Evaluation of SiO_2/(001)Si interface roughness using high-resolution transmission electron microscopy and simulation

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The high-resolution transmission-electron-microscopy (HRTEM) image of the SiO_2/(001)Si interface varies with TEM specimen thickness. The most noticeable feature is the periodic image observed between the lattice fringe of the *c*-Si substrate and the granular image of SiO_2 for the thick region of the specimen. Optical-diffraction-pattern observations clarified that each 111 spot obtained from the periodic image of the thick specimen is split into two spots. Simulation has revealed that both the periodic image and the 111 spot splitting are reproduced by interface roughness with Si protrusions delineated by {111} facets. The height of the Si protrusion can be evaluated by comparing the HRTEM images and the optical-diffraction pattern taken from the thick region of the specimen with simulated ones. For the interface formed by dry oxidation at 950 °C, the Si protrusion is higher than 6 monolayers.

I. INTRODUCTION

The morphology of the SiO_2/Si interface has been studied using a variety of methods, including low-energy electron diffraction (LEED) microscopy,[1] scanning tunneling microscopy (STM),[2] and high-resolution transmission electron microscopy (HRTEM).[3–8] When we use surface-analyzing techniques, such as LEED and STM, the oxide layer has to be removed to obtain the bare Si surface. This etching process will possibly modify the atomic-level interface structure because the SiO_2/Si interface contains a suboxide layer.[9–11]

HRTEM is one of the methods by which we can observe the interface structure without removing the oxide. A number of papers on SiO_2/Si interface morphology and how it relates to electrical properties have been published.[3–8] It is generally understood that the interface morphology observed by HRTEM depends on the crystallographic orientation of the *c*-Si substrate; that is, the SiO_2/(111)Si interface is observed to be quite flat,[3] whereas the SiO_2/(001)Si interface is roughened.[4–7] Current-voltage (*I-V*) characteristics of oxides reveal that large *c*-Si protrusions induce current though the oxide for lower electrical fields.[6] A mobility measurement of a metal-oxide-semiconductor (MOS) transistor showed that the interface roughness should be larger than that observed using HRTEM.[7] Recent observation of the SiO_2/(001)Si interface formed by oxidation of the MBE-grown Si (MBE denotes molecular-beam epitaxy) surface found that a periodic image is observed between the granular image of amorphous SiO_2 and the lattice image of the *c*-Si substrate.[8] This periodic image shows a strong 110 periodicity. Simulation of the HRTEM image and other experimental results suggest that the periodic image is attributable to tridymite growing epitaxially over the atomically flat (001) Si surface and consisting of a number of stacking faults in the plane perpendicular to the *c* axis of the tridymite.

In spite of these efforts, the SiO_2/Si interface morphology is still unclear. This is mainly due to the difficulty of analyzing the HRTEM image. The HRTEM image is not a simple projected image of the potential inside the specimen, but is, instead, formed by (a) electrons multiply scattered inside the specimen, and (b) application of further modulation due to the incompleteness of the HRTEM optical system. Therefore, HRTEM is drastically affected by changes in defocusing and specimen thickness. In addition, since scattering conditions at the interface are different from those in the crystalline bulk due to the termination of the periodic atom arrangement at the interface, it is probable that the lattice fringe of the crystalline material near the interface is modified from that of bulk *c*-Si. Therefore, it is quite difficult to deduce the interface morphology from the HRTEM image. Since the effect of multiple scattering inside the specimen is small for thin specimens, the structural analysis of crystalline material is usually done by comparing the image obtained from a very thin region of the specimen to simulated images. However, in the case of the SiO_2/Si interface, the interface morphology would be modified by the ion-beam-induced annealing that it undergoes during specimen preparation, because the system may consist of a thermally inequilibrated suboxide region.[9–11] Therefore, evaluation of the interface-roughness height using specimens as thick as possible (to reduce the effect of the ion bombardment) is required.

In this paper we propose a method of evaluating the interface-roughness height by comparing HRTEM images and the optical-diffraction patterns obtained experimentally from specimens of varying thickness to simulated HRTEM images and optical-diffraction patterns calculated by using an interface structural model. The SiO_2/(001)Si interface was observed using HRTEM in several regions of the same TEM specimen, revealing that the HRTEM image near the interface varies with specimen thickness. In particular, it was found that a thin layer exhibiting 110 periodicity can be observed between a bulk *c*-Si lattice image and a granular image of SiO_2 for

thick regions of the specimen. Optical-diffraction patterns were taken from the interface region of the HRTEM images to reveal the spatial frequency of the scattered waves that form the image. Observation of optical-diffraction patterns revealed that 111 spots of *c*-Si split into two spots for the region where the HRTEM image with 110 periodicity is observed. The cause of these experimental facts was examined by comparing them with the stimulated images. Both the HRTEM images and the optical-diffraction patterns were compared to estimate the consistency of the HRTEM images obtained by simulation and by experiment accurately. As a result, we have revealed that both the periodic image and 111 spot splitting observed for the thick region of the TEM specimen can be reproduced by interface roughness delineated by the {111} facets with heights of more than 6 monolayers (ML). By using this method, we can estimate the interface-roughness height by using HRTEM images obtained from the thick region of the specimen where the damage due to ion bombardment is expected to be low.

