Influence of Si deposition on the electromigration induced step bunching instability on Si(111)

B.J. Gibbons, J. Noffsinger\textsuperscript{a)}, and J.P. Pelz\textsuperscript{b)}

\textit{Department of Physics, The Ohio State University, Columbus, OH 43210}

Abstract

Effects of Si deposition on electromigration induced step bunching on the Si(111) – (1x1) surface were studied for “Temperature Regimes” I (~850 – 950 °C) and II (~1040 – 1190 °C) on “dimpled samples” that have a range of initial surface miscut angles (±0.5°). We find that a step-down electric current is required to induce bunching under both net sublimation and depositions conditions in temperature Regime I, in agreement with previous reports. However, for temperature “Regime II” we observe that step-up current is required to induce step bunching for \textit{both} net deposition and net sublimation conditions, in contradiction with the report of Métois \textit{et al.} [Surf. Sci. 440 (1999) 407] and suggested “step permeability” model of Stoyanov [Surf. Sci. 416 (1998) 200]. We further observe a strong reduction in the number of crossing steps on the wide terraces for near equilibrium Si flux conditions. We also report a systematic, nearly linear dependence of the step bunching rate on the initial sample miscut angle in both Regimes I and II, which is independent of net deposition/sublimation conditions.

\textsuperscript{a)} Permanent address: \textit{Department of Physics & Astronomy, The University of Kansas, Lawrence, KS 66045.}
\textsuperscript{b)} Corresponding author. Tel.: 614-292-8388; fax: 614-292-7557.  
\textit{E-mail address: jpelz@mps.ohio-state.edu} (J.P. Pelz)
Keywords: Atomic Force Microscopy, Diffusions and migration, Evaporation and sublimation, Growth, Step formation and bunching, Surface diffusion, Surface structure, morphology, roughness, and topography.

Introduction

Understanding the processes that govern the motion of vicinal surface steps has been a longstanding problem of great fundamental interest in surface science. For semiconductors, understanding the behavior of surface steps is technologically critical for the growth of epitaxial overlayers and for device processing. Recently there has been great interest in processes that lead to spontaneous rearrangement of steps into surface structures of size ranging from a few nm up to many \( \mu \text{m} \) [1–5].

Since 1988 [6] it has been known that heating a slightly-miscut Si(111) with direct current (DC) in an ultra-high vacuum (UHV) environment leads to large scale changes in surface morphology such as the formation of step bunches. The earliest reports of these “surface electromigration” phenomena by Latyshev and coworkers [6] showed that step bunching on Si(111) only results for one direction of the current relative to the vicinal “step-up” or step-down” direction (see Fig. 1(a)), but also that this required current direction reverses multiple times with increasing temperature. In temperature “Regime I” (~850 – 950 °C) and “Regime III” (~1200 – 1300 °C) bunching occurs only for step-down heating current, while in “Regime II” (~1040 – 1190 °C) and “Regime IV” (>1320 °C) bunching occurs only for step-up heating
current. In all temperature regimes the opposite current direction maintains an initially-vicinal surface and accelerates relaxation of an initially step-bunched surface [7].

Still controversial is the physical origin of these temperature-dependent reversals of the current direction required for step bunching. It is generally accepted that the diffusing surface species (thought to be individual Si adatoms for Si(111)) each have an effective charge $q_{\text{eff}}$ that causes them to drift (or flow) either parallel (for $q_{\text{eff}} > 0$) or antiparallel (for $q_{\text{eff}} < 0$) to the applied electric field. It is also generally agreed that a step-down adatom flow will cause a bunching instability provided steps have a sufficiently small attachment probability and are sufficiently “impermeable” (i.e., a diffusing surface atom must incorporate into a step before it can cross onto the adjacent terrace [8]). One general model proposed that $q_{\text{eff}}$ changes sign as the temperature is increased so that adatom flow is parallel to the applied electric current in Regime I and Regime III, and anti-parallel in Regime II and Regime IV [9, 10]. In this case there will be step-down adatom flow (and hence bunching) only for step-down (step-up) electric current in Regimes I and III (Regimes II and IV). Another general view is that adatom flow is always parallel to the applied electric field (i.e., $q_{\text{eff}}$ is always positive) but that temperature-dependent changes to the step permeability [11, 12] or to the relative adatom diffusivity close to a step edge [13] lead to bunching even for step-up adatom flow. Here we focus on the model proposed by Stoyanov [11] and supporting experimental evidence [12] which hold that significantly increased step permeability in Regime II causes step bunching for a step-up adatom flow in that regime. A key prediction of this model is that permeable steps will bunch for step-up adatom flow only under “net sublimation” conditions (when the Si sublimation rate $R_{\text{sub}}$ is larger than an applied Si deposition rate $R_{\text{dep}}$) while a step-down flow is required to produce bunching under “net

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1 As addressed later, so called “in-phase step wandering” has sometimes been observed for step-down current in regime II.
deposition” conditions ($R_{\text{sub}} < R_{\text{dep}}$). Métois et al. [12] reported observations that bunching in Regime II indeed follows this predicted pattern, with step-up current required for bunching under net sublimation, and step-down current required under net deposition conditions. This would appear to rule out competing models [9, 10, 13] which predict that the required current direction for step bunching should be the same under either net sublimation or net deposition conditions.

