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Anomalous dynamic scaling on the ion-sputtered Si(111) surface

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The time evolution of an ion-sputtered Si(111) surface was investigated between 300 and 650 °C using the high-resolution low-energy electron diffraction technique. Below 450 °C, a (1×1) rough phase shows an anomalous dynamic-scaling behavior on the short-range scale where the measured height-height correlation has a form of $\sim\ln(t)r^{2\alpha}$, which grows in time with a roughness exponent $\alpha=1.15\pm0.08$. Such a behavior is consistent with the prediction by a recent dynamic-scaling theory. Above 530 °C, we found that the (7×7) phase does not undergo roughening evolution. The dramatic morphology change with temperature indicates a dynamic phase transition.

I. INTRODUCTION

The study of film-growth morphology has had a long history. Recent interest in far-from-equilibrium growth has been stimulated very much by the theoretical development of the dynamic-scaling approach.[1] In dynamic scaling, the morphology of a growing interface is proposed to have both short-range space scaling and long-range time scaling. The space scaling is characterized by a scale-invariant height-height correlation, $\langle[Z(\mathbf{r},t)-Z(0,t)]^2\rangle\sim r^{2\alpha}$, where $Z(\mathbf{r},t)$ represents the surface height of the planar position $\mathbf{r}$ at growth time t, and the exponent α describes surface roughness. The time scaling shows that the surface roughness grows with time in a form of power law $w\sim t^{\beta}$, where w is the interface width (root-mean-square surface height fluctuation). Removal of materials from a surface is perhaps as important as deposition on a surface from both theoretical and practical points of view. Thus far very little work has been reported on the study of etched rough surfaces using the dynamic-scaling approach. Recently, two very important experiments[2,3] showed certain scaling features during sputter etching processes. However, the full dynamic scaling behavior in both time-evolution and scale-invariant characteristics has not been completely demonstrated in these measurements. In this paper, we report a study of the ion-sputtered Si(111) surface using the high-resolution low-energy electron-diffraction (HRLEED) technique. The time evolution of the sputter-induced roughening was observed to show a non-self-affine scaling behavior. The phenomenon is explained by a recent dynamic-scaling theory.

II. EXPERIMENTS

The experiment was performed in an UHV chamber equipped with a HRLEED system,[4] an Auger-electron spectrometer (AES) and a sputter ion gun. A clean and long-range ordered Si(111)-7×7 surface was prepared in UHV by flashing the sample up to 1250 °C. The sputtering was performed with a normal-incident 0.5-keV Ar^+-ion bombardment at sample temperatures between 300 and 650 °C. The Ar^+ flux was estimated to be 0.2 $\mu\mathrm{A/mm}^2$ for an Ar pressure of $\sim2.2\times10^{-6}$ torr, with an estimated sputter-etching rate of ~1 layer/min. The sputtered surface morphology was measured by HRLEED after the sample was quickly quenched to room temperature. Every sputtering started from a flat surface that had a sharp 7×7 diffraction pattern, and no step could be detected within the resolution of the HRLEED.

III. RESULTS

A. Temperature-dependent morphologies during sputtering

We first studied the sample temperature-dependent sputtering processes in which different HRLEED diffraction patterns were observed. At $300\,^\circ\mathrm{C}<T\leq450\,^\circ\mathrm{C}$, we found a (1×1) diffraction pattern. (Below 300 °C, the HRLEED intensity was too low to be measured.) Between 450 and 530 °C, the diffraction showed a mixture of (1×1) and $(\sqrt{3}\times\sqrt{3})R30^\circ$ patterns. Above 530 °C, (7×7) spots showed up and their intensities were enhanced significantly at higher temperatures. The $(\sqrt{3}\times\sqrt{3})R30^\circ$ structure has been observed previously on highly stepped Si(111) surfaces with terraces narrower than the width of a (7×7) unit cell.[5] This suggest that a highly stepped surface may be produced by sputtering at temperatures lower than 530 °C. More quantitative evidence for the existence of steps was shown in the measured full width at half maximum (FWHM) of the HRLEED line shapes as a function of the incident electron-beam energy, E. Figure 1 is the plot of the FWHM of the (00) beam line shape scanned along the $[11\bar{2}]$ direction as a function of $k_\perp c$ at different sample temperatures with a fixed sputtering duration $t=20$ min. $k_\perp$ is the diffraction momentum transfer perpendicular to the surface, and c is the surface layer spacing (3.135 Å) of Si(111). At temperatures not greater than 450 °C, as shown in Figs. 1(a)–1(c), the plot exhibits a significant oscillatory behavior with the minimal positions at $k_\perp c\approx6\pi$, 8π, and 10π, and maximal positions at 7π and 9π. It is well known that such an oscillation is the result of the diffraction from a stepped surface,[6] where the constructive (in-phase) diffraction occurs at $k_\perp c=2m\pi$ and the