II. EXPERIMENT

Boron-doped (001)Si wafers (4.5–5.5 Ω cm) were used throughout the experiments. RCA cleaning followed by etching with a $HF+NH_4F$ solution were performed as a preoxidation cleaning of the wafer. The oxidation was done in a dry-oxygen-flow furnace heated at 950 °C for a certain time to form oxide layers with desired thicknesses (about 30 nm). After oxidation, the furnace was gradually cooled down to room temperature. The flow gas was changed from oxygen to Ar at the beginning of the cooling-down period. It took about 1 h to lower the wafer temperature from 950 °C to room temperature. In order to protect the interface layer from damage during TEM specimen preparation, polycrystalline Si was deposited on top of the oxide layer by chemical-vapor deposition.

TEM specimens for cross-sectional observation were made by slicing a pair of wafers bonded together with epoxy glue along a direction parallel to the ⟨110⟩ directions of *c*-Si. The sliced specimen was thinned mechanically to several tens of micrometers. Further thinning of the specimen was performed by Ar-ion mill-ing. HRTEM observation was made on the edge of the perforation formed by the ion milling by using a JEM 2000EX device operated at 200 keV from the direction along a ⟨110⟩ direction of *c*-Si substrate.

III. RESULTS OF HRTEM OBSERVATION

HRTEM images taken near the SiO_2/(001)Si interface of two different specimen thicknesses are shown in Fig. 1. Variation of the specimen thickness can be determined from the thickness fringes that are observed in a HRTEM image. The thickness of the "thin" specimen is thinner than the region exhibiting the first thickness fringe, while the "thick" specimen is thicker than that. According to our simulation, the first thickness fringe of *c*-Si appears at a thickness of about 13 nm for observation along the ⟨110⟩ direction at 200 keV. However, we note that the first thickness fringe for a 13-nm-thick specimen is not always observed because of the steep increase in specimen thickness. In order to confirm that the thickness fringe observed in the thinnest region of the specimen is the first thickness fringe observed for a 13-nm-thick specimen, we have examined the intensity of the 002 spots of the optical-diffraction pattern taken from the HRTEM image of the *c*-Si substrate, as will be mentioned later.

For the thin region, the lattice fringe is characteristic of the *c*-Si substrate below the interface, and changes abruptly to the granular image representative of the amorphous SiO_2. Interface roughness is clearly observed. Conversely, for the thick region, a periodic image different from that of the *c*-Si substrate is observed at the interface. This specific layer is about 4 ML thick, exhibits strong 110 periodicity, and is observed along the roughened interface. The image with 110 periodicity is observed in the region where the specimen is thicker than that corresponding to the first thickness fringe for various defocusing conditions.

There are two possible causes for the periodic image at the interface. One is the existence of a crystalline phase between *c*-Si and *a*-SiO_2. The other is the result of a special diffraction condition at the interface due to termination of the periodic potential of *c*-Si and/or the effect of the interface roughness. It has been reported that a similar type of image is observed for the interface formed over the atomically flat (001)Si surface prepared by MBE; this is evidence of the existence of epitaxially grown tridymite.[8] In our case, however, since the SiO_2/(001)Si interface is apparently roughened, as already shown in the figures, the tridymite model, which requires the atomically flat interface, cannot be applied. As is discussed in detail in the following section, we ascribe the 110 periodic image to interface roughness and termination effects.

