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Magnetic Field Dependence of Critical Current Density and Microstructure in $Sm_{1+x}Ba_{2-x}Cu_3O_y$ Films on Metallic Substrates

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***Abstract*—We have reported high-J_c $Sm_{1+x}Ba_{2-x}Cu_3O_y$ (SmBCO) films on single crystal MgO prepared by pulsed laser deposition (PLD). In this study, we fabricate SmBCO films on metallic substrates using low temperature growth (LTG) technique, to investigate the availability of the LTG technique for coated conductors. The LTG-SmBCO films are deposited on $PLD-CeO_2/IBAD-YSZ/Hastelloy$ tapes. The critical current density (J_c) is 4.7 MA/cm^2 (77 K, $B//c$, $B = 0$ T) and 0.23 MA/cm^2 (77 K, $B//c$, $B = 5$ T). Furthermore, at 65 K and $B = 17$ T, the J_c of LTG-SmBCO film ($B//c$) reaches 0.03 MA/cm^2. These values are higher than those of conventional PLD-YBCO and PLD-SmBCO films. From these results, we suggest that the LTG is an effective technique for improving the magnetic field dependence of J_c of coated conductors.**

***Index Terms*—Critical current, films, flux pinning, low temperature growth, $SmBa_2Cu_3O_y$.**

I. Introduction

$REBa_2Cu_3O_y$ (REBCO, rare earth (RE) element) coated conductors have an important potential for use in high magnetic field applications such as superconducting magnetic energy storage (SMES) and nuclear fusion. For these applications, high-critical current density (J_c) under a high magnetic field and small anisotropic J_c for a magnetic field direction are required. Recent experiments have showed the improvement of the magnetic field dependence of J_c by introduction of artificial flux-pinning centers into $YBa_2Cu_3O_y$ (YBCO) coated conductors [1]–[3]. We have reported high-J_c $Sm_{1+x}Ba_{2-x}Cu_3O_y$ (SmBCO) films on single crystal MgO prepared by a low-temperature growth (LTG) technique. The LTG-SmBCO ($x = 0.04$) films had $J_c = 0.28$ MA/cm^2 (77 K, $B//c$, $B = 5$ T) [4]. The magnetic field angular dependence of J_c and composition analysis showed that the LTG-SmBCO films had a high density of dislocations [5] and nanosize low-T_c phases within a high-T_c matrix. Moreover, we introduced nanoparticle superconductors with a low-T_c into LTG-SmBCO "LTG-SmBCO+nanoparticle" films on single crystal MgO, to improve J_c under a high magnetic field, and the film showed extremely high J_c, 0.1 MA/cm^2 (77 K, $B//c$, $B = 9$ T) [6], [7]. The composition analysis indicated that the LTG-SmBCO+nanoparticle films had nanosize low-T_c phases and low-T_c network dispersed in the high-T_c matrix [6]. However, for practical applications of REBCO film, it needs to be fabricated on metallic substrates. In this study, we fabricated SmBCO films on metallic substrates using a LTG technique to investigate the availability of LTG technique for coated conductors. We will report the magnetic field dependence of J_c and microstructures of the SmBCO films on metallic substrates.

II. Experimental Procedure

The LTG-SmBCO films with a thickness of 500 nm were prepared by usual pulsed laser deposition technique on Hastelloy tape using the sintered targets. The PLD-SmBCO films were deposited at a substrate temperature (T_s) of 860°C on $PLD-CeO_2$/ion beam assisted deposition (IBAD)-yttrium stabilized zirconium (YSZ)/Hastelloy (hereafter referred to as IBAD). The LTG technique consisted of two steps: firstly, the deposition of a $Sm_{1.08}Ba_{1.92}Cu_3O_y$ seed layer with a thickness of 100 nm at 860°C on IBAD, and secondly the growth of a $Sm_{1.04}Ba_{1.96}Cu_3O_y$ upper layer with a thickness of 400 nm at 780°C on the seed layer. Deposition conditions are listed in Table I. The crystal structure and in-plane alignment of the SmBCO films were examined by X-ray $\theta - 2\theta$ diffraction and ϕ-scan using the (102) plane of SmBCO, respectively. To determine the dislocation density, we etched 500-nm-thick SmBCO films in 0.4 vol.% Br-methanol solution. The surface morphologies of both as-grown and etched films were observed by resonance-mode atomic force microscopy (AFM). The microstructure and the composition were analysed by transmission electron microscopy (TEM), equipped with an energy dispersive X-ray spectroscopy (EDX) system. Resistivity and