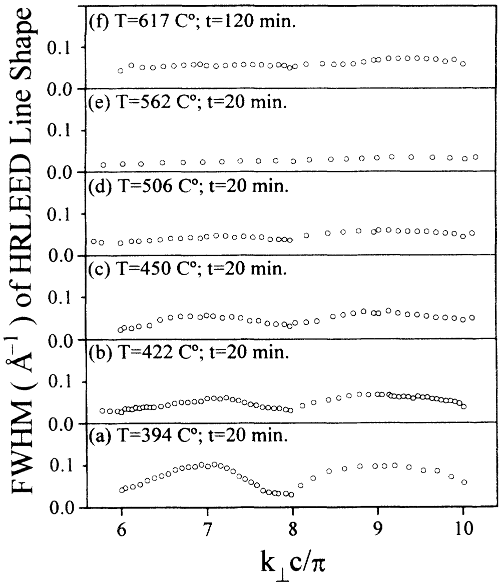

FIG. 1. The FWHM of the (00) beam along the $[11\bar{2}]$ direction is plotted as a function of $k_\perp c$ at different sputtering temperatures for a fixed 20-min sputtering [except (f), where 120 min was used]. The uncertainty in the measurement is comparable with the size of the data points.

destructive (out-of-phase) diffraction occurs at $k_\perp c = (2m+1)\pi$ with $m = 0, 1, 2, \ldots$. The oscillation amplitude is proportional to the surface step density.[6] As shown in Fig. 1, the lower the sputtering temperature, the higher the step density. For example, the average terraces are about 70, 120, and 150 Å at 394, 450, and 506 °C, respectively. The oscillation disappears at 562 °C [Fig. 1(e)]. At 617 °C [Fig. 1(f)], the nonoscillatory behavior persists even for a longer sputtering time, $t = 120$ min.

An immediate conclusion is that the sputtering produces different morphologies at different temperatures. Above 530 °C, the high atomic mobility and the large terrace size allow the surface atoms to rearrange themselves to form the (7×7) phase. In contrast, at $T \leq 450$ °C, the slow diffusion and the narrow terraces inhibit any reconstruction. At the intermediate temperatures between 450 and 530 °C, the metastable $(\sqrt{3}\times\sqrt{3})R30°$ reconstruction becomes favorable. The dramatic morphology change with temperature suggests a dynamic phase transition driven by dynamic and geometric conditions.

B. Time-dependent morphologies during sputtering

The above temperature-dependent phenomenon is consistent with the previous low-energy ion-sputtering experiment,[7] where, at 550 °C, large Si(111)-7×7 domains were observed during layer-by-layer sputtering, while at 250 °C a disordered phase with no long-range reconstruction eventually replaces the (7×7) phase. This earlier experiment focused on the initial stage of the removal of the first few monolayers, while our study concentrated on the morphology evolution during long-time sputtering. We studied the time-dependent sputtering process where the sample was held at a fixed low temperature and each sputtering was performed from an initially flat surface. Figure 2 shows the (00) beam line shapes as a function of sputtering time at $T = 450$ °C. Diffraction was measured at $E = 47.0$ eV, which corresponds to an out-of-phase condition $k_\perp c \approx 7\pi$. At $t = 0$ (before sputtering), the sharp line shape contains only a δ-like profile (convoluted with the HRLEED resolution function), showing that the surface is step free within the resolution of the instrument which has a maximum resolvable terrace size of at least 1000 Å in real space.[8] At the sputtering time of $t = 10$ min, the growing diffuse component broadens the line shape, while the δ component is significantly reduced. The type of "δ+diffuse" line shape is characteristic of a rough surface with a finite interface width w.[9] Due to the coherent electron diffraction, a sharp central δ peak appears when the surface looks smooth on the long-range scale, and it decays quickly with increasing w as $\sim e^{-w^2}$. Since the surface is rough on the short-range scale, a broad diffuse line shape also shows up, resulting from interference from steps.

