YBCO-buffered NdBCO film with higher thermal stability in seeding REBCO growth

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Abstract

In this work, for the first time, we report a strengthened superheating effect caused by a buffering \(\text{YBa}_2\text{Cu}_3\text{O}_y\) (Y123 or YBCO) layer in a \(\text{Nd}_{1+x}\text{Ba}_{2-x}\text{Cu}_3\text{O}_{7-y}\) (Nd123 or NdBCO) thin film with a MgO substrate (i.e., NdBCO/YBCO/MgO thin film). In cold-seeding melt-textured (MT) growth the NdBCO/YBCO/MgO film presented an even higher superheating level, about 20\(^\circ\)C higher than that of non-buffered NdBCO film (i.e., NdBCO/MgO film). Using this NdBCO/YBCO/MgO film as the seed and under a maximum processing temperature (\(T_{\text{max}}\)) of up to 1120\(^\circ\)C, we succeeded in growing various RE\(_{1+x}\)Ba\(_{2-x}\)Cu\(_3\)O\(_{7-y}\) (REBCO, RE = Gd, Sm, Nd, RE refers to rare elements) bulk superconductors. Pole figure (x-ray \(\Phi\)-scan) measurement reveals that the NdBCO/YBCO/MgO film has better in-plane alignment than NdBCO/MgO film, indicating that the induced intermediate layer improves the crystallinity of the NdBCO film, which could be the main origin of the enhanced thermal stability. In short, possessing higher thermal stability and enduring a higher \(T_{\text{max}}\) in the MT process, the NdBCO/YBCO/MgO film is beneficial to the growth of bulk superconductors in two aspects: (1) broad application for REBCO materials of high peritectic temperature (\(T_p\)); (2) effective suppression against heterogeneous nucleation, which is of great assistance in growing large and high-performance REBCO crystals.

(Some figures may appear in colour only in the online journal)

1. Introduction

Nowadays, top-seeded melt-texture growth (TSMTG) is commonly used to obtain large single domain REBCO bulks. In this method, the seed crystal or film is placed on the top of the precursor to induce the epitaxial nucleation. The seed is required to (1) have a similar crystal structure to REBCO and (2) remain stable during the whole TSMTG process. SmBCO and NdBCO crystals have been commonly used as seeds. However, since their peritectic temperatures are not high enough, the members they can seed in the REBCO family are limited [1]. Mg doping in NdBCO fascinatingly breaks this limitation, raising its \(T_p\) by 20\(^\circ\)C over the \(T_p\) of pure NdBCO [2].

In our previous work, a superheating phenomenon was demonstrated in REBCO thin films which were deposited on an MgO substrate. Firstly, a YBCO thin film, possessing a \(T_p\) of 1010\(^\circ\)C, was found to be able to endure a high processing temperature (1057\(^\circ\)C) to induce an epitaxial growth of a NdBCO \((T_p = 1085\,^\circ\text{C})\) thick film in liquid phase epitaxial growth [3]. In the second example, where the superheating nature appears more evidently, a YBCO bulk was induced by...
using its homoseed, a YBCO/MgO film, after holding at a temperature of 30 °C above its \( T_p \) for 1.5 h \[4\]. Furthermore, the superheating phenomenon was clearly identified to exist in all commonly used YBCO-film/substrate structure (where the substrate is MgO, LaAlO\(_3\) or SrTiO\(_3\)) \[5\]. All these facts suggest a universal superheating mode, which absolutely relates to a distinctive construction feature, i.e., a free surface with low surface energy and a semi-coherent interface between film and substrate. So far, most kinds of REBCO bulks have been successfully seeded by NdBCO/MgO thin films, which will potentially become popular because NdBCO has the highest \( T_p \) among the commonly applied REBCO materials \[6\]. However, as it is in the structure of a thin film, NdBCO has a superheating upper limit at about 1098 °C in MT growth, only 14 °C higher than its \( T_p \), which is lower than that of a Y123 thin film \[7\]. Compared with the YBCO system, due to a lower liquidus slope, there is a higher Nd supersaturation for the RE123 growth in the same undercooling. Similarly, there is a higher Nd supersaturation for \( \text{RE}_2\text{BaCuO}_5 \) (RE211) growth in the same superheating, which is equivalent to a higher growth rate of the \( \text{Nd}_4\text{Ba}_2\text{Cu}_2\text{O}_7 \) (Nd422, with a cation compositional ratio of RE211) and higher dissolution rate of the Nd123 \[7\]. In addition, the manufacturer Theva GmbH has reported that the growth window is rather small to obtain high-quality NdBCO/MgO films \[8\]. Both of these demerits preclude NdBCO/MgO film from being used to seed the growth of high-\( T_p \) REBCO bulks, e.g., RE = Sm or Nd. Recently, on growing a YBCO buffer between the NdBCO film and the MgO substrate, namely, a NdBCO/YBCO/MgO film, the manufacturer recognized its effect on broadening the growth window of high-quality NdBCO films \[8\].

