this observation, it may be possible to sustain the high J_c (H, T) reported in this work to thicker films needed for high-I_c HTS wires.

The growth rate used for deposition of the films is listed in the paper and is 30nm/min. We have not yet explored higher growth rates and this will be subject of our future work. Please note that it has been shown by others that even at very high growth rates of 100nm/sec, the self-assembled BMO columnar nanorod microstructure is maintained - See reference: "Ultra-fast growth (up to 100 nm s^{-1}) of heavily doped $EuBa_2Cu_3O_7$ film with highly aligned $BaHfO_3$ nanocolumn structure," Yue Wu, Guangyu Jiang, Jiamin Zhu, Donghong Wu, Xueling Quan, Jiangtao Shi, Hongli Suo and Yue Zhao, Supercond. Sci. Technol. 34 (2021) 05LT01; https://doi.org/10.1088/1361-6668/abece6.

We hope that once this paper is published efforts by wire manufacturers to further optimize their deposition conditions will yield such performance in thicker films and in long wires. We may also focus on thicker films in future work but we are limited with the laser we have (industry has better and more powerful lasers).

Lastly, as was pointed out in the paper, the 0.2 μm thick films reported in this paper, even at this thickness, have comparable I_c (A/4mm) as commercial HTS wires having YBCO layers that are 2-3 μm thick!

-The references used to categorize the achievements of today, are quite old (more than 10 years) and they miss the results achieved by the community since then in several of the growth techniques (PLD, MOCVD, MOD, Reactive Evaporation, RCE-DR), also for industrial achievements.

Stangl's reference (published in 2021) and the comparison data included in that paper are all recent. We have now included Miura's reference in. the paper. Here we are focusing on the highest performance results and have included ALL papers we could find with such high-performance results.

-The TEM images from Figure 5 are not matching the quality of the images required for this investigation and published by many authors. Nowadays, there are good microscopes available for better analysis of the microstructure. The nanorods are difficult to be seen, their interfaces are not appreciated, their size and distance between each other is impossible to be determined by one of that images and statistics are required. Also the density of nanorods cannot be determined by the TEM images supplied.

We have added 4-5 new figures. We have the highest resolution microscope available TODAY, the Thermofisher Spectra. All aspects of the nanorods are now clear. Several additional details are summarized below:

- The areal density of BZO nanorods was 2.33 x 10^{13} / cm^2, an order of magnitude higher than what we observed previously in PLD YBCO films with nominal 2vol% BZO additions [48].
- The average spacing of BZO nanorods was also less at 15nm as opposed to 20nm.
- Another key finding was that under the deposition conditions used, BZO nanorods contain Y and Gd, which we did not observe in YBCO films with 2 vol% BZO additions [48]. The presence of Y and Gd in BZO nanorods may also be a result of the higher effective deposition temperatures.
- No atomic ordering of Y and Gd atoms in the REBCO lattice was observed.

-Additionally, the TEM technique is not the adequate technique to determine the thickness of a film, given the very small area under observation, though it can by an additional prove. Instead, perfilometry should be more adequate given the relevance of the thickness value in determining the value of the critical current density (thickness inversely proportion to Jc).

We fully agree with the reviewer that it is best to measure film thickness by profilometry. In TEM, if the film is very rough one can obtain a lower thickness in some local areas. If the film is smooth and has a uniform thickness (as is the case here), one can obtain a slightly higher thickness, since TEM thin foils are made at a very shallow angle through the film thickness to obtain an electron transparent region for microscopy and hence and a higher film thickness can be observed. The profilometer scan included in the supplementary materials nicely shows that the film thickness is 200 nm. We have now used the thickness determined from the profilometer scan for the film thickness as it more representative of the film thickness.

-Figure 6 compares the results from the manuscript with quite old results from many other authors. These should be now updated with present results from the community. Additionally, a consideration on the thickness should be done. It is easier to optimize thin films than thick films.

The paper from Stangl et al. was from 2021 and it included the highest-performance results from the community until then. The additional new high-performance result beyond that is from Miura et al. in 2022 and this is now included as a comparison in Figure 6 for PLD samples and also MOD samples.

-The authors discuss the results of Jc(T,H) by the different types of defects present in the films. Many authors have explored this subject before, some have done a strong effort on developing models to identify the different types of defects. There is no reference on those, neither what model authors employ to propose what they claim. The fact that oxygen vacancies can be point defects is clear for the community nowadays, also that they can be localized inside other defects. All this discussion is incomplete in the manuscript.

