Epitaxial insulator for bottom-gate field-effect devices based on TiO₂

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Abstract

Field-effect devices with bottom-gate structure based on anatase TiO₂ active channels were fabricated. The key factor to achieve this was the quality of epitaxial insulator layers, viz.; the insulating property and their crystallinity, because they work not only as a well insulator for field-effect devices but also as a template for the subsequent growth of epitaxial active layers. Our devices showed typical transistor actions. On-to-off current ratio exceeded 10^4 and the field effect mobility of $0.04 \text{ cm}^2/\text{Vs}$ were obtained. Interestingly, the device characteristics were found to be sensitive to ambient and light, suggesting their potential for manipulating the fruitful TiO₂ surface functions by tuning the gate voltage.

Keyword: Electric field effect, epitaxy of thin film, Titanium dioxide

1. Introduction

Recent development of oxide electronics attracts much attention from both basic and applied viewpoints due to the wide variety of their physical and chemical properties [1]. Field-effect technique with oxide active channel has been applied for the control of oxide physical properties through their carrier concentration [2-8]. This enables to control carrier concentrations cleanly and reversively. We, indeed, have already reported that field-effect transistors based on TiO_2 active channels with top-gate device structure and showed the potential for the carrier control of TiO_2 by field-effect [9]. In order to utilize fruitful surface chemical phenomena of TiO_2 [10], e.g., photocatalytic properties and photo-induced hydrophilic conversion, field-effect devices with exposed TiO_2 active surface are required, i.e., bottom-gate device structure. To achieve this, one of the key requirements is the epitaxial insulator. It is required to fulfill both insulating properties for field-effect device and high crystallinity for the template of the subsequent growth of epitaxial active layers. In this study we have reported the fabrication of field-effect devices with bottom-gate structure based on anatase TiO_2 channels and shown their potential for the tuning of surface functions by the gate voltage.

2. Experimental

Our device structure is schematically illustrated in Fig.1(a). Nb-doped (0.5 wt%) SrTiO_3 single crystals (001) (available from Shinkosha Co.) were used as substrates, which worked as a gate electrode. Epitaxial LaAlO_3 layers, which worked as an insulator,

were grown by pulsed laser deposition (PLD). A LaAlO_3 single crystal was ablated by KrF excimer laser ($\lambda = 248 \text{ nm}$, $\sim 1.5 \text{ J/cm}^2$, 5 Hz) to deposit films onto a substrate kept at 700°C . Oxygen partial pressure during LaAlO_3 growth was varied from $1 \times 10^{-5} \text{ Torr}$ to $1 \times 10^{-2} \text{ Torr}$. Subsequent to the LaAlO_3 growth, anatase TiO_2 was epitaxially grown also by PLD. Substrate temperature, repetition rate and laser fluence of KrF excimer laser, and oxygen partial pressure were 650°C , 5 Hz and $\sim 1.5 \text{ J/cm}^2$, $1 \times 10^{-3} \text{ Torr}$, respectively. After TiO_2 growth, the sample was post annealed at 400°C for 2 h in atmospheric O_2 . Finally, Al electrodes were formed by vacuum evaporation. They were patterned through stainless steel stencil masks ($W/L = 500 \text{ }\mu\text{m}/200 \text{ }\mu\text{m}$). Typical thicknesses of LaAlO_3 , TiO_2 , and Al were 500 nm, 10 nm, 20 nm, respectively. Transistor characteristics were evaluated with HP4155B semiconductor parameter analyzer at room temperature in various ambient.

3. Results and discussions

Figures 1(b)-(d) show reflection high-energy electron diffraction (RHEED) pattern, atomic force microscope (AFM) image and X-ray diffraction (XRD) patterns of LaAlO_3 epitaxial insulator grown at oxygen partial pressure of $1 \times 10^{-4} \text{ Torr}$. The streak pattern of RHEED shows that LaAlO_3 film grew epitaxially in plane, although the pattern was relatively diffuse. The XRD pattern shows c-axis oriented LaAlO_3 . The full width of half maximum of the rocking curve of $\text{LaAlO}_3(002)$ was 0.7° . RHEED and XRD results clearly shows that LaAlO_3 films grow epitaxially on $\text{Nb:SrTiO}_3(001)$, with a fairly good crystallinity of LaAlO_3 film. The AFM image of LaAlO_3 surface reveal a smooth

morphology with root mean square roughness of about 1 nm, although the surface morphology was not atomically flat. These results further support an epitaxial growth of LaAlO₃. Similar quality of the LaAlO₃ films grown at oxygen partial pressure of 1×10^{-3} Torr and 1×10^{-5} Torr has been achieved. The films grown at 1×10^{-2} Torr, however, showed a halo RHEED pattern, indicating the LaAlO₃ was amorphous.

