Synthesis of superconducting (Cu, C)-Ba-O films on SrCuO$_2$ buffer by pulsed laser deposition

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Abstract. (Cu, C)-1201 films have been grown on SrCuO$_2$ buffer layer over SrTiO$_3$ (100) by pulse laser deposition. Effects of the insertion of SrCuO$_2$ buffer on growth mode of the 1201 phase has been investigated by comparing in-situ RHEED pattern of the films growth on SrCuO$_2$/SrTiO$_3$, SrTiO$_3$ and NdGaO$_3$. (Cu, C)-1201 films directly grown on SrTiO$_3$ and NdGaO$_3$ exhibit diffusive streak patterns in the beginning of the film growth. On the other hand, the films on the buffer show sharp streaks in the beginning from the film growth on buffer. a-axis parameters of the films on SrTiO$_3$ and NdGaO$_3$ estimated by RHEED patterns drastically expanded from 3.90 ± 0.06 Å and 3.93 ± 0.10 Å at 10 nm thick to 4.07 ± 0.05 Å and 3.99 ± 0.03 Å at 40 nm thick, respectively. The films on 10 nm thick buffer gradually expanded from 3.95 ± 0.05 Å at 10 nm thick to 4.01 ± 0.05 Å at 40 nm thick. The films on 10 nm and 20 nm thick buffer layers showed the rises of superconducting onset temperature $T_{c-onset}$ to 72 K and 62 K, respectively, whereas $T_{c-onset}$ of the films directly grown on SrTiO$_3$ was about 50 K. Due to the growth mode without misfit dislocations in the beginning of the film growth, the SrCuO$_2$ buffer could strain the 1201 phase. These results indicated that the interfacial strain should be important for (Cu, C)-1201 films, and superlattice based on it.

1. Introduction

Multilayer cuprate (Cu, C)Ba$_2$Ca$_{n-1}$Cu$_n$O$_x$, [(Cu, C)-12(n-1)n] is a series of the attractive high temperature superconductor (HTS). For the Cu- and (Cu, C)-system with n > 3, presence of two or more crystallographically inequivalent CuO$_2$ planes in a unit cell HTS causes novel properties, including a coexistence of superconductivity and antiferromagnetism within a unit cell [1], maintaining high superconducting critical temperature $T_c$ even in the heavily over-doped region for (Cu, C)-1223 [2] and potential of multi-component superconductivity [3]. Most of the sample of Cu- and (Cu, C)-systems were, so far, prepared by means of sintering under high pressures and high temperatures. Epitaxial growth of thin films by several kinds of techniques enables to stabilize the infinite-layer-related structure. Furthermore, artificial superlattice technique demonstrated by Balestrino et al. has opened a new field of research in ultra-thin or the large n phases [4-6]. For sputtered films, superconducting Ba-Ca-Cu-C-O phase with the structure related to the (Cu, C)-systems containing Ba oxycarbonate compound as charge reservoir has been obtained [7, 8]. In most cases, control of CO$_2$ gas pressure during the film growth is a key parameters for structural stabilization as well as development of superconducting.
In our previous works, we attempted to fabricate (Cu, C)-1201 films by pulse laser deposition (PLD) with a precise control of CO₂ introduction, and that superconducting 1201 films were successfully obtained at noteworthy low temperature of 500 °C ~ 600 °C [9]. (Cu, C)₃Ba₂Ca₂Cu₃Oₓ₋δ [(Cu, C)-1201] should be one of the suitable starting materials to explore the multi-layer (Cu, C)-12(n-1)n by means of the artificial superlattice method.

In spite of these attractive points, the surface smoothness of (Cu, C)-1201 films so far directly grown on SrTiO₃ were rather rough. The grain growth of those films should originate in the lattice mismatch between the films and substrate. From this point of view, the insertion of SrCuO₂ with infinite layer structure as a buffer layer between the film and substrate should be useful for suppressing interfacial defects, since the a-axis parameter 3.926 Å [10] of SrCuO₂ is the intermediate length between 3.905 Å of SrTiO₃ and 3.959 Å of (Cu, C)-1201 reported in Ref. [11].

In the present study, we have investigated the effect of the lattice mismatch on growth mode and transport properties of the (Cu, C)-1201 films by growing them on SrCuO₂ buffer / SrTiO₃, and directly on SrTiO₃ and NdGaO₃. Their crystal structures were compared by means of in-situ reflection high energy electron diffraction (RHEED). The obtained results revealed that the relatively large lattice mismatch between the 1201 films and SrTiO₃ and NdGaO₃ substrates resulted in a poor crystallinity of the films in the beginning of the film growth, though the mismatch was quickly relaxed in the subsequent stage of the growth. On the other hand, the reduced mismatch by the insertion of the SrCuO₂ buffer resulted in a good crystallinity even for the each stage of the film growth. Interfacial strain between the 1201 film and SrCuO₂ buffer was gradually relaxed with the film growth. The strained thin layer of (Cu, C)-1201 exhibited Tc onset up to 70 K, which is higher than those directly grown on SrTiO₃ and NdGaO₃. It suggests that the effect of gradual strain in the interface should be useful for the emergence of higher superconductivity for (Cu, C)-1201.

