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# Improvement of Surface Morphology of $Bi_2Sr_2CaCu_2O_x$ Thin Films by using Nano-structured $SrTiO_3$ Thin Films

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**Abstract**. Surface morphology of $Bi_2Sr_2CaCu_2O_x$ (BSCCO) thin films prepared by metal-organic decomposition (MOD) method is successfully improved by using nano-structured $SrTiO_3$ (STO) buffer layer prepared by MOD method. The nano-structured STO thin films were obtained by 2-step thermal treatment at 870ºC for 2 hours followed by high temperature of 1000ºC in $O_2$ atmosphere for 1 hour. In the case of BSCCO thin films on conventional STO substrates, many islands consist of additional phases and large-size steps were observed. In contrast, segregations are extremely suppressed, and terrace structures with a-b plane of BSCCO were clearly observed. The BSCCO thin films on the nano-structured STO showed a critical temperature of 80 K. This result indicates that a control of nucleation is one of the most important factors to obtained high quality thin films in the case of hetero-epitaxy.

## 1. Introduction

Superconducting devices using high-Tc superconductors have not been fully realized even after a period of 15 years. One of the most serious problems is the thin film preparation technology, although various kinds of techniques such as a molecular beam epitaxy method, an metal-organic chemical vapor epitaxy method, a laser ablation method, a sputtering method, etc. have been developed. Since oxide high-Tc superconducting materials have a complicated structure which contains more than three or four elements, it is hard to obtain high quality thin films without a twin structure, segregations of additional phases, defects, etc. In addition, those materials should be prepared on a different kind of single crystal substrates, so-called “hetero-epitaxy”, which make also difficult to obtain single high quality thin films.

Recently, a metal-organic decomposition method (MOD) which is one of the simplest techniques to prepare oxide thin films is attracting attentions in a field of the high-Tc superconductors, since it allows us to prepare large-size thin films as large as a maximum size of available substrates and to get higher critical current density of > $10^7$ A/cm$^2$. However, problems mentioned above still exist. In this work, to solve those problems, we tried to control the crystal growth by not varying the growth conditions such as annealing temperature, atmosphere, etc., but by utilizing nano-structures fabricated on the substrate. The nano-structure having numerous steps, which act as kinks, helps nucleation of high-Tc superconducting materials without precipitations of additional phases. In this paper, we report on $Bi_2Sr_2CaCu_2O_x$ (BSCCO) thin films prepared by the MOD method on nano-structured $SrTiO_3$ (STO) by the MOD method.

## 2. MOD process

BSCCO thin films were prepared by using a metal-organic solution (SKBSCCO-008, Kojundo Chemical Lab. - Symetrix) [1] and STO by (STO008, Kojundo Chemical Lab.). The MO solution for BSCCO was spin coated on STO (001) substrates or STO thin films by 2-step process using 500 rpm for 5 s and 3000 rpm for 1 min. After the drying process on a hot plate at 150ºC for 2 min, the films were annealed in $O_2$ at 870ºC for 2 h and in $O_2$ + $N_2$ (10:90) atmosphere at 885ºC for 2 h. Thickness of the films was approximately 50 nm. STO films were prepared on STO (001) substrates by almost same process, the spin-coating, the drying at 150ºC and the thermal treatment at 870ºC for 2 h. To obtain nano-structure, the STO thin films were annealed at high temperature of 1000ºC in $O_2$ atmosphere for 1 h.

## 3. Nano-structured STO films

Figure 1 shows scanning electron microscope (SEM) images of STO thin films. Figure 1(a) is a sample annealed at 870ºC and Fig.1 (b) is that treated at 1000ºC after annealing at 870ºC. In Fig.1(a), a glandular structure with a grain size of ~ 100nm is observed, and it is found that each grain have a round shape. On the other hand, by annealing at 1000ºC the round-shaped grains became rectangular-shaped faceted structure as shown in Fig.1 (b). We expect that this faceted structure acts as kinks. Another choice of making kinks is utilizing off-axis substrates. In fact, untwined BSCCO thin films were successfully obtained as a result of a control of the nucleation at step edges [2-4], and it is also effective to suppress segregations during the thin film growth.