Figure 2 shows histograms of breakdown electric field of LaAlO₃ epitaxial insulator grown at oxygen partial pressure of 1×10^{-4} Torr evaluated by Al/LaAlO₃/Nb:SrTiO₃ capacitor structure before and after post annealing at 400°C for 2 h in atmospheric O₂. The area of the Al electrode was 1.3×10^{-3} cm². For as-grown LaAlO₃, the values and the deviations of breakdown electric field were low and wide, respectively. For post annealed LaAlO₃, in contrast, the values were improved to about 2 MV/cm and the deviations became narrow. Although the insulating properties of epitaxial LaAlO₃ even after post annealing were inferior to those of amorphous LaAlO₃ [11], post annealing after LaAlO₃ growth resulted in the improvement of the device yield.

We have also fabricated thin LaAlO₃ films (50 nm thick). The surface morphology was step and terrace structure, indicating atomically flat surface, which was expected to result in fabricating well-defined insulator/semiconductor interface. Insulating properties, however, were not sufficient to operate FETs, i.e., very leaky, mainly due to the current through the grain boundaries in epitaxial LaAlO₃. Thin LaAlO₃ enhanced the quality of surface morphology, but did not meet the insulating properties required for a meaningful field effect device.

Figure 3 shows FET characteristics of TiO₂ FET with bottom-gate structure in air. LaAlO₃ epitaxial insulator was grown at oxygen partial pressure of 1×10^{-4} Torr and post annealed. Typical accumulation type FET actions were observed. Drain voltage (V_d) - drain current (I_d) curves clearly shows pinch-off points and current saturation. Gate voltage (V_g) - I_d curves shows the on-to-off current ratio exceeding 10^4 . Leakage current (I_g) was relatively high, but it was satisfactorily lower than the I_d . Field-effect mobility at saturated regime was $0.04 \text{ cm}^2/\text{Vs}$. The maximum carrier density deduced from the conductivity and the mobility was of the order of 10^{18} cm^{-3} .

FET characteristics obtained in this study was inferior to that of top-gate device structure [12]. It seemed to be resulted from two factors; the quality of TiO₂/LaAlO₃ interface and the crystallinity of TiO₂. Judging from surface morphology of epitaxial LaAlO₃ insulator, TiO₂/LaAlO₃ interface was not atomically sharp. Thus, the interface was considered to contain many defects, which can act as trapping and scattering sites of carriers. Field-effect carrier control is basically interface phenomena, thus the quality of the interface was critical. Moreover, the crystallinity of TiO₂ active layers was not so high, because the crystallinity of LaAlO₃ beneath the TiO₂ was not so high, resulting in low carrier mobility. More improvement in quality of the epitaxial insulator, surface morphology and the crystallinity is, therefore, required, which further, leads to an improved responses to the ambient and light as mentioned below.

Figure 4 shows FET characteristics under light (Xe lamp, 150 mW/cm^2) and/or oxygen ambient. As shown in Fig. 4(a), under oxygen ambient, I_d was slightly reduced

compared with that under vacuum (1×10^{-6} Torr) irrespective of light illumination. This result was expected as the carriers may be trapped by adsorbed oxygen [13]. Under light illumination, I_d was increased compared to that in dark, but still modulated by the gate voltage, indicating that field effect control was feasible under light illumination. These responses to ambient and light were reversible. The effects of light illumination almost diminished in a few tens of seconds after the turning off the light, while the effect of adsorbed oxygen remained for a few hours after evacuation. Figure 4 (b) shows I_d response to alternating oxygen pressure under light illumination. Oxygen pressure was repeatedly changed from 1×10^{-6} Torr to 760 Torr and vice versa. I_d definitely responded to oxygen pressure. These results indicate the device potential for gas or light sensor application, where the dynamic range of the sensitivity can be controlled by gate voltage.

4. Conclusion

In summary, we have succeeded in fabricating the field-effect transistors based on TiO_2 active channels with bottom-gate structure. One of the key points to achieve this is the growth of high-quality epitaxial insulator, which fulfills both insulating properties and high crystallinity, required for FET. Fairly good epitaxial LaAlO_3 was obtained. Our bottom-gate devices showed typical transistor actions and clear responses to light and/or ambient, suggesting the sensor application with sensitivity tuned by the gate voltage.

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Figure captions

- Fig. 1 (a) Schematic illustration of the device structure. (b) RHEED image, (c) AFM image ($1 \times 1 \mu\text{m}^2$), and (d) XRD patterns of epitaxial LaAlO_3 film. The inset of (d) is a rocking curve of $\text{LaAlO}_3(002)$ diffraction.
- Fig. 2 Histograms of breakdown electric field of epitaxial LaAlO_3 film (a) before and (b) after post annealing.
- Fig. 3 Transistor characteristics of bottom-gate anatase FET.
- Fig. 4 (a) V_g - I_d curves of anatase TiO_2 FET under various conditions. Open and filled markers denote under Xe light illumination and dark condition, respectively. Triangle and circle markers denote ambient condition; 1×10^{-6} Torr and 760 Torr O_2 ambient, respectively. (b) Drain current responses to oxygen pressure under Xe lamp illumination.

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