- Polar Coincident Site Lattice Figure - Multiple Domain Growth of Bismuth Cuprate Superconducting Film

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Bismuth cuprate superconducting film has a large-scale periodicity, supercell, other than an ordinal lattice cell. Although the supercell lies along b axis in bulk material, the structure lies along both a and b axes on thin film and shows asymmetric structure in reciprocal space along c axis. The asymmetric structure was described with triangle function, which modulates the structure to form the supercell. In the terms of the double domain along a and b axes were observed on three dimension X-ray reciprocal space mapping. Bismuth oxide film ($\text{Bi}_2\text{Sr}_2\text{Ca}_1\text{Cu}_2\text{O}_x$) grew with the relation of $\text{Bi-2212}[001]$ parallel to $\text{MgO}[110]$ (45° rotation growth), but the bismuth cuprate superconducting film often grows with lateral rotation from 45° growth. To describe such a relation, coincidence site lattice was visually expressed on polar coordinate system.

Key words: high T_c superconductor, thin film, supercell, reciprocal space mapping

1. INTRODUCTION

High T_c superconductor, bismuth cuprate oxides, is well known for structural modulation, supercell. The supercell is incommensurate modulation consisting of several unit cells along b axis with periods of 4.7 unit cells [1-4]. The supercell (SC) of $\text{Bi}_2\text{Sr}_2\text{Ca}_1\text{Cu}_2\text{O}_x$ (Bi-2212) is expressed as $\text{SC} = 4.7\mathbf{b} + 1.0\mathbf{c}$, where $\mathbf{b}$ and $\mathbf{c}$ are unit cell vectors along b and c axes. The modulation is incommensurate along b axis but is commensurate along c axis in bulk samples previously reported.

While bulk samples show symmetric structure of SC by high-resolution electron microscopy, electron diffraction, x-ray diffraction, and neutron diffraction [5-9], thin films show asymmetric peaks on X-ray Reciprocal Space Mapping (XRSM) [10]. The symmetric peaks observed on bulk samples can be generated by modulation vector as a sinus function modulating unit cells through its crystal, and the asymmetric peaks are described by triangle function [11]. Another XRSM shows the SC modulations exist along both a and b axes. An epitaxial Bi-2212 film deposited on $\text{MgO}(001)$ substrate grows with relation of $\text{Bi-2212}[100]$ parallel to $\text{MgO}[110]$ (45° rotation), indicating Bi-2212 grows epitaxially on MgO substrate in term of unit cells. However, from the view of long periodicity the SC structure has two domains along a and b axes.

Aliovalent substitution and extra oxygen had been

Fig.1 Schematic of X-ray Reciprocal Space Mapping (XRSM). X-ray irradiates a sample with an incident beam and is diffracted to the left. An ordinal cross sectional XRSM (black plane) and a plan view XRSM (top gray plane).

carried out to investigate the origin of SC structure [12-14]. The aliovalent substitution replaces cations with different radius, and the oxygen doping changes oxygen content by annealing or by high-pressure oxidization. The aliovalent substitution of cations changes also oxygen content by replacing divalent strontium with trivalent lanthanum, for instance, so that the aliovalent substitution might affect oxygen content as well as inducing strain into crystal. To emphasize the effect of strain, multilayered and thick films were prepared and investigated in detail [15,16]. In multilayered structure the strain must be prominent, to the contrary, the strain must be released in thick film. Interestingly in the thick

film other domains grow on MgO(001) substrates with $\sim 30^\circ$ lateral rotation relative to initial 45° rotation [16].

In this study, 45° rotation growth was addressed by the coincidence site lattice [17-19] expanded on polar coordinate system for visualization, and to verify double domain growth in terms of SC structure plan view XRSM was taken on the plane vertically crossing ordinal cross sectional XRSM around Bi-2212(0020) peak. By continuously taking slices of plan view XRSM, three dimensional XRSM was also presented.

2. EXPERIMENTAL

Bi-2212 thin films were prepared by pulsed laser deposition using “slower Q-switched Nd:YAG laser” [20] at the repetition rate of 2 Hz generated by 10 Hz modulated flash lamp with secondary function generation. Targets were prepared by stoichiometric ratio of Bi_2O_3 , SrCO_3 , CaCO_3 and CuO powders, which were ground and calcined for total four hours in oxygen atmosphere at 800°C . The mixture was pressed as pellets at the pressure of 400 kgf/cm^2 , and sintered again in oxygen atmosphere at 800°C .

