Improvement of anisotropy of superconducting properties in Y-rich YBa$_2$Cu$_3$O$_y$ film in magnetic fields

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Abstract. In order to put the REBa$_2$Cu$_3$O$_y$ superconductor into practical use, it is necessary to more isotropically improve the critical current density ($J_c$) with respect to an applied magnetic field. Therefore, artificial pinning centers (APCs) which are like a particle shape should be introduced into the superconductor. In this study, nano-particle APCs were introduced by fabricating Y$_{1+x}$Ba$_2$Cu$_3$O$_y$ ($x = 0 \sim 1.0$) thin films with excess Y composition, and effect of $x$, substrate temperature ($T_s$) and pulsed laser repetition frequency ($f_L$) on the isotropy of $J_c$ in the magnetic field was evaluated. As a result, these conditions proved to have a strong influence on isotropic improvement of $J_c$ in the magnetic field.

1. Introduction
Since a REBa$_2$Cu$_3$O$_y$ (REBCO, RE=Rare Earth) superconductor has high critical temperature ($T_c$) which is over 90 K, cooling cost can be suppressed and it is expected to put into a practical use such as superconducting magnet coil. However, when applying REBCO wire as a coil, the magnetic field in the coil is not applied to the wire in a certain direction but is applied from various directions [1]. From this requirement, the REBCO superconductor having a large anisotropy for magnetic field angular dependence of $J_c$ ($J_c(\theta)$) is required isotropic improvement of $J_c$ ($\theta$). As a solution, isotropic improvement of $J_c$ ($\theta$) has been reported by introducing artificial pinning centers (APCs) in nano-particle shape such as RE$_2$O$_3$ or RE$_2$BaCuO$_y$ (RE211) in a superconductor [2, 3]. As a method of introducing them, it is reported tuning a composition of RE in a target when preparing REBCO wires by pulsed laser deposition (PLD) method [4]. Additionally, substrate temperature ($T_s$) and pulsed laser repetition frequency ($f_L$) are one of the important deposition parameters when we fabricate the films by PLD method.

In this study, Our goal is to achieve a REBCO film that has isotropic enhancement of $J_c(\theta)$ at sub-cooling temperature of liquid nitrogen (65 K). We fabricated Y$_{1+x}$Ba$_2$Cu$_3$O$_y$ (Y$_{1+x}$BCO; $x = 0 \sim 1.0$) films by PLD method. We aim to control the size and number density of nanoparticles not only $x$ but also deposition conditions, and also examine what kind of modulations the microstructure of the film might have on these conditions by $J_c(\theta)$ in magnetic field and so on.

2. Experimental details
We fabricated Y$_{1+x}$BCO thin films on IBAD-MgO metal substrate by PLD method using a KrF excimer laser ($\lambda = 248$ nm). The oxygen partial pressure, the energy density irradiated on the target, the film
thicknes were 800 mTorr, 1.4 J/cm², 300 nm, respectively. The critical current density in self-field (at 65 K in 3 T of YBCO films) depended on excess Y of x. Table 1 shows the critical temperature (Tc) and critical current density (Jc) of Y1+xBCO thin films with different excess Y of x.

| x  | Tc [K] | Jc [MA/cm²] |
|----|--------|-------------|
| 0  | 90.6   | 2.47        |
| 0.2| 90.4   | 3.27        |
| 0.4| 88.9   | 2.87        |
| 0.8| 89.1   | 1.00        |
| 1.0| 89.1   | 0.83        |

Figure 1. Magnetic Field angular dependence of Jc on Y1+xBCO (x = 0 - 1.0).

3. Results and discussion

3.1. Dependence of excess Y on superconducting properties in magnetic field and microstructure
We fabricated Y1+xBCO (x = 0 ~ 1.0) films at Tc = 840°C and fL = 20 Hz. Table 1 shows Tc and critical current density in self-field (Jc,x). From this result, Tc decreased by about 1.5 K when x is 0.4 or more, Jc,x became the maximum value at x = 0.2, and declined with increasing x larger than 0.2. Figure 1 indicates x dependence of Jc(θ) at 65 K and 3 T. It is the same tendency with Jc,x and became the maximum at x = 0.2 at Jc(θ = 0°). Thus, x = 0.2 film contains the nanoparticles which has optimum size and number density to improve isotropic Jc.

