

## Experimental Evidence of a Gaussian Roughness at Si(111)/SiO<sub>2</sub> Interfaces

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With a plan-view transmission electron microscope technique to unambiguously image the “physical” interface position between Si and furnace grown SiO<sub>2</sub> layers, we first show experimental evidence that the height-height autocorrelation function is a Gaussian function at Si(111)/SiO<sub>2</sub> interfaces. With a simple kinetic model, we have found that this Gaussian autocorrelation function is a natural consequence of step motion during silicon oxidation. This result puts interfacial roughness measurements on a firmer foundation in the future. [S0031-9007(98)07785-0]

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Many techniques which are applied to study interfacial roughness rely on models of the interface profile. In particular, roughness at the Si/SiO<sub>2</sub> interfaces, which has great practical importance [1,2], is studied in this way by these techniques. Among them, spectroscopic ellipsometry and second-order harmonic scattering are very indirect and models are needed to fit the experimental data [3]. Although scanning tunneling microscopy (STM) and atomic-force microscopy (AFM) have the advantage of direct imaging [4], the silicon dioxide layer has to be removed first so that the interface is exposed to the probe of STM or AFM, a process which could inflict damages to the interface. STM and AFM images are also often hard to explain in terms of atomic structure. The resolution limit also decides that the frequency range which STM and especially AFM can view falls out of the range that is of real interest and importance [5]. Diffuse x-ray scattering provides a very promising way to study Si/SiO<sub>2</sub> interface roughness with atomic sensitivities [6] and yet it needs assumptions of the statistics of the interface height distribution and the form of the height correlation function [7]. Using a transmission electron microscopy direct-imaging method, we have for the first time experimentally measured the correlation function at SiO<sub>2</sub>/Si(111) buried interfaces without any presumptions on the statistics or the functional form of the height-height correlation function.

A self-affine power law has often been assumed for the height-height autocorrelation function according to the dynamic scaling hypothesis, which has the form [7]

$$\langle [h(x', y') - h(x, y)]^2 \rangle = AR^{2h}, \quad (1)$$

where  $R = [(x' - x)^2 + (y' - y)^2]^{1/2}$ ,  $h$  is the roughness exponent, and  $A$  is a constant. An alternative form of the height-height autocorrelation function has also been suggested [7]:

$$\langle [h(x', y') - h(x, y)]^2 \rangle = 2\sigma^2 \{1 - \exp[-(R/\xi)^{2h}]\}, \quad (2)$$

where  $R = [(x' - x)^2 + (y' - y)^2]^{1/2}$ ,  $\sigma$  is the root-mean-square roughness,  $h$  is the roughness exponent, and  $\xi$  is the correlation length, which sometimes is also called

the effective cutoff length. Equation (2) was suggested based on the idea that the height-height correlation function behaves like a power law near the origin point and vanishes at long distances. Both Eqs. (1) and (2) have been widely used to fit x-ray scattering data to extract roughness parameters [8], and yet surprisingly, there has been no direct experimental evidence to corroborate either of them. In this Letter, we present clear experimental evidence that agrees with Eq. (2).

We previously developed a transmission electron microscopy (TEM) technique to investigate Si/SiO<sub>2</sub> interface roughness [9]. Reflections far from ideal Bragg conditions, which are similar to crystal truncation rods in x-ray scattering, are utilized to reach atomic sensitivities to interface roughness. One advantage of this TEM technique is that both diffraction and image modes can be used to study interface roughness. Hence, the model of roughness needed in fitting diffraction data can be validated through directly imaging the interface roughness on the same area studied under the diffraction mode. Since SiO<sub>2</sub> layers are amorphous, Si/SiO<sub>2</sub> interfaces can be viewed by TEM directly without their removal. This is important for studies on the effect of silicon oxidation on the Si/SiO<sub>2</sub> interface properties. According to electron diffraction theory, the local image intensity shall be a function of the deviation parameter and the local specimen thickness  $t(x, y)$  in the form of [5,10]

$$I(x, y) = \frac{1}{s_{\text{eff}}^2 \xi_g^2} \sin^2[\pi t(x, y) s_{\text{eff}}], \quad (3)$$

where  $s_{\text{eff}} = \sqrt{s^2 + 1/\xi_g^2}$ , and  $s$  is the deviation parameter that measures the distance in reciprocal space between the Bragg reflection and the Ewald sphere and is calibrated from the tilting angle of the sample holder starting from an ideal Bragg condition.  $\xi_g$  is the extinction distance which is inversely proportional to the strength of Bragg scattering. Equation (3) shows that the intensity can be made arbitrarily sensitive to thickness change by increasing the deviation parameter  $s$  through taking images with reflections that are far away from an ideal Bragg condition

