Extended Abstracts of the 1999 International Conference on Solid State Devices and Materials, Tokyo, 1999, pp. 310-311

Mechanism of Layer-by-Layer Oxidation of Si (001) Surfaces Proceeding by Two-Dimensional Oxide-Island Nucleation at SiO_2/Si Interfaces

Heiji Watanabe, Toshio Baba, and Masakazu Ichikawa[1]

Fundamental Research Laboratories, NEC Corporation,
34 Miyukigaoka, Tsukuba, Ibaraki 305-8501, Japan
Phone: +81-298-50-1512 Fax: +81-298- 56-6139 e-mail: watanabe@frl.cl.nec.co.jp
[1]Joint Research Center for Atom Technology, Angstrom Technology Partnership,
1-1-4 Higashi, Tsukuba, Ibaraki 305-0046, Japan

1. Introduction

The layer-by-layer oxidation of Si surfaces has attracted a great deal of attention because layer-by-layer oxidation is an ideal way to realize an ultrathin gate oxide for MOS devices [1-2]. We have found that scanning reflection electron microscopy (SREM) can reveal atomic-structures at buried SiO_2/Si(001) interfaces, and have observed a periodic reversal of terrace contrast in SREM images during initial oxidation [3]. From our SREM observations, we confirmed the layer-by-layer oxidation of Si(001) surfaces, and proposed a mechanism by which the oxidation proceeds by two-dimensional (2D) oxide-island nucleation at the interfaces. However, this oxidation mechanism has not been directly confirmed by SREM due to the resolution limit of this technique.

In this study, to clarify the layer-by-layer oxidation mechanism, we investigated SiO_2/Si(001) interfaces by using reflection high-energy electron diffraction (RHEED) and scanning tunneling microscopy (STM) techniques.

2. RHEED intensity oscillation during oxidation

The spot intensity oscillation in RHEED patterns has been reported for various kinds of epitaxial growth that proceed by 2D island nucleation [4]. We used RHEED to investigate the oxidation of Si surfaces. Figures 1(a) and (b), respectively, show the change in the specular spot intensity and the oxygen Auger peak intensity as a function of the oxidation time. A Si(001)-2x1 surface was oxidized at room temperature under 1 x 10^{-7} oxygen gas pressure. Under these conditions, the first-layer oxidation was complete within 15 min [3]. The spot intensity reached a minimum after about 3 min of oxidation, and then gradually recovered and saturated after 15 min. In addition, we investigated the streaky spot profile obtained from the SiO_2/Si(001) interface, and confirmed that the diffuse-scattering originating from atomic-scale interfacial roughness increased at about 3 min (Fig. 2). This oxidation time corresponded to the 0.5-ML oxidation of the first layer (see Fig. 1(b) and ref.[3]). From these results, we can conclude that the oscillation phenomena shown in Figs. 1 and 2 are attributed to the RHEED oscillation phenomena during the first-layer oxidation, which is consistent with our layer-by-layer oxidation model based on 2D oxide-island nucleation.

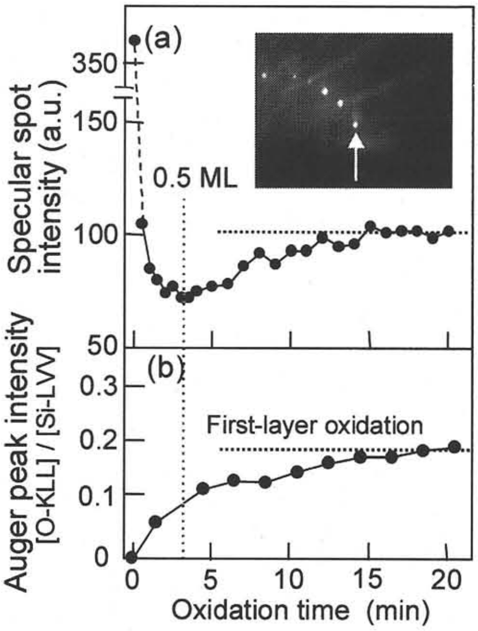

Fig. 1. First-layer oxidation of Si(001)-2x1 surfaces. (a) shows the change in a specular spot intensity in RHEED patterns, and (b) shows an increase in the oxygen Auger peak intensity [O-KLL] normalized by the Si peak [Si-LVV].