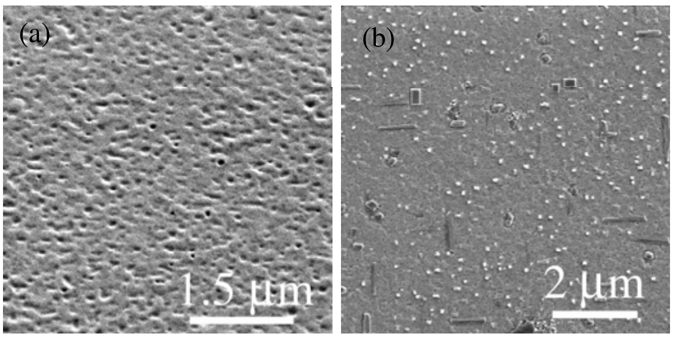


Fig.1 SEM images of STO thin films prepared at (a) 870ºC and (b) 870º + 1000ºC.

## 4. BSCCO Thin Films on the Nano-structured STO Thin Films

Figure 2(a) shows a BSCCO thin film prepared on a commercial STO substrate. The growth condition was optimized as reported in Ref.1 and therefore this film shows a good electrical properties with a critical temperature of 84 K and a critical current density of ~ $10^6$ A/cm$^2$ at 15 K. However many islands consist of additional phases and large steps are observed in this film. These three-dimensional structures cause defects in superconducting device structures. In contrast, in Fig.2 (b), terrace structures with facets of BSCCO are clearly observed and segregations are extremely suppressed as we expected. We consider that this improvement of morphology is owing to an introduction of nucleation center, nano-structured STO layer. Therefore, we believe that a control of nucleation is one of the most important factors to obtained high quality thin films in the case of hetero-epitaxy.

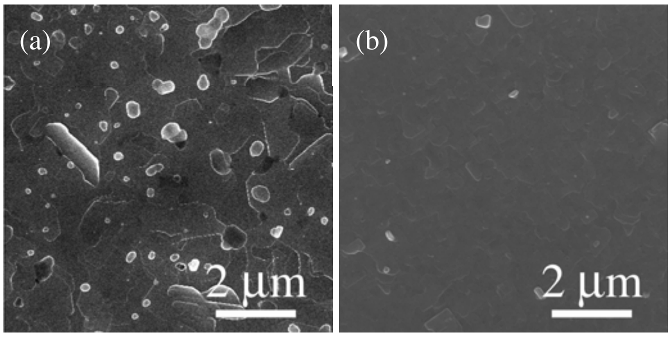


Fig.2 SEM images of BSCCO thin films prepared on (a) the STO substrate and (b) the nano-structured STO thin film.

