

Annealing-ambient dependence of LaAlO₃/SiO₂/Si gate stack structures studied by synchrotron radiation photoemission spectroscopy

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Introduction

Recently high dielectric constant (high-*k*) materials such as Al₂O₃, ZrO₂, HfO₂, rare-earth oxides, *etc.* have been intensively studied as alternatives to SiO₂ which has long been used as gate dielectrics in ultra-large scale integration (ULSI) metal-oxide-semiconductor field-effect transistors (MOSFETs) [1]. Among them, LaAlO₃ is regarded as a promising candidate for high-*k* gate dielectrics in MOSFETs [2].

Although thermal stability of LaAlO₃ under various conditions has been reported [3], the detailed behavior of LaAlO₃ thin films on Si during annealing has not been investigated so far. Thus, we have studied the annealing-ambient dependence of LaAlO₃/Si in terms of photoemission spectroscopy.

Experimental

LaAlO₃ 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 nominal thickness of each sample was set at about 3 nm. Synchrotron radiation photoemission spectroscopy measurements were carried out at an undulator beamline BL-2C. Annealing of LaAlO₃ thin films was performed at 10⁻⁷ Torr (base pressure of the annealing chamber), N₂ 10 Torr, and 100 Torr by the direct current flowing method through the samples for 3 min at 850 °C before the photoemission measurements.

Results and discussion

Figure 1(a) shows La 4*d* and Si 2*p* core level spectra for as-grown and 850 °C-annealed LaAlO₃ thin films at 10⁻⁷ Torr, 10 Torr, and 100 Torr N₂. La 4*d* and Si 2*p* oxide peak intensities dramatically decrease after the annealing under base pressure. This indicates that the annealing results in the thermal decomposition involving the reduction of LaAlO₃. On the other hand, the intensity ratio of La 4*d* to Si 2*p* substrate peak does not decrease after the N₂-ambient annealing, which indicates the suppression of reduction reaction. Furthermore, Si oxide peak intensity increases with introducing the N₂ ambient. This behavior can be explained by following two different scenarios;

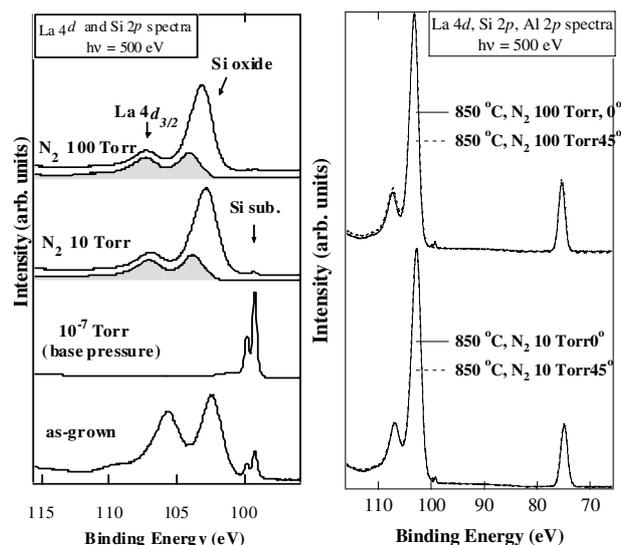


Fig. 1 (a) La 4*d* and Si 2*p* core level spectra for as-grown and 850 °C-annealed LaAlO₃ thin films at 10⁻⁷ Torr, 10 Torr, and 100 Torr N₂. (b) La 4*d* and Si 2*p* core level spectra with the take-off angle of 0° and 45° after annealing in N₂ ambient.

- (1) Residual oxygen in ambient N₂ diffuses into the SiO₂/Si interface through LaAlO₃, which results in the oxidation of buried Si substrates.
- (2) The diffusion of Si derived from substrates or the formation of SiO gas at the SiO₂/Si interface leads to the formation of LaAlSiO_x.

Figure 1(b) shows La 4*d* and Si 2*p* core level spectra with the take-off angle of 0° (bulk sensitive) and 45° (surface sensitive) after annealing in the N₂ ambient. No take-off angle dependence is observed, which indicates that the layered structure is not maintained. Since the former scenario should involve the LaAlO₃/SiO₂ layered structure, these results suggest the possibility of the later scenario.

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).

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