We have now included reference to our own previous papers as well as others, in particular that of Miura et al. for: "*Thermodynamic approach for enhancing superconducting critical current performance*," Masashi Miura, Go Tsuchiya, Takumu Harada, Keita Sakuma, Hodaka Kurokawa, Naoto Sekiya, Yasuyuki Kato, Ryuji Yoshida, Takeharu Kato, Koichi Nakaoka, Teruo Izumi, Fuyuki Nabeshima, Atsutaka Maeda, Tatsumori Okada, Satoshi Awaji, Leonardo Civale and Boris Maiorov, Miura et al. **NPG Asia Materials** (2022) 14:85, https://doi.org/10.1038/s41427-022-00432-1. W.r.t. type of defects and how they affect pinning in different (H,T), see comments to question below.

-The authors calculate the microstrain of the layers, but they are not extracting additional information a part from commenting that they are of similar values as other PLD films. Angular dependent transport measurements to determine the anisotropy of the critical current densities at different magnetic fields and temperatures could help in the understanding of vortex pinning of these films.

We appreciate that angular transport measurements are helpful and we have published these extensively previously. See references - Ref. 15, 16, 22, 24, 44. These particular measurements were not a focus of the current work. What we have observed previously, namely, that BZO nanocolumns affect J_c most for H//c configurations, stacking faults for H//ab configurations and point defects have a universal enhancement in J_c holds true for these films as well. Splay in the alignment of the BZO nanocolumns results in a broader H//c peak. All of this was reported previously by us and others.

-Finally, the results obtained by the authors are not so far from those published by Miura et al. NPG Asia Materials (2022) 14:85, in this later case for MOD growth technique. They demonstrated 130 MA/cm2 and pinning forces of 3 TN/m^3 at 4.2 K. The later demonstrated that those samples were in the overdoped state by directly measuring the charge carrier density. Given the fact that the authors want to claim on a general breakthrough for the community, other growth techniques should have been checked and properly referenced.

We have compared our results with that reported by Miura et al. The results reported here are still a massive improvement in J_c and F_p at all H, T. Also, to compare directly with Miura's results, we now report our 4.2K data also. In addition, the films in this work are underdoped and Miura's films are heavily overdoped. We have found in this work, all details of a fabrication are important in dictating J_c and can affect high-performance. We hope that research groups who are experts in other fabrication processes such as CSD/MOD, MOCVD, etc. will be motivated by these results to further optimize their film fabrication and deposition processes to obtain similar or higher performance.

Given the significant new results, carrier concentration measurements, profilometer characterization as well as detailed HAADF-STEM characterization of microstructure as details of film synthesis, we hope that the paper is now acceptable to publication. We believe that all key suggestions by the reviewers have been fully addressed and have made the paper better.

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The authors have addressed my concerns in the revision of their manuscript, which I now feel suitable for publication.

Reviewer #2 (Remarks to the Author):

This manuscript has addressed most of my concerns, and I recommend the publication as is.

Reviewer #3 (Remarks to the Author):

This reviewer acknowledges the modifications to the manuscript in this second version. The authors have achieved most of the points raised. In particular, I would like to rate very positively the detailed and outstanding TEM work now included, the additional measurements of charge carrier density which clarifies the doping state and the description of the growth system explained in the methods. The manuscript has now relevant insights on the process and characterization of the layers which supports the high values of the critical currents achieved. There are just few minor changes that should be attained before publication.

1. The title of the manuscript suggests that the work presented is done on wires which is not the case. Therefore, the title should be modified to "Ultra-high Critical Current Density and Pinning Force in REBCO-based, Nanostructured Superconducting Films". The future implications of this work to wire manufacturing is already described in the abstract.

2. In the abstract where it says "The key to making the price/performance metric favourable is to significantly enhance the performance", should say "One of the keys to making the price/performance metric favourable is to significantly enhance the performance", since there are other ways to also enhance this metric which are being followed by other researchers like increasing thickness, growth rate, double side tapes, yield, manufacturing throughput, substrate thickness, …

3. I notice that there has been an increase of the values of the performance in this second version that the authors have not explained. They should explain to the referees where the increase comes from.

4. There are two relevant review articles not referenced in this manuscript that should be cited: "J. L. MacManus-Driscoll et al, Nature Reviews Materials 6, 587 (2021), Processing and application of high-temperature superconducting coated conductors" and "T. Puig et al, Nature Reviews Physics 6, 132 (2024), Impact of high growth rates on the microstructure and vortex pinning of high-temperature superconducting coated conductors", both describing and discussing the relevant achievements in coated conductors fabrication and performances.

5. The c-axis parameter of the nanocomposite film (11.778 A) is larger than that of the REBCO film (11.722 A), a comment and a possible explanation should be given.