Here we report a series of measurements made under clean UHV conditions, which clearly show that changing from net sublimation to net deposition conditions has no effect on the required current direction for step bunching in Regime II, in disagreement with the step permeability model [11] and supporting experiments [12]. Our measurements utilize a Si(111) wafer with a large-radius spherical “dimple” [14–19] so that behavior for step-up and step-down current flow could be observed on different parts of a wafer annealed under identical experimental conditions. We do observe that the net deposition or sublimation has a strong effect on the density of “crossing steps” on the terraces between the step bunches, in agreement with basic considerations and the observations of Stoyanov et al. [20]. We also report a systematic, nearly linear dependence of the rate of step bunching on the initial sample miscut angle (in both Regime I and Regime II) which may be useful to test different models.

**Experiment**

Samples were cut from an n-type Si(111) wafer (0.075 – 0.2 Ω·cm) into 25 x 6 x 0.4 mm$^3$ rectangular strips with the long edge parallel to $\langle 1\bar{1}2 \rangle$. Shallow spherical “dimples” of radius of ~76 mm were ground into the sample surface providing a ±0.5° range of surface miscuts angles and cleaned as described elsewhere [14–16]. To minimize contamination during annealing, unmounted wafer samples where introduced through a load-lock into our UHV annealing chamber (base pressure <1 x 10$^{-10}$ Torr) and inserted in-situ into a well-outgassed sample holder.
consisting of two ~2.5 cm long Mo heating supports held far from anything else in the system. Samples were outgassed in UHV for ~2 hours at 780 °C by passing 60 Hz alternating-current (AC) through the sample, and then cleaned by “flashing” repeatedly at ~1220 °C (each flash lasting ~5 – 10s) for a total time ~2.5 – 5 minutes. Samples were then annealed for 3.0 h at 940 °C or 1090 °C under various deposition conditions. Si deposition (up to ~0.25 Å/s) was done using a commercial rod-fed electron beam evaporator (Tectra GmbH e-flux evaporator) held ~10 cm from the Si wafer. The Si deposition rate was monitored by a quartz crystal that was shielded from the hot sample to ensure it only received Si flux from the deposition source. Temperature was monitored using an optical pyrometer. The pressure remained below ~4\times 10^{-10} \text{Torr} during long anneals in the absence of Si flux, and below ~1\times 10^{-9} \text{Torr} for the cases of highest Si deposition flux.

After annealing under a given sublimation/deposition condition, samples were transferred out of the UHV and characterized \textit{ex-situ} with atomic force microscopy (AFM), as shown in Figs. 1 and 2. On samples annealed with the electron beam evaporator operating, we did observe widely-spaced “pinning sites” [21, 22] but these only affected step bunching within 5 – 10 \mu m of the pinning site (see Fig. 3(a)). As done previously [10, 21] we used the shape of pinned steps close to pinning sites (see Fig. 3(b)) and 3(c)) to accurately determine whether a given annealing condition produced net sublimation or net deposition conditions. We could estimate the sublimation rate at a given temperature by measuring the deposition flux that produced near zero net sublimation/deposition rate $R_{\text{net}} \equiv (R_{\text{dep}} - R_{\text{sub}})$.

\textbf{Results and Discussion}

Fig. 1(a) shows a schematic sample cross-section near the dimple bottom, with the applied electric field (and hence the applied current direction) directed from right to left. All
images shown here correspond to this geometry, so the left (right) side of the dimple corresponds to step-up (step-down) current. Figs. 1(b) and 1(c) show step-up and step-down areas of a sample annealed for 3 hours at 940 °C (in regime I) in the absence of the Si deposition flux (free sublimation with $R_{\text{sub}} \cong 0.002 \text{ Å/s}$), while Figs. 1(d) and 1(e) show step-up and step-down regions of a different sample annealed at the same temperature but under a Si flux $R_{\text{dep}} \cong 0.012 \text{ Å/s}$, and so $R_{\text{net}} \cong +0.01 \text{ Å/s}$. Step bunching occurs only for step-down current under both net sublimation and net deposition conditions, consistent with past reports for Regime I [10, 12].

We also observe in Fig. 1(c) (free sublimation) that the lowest step in each bunch has separated slightly from the bunch, as observed by Homma et al [23], who suggest this is due to step-flow growth when free adatoms on the wide terraces attach to the bounding step edges during the sample quench. Fig. 1(e) shows slightly different behavior under net deposition, where now a number of “crossing steps” [24] can be observed on the wide terraces between bunches. During annealing, the net adatom deposition flux on the terraces causes significant step-flow growth of the lowest step of each bunch, which continuously break away from the bottom of one bunch and cross over to the top edge of the adjacent bunch. As discussed later, crossing steps should in general exist under either net sublimation or net deposition conditions [24], but not close to balanced deposition/sublimation conditions.

