Magnetic Field Dependence of Critical Current Density in $Sm_{1+x}Ba_{2-x}Cu_3O_{6+\delta}$ Films Prepared by Pulsed Laser Deposition

K. Sudoh, Y. Ichino, Y. Yoshida, Y. Takai, and I. Hirabayashi

***Abstract*—We have investigated $Sm_{1+x}Ba_{2-x}Cu_3O_{6+\delta}$ (SmBCO) films deposited by pulsed laser deposition technique using stoichiometric and Sm-rich SmBCO ($x = 0.08$) laser targets. In the SmBCO films deposited using the target of $x = 0.08$, the critical current density (J_c) at 77 K with zero applied field was obtained 4.3 MA/cm^2 at film thickness of 1.1 μm.To study the pinning mechanism of SmBCO films, we investigated the magnetic field (0 $\sim$ 9 T) dependence of J_c for $x = 0$ and $x = 0.08$ films. From this result, we found the different behavior of the J_c between $x = 0$ and $x = 0.08$ films. We examined the difference of pinning mechanism from the scaling law and the microstructure of film using transmission electron microscopy.**

***Index Terms*—Critical current density, pinning mechanism, pulsed laser deposition, $Sm_{1+x}Ba_{2-x}Cu_3O_{6+\delta}$ film.**

I. Introduction

To obtain coated conductors based on $REBa_2Cu_3O_{7-y}$ (REBCO) superconductors with high critical current density (J_c) and critical current, considerable effort has been made in the last few years. $Sm_{1+x}Ba_{2-x}Cu_3O_{6+\delta}$ (SmBCO) superconducting materials have higher critical temperatures and irreversible fields, compared with $YBa_2Cu_3O_{7-y}$ (YBCO) superconductor. Unlike YBCO superconductor, SmBCO superconductor exhibits the substitution of Sm^{3+} into the Ba^{2+} site, denoted by the parameter x in the formula $Sm_{1+x}Ba_{2-x}Cu_3O_{6+\delta}$. It was reported that the separation of the superconducting phase and the nonsuperconducting phase in the $Nd_{1+x}Ba_{2-x}Cu_3O_{6+\delta}$ (NdBCO) was caused by the substitution, such as the formation of the solid solution by the spinodal decomposition [1]–[5]. Because of the superior characteristics of NdBCO superconductor, several groups have studied NdBCO films and successfully fabricated high quality NdBCO films by using pulsed laser deposition (PLD) [6]–[8] and sputtering [9]–[11]. However, there are very few studies on the other REBCO films. In case of SmBCO films, several studies have been reported on the relationship between the critical temperature (T_c) and the deposition conditions [12], [13], the composition analysis of Sm/Ba [14] and also the growth mechanism [15]–[17].

We fabricated high-quality c-axis oriented SmBCO films on MgO (100) substrates by PLD using $x = 0$ and $x = 0.08$ laser targets. In this work, we investigate the pinning mechanism for difference of the Sm/Ba substitution quantity.

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II. Experimental

The deposition conditions, the film orientation and the T_c of SmBCO films have been reported in detail elsewhere [18]. SmBCO targets were sintered by the conventional solid reaction methods, starting from quantities of Sm_2O_3, BaO_2 and CuO powders. Appropriate amounts were accurately weighed and thoroughly mixed. The mixed powder was first fired at 700°C. The reacted product was reground and pressed into pellets. Three sintering steps were carried out at 850, 900 and 920°C, respectively, for 24 hour each time with intermediate regrinding and repressing.

SmBCO films were grown on MgO (100) substrates deposited by PLD using a 193 nm ArF excimer laser. The energy density of the laser beam was operated about 1 J/cm^2 at a repetition rate of 10 Hz. The substrate temperature was maintained at 850°C and 810°C. The oxygen partial pressure in the deposition was adjusted 0.4 Torr. The substrate was placed at 50 mm above the laser target. At the end of the deposition the vacuum chamber was filled with pure oxygen up to 20 Torr and the heater was switched off.

The superconducting properties of SmBCO films were measured by usual four-probe point method. Magnetic field from 0 T up to 9 T at 77 K was applied parallel to the c-axis of the SmBCO films. For transport critical current density (J_c) measurement, samples were patterned to produce microbridge of 40-100 μm width and 200 μm length using a KrF excimer laser. Before the J_c and the T_c measurements, the SmBCO films were annealed at 350°C for 1 h in the flowing oxygen.

