Carrier Density Dependence of the Critical Temperature in BaHfO₃ doped SmBa₂Cu₃O₉ Films

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Abstract. BaHfO₃ (BHO) doped SmBa₂Cu₃O₉ (SmBCO) films show high critical current density (Jc) under magnetic fields. However, excessive addition of BHO causes degradation of superconducting properties. In this study, SmBCO films with different BHO contents were prepared, and the Tc, carrier density and c-axis length were measured. As a result, it was clarified that as the BHO content increases, the c-axis length of SmBCO extends, and Tc and the carrier density decrease. In order to increase the carrier density, we optimized the oxygen annealing condition. As a result, the Tc and the carrier density were increased by annealing at low temperature for a long time.

1. Introduction
Superconducting magnetic energy storage (SMES) and magnetic resonance imaging (MRI) are used under magnetic field, and the improvement of these performances in higher magnetic fields is expected. However, in the superconductors, Jc decreases under magnetic fields. Introducing artificial pinning centers (APCs) in superconductors is one of the promising methods for improving Jc under magnetic field. BaMO₃ (BMO: M=Zr, Sn, Hf) forms nanorods in REBa₂Cu₃O₇ (REBCO: RE=Rare Earth) films fabricated by vapor phase epitaxy [1]. Doping BMO in REBCO films can suppress decrease of the Jc under magnetic fields because BMO nanorods behave as APCs. The number density of nanorods in REBCO films increase with increasing BHO content, and the irreversible magnetic field is expected to be improved. However, the excess BMO addition in REBCO films cause decrease of the Tc [2]. As the number density of BMO nanorods increases, the lattice strain increases, and the c-axis length of REBCO is extended [3]. The extension of c-axis length causes the oxygen deficiency [4].

In this study, we aimed to improve superconducting properties of REBCO films with a large amount of BMO at 77 K high field which can be obtained by cooling with liquid nitrogen. At first, we measured the Tc, c-axis length and carrier density of SmBCO films with different BHO contents. Then, in order to improve the Tc of the films, which contains large volume fraction of BHO of 15.3 vol.%, and we optimized the annealing condition such as annealing temperature and holding time.

2. Experimental details
We fabricated SmBCO films on LaAlO₃(100) substrates by pulse laser deposition (PLD) method using a Nd:YAG pulsed laser. The O₂ partial pressure during the deposition, the distance between the substrate and the targets, and the laser energy density were 53 Pa, 42.5 mm and 2.0 J/cm², respectively. Alternating targets technique was used to introduce various contents of BHO of 0 ~ 15.2 vol.% into
SmBCO films [3]. After the deposition, oxygen was introduced into the chamber, and the substrate was rapidly cooled down to room temperature. In order to suppress the oxygen deficiency of the REBCO films, post annealing in an oxygen atmosphere was performed. The annealing temperature was varied from 150 to 450°C and the annealing time was examined from 1.5 to 6 hours.

The crystal orientation of the films were determined by X-ray diffraction (XRD) method, and inductively coupled plasma (ICP) emission spectroscopy was used to determine the film thickness and the BHO content. The $J_c$ and the $T_c$ were measured using DC four-terminal method by physical properties measurement system (PPMS). The carrier density was calculated based on Hall coefficient obtained by Hall effect measurement at 100 K.

3. Results and discussion

3.1. Dependence of $T_c$ and carrier density on BHO content

After deposition, SmBCO films with various BHO contents were post-annealed in an oxygen atmosphere at 350°C for 3 hours, which is an optimum post annealing condition for a pure SmBCO film. Figure 1 shows Magnetic field dependence of $J_c$ at 77 K, $B//c$ for BHO doped SmBCO and non-doped SmBCO films. The $J_c$ decreased with increasing BHO content. Figure 2 shows BHO content dependence of matching field ($B_φ$). Here, the $B_φ$ is defined by the end of the apparent magnetic field of the plateau in the $J_c$-$B$ curve shown in Figure 1. It indicates that the number density of BHO nanorods increases for the higher BHO content because $B_φ$ is proportional to nanorod density in the single vortex pinning state.

3.2. Dependence of $T_c$ on BHO content

Figure 3 shows BHO content dependence of $c$-axis length, carrier density and $T_c$.
Figure 3 shows BHO content dependence of c-axis length, carrier density, and $T_c$. The carrier density varied in the range of $2.15 \times 10^{21} / \text{cm}^3$ to $3.95 \times 10^{21} / \text{cm}^3$, which is equivalent to the reported values of YBa$_2$Cu$_3$O$_7$ films [5,6]. Although $B_h$ increased with increasing BHO content, $T_c$ decreased. In Figure 3, the carrier density decreases and the c-axis length is extended with decreasing the $T_c$. These indicate that BHO nanorods extend c-axis length of SmBCO by lattice strain, and extension of c-axis length form oxygen vacancies in SmBCO. As a result, we clarified that the lower density of the hole carrier suppresses the $T_c$ in the SmBCO films with the higher BHO contents possibly due to interfacial strain between SmBCO and BHO.

3.2. Annealing temperature dependence of $T_c$ and carrier density
The SmBCO films with the higher BHO contents show poor properties due to a large amount of oxygen deficiency. Therefore, in order to improve the superconducting properties, it is necessary to dope sufficient oxygen and increase the carrier density by optimizing post annealing condition in an oxygen atmosphere. Figure 4 shows annealing temperature ($T_a$) dependence of c-axis length, carrier density and $T_c$. The post annealing time ($t_a$) is fixed at 3 hours. $T_c$ and carrier density increased with increasing $T_a$ up to 200°C, and then monotonically decreased. The c-axis length showed the opposite trend. The optimum annealing condition of pure SmBCO film was 350°C, while the optimum annealing condition of the BHO 15.3 vol.% doped SmBCO film was 200°C. In BHO heavy doped SmBCO films, it is considered that oxygen is easy to desorb from SmBCO matrix at high $T_a$ because the c-axis length of 15.3vol.% BHO doped film is elongated by lattice strain.

3.3. Annealing time dependence of $T_c$ and carrier density
It is expected that oxygen annealing at low $T_a$ needs a long time to reach the equilibrium oxygen content. To optimize $t_a$, the $T_a$ was kept at 200°C, and $t_a$ was varied from 0 to 6 hours. Figure 5 shows $t_a$ dependence of c-axis length, carrier density and $T_c$ of SmBCO films with 15.3 vol.% BHO. As the
annealing time increases, $T_c$ and carrier density increase and the $c$-axis length become shorter. As a result, the $T_c$ of the SmBCO film with BHO of 15.3 vol.% was improved from 85.4 K to 86.7 K.

4. Summary
In this study, we found $T_c$ decreased because of oxygen deficiency by measuring $c$-axis length, carrier density and $T_c$, therefore we attempted to increase $T_c$ by optimizing annealing conditions of BHO heavy doped SmBCO films. The optimum annealing condition of BHO heavy doped SmBCO thin films was different from that of pure SmBCO thin films, and annealing at a low temperature for a long time was necessary compared with non-doped SmBCO thin film. In addition, it is suggested that higher $T_c$ can be obtained by performing further hole doping.

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