

Roughening in Plasma Etch Fronts of Si(100)

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A novel etch front roughening phenomenon has been observed in the plasma etching of Si(100). The morphology exhibits a network structure with holes which coarsen with etch time, and a wavelength selection with a characteristic spatial frequency decreasing with time. The average local slope is invariant while the vertical roughness grows as $w \sim t^\beta$, with $\beta = 0.91 \pm 0.03$. We suggest a nonlocal Langevin equation based on the redistribution of the reactant gas flux by local morphological features. Numerical calculations give results consistent with our experiments. [S0031-9007(99)09388-6]

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Kinetic roughening of film growth/etch fronts has attracted considerable attention in recent years. Surface and interface roughness controls many important physical and chemical properties of films. Very often the phenomenon of growth/etch front roughening occurs under far-from-equilibrium conditions. Recently, by employing the concept of dynamic scaling combined with analytical treatments including the use of stochastic differential equations, considerable progress has been made in the basic understanding of this phenomenon [1,2]. One outstanding outcome of the dynamic scaling hypothesis is the existence of *universality* in which the essential features of the roughness evolution depend only on certain symmetries and the dimensionality of the system but not on the detailed interactions. Classification of the universality classes of roughness evolution is a main focus of many researchers.

Unlike the study of growth front roughening, such as in molecular beam epitaxy, very little work has been reported on plasma etch front roughening [3–5] despite the widespread use of this technique. Plasma etching is a major tool in thin film patterning, which is an important process for microelectronic fabrication. For plasma etching of a Si surface, for example, a reactive gas such as SF₆, or CF₄ mixed with a small percentage of O₂ or SF₆ is used. The chemistry of etching is well known [6]. In the plasma etching configuration, the etching (removal of Si from the surface) is due to the chemical reaction between the etchant gas and the sample and is not due to the physical sputtering of the sample. Active fluorine atoms are released from the plasma and react with the Si surface to form volatile fluoride species. Unlike molecular beam epitaxy, the etchant (F atoms) reaches the surface from all directions. The etch front is amorphous (containing fluorinated Si) and the morphology is isotropic.

In an earlier experiment [5], it was observed that the interface width (root-mean-square roughness) w of the etch front increased linearly with etch time, and the lateral correlation length ξ grew as a power law in time, $\xi \sim t^{0.66}$. In this Letter, we show that, when we extend the measurements to a much larger length scale both in the vertical (interface width) and lateral (lateral correlation length) di-

rection, we arrive at a very different conclusion not only quantitatively but also qualitatively. A possible novel universality class of roughening may emerge.

The experiments were conducted in a Technical PlanarEtch II. The high-frequency (30 kHz) plasma generator can provide up to 500 W of continuous power, dynamically regulated to maintain preset power levels during operation. The samples used were commercial *n*-type Si(100) wafers with 10–16 Ω cm resistivities. The etching was performed in the etchant gases, which are CF₄ (39.0 ± 1.0 SCCM) and (SCCM denotes cm³/min at STP) and 4% O₂, under a pressure of 300 ± 5 mTorr. The plasma power used was 102 ± 2 W. Under such conditions, the etching is due to chemical reaction between the etchant gas and the inserted sample, and the etching is isotropic [6]. The samples were etched over a time period ranging from 1 to 100 min. For each run, the etching always started with a fresh Si wafer at room temperature, and then proceeded without interruption to a predetermined time. The average etch rate was 0.15 ± 0.02 $\mu\text{m}/\text{min}$.

An atomic force microscopy (AFM) image of 256×256 data points was obtained for each sample after etching. Some representative 10×10 μm^2 AFM images of the etched Si(100) samples for etch durations of $t = 20, 30, 40,$ and 60 min are presented in Fig. 1. The surface morphology for $t = 20$ min is relatively flat, with randomly distributed holes and grains. With increasing etch time, the holes become bigger and then coarsen together to form bigger holes as seen for $t \geq 30$ min. The similarity in the surface morphologies for $t = 30, 40,$ and 60 min implies that the etched surface possesses a self-similar structure in time scale.

