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Chemical structures of ultrathin SiO$_2$/Si(100) interfaces revealed by ARPES

K. Kimura[1], M. Oshima[1], J.-H. Oh[2], K. Ono[3], M. Niwa[4], K. Usuda[4], and N. Hirashita[4]

[1]Department of Applied Chemistry, The University of Tokyo, Bunkyou-ku, Tokyo 113-8656, Japan

[2]Hiroshima University, Higashi-Hiroshima, Hiroshima 739-8526, Japan

[3]High-Energy Accelerator Organization (KEK), Tsukuba, Ibaraki 305-0801, Japan

[4]Semiconductor Technology Academic Research Center, Kanagawa 222-0033, Japan

1. Introduction

Extensive studies have been performed on formation and analysis of ultrathin SiO$_2$ films on Si(100) with atomically abrupt interfaces for ULSI gate dielectrics applications where MOSFET normal operation was confirmed for only 0.8 nm thick SiO$_2$ films. However, the atomic-scale understanding of the interface structures is still lacking. The intriguing nature of the interfaces is manifested by the existence of a transition layer at the SiO$_2$/Si interfaces. Although many interfacial structure models[1] have been proposed for SiO$_2$/Si(111) and (100), the contradicting discussions on the chemical abruptness including sub-oxide component distribution, and the thickness of transition layers are still in progress. Recently Oh *et al.* proposed a three-layer model for a 0.6 nm thick SiO$_2$ film grown on the Si(100) substrate based on high-resolution angle-resolved photoelectron spectroscopy (PES) results.[2] In this study, we have performed high-resolution PES of ultrathin SiO$_2$ films in order to investigate the evolution of the transition layer in the course of thermal oxidation.

2. Experimental

High-resolution PES experiments were carried out using an angle-resolved photoelectron spectrometer system (VG ARUPS-10) at BL-1C of the Photon Factory. A clean Si(100) 2x1 substrate was oxidized by highly pure O$_2$ gas at a pressure of 5x10^{-8}Torr for 6000, 2000, 1000, 400, and 200 sec (300, 100, 50, 20, and 10L in total) at 600°C. The total thicknesses of the oxide layer were estimated to be 0.5-0.2 nm by a simple Si *2p* intensity analysis. The Si *2p* photoelectron spectra were measured by changing the polar emission angle (θ) from 0° to 70°, with steps 2.5°. The total energy resolution was set to 70meV at the photon energy of 130eV. The angular resolution was set to 2° for all measurements.

3. Results and discussion

Figure 1(a) shows Si *2p* spectra of SiO$_2$/Si(100) (50 L at 600°C) taken at (a) θ=0° and (b) θ=60°. For quantitative analyses, all the spectra were fitted by a standard least-square fitting procedure with spin orbit doublets of Voigt functions. These spectra were distinctly deconvoluted into B and Si^{1+}-Si^{4+} components. The accurately determined core-level shifts were 1.00, 1.79, 2.60, and 3.58eV for the Si^{1+}-Si^{4+} components, respectively. Furthermore, two extra components, α and β, were required to obtain reasonably good fits, with binding energy shifts of about -0.28eV and 0.27eV, respectively.
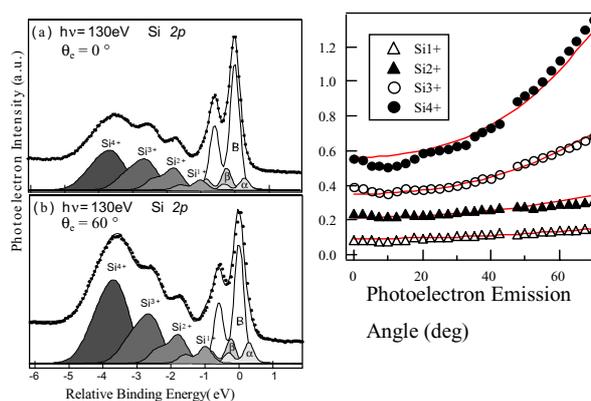
Fig. 1. Si *2p* photoelectron spectra (left) and dependence of relative intensities of Si sub-oxide components on photoelectron emission polar angle (right) from the 50L SiO$_2$/Si(100) sample.

In order to determine the depth distribution of sub-oxide components, Si *2p* photoelectron intensities of Si^{1+}-Si^{4+} components are plotted as a function of θ, from 0° to 70°, as shown in Fig. 1(b). As θ increases, the normalized intensities increase. These results can be well explained by the three-layer non-abrupt interface model.

We, then, investigated various SiO$_2$/Si(100) interfaces with changing O$_2$ dosage from 10 L to 300 L, and found that the three-layer model fits the results for 50 L to 300 L very well. At 10 L and 20 L, however, the intensity ratios are not explained simply by the model. At the initial stage of Si(100) oxidation, Si^{1+} and Si^{4+} exist together on the surface, suggesting that oxidation proceeds fast at some part of the surface, resulting in formation of Si^{4+}, while on the other part oxidation starts slowly, resulting in formation of Si^{1+}.

Thus, the initial oxidation features can be well explained by the non-abrupt interface model for 50 L to 300 L. From the detailed analysis of sub-oxide component intensities, it is found that the content of Si^{3+} at the third layer and that of Si^{1+} at the first layer increase as oxidation proceeds, and finally both reach 50 %. This means that the interface becomes abrupt based on the statistical cross-linking model[1]. These results provide a good insight for formation of ultrathin gate insulator films.

References

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