6. In the new TEM section, the cross section analysis shows that Y is included in the BZO nanorods but not Gd, however in the planar view it is shown that both, Y and Gd are incorporated in the nanorods. What is the reason of this disagreement? How is the c-axis parameter of BZO nanorords modified?
In fact in T. Schneller et al Solid State Ionics. 2003, 164, 131-136, it was demonstrated that the BZO structure could admit a 20% of Y, to generate $BaZr_{0.8}Y_{0.2}O_{3-x}$.

7. I would appreciate some more description of the TEM like that there are many SFs surrounding the nanorods and that they hardly have continuation from one site to the other of the nanorod, so that they plausibly emerge from them to decrease the interface strain.

8. The authors comment about the possible existence of a transient liquid to explain the faceted nanorods. However, this is very speculative. On one hand, although there were some early opinions on the direction that TFA-CSD films could grow from a liquid, it was never confirmed and the low growth rates of TFA didn't support it. For a recent review on TFA, notice X. Obradors et al SUST 37, 053001 (2024).
Only recently, with TLAG CSD novel approach, it has been demonstrated that a transient liquid does exist, in this case also in agreement with very high growth rates in the range of 100-1000 nm/s (L. Soler, et al Nat Comm 11, 344 (2020), S. Rasi et al, Adv Science 2203834 (2022).
Facets of BZO were observed in TFA as a consequence of energy minimization, also in decorated islands, as explained in A. Abellan, et al Acta Materialia 59 (2011) 2075–2082 and A. Llorder et al, 11 329 (2912). Also island and nanowire facets have been largely investigated in literature, for CSD case there is this review X. Obradors et al, Chem Soc. Rev. 43, 2200 (2014).
I suspect that the faceted nanorords observed in this manuscript can probably come from surface and interface energy minimization.

9. Figure 8 should clarify the ranges of positive and negative strain or show some particular examples. I think this very nice results should be reinforced.

10. I congratulate the authors to reach the values of critical current achieved still not being in the overdoped state. I'm looking forward to see which values could be reached if these films could be overdoped.

11. I appreciate all the included information on the growth system in the methods section. What do you mean by a laser-MBE? you use the laser for PLD but the system has also MBE capabilities? Or do you use those for your growth?

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The authors have addressed my concerns in the revision of their manuscript, which I now feel suitable for publication.

We thank the reviewer for accepting the manuscript for publication.

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This manuscript has addressed most of my concerns, and I recommend the publication as is.

We thank the reviewer for accepting the manuscript for publication.

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This reviewer acknowledges the modifications to the manuscript in this second version. The authors have achieved most of the points raised. In particular, I would like to rate very positively the detailed and outstanding TEM work now included, the additional measurements of charge carrier density which clarifies the doping state and the description of the growth system explained in the methods. The manuscript has now relevant insights on the process and characterization of the layers which supports the high values of the critical currents achieved. There are just few minor changes that should be attained before publication.

We thank the reviewer for acknowledging the extensive modifications made to the paper and for the complimentary remarks.

1. The title of the manuscript suggests that the work presented is done on wires which is not the case. Therefore, the title should be modified to “Ultra-high Critical Current Density and Pinning Force in REBCO-based, Nanostructured Superconducting Films”. The future implications of this work to wire manufacturing is already described in the abstract.

We agree that the word "wires" may give the impression of a long sample. We have therefore modified the title to - " *Ultra-high Critical Current Density and Pinning Force in Nanostructured, Superconducting, REBCO-based, Coated Conductor*". Since all work is reported on a coated conductor sample, this is an appropriate title.

2. In the abstract where it says “The key to making the price/performance metric favourable is to significantly enhance the performance”, should say “One of the keys to making the price/performance metric favourable is to significantly enhance the performance”, since there are other ways to also enhance this metric which are being followed by other researchers like increasing thickness, growth rate, double side tapes, yield, manufacturing throughput, substrate thickness, ...

We agree with the reviewer and have changed the sentence in the manuscript to "A key route to...", implying that there could be other ways as well.

3. I notice that there has been an increase of the values of the performance in this second version that the authors have not explained. They should explain to the referees where the increase comes from.

As mentioned in the point-by-point response to the first set of review comments, as suggested by reviewer #3, we have now done a profilometer scan to determine the film thickness.

We fully agreed with the reviewer that it is best to measure film thickness by profilometry. In TEM, if the film is very rough one can obtain a lower thickness in some local areas. If the film is smooth and has a uniform thickness (as is the case here), one can obtain a slightly higher thickness, since TEM thin foils are made at a very shallow angle through the film thickness to obtain an electron transparent region for microscopy and hence and a higher film thickness can be observed. The profilometer scan included in the supplementary materials nicely shows that the film thickness is 200 nm. We have now used the thickness determined from the profilometer scan for the film thickness as it more representative of the film thickness. Previously, from TEM, the thickness locally was 217nm.