To examine the possibility of dynamic scaling behavior, we have performed a quantitative study of the morphology of the etch front. In order to obtain reliable statistics, it is required that the scan size extend over at least 10ξ , where ξ is the lateral correlation length [7]. Since the feature size of the morphology grows rapidly, the actual data were taken from AFM images with scanning areas ranging from 1×1 μm^2 up to 70×70 μm^2 . The

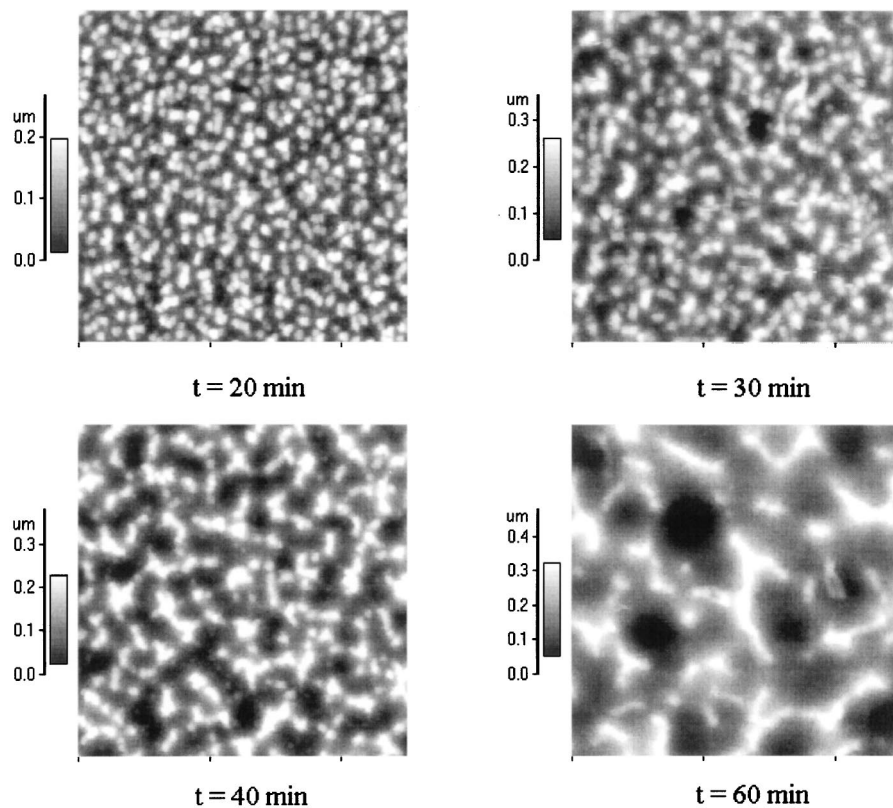


FIG. 1. AFM images of plasma etched Si(100) substrates at room temperature for $t = 20, 30, 40,$ and 60 min. The scan sizes are $10 \times 10 \mu\text{m}^2$.

