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Fabrication and critical current properties in Nd:YAG-PLD
REBa$_2$Cu$_3$O$_y$ (RE=Y and Er) thin films

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Abstract

We report optimization of substrate temperature ($T_s$) regions for achievement of the bi-axially oriented growth without the a-axis oriented domains, critical current properties and crystallinities in REBa$_2$Cu$_3$O$_y$ (RE123) films (RE= Y and Er) grown by pulsed laser deposition using a Nd:YAG laser. Appropriate $T_s$ regions for achieving both high critical temperature and self-field critical current density in Y123 and Er123 films were found to be similar. In the case of Y123, infield critical current densities and inplane lattice parameters for the Y123 films grown in a lower $T_s$ region were slightly changed compared with those for the films grown at a higher $T_s$ region. Our findings in the present study give us fundamental information to understand intrinsic effects of introduction of nanorods in the RE123 films containing nanorods.

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Keywords: Nd:YAG laser; pulsed laser deposition; RE123; thin film; critical current property

1. Introduction

A pulsed-laser-deposition (PLD) method using excimer lasers as an energy source for deposition has been intensively utilized for growth of REBa$_2$Cu$_3$O$_y$ (RE123) thin films as a practical production process of coated conductors of RE123. This is because RE123 films with high superconducting performances have been obtained easily by the excimer-laser PLD in comparison with the cases by other light sources. However, deposition apparatuses based on the excimer-laser PLD require halogen and noble gases, leading to high running costs and dangerous operation for the production of the coated conductors. From the viewpoints of low-cost production and safety of the production process, a Nd:YAG laser is a promising candidate as a practical laser source in the PLD system [1,2]. This is because the Nd:YAG laser can be generated only by electric source without gases and, therefore, reduction of the running cost is also expected.

Recently, introduction of nanorods into RE123 films by PLD based on the excimer laser and drastic improvement of their infield critical current densities ($J_c$), which is due to the introduction of vortex pinning centres, have been intensively reported[3-7]. Furthermore, changes in infield $J_c$ [8] and irreversibility line (IL) [9] for the change in deposition temperature (or substrate temperature, $T_s$) were observed in spite of nanorod-doped RE123 films obtained using a target with a certain ratio of the nanorod-material addition to RE123. When one applies a concept of the above

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experiments performed by the excimer-laser PLD to the films fabricated by the Nd:YAG-laser PLD, optimization of growth conditions and understanding of \( J_c \) and IL depending on the deposition conditions are important even in "nanorod-free" RE123 films as a fundamental research for quantitative evaluation of the addition of nanorods into the RE123 films. In the present study, we attempted to fabricate pure Y123 and Er123 films at various \( T_s \) by the low-cost PLD process using the Nd:YAG laser and clarified the dependences on \( T_s \) of their \( J_c \), ILs and crystallinities.

2. Experimental details

Pure Y123 and Er123 thin films were deposited on (100) single crystal substrates of SrTiO\(_3\) by PLD using a Q-switched Nd:YAG-laser with its fourth harmonics (\( \lambda = 266 \text{ nm} \)). Density of the laser energy is approximately 2 J/cm\(^2\) on the target. The distance between the target and substrate was fixed to be 25 mm. During the deposition for 30 min, the substrate temperature (\( T_s \)) was kept at 790-920°C in the oxygen pressure of 20 Pa by the flowing of O\(_2\) gas. Thickness of thus obtained films was roughly 350 nm, which was determined from cross-section by a scanning electron microscope. After deposition, the films were rapidly cooled down to 450°C, then the deposition chamber was filled with oxygen gas up to 0.2 atm. Furthermore, in-situ oxygen-annealing was performed by cooling from 450°C to 250°C with a cooling rate of 5°C/min.