With this special NdBCO-film/YBCO-buffer-layer/MgO-substrate structure, better crystallinity and higher thermal stability of the NdBCO film can be expected. In this work, using NdBCO/YBCO/MgO thin films as seeds in the MT process, a temperature as high as 1120 °C was tolerated, which was remarkably 35 °C higher than the \( T_p \) of NdBCO. Seeded by this NdBCO/YBCO/MgO film, in particular, a fully grown NdBCO bulk without Ag addition was successfully obtained.

2. Experiments

The NdBCO/YBCO/MgO thin film, with a 600 nm NdBCO film and a 25 nm YBCO buffer layer, was provided by the company Theva GmbH. The in-plane alignments for both the NdBCO/YBCO/MgO and NdBCO/MgO thin films (also made by Theva GmbH) were tested by pole figure analysis using Cu Ka radiation, indicating the orientation relationship between the film and the MgO substrate. The Nd123 and Nd422 precursor powders were prepared by grinding and calcining stoichiometric mixtures of \( \text{Nd}_2\text{O}_3 \), BaCO\(_3\) and CuO raw powders repeatedly. All the synthesized powders were characterized by the x-ray diffraction (XRD) method. A sample with the nominal compositions of Nd123 + 20 mol%Nd422 + 1 wt%CeO\(_2\) was prepared and pressed uniaxially into a pellet of 20 mm in diameter. The growth of the NdBCO sample was performed in air using a typical TSMTG method. The NdBCO/YBCO/MgO thin film was placed on top of the pellet as a seed, to control the growth of a single grain in the (001) orientation. The heating profile was as follows: the sample was heated from room temperature to 950 °C in 5 h, held at 950 °C for 4 h, and then heated to 1117 °C in 2 h. After being held at 1117 °C for 1.5 h, the sample was cooled rapidly to 1083 °C in 20 min, and then slowly cooled down at a rate of 0.25 °C h\(^{-1}\) before quenching.

3. Results and discussion

By cold-seeding a NdBCO/YBCO/MgO thin film to induce a homo-epitaxy for the first time, we report a success on the full growth of a single-grain NdBCO bulk without Ag addition. Remarkably, this NdBCO/YBCO/MgO film seed has endured the \( T_{\text{max}} \) of 1117 °C for 1.5 h, which is about 20 °C higher than the \( T_p \) of NdBCO. Seeded by this NdBCO/YBCO/MgO film, in particular, a fully grown NdBCO bulk without Ag addition was successfully obtained.

Figure 1 shows the top view of the as-grown NdBCO bulk seeded by NdBCO/YBCO/MgO thin film, using a \( T_{\text{max}} \) of 1117 °C.

Figure 2 shows the pole figure analysis for the two kinds of NdBCO films with and without a YBCO buffer layer. A noticeable improvement of crystallinity in the YBCO-buffered NdBCO film is exhibited.

The pole figure measurement result from the NdBCO thin film (figure 2(a)) presents a poor in-plane alignment. There are small unexpected peaks (satellite peaks) at ±17.5° around the strong peaks, indicating that the (100) direction of the NdBCO thin film orients to the (100) direction of the MgO substrate in a 0° and (0° ± 17.5°) relationship. In comparison, the result from the NdBCO/YBCO/MgO thin film (figure 2(b)) reveals a good four-fold symmetry, indicating that there is only one in-plane orientation in the NdBCO/YBCO/MgO thin film.
Figure 2. X-ray pole figures of (a) NdBCO/MgO thin film and (b) NdBCO/YBCO/MgO thin film with a 25 nm YBCO buffer layer.