larger area scans were performed on samples that were etched for longer times. In Fig. 2 we plot, in log-log scale, the equal-time height-height correlation functions $H(r, t)$, defined as $H(r) = \langle [h(\vec{r}) - h(0)]^2 \rangle$ (a function of the magnitude of the position \vec{r} for a fixed value of t). Here, $h(\vec{r})$ is the surface height at position $\vec{r} = (x, y)$ on the surface. The notation $\langle \dots \rangle$ means an average over all possible choices of the origin, and an ensemble average over all possible surface configurations. The scaling hypothesis requires that $H(r) \propto r^{2\alpha}$ for $r \ll \xi$, and $H(r) = 2w^2$ for $r \gg \xi$ [1,2]. Here, ξ is the correlation length, within which the surface heights of any two points are correlated, and α is the roughness exponent. The value of α , which lies between 0 and 1, describes how wiggly the surface is. We found that the roughness exponent α is 0.96 ± 0.06 . The interface width w versus t is plotted in Fig. 3. We found that $\beta = 0.91 \pm 0.03$, which is close to the β value reported in Ref. [5]. The average local surface slope ρ , which can be obtained from the intersection of $H(r)$ with the vertical axis shown in Fig. 2 [8], was found to be 0.19 ± 0.06 . The lateral correlation length ξ can be determined from the autocorrelation function $C(r) = \langle h(r)h(0) \rangle$, in which ξ is defined through $C(\xi) = C(0)/e$, where e is the base of the natural logarithm. Figure 3 also shows ξ as a function of the etching time t , which gives the reciprocal dynamic exponent $z^{-1} = 0.95 \pm 0.08$. This result is very different from the value 0.66 reported in Ref. [5].

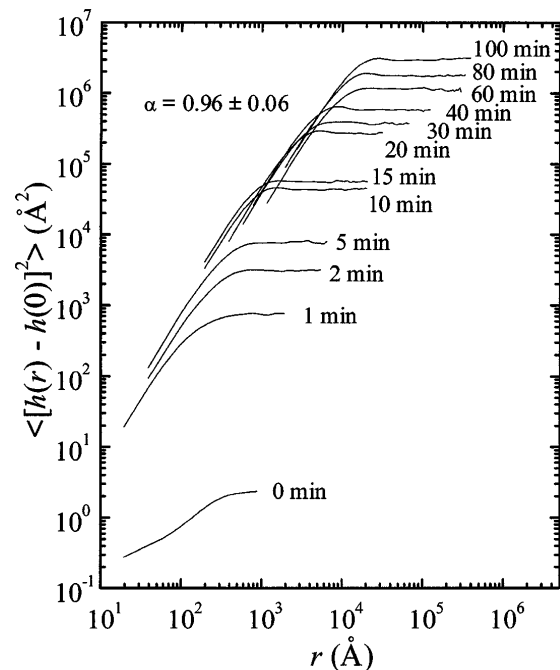


FIG. 2. The height-height correlation function $H(r, t)$ as a function of the distance r is plotted for different etch times. All $H(r, t)$ approximately overlap in the short-range spatial scaling regime ($r \ll \xi$) and the slope gives 2α and $\alpha = 0.96 \pm 0.06$. The intersections of the H curves with the vertical axis have almost the same value which is related to the average local surface slope $\rho = 0.19 \pm 0.06$.

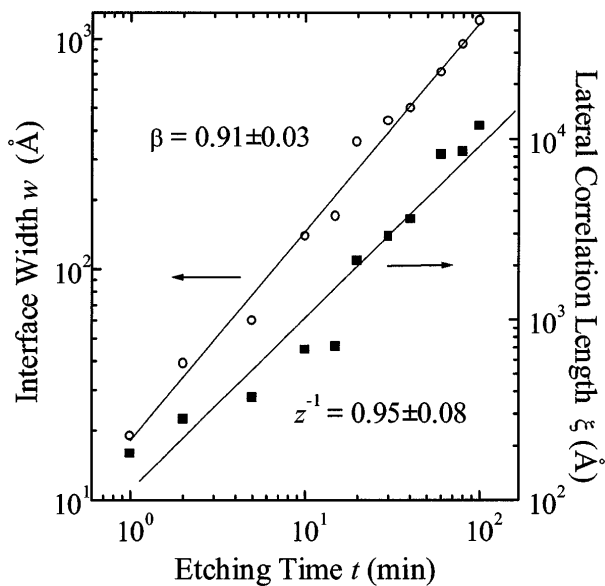


FIG. 3. A plot of interface width w and lateral correlation length ξ versus etch time t . The open circles represent the interface width, and the filled squares represent the lateral correlation length. The growth exponent β and the inverse of the dynamic exponent z are 0.91 ± 0.03 and 0.95 ± 0.08 , respectively.

