# Application of Nd:YAG laser to preparation of $REBa_2Cu_3O_z$ films for wire processing (RE: Y or rare-earth element)

**G I P De Silva[1,4], T Maeda[1,3], S Horii[1], Y Ichino[2] and Y Yoshida[2]**
[1] Kochi University of Technology, Tosayamada-cho, Kochi 782-8502, JAPAN
[2] Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, JAPAN
E-mail: maeda.toshihiko@kochi-tech.ac.jp

**Abstract**. Fourth harmonics of Nd:YAG (neodymium-doped yttrium-aluminum-garnet) laser at 266 nm was used for pulsed-laser-deposition (PLD) method to prepare $(Y_{1-x}Ho_x)Ba_2Cu_3O_z$ ($x$=0, 0.3, 0.5, 0.7 and 1) films on (100) $SrTiO_3$ (STO) single crystal substrates. Ho was selected as the substituting element for Y site because the ionic radius of $Ho^{3+}$ is almost identical to that of $Y^{3+}$ and as a result lattice distortion may not occur. It may be possible to generate some pinning effect as a result of random electron potential energies induced by the randomly occupied (Y,Ho) sites. Texture quality and current characteristics of the obtained films were analyzed with the intension of extending this work towards the cost effective coated-conductor fabrication.

## 1. Introduction

In coated-conductor fabrication, many kinds of deposition process have been used to prepare $REBa_2Cu_3O_z$ (REBCO; RE: Y or rare-earth element) films. Among them, pulsed-laser-deposition (PLD) method, which is one of laser-ablation processes, has been extensively used for many researches. KrF excimer laser, especially, is popular choice among the PLD community because its short wavelength of 248 nm causes high pulse laser energy output [1,2]. However, some important challenges or fundamental problems hindering coated-conductor applications need to be scientifically solved, i.e., reducing the fabrication cost with high reproducibility and finding the most effective and economic approaches to enhance the superconducting critical current density ($J_c$) in high magnetic fields which is the most significant characteristic for the coated-conductor application of high-temperature superconductors (HTSC's) [3-5].

Possible solutions for these problems are using Nd:YAG (neodymium doped yttrium aluminium garnet; Nd:$Y_3Al_5O_{12}$)-type laser [6,7] which has relatively lower installation cost and longer lifetime compared to the excimer lasers and investigeting substitution effect of some rare-earth elements for Y sites of $YBa_2Cu_3O_z$ (YBCO). For the former, there have been few reports on preparation of PLD-REBCO coated conductors using Nd:YAG laser [8-10], and for the latter, partial substitution of some rare-earth (RE) elements for Y sites of YBCO is known to be one of effective procedures to increase $J_c$ in an external magnetic field. The replacement of Y by RE causes local distortion of the crystal structure and electron density and as a result strain induced pinning improvements may occur [11]. This effect is expected to depend on valence and size of $RE^{3+}$ ion.

---

[3] To whom any correspondence should be addressed.

[4] Present address: Dept. Mater. Sci. & Eng., Univ. Moratuwa, Katubedda, Moratuwa, Sri Lanka.

In the present work, $Ho^{3+}$ is selected as the dopant because the ionic radius of $Ho^{3+}$ is almost similar to that of $Y^{3+}$ ($Y^{3+}$: 0.1019 nm, $Ho^{3+}$: 0.1015 nm [12]) and therefore lattice distortion may not occur. $(Y,Ho)Ba_2Cu_3O_z$ films are prepared applying improved Q-switched Nd:YAG pulse laser (LOTIS TII Ltd.; model LS2147) at 266 nm to the PLD process with the ultimate objective of high $J_c$ coated-conductor fabrication. $(Y,Ho)Ba_2Cu_3O_z$ thin films are deposited on (100) $SrTiO_3$ (STO) single-crystal substrates. Texture quality and current characteristics of the prepared films are analyzed with respect to the Y:Ho ratio.