The degrees of orientation for the obtained films were evaluated by the x-ray diffraction analysis. In order to measure transport properties, the films were patterned into a micro-stripline-shape with 100 \( \mu \text{m} \) in width and 1.5 mm in length. Transport properties were measured by a four-probe method under various magnetic fields. The transport current was always perpendicular to the direction of the applied magnetic field (\( B \)). To clarify the change in crystallinity for the obtained films, the reciprocal space mapping measurements were examined at (109) and (108) peaks for Y123 films with \( T_s = 850, 870 \) and 890°C.

![Fig. 1. \( T_s \) dependence of the ratio of the \( a \)-axis oriented domains to the \( c \)-axis oriented domains for the (a) Y123 and (b) Er123 films.](image)

3. Results and discussion

Figs. 1a and 1b show relationship between \( T_c \) and \( I_{500}/(I_{005}+I_{200}) \) for the Y123 and Er123 films, respectively. Note that the value of \( I_{500}/(I_{005}+I_{200}) \) represents a ratio of the \( a \)-axis-oriented domains to all the domains at which x-ray was irradiated for the \( \theta-2\theta \) scanning. We fabricated the samples in two different sequences of the deposition experiments as a function of \( T_s \), and their results were plotted in Fig. 1. In the case of Y123, remarkable co-existence of the \( a \)-axis oriented domains was appeared below 850°C. On the other hand, the co-existence was appeared below 810°C for Er123. When one focuses on a higher \( T_s \) region, it was deduced from surface morphology by a scanning electron microscope that re-evaporation on the Y123 and Er123 films was occurred above 910°C and 900°C, respectively. That is, \( T_s \) regions in which RE123 films consisting of only the \( c \)-axis oriented domains without the \( a \)-axis-oriented domains and the re-evaporation can be obtained were quite different between Y123 and Er123 and the \( T_s \) region for Er123 (~90 °C) was larger than that for Y123 (~50°C).

Fig. 2a shows \( T_s \) dependence of \( T_c \) for the Y123 and Er123 films. In both Er123 and Y123 films, \( T_s \) regions for achieving \( T_c > 85 \text{ K} \) were almost coincident with those for suppression of the co-existence of the \( a \)-axis-oriented domains. In detail, their \( T_s \) decreased with \( T_c \) for \( T_c >850\text{°C} \). This is in a contrast to the behaviour in ex-situ oxygen-annealed Er123 films grown by the excimer-laser PLD[10]. At the current stage, origin of these \( T_s \)-dependent \( T_c \) is controversial because both contributions of crystallographic and compositional effects should be taking into account as
determination factors of \( T_c \). In order to overcome this issue, clarification of the changes in \( T_c \) using the films through the ex-situ oxygen-annealing process is required.

Fig. 2b shows ILs for the Y123 films grown at various \( T_s \) from 850°C to 890°C, at which Y123 films without the \( \alpha \)-axis-oriented domains and re-evaporation could be obtained. The ILs for the Y123 films were clearly overlapped one another, and this \( T_c \)-independent behaviour was corresponding to that for the Er123 grown by the excimer-laser PLD[9]. To understand the behaviours of these ILs quantitatively, exponents, \( \alpha \), in \( H_{irr} \propto (1-T/T_c)^\alpha \) were determined from the ILs of Y123 shown in Fig. 2b. The \( \alpha \) values for all the films exhibited 1.14-1.20, and were almost constant for the change in \( T_s \). Moreover, these values were coincident well with those for the Er123 films grown by the excimer-laser PLD[9]. This means that the ILs for the Y123 films were independent of \( T_s \) and the behaviours of ILs for the pure RE123 (RE=Y and Er) films were intrinsically independent of the type of laser source for PLD.

