Angular dependence of $J_c$ in YBCO films with c-axis correlated nano-rods and in-plane-distributed nano-particles

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Abstract. The quasi-multilayered films consisting of YBa$_2$Cu$_3$O$_y$ layers with BaSnO$_3$ nano-rods and the pseudo layers of in-plane-distributed BaSnO$_3$ nano-particles were fabricated using a PLD method, in order to clarify the pinning landscape simultaneously improving the critical current densities, $J_s$, both at $B \parallel c$ and at $B \parallel ab$. The insertion of the pseudo layers into YBa$_2$Cu$_3$O$_y$ films contributes to the enhancement of $J_c$ at $B \parallel ab$ but it is a tendency to reduce the $J_c$ at $B \parallel c$. When the density of the nano-particles per layer is decreased, by contrast, the $J_c$ at $B \parallel ab$ can be enlarged without the reduction of the $c$-axis peak. This is attributed to the fragmentation of the channel for flux creep motion through the pseudo layers. Furthermore, the $J_c$ in tilted magnetic fields off the $c$-axis enhances as well as $c$-axis peak in a high magnetic field, when BaSnO$_3$ nanoparticles are increased.

1. Introduction

To improve a critical current density $J_c$ in magnetic field for REBa$_2$Cu$_3$O$_y$ (REBCO, RE: rare earth elements) based coated conductors, an introduction of additional crystalline defects and impurities into REBCO films is a promising way, where nano-sized defects work as pinning centers (PCs) and effectively arrest flux lines penetrating in superconducting materials [1]. Nano-rods, which are formed through self-assembled stacks of BaMO$_3$ (M = Zr, Sn, Hf, etc.) along the c-axis into REBCO films, belongs to one-dimensional (1D) PCs and effectively interrupt the motion of flux lines when the magnetic field is applied parallel to the c-axis [2, 3]. Planar defects consisting of continuous impurity layers along the ab-plane are two-dimensional (2D) PCs and act as ab-correlated PCs, leading to a weak field dependence of $J_c$ at $B \parallel ab$ [4]. Nano-particles classified as three-dimensional (3D) PCs, on the other hand, have the morphology with no correlated orientation for flux pinning, leading to the isotropic pinning force against any orientations of magnetic field [5].

In recent years, further improvement of flux pinning including a reduction of the anisotropy of $J_c$ has been attempted by using a combination of different types of PCs, which is referred to as hybrid flux pinning [6, 7]. The hybrid flux pinning by using nano-rods and nano-particles enhances not only the $J_c$ peak centered at $B \parallel c$ but also the $J_c$ in a wide angular range of magnetic field [7], where nano-particles play an important role in the prevention of the sliding motion of flux lines for magnetic fields tilted off the direction of nano-rods and in the suppression of the motion of double kinks of flux lines due to thermal fluctuation [8]. The combination of 1D and 3D PCs, however, does not sufficiently reduce the anisotropy of $J_c$, where the c-axis $J_c$-peak is larger than the $J_c$ peak at $B \parallel ab$ in most cases. The combination of 1D and 2D PCs consisting of c-axis-aligned nano-rods and ab-plane-aligned planar...
nanostructures, on the other hand, can contribute to the improvement of $J_c$ both at $B \parallel c$ and at $B \parallel ab$ [6, 9]. The insertion of planar defects along the $ab$-plane, however, tends to reduce the $c$-axis peak in the $J_c$ anisotropy caused by nano-rods, since flux lines along the $c$-axis easily move along the $ab$-aligned nanostructure layers for $B \parallel c$ [4] or the structure of flux lines changes from conventional Abrikosov vortices to pancake vortices [10].

In this work, we investigated the hybrid flux pinning structure which improves the $J_c$ in all magnetic field orientations, i.e. at $B \parallel c$, at $B \parallel ab$, and in the intermediate-angle region, by preparing the quasi-multilayered films consisting of YBCO layers with BaSnO$_3$ nano-rods and the pseudo layers of in-plane-distributed BaSnO$_3$ nano-particles using a pulsed laser deposition (PLD) method. Figure 1 schematically illustrates distribution of the nano-rods and nano-particles in the multilayered films. The nano-rods consisting of BaSnO$_3$ contribute to the enhancement of $J_c$ at $B \parallel c$, while the in-plane-distributed BaSnO$_3$ nano-particles act as PCs with features of both 2D and 3D PCs: the nano-particles aligned along the $ab$-plane can be expected to enhance the $J_c$ at $B \parallel ab$ and in the intermediate-angle region, but without reducing the $c$-axis peak.