[9]. However, the signal-noise ratio is reduced with the increase of the deviation parameter. Hence, noise will set a limit on the minimum detectable thickness change. In this study, background noise was minimized with a very small objective aperture of size  $1 \mu\text{m}$  for dark-field imaging. The final noise-controlled spatial resolution limit is about  $0.4 \text{ nm}^{-1}$  in this study. The maximum deviation parameter we could get was about  $0.6 \text{ nm}^{-1}$  for Si(111).

Because of the size of current metal-oxide-semiconductor devices, a typical silicon dioxide layer is about  $50 \text{ \AA}$  thick, which falls into the thin oxide regime. There are currently two lines on studying silicon oxidation: One is to investigate roughness formed under different oxidation conditions and thermal recipes, often very qualitative due to experimental methods [11]; the other is to study oxidation kinetics in a thin oxide regime. There has been much work trying to build a bridge between these two aspects [12]. However, most of them were done under UHV conditions which are dramatically different from industrial conditions [13]. Although these studies have improved our understanding of silicon oxidation, they are inadequate to answer how roughness is formed under industrial oxidation conditions and how interface can be improved through processing. One motivation of our study is to understand the relation between interface roughness at Si/SiO<sub>2</sub> interfaces formed and oxidation conditions in the thin oxide regime. This problem involves several issues: oxidation kinetics, roughening mechanisms, interface growth, and characterization. These issues are closely intertwined. In this Letter, focusing on interface growth and characterization, we report experimental evidence of a Gaussian interface roughness at Si(111)/SiO<sub>2</sub> interfaces.

P-doped  $1 \Omega \text{ cm}$  Si(111) disks of  $3 \text{ mm}$  diameter were thinned, to about  $50 \mu\text{m}$  at center, from both sides with a VCR mechanical dimpler. A chemical etch of composition  $2(\text{HF}):1(\text{HNO}_3):1(\text{H}_2\text{O})$  was used to prepare electron transparent areas. Note that the high relative concentration of acid to water usefully leads to a macroscopically rougher etching process which generates many useful thin areas for TEM observations. Before observation by a transmission electron microscope, these thinned silicon disks were first oxidized at a temperature about  $1100 \text{ }^\circ\text{C}$  for 5 hours, to form a "sacrificial" oxide of  $\sim 0.5 \mu\text{m}$ . The sacrificial oxide layer was then removed chemically. Two well-controlled starting surfaces were obtained through this process. Then samples were oxidized for 30 min in 1 atmosphere pressure oxygen. Annealing was performed on some samples. [For Si(111), we found annealing at  $900 \text{ }^\circ\text{C}$  does not have an obvious effect on roughness [10,14].] The thickness of the SiO<sub>2</sub> layer was about  $50 \text{ \AA}$ . After the final oxidation, samples were subjected to TEM observations with oxide layers on them.

All transmission electron microscope investigations were done on a Philips CM-12 microscope with an accelerating voltage of  $120 \text{ kV}$ . Dark-field images of Si/SiO<sub>2</sub> interfaces were taken at a series of reflections.

Samples were tilted starting from a  $11\bar{1}$  Bragg reflection,  $1^\circ$  each step so that dark-field images were obtained for a series of deviation parameter values [9]. The starting position and the position of each tilt were recorded. The deviation parameter was calibrated within an error of  $0.06 \text{ nm}^{-1}$  for each reflection. All the images were recorded by a cooled  $1024 \times 1024$  pixel CCD camera and then analyzed by Gatan DigitalMicrograph<sup>TM</sup> software. A modified Wiener filter was used on recorded images to further reduce image noise [10]. Interface heights are calibrated from the deviation parameter value based on Equation (3). An interface height map finally obtained includes all the information regarding interface roughness within a frequency band sampled by the microscope. The root-mean-square roughness and the height-height correlation function were directly retrieved from this map with DigitalMicrograph<sup>TM</sup>. The frequency range in this study was from  $0.001$  to  $0.4 \text{ nm}^{-1}$ . Through the whole process, no models or assumptions on the interface roughness were needed.