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TABLE I
THE DEPOSITIONAL CONDITION OF THE SMBCO FILMS

Parameters	Conditions
Target	seed layer : $Sm_{1.08}Ba_{1.92}Cu_3O_y$
	upper layer : $Sm_{1.04}Ba_{1.96}Cu_3O_y$
Substrate	Hastelloy C276 (60 μm)
Substrate Temperature (T_s)	seed layer : 860°C
	upper layer : 780°C
Oxygen Pressure (pO_2)	400 mTorr
Laser Source	ArF (λ= 193 nm)
Laser Energy	1.0 J/cm^2
Laser Repetition Rate	10 Hz

TABLE II
SAMPLE DATA FOR PLD AND LTG-SMBCO FILMS ON IBAD

Material	$\delta\phi$ (deg.)	T_c(K)	$J_c^{\,s.f}$ (MA/cm^2)
PLD-SmBCO	7.88	92.1	3.0
LTG-SmBCO	7.10	93.0	4.7

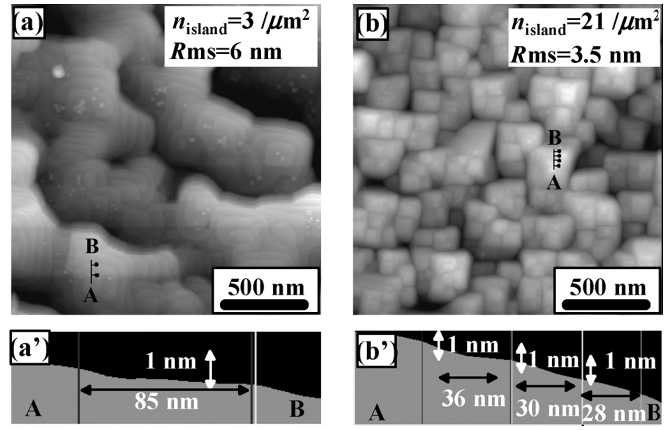

Fig. 1. Surface morphologies of various SmBCO films on IBAD (scan size 2×2 μm^2). (a) top-view and (a') line scan profile of PLD-SmBCO film. (b) top-view and (b') line scan profile of LTG-SmBCO film.

critical current at various temperatures and magnetic fields were measured using a physical property measurement system (PPMS) by a standard four-probe method in a bridge geometry patterned by photolithography. Zero dissipative resistance and critical current density were determined for an electrical field criterion of 1 $\mu V/cm$.

III. RESULTS AND DISCUSSION

A. Crystal Quality and Surface Morphology

Table II shows the crystal quality and superconducting properties of SmBCO films on IBAD fabricated by PLD and LTG techniques. All the SmBCO films presented complete c-axis orientation and cube-on-cube texture on IBAD. However, the PLD-SmBCO film deposited at above 860°C, $BaCeO_3$ formed at a SmBCO $film/CeO_2$ buffer interface. On the other hand, because the seed layer was exposed to high temperature a shorter time compared with PLD-SmBCO film, the LTG-SmBCO film had no $BaCeO_3$. In fact, Ce diffuses when exposing it at a high temperature for a long time after fabricating, and the amount of $BaCeO_3$ increases with increasing time. The LTG-SmBCO film showed high-crystal quality compared with the PLD-SmBCO film. Moreover, T_c and self-field $J_c(J_c^{s.f.})$ of LTG-SmBCO film are higher than those of the PLD-SmBCO film. Based on the case of the LTG-SmBCO films on single crystal MgO, we considered that the excellent crystal quality of the LTG-SmBCO film on IBAD is attributed to the homo-epitaxial growth on seed layer and a small quantity of Cu_2O precipitates [8].