The microscopic structure and the substitution quantity x of the SmBCO films were observed by transmission electron microscopy (TEM), equipped with an energy dispersive X-ray spectroscopy (EDX) system. Cross section samples for TEM were prepared by ion milling. The orientation of the SmBCO

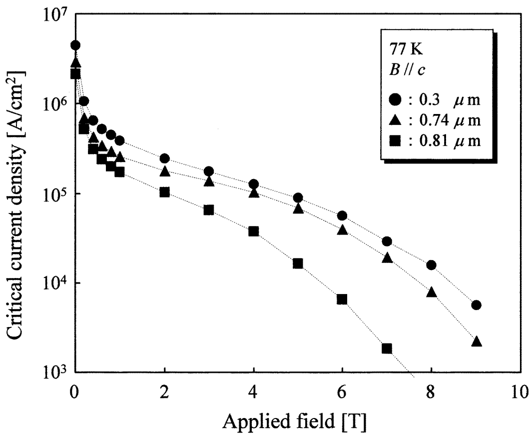


Fig. 1. Magnetic field dependence of critical current density for $x = 0$ SmBCO films of various film thickness. Closed circles, triangles, and squares correspond to the film thickness of 0.3, 0.74, and 0.81 μm, respectively.

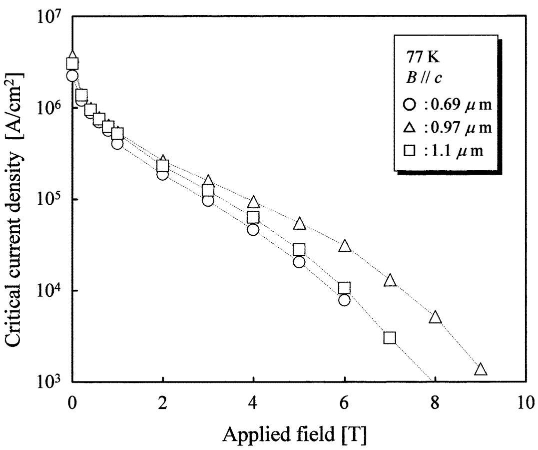


Fig. 2. Magnetic field dependence of critical current density for $x = 0.08$ SmBCO films of various film thickness. Open circles, triangles, and squares correspond to the film thickness of 0.69, 0.97, and 1.1 μm, respectively.

films was examined by X-ray $\theta - 2\theta$ diffraction and Φ-scan using Cu-$k\alpha$ radiation. The film thickness of SmBCO films was evaluated by inductively coupled plasma (ICP) spectrometry.

III. Result and Discussion

To investigate the pinning mechanism of SmBCO film, the SmBCO films on MgO substrates were fabricated using $x = 0$ and $x = 0.08$ laser targets. As reported elsewhere [18], [19], we have obtained c-axis oriented and cube-on-cube textured SmBCO films with the zero resistance temperatures of $\sim$ 91 K and the critical current densities of $\sim$ 4 $\mathrm{MA/cm^2}$.

Fig. 1 shows the magnetic field dependence of the J_c for the SmBCO films of various film thickness deposited with the $x = 0$ target. The applied magnetic field was parallel to the c-axis of the SmBCO film at 77 K. The J_c decreases with increasing the magnetic field, which is similar to the results observed for YBCO films [20] and NdBCO films [21], [22]. The J_c of SmBCO film with film thickness of 0.3 μm is still as high as 1.5×10^4 $\mathrm{A/cm^2}$ at 8 T and 77 K.

On the other hand, the magnetic field dependence of the J_c showed different behavior for the SmBCO films deposited with the $x = 0.08$ target, as shown in Fig. 2. In particular, it is found that the J_c of the $x = 0.08$ films showed higher J_c values in low magnetic field, compared with that of the $x = 0$ films. From these results, it is considered that the $x = 0.08$ films have strong flux pinning site in low magnetic field.

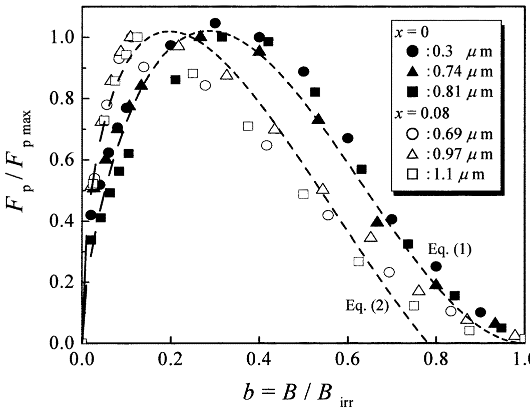


Fig. 3. Scaling property of the flux pinning force density, $F_p = J_c \times B$. Relationship between normalized magnetic field and normalized F_p of $x = 0$ and $x = 0.08$ films. The dashed lines show the curve fitting by (1) and (2).