Figure 1 shows a typical image of Si(111)/SiO<sub>2</sub> interfaces. Those fringes are height contours. They not only indicate the position of thickness changes but also tell the height at each position. For convenience, we measured a normalized height-height autocorrelation function from the interface height image, which is related to the height-height correlation in Eq. (2) as

$$C(R) = \frac{\langle h(R)h(0) \rangle}{\sigma^2} = 1 - \frac{1}{2\sigma^2} \langle [h(x',y') - h(x,y)]^2 \rangle, \quad (4)$$

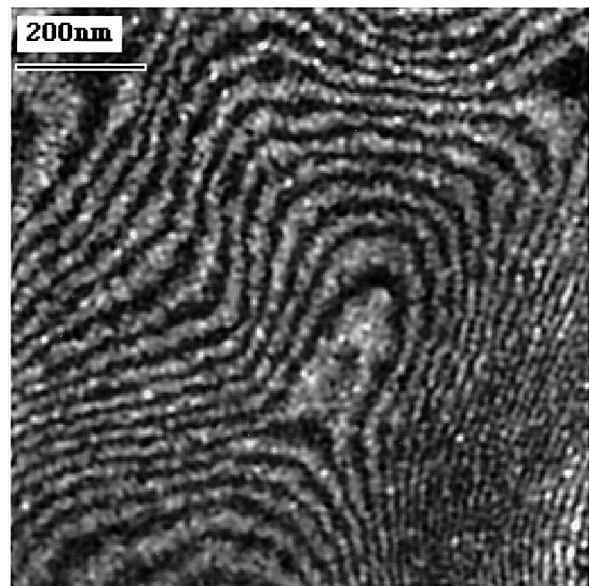


FIG. 1. A TEM dark-field image of one area of Si(111)/SiO<sub>2</sub> interfaces. Thickness fringes indicate interface heights and locations. An interface height map can be obtained with information in this image. The root-mean-square roughness, the correlation length, and the height-height correlation function can then be measured.

where  $R = [(x' - x)^2 + (y' - y)^2]^{1/2}$ . Hence, the assumption that the height-height correlation function has the form of Eq. (2) is equivalent to the assumption that the height-height autocorrelation function takes the form of

$$C(R) = \exp[-(R/\xi)^{2h}]. \quad (5)$$

One of the retrieved autocorrelation functions is shown in Fig. 2. Figure 3 gives our plotted data in a  $\ln\{-\ln[C(R)]\} \sim \ln R$  plot with error bars, excluding the trivial origin point. Those data were taken from different areas on a sample and often along different directions. It is shown that all data fall on the same line within the range of experimental error, which means that the height-height autocorrelation function takes the form of Eq. (5). Hence, for the first time, we obtained strong experimental evidence that the height-height correlation function for interface roughness at Si(111)/SiO<sub>2</sub> interfaces has the form of Eq. (2). Furthermore, from the linear curve we got we measured the roughness exponent as  $0.99 \pm 0.01$ . Hence, we showed a Gaussian roughness at Si(111)/SiO<sub>2</sub> interfaces. Hereby it also merits mention that our data cannot be put into other functional forms that have been suggested, such as self-affine power law or Lorentzian function.

A model has been developed to explain this roughness [14], in which a Langevin equation with the form of

$$\frac{\partial h(x, y)}{\partial t} = \mu \nabla^2 h(x, y) + \eta(x, y, t) \quad (6)$$

is used to model interface growth during silicon oxidation, where  $t$  is the oxidation time,  $h(x, y)$  is the interface height,  $\mu$  is the diffusion coefficient, and  $\eta(x, y, t)$  is a noise term describing the oxidation of silicon atoms. The first term on the right-hand side of Eq. (6) addresses the thermodynamic smoothing of step motion and the second term corresponds to the roughening process of silicon oxidation. The noise term has to satisfy the following conditions to ensure Eq. (6) yields the right equilibrium probability distribution:

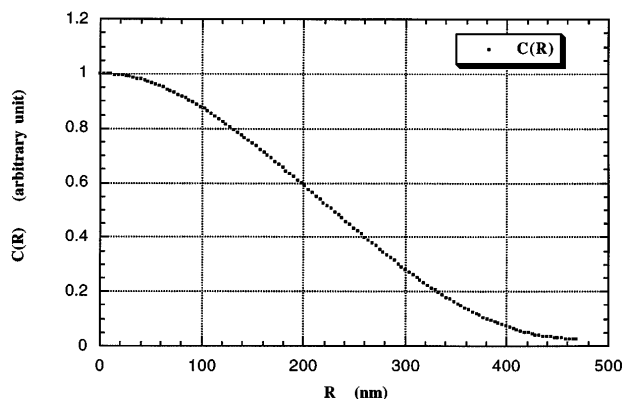


FIG. 2. A set of data points of measured height-height autocorrelation function from our Si(111)/SiO<sub>2</sub> interfaces shows a Gaussian-like behavior.