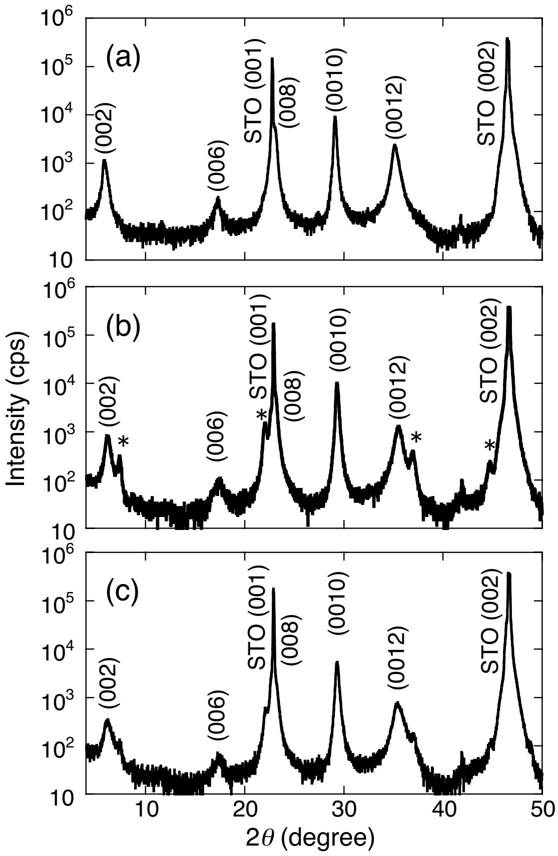


Fig.3 XRD patterns of BSCCO thin films grown on (a) the STO substrate and (b)-(c) the nano-structured STO buffer layer. (a) and (b) were annealed for 2h at 870ºC in $O_2$ and at 885ºC in $O_2$ + $N_2$ (10:90) and (c) was annealed for 4h.

Figure 3 shows X-ray diffraction patterns of BSCCO thin films grown (a) directly on the STO substrate and (b)-(c) on the nano-structured STO buffer layer. BSCCO thin films shown in Fig.3 (a) and Fig.3 (b) were prepared by the same condition, 870ºC in $O_2$ for 2 h and at 885ºC in $O_2$ + $N_2$ (10:90) atmosphere for 2 h, optimized for the BSCCO on the STO substrate. In this case, 2201 phase appears only in the BSCCO thin film on the nano-structured STO buffer layer as depicted by asterisks in Fig.3 (b), while a single phase 2212 is obtained on the STO substrate as shown in Fig.3 (a). This fact suggests that 2201 nuclei also grow while the nucleation of 2212 phase is strongly enhanced by the nano-structured STO. Considering that 2212 phase is the most preferable in the annealing temperature used here and with the stoichiometric chemical composition of the MOD liquid, the 2201 phase formed on the nano-structured STO should become 2212 phase by a longer annealing. In fact, the 2201 phase can be suppressed by a longer annealing at 870ºC in $O_2$ for 4 h and at 885ºC in $O_2$ + $N_2$ (10:90) atmosphere for 4 h as shown in Fig.3 (c).

Finally, R-T curves of the samples are shown in Fig.4. These samples are the same samples as shown in Fig.3. The BSCCO thin film on the nano-structured STO showed semiconductor-like behavior above 100K and has a critical temperature (Tc) of 70 K although the BSCCO thin film prepared directly on the STO substrate is 84 K. This degradation of the Tc is consistent with the result of XRD pattern showing the existence of 2201 phase. On the other hand, Tc was improved by the longer annealing up to 80 K as shown in Fig.4 (c). This improvement is also consistent with the XRD analysis showing suppression of 2201 phase.

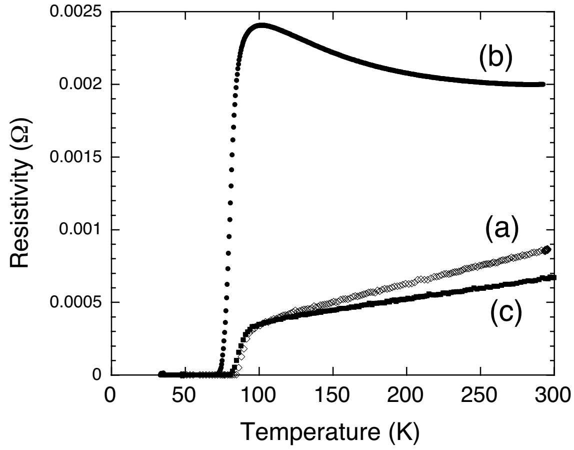


Fig.4 R-T curves of BSCCO thin films grown on (a) the STO substrate and (b)-(c) the nano-structured STO buffer layer. (a) and (b) were annealed for 2h at 870ºC in $O_2$ and at 885ºC in $O_2$ + $N_2$ (10:90) and (c) was annealed for 4h.

## 5. Summary

We have succeeded to prepare nano-structured STO and BSCCO thin films by the MOD method. The morphology of BSCCO thin films was successfully improved by using nano-structured STO thin films. This result indicates that nano-structured substrates are more effective to prevent segregations and obtain flat surfaces than atomically flat substrates.

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