

Low-Temperature Growth of $\text{YMnO}_3/\text{Y}_2\text{O}_3/\text{Si}$ Capacitor

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We proposed a $(0001)\text{YMnO}_3/(111)\text{Y}_2\text{O}_3/(111)\text{Si}$ structure for a metal/ferroelectric/insulator/semiconductor-field effect transistor (MFIS-FET). We successfully obtained epitaxial YMnO_3 films on epitaxial $\text{Y}_2\text{O}_3/\text{Si}$ and reported the crystallinity and the dielectric properties of the epitaxial- $\text{YMnO}_3/\text{Y}_2\text{O}_3/\text{Si}$ were superior to those of preferentially oriented $\text{YMnO}_3/\text{Y}_2\text{O}_3/\text{Si}$. Although the MFIS capacitor showed a relatively long retention time, the composition of the YMnO_3 layer is distributed along the film thickness due to the Mn evaporation that occurred by radiation heat from the substrate heater. Therefore, the density of the YMnO_3 target was improved and the substrate heating system was changed from lamp to semiconductor laser heating systems. Eventually, the compositional distribution was fixed and the optimal deposition temperature of YMnO_3 on Pt/Sapphire could be lowered from 800 to 700°C. The epitaxial $\text{YMnO}_3/\text{Y}_2\text{O}_3/\text{Si}$ deposited at 740°C with the new deposition system showed a ferroelectric capacitance-voltage(C-V) hysteresis loop with the memory window of 2.1V.

Key words: YMnO_3 , Y_2O_3 , epitaxial films, MFIS capacitor, low temperature growth

1. INTRODUCTION

Ferroelectric gate field-effect transistors (FETs)¹⁻⁴ have been investigated as potential future nonvolatile memory devices, because they have several advantages, such as high-density integration and nondestructive readable operation as compared to capacitor-type ferroelectric memories. We proposed a $(0001)\text{YMnO}_3/(111)\text{Y}_2\text{O}_3/(111)\text{Si}$ structure for the metal/ferroelectric/insulator/semiconductor (MFIS) type ferroelectric gate FETs.⁵⁻⁸ The guiding principle for obtaining long retention of the MFIS structure is to find the ferroelectric material that has small spontaneous polarization and small dielectric constant in order to effectively polarize the ferroelectric layer and decrease the depolarization field. The use of YMnO_3 for the ferroelectric gate FETs has several advantages compared to the use of other ferroelectric materials, because it has a small dielectric constant of 20 and small polarization of $5.5 \mu\text{C}/\text{cm}^2$.⁹⁻¹³ Recently, we successfully obtained epitaxial YMnO_3 films on epitaxial $\text{Y}_2\text{O}_3/\text{Si}$ and reported that the crystallinity and the dielectric properties of the epitaxially grown $\text{YMnO}_3/\text{Y}_2\text{O}_3/\text{Si}$ were superior to those of oriented $\text{YMnO}_3/\text{Y}_2\text{O}_3/\text{Si}$.¹⁴ The MFIS capacitor showed a relatively long retention time of over 10^4 sec. However, the composition of the YMnO_3 layer was not uniform along the film thickness due to Mn evaporation that occurred by radiation heat from the substrate heater. Therefore, we tried to improve the density of the YMnO_3 target and the substrate heating system of the pulsed laser deposition (PLD) system was changed from a conventional lamp heating system to one using a semiconductor laser. The wavelength of the laser for ablation was also changed from KrF ($\lambda=248$ nm, 5.0 eV) to ArF ($\lambda=193$ nm, 6.4 eV)

because the band gap energies of YMnO_3 and Y_2O_3 are about 4.3 eV and 5.6 eV, respectively.

As a result, stoichiometric YMnO_3 films on epitaxial Pt/Sapphire substrate were obtained in the wide deposition temperature range from 700 to 800°C. All the films were confirmed as having epitaxial crystallographic relationships with the epitaxial Pt/Sapphire substrate. Especially, the film deposited at 740°C had the most excellent crystallinity and very smooth surface morphology compared to the films deposited at 800°C with the previous target, PLD system and laser for ablation¹⁵.