2. Experimental details

The quasi-multilayered films consisting of YBCO layers with BaSnO$_3$ nano-rods and the pseudo layers of in-plane-distributed BaSnO$_3$ nano-particles were fabricated on (100) SrTiO$_3$ substrates by a PLD technique alternating a 2.0 vol.% BaSnO$_3$ doped YBCO target and a BaSnO$_3$ target. The repetition rate of the laser was 2 Hz for the deposition of YBCO layers with BaSnO$_3$, while the pseudo layers of BaSnO$_3$ were deposited at 1 Hz. The growth of BaSnO$_3$ nano-rods in the YBCO layer would be continuous along the film-thickness direction even in the multilayering process [11]. The deposition of BaSnO$_3$ by several laser pulses, on the other hand, provides not a BaSnO$_3$ layer but in-plane-aligned BaSnO$_3$ nano-particles [12]. During the deposition, the substrate was heated at 760 °C and the oxygen partial pressure was 300 mTorr. After the deposition, the films were cooled down naturally from 760 °C to 500 °C and from 200 °C to room temperature in 600 Torr oxygen, whereas the cooling rate was controlled at 10 °C/min from 500 °C to 200 °C. A series of the quasi-multilayered films in this work is listed in Table 1. We refer to a sample as B($m$, $n$), where $m$ and $n$ denote the number of laser pulses on the BaSnO$_3$ target and the total number of BaSnO$_3$ / YBCO + BaSnO$_3$ bilayers, respectively. A layer of YBCO + BaSnO$_3$ was always deposited on the top of the multilayered films and the total number of laser pulses on the BaSnO$_3$ doped YBCO target was kept constant at 3600 pulses, resulting in the film thickness of about 300 nm.

| Sample | Number of laser pulses on BaSnO$_3$ target, $m$ | Number of bilayers, $n$ | $T_c$, [K] |
|--------|---------------------------------------------|------------------------|----------|
| B(3, 5) | 3                                           | 5                      | 89.2     |
| B(6, 5) | 6                                           | 5                      | 89.8     |
| B(3, 10)| 3                                           | 10                     | 89.7     |
| B(6, 10)| 6                                           | 10                     | 90.0     |
| BSO2   | -                                           | -                      | 89.4     |

Figure 1. Schematic illustration of quasi-multilayered films consisting of YBCO layers with BaSnO$_3$ nano-rods and the pseudo layers of in-plane-distributed BaSnO$_3$ nano-particles.

Table 1 Samples in this work.
Simultaneously, a 2.0 Vol.% BaSnO$_3$ doped YBCO film was prepared as a reference sample, which are referred to as BSO2.

Transport properties were measured using a four-probe method, where the films were patterned into bridge geometry with about 40 μm width and 1 mm length using a standard photolithography. The value of $J_c$ was defined by electric field criterion of 1 μV/cm. The angular dependence of $J_c$ was evaluated as a function of the angle $\theta$ between the magnetic field and the $c$-axis, where the magnetic field was always applied perpendicular to the direction of the current (the maximum Lorentz force configuration).

3. Results and discussion

Firstly, we compare the angular behaviour of $J_c$ between the multi-layered films with different $m$, in order to clarify the thinning-out effect of the in-plane-distributed nano-particles on the $c$-axis peak formed by the nano-rods. Figure 2(a) shows the $J_c$ as a function of magnetic field orientation at 77 K and 1 T for the 5-bilayer films with different $m$, compared with BSO2. Two peaks can be observed in the angular dependences of $J_c$ for all the samples. One of the peaks emerges at $\theta = 0^\circ$, which arises from the $c$-axis correlated pinning due to the nano-rods. Another is the steep peak at $\theta = 90^\circ$, which is generally attributed to the stacking faults or to the intrinsic pinning due to the CuO$_2$ planes. The $J_c$ peak at $\theta = 90^\circ$ is also affected by the structure of multilayered films [5] and is more evolved with increasing $m$, as shown in Figure 2(a). This is because the BaSnO$_3$ nano-particles aligned along the in-plane direction of the film effectively work as correlated PCs along the $ab$-plane. It is noteworthy that the multilayered films with larger $m$ more enlarge the $J_c$ in the intermediate angle region between $B \parallel c$ and $B \parallel ab$. This suggests that the in-plane-distributed BaSnO$_3$ do not grow into layers but into particles: each nano-particles is isolated enough to work as 3D-PCs.