## 2. Experimental

Targets for the PLD process were prepared by a solid-state reaction of commercially available powders of $Y_2O_3$, $Ho_2O_3$, $BaCO_3$ and CuO. Target composition was set at $x$=0, 0.3, 0.5, 0.7 and 1 in $(Y_{1-x}Ho_x)Ba_2Cu_3O_z$. 0.5 mm thick mirror-polished (100) $SrTiO_3$ (STO) single crystals were used as substrates. Prior to deposition, these substrates were immersed in 1M $HNO_3$ for 10 min and subsequently subjected to pre-annealing treatment at 850 °C for 6 hours in air. Thin films of $(Y,Ho)Ba_2Cu_3O_z$ films were grown by the PLD method using 266 nm radiation of the Nd:YAG laser. At first, deposition chamber was evacuated down to 0.01 Pa. Repetition rate of the laser pulse and pulse duration were respectively set at 10 Hz and 18 ns. The target-to-substrate distance (T-S distance; $d_{TS}$) was adjusted between 30 and 40 mm. The displayed temperature which corresponds to substrate temperature during the film deposition ($T_{sub}$), and oxygen gas pressure were set at 800-870 °C and 40-50 Pa, respectively. Pump energy was adjusted between 35 and 45 J. Laser spot area at the surface of the target was evaluated as 0.021 $cm^2$ which corresponded to laser energy density at the surface of the target ($E$). Target rotation speed of 20 rpm and target-to-lens distance of 50 cm were kept constant. After deposition, post-annealing at 450°C for 30 min in a flowing oxygen gas was carried out in order to make the as-deposited films superconducting.

Degree of c-axis orientation (<001> of REBCO perpendicular to the substrate surface) was analyzed by means of $\theta$-$2\theta$ scanning of X-ray diffractometry (XRD) and in-plane texturing (<100> or <010> of REBCO parallel to <100> of STO; "cube-on-cube" configuration) was assessed by XRD $\phi$-scan measurement. CuK$\alpha$ radiation was used for both of the XRD measurements. Film thickness was estimated using a cross-sectional view of fractured samples taken by means of field-emission scanning-electron-microscopy (FE-SEM), and film deposition rate was calculated using the thickness values. Critical temperature ($T_c$) and critical current ($I_c$) were measured using a standard four-probe method.

## 3. Results and discussion

### 3.1. Texture quality

It is well known that two types of c-axis oriented REBCO grains are deposited on (100) STO single-crystal substrates. One is the "cube-on-cube" configuration and another one is "45° rotated" configuration in which each grain is 45° rotated around <001> axis of REBCO from the "cube-on-cube" configuration [13]. In this study, PLD parameters were varied in order to obtain the optimum PLD condition which corresponds to c-axis oriented thin film with improved in-plane texture ("cube-on-cube" configuration) because degree of the in-plane texturing plays a significant role in enhancing $J_c$. The determined optimum sets of parameters for each target composition are listed in Table 1.

Highly c-axis oriented thin films were prepared for all films. However, all contained both "cube-on-cube" and a small amount of "45° rotated" configurations. ("cube-on-cube" configuration is predominant). Figure 1 shows the XRD $\phi$-scan profile of a sample with $x$=0.3.

### 3.2. Current characteristics

The substrate materials with various crystal structures and lattice parameters influence on the introduction of crystalline defects in REBCO thin films during crystal growth. Research reports [14] have shown that small misfit between YBCO and STO single-crystal causes better crystalline quality

**Table 1.** Optimized PLD parameters for $(Y_{1-x}Ho_x)BCO$ films on STO substrates

| Composition | $x$=0 | $x$=0.3 | $x$=0.5 | $x$=0.7 | $x$=1 |
|---|---|---|---|---|---|
| [a]Displayed temp. (°C) | 840 | 840 | 850 | 857 | 865 |
| $O_2$ pressure (Pa) | 45 | 45 | 45 | 45 | 45 |
| T-S distance (mm) | 37 | 37 | 37 | 37 | 37 |
| Energy density ($Jcm^{-2}$) | 1.8 | 1.8 | 1.8 | 1.8 | 1.8 |
| Deposition time (min) | 20 | 20 | 20 | 20 | 20 |

[a]The temperature displayed in the PLD machine ($T_s$ is lower than this value). The relationship between $T_s$ and displayed temperature is not detected yet.