Fig. 1 shows the surface morphology of SmBCO films fabricated by PLD and LTG techniques. We found that the PLD-SmBCO film was grown up by the spiral growth mode, on the other hand, the LTG-SmBCO film was grown up by 2D island growth mode. The LTG-SmBCO film has a very smooth surface compared with the PLD-SmBCO film. This result is important for thicker films, because a rough surface causes a misaligned growth. In addition, Figs. 1(a) and 1(b) shows that the island density (n_{island}) of LTG-SmBCO film is higher than that of PLD-SmBCO film. As displayed in Figs. 1(a') and 1(b'), the height of the spiral and 2D steps are about 1 unit cell of SmBCO crystal, and the LTG-SmBCO film has short step width compared with the PLD-SmBCO film. The LTG and PLD-SmBCO films have step width of ~36, ~85 nm, respectively. Here, we discuss the relationship between T_s and crystal growth. Since the PLD-SmBCO films were fabricated at a high-T_s, i.e., a low supersaturation, the films were grown up by spiral growth originated from screw dislocations. On the other hand, the LTG-SmBCO films were fabricated at a low-T_s, i.e., a high supersaturation, the nucleation was easier than that of other processes. Besides, in such a high supersaturation, the growth rates of spiral and 2D growths were almost equal. Note that when the screw dislocation density was very small, 2D growth was dominated in the LTG-SmBCO films. In general, the critical radius for nuclei and step width in such a high supersaturation were small comparing with the case of a low supersaturation [9]. As a result, the films fabricated at a high supersaturation such as the LTG-SmBCO film has a high-n_{island} and short step width, which is consistent with Fig. 1(b').

B. Critical Current Density and Its Field Dependence

The effect of LTG technique on the magnetic field dependence of $J_c(J_c - B)$ at both 65 and 77 K is shown in Fig. 2. At 77 K the LTG-SmBCO film shows $J_c = 0.24\ MA/cm^2$ at $B = 5\ T\ (B//c)$, this value is almost equal to that of NbTi wire measured at 4.2 K and as high as film on single crystal MgO. Additionally, at 65 K the LTG-SmBCO film has 0.03 MA/cm^2 at $B = 17\ T\ (B//c)$, which is higher than $(NbTi)_3Sn$ tube at 4.2 K. We considered that the LTG-SmBCO film has improved $J_c - B$ property under a high magnetic field by enhancement of pinning center and irreversibility field (B_{irr}).

B_{irr} is one of the factors affecting the $J_c - B$ property. Fig. 3 shows B_{irr} curve for PLD and LTG-SmBCO films. B_{irr} is determined by a resistivity criterion of 0.1 $\mu\Omega cm$, which corresponds to the $J_c = 10\ A/cm^2$ defined by the 1 $\mu V/cm$. From Table II, T_c of the LTG-SmBCO film is higher than the PLD-SmBCO film, therefore the B_{irr} of LTG-SmBCO film shows high-B_{irr} under a high temperature. Moreover, at below 77 K the B_{irr} of the LTG-SmCBO film is markedly higher than that of PLD-SmBCO film. The LTG and PLD-SmBCO films have $B_{irr} = 11.32$, 10.01 T at 77 K, respectively. We speculated that

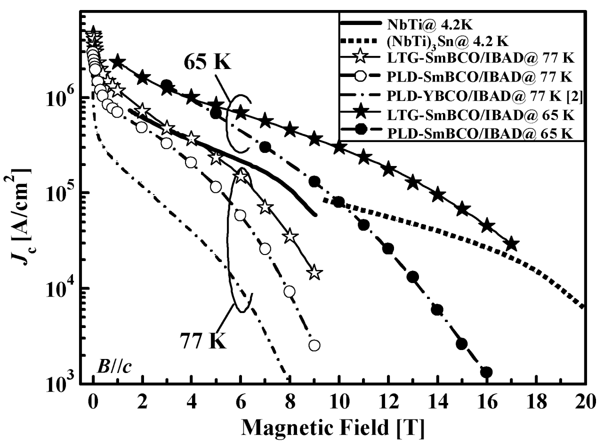

Fig. 2. The magnetic field dependence of J_c measured for $B//c$ at both 65 and 77 K for LTG-SmBCO film on IBAD, compared with PLD-SmBCO, PLD-YBCO films [2], NbTi and $(NbTi)_3Sn$. Opened and filled symbols show J_c at 77 and 65 K, respectively.

