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Fabrication and Crystal Structure of [$BiMO_3/CaBO_3$] (M=Fe, $Fe_{1-x}Mn_x$ B=Fe, Mn) Superlattices Grown by Pulsed Laser Deposition Method

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Abstract: The aim of our study is to synthesize the novel materials which shows ferromagentic (FM) and ferroelectric (FE) properties with the magnetoelectgric (ME) effect at room temperature. We expect an induced ferromagnetic ordering near the interface in the [$BiMO_3/CaBO_3$] (M=Fe, $Fe_{1-x}Mn_x$ B=Fe, Mn) superlattices. The [7 units BFMO / 7 units CFO]$_{14}$ superlattices were grown on STO(100) substrate using the PLD method. The streaky RHEED pattern demonstrated two dimensional flat surface of the superlattice. The surface morphology shows step-terraces structure. From the XRD spectrum of [BFMO/CFO] superlattice, the satellite peaks from -1 to +1, and Laue oscillations were clearly observed. The satellite peaks was fitted to substrate in-plain. The strong compressed stress was forced in BFO and BFMO layer in superlattice. The saturated magnetization of the superlattice at 300K was approximately 0.055μ_B / $Fe_{1-x}Mn_x$ atom with expected Curie temperature around 450K.

1. Introduction

The aim of our study is to synthesize the novel materials which shows ferromagentic (FM) and ferroelectric (FE) properties with the giant magnetoelectgric (ME) effect at room temperature. We expect an induced FM ordering near the interface in the [$BiMO_3/CaBO_3$] (M=Fe, $Fe_{1-x}Mn_x$ B=Fe, Mn) superlattices. The Fe^{3+} has five electrons in *3d* orbital showing the largest spin values. Since the magnetic ordering of $BiFeO_3$ is antiferromagnetic[1],[2], the induced ferromagnetic ordering is expected due to the electron transfer. When the electron transfers, we expect that the superexchange interaction between iron ions in BFO changes from antiferromagnetic to ferromagnetic according to the Kanomori-Goodenough rule. In this research, single layer growth of $LaFeO_3$(LFO), $CaFeO_3$(CFO), $CaMnO_3$(CMO), BFO and Mn-doped BFO(BFMO) was demonstrated, and [$BiMO_3/CaBO_3$] superlattices was synthesized. In the superlattice, we expect that the charge transfer through the interface induces the FM ordering around the interface.

2. Experimental

For the purpose of the study, the superlattices of BFMO and $CaBO_3$(B=Fe, Mn) were grown on Nb doped and non-doped $SrTiO_3$(STO)(100) substrates. The STO(100) substrates was ultrasonically cleaned in acetone and ethanol. The cleaned substrate was soaked in pure water for 30 min. The substrate surface was etched by a Buffered HF(BHF) (Daikin Industries, Ltd., pH=5.0) for 45 sec, immediately rinsed by pure water. The etched substrate was annealed at 920 °C for 6 h in air. In the superlattices, The [7 units BFO, or BFMO / 7 units CFO, or CMO]$_{14}$ superlattices were tried to be grown by the pulsed laser deposition (PLD) method using excimer laser of KrF 248 nm. The deposited condition was as fallows; 670ºC heater temp., 2.7~2.8 J/cm^2 energy density at the target, 4 Hz repetition rate, and 20Pa oxygen atmosphere during growth. For accurate deposition of each film thickness, the growth rate ratios of BFO, BFMO, CFO, CMO to LFO or STO were investigated in advance. From the results LFO, CFO, CMO, BFO and BFMO single layer growth, we concluded that the estimation of the deposition rate from the RHEED oscillation

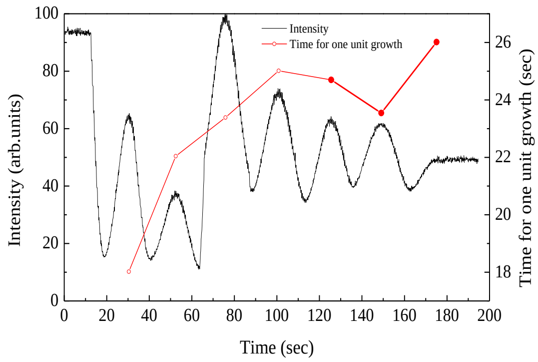

Fig.1 The RHEED oscillation of initial seven units LFO buffer layer growth.

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of LFO thin film. The best for accurate control of thickness of LFO, CFO, CMO, BFO and BFMO layers when the superlattices are prepared.

3. Results and Discussion

Fig.1 shows The RHEED oscillation and growth rate of initial seven units of LFO at repetition rate of 2Hz. At first seven LFO layers were deposited monitoring the RHEED oscillation, and then the rate was calculated form the last stable three oscillations.