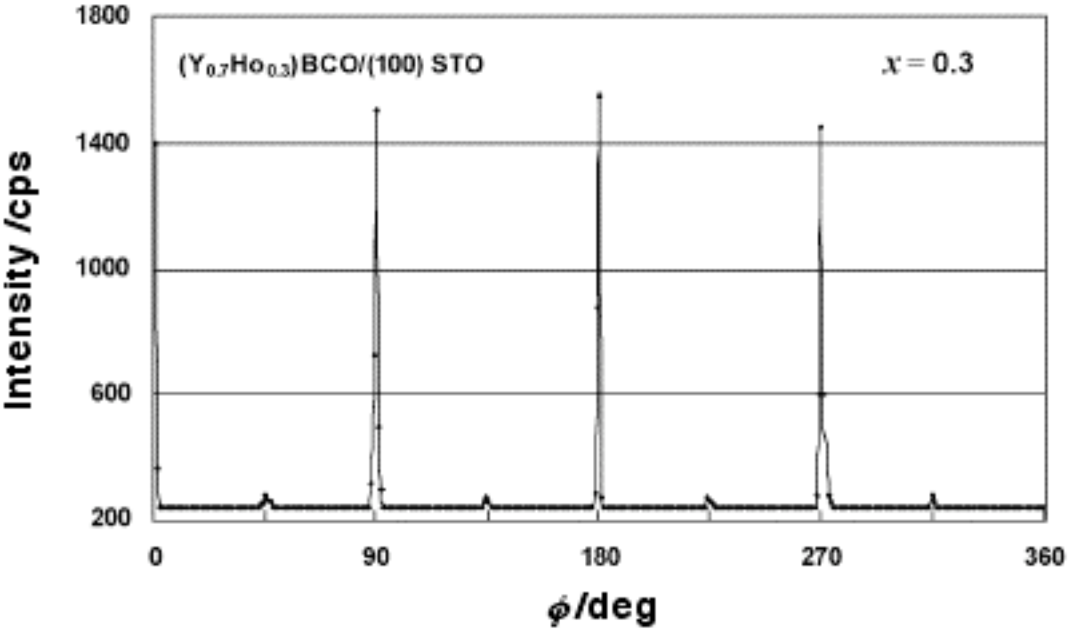

**Fig. 1.** XRD $\phi$-scan profile for sample with $x$ = 0.3 prepared on STO substrate.

and this results in intrinsic pinning properties. Therefore in the present study STO single-crystals were selected with the aim of improving $J_c$.

The $J_c$ in epitaxial REBCO films strongly depends on the film thickness. Several reports reveal that the nanostructure of pulsed-laser-deposited epitaxial REBCO films is evolving during growth due to high dislocation mobility and this causes degradation of the $J_c$ in thick films [15,16]. As an initial step, therefore, relatively thin films with thickness of 200-250 nm were fabricated. Figure 2 shows the FE-