4. There are two relevant review articles not referenced in this manuscript that should be cited: "J. L. MacManus-Driscoll et al, Nature Reviews Materials 6, 587 (2021), Processing and application of high-temperature superconducting coated conductors" and "T. Puig et al, Nature Reviews Physics 6, 132 (2024), Impact of high growth rates on the microstructure and vortex pinning of high-temperature superconducting coated conductors", both describing and discussing the relevant achievements in coated conductors fabrication and performances.

We have added these suggested references.

5. The c-axis parameter of the nanocomposite film (11.778 A) is larger than that of the REBCO film (11.722 A), a comment and a possible explanation should be given.

It is well-known that incorporation of self-assembled BZO nanocolumns results in a tensile strain in the REBCO film resulting in an increased c-axis lattice parameter. With increased BZO incorporation, the tensile strain increases and so does the c-axis lattice parameter.

The reason for increased c-axis lattice parameter for the nanocomposite film is now included in the paper.

6. In the new TEM section, the cross section analysis shows that Y is included in the BZO nanorods but not Gd, however in the planar view it is shown that both, Y and Gd are incorporated in the nanorods. What is the reason of this disagreement? How is the c-axis parameter of BZO nanorords modified?
In fact in T. Schneller et al Solid State Ionics. 2003, 164, 131-136, it was demonstrated that the BZO structure could admit a 20% of Y, to generate $BaZr_{0.8}Y_{0.2}O_{3-x}$.

In the cross-section TEM, the TEM foil thickness is much greater than the diameter of the BZO nanorod, so one is picking up the REBCO matrix as well. The Gd concentration in the BZO is less than the Gd concentration and so in cross-section EDS maps, Gd shows up less. In the plan-view analysis, the BZO nanorod is going through the thickness and so is a more accurate examination of of the composition within BZO nanorods. Clearly, both Y and Gd are present in BZO nanorods. We are not specifying exact concentrations of Y and Gd since there can be significant error in the computation of at% if the foil thickness is not very accurately and exactly known.

We have included a comment in the paper regarding this.

We have not focused on an accurate determination to gauge the change in c-axis parameter of BZO with Y and Gd additions. Thank you for pointing out the work of T. Schneller et al Solid State Ionics. 2003, 164, 131-136, indicating 20% Y incorporation. We suspect that significant amounts of other RE atoms such as Gd can also be incorporated into BZO at the right temperatures.

7. I would appreciate some more description of the TEM like that there are many SFs surrounding the nanorods and that they hardly have continuation from one site to the other of the nanorod, so that they plausibly emerge from them to decrease the interface strain.

We have added a comment as suggested by the reviewer.

8. The authors comment about the possible existence of a transient liquid to explain the faceted nanorods. However, this is very speculative. On one hand, although there were some early opinions on the direction that TFA-CSD films could grow from a liquid, it was never confirmed and the low growth rates of TFA didn't support it. For a recent review on TFA, notice X. Obradors et al SUST 37, 053001 (2024).
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I suspect that the faceted nanorords observed in this manuscript can probably come from surface and interface energy minimization.

There has been extensive work done on incorporating self-assembled BZO nanorods using PLD deposition in REBCO films. Most plan-view TEM images show a non-faceted BZO nanorod cross-section, including in our original work. Surface and interface energy minimization would have also occurred in these samples, but perhaps the temperature was not high-enough for either greater solid-state diffusion to form a facet and/or a transient liquid to accelerate the diffusion of atoms was not present during growth.

So, we agree with the reviewer that a transient liquid phase is not the only mechanism for surface and interface energy minimization to result in a facet. A comment to this effect is now included in the paper.

9. Figure 8 should clarify the ranges of positive and negative strain or show some particular examples. I think this very nice results should be reinforced.

We have changed the labels for the strain in the paper to clarify the range of strains as suggested by the reviewer.

10. I congratulate the authors to reach the values of critical current achieved still not being in the overdoped state. I'm looking forward to see which values could be reached if these films could be overdoped.

We thank the reviewer for his/her comments.

11. I appreciate all the included information on the growth system in the methods section. What do you mean by a laser-MBE? you use the laser for PLD but the system has also MBE capabilities? Or do you use those for your growth?

Laser-MBE is essentially Pulsed laser deposition (PLD), with the distinction that the PLD system also has the capabilities of being UHV system and having in-situ RHEED capabilities. Else, for all practical purposes, it is a PLD system.

REVIEWERS' COMMENTS

Reviewer #3 (Remarks to the Author):

The authors have addressed my comments also in this second revision of their manuscript. I now recommend the manuscript for publication.