

Influence of Thermal Annealing on Chemical Structure of Lanthanum oxide/Si Interfacial Transition Layer

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1. Introduction

La₂O₃ have been studied extensively as an alternative to silicon oxide in the future generation of MOSFETs.[1-4] In general the lanthanum silicate layer is intentionally or inevitably formed between the lanthanum oxide and Si substrate as a transition layer. The SiO₂/Si interfacial transition layer is also formed between this silicate layer and Si substrate. Because the interface state density and the carrier transport in channel region are seriously affected by the structure of these transition layers(abbreviated as TLs hereafter), it is important to clarify the chemical structure of these TLs in the depth direction. However, in our previous study we could not determine the chemical structures of these TLs because Si 2p core level have almost the same binding energy with La 3d core level and we inevitably used Si 2s spectra, whose full width at half maximum(FWHM) is not small enough for the determination of the chemical structures of these TLs, instead of Si 2p spectra. Hard X-ray (5.9534 keV photons) photoelectron spectroscopy system newly developed at SPring-8 enabled us to detect Si 1s spectra with enough energy resolution for the determination of the chemical structure of these TLs. The chemical structures of these TLs can be easily determined with atomic-scale depth resolution if we analyze angle-resolved photoelectron spectra based on high resolution Rutherford backscattering(HRBS) spectra.[4,5]

2. Experimental Details

Approximately 4-nm-thick lanthanum oxide films were deposited on Si(100) substrate at room temperature by electron beam evaporation of La₂O₃ and subsequently annealed in nitrogen atmosphere under atmospheric pressure at 300°C, 400°C and 500°C. 5.9534 keV photons excited La 3d, Si 1s and O 1s spectra arising from these films were measured at photoelectron take-off angle of 8, 15, 30, 40, 55 and 80 degrees using high resolution electron energy analyzer ESCA-2002 at undulator beam line(BL47XU) of SPring-8.

3. Experimental Results and Discussions

Figure 1 shows annealing-induced changes in the atomic composition of lanthanum oxide films. According to this figure the increase in the annealing temperature from 300°C to 400°C results in the

formation of 0.63-nm-thick silicon oxide layer at the interface. Further increase in the annealing temperature from 400°C to 500°C results in the inward diffusion of lanthanum atoms and outward diffusion of silicon atoms. Figure 2 shows La 3d, Si 1s and O 1s spectra measured at photoelectron take-off angle of 50 degrees with annealing temperature as a parameter. According to this figure lanthanum silicate is formed by increasing the annealing temperature from 300°C to 400°C and 500°C. Figure 3 shows the La 3d spectral intensity N_{La} arising from La₂O₃ and Si 1s spectral intensity N_{SiO_2} arising from SiO₂ as a function of photoelectron take-off angle. Here, N_{La} and N_{SiO_2} are normalized by Si 1s spectral intensity N_{Si} arising from Si substrate. Solid and dashed lines in Fig. 3 were calculated by assuming that La₂O₃ is formed and remaining oxygen atoms form SiO₂ and photoelectric cross section of La 3d is two times larger than that calculated.[6] Therefore, the dependence of the compositional depth profile on the annealing temperature shown in Fig. 1 implies that La₂O₃ diffuses inward and SiO₂ diffuses outward to form lanthanum silicate between La₂O₃ and Si substrate. In Fig. 2 the chemical structure of the La₂O₃/Si interfacial TL after annealed at 300°C are shown to consist of Si⁴⁺, Si³⁺, Si¹⁺ in addition to Si^{x+}, which localizes at La₂O₃/SiO₂ interface. The total amount of Si¹⁺ and Si³⁺ was found to be $4.7 \times 10^{18} \text{ m}^{-2}$, which is close to the areal density of Si atoms at the SiO₂/Si interface and implies the abrupt compositional transition at the interface.

4. Conclusion

Measurement of angle-resolved Si 1s spectra using hard X-ray photoelectron spectroscopy system newly developed at SPring-8 combined with HRBS enabled us to clarify the dependence of the chemical structures of lanthanum oxide/Si interfacial transition layer on the annealing temperature.

References

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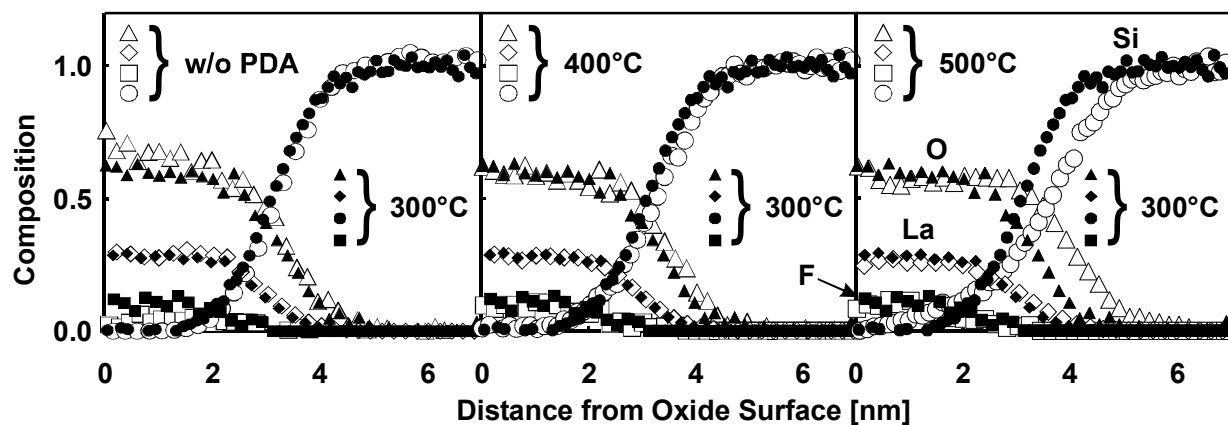