The insertion of the impurity layers aligned along the $ab$-plane, on the other hand, tends to weaken the $c$-axis peak [10, 13]. For B(6, 5), the $J_c$ peak at $\theta = 0^\circ$ is lower than that for BSO2. This is because the in-plane-distributed BaSnO$_3$ nano-particles for B(6, 5) easily induce the flux creep motion of the interstitial flux lines between nano-particles [4], resulting in the negative effect for the flux pinning at $B \parallel c$ (see figure 3). Note that the dimensionality of the YBCO system, which significantly affects the flux pinning by linear defects [10], would not be modified by the insertion of the impurity layers in this work, since the thickness of the bilayer is about 30-60 nm and the impurity deposition did not form complete non-superconducting-layers.

When $m$ is decreased, by contrast, no deterioration in the $c$-axis peak is identified for B(3,5), resulting in the upward shift in the $J_c$ over the entire magnetic field directions compared to BSO2. The influence of $m$ on the $J_c$ becomes more evident when the number of bilayers is increased to $n = 10$, as shown in Figure 4. These results suggest that the thinning of in-plane-distributed nano-particles effectively suppresses the flux creep motion between the nano-particles, resulting in the improvement of the $J_c$ at

![Figure 2](image1.png)

**Figure 2.** Angular dependence of $J_c$ for multilayered films with 5 bilayers and different laser pulses on BaSnO$_3$ target at 77 K for (a) 1 T and (b) 3 T.
Figure 3. Schematic illustrations of possible flux pinning states in the presence of nano-rods and in-plane-distributed nano-particles. As the number of $m$ is larger, the density of the nano-particles per layer increases, which easily induces the flux creep motion between the nano-particles as shown by blue arrows.

Figure 4. Angular dependence of $J_c$ for multilayered films with 10 bilayers and different laser pulses on BaSnO$_3$ target at 77 K for (a) 1 T and (b) 3 T.

$B \parallel ab$ without a reduction of the $c$-axis peak. In increasing the magnetic field, the influence of the nano-particle pinning becomes apparent even on the $J_c$ at $B \parallel c$: the $J_c$ around $\theta = 0^\circ$ for the multilayered film with $m = 3$ exceeds that for BSO2. When flux lines outnumber the nano-rods in high magnetic field, interstitial flux lines existing between the nano-rods are effectively trapped by the in-plane-distributed BaSnO$_3$ nano-rods [13].

To explore further optimization for the flux pinning effect of the multilayered films, we investigated the influence of the bilayer number $n$ on the angular behaviour of $J_c$ for the multilayered films with $m = 3$, as shown in figure 5. The multilayered films with $m = 3$ show upward shift in $J_c$ compared to BSO2 in overall orientations of magnetic field, although the $c$-axis peak for the multilayered films is equal to or slightly higher than that of BSO2 for 1 T. When the number of bilayer $n$ increases, the upward shift in $J_c$ becomes more remarkable at $B \parallel ab$ and in the intermediate angle region. The increase in $J_c$ at $B \parallel ab$ is due to a large number of the bilayers which work as pseudo planar PCs. In the intermediate angle region, on the other hand, the nano-particles play a role in 3D PCs [7, 13], which can pin the segments of flux lines released from the nano-rods along the $c$-axis. Therefore, a large number of bilayers is effective for the improvement of $J_c$ both in high magnetic field and in the intermediate angle region.