In this study, this newly developed PLD system and the YMnO_3 target were used to fabricate $\text{YMnO}_3/\text{Y}_2\text{O}_3/\text{Si}$ MFIS capacitors.

2. EXPERIMENT

A pulsed laser deposition (PLD) system with a laser substrate heating system was used for the deposition of Y_2O_3 and YMnO_3 films. The thickness of the epitaxial Y_2O_3 films on Si varied from 20 to 45 nm. The YMnO_3 films with a fixed thickness of 100 nm were deposited on Y_2O_3 (111)/Si (111) at the substrate temperature from 660 to 740°C. The oxygen gas pressure was fixed at 5×10^{-3} Torr. An rf-magnetron sputtering deposition system was used to deposit of the Pt top electrode. The crystal structure of films was analyzed by X-ray diffraction (XRD, PHILIPS X'pert) using Cu $K\alpha$ radiation and reflection high-energy electron diffraction (RHEED). The leakage current density was measured using a pico-ampere meter (HP, 4140B). The capacitance-voltage(C-V) property was measured using an LCR meter (HP, 4284A).

3. RESULTS AND DISCUSSION

3.1 Fabrication of Y_2O_3 thin films on Si

Because the YMnO_3 has a unipolarization axis along the $[0001]$ direction,⁹⁻¹³ the crystallinity and orientation distribution along $[0001]$ should be responsible for the squareness of the P-E hysteresis loop. Therefore, the epitaxial growth of $(111)\text{Y}_2\text{O}_3/(111)\text{Si}$ is required as a substrate to obtain the excellent ferroelectric properties of epitaxial YMnO_3 . To fabricate the epitaxially grown Y_2O_3 films, the 3 mono-layer thick-initial Y_2O_3 layer was deposited at 450°C in high vacuum below 1×10^{-8} Torr to prevent the formation of surface oxidation of Si substrate. Then, the substrate temperature and the oxygen pressure were increased to 700°C and 1×10^{-6} Torr for the deposition of the main Y_2O_3 layer. Finally, the film was kept at 740°C in the oxygen pressure of 5×10^{-3} Torr, which is the deposition condition of the YMnO_3 layer.

Figure 1(a) and (b) show the XRD patterns of 2θ - θ scan and phi-scan, respectively, for the Y_2O_3 film on Si with a thickness of 45 nm. The epitaxial growth of $(111)\text{Y}_2\text{O}_3$ film was confirmed.

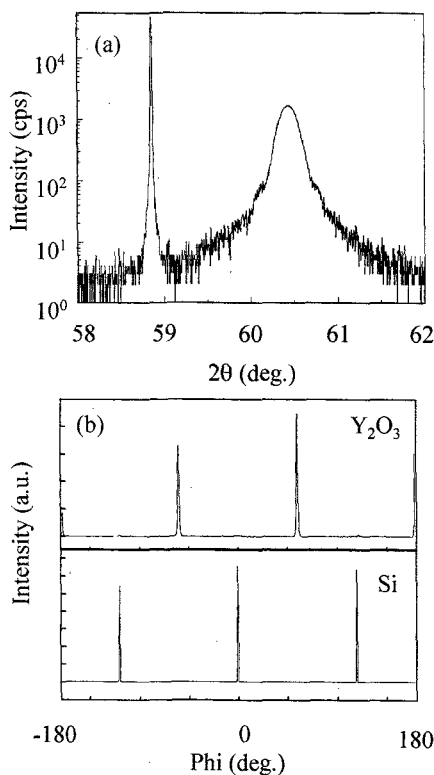


Fig. 1. (a) XRD pattern of $\text{Y}_2\text{O}_3(111)/\text{Si}(111)$, (b) phi-scan.

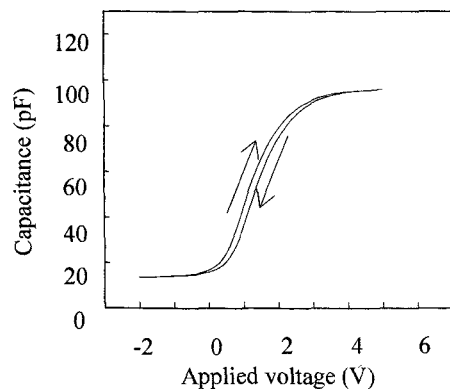


Fig. 2. C-V characteristic of the $\text{Y}_2\text{O}_3/\text{Si}$ capacitor.