2. Experimental
The buffers of infinite-layer SrCuO₂ which has a similar crystal structure to the (Cu, C)-1201 and intermediate a-axis lattice parameter of 3.926 Å between SrTiO₃ and the 1201 phase were grown on (100) SrTiO₃ at growth temperature of 500 °C by PLD. The (Cu, C)-1201 films were grown on (100) SrTiO₃, (001) NdGaO₃ substrates or SrCuO₂ buffer at temperature of 500 °C ~ 600 °C by PLD under the following conditions: Ar+O₂+CO₂ mixture as atmosphere gas, total pressure of Ar+O₂+CO₂ was 80 mTorr, O₂ partial pressure was fixed at 5 mTorr, CO₂ partial pressure in the range of 0.10 ~ 0.25 mTorr, and laser power and its repetition frequency of 100 mJ/pulse and 1.5 Hz, respectively. The BaCu₀.₇₅Oₓ pelat was used as the target.

Crystal structure of the films was characterized by in-situ reflection high energy electron diffraction (RHEED), and θ-2θ scan of X-ray diffractometry (XRD) with Cu Kα radiation. A surface morphology of the films was examined by in-situ RHEED and atomic force microscopy (AFM). After the deposition of the film, amorphous CaCuO₂ passivation layer was deposited on the films in-situ to avoid chemical degradation. Transport properties were measured by the standard four-probe method immediately after taking out the specimens.

3. Results and discussion
Figure 1 shows RHEED patterns of (Cu, C)-1201 films on SrTiO₃, NdGaO₃ and SrCuO₂ / SrTiO₃ taken along [100] azimuth direction. 10 nm thick films directly grown on SrTiO₃ and NdGaO₃ exhibited rather broad streaks, as shown Figure 1 (a). On the other hand, as shown Figure 1 (c) 10 nm thick (Cu, C)-1201 film on 10 nm thick SrCuO₂ buffer showed sharper streaks. These indicate that the insertion of the buffer improves crystallinity of the (Cu, C)-1201 film in the beginning of the growth. Figure 1 (b) and (d) show RHEED patterns of 40 nm thick films directly on the substrate and on the buffer. The streaks of the films directly grown on the substrates got sharper. This gradual improvement of crystallinity is frequently observed in films grown on the mismatched substrate. In contrast with 10 nm thick specimen, the 40 nm thick (Cu, C)-1201 on the buffer showed broadened streaks, which was due to an increase of surface roughness as a function of thickness.
As shown Figure 2, a-axis lattice parameters of 10 nm thick films directly on SrTiO$_3$ and NdGaO$_3$ were 3.90 ± 0.06 Å and 3.93 ± 0.10 Å, respectively. In the beginning of film growth, the a-axis lattice parameters of the film were close to that of substrates. Increase of the thickness of these films to 40 nm resulted in expansion of a-axis lattice parameters: 4.07 ± 0.05 Å and 3.99 ± 0.03 Å on SrTiO$_3$ and NdGaO$_3$, respectively. The change of a-axis lattice parameters occurred at film thickness of 20 nm accompanied with sharpening the streak pattern. The (Cu, C)-1201 films with 10 nm thickness on the buffer layer show a-axis lattice parameter of 3.95 ± 0.05 Å. With increasing of the film thickness to 40 nm, a-axis lattice parameter gradually expand to 4.01 ± 0.05 Å which is small compared with that of the 40 nm thick films directly grown on substrates.

The 40 nm and 80 nm thick (Cu, C)-1201 film directly grown on SrTiO$_3$ have the superconducting onset temperature 20 K and 50 K, respectively. Superconducting transition of those films on NdGaO$_3$...
was not observed. (Cu, C)-1201 films on the 10 nm and 20 nm thick buffer layers had the superconducting onset temperature 72 K and 62 K, respectively, as shown in Figure 3. Figure 4 shows the XRD pattern of 80 nm thick (Cu, C)-1201 film on 20 nm thick SrCuO$_2$ buffer with $T_c$ onset $\sim$ 62 K. The film exhibited c-axis preferred orientation and c-axis lattice parameter of 8.18 Å which coincided to that of (Cu, C)-1201 phase.

The abrupt expansion of lattice parameter accompanied with sharpening of RHEED streaks of the 20nm thick (Cu, C)-1201 film directly grown on SrTiO$_3$ indicates that the mismatch is relaxed around this thickness. On the other hand, the gradual increase a-axis parameter of the film growth on SrCuO$_2$ buffer revealed that interfacial strain existed in them. Considering that 10 nm thick films on the buffer showed the sharp RHEED streaks, in the thin region of these films adjacent to the buffer good crystallinity and compressive stress which depends on thickness of the buffer should coexist. Since the (Cu, C)-1201 phase is hole doped HTS, and the (Cu, C)-1201 films fabricated so far were in the underdoped state [12], a contraction of a-axis of this phase should result in a rise of hole concentration as well as $T_c$. These results reveal that the insertion of infinite layer SrCuO$_2$ buffer with smaller lattice mismatch is useful to improve crystallinity of very thin layer of (Cu, C)-1201 and to introduce interfacial strain beneficial to superconducting properties.

4. Conclusion

(Cu, C)-1201 films were grown on SrTiO$_3$ and NdGaO$_3$ and SrCuO$_2$ / SrTiO$_3$ by PLD. The obtained results revealed that effect of the insertion of SrCuO$_2$ buffer on the growth mode occur good crystallinity of the films resulted in gradually expanding lattice parameter without misfit dislocation and its gradual compressive strain should result in a rise of hole concentration as well as $T_c$. Insertion of buffer is one of useful ways for obtaining good crystallinity and smooth surface which are requirement of artificial superlattice method. The effect of the compressive strain should influence transport properties and higher superconducting transition temperature for ultra thin film.

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