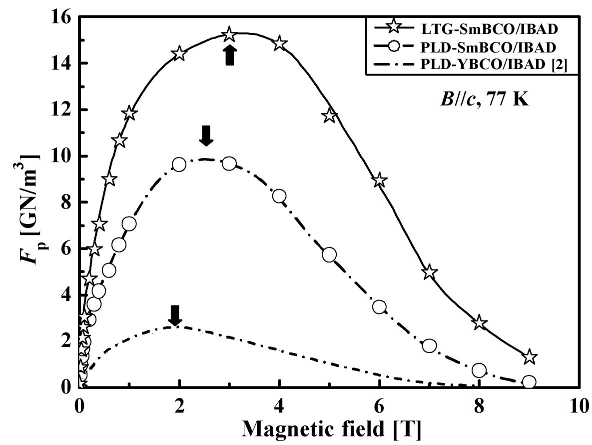

Fig. 4. F_p for PLD and LTG-SmBCO films on IBAD at 77 K as a function of magnetic field.

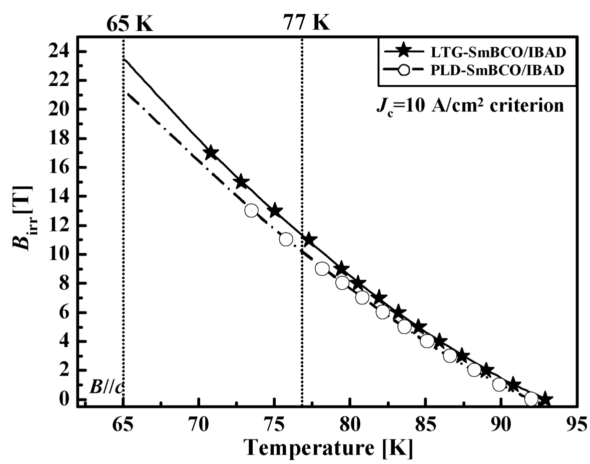

Fig. 3. B_{irr} for $B//c$ in PLD and LTG-SmBCO films on IBAD.

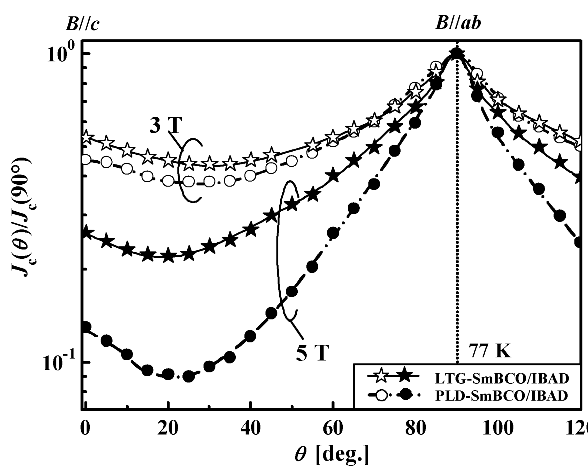

Fig. 5. Magnetic field angular dependence of J_c at 77 K for LTG-SmBCO film on IBAD compared with that of PLD-SmBCO film. As indicated in figure, $\theta = 0°$ corresponds to $B//c$, and $\theta = 90°$ to $B//ab$. Opened and filled symbols show J_c at $B = 3$ and 5 T.

the B_{irr} of the LTG-SmBCO films was improved difference of pinning center. Thereby, we noticed the $J_c - B$ property correlated with B_{irr}.

In order to investigate the pinning property, we calculated the pinning force $(F_p = J_c \times B)$. Fig. 4 displays F_p as a function of magnetic field for PLD and LTG-SmBCO films on IBAD at 77 K, compared with PLD-YBCO films [2]. The LTG-SmBCO film shows striking improvement of F_p at both low and high magnetic field compared with PLD-SmBCO and PLD-YBCO films, the maximum of F_p reaches 15 GN/m^3 at 77 K and $B = 3$ T. We can consider that the pinning center is improved by using a LTG technique has enhanced pinning property in the LTG-SmBCO film.

We measured the angular dependence of J_c at 77 K for PLD and LTG-SmBCO films on IBAD to investigate the pinning center configuration. Fig. 5 shows the $J_c - B - \theta$ curve at $B = 3$ and 5 T. Under these magnetic fields, the LTG-SmBCO film shows isotropic $J_c - B - \theta$ compared with the PLD-SmBCO film, as well as the LTG-SmBCO film on single crystal. This result supports to the developing possibility of high magnetic field application such as SMES of solenoid type.

C. *Pinning Centers in LTG-SmBCO Film on IBAD*

By a lot of experiments for pinning center, many kind of pinning centers in SmBCO films have been considered. In this section, we will discuss about the low-T_c phase playing as field-induced pinning centers and linear defects such as dislocation.