Table 1. Various kinds of REBCO bulk seeded by the NdBCO/YBCO/MgO thin film and the $T_{\text{max}}$ of each process we succeeded in.

| RE123 systems | $T_{\text{max}}$ ($^\circ\text{C}$) | Mixed RE123 systems | $T_{\text{max}}$ ($^\circ\text{C}$) |
|---------------|---------------------------------|---------------------|---------------------------------|
| SmBCO        | 1115                             | Ag–GdBCO            | 1115                              |
| GdBBCO       | 1120                             | Ag–NdBCO            | 1115                              |
| NdBCO        | 1117                             |                     |                                  |

i.e., the (100) direction of the NdBCO/YBCO thin film orients well in a $0^\circ$ relationship to the (100) direction of the MgO substrate. Convincingly, it is the YBCO intermediate layer that plays a role in improving the in-plane alignment of the NdBCO film. Similarly to the sandwich structure of REBCO/buffer/substrate in REBCO coated conductors, the YBCO buffer layer in NdBCO/YBCO/MgO thin film performs functions in (1) compensating the lattice misfit between NdBCO and MgO and (2) alleviating the problems related to the interface with the substrate [10].

As we know, the melting always initiates at defect sites (e.g., free surfaces and internal grain boundaries) because of their high free energy; defects of grain boundaries in a film play an important role in the thermal stability. Due to the excellent in-plane alignment, there are many fewer grain boundaries in the NdBCO/YBCO/MgO thin film than in NdBCO/MgO thin film, which predominantly takes the responsibility for the enhanced superheating capability in the NdBCO/YBCO/MgO thin film.

Considering the similar crystal structure in the whole REBCO family, it is reasonable to assume that almost all kinds of REBCO bulks could be seeded by NdBCO/YBCO/MgO thin films. Table 1 lists the various kinds of REBCO bulk effectively seeded by the NdBCO/YBCO/MgO thin film and the $T_{\text{max}}$ of each process we succeeded in. Figure 3, as one more distinctive result, shows the top view of a GdBBCO single grain induced by the YBCO-buffered NdBCO-film seed undergoing a $T_{\text{max}}$ of 1120 $^\circ\text{C}$, which is a $T_{\text{max}}$-tolerating record in the homo-seeding MT growth of REBCO bulk as far as we know.

Figure 4 shows the types of film seed that have been practically used and their reported $T_{\text{max}}$ values in the MT processes.

Figure 3. A GdBBCO single grain seeded by the NdBCO/YBCO/MgO thin film using a $T_{\text{max}}$ of 1120 $^\circ\text{C}$.

Figure 4. The types of film seed that have been practically used and their reported $T_{\text{max}}$ values in the MT processes.

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There are three kinds of surface-related defect that are relative to the thermal stability of a film: (1) the hetero-structure interface, like the MgO/NdBCO interface; (2) inner grain boundaries in the film; (3) the exposed surface with dangling bonds. As the YBCO buffer layer induces better in-plane alignment (as shown in figure 2), it reduces the inner grain boundaries in the NdBCO film. But there is some negative effect too. Because of the great difference in the intrinsic $T_p$ values between YBCO and NdBCO, the YBCO buffer decomposes earlier than the NdBCO. In the MT heating process, the YBCO buffer decomposes into the Y211 solid and the Ba–Cu–O (BCO) liquid, forming a BCO-liquid/NdBCO hetero-structure interface, which has a higher surface energy. However, the small Nd123 grains in the film might become coarsened during the long time of heating, which also reduces the grain boundaries and slows down the nucleation rate according to Griffith et al [15]. Besides, the over-saturation of the rare earth element in the BCO liquid could suppress the decomposition of the Nd123 grain, making the film seed stable in the MT process [4]. In other words, the reduced inner grain boundaries induced by YBCO buffer layer play a dominant role in the enhanced thermal stability of the NdBCO/YBCO/MgO film. The performance of the NdBCO/YBCO/MgO film at temperatures near the melting point and how it differs from the NdBCO/MgO film are still under investigation and will be discussed in another paper.

4. Conclusion

Using a NdBCO/YBCO/MgO thin film as a seed and taking a $T_{\text{max}}$ of 1117 °C in the MT process, a NdBCO bulk superconductor with a full growth was obtained. According to the x-ray pole figure results, NdBCO/YBCO/MgO thin film has better crystallinity than NdBCO/MgO thin film, which leads to the fact that the NdBCO/YBCO/MgO thin film possesses a stronger superheating effect than NdBCO/MgO thin film. Therefore a higher $T_{\text{max}}$ could be used during the MT growth seeded by the NdBCO/YBCO/MgO thin film, which could suppress the heterogeneous nucleation, broaden the growth window, and benefit the growth of large and high-performance REBCO crystals.

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