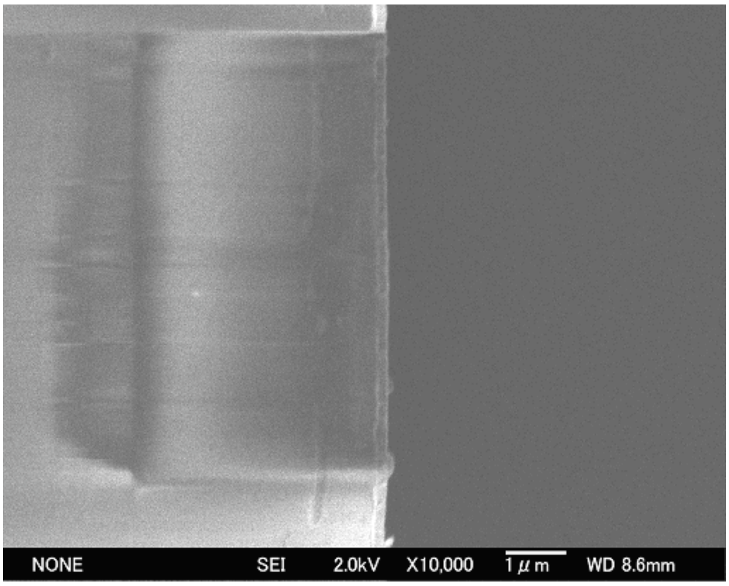

**Fig. 2.** FE-SEM cross-sectional image of sample with $x$=0.3.

**Table 2.** $T_c$ and thickness for $(Y_{1-x}Ho_x)$BCO films on STO substrates

| Composition | $x = 0$ | $x = 0.3$ | $x = 0.5$ | $x = 0.7$ | $x = 1$ |
|---|---|---|---|---|---|
| $T_c$ (K) | 90 | 87 | 89 | 89 | 89 |
| Thickness (nm) | 250 | 200 | 200 | 250 | 200 |

SEM cross-sectional image of the sample with $x$=0.3. Average film deposition rate was calculated to be 11 nm/min using the thickness values.

$T_c$ values for all films were obtained in the range of 87-90 K as listed in Table 2 which were thought to be practically sufficient values for coated conductors. Temperature dependence of resistivity around $T_c$ for sample with $x$=0 is shown in Fig. 3. $I_c$ was measured at 77.3 K for narrow bridge in patterned samples using the criterion of 10 μV/cm and those values are listed in Table 3. $J_c$ values were determined to be around 1 MA/cm$^2$ (0.8-1.5 MA/cm$^2$) except for $x$=0. This extremely low $J_c$ of $x$=0.5 could be caused by some experimental error such as a failure in oxygen annealing. However, according to the other $J_c$ values appeared in Fig. 4, it seems that $J_c$ reaches the highest value at the most disordered sample of $x$=0.5 and decreases as the samples become purer. This result shows that Y:Ho ratio influences $J_c$ as can be seen in Fig. 4. It is likely that different electron potential energies in randomly distributed $Y^{3+}$ and $Ho^{3+}$ sites have some effect on flux pinning.

Even though these $J_c$ values are not so high at present, it is expected that these can be increased to much higher values by adjusting the parameters such as wavelengths (second harmonics: 532nm; third harmonics: 355nm), pulse repetition rate and pulse duration (FWHM).

In the next stage of this work, furthermore, influence of film thickness on $J_c$ should be studied will be studied with the aim of finding optimum thickness which corresponds to the maximum $I_c$ because

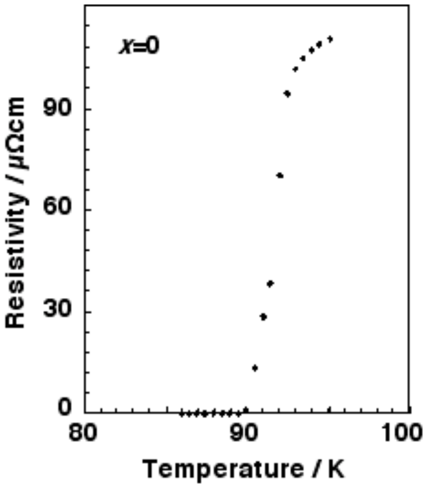

**Fig. 3.** Temperature dependence of resistivity for sample of $x$=0.

**Table 3.** $I_c$ and $J_c$ of $(Y_{1-x}Ho_x)$BCO films on STO substrates.

| Composition | $x = 0$ | $x = 0.3$ | $x = 0.5$ | $x = 0.7$ | $x = 1$ |
|---|---|---|---|---|---|
| $I_c$ (mA) ;77.3 K, self field | 310 | 316 | 27 | 367 | 186 |
| $J_c$ MA/cm$^2$ | 1.20 | 1.45 | 0.14 | 1.26 | 0.83 |

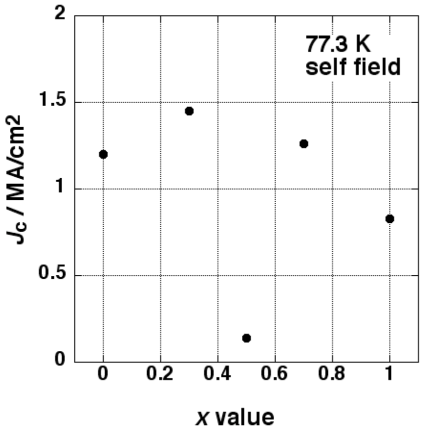

**Fig. 4.** Y:Ho ratio dependence of $J_c$.

HTS coated conductors are required to transport large amounts of current suitable for electric power applications.