The existence of the δ component at $t = 10$ min indicates that at the initial stage the sputtered surface has a small interface width and is not rough enough to have a multilayer step structure. However, the situation quickly changes at a longer sputtering time. Starting at $t = 30$ min, as shown in Fig. 2, the δ component completely disappears and, most importantly, the remaining diffuse profile broadens and its intensity decreases continuously with increasing sputtering time. The evolution does not seem to be saturated even at a much longer time, $t > 120$ min. Similar characteristics can also be observed at other

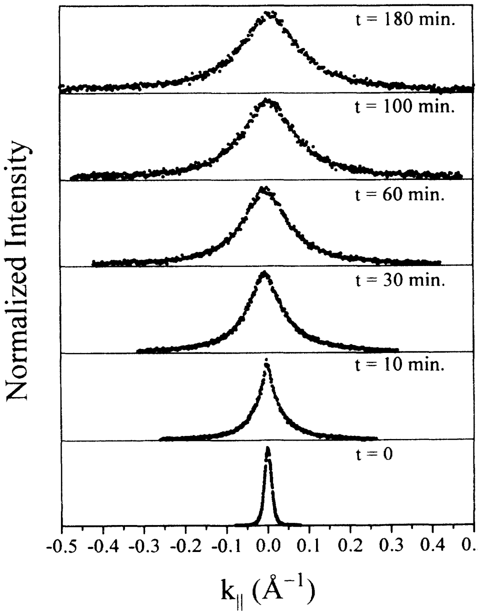

FIG. 2. The normalized angular profiles of the (00) beam scanned along the $[11\bar{2}]$ direction at $k_\perp c \approx 7\pi$ are plotted as a function of sputtering time at $T = 450$ °C, where $k_{\parallel}$ is the diffraction momentum transfer parallel to the surface. The intensity has been scaled up by the factor shown near the peak of each profile.

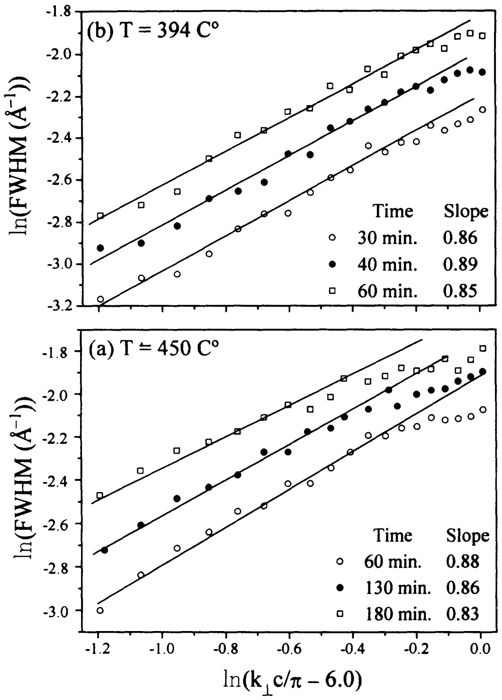

FIG. 3. The ln-ln plots of FWHM vs $k_{\perp}c$ along the $[11\bar{2}]$ direction for the (00) beam at different sputtering times are shown for (a) $T=450\,^{\circ}\mathrm{C}$ and (b) $T=394\,^{\circ}\mathrm{C}$. The diffraction condition $k_{\perp}c$ ranges from 6.3π to 7.0π. The uncertainty is about the size of the data points.

diffraction conditions. Figure 3(a) is the plot of the FWHM vs $k_{\perp}c$ at 450 °C for different sputtering times, with $k_{\perp}c$ varying from 6.3π to 7.0π. The same behavior occurred at a lower temperature of 394 °C, as shown in Fig. 3(b). The FWHM shown in Fig. 3 increases with sputtering time. Such a time evolution can be demonstrated more clearly by further plotting the curve of FWHM vs t at $k_{\perp}c=7.0\pi$. It is plotted in Fig. 4 as open circles for $T=450\,^{\circ}\mathrm{C}$, and solid circles for $T=394\,^{\circ}\mathrm{C}$.

