Thermal decomposition 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 dielectric constant (high-k) gate dielectrics dielectrics in metal-oxide-semiconductor field-effect transistors (MOSFETs) [2].

Although thermal stability of LaAlO3 under various conditions has been reported [3], the behavior of LaAlO3 thin films on Si during ultrahigh vacuum (UHV) annealing has not been clarified. Thus, we have studied the changes in chemical states of LaAlO3/Si interface after UHV annealing.

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 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 LaAlO3 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 2s core level spectra for as-grown, 700 °C-, 800 °C-, and 850 °C-annealed LaAlO3 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 2s spectra, which indicates the formation of LaAlSiO4 due to the interdiffusion of the interfacial SiO2 layer and the top LaAlO3 layer during annealing. This is supported by the fact that the peak positions of Al 2p core level for 700 °C- and 800 °C-annealed LaAlO3 shift toward higher binding energy than that for as-grown LaAlO3, 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 oxide (volatile SiO). Furthermore, this reduction reaction involves the decomposition of LaAlSiO4. After annealing at 850 °C, Al 2p spectrum contains three components, such as Al2O3, LaAlO3, and Al metal peaks. The existence of Al metal after UHV annealing at 850 °C is consistent with the results in HfO2-Al2O3 system [4]. In this stage, desorption of Al-O component (may be Al2O3) as well as SiO gas may cause the decomposition of LaAlSiO4. 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


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