Extended Abstracts of the 2001 International Conference on Solid State Devices and Materials, Tokyo, 2001, pp. 224–225

B-6-2

Atomic-Scale Depth Profiling of Oxides/Si(111) and Oxynitrides/Si(100) Interface

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1. Introduction

0.8-nm-thick silicon oxide, which was used as gate insulator in MOSFET operated at room temperature[1], consists mostly of structural transition layer[2], the quality of which must be improved using atomic oxygen. Our studies on the oxidation mechanism of Si(111) surface using atomic oxygen revealed that the oxidation rate becomes extremely small if the amount of Si^{2+} and Si^{3+} localized at the interface takes their maximum value[3]. However, the mechanism for this phenomena is not clarified yet.

The silicon oxynitrides are also important as ultrathin gate dielectrics in future MOSFETs due to their improved reliability and ability to suppress boron penetration. Our studies on the chemical structures of oxynitrides/Si(100) interface formed by the wet oxidation followed by the annealing in NO ambient revealed that the most of nitrogen atoms localize near the interface and have bonding with two Si atoms at the interface and one Si atom, which has bonding with three oxygen atoms in the overlayer.[4] However, the effect of nitridation on the distribution of intermediate oxidation states of Si is not clarified yet.

It is the purpose of the present paper to demonstrate the atomic-scale depth profiling of the chemical structures of the SiO_2/Si(111) interface and oxynitrides/Si(100) interface structures by applying the maximum entropy method[5,6] to the angle resolved Si 2p photoelectron spectra under the consideration of the elastic scattering of Si 2p photoelectrons in silicon oxide films.[7]

2. Experimental Details

The oxide films studied were prepared as follows. The atomically flat hydrogen terminated Si(111) surfaces were oxidized at 400°C using atomic oxygen produced in microwave (5.0 W/cm^2 at 2.45 GHz) excited Kr/O_2 mixture(Kr/O_2 = 97/3, 1 Torr) gas plasma. Oxynitride films containing maximum nitrogen concentrations of 1, 2, 3, 5 and 6 at.% were grown on p-type Si(100) by the wet oxidation followed by the annealing in NO ambient at 900°C.

Angle resolved Si 2p spectra excited by AlKα radiation arising from these films were measured with photoelectron acceptance angle of 3.3° at the entrance of electron energy analyzer. using high resolution and highly sensitive ESCA-300[8].

3. Experimental Results and Discussions

Figure 1 shows the dependence of Si 2p photoelectron spectral intensities NI arising from intermediate oxidation states Si^{1+}, Si^{2+}, Si^{3+} and those NO arising from SiO_2 in silicon oxide films formed using atomic oxygen normalized by Si 2p spectral intensities NS arising from Si substrate on photoelectron take-off angle for the interface containing maximum amount of Si^{3+}, Si^{1+} and Si^{2+}. Figure 2 shows Gaussian distribution of the amount of Si^{1+}, Si^{2+}, Si^{3+} and SiO_2 obtained by applying maximum entropy method to Fig. 1. The solid, dotted and dashed curves in Fig. 1 are calculated using the depth profile in Fig. 2. According to Fig. 2, the interface containing the maximum amount of Si^{1+} is extremely abrupt than the interface containing the maximum amount of Si^{3+} or Si^{2+}. This must be the reason for the observation of extremely small oxidation rate at the interface containing the maximum amount of Si^{3+} or Si^{2+}.

Figure 3 shows the dependence of Si 2p photoelectron spectral intensities NI arising from Si^{1+}, Si^{2+}, Si^{3+} and those NO arising from SiO_2 in silicon oxide films formed using molecular oxygen normalized by Si 2p spectral intensities NS arising from Si substrate on photoelectron take-off angle for interface containing maximum amount of Si^{3+} and Si^{1+}. Figure 4 shows Gaussian distribution of the amount of Si^{1+}, Si^{2+}, Si^{3+} and SiO_2 obtained by applying maximum entropy method to Fig. 3. The solid, dotted and dashed curves in Fig. 3 are calculated using the depth profile in Fig. 4. It is found from the comparison between Figs. 2 and 4 that the interfaces formed using atomic oxygen are more abrupt than the interfaces formed using molecular oxygen.

Figure 5 shows the dependence of Si 2p photoelectron spectral intensities NO arising from SiO_2, those NI arising from intermediate oxidation states SiO_x and those NN arising from oxynitrides SiO_xN_y normalized by Si 2p spectral intensities NS arising from Si substrate on photoelectron take-off angle for oxynitiride containing maximum nitrogen concentrations of 3 and 6 at.%. Figure 6 shows Gaussian distribution of the amount of SiO_2, SiO_x and SiO_xN_y obtained by applying maximum entropy method to Fig. 5. The solid, dotted and dashed curves in Fig. 5 are calculated using the depth profile in Fig. 6. According to Fig. 6, the amount of intermediate oxidation states of Si is weakly affected by the incorporation of nitrogen atoms at SiO_2/Si interface.

4. Conclusion

In conclusion, the atomic-scale depth profiling of SiO_2/Si(111) interface and silicon oxynitrides/Si(100) interface structures are demonstrated for the first time by applying maximum entropy method to the angle-resolved Si 2p spectral intensities arising from intermediate oxidation states of Si and those arising from silicon oxynitrides normalized by Si 2p spectral intensities arising from Si substrate under the consideration of the elastic scattering of Si 2p photoelectrons in silicon oxide films. The present depth profiling analysis must be effective for the study on the high-k dielectrics/Si interface structures.