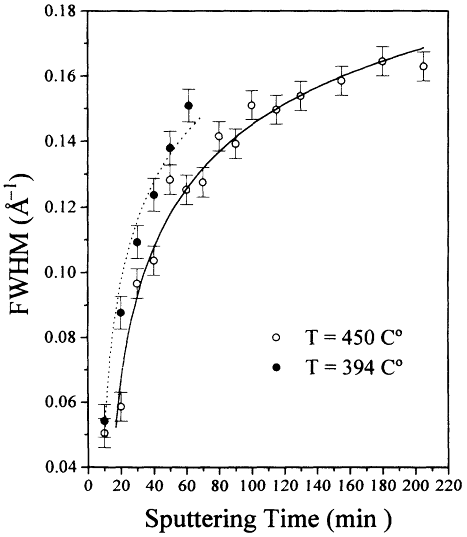

FIG. 4. The evolution of the FWHM of the (00) beam at $k_{\perp}c=7.0\pi$ as a function of the sputtering time is plotted as open circles for $T=450\,^{\circ}\mathrm{C}$ and filled circles for $T=394\,^{\circ}\mathrm{C}$. The fitted results using the form of FWHM $=A\sqrt{\ln(t/\tau)}$ are plotted as the solid curve for $T=450\,^{\circ}\mathrm{C}$ and the dashed curve for $T=394\,^{\circ}\mathrm{C}$.

C. Linear diffusion dynamics and scaling analysis

It has been suggested[7] that a close analogy exists between sputtering and epitaxial growth. In sputtering processes, fluctuation and surface diffusion are the two major competing factors. If linear diffusion plays a dominant role, the sputter etching process can be assumed to obey a Langevin equation which has been originally applied to the molecular-beam epitaxy (MBE) process[10,11,12]

$$\frac{\partial}{\partial t}Z(\mathbf{r},t)=R-\kappa\nabla^{4}Z(\mathbf{r},t) + \text{lattice pinning potential} + \eta\,. \qquad (1)$$

where R is the etching rate and η is the white-noise-simulating random fluctuation during sputtering. The lattice pinning potential[11] favors the discrete values of the heights in a crystalline surface. The ∇^4 term in Eq. (1) represents the linear diffusion, with the coefficient κ governed by the Arrhenius law $\kappa \sim \exp(-E_a/k_BT)$, where E_a is the activation energy for nearest-neighbor hopping.[11,12] Note that κ defined in front of the ∇^4 term in Eq. (1) is not the same as the diffusion coefficient associated with the ∇^2 term in the conventional Langevin equation.[13]

Equation (1) reveals two morphologies, i.e., a smooth phase at high temperatures and a rough phase at low temperatures. The transition between them may look like a Kosterlitz-Thouless transition,[14] although this transition is purely dynamic and controlled by the diffusion coefficient κ.[11] At high temperatures, the pinning potential with large κ is dominant, and the surface forms a flat phase. In contrast, at low temperatures the diffusion is so slow that the fluctuation prevails to produce a rough phase. Thus our observation shown in Fig. 1 is qualitatively consistent with the above prediction.

The most important prediction from Eq. (1) is the dynamic-scaling behavior that exists in the low-temperature phase. After the pinning potential in Eq. (1) is renormalized to zero at low temperature, the height-height correlation function can be obtained analytically.[10,11] The explicit form has been derived by Amar, Lam, and Family:[15]

$$H(\mathbf{r},t)=\langle[Z(\mathbf{r},t)-Z(0,t)]^{2}\rangle \sim \frac{1}{\kappa}\int_{0}^{1/b_c}\frac{dq}{q^{3}}[1-J_{0}(qr)](1-e^{-2\kappa t q^{4}})\,, \qquad (2)$$

where the system is assumed to be infinitely large with $d=2+1$, $J_0(x)$ denotes the zeroth order Bessel function, and b_c is the short length-scale cutoff with an order of the lattice constant. The asymptotic form of Eq. (2) shows dynamic-scaling behavior. The time scaling exists in the long-range scale $r \gg \xi$, where $H(\mathbf{r},t)=2w^{2}\sim t^{1/2}$, and the lateral correlation length, $\xi \sim t^{1/4}$, which is a distance within which the surface height fluctuations are correlated but beyond which the variations spread and are not correlated. Space scaling exists in the short-range scale $r \ll \xi$, where the leading term in Eq. (2) has a scale-invariant form $H(\mathbf{r},t) \sim (t/\rho)^{2\alpha} \sim r^2\kappa^{-1}\ln(t)$, with $\alpha = 1$ and $\rho^{-1} \propto \sim \sqrt{\kappa^{-1}\ln(t)}$. In crystalline surfaces, ρ^{-1} is proportional to the average local step density.[9] (For noncrystalline surfaces, $\rho^{-1} \propto$ average local surface slope). Therefore, in this model, the local morphology is not stationary and the step density grows as $\sqrt{\ln(t)}$. This phenomenon arises from the fact that, at sufficiently low temperatures, the atomic mobility is so low that the disordered atoms can only relax to nearby kink sites before they are subsequently sputtered. The diffusion is thus unable to compete with the fluctuation, and a balance between them cannot be established completely on the short-range scale, which leads to a local roughening evolution with the growing step density.