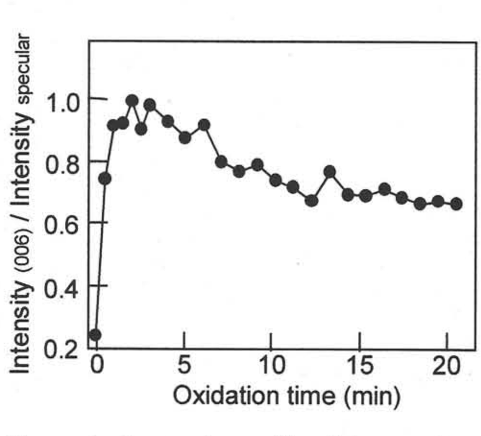

Fig. 2. Change in the streaky profile of the specular spot during oxidation. The intensity ratio between the diffuse-scattering around the (006) reflection and the specular spot intensity is plotted as a function of oxidation time.

3. STM observation of 2D oxide islands at SiO_2/Si(001)

Next, we used STM to directly observe 2D oxide islands nucleated at the interfaces. Since SREM observation reveals the oxidation kinetics of each layer [3], we could prepare SiO_2/Si samples with an oxide thickness of 2 ML or 2.5 ML. Figure 3(a) is a SREM image obtained after 2-ML oxidation, where we can observe the interfacial terrace contrast. After 2.5-ML oxidation, the terrace contrast disappeared (Fig. 3(b)). The oxide layers were carefully removed with a HF solution (1%), then we observed the interfacial morphology by STM. Figures 3(c) and (d) show the SiO_2(2 ML)/Si(001) interfacial morphology. We could recognize interfacial steps and atomically-flat terraces. In contrast to the 2-ML oxidation, we observed increased roughness for 2.5-ML oxidation (Figs. 3(e)) and (f)). Note that the typical corrugation has a single-atomic height. These results indicate that the STM image obtained after 2.5-ML oxidation directly revealed 2D oxide islands nucleated at the SiO_2/Si(001) interface during the third-layer oxidation. We also found that the islands were around 3–5 nm in diameter, which is consistent with our oxidation model and estimation from the RHEED spot analysis [5].

4. Conclusion

We have investigated the oxidation of Si(001) surfaces by using RHEED and STM. We observed both RHEED intensity oscillation and 2D oxide-island nucleation at the interfaces. These results confirm the validity of our layer-by-layer oxidation model in which oxidation proceeds by oxide-island nucleation and lateral island growth.

Acknowledgment

This work was partly supported by NEDO.

References

[1] J. M. Gibson and M. Y. Lanzerotti, Nature **340**, 128 (1989).
[2] K. Ohishi and T. Hattori, Jpn. J. Appl. Phys., Part 2 **33**, L675 (1994), M. Ohashi and T. Hattori, Jpn. J. Appl. Phys., Part 2 **36**, L397 (1997).
[3] H. Watanabe, K. Kato, T. Uda, K. Fujita, M. Ichikawa, Y. Kawamura and K. Terakura, Phys. Rev. Lett. **80**, 345 (1998).
[4] J. J. Harris and B. A. Joyce, Surf. Sci. **103**, 90 (1981).
[5] N. Miyata, H. Watanabe and M. Ichikawa, Phys. Rev. **B58**, 13670 (1998).