$$\langle \eta(x, y, t) \rangle = 0, \quad (7)$$

$$\langle \eta(x, y, t) \eta(x', y', t') \rangle \propto \lambda \delta(x - x') \delta(y - y') \delta(t - t'),$$

where  $\langle \dots \rangle$  is an ensemble average and  $\lambda$  is the oxidation rate. Solving Eq. (6) with condition (7), we obtain the normalized height-height autocorrelation function  $C(R)$  as

$$C(R, t) = \exp\left(-\frac{R^2}{8\mu t}\right), \quad (8)$$

where  $t$  is the oxidation time [15]. The physics behind this solution is the following: in an extreme case of oxidation without step motion, which means  $\mu = 0$ , spatially uncorrelated interfaces would be generated; hence the correlation length is zero. It is exactly the case described by the random deposition model [16]; however, step motions at Si(111)/SiO<sub>2</sub> interfaces smoothen the interface and introduce spatial correlation in the interface roughness. Consequently, a roughness with a Gaussian height-height autocorrelation function results. Therefore, the Gaussian roughness we measured is a natural consequence of step motions described by the first term on the right-hand side of Eq. (6). It is worth mentioning that previous Monte Carlo simulations on similar systems were consistent with our analytical solution (8) [17]. The physics picture we present here also agrees with our experimental results on an annealing effect [14].

It must be noticed that images in TEM plan-view mode actually record two overlapping interfaces. However, our way of making samples determines that the two interfaces of a sample are statistically identical and yet uncorrelated. In this case, it can be easily shown that the stochastic nature of each interface will remain after the overlapping of these two interfaces. Hence, our method of measuring roughness is still valid although we are actually looking at two overlapping interfaces. There is also a small deviation from the Gaussian form for  $R$  smaller than 350 nm. This deviation may come from the fact that there

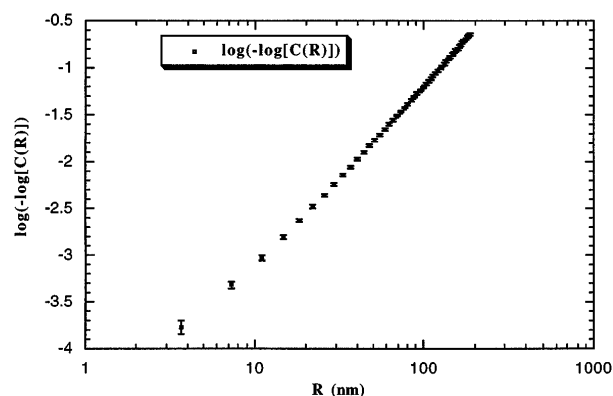


FIG. 3. A plot of  $\log\{-\log[C(R)]\}$  as a function of  $R$  in a log scale. All the data points fall on a line indicating that the height-height correlation function on Si(111)/SiO<sub>2</sub> interfaces takes the form of Eq. (2). Further measurement on  $h$  shows a Gaussian roughness.

is always a high frequency cutoff when Fourier transform is carried out in the process of measuring the correlation function, and, hence, there shall be a deviation for small distance.

Through back-transformation, Salditt *et al.* reported a way to measure the height-height correlation function with diffuse x ray without a presumption of the functional form [18]. However, the statistics of the interface height distribution was still assumed to be a Gaussian form. In this paper, we have not touched the nature of the statistics of the interface height distribution. However, we point out that we can even further investigate the statistics by measuring higher order moments of the interface height difference from our images. Because of the length limit, it shall be a topic for another paper.

Our study also implies that dynamics is the origin of interface roughness growth. It is natural to infer that a different dynamic process will yield roughness with different characters. On the other hand, roughness also may serve as a window to processing dynamics. It may be a new approach to gain insight on oxidation kinetics from our measured roughness in the future.

In conclusion, we validated Eq. (2) as a correct form for the height-height correlation function of the interface roughness of Si(111)/SiO<sub>2</sub>. The roughness exponent was measured as  $0.99 \pm 0.01$ , which means a Gaussian roughness. By modeling interface growth during silicon oxidation, we show that this Gaussian roughness is a natural consequence of step motions at the interface. Our result provides a firmer foundation for roughness measurements at Si/SiO<sub>2</sub> interfaces in the future.

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