The scaling characteristics with $\alpha = 1$ and $\rho^{-1} \propto \sqrt{\ln(t)}$ are distinctly different from the conventional dynamic scaling hypothesis[1] in which the scale-invariant local structure is both self-affine $(0 \le \alpha < 1)$ (Ref. 16) and time invariant $(\rho^{-1} = \text{const})$. The local time-invariant characteristic with a self-affine surface morphology can be the result of the nonlinear diffusion dynamics.[10–12] The time-invariant growth has been observed recently in MBE experiments.[17,18] Our present experiment cannot be explained by the conventional dynamic-scaling theory, but is consistent with the anomalous dynamic-scaling behavior[15] predicted by Eq. (2). The scale-invariant behavior is clearly demonstrated in Fig. 3, where all plots at different times have log-scaled linear relations with almost equal slopes, except at the region close to the out-of-phase diffraction condition, $k_\perp c = 7\pi$, i.e., $\ln(k_\perp c/\pi - 6) = 0$. The FWHM thus varies with $(k_\perp c - 6\pi)$ in the form of a power law. It has been shown rigorously[9] that for a scale-invariant surface structure described by $H(r,t) \sim (r/\rho)^{2\alpha}$, the relationship of FWHM vs $k_\perp c$ in the corresponding diffraction line shape should exhibit a power law FWHM $\sim \rho^{-1}|k_\perp c - 2n\pi|^{1/\alpha}$, where n is an integer such that $|k_\perp c - 2n\pi| \le \pi$. In Fig. 3, the power-law behavior of the FWHM, $|k_\perp c - 6\pi|^{1/\alpha}$, is preserved during sputtering, while the roughening evolution manifests itself in the growing amplitude of the FWHM which is proportional to the step density ($\propto \rho^{-1}$). In Fig. 3, the deviation from the power law at the near out-of-phase condition is a result of the discrete lattice effect that breaks down the scaling behavior on the atomic length scale.[9]

According to the relation $\alpha = 1/\text{slope}$, the roughness exponent α can be extracted from the measured power-law relation. We estimated $\alpha = 1.15 \pm 0.08$. Judging from the measured $\alpha(\sim 1)$ and the time-dependent FWHM shown in Figs. 2–4, we conclude that the local morphology is neither self-affine nor stationary during sputtering. It is therefore consistent with anomalous scaling characteristics given by Eq. (2). Quantitatively, we can further demonstrate that the evolution of FWHM agrees well with the time-dependent relation FWHM $\propto \rho^{-1} \propto \sqrt{\kappa^{-1}\ln(t)}$, as predicted from Eq. (2). The experimental data in Fig. 4 were fitted by an equivalent form of FWHM $= A\sqrt{\ln(t/\tau)}$, where A and τ are adjustable fitting parameters (the constant τ has a unit of time). The fitted result is plotted in Fig. 4 as the solid curve for $T = 450\,^\circ$C, and the dashed curve for $T = 394\,^\circ$C.

IV. SUMMARY AND DISCUSSION

The sputtering process produces both a smooth Si(111)(7×7) phase at high temperatures and a rough (1×1) phase at low temperatures. The dramatic morphology change indicates a dynamic phase transition occurring between 450 and 530 °C. Below 450 °C, the sputter-induced roughening shows an anomalous dynamic-scaling behavior, which agrees quantitatively with the prediction based on linear diffusion dynamics. This result suggests that the dynamic-scaling approach designed to characterize the growth processes can be readily applied to the etching and erosion processes.

Recently, a number of MBE experiments have been performed to study the dynamic-scaling phenomenon. However, the anomalous scaling due to linear dynamics in MBE growth has not been observed. Apart from experimental difficulties, the reason for this may arise from the fact that the growth in the chosen systems is dominated either by nonlinear dynamics[17] or by the facet formation.[18] The use of the sputtering process is therefore an alternative approach to avoid these problems. We believe that our experiment can stimulate more studies of dynamic scaling through different approaches.

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