The optical-diffraction patterns are shown in Fig. 1 in the same row as the corresponding HRTEM images. The optical-diffraction pattern is obtained by using a laser

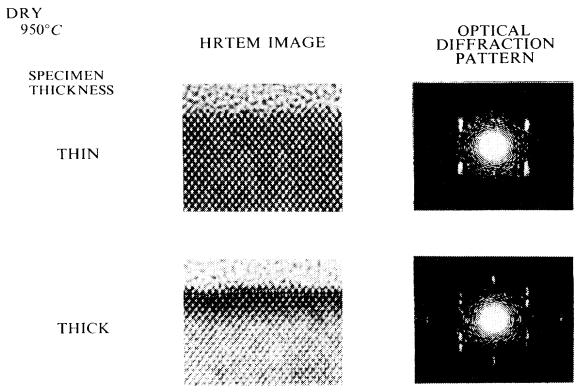

FIG. 1. HRTEM images near SiO_2/(001)Si interface and corresponding optical-diffraction patterns for thin and thick specimens.

beam diffracted by employing a negative film of a HRTEM image as a diffraction grating, so we can know the spatial frequency of the scattered waves which formed the HRTEM image. Since our interest is in the HRTEM image in the vicinity of the SiO_2/(001)Si interface, we irradiated the negative film with the laser beam only at the image near the interface region.

The optical diffraction taken from the thin region of the specimen shows a streak along the direction perpendicular to the interface. This is understood to be the termination effect of the periodic lattice image of *c*-Si at the interface. The optical diffraction provides information on the specimen thickness as well. The 002 spots are not observed in the optical-diffraction pattern for the thin region. Thus, we can conclude that this region is thinner than the first thickness fringe region because 002 reflections are produced by the multiple scattering of electrons inside the specimen and its intensity is quite low for the thin region.

For the thick region, it is noticeable that the optical-diffraction pattern taken from the HRTEM image with 110 periodicity observed at the interface exhibits 111 spot splitting along the direction perpendicular to the interface. Neither of the split spots correspond to the 111 spots of *c*-Si, but they are just above and below the exact 111 positions. Furthermore, the 002 reflection observed in the optical-diffraction pattern of the thick specimen indicates that this region is thicker than the first-thickness-fringe region.

IV. SIMULATION

The HRTEM image is formed by electrons that have undergone multiple scattering inside the specimen and it is affected by the drawbacks of an optical system such as an electron microscope, namely lens aberration and current fluctuation. Therefore, HRTEM images cannot be simply projected images of potential inside the specimen. In order to interpret the HRTEM images and to evaluate the validity of the model used for the simulation correctly, it is necessary to simulate the multiple scattering and the effect of the HRTEM optical system, and then to compare that simulation with the experimental results.

In this section we will show the results of simulating HRTEM images and optical-diffraction patterns by using a simple interface structural model. The multiple scattering of electrons is simulated by using the multislice method,[12] and the effect of aberrations of the HRTEM optical system is simulated by using the method proposed by Horiuchi.[13] However, the effect of the noncollimated incident beam is taken into account in an original method. Since the incident electron beam is focused on the specimen surface to obtain sufficient intensity, an electron has a nonzero momentum toward the direction parallel to the specimen surface as a result of nonzero irradiating angle. Therefore, the probability density of the electron is not uniform over all the specimen surface, but it is presented as a wave packet. We approximated the shape of the wave packet by a Gaussian function and calculated the HRTEM images by assuming that the electrons with Gaussian probability density are struck at the same place in the interface region of the structural model. With this method we are not required to consider periodic boundary conditions and can use any interface structural models. We have confirmed that the HRTEM image simulated by this method is not changed by moving the irradiating position of the electron, except for the intensity.

The extent of a wave packet, x_{12}, is given by the following relation as a function of irradiation angle α and the wavelength of the incident electron λ,[13]

$$x_{12} < \frac{\lambda}{2\alpha} .$$

If the electron wavelength is 2.51×10^{-3} nm for 200 keV, and the irradiation angle is 0.5 mrad, then

$$x_{12} < 2.5 \text{ nm} .$$

We used this extent of the wave packet (2.5 nm) as the standard deviation of the Gaussian function in the HRTEM image simulation.

For the HRTEM image simulation of the interface, it is not possible to simplify the calculation by taking advantage of periodicity. Choice of model size for the simulation is most important for a reasonable simulation. If the model size is too small, since the structure factor of the model has broad peaks, the simulated image becomes that of the small crystalline cluster rather than that of *c*-Si. On the other hand, a large model consumes a great deal of calculational time. Optimization of the model size has been done by comparing the simulated results of *c*-Si obtained using this method with the result obtained using the conventional periodic calculation using the unit cell of *c*-Si. We have found that an (8×8)-cell model gives a reasonable result. This size of the model can include almost the entire extent of the Gaussian function.

The interface has been described by a termination of the periodic atom arrangement. The contribution of SiO_2 to the image and the displacement of atoms in *c*-Si due to SiO_2 stacking has not been taken into account. As the interface models, we have adopted a flat (001) model and {111}-facet models so that we may know the effect of the termination of the periodic atom arrangement on the *c*-Si lattice fringe near the interface, and of the additional effect due to interface roughness. A schematic drawing of the {111}-facet model is shown in Fig. 2. The *c*-Si surface is delineated by {111} facets. The distortion-energy calculation showed that this structure has a lower distortion energy than the flat (001) interface.[14] In order to simplify the modeling, the heights of the Si protrusions are set to be the same while they are displayed randomly in the direction perpendicular to the observed direction to avoid artificial alignment of the roughness. Three {111}-facet models with Si protrusions 4, 6, and 8 ML high have been used in the present work.