From the magnetic field dependence of J_c data of Figs. 1 and 2, the scaling property of the flux pinning force density ($F_p = J_c \times B$) is calculated as shown in Fig. 3. The irreversibility field (B_{irr}) is defined as the magnetic field in $J_c = 10^3$ $\mathrm{A/cm^2}$. The flux pinning force density exhibits clear scaling behavior with the magnetic field normalized by B_{irr} and the maximum pinning force density $F_{p\,\mathrm{max}}$.

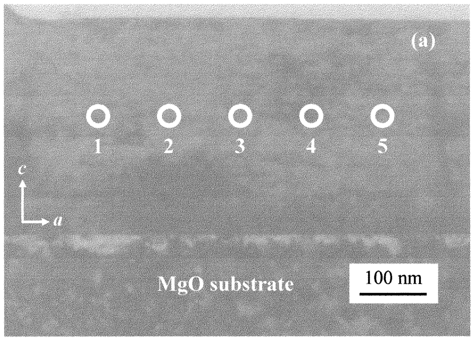
From this result, it is observed that the magnetic field dependence of J_c has weak dependence on the film thickness. However, it is confirmed that the different of pinning mechanism between $x = 0$ and $x = 0.08$ films. In particular, it is observed that the peak of scaling property for $x = 0.08$ films shift the low magnetic field region compared with that of $x = 0$ films. Dashed line in Fig. 3 shown curve fitting by (1) and (2) [23]. From these equations, the experimental $F_p/F_{p\,\mathrm{max}}$ data agree with the theoretical result.

$$F_p \propto b^{1/2}(1-b)^2 + b(1-b)^2 \quad (1)$$

$$F_p \propto b^{1/2}(1-b)^2 + b^{1/2}(1-2b)(2). \quad (2)$$

It is reported that the critical Lorentz force are proportional to $b^{1/2}(1-2b)$, $b^{1/2}(1-b)^2$ and $b(1-b)^2$ for the volume pinning, the plane pinning and the point pinning, respectively [23]. In our case, the best fit to the data results in the volume pinning and the plane pinning for $x = 0.08$ films and the plane pinning and the point pinning for $x = 0$ films.

These differences can be understood given the difference in defect structure between both samples. So, we investigated the defect structure of the $x = 0.08$ film using cross section TEM image. Fig. 4(a) shows a cross section TEM image of the SmBCO film deposited with $x = 0.08$ target. Open circles along the ab plane in this figure indicate the analyzed spots with TEM-EDX. Fig. 4(b) shows the variation of substitution quantity x in the SmBCO film corresponding to the analyzed spots of Fig. 4(a). A horizontal axis represents movement distance parallel to the surface of MgO substrate. And, the vertical axis represents the substitution quantity x of Sm/Ba.From



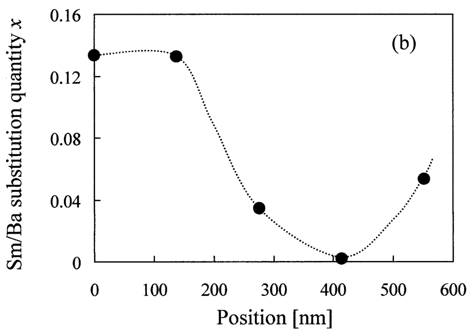


Fig. 4. (a) Cross-sectional TEM image of SmBCO film deposited with $x = 0.08$ target. (b) The variation of substitution quantity x in the SmBCO film measured by TEM-EDX analysis.

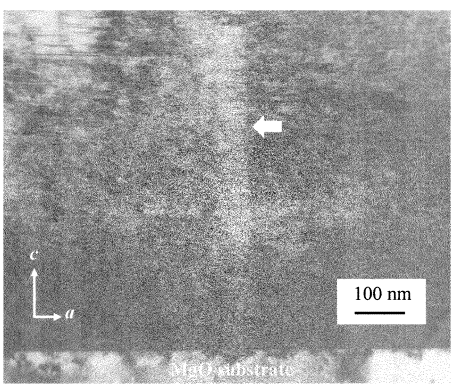


Fig. 5. Cross-sectional TEM image of SmBCO film deposited with $x = 0.08$ target. The arrow indicates the grain boundary along c-axis direction of SmBCO film.

this result, the substitution quantity x fluctuates between 0 and 0.14. Furthermore, it is considered that the Sm-rich phase is dispersed at a distance of 20 nm, which is similar to the results observed for the NdBCO films [4], [24]. This implies that the superconducting phase and nonsuperconducting phase mingle with each other. We assume that the fluctuation of the Sm/Ba ratio caused the difference of scaling property in the low magnetic field.