Bi-2212 films were deposited on MgO(100) substrates at a temperature of 750°C with a substrate-target distance of 40 mm. A thick film $\sim 5,000 \text{ \AA}$ was also prepared in the same conditions. Epitaxial growth was verified by mainly x-ray diffraction (XRD) such as θ - 2θ and ϕ scan, and deposition rate was estimated using film thicknesses obtained by x-ray reflection (XRR). X-ray reciprocal space mapping (XRSM) was taken on a cross section ($\mathbf{b}^* \times \mathbf{c}^*$ plane) and a plan view ($\mathbf{a}^* \times \mathbf{b}^*$ plane) to observe SC peaks generated by SC structure, as shown in Fig.1. The series of plan view XRSMs were taken to visualize SC structure in three dimensions. Resistivity was measured by the Van der Pauw method with silver electrodes, and showed the transition temperature ($R=0$) of 70 K.

3. RESULTS AND DISCUSSION

XRD θ - 2θ and ϕ scan verified an epitaxial growth of Bi-2212 film with the relation of Bi-2212[001] parallel to MgO[001] and Bi-2212[100] parallel to MgO[110] (45° rotation), and XRR showed the film thickness of $\sim 1,000 \text{ \AA}$. The lattice mismatch of Bi-2212[100] is about 25 % to MgO[100] (cubic on cubic) and 5 % to MgO[110] (45° rotation), respectively. The thick film with thickness of $\sim 5,000 \text{ \AA}$ showed lateral rotation other than 45° rotation growth commonly obtained on MgO(001) substrate.

A cross sectional XRSM was taken around Bi-2212(2022) including a substrate peak of MgO(113) so that the lattice constants of Bi-2212 can be verified by those of MgO substrate. The two peaks lied on MgO[110] $\times$ MgO[001] plane, which is equivalent to $\mathbf{b}^* \times \mathbf{c}^*$ plane of Bi-2212 film. The cross sectional XRSM observes a plane of reciprocal lattice by using $\omega \times (\omega - 2\theta)$ axes, so that reciprocal lattice units (rlu), Qx and Qy can be expressed as,

$$Q_x = \frac{2 \sin \theta}{\lambda} \sin(\omega - \theta) ; \quad Q_y = \frac{2 \sin \theta}{\lambda} \cos(\omega - \theta),$$

where λ is the wave length of incident X-ray ($\text{CuK}\alpha_1 = 1.5406 \text{ \AA}$). Since these are rectangular coordinates, the lattice constants can easily be estimated on Qx-Qy

Fig.2 XRSM on MgO[110] $\times$ MgO[001] plane, which is equivalent to the $\mathbf{b}^* \times \mathbf{c}^*$ plane of the Bi-2212 film. Bi-2212(2022) peak was observed with the substrate of MgO(113) peak.

Fig.3 Polar CSL figure of Bi-2212 film grown on MgO(001) substrate (a) within domain size of 10 Å, and (b) within 20 Å. 45° rotation growth is dominant.

coordinate. The Bi-2212 thin film on a MgO(100) substrate showed the lattice constants, $b \sim 5.43 \text{ \AA}$, and $c \sim 30.88 \text{ \AA}$, which were estimated by XRSM observed around Bi-2212(2022) peak as shown in Fig.2.

Lattice mismatch is often mentioned for epitaxial growth, where the lattice coherent strain [21] e_ℓ is expressed as

$$e_\ell = 2 \frac{a_{\text{Bi}} - a_{\text{MgO}}}{a_{\text{Bi}} + a_{\text{MgO}}},$$

where a_{Bi} and a_{MgO} are lattice constants of Bi-2212 and MgO substrate, respectively. For an epitaxial growth the lattice coherent must be reasonably small. However, an epitaxial growth exists even with a large mismatch [22]. The lattice coherent strain is considered to be combinations of one each unit cells between a film and substrate. Epitaxial growth with a large lattice coherent strain is often described by domain epitaxial growth consisting of $(m \times n)$ unit cells of film growing on $(k \times \ell)$ ones of a substrate, and two lattices have a coincident site lattice (CSL) between $(k \times \ell)$ and $(m \times n)$ unit cells [17]. Domain coherent strain is often shown with only some combinations of k , ℓ , m and n in table.