In Fig. 3 simulated HRTEM images for the {111}-facet models with various roughness heights are shown for two different specimen thicknesses together with the experimental results. Since the absolute defocusing values of the HRTEM images are not obvious for the experiment, we chose the defocusing values for the simulation by comparing a through-focus set of HRTEM im-

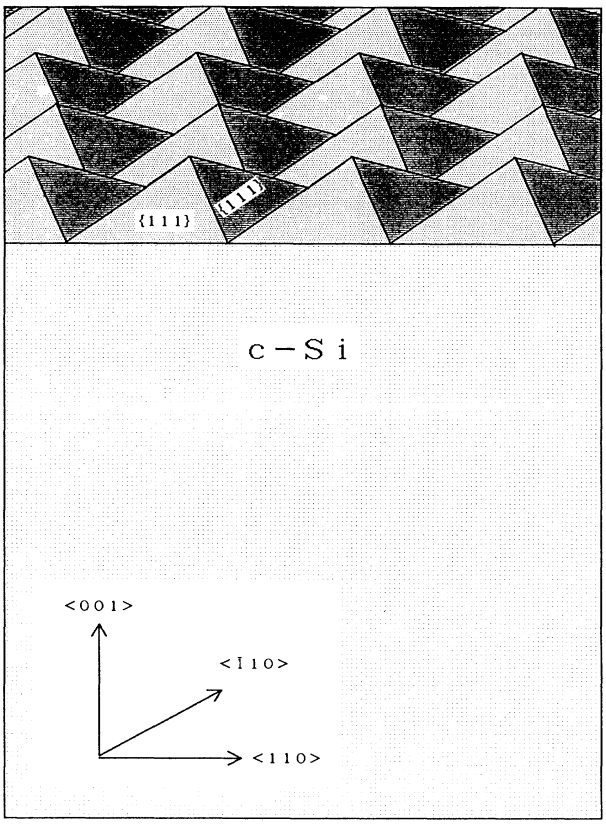

FIG. 2. Schematic drawing of the interface morphology of the {111}-facet model.

ages with simulated images of various defocusing values. The defocusing values for the HRTEM image simulation of the thin and thick regions are −30 and −40 nm, respectively. The thickness of the specimen is governed by the thickness fringes in the HRTEM images, and we used 5 and 23 nm for thin and thick regions, respectively.

For the thin models, there is no large change in the HRTEM image, even near the interface. The *c*-Si lattice fringe continues toward the interface. Image contrast becomes vague for the region with roughness. This feature is very consistent with the experimental results. The apparent roughness height is smaller than the actual roughness heights of the models.

On the other hand, for the thick models, the HRTEM image exhibiting strong 110 periodicity is reproduced in the roughened region. The best correspondence of experimental image with simulated ones is obtained when the roughness height is more than 6 ML. For the 8-ML roughness height, a normal *c*-Si lattice image appears above the 110 period image. This is also in good agreement with the experimental observation. Thus, the observed periodic image at the interface can be reproduced by the simulation without assuming the presence of crystalline-phase SiO_2 at the interface.

The simulated optical-diffraction patterns for the {111}-facet models are compared in Fig. 4 with the corresponding experimental results. Although only the streak of each spot along the direction perpendicular to the interface is observed for the thin models, 111 spot splitting is not reproduced. For the thick models, 111 spot splitting is clearly reproduced. This is also evidence of good agreement between simulation and experiment.