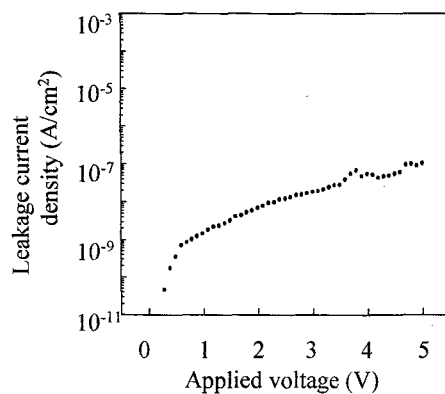


Fig. 3. I-V characteristic of the $\text{Y}_2\text{O}_3/\text{Si}$ capacitor.

Figure 2 shows C-V property of the epitaxial $\text{Y}_2\text{O}_3/\text{Si}$ capacitor. The C-V curve indicates the existence of the small charge injection, and the dielectric constant is calculated to be approximately 12.5, which is slightly lower than that of bulk Y_2O_3 ($\epsilon=15$). This indicates that the SiO_2 layer was formed at the $\text{Y}_2\text{O}_3/\text{Si}$ interface during initial layer deposition. Although the deposition of the initial layer was performed in high vacuum below 1×10^{-8} Torr, the formation of the atomic oxygen radical included in the plume might be responsible for the oxidation.

Figure 3 shows the I-V property of the epitaxial $\text{Y}_2\text{O}_3/\text{Si}$ with the thickness of 45 nm. The leakage current density at the applied voltage below 3V is recognized as low as 10^{-8} A/cm^2 . This results from the existence SiO_2 at the interface.

3.2 Fabrication $\text{YMnO}_3/\text{Y}_2\text{O}_3/\text{Si}$

As mentioned in the introduction, in a past study we deposited YMnO_3 films at 800°C when the lamp substrate heating system, KrF laser for ablation, and low density target were used for PLD deposition. The deposition conditions of epitaxial $(111)\text{Y}_2\text{O}_3$ films for previous study are reported elsewhere.¹⁴ To examine if the newly developed PLD system and the target with higher density also works to lower the substrate temperature for the MFIS structure, 100 nm-thick

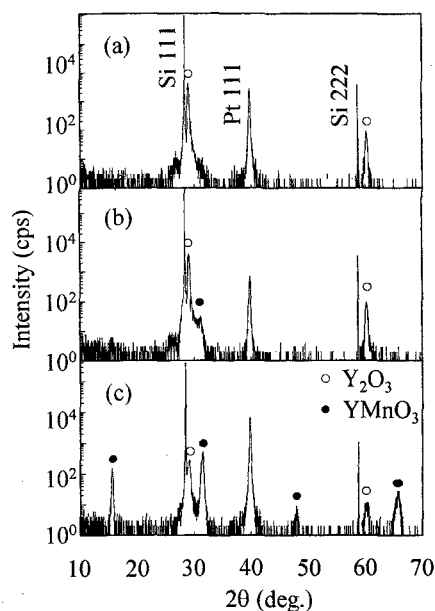


Fig. 4. XRD patterns of YMnO₃ films grown on Y₂O₃/Si substrate deposited at 660°C (a), 700°C (b), and 740°C (c).

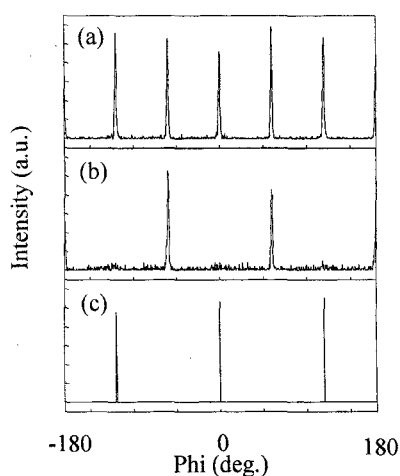


Fig. 5. XRD phi-scan of epitaxial YMnO₃/Y₂O₃/Si capacitor. (a) YMnO₃, (b) Y₂O₃, and (c) Si.