It is seen that our experimental exponents α ($= 0.96 \pm 0.06$), β ($= 0.91 \pm 0.03$), and z ($= 1.05 \pm 0.09$) are consistent with the dynamic scaling relationship, $z = \alpha/\beta$. However, the exponents obtained from our experiments are different from those predicted by existing universality classes [1,2]. The near-unity scaling exponents imply that the surface morphology, in this case, is not self-affine. It is remarkable that, while the interface width and correlation length have grown more than 2 decades, the local structure, defined by the value of α and the average magnitude of the local surface slope ρ , is invariant. This is seen in Fig. 2. For small r , the slopes of $H(r)$ are about the same, and all $H(r)$ curves can be extrapolated to intersect at almost the same point on the vertical axis [8], indicating that ρ is a constant.

We also analyzed the power spectra of the surfaces. Figure 4 shows the circularly averaged power spectra obtained from AFM images for $t = 20, 40$, and 100 min. A clear maximum value of the power spectrum, located at a nonzero position k_0 , indicates the existence of wavelength selection. As the etch time increases, k_0 decreases, illustrating the coarsening process. We observed that $k_0 \sim t^{-\gamma}$, where $\gamma = 1.3 \pm 0.3$.

These results are in contrast to those obtained from the evaporation of Si on Si(111) [9] and ion sputtering of Si(111) [10], where neither an invariant local slope nor wavelength selection were observed. We must point out that, in the present experiment, the action (etching) is perpendicular to the local surface rather than the average surface (in contrast to evaporation deposition or ion sputtering). The local slope selection and wavelength selection observed in our experiment are similar to the

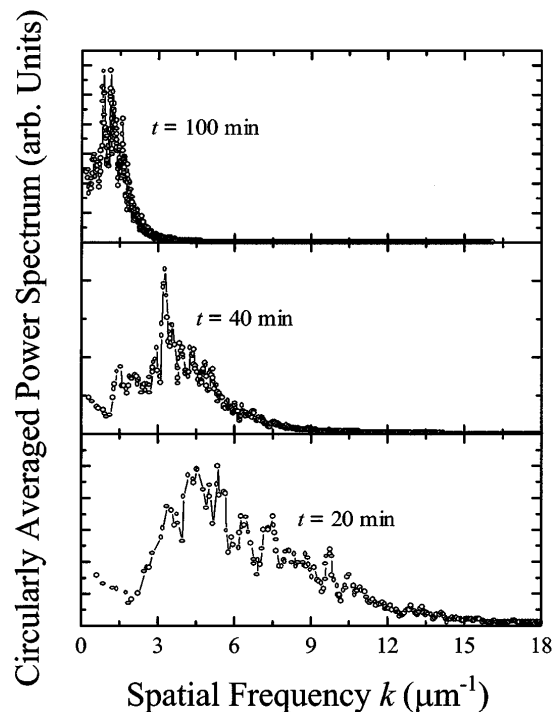


FIG. 4. The circular average of the power spectra analyzed from AFM images of the Si(100) surface etched for $t = 20, 40$, and 100 min.

results of mound formation caused by the step barrier [11]. However, in mound formation, the observed γ value was between 0.16 and 0.26, and β was less than 0.5 [11]. These values are much smaller than what we obtained in the present work. The value of α and z , and the unusually high growth exponents β and γ , suggest that this etching process might belong to a new universality class.