For the flat model, neither simulated HRTEM images

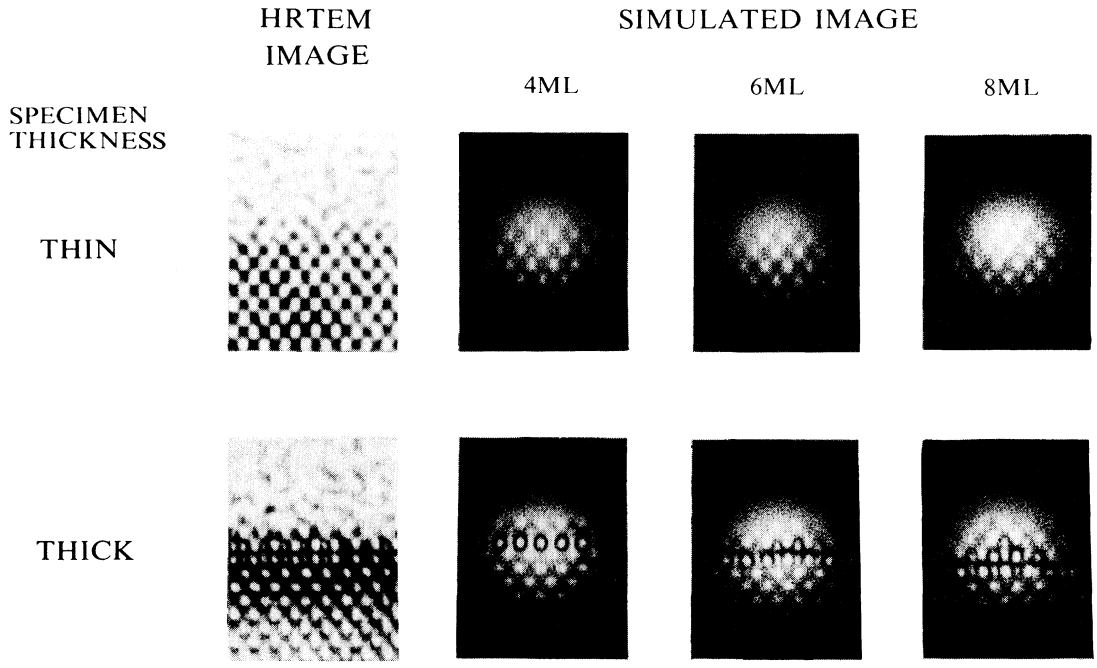

FIG. 3. Result of HRTEM image simulation using the {111}-facet models with different roughness heights (4, 6, and 8 ML).

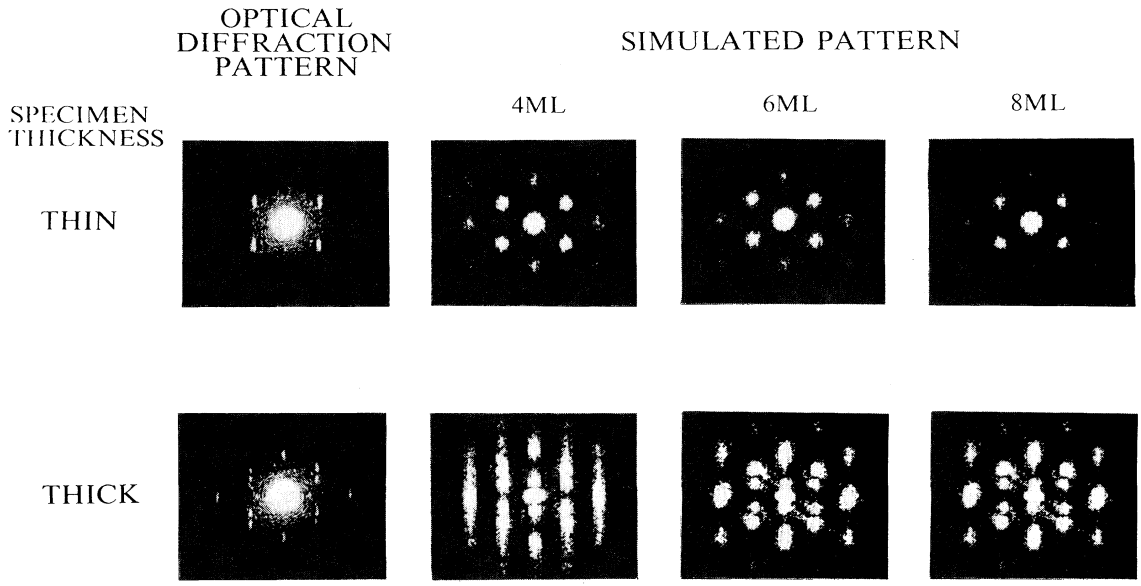

FIG. 4. Result of optical-diffraction-pattern simulation using the {111}-facet model with different roughness heights (4, 6, and 8 ML).