YMnO₃ films were deposited at the substrate temperature ranging from 660 to 740°C on the epitaxial Y₂O₃/Si. The thickness of the Y₂O₃ film was fixed as 20 nm, to increase the applied voltage to YMnO₃ layer. Fig. 4 shows the XRD patterns of the YMnO₃/Y₂O₃/Si capacitor deposited at various substrate temperatures. Although definite diffraction peaks from YMnO₃ were not observed at the substrate temperatures of 660 and 700°C, the clean (0001)YMnO₃ diffractions were recognized from the film on deposited at 740°C. The full width at half maximums (FWHM) of the XRD rocking curve using YMnO₃ 0004 diffraction, which shows the degree of orientation distribution along [0001] YMnO₃ films was, 1.52°. Although the value is lower than that of epitaxial YMnO₃ films (0.49°) on epitaxial Pt/Sapphire substrate¹⁵, distinct epitaxial growth was

recognized from the XRD phi-scan (Fig. 5). The epitaxial growth temperature could be lowered from 800°C to 740°C when the newly developed PLD system and the YMnO₃ target were used for deposition.

Figure 6 shows the C-V property of the epitaxial YMnO₃/Y₂O₃/Si capacitor. The ferroelectric type (counterclockwise rotation) C-V hysteresis loop with the memory window of 2.1 V is recognized. As we have reported, the result of conventional C-V measurement often includes the effect of interfacial polarization and rearrangement of the space charge.⁸ To eliminate these effects, a shorter charging time should be used for accurate evaluation of the ferroelectricity of the MFIS capacitor using C-V measurement. For this purpose, we developed pulsed C-V measurement.⁸ Fig. 6 shows the result of pulsed C-V measurement for the epitaxial YMnO₃/Y₂O₃/Si capacitor. The ferroelectric polarization switching type C-V characteristic, which is real evidence showing that ferroelectricity is obtained.

Although the epitaxial YMnO₃/Y₂O₃/Si capacitor reported in this work has several issues especially at the Y₂O₃/Si interface, the growth temperature can be lowered from 800°C to 740°C without losing ferroelectricity.

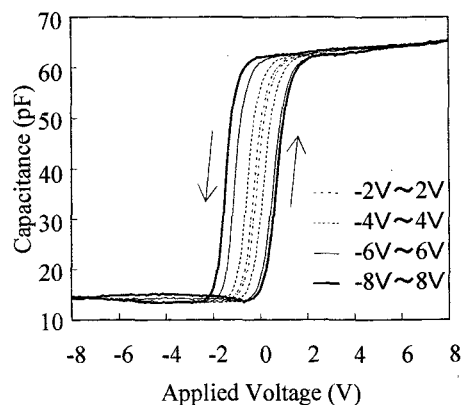


Fig. 6. C-V properties of YMnO₃/Y₂O₃/Si capacitors.

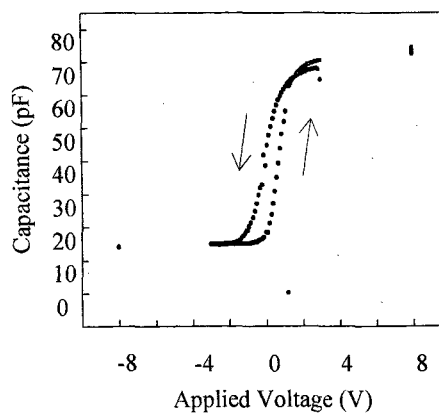


Fig. 7. Pulsed C-V property of the YMnO₃/Y₂O₃/Si capacitor.

4. CONCLUSION

To improve the stoichiometry of the $YMnO_3$ film, the $YMnO_3$ target was improved and the substrate heating system and the laser for ablation were changed.

The epitaxial $(0001)YMnO_3/(111)Y_2O_3/(111)Si$ capacitors were fabricated at $740^\circ C$, which is a lower temperature than capacitors fabricated using previous PLD system and target. A ferroelectric type C-V hysteresis loop with a memory window of 2.1 V was obtained. By evaluating pulsed C-V measurement, distinct ferroelectricity without the effect of the interfacial polarization and the rearrangement of the space charge was recognized.

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