## 4. Conclusion

Forth harmonics of Nd:YAG pulse laser was applied to the PLD process in order to prepare (Y,Ho)$Ba_2Cu_3O_z$ films focusing on coated-conductor fabrication. Highly biaxially-textured films were successfully grown on (100) STO substrates. For all films, $T_c$ values were measured to be around 90 K.

Present results show that Nd:YAG-PLD can be one of acceptable processes for cost-effective coated-conductor fabrication in the near future. For the effects of disordering in the rare-earth sites on $J_c$, while it was likely that the most disordered sample of $x$=0.5 had the highest $J_c$, we could not confirm that because of some experimental error.

## Acknowledgement

The authors would like to thank Prof. K. Matsumoto for fruitful discussion and also to thank Prof. T. Doi of Kagoshima University for his kind help in critical-current measurement. They also thank Mr. K. Kanai and Ms. M. Yamaki of Kochi University of Technology for their kind support in PLD experiments.

## References

[1] Chrisey D B and Hubler G K 1994 *Pulsed Laser Deposition of Thin Films* (New York: John Wiley and Sons, Inc) p 25
[2] Michael N, Ashfold R, Claeyssens F, Gareth M and Simon J 2004 *Chem. Soc. Rev.* **33** 23-31
[3] Shiohara Y, Yoshizumi1 M, Izumi T, Yamada Y 2008 *Supercond. Sci. Technol.* **21** 034002
[4] Inoue M, Kiss T, Motoyama K, Nakamura T, Fujiwara T, Mitsui D, Awaji S, Watanabe K, Ibi A., Takahashi K, Miyata S, Yamada Y and Shiohara Y 2007 *Physica C* **463-465** 674-677
[5] Matsushita T, Kiuchi M, Kimura K, Takayama S, Yamada Y, Shiohara Y, Fujikami J, Hayashi K and Sato K 2007 *Physica C* **463-465** 686-691
[6] Park J S, Kim C O, Lee Y P and Prokhorov V G 2003 *Supercond. Sci. Technol.* **16** 893
[7] Bowman R M, Pegrum C M, Goodyear S W and Edwards J A 1991 *Supercond. Sci. Technol.* **4** 427
[8] Kiss T, Enpuku K, Mathumura T, Iriyama Y, Nakamura T and Takeo M 1996 *IEICE Transactions on Electrics* **E79C** 1269
[9] Kusumori T and Muto H 2000 *Physica C* **57-61** 337
[10] De Silva G I P and Maeda T 2009 *J. Mater. Sci. Soc. Jpn.* **46** 236-241
[11] MacManus-Driscoll J L, Foltyn S R, Jia Q X et al 2004 *Appl. Phys. Lett.* **84** 5329
[12] Shannon R D 1976 *Acta Cryst.* **A32** 751
[13] Maeda T, Ohashi Y, Watanabe T, Iwai H, Hirabayashi I and Mimura M 2003 *Physica C* **392-396** 821-829
[14] Horide T, Matsumoto K, Osamura K, Ichinose A, Mukaida M, Yoshida Y and Horii S 2004 *Physica C* **412–414** 1291–1295
[15] Cherpak Y V, Svetchnikov V L, Semenov A V, Moskaliuk V O, Tretiatchenko C G, Flis V S, and Pan V M 2008 *J. Phys.*: Conf. Ser. **97** 012259
[16] Cherpak Y V, Moskaliuk V O, Semenov A V, Svetchnikov V L, Tretiatchenko C G and Pan V M 2007 *Supercond. Sci. Technol.* **20** 1159–1164