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Thermal decomposition of $LaAlO_3/SiO_2/Si$ gate stack structures studied by synchrotron radiation photoemission spectroscopy

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Introduction

Recently high dielectric constant (high-*k*) materials such as Al_2O_3, ZrO_2, HfO_2, rare-earth oxides, *etc*. have been intensively studied as alternatives to SiO_2 which has long been used as gate dielectrics in ultra-large scale integration (ULSI) metal-oxide-semiconductor field-effect transistors (MOSFETs) [1].

Among them, $LaAlO_3$ is regarded as a promising candidate for high dielectric constant (high-*k*) gate dielectrics dielectrics in metal-oxide-semiconductor field-effect transistors (MOSFETs) [2].

Although thermal stability of $LaAlO_3$ under various conditions has been reported [3], the behavior of $LaAlO_3$ thin films on Si during ultrahigh vacuum (UHV) annealing has not been clarified. Thus, we have studied the changes in chemical states of $LaAlO_3$/Si interface after UHV annealing.

Experimental

$LaAlO_3$ thin films were prepared by laser molecular beam epitaxy method on clean *n*-type Si (100) substrates at the growth temperature of 300 °C using a Nd:YAG laser. Ambient oxygen pressure during deposition was 10^{-6} Torr. The total thickness of each sample was set at about 3 nm. Synchrotron radiation photoemission spectroscopy measurements were carried out at an undulator beamline BL-2C of the Photon Factory in High-Energy Accelerator Organization (KEK). Annealing of $LaAlO_3$ thin films was performed under UHV by the direct current flowing method through the samples for 3 min at each temperature before the photoemission measurements.

Results and discussion

Figure 1(a) shows the Si 2*s* core level spectra for as-grown, 700 °C-, 800 °C-, and 850 °C-annealed $LaAlO_3$ thin films. The peak intensity ratio of Si oxide to Si substrate is larger for 700 °C- and 800 °C-annealed samples than that for as-grown one in Si 2*s* spectra, which indicates the formation of $LaAlSiO_x$ due to the interdiffusion of the interfacial SiO_2 layer and the top $LaAlO_3$ layer during annealing. This is supported by the fact that the peak positions of Al 2*p* core level for 700 °C- and 800 °C-annealed $LaAlO_3$ shift toward higher binding energy than that for as-grown $LaAlO_3$, as shown in Fig. 1(b).

After 850 °C annealing, intensity of Si oxide peak drastically decreases. This is due to the desorption of Si

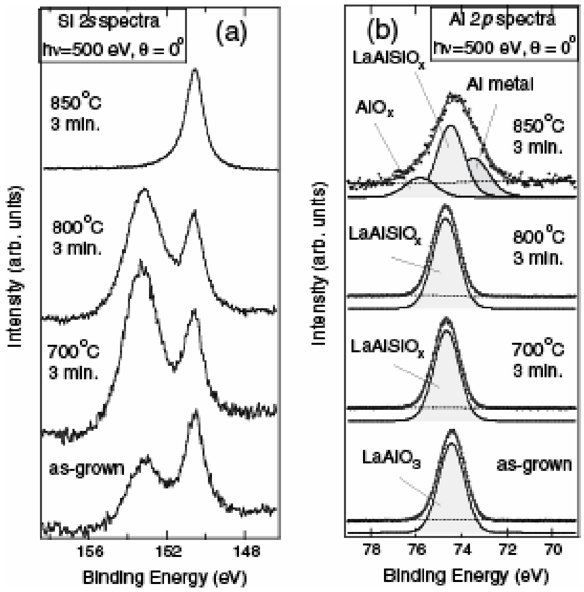

Fig. 1 (a) Si 2*s* and (b) Al 2*p* spectra for as-grown, 700°C-, 800°C-, and 850 °C-annealed $LaAlO_3$ thin films on Si substrates. Each UHV annealing was performed for 3 min.

oxide (volatile SiO). Furthermore, this reduction reaction involves the decomposition of $LaAlSiO_x$. After annealing at 850 °C, Al 2*p* spectrum contains three components, such as AlO_x, $LaAlO_x$, and Al metal peaks. The existence of Al metal after UHV annealing at 850 °C is consistent with the results in HfO_2-Al_2O_3 system [4]. In this stage, desorption of Al-O component (may be Al_2O) as well as SiO gas may cause the decomposition of $LaAlSiO_x$. Flat surface is observed for the 800°C-annealed film from the atomic force microscopy measurements,, while clusters with the hight of several nanometers are observed for 850 °C-annealed one. Combined with the angle-dependent photoemission spectroscopy of Al 2p core level spectra, Al-metal is most probable clusters.

References

[1] G. D. Wilk et al., J. Appl. Phys. 89, 5243 (2001).
[2] A. D. Li et al., Appl. Phys. Lett. 83, 3540 (2003).
[3] X. B. Lu et al., Appl. Phys. Lett. 84, 2620 (2004).
[4] H. S. Chang et al., Appl. Phys. Lett. 84, 28 (2004).

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