Fig. 2 shows the RHEED pattern and the surface image of the [BFMO/CFO] superlattices after deposition (a) Using the STO(100) substrate (b) Using the STO(110) substrate. In the Fig.2 (a), the streaky RHEED pattern demonstrated two dimensional flat surface of the superlattice. The step-terraces structure was clearly observed, the grains height was 1.4 nm. In the Fig.2 (b), the RHEED pattern was 3D spots, the surface was rough. The step-terraces structure was observed, the surface morphology was composed of linear structure with the height of 3.0 nm grains.

In the Fig.3 XRD 2θ-θ spectrum of [BFMO/CFO] superlattice on Nb-STO(100), the satellite peaks from -1 to +1, and Laue oscillations were clearly observed around STO(002) substrate peak. The average lattice constant and [BFMO/CFO] total film thickness were calculated at 0.390nm, 76.97 nm from 0 peak and Laue oscillations. The full width at half maximum value of rocking curve was 0.0572°.

Fig.4 shows the reciprocal space mappings around STO(003), STO(103) and STO(113). The satellite peaks as well as Laue oscillation were clearly observed, and the superlattice was fitted to that of substrate in-plain. Although in-plain lattice in single layer, BFO and BFMO was not fitted, the strong compressed stress was forced in BFO and BFMO layer in superlattice. The saturated magnetization of the superlattice at 300K was approximately 0.055μ_B / $Fe_{1-x}Mn_x$ atom with expected Curie temperature around 450K.

The results of the other superlattices including an interface structure will be discussed.

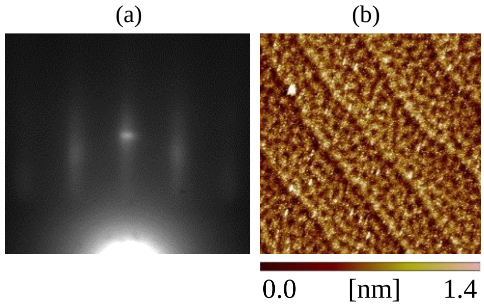

Fig.2 (a)The RHEED pattern and (b)the 5×5μm^2 SPM surface image of the [BFMO/CFO] superlattice.

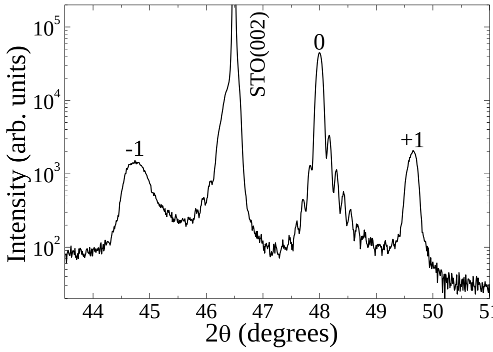

Fig.3 The XRD pattern of the [BFMO/CFO] superlattice grown on the STO(100) substrate around the STO(002) substrate peak. The Laue oscillations was clealy observed from around 0 peak. The thickness of [BFMO/CFO] total thin film thickness was calculated at 76.97nm from Laue osc.

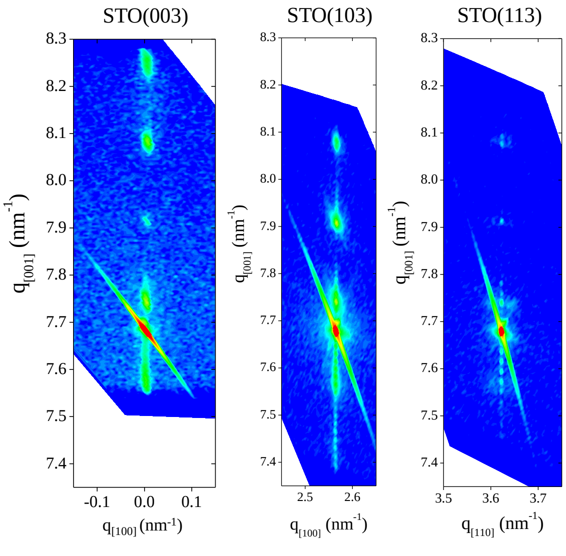

Fig.4 The Reciprocal space maps(RSMs around the STO(003), STO(103), SYO(113). The in-plane lattice parameter of the superlattice was fitted to STO substrate peak.

4. Summary

The [7 units BFMO / 7 units CFO]$_{14}$ superlattices were grown on STO(100) substrate using the PLD method. The streaky RHEED pattern demonstrated two dimensional flat surface of the superlattice. The surface morphology shows step-terraces structure. From the XRD spectrum of [BFMO/CFO] superlattice, the satellite peaks from -1 to +1, and Laue oscillations were clearly observed. The satellite peaks was fitted to substrate in-plain. The strong compressed stress was forced in BFO and BFMO layer in superlattice.

5. Reference

[1] T. Zhao : "Electrical control of antiferromagnetic domains in multiferroic $BiFeO_3$ films at room temperature", Nature Materials 5, 823-829, (2006)

[2] J. Wang : "Epitaxial $BiFeO_3$ Multiferroic Thin Film Heterostructures" , Science 299, 1719-1722, (2003)