Fig. 6 shows the etched surface morphologies of PLD and LTG-SmBCO films on IBAD. As shown in Fig. 6, we found that the dislocation density (n_{disl}) of the LTG-SmBCO film is higher than that of PLD-SmBCO film. From our previous experimental result, n_{disl} increases with increasing n_{island}, because the mechanism for edge dislocation formation was mainly related to merging of misaligned growth front [5]. In fact, because the LTG-SmBCO film on IBAD had a high-n_{island} (see Fig. 1(b)), that has a high-n_{disl}. Additionally, we considered that the c-axis angular peaks (see Fig. 5) in the LTG-SmBCO film on IBAD were due to high-n_{disl}.

As another kind of pinning center, we speculated that the improved $J_c - B$ (Fig. 2), B_{irr} (Fig. 3) and $J_c - B - \theta$ (Fig. 5) under a high magnetic field in the LTG-SmBCO film on IBAD were due to mainly nanosize low-T_c phase within high-T_c matrix, because these behaviors were the same as in the case of

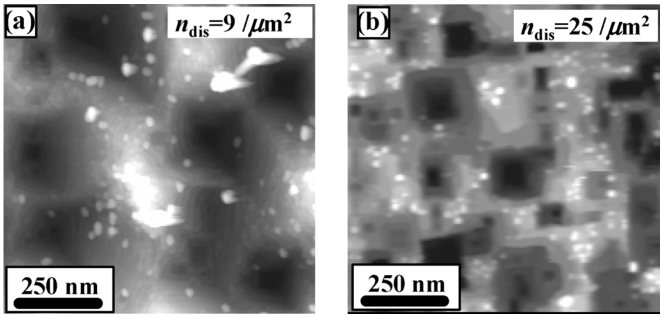

Fig. 6. The 30s-etched surface morphology of (a) PLD and (b) LTG-SmBCO film on IBAD (scan size 1×1 μm^2).

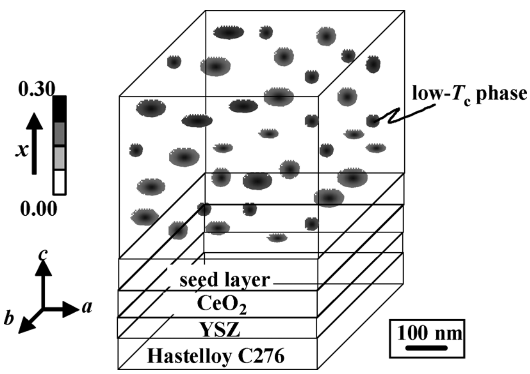

Fig. 7. Schematic drawing of 3D compositional distribution of LTG-SmBCO films on IBAD which were inferred from microstructural observation based on the TEM-EDX in LTG-SmBCO film on single crystal MgO [4].

film on single crystal MgO that had nanosize low-T_c phases [4]. Fig. 7 shows schematic drawing of 3D compositional distribution of LTG-SmBCO film on IBAD which was inferred from microstructural observations based on the TEM-EDX in LTG-SmBCO film on single crystal. If there are low-T_c phases with x of 0.2–0.3, under a high magnetic field, low-T_c phases would become a normal state, and then can be able to contribute to flux pinning, and therefore these should derive to enhance the magnetic field dependence of properties. In fact, as shown in Fig. 8, above $x = 0.2$ films at 77 K were normal state. Therefore, not only the density of low-T_c phases and the size but also the Sm/Ba composition within the low-T_c phases are important in the improvement of $J_c - B$ and $J_c - B - \theta$ properties.

IV. Conclusion

We have fabricated SmBCO films on metallic substrates using a low temperature growth (LTG) technique. The LTG-SmBCO film on metallic substrate showed an excellent crystal quality, high-T_c, high-$J_c^{s.f.}$, enhancement of $J_c - B$, improvement of B_{irr} and $J_c - B - \theta$ compared with that of conventional PLD-SmBCO and PLD-YBCO films, as well as the LTG-SmBCO film on single crystal MgO. Additionally, the low-temperature deposition adopted in the LTG technique