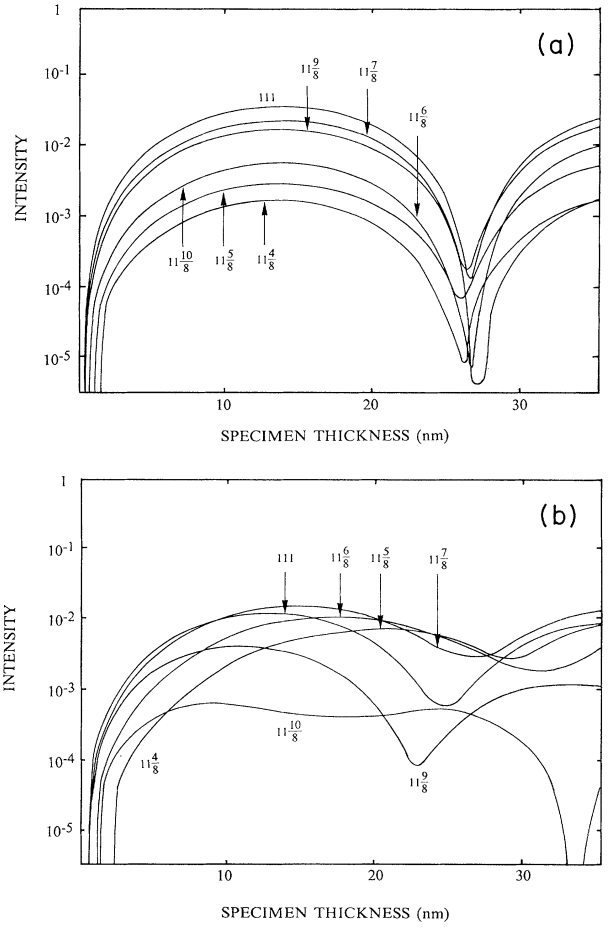

FIG. 5. Specimen-thickness dependence of the scattered-electron wave intensity around the 111 spot of *c*-Si. (a) The flat model; (b) the {111}-facet model with a roughness height of 6 ML.

nor the simulated optical-diffraction pattern show the specimen-thickness dependence observed experimentally. Thus, the HRTEM image with 110 periodicity and 111 spot splitting in the optical-diffraction pattern not only show the effect of the termination of the periodic potential of *c*-Si at the interface, but also the effect of interface roughness on the electron scattering inside the specimen.

In order to examine the cause of these abnormal phenomena for the roughened interface, the specimen-thickness dependence of the scattered-wave intensities has been calculated as shown in Fig. 5 for both the flat model and the {111}-facet model with a 6-ML roughness height. Because our interest is the cause of the 111 spot splitting, the intensity of scattered waves only around the 111 spot is shown. For the flat model [Fig. 5(a)], the intensity of the 111 spot is always stronger than the others. On the other hand, the spatial frequency of the most intense wave changes from 111 to $11\frac{5}{8}$ when the specimen thickness is around 20 nm for the roughened interface [Fig. 5(b)]. The most intense position of the scattered waves around the $11\bar{1}$ spot also changes from $11\bar{1}$ to $11\frac{\bar{10}}{8}$. However, the splitting of the 111 spots observed in optical diffraction does not occur solely via multiple scattering of electrons near the roughened interface. Therefore, the splitting of the 111 spots in the optical-diffraction pattern is brought about by the effects of the electron asymmetrically scattered by the roughened interface and those of the aberrations of the HRTEM optical system.

V. DISCUSSION

As stated above, both the HRTEM image with 110 periodicity and the splitting of 111 spots can be reproduced by the simulation using the model with {111} facets higher than 6 ML. These results are possible indi-

cations that the actual SiO_2/(001)Si interface has Si protrusions delineated by {111} facets or is formed by Si protrusions of similar shape with equally low interface free energy. The driving force that produces the roughness should be the high distortion energy necessary to form the flat (001) interface due to the large lattice mismatch between Si and SiO_2.[14] Modification of the interface morphology would reduce the interface distortion energy by forming the roughness.

We have also shown that the actual height of the roughness is much larger than the roughness observed in the HRTEM images. This result agrees well with the previously reported result obtained by comparing the electrical properties of transistors and HRTEM images.[7]

Exactly when the interface roughness is produced during oxidation has not been clarified by this experiment. In the oxidation process that we used, the specimen underwent dilute Ar oxidation at the end of the oxidation process because the oxygen gas did not abruptly change into pure Ar gas. Since the fresh oxide is produced at a SiO_2/Si interface, the interface structure we observed with use of HRTEM is possibly produced during this cooling-down period. Considering the SiO_2/(001)Si interface structure from the standpoint of the interface distortion energy, it is possible that the size of the Si protrusions increases during the cooling-down period, because there is sufficient time for the interface structure to be modified to lower the interface energy due to the slow oxidation rate in the dilute Ar ambient. Therefore, we believe that control of the cooling-down process must be the key factor in controlling the interface roughness.