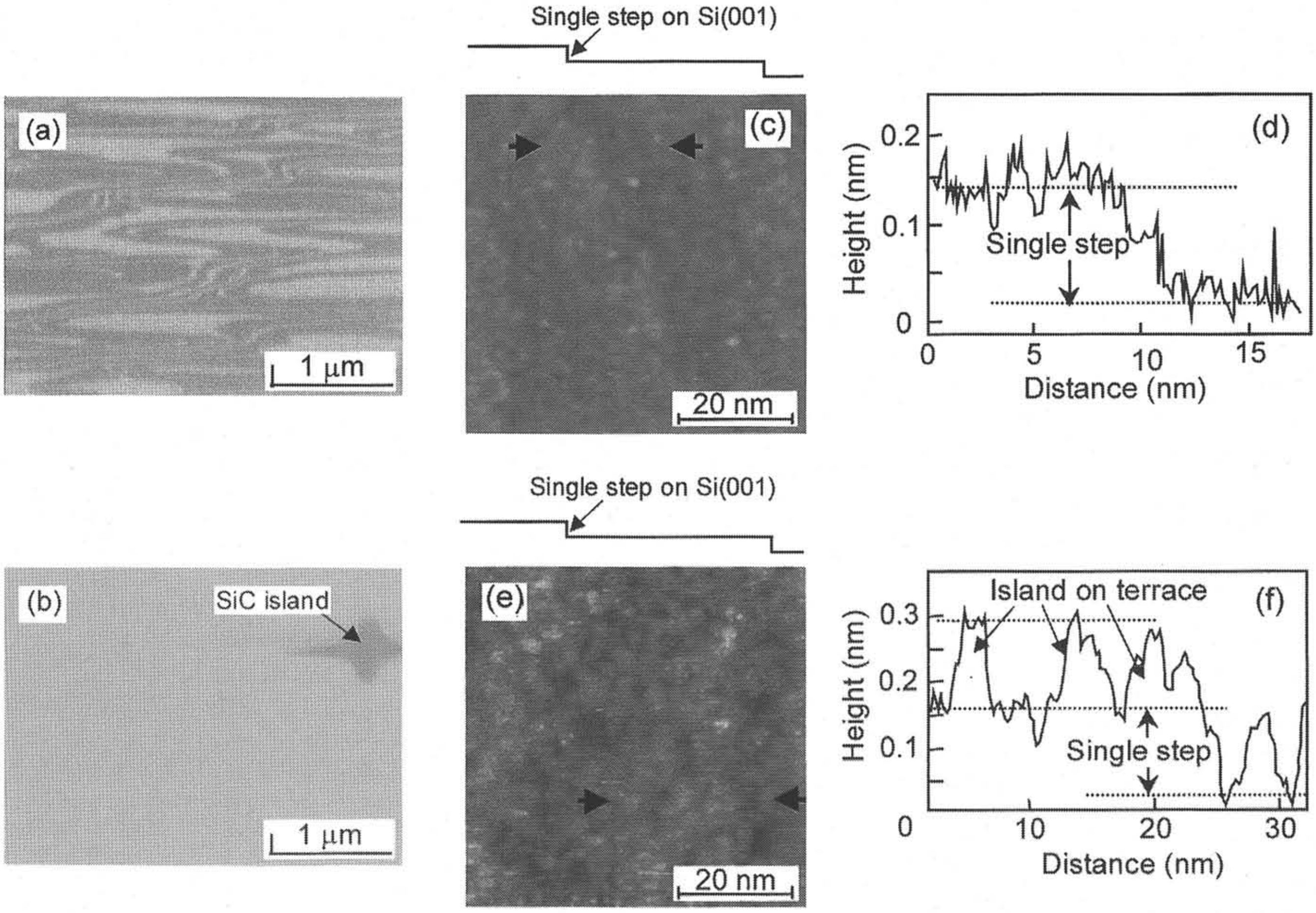

Fig. 3. Direct STM observation of 2D oxide islands. The Si(001)-2x1 surfaces were oxidized to 2 ML or 2.5 ML. The oxide thickness was accurately monitored through the interfacial SREM images shown in (a) and (b). (c) and (d) show the results of STM observation after 2-ML oxidation, and (e) and (f) show the results after 2.5-ML oxidation.