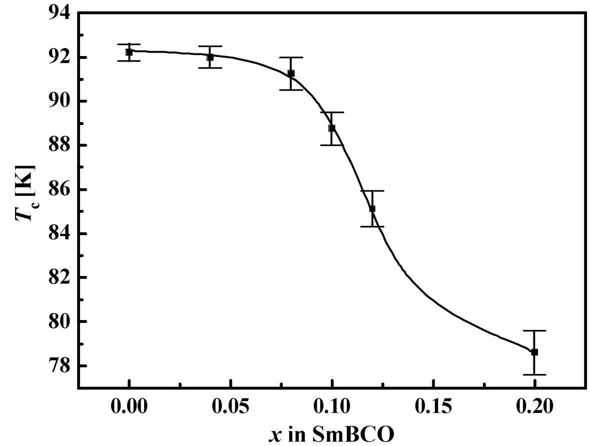

Fig. 8. T_c of SmBCO film on MgO with $x = 0$–0.2. T_c decreases with increasing x in SmBCO film.

results in protects formation of $BaCeO_3$, that is expected to prevent the diffusion of metal. From these results, we suggest that LTG is an effective technique for improving the magnetic field dependence of J_c of coated conductors.

References

[1] J. L. Macmanus-Driscoll, S. R. Foltyn, Q. X. Jia, H. Wang, A. Serquis, L. Civale, B. Maiorov, M. E. Hawley, M. P. Maley, and D. E. Peterson, "Strongly enhanced critical current density in superconducting coated conductors of $YBa_2Cu_3O_{7-\delta} + BaZrO_3$," *Nature Materials*, vol. 3, pp. 439–443, 2004.

[2] Y. Yamada, K. Takahashi, H. Kobayashi, M. Konishi, T. Watanabe, A. Ibi, T. Muroga, S. Miyata, T. Kato, T. Hirayama, and Y. Shiohara, "Epitaxial nanostructure and defects effective for pinning in $Y(RE)Ba_2Cu_3O_{7-x}$ coated conductors," *Appl. Phys. Lett.*, vol. 87, p. 132502, 2005.

[3] S. Kang, A. Goyal, J. Li, A. A. Gapud, P. M. Martin, L. Heatherly, J. R. Thompson, D. K. Christen, F. A. List, M. Paranthaman, and D. F. Lee, "High-performance high- T_c superconducting wire," *Science*, vol. 311, pp. 1911–1913, 2006.

[4] M. Miura, M. Itoh, Y. Ichino, Y. Yoshida, Y. Takai, K. Matsumoto, A. Ichinose, S. Horii, and M. Mukaida, "Effect of Sm/Ba substitution on the J_c in magnetic field of SmBCO thin films by low temperature growth technique," *IEEE Trans. Appl. Supercond.*, vol. 15, pp. 3078–3081, 2005.

[5] M. Miura, Y. Yoshida, Y. Ichino, T. Ozaki, Y. Takai, K. Matsumoto, A. Ichinose, S. Horii, and M. Mukaida, "Dislocation density and critical current density in $Sm_{1+x}B_{2-x}Cu_3O_y$ films prepared by various fabrication processes," *Jpn. J. Appl. Phys.*, vol. 45, pp. L701–L704, 2006.

[6] M. Miura, Y. Yoshida, Y. Ichino, Y. Takai, K. Matsumoto, A. Ichinose, S. Horii, and M. Mukaida, "Enhancement of flux-pinning in epitaxial SmBCO films by introduction of low- T_c nanoparticles," *Jpn. J. Appl. Phys.*, vol. 45, pp. L11–L13, 2006.

[7] M. Miura, Y. Ichino, Y. Yoshida, Y. Takai, K. Matsumoto, A. Ichinose, S. Horii, and M. Mukaida, "Addition of low- T_c-nanoparticles dispersions to enhance flux pinning of $Sm_{1+x}Ba_{2-x}Cu_3O_y$ films," *Physica C*, vol. 445–448, pp. 643–647, 2006.

[8] M. Miura, M. Itoh, Y. Yoshida, Y. Ichino, Y. Takai, K. Matsumoto, A. Ichinose, S. Horii, and M. Mukaida, "In-plane alignment and superconducting properties in high- J_c $Sm_{1+x}Ba_{2-x}Cu_3O_{6+\delta}$ thin films," *Physica C*, vol. 426–431, pp. 985–989, 2005.

[9] T. Nishinaga and H. J. Sheel, "Crystal growth aspect of high- T_c superconductor," *Advance in Superconductivity VIII*, pp. 33–38, 1996.