Fig 1. Influence of annealing temperature on composition depth profile of lanthanum oxide/Si(100)

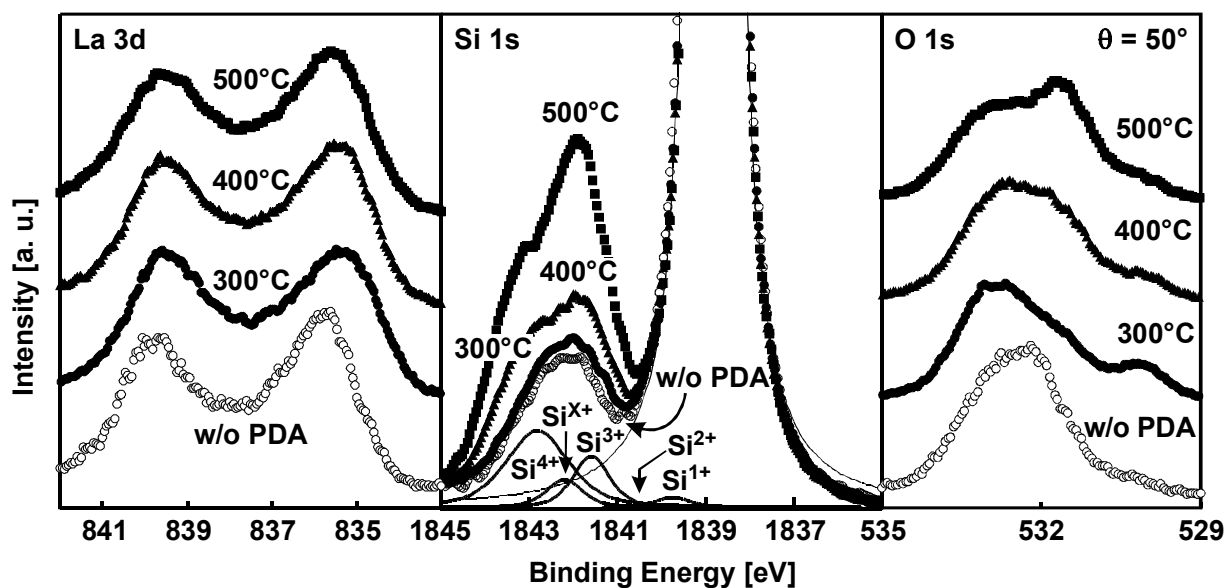


Fig 2. La 3d, Si 1s and O 1s photoelectron spectra with annealing temperature as a parameter

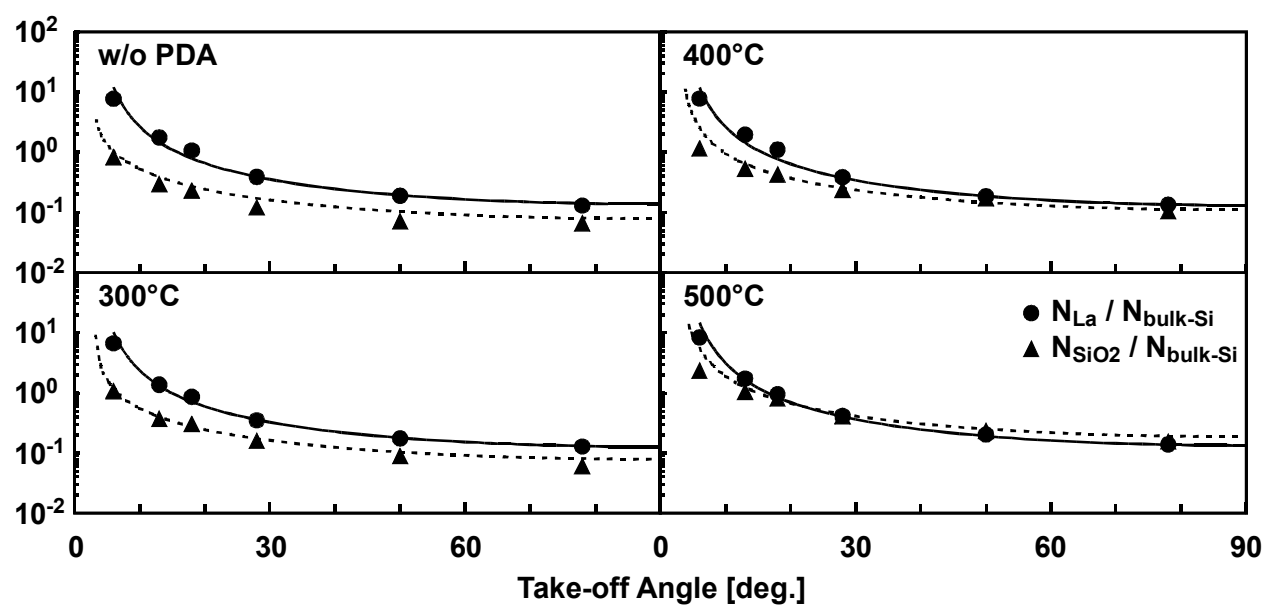


Fig 3. Dependence of $N_{\text{La}} / N_{\text{bulk-Si}}$ and $N_{\text{SiO}_2} / N_{\text{bulk-Si}}$ on photoelectron take-off angle