In Figure 6, we compare the angular dependence of $J_c$ between the multilayered films with $m \times n = 30$, which means the same amount of nano-particles regardless of the different multilayer structures. The $J_c$ in a wide range of magnetic field orientations can be improved by thinning out the nano-particles on each layer and by increasing the number of bilayers, even if the amount of nano-particles is the same. In other words, tuning of spatial distribution of nano-particles would be one of the keys to further enhance the overall $J_c$ for the hybrid flux pinning. Note that the $J_c$ peaks at $B \parallel ab$ for B(3, 10) and
Figure 5. Angular dependence of $J_c$ for multilayered films with $m = 3$ and different numbers of bilayers $n$ at 77 K for (a) 1 T and (b) 3 T.

Figure 6. Angular dependence of $J_c$ for multilayered films with $m \times n = 30$ at 77 K for (a) 1 T and (b) 3 T. B(6, 5) nearly overlap each other for both 1 T and 3 T. The in-plane-distributed nano-particles contribute to effective flux pinning at $B \parallel ab$. The same peak behaviour of $J_c$, however, is not always provided for $B \parallel ab$ by the different multi-layered structures even if the amount of nano-particles is the same [13].

4. Conclusions
In order to clarify the pinning structure simultaneously attaining high $J_c$ both at $B \parallel c$ and at $B \parallel ab$, we investigated the influence of the structure of the multilayered YBCO films with BaSnO$_3$ nano-rods and in-plane-distributed BaSnO$_3$ nano-particles on the angular behaviour of $J_c$, where the number of nano-particles per layer and the number of bilayers were controlled. The in-plane-distributed nano-particles can contribute not only to improving the $J_c$ at $B \parallel ab$ but also to keeping the c-axis peak due to the nano-rods by thinning out the nano-particles per layer. In addition, the $J_c$ can be further evolved in high magnetic field and in a wide range of magnetic field orientation by increasing the number of bilayers consisting of YBCO layer and the nano-particles. The formation of the in-plane-distributed nano-particles for the hybrid flux pinning structure is one of the important factors for the improvement of $J_c$ in all magnetic field orientations.

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References

[1] Matsumoto K and Mele P, 2010 Supercond. Sci. Technol. 23 014001
[2] Mele P, Matsumoto K, Horide T, Ichinose A, Mukaida M, Yoshida Y, Horii S and Kita R, 2008 Supercond. Sci. Technol. 21 032002
[3] Tobita H, Notoh K, Higashikawa K, Inoue M, Kiss T, Kato T, Hirayama T, Yoshizumi M, Izumi T and Shiohara Y, 2012 Supercond. Sci. Technol. 25 062002
[4] Yang H, Wang H, Maiorov B, Lee J, Talbayev D, Hinton M J, Feldmann D M, MacManus-Driscoll J L, Taylor A J, Civale L, Lemberger T R and Jia Q X, 2009 J. Appl. Phys. 106 093914
[5] Gapud A A, Kumar D, Viswanathan S K, Cantoni C, Varela M, Abiade J, Pennycook S J and Christen D K, 2005 Supercond. Sci. Technol. 18 1502
[6] Wu J Z, Shi J J, Bacal F J, Emergo R, Wilt J and Haugan T J, 2015 Supercond. Sci. Technol. 28 125009
[7] Horide T, Kawamura T, Matsumoto K, Ichinose A, Yoshizumi M, Izumi T and Shiohara Y, 2013 Supercond. Sci. Technol. 26 075019
[8] Maiorov B, Baily S A, Zhou H, Ugurlu O, Kennison J A, Dowden P C, Holesinger T G, Foltyn S R and Civale L, 2009 Nat. Mater. 8 398
[9] Ishikawa K, Ichino Y, Yoshida Y, Yoshizumi M, Izumi T and Kato T, 2015 J. Cryo. Super. Soc. Jpn. 50 218 (in Japanese)
[10] Holzapfel B, Kreiselmeyer G, Kraus M, Bouffard S, Klaumünzer S, Schultz L and Saemann-Ischenko G, 1993 Phys. Rev. B 48 600
[11] Tsuruta A, Yoshida Y, Ichino Y, Ichinose A, Matsumoto K and Awaji S, 2014 Supercond. Sci. Technol. 27 065001
[12] Huhtinen H, Schlesier K and Paturi P, 2009 Supercond. Sci. Technol. 22 075019
[13] Sueyoshi T, Kotaki T, Uraguchi Y, Suenaga M, Makihara T, Fujiyoshi T and Ishikawa N, 2016 Physica C 530 72