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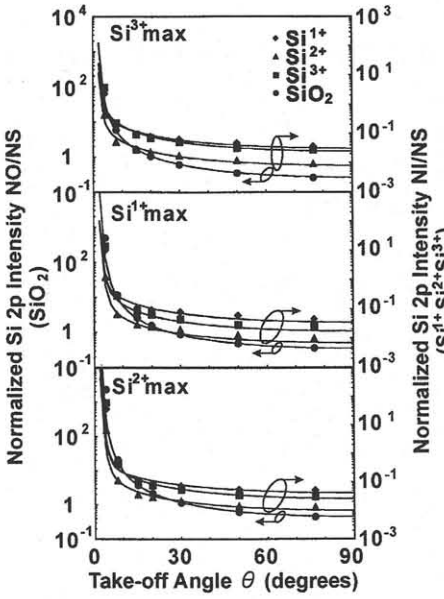

Fig. 1 Dependence of normalized Si 2p spectral intensities arising from Si^{1+}, Si^{2+}, and Si^{3+} and SiO_2 in oxide formed using atomic oxygen on photoelectron take-off angle. The solid curves are calculated using the depth profile in shown in Fig. 2.

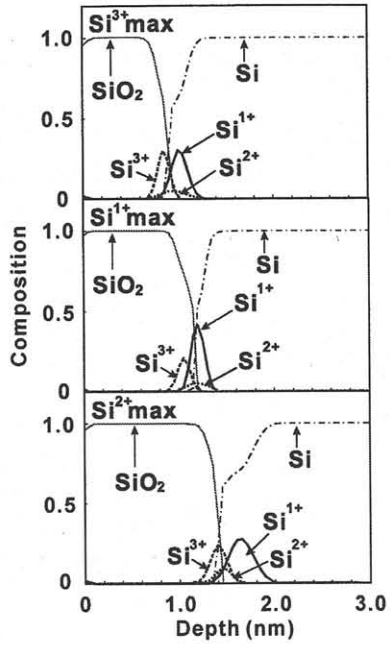

Fig. 2 Compositional depth profile obtained from the analysis of Fig. 1.

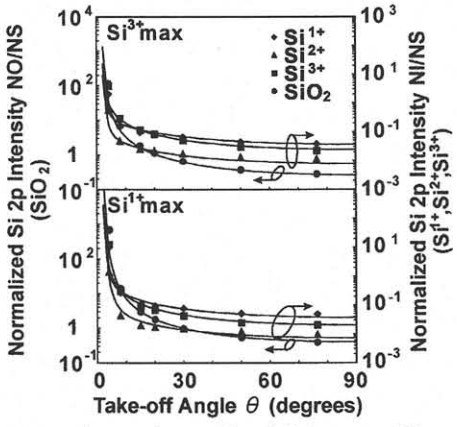

Fig. 3 Dependence of normalized Si 2p spectral intensities arising from Si^{1+}, Si^{2+}, and Si^{3+} and SiO_2 in oxide formed using molecular oxygen on photoelectron take-off angle. The solid curves are calculated using the depth profile shown in Fig. 4.

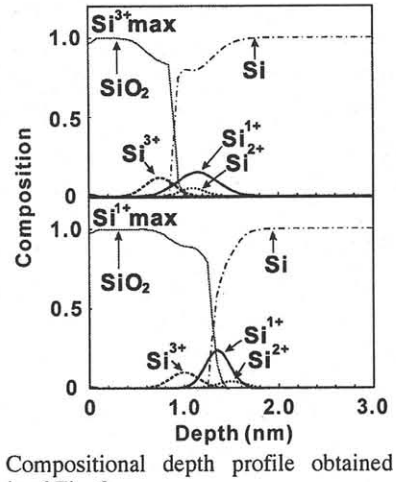

Fig. 4 Compositional depth profile obtained from the analysis of Fig. 3 .

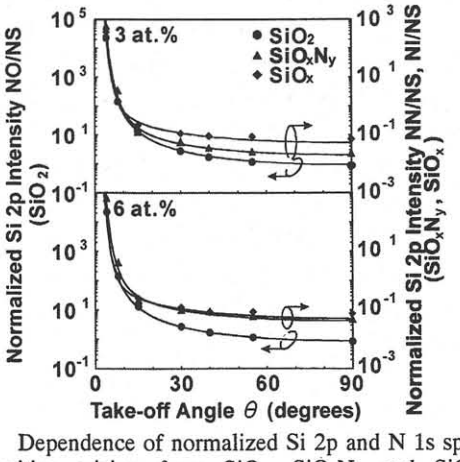

Fig. 5 Dependence of normalized Si 2p and N 1s spectral intensities arising from SiO_x, SiO_xN_y and SiO_2 in oxynitrides on photoelectron take-off angle. The solid curves are calculated using depth profile shown in Fig. 6.

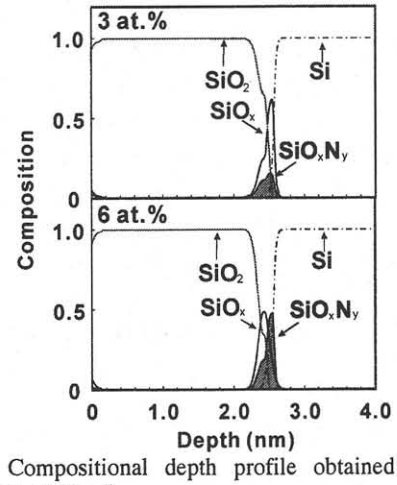

Fig. 6 Compositional depth profile obtained from the analysis of Fig. 5 .