On the other hand, we investigated the plane pinning shown in Fig. 3. Fig. 5 shows cross section TEM image of the SmBCO film deposited with $x = 0.08$ target. The arrow in the image shows the grain boundary along c-axis direction of SmBCO film. It is considered that the grain boundary caused by the tilt of the grain due to the lattice mismatch between the SmBCO film and the MgO substrate. It is assumed that these defects exerted as the plane pinning for $x = 0$ and $x = 0.08$ films.

IV. Conclusion

Pinning mechanism of SmBCO films were experimentally studied. From the scaling property of the magnetic field dependence of J_c, the different pinning mechanisms between $x = 0$ and $x = 0.08$ films is confirmed. We assume that the different behavior in the low magnetic field is influenced by the fluctuation of the substitution quantity x in the SmBCO film deposited with $x = 0.08$ laser target.

Also, from the cross section TEM image, we observed grain boundary along c-axis direction of SmBCO film. It is considered that these defects exerted as the plane pinning of $x = 0$ and $x = 0.08$ films.

References

[1] S. I. Yoo and R. W. McCallum, “Phase diagram in the Nd-Ba-Cu-O system,” *Physica C*, vol. 210, p. 147, 1993.

[2] S. I. Yoo, M. Murakami, N. Sakai, T. Higuchi, and S. Tanaka, “Enhanced T_c and strong flux pinning in melt-processed $NdBa_2Cu_3O_y$ superconductors,” *Jpn. J. Appl. Phys.*, vol. 33, p. L1000, 1994.

[3] E. Goodilin, M. Kambara, T. Umeda, and Y. Shiohara, “Construction of a quasiternary phase diagram in the $NdO_{1.5}$ -BaO-CuO system in the air atmosphere part II. Phase equilibria of the neodymium-rich $Nd_{1+x}Ba_{2-x}Cu_3O_z$ solid solution,” *Physica C*, vol. 289, p. 251, 1997.

[4] T. Hirayama, Y. Ikuhara, M. Nakamura, Y. Yamada, and Y. Shiohara, “Periodic fluctuation of Ba/Nd ratio in single crystals of high-J_c $NdBa_2Cu_3O_{7-\delta}$ superconductor,” *J. Mater. Res.*, vol. 12, p. 293, 1997.

[5] M. Kambara, K. Miyake, K. Murata, T. Izumi, Y. Shiohara, and T. Umeda, “Non-steady state solidification of the Nd-123 superconducting oxides,” *Physica C*, vol. 330, p. 191, 2000.

[6] M. Badaye, F. Wang, Y. Kanke, and K. Fukushima, “High-quality c-axis oriented superconducting $Nd_{1+x}Ba_{2-x}Cu_3O_{7-\delta}$ thin films deposited by the laser ablation method,” *Appl. Phys. Lett.*, vol. 66, p. 2131, 1995.

[7] H. Zama, K. Ishikawa, T. Suzuki, and T. Morishita, “New growth conditions for the c-Axis oriented $NdBa_2Cu_3O_x$ films by pulsed laser deposition: Lower substrate temperature and higher oxygen pressure,” *Jpn. J. Appl. Phys.*, vol. 38, p. L923, 1999.

[8] Y. Li and K. Tanabe, “Effect of target composition on properties of $NdBa_2Cu_3O_{7-\delta}$ superconducting thin films grown by pulsed laser deposition: A comparative study,” *J. Appl. Phys*, vol. 83, p. 7744, 1998.

[9] I. S. Kim, K. W. Lee, Y. K. Park, and J. C. Park, “Properties of $NdBa_2Cu_3O_{7-\delta}$ thin film prepared by off-axis rf sputtering,” *Appl. Phys. Lett.*, vol. 68, p. 1859, 1996.

[10] M. Suzuki, Y. Soman, E. Tarte, P. Berghuis, and J. Evetts, “Preparation and properties of c-axis-oriented $NdBa_2Cu_3O_{7-\delta}$ films by dc sputtering,” *Physica C*, vol. 312, p. 191, 1999.

[11] Y. Hakuraku, Z. Mori, S. Koba, N. Yokoyama, T. Doi, and T. Inoue, “Critical parameters in the sputter-deposition of $NdBa_2Cu_3O_{7-\epsilon}$ thin films,” *Supercond. Sci. Technol.*, vol. 12, p. 481, 1999.