The average height of the SiO_2/(001)Si interface roughness of our sample needs to be at least 6 ML. As mentioned previously, atoms just above {111} facets are localized and contribute to HRTEM image formation.[15] The effect of the localization is to lower the apparent height of the Si protrusions. In addition, simulation shows that the interface is observed to be flat for the thick region if the height of the roughness is uniform. In the actual HRTEM images of the SiO_2/(001)Si interface, we can clearly observe the interface roughness. Thus there are Si protrusions of various heights at the actual interface. It is supposed from this evidence that the interface-roughness height is much larger than 6 ML.

VI. CONCLUSIONS

The specimen-thickness dependence of the HRTEM images and their optical-diffraction patterns is reproduced by simulation using the {111}-facet model, in which the interface is formed by Si protrusions delineated by {111} facets. Particularly for thick specimens, the 110 period image observed between the *c*-Si lattice fringe and the SiO_2 granular image is reproduced by the interface roughness without assuming the existence of crystalline-phase SiO_2. We obtain good agreement between simulated and experimental results when the height of the Si protrusions is more than 6 ML. These results suggest that the actual interface would be delineated by {111} facets or formed by Si protrusions of similar shape. Furthermore, the roughness observed in the HRTEM image would be smaller than the actual roughness height.

The interface roughness lowers the interface energy because the flat (001) interface produces high distortion energy while the interface delineated with {111} facets has the lower distortion energy. Thus, the SiO_2/(001)Si interface tends to be roughened even though the initial Si surface is atomically flat.

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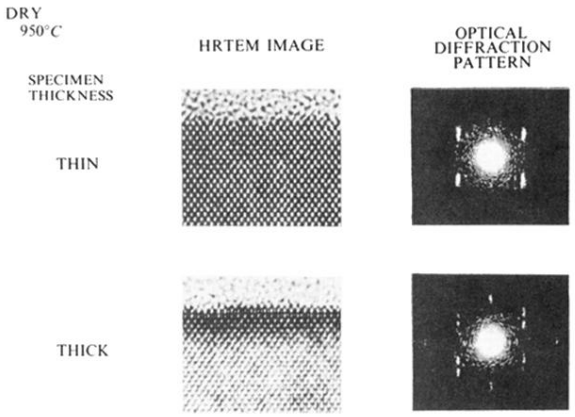

FIG. 1. HRTEM images near SiO_2/(001)Si interface and corresponding optical-diffraction patterns for thin and thick specimens.

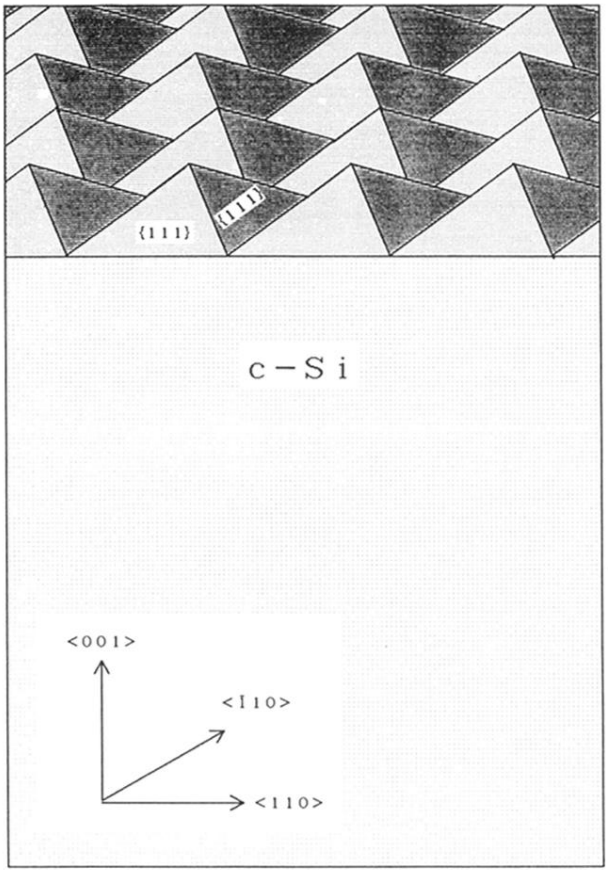

FIG. 2. Schematic drawing of the interface morphology of the {111}-facet model.