In order to visualize domain coherent strain, a polar CSL has been proposed with all the combinations of $(k \times \ell)$ and $(m \times n)$ [22]. Domain coherent strain e_d is defined as

Fig.4 Cross sectional XRSM on ($\mathbf{b}^* \times \mathbf{c}^*$) plane of Bi-2212(0020) peak. Asymmetric intensity distribution of SC satellite peaks are observed around the main peak of Bi-2212(0020).

$$e_d = 2 \frac{\sqrt{m^2 + n^2} a_{\text{film}} - \sqrt{k^2 + \ell^2} a_{\text{sub}}}{\sqrt{m^2 + n^2} a_{\text{film}} + \sqrt{k^2 + \ell^2} a_{\text{sub}}},$$

where a_{film} and a_{sub} are lattice constants of film and substrate, respectively. Using polar coordinate, r and θ are defined as

$$r = \frac{\sqrt{k^2 + \ell^2} a_{\text{sub}} + \sqrt{m^2 + n^2} a_{\text{film}}}{2},$$

$$\phi = \tan^{-1}(n/m) - \tan^{-1}(\ell/k),$$

where r is the domain size and ϕ is the angle of lateral rotation between two lattice cells. The polar CSL is expressed on cylindrical polar coordinates r and θ with height z as domain matching, so that the figure shows domain size as the distance from the center point and in-plane epitaxial relation as angle of ϕ .

In spite of small lattice mismatch of 5% with 45° rotation growth, Bi-2212 film often grows with other domains than 45° rotation growth. Using a polar CSL, one can bird's-view domain coherent strain as well as lattice coherent strain. Figure 3(a) is the polar CSL figures of Bi-2212 film grown on MgO(001) substrate within domain size of 10 Å, which clearly shows the dominant is 45° rotation growth. As shown in Fig.3(b) with increasing domain size, CSL peaks are scattered around the angle ϕ , which is lateral rotation angle between film and substrate. For Bi-2212 film grown on MgO(001) 45° rotation growth is dominant, though it is possible to grow other domain in term of domain growth.

The SC structure was observed on XRSM taken around Bi-2212(0020) peak on ($\mathbf{b}^* \times \mathbf{c}^*$) plane as shown in Fig.4, and the SC size can be estimate in the same way to estimate lattice constants using the distance between main and SC peak on the Qx-Qy coordinate. The SC size in Bi-2212 film with thickness of ~1,000 Å was estimated to be $\text{SCb} \sim 4.2b$ and $\text{SCc} \sim 0.9c$ along $\mathbf{b}$ and $\mathbf{c}$ axes, respectively. Since the same mapping was obtained even on the plane laterally rotated 90° ($\mathbf{a}^* \times \mathbf{c}^*$ plane), unlike bulk sample the SC structure lies along both $\mathbf{a}$ and $\mathbf{b}$ axes; double domain in terms of SC structure. To verify the double domain of SC structure, continuous slice of plan view XRSM were taken on ($\mathbf{a}^*$

Fig.5 Slices of plan view XRSMs. Continuous plan view XRSM are taken along $\mathbf{c}^*$ axis of Bi-2212 film.

Fig.6 Three dimensional XRSM. 3D XRSM image is visualized from slices of plan view XRSMs by using an image processing software Osirix. Four SC satellite peaks are clearly observed on Bi-2212 film.

$\times \mathbf{b}^*$) plane of Bi-2212 film as shown in Fig.5. An image processing software “Osirix” dedicated to DICOM (Digital Imaging and Communications in Medicine) was employed to visualize three dimensional image from slices of plan view XRSM. Three dimensional (3D) XRSM was obtained as shown in Fig.6. Four SC satellite peaks were clearly observed under the main peak of Bi-2212(0020) peak at $\sim (\pm 0.2, \pm 0.2, 19)$.

In summary, Bi-2212 films were prepared by PLD method with slower Q-switched YAG laser. XRD 0-20 and ϕ scan verified the epitaxial growth of Bi-2212 film, though lateral rotation growth is often reported on Bi-2212 film. To describe the lateral rotation growth including 45° rotation growth, a polar CSL figure was proposed. To verify the SC structure lay along $\mathbf{a}$ and $\mathbf{b}$ axes, slices of plan view XRSMs were taken around Bi-2212(0020) peak, and 3D-XRSM clearly showed the SC structure lay along both $\mathbf{a}$ and $\mathbf{b}$ axes.

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