Recently, Hwang *et al.* [12] performed a detailed study of the scattering of F atoms on a fluorinated surface. They showed that a redistribution of the F flux occurred at the surface due to scattering and trapping desorption. We consider here a nonlocal model that includes the redistribution of the etchant at the surface as shown in Fig. 5. The etchant particle from the gas phase hits the surface at point A with a certain sticking coefficient s . The etchant particle either sticks on the surface and

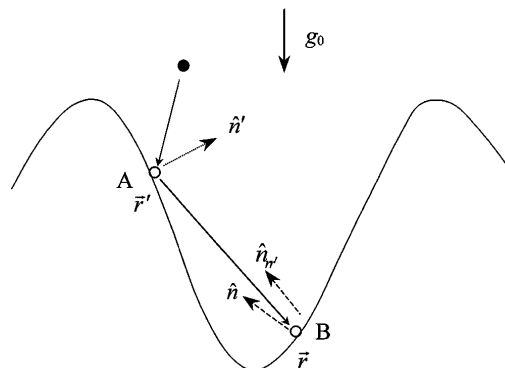


FIG. 5. A schematic showing the etchant redistribution.

reacts with the surface or it is reemitted from the surface according to a certain reemission mode (for details, see Ref. [13]), and reaches point B . Therefore, the flux of the etchant would redistribute according to the reemission mechanism. If the reemission plays an important role, clearly we can expect from Fig. 5 that the valley of the surface will receive more redistributed etchant than the peak of the surface. Therefore, the valley will have a higher etching rate than the peak, and that causes the

instability of the morphology evolution. The equation for plasma etch fronts can be written as

$$\frac{\partial h}{\partial t} = \nu \nabla^2 h - k \sqrt{1 + (\nabla h)^2} g_r(\vec{r}, t) + \eta(\vec{r}, t), \quad (1)$$

where the first term on the right-hand side is due to the surface tension, k is a constant associated with the reaction rate and sticking coefficient, $\eta(\vec{r}, t)$ is the intrinsic white Gaussian noise, and $g_r(\vec{r}, t)$ is the reemitted flux, satisfying the following equation (to the first order) [4]:

$$g = g_0 + g_r = g_0 + (1 - s) \int Z(\vec{r}, \vec{r}', t) g_0(\vec{r}', t) \frac{(\hat{n}_{r'} \cdot \hat{n}) P(\vec{r}', \vec{r}, t)}{(\vec{r} - \vec{r}')^2 + (h - h')^2} dA', \quad (2)$$

where g_0 is the incident flux from the plasma, $Z(\vec{r}, \vec{r}', t)$ is the self-shadowing factor, and \hat{n} and \hat{n}' are the surface normals at \vec{r} and \vec{r}' , respectively. Also, $\hat{n}_{r'}$ is the unit vector pointing from the point on the surface at \vec{r} to the point on the surface at \vec{r}' (see Fig. 5). $P(\vec{r}', \vec{r}, t)$ is the redistribution function. For thermal reemission, $P(\vec{r}', \vec{r}, t) = (\hat{n}_{r'} \cdot \hat{n}')/\pi$. The second term on the right-hand side of Eq. (2) integrates over the whole surface except the position \vec{r} . In fact, due to the self-shadowing effect, the main contribution to the integration is within a radius on the order of $\xi(t)$.

We performed a numerical integration of Eq. (1) for single reemission etching, i.e., the first order iteration. A size of 128×128 square lattice was used in this calculation. For thermal reemission, the scaling exponents we obtained were $\alpha = 0.98$, $\beta = 0.96$. [Because of the small system size in our numerical calculation, we could not extract the value of γ reliably. Additional work on a large size (1024×1024) Monte Carlo simulation reveals that the system exhibits a clear wavelength selection and gives $\gamma \approx 1$, which is very close to our experimental result [13].] The reemitted gas plays a very important role in roughening. In the calculation, the noise is turned off after a short period, and the system evolves deterministically. However, the noise should be important only in the early stages of evolution because, for late times, unstable growth dominates the process. Another factor in this model is the method of reemission; two possibilities are reflection (particles bounce off the surface) and diffuse reemission (according to a particular reemission distribution function) [13]. Our numerical calculations indicate that the exponents obtained do not depend significantly on the mode of reemission or the distribution function, but the morphology does depend on these factors, which implies that the proposed mechanism could be universal.

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