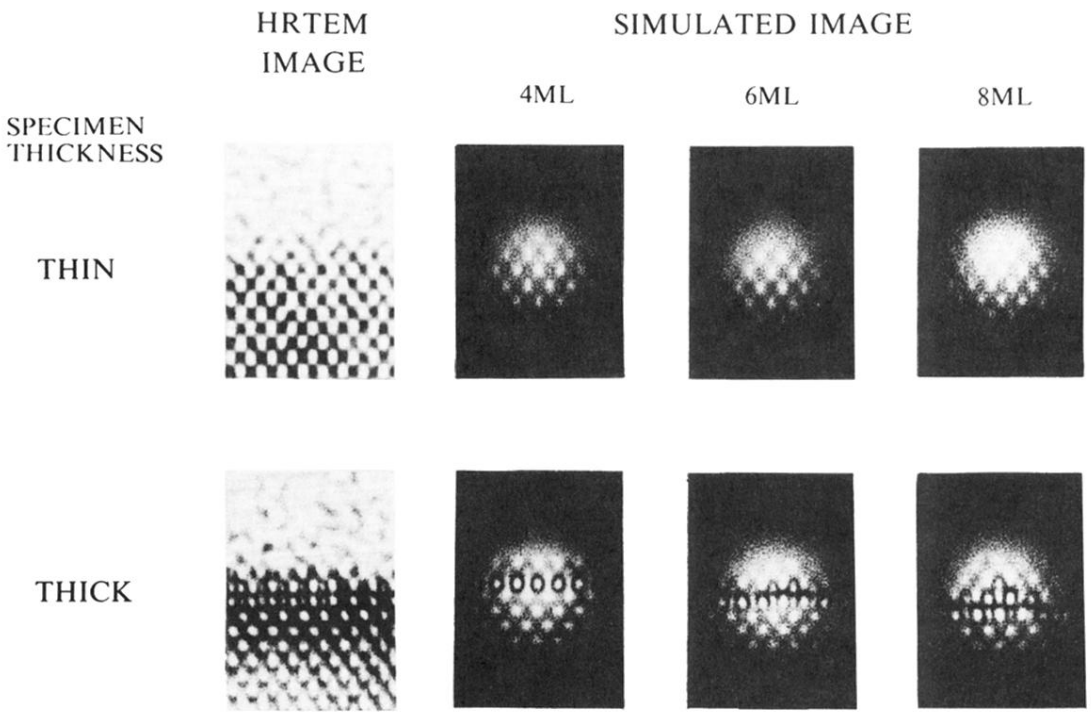

FIG. 3. Result of HRTEM image simulation using the {111}-facet models with different roughness heights (4, 6, and 8 ML).

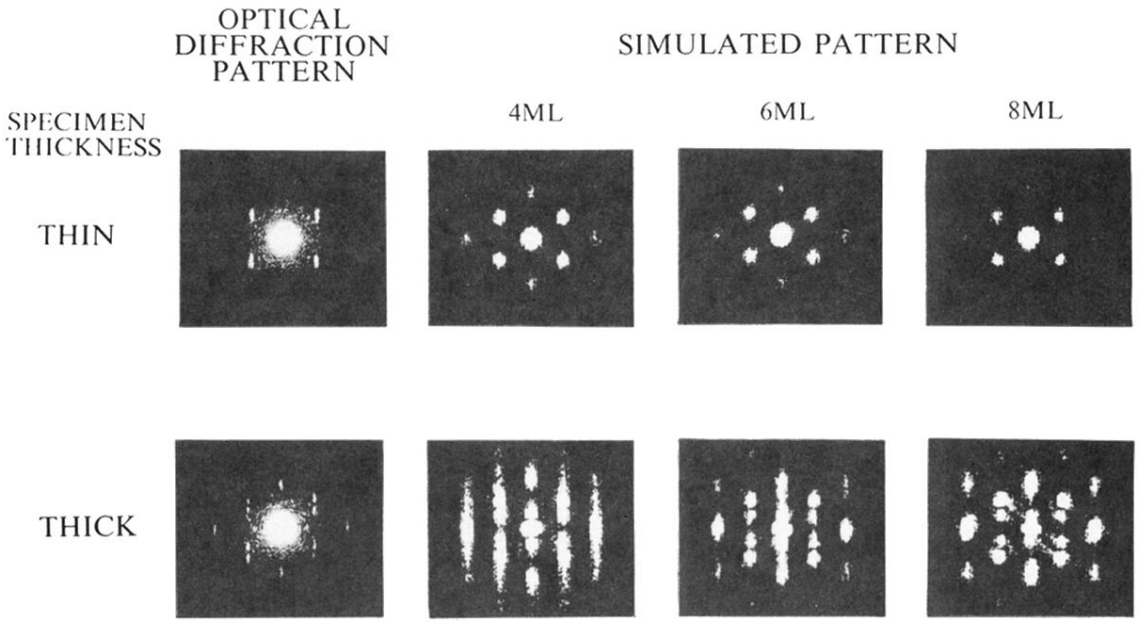

FIG. 4. Result of optical-diffraction-pattern simulation using the {111}-facet model with different roughness heights (4, 6, and 8 ML).