[12] A. Di Trolio, A. Morone, S. Orlando, U. Gambardella, and S. Pace, “Analysis of growth conditions of SmBaCuO thin films,” *IEEE Trans. Appl. Supercond.*, vol. 9, p. 1583, 1999.

[13] H. M. Appelboom, V. C. Matijasevic, F. Mathu, G. Rietveld, B. Anczykowski, W. J. A. M. Peterse, F. Tuinstra, J. E. Mooij, W. G. Sloof, H. A. Rijken, S. S. Klein, and L. J. van IJzendoorn, “Sm-Ba-Cu-O films grown at low temperature and pressure,” *Physica C*, vol. 214, p. 323, 1993.

[14] A. Di Trolio, A. Morone, N. De Cesare, E. Perillo, and G. Spadaccini, “Compositional analysis of $SmBa_2Cu_3O_{7-x}$ films produced by laser ablation,” *Applied Surface Science*, vol. 154–155, p. 244, 2000.

[15] B. Stäuble-Pümpin, V. C. Matijasevic, B. Ilge, J. E. Mooij, W. J. A. M. Peterse, P. M. L. O. Scholte, F. Tuinstra, H. J. Venvik, D. S. Wai, C. Træholt, J. G. Wen, and H. W. Zandbergen, “Growth mechanisms of coevaporated $SmBa_2Cu_3O_y$ thin films,” *Phys. Rev. B*, vol. 52, p. 7604, 1995.

[16] Q. D. Jiang, D.-M. Smilgies, R. Feidenhans, M. Cardona, and J. Zegenhagen, "Surface morphology and in-plane-epitaxy of $SmBa_2Cu_3O_{7-\delta}$ films on $SrTiO_3$ (001) substrates studied by STM and grazing incidence X-ray diffraction," *Solid. State. Commun.*, vol. 98, p. 157, 1996.

[17] V. C. Matijasevic, B. Ilge, B. Stäuble-Pümpin, G. Rietveld, F. Tuinstra, and J. E. Mooij, "Nucleation of a complex oxide during epitaxial film growth: $SmBa_2Cu_3O_y$ on $SrTiO_3$," *Phys. Rev. Lett.*, vol. 76, p. 4765, 1996.

[18] K. Sudoh, Y. Yoshida, and Y. Takai, "Effect of deposition conditions and solid solution on the $Sm_{1+x}Ba_{2-x}Cu_3O_{6+\delta}$ thin films prepared by pulsed laser deposition," Physica C., to be published.

[19] K. Sudoh, Y. Ichino, M. Itoh, Y. Yoshida, Y. Takai, and I. Hirabahashi, "Correlation between film thickness and critical current density of $Sm_{1+x}Ba_{2-x}Cu_3O_{6+\delta}$ films deposited by pulsed laser deposition," *Jpn. J. Appl. Phys.*, vol. 41, p. L983, 2002.

[20] P. C. McIntyre, M. J. Cima, D. H. Liebenberg, and T. L. Francavilla, "Mixed-state behavior in large magnetic fields of high-temperature superconducting films prepared by metalorganic deposition," *Appl. Phys. Lett.*, vol. 58, p. 2033, 1991.

[21] Y. Li and K. Tanabe, "Influence of deposition parameters on superconductivity and surface morphology of $NdBa_2Cu_3O_{7-\delta}$ films grown by pulsed laser deposition with a single crystal target," *Physica C*, vol. 324, p. 198, 1999.

[22] C. Cantoni, D. P. Norton, D. M. Kroeger, M. Paranthaman, D. K. Christen, D. Verebelyi, R. Feenstra, D. F. Lee, and E. D. Specht, "Phase stability for the *in situ* growth of $Nd_{1+x}Ba_{2-x}Cu_3O_y$ films using pulsed-laser deposition," *Appl. Phys. Lett.*, vol. 74, p. 96, 1999.

[23] D. Dew-Hughes, "Flux pinning mechanisms in type II superconductors," *Philo. Mag.*, vol. 30, p. 293, 1974.

[24] Y. Kumagai, Y. Yoshida, M. Iwata, M. Hasegawa, Y. Sugawara, T. Hirayama, Y. Ikuhara, I. Hirabayashi, and Y. Takai, "Fabrication and characterization of $Nd_{1+x}Ba_{2-x}Cu_3O_{7-y}$ thin films deposited by metal-organic chemical vapor deposition using liquid state sources," *Physica C*, vol. 304, p. 35, 1998.