![Fig. 2. (a) \( T_s \) dependence of \( T_c \) for the Y123 and Er123 films; (b) Irreversibility lines for the Y123 films grown at various \( T_c \).](image)

In order to evaluate another critical current property for the obtained Y123 films, magnetic field dependence of \( J_c \) for \( B//c \) was measured at 77 K and were plotted in Fig. 3a. Self-field \( J_s \) for all the obtained Y123 films deposited at \( T_s=850-890°C \) showed \( \sim 10^6 \) MA/cm\(^2\) in common, whereas difference in the \( J_c-B \) curves emerged under higher fields. In detail, the \( J_c \) at 3 T for the film with \( T_s=850°C \) was approximately three times smaller compared with those for the films with \( T_s=870°C \) and 890°C. The \( J_c \) values under self-field for the excimer-laser PLD Er123 films were reported to be almost constant and be insensitive to \( T_s \) [11], which is coincident qualitatively with our result in the Y123 films grown by the Nd:YAG-laser PLD. This strongly suggests that the Nd:YAG-laser PLD is as useful as the excimer-laser PLD as a fabrication process of RE123 films.

![Fig. 3. (a) \( J_c-B \) curves for the Y123 films deposited at \( T_s = 850, 870 \) and 890°C. (b)Line profiles of the (108) peaks, which were obtained along \( Q_x \) direction from the reciprocal space mapping, for the Y123 films with \( T_s = 850, 870 \) and 890°C.](image)
Finally, we would like to comment on the $T_s$ dependence of crystallinity for the Y123 films grown by the Nd:YAG-laser PLD. Fig. 3b shows line profiles of the (108) peaks along the $Q_x$ direction, which were obtained from the reciprocal space mappings of the Y123 films with $T_s = 850, 870$ and $890^\circ C$ as representatives of the growth at lower, middle and higher $T_s$, respectively. One can clearly see two peaks along the $Q_x$ direction for all these three films, which are derived from the $a$-axis and $b$-axis lengths of the grown Y123 films. Therefore, the observed two peaks were due to the (108) and (018), respectively. Incidentally, the peaks due to the (103) reflection of the single crystalline substrate were observed at $Q_x = 0.256$ in common for all the three films, and were independent of $T_s$. On the other hand, the positions of these peaks were systematically shifted toward the lower $Q_x$ region, indicating that the lengths of the $a$- and $b$-axes intrinsically increased with the decrease in $T_s$ in spite of the same condition of the in-situ oxygen post-annealing. Moreover, values of full widths at half maxima (FWHM) of these two peaks for $T_s = 850^\circ C$ were larger compared with those for $T_s = 870$ and $890^\circ C$, which is suggesting that crystallinity and/or homogeneity of $y$ in the Y123 films were slightly degraded for $T_s = 850^\circ C$. If the inhomogeneous distribution of $y$ is introduced into the films, underdoped regions with slightly lower-$T_c$ and relatively deteriorated infield $J_c$ will be co-existed in the films. This probably leads to serious reduction of infield $J_c$ rather than the reduction of $T_c$ and self-field $J_c$. In practice, this speculation is qualitatively consistent with the results in Fig. 3a. When one investigates intrinsic effects of the introduction of nanorods as a vortex-pinning centre for the nanorod-doped RE123 films, understanding of intrinsic changes in critical current properties as a function of $T_s$ on the nanorod-free RE123 films is required with taking the crystallographic and/or compositional changes of the matrix into account.

4. Conclusion

We attempted to fabricate the Y123 and Er123 films by the Nd:YAG-laser PLD at various substrate temperatures for clarifying the $T_s$ dependences of $T_c$, $J_c$ and crystallinities. The simultaneous accomplishment of high $T_c$ and $c$-axis orientation without the $a$-axis-oriented domain was realized in the $T_s$ region from $850^\circ C$ to $910^\circ C$ for both the Y123 and Er123. Although infield $J_c$ for $B/\!c$ was deteriorated under 5 T in the Y123 film with $T_s = 850^\circ C$, self-field $J_c$, IL were independent of the change in $T_s$. From the reciprocal space mapping measurement at the (108) peaks revealed that the $a$-axis and $b$-axis lengths of the grown Y123 films systematically decreased with the increase in $T_s$, and the reduction of crystallinity was suggested for the film with $T_s = 850^\circ C$. To understand intrinsic vortex-pinning effects of nanorods in the nanorod-doped RE123 films, the present study using the “pure” films gives us fundamental and useful information on the $T_s$-dependent behaviours of films without the doping of nanorods.

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