Figure 2 shows TEM cross sectional image and diameter histogram of nanoparticles of x = 0.2 film. We measured the diameter of the nanoparticles from Fig2 (a). One can see that many nanoparticles which are Y-rich composition. The particle sizes are distributed in the range from 3 ~ 12 nm and average diameter (dav) is 8.03 nm. It is considered that these nano-particles trap the quantized flux at 65 K. From these results, we found Y1.2BCO (x = 0.2) thin film exhibited most isotropic Jc(θ).

3.2. Tc Dependence of superconducting properties in magnetic field
We evaluated Jc(θ) in magnetic field the Y1.2BCO thin films prepared at various Tc. Figure 3 shows Jc(θ) at 65 K in 3 T of Y1.2BCO thin films fabricated at Tc = 820 ~ 860°C and fL = 20 Hz. This figure indicates...
Therefore, it is shown that higher density nanoparticles are introduced into the film due to the lowering of deposition rate. In addition, the density of nanoparticles became higher by lowering deposition rate. Additionally, it is shown that higher density nanoparticles are introduced into the film due to the lowering deposition rate. In addition, the density of nanoparticles became higher by lowering deposition rate.

3.3. Influence of f\textsubscript{L} on the isotropy of J\textsubscript{c} in the magnetic field

The influence of the laser repetition frequency f\textsubscript{L} on J\textsubscript{c}(\theta) in magnetic field was evaluated. Figure 4 shows J\textsubscript{c}(\theta) at 65 K, 1 and 3 T of Y\textsubscript{1.2}BCO thin film fabricated at f\textsubscript{L} = 5 ~ 20 Hz and T\textsubscript{s} = 840\degree C. As compared with f\textsubscript{L} = 20 Hz, f\textsubscript{L} = 5 and 10 Hz improved J\textsubscript{c}(\theta) for all magnetic field applied angles, and we confirmed the J\textsubscript{c} dome around B//c appears as increasing with f\textsubscript{L} at 1 T, while the dimple appears by decreasing with f\textsubscript{L} at 3 T around B//c. Matsui et al. reported that such shapes appeared by introducing small-size and high-number density defects induced by light ions irradiation to a YBCO thin film[6]. Therefore, it is shown that higher density nanoparticles are introduced into the film due to the lowering f\textsubscript{L}. Additionally, J\textsubscript{c}s of B//ab for f\textsubscript{L} = 5 and 10 Hz were higher than that of the f\textsubscript{L} = 20 Hz sample. Thus, there is a possibility that ab correlated APCs are introduced by low f\textsubscript{L}.

4. Summary

In this study, we fabricated Y\textsubscript{1.2}BCO thin films with the x range from 0 to 1.0 for isotropic improvement of J\textsubscript{c}(\theta) and evaluated effects of T\textsubscript{s} and f\textsubscript{L}s. As a result, for x, T\textsubscript{s}, J\textsubscript{c}(x,T) showed the largest at x = 0.2. T\textsubscript{s} dependence of J\textsubscript{c}(\theta) at Y\textsubscript{1.2}BCO thin film was evaluated, and we confirmed J\textsubscript{c}(\theta) curves changed by T\textsubscript{s}. It indicates that diameter of nanoparticles changes with T\textsubscript{s}. We finally demonstrated about J\textsubscript{c}(\theta) of Y\textsubscript{1.2}BCO thin films which were fabricated in f\textsubscript{L} = 5 ~ 20 Hz, so we achieved isotropic improvement of J\textsubscript{c}(\theta). This fact that density of nanoparticles became higher by lowering deposition rate. In addition, we suggested that ab-correlated APC which enhanced J\textsubscript{c} of B//ab were introduced by low deposition rate.

These results proved that the deposition conditions have an influence on the isotropy of J\textsubscript{c}(\theta) and also change the nanoparticle size and density.

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