Figure 2 shows the bunching behavior at 1090 °C (well into Regime II) where under free sublimation conditions step bunching is known to exist only for step-up current [6, 10, 12, 23, 25, 26, 27]. Figs. 2(a) and 2(b) show that we also observe bunching only for step up current under free sublimation ($R_{\text{sub}} \cong 0.16 \text{ Å/s}$), consistent with these previous reports. Figs. 2(c) and 2(d) show bunching behavior at 1090 °C near “balanced” sublimation/deposition conditions ($R_{\text{dep}} \cong R_{\text{sub}} \cong 0.16 \text{ Å/s}$), where again bunching is only observed for step-up current. In this near-
balanced case however, we do observe a dramatic reduction in the number of crossing steps on
the bunched surface, consistent with the report of Stoyanov et al. [26]. We will return to this
point later.

Figs. 2(e) and 2(f) show bunching behavior at 1090 °C under net-deposition conditions,
with $R_{\text{dep}} \cong 0.25$ Å/s and hence $R_{\text{net}} \cong 0.09$ Å/s. Here again bunching is clearly observed only for
step-up current, in direct disagreement with the report [12] that net deposition conditions in
Regime II cause bunching to occur only for step-down current. We have repeated this
measurement more than 5 times under a variety of net deposition/sublimation conditions, and so
far have never observed step bunching for step-down current in Regime II. Our observations are
inconsistent with the step-permeability model [11] which predicts a reversal of the required
current direction for step bunching in Regime II. Our observations are consistent with the model
which assumes sign reversal of the effective charge in Regime 2 [9, 10], and the recent model
which assumes diffusion near step edges becomes very fast in Regime II [13]. Both of these
models predict that net deposition/sublimation have no effect on the required current direction
for bunching.

Fig. 2(c) shows a marked reduction in the density of crossing steps under near-balanced
conditions ($R_{\text{dep}} \cong R_{\text{sub}}$), consistent with the report of Stoyanov et al. [26]. We can understand
this within the “simultaneous bunching/debunching instability” model of Kandel and Weeks [24]
in the following way. The applied electric field causes the adatom concentration to vary across a
wide terrace between bunches as shown schematically in Fig. 4, assuming here that surface
electromigration causes the adatoms “pile-up” on the down-hill side of the wide terrace. In
strong net sublimation [Fig. 4(b)], the adatom concentration is below the equilibrium value $c_{\text{eq}}$ on
both sides of the large terrace. This will result a net loss of adatoms from the top step in the right
bunch, causing it to break away from the right bunch and cross over to the left bunch. In strong net deposition [Fig. 4(d)], the adatom concentration is above \( c_{eq} \) on both sides of the large terrace, so now the bottom step of the left bunch is unstable against crossing over. However, for a certain range of near-balanced conditions the concentration will be above \( c_{eq} \) on the right side and below \( c_{eq} \) on the left side, causing stability for the bounding steps on both sides of the wide terrace. We are currently exploring the possibility of using the dependence of the crossing step density on \( R_{\text{net}} \) to quantify the adatom pile-up during DC heating.

Lastly, we observe a clear trend in the bunching rate with sample miscut, by inspecting regions within the dimple which have different miscut. Figure 5 shows a plot of the average bunch height as a function of local miscut angle for several samples annealed for 3.0 h at 1090 °C and 940 °C under various deposition conditions. Each data point is the calculated average of numerous bunches at a given miscut, and the error bars represent the standard deviation of the mean. It is clear that the average bunch height increases systematically (and approximately linearly) with sample miscut. As a result, the average size of the large terrace between bunches is similar for all miscuts in this range. Interestingly, the bunching rate does not depend strongly on the deposition conditions, consistent with the images shown in Fig. 2. This measured dependence of bunching rate on initial miscut angle may prove useful in comparing different step-bunching models.

Finally, we note that in-phase step wandering (IPSW) has been reported in regime II for a step-down directed heating current after long heating times [18]. In our study, we observed IPSW on two samples out of the eight we studied. The IPSW occurred on those two samples at regions of step-down current and fairly high local miscut (≥ 0.3°), at near balanced conditions (\( R_{\text{net}} \approx -0.015 \) and 0.005 Å/s). We note that the ~3 h annealing time in the present study was
short compared to the ~ 4 – 48 h times use in the previous reports of IPSW [18], making it hard to determine whether the net deposition/sublimation conditions affect the formation of IPSW.

**Conclusion**

In conclusion we have studied the bunching behavior of Si(111) in regimes I and II under various Si flux deposition conditions. In Regime 2, we observe *no reversal* in the required current direction for step bunching under net deposition conditions, which argues against the step permeability explanation [11, 12] of the bunching behavior in Regime II. We also observed a minimum in crossing step density for near-balanced deposition/sublimation conditions, and a systematic near-linear increase in the bunching rate on initial sample miscut.

**Acknowledgements**

We thank S. Schaepe for assistance and discussions. This work was supported by NSF Grant No. DMR-0074416. J. Noffsinger was supported by the NSF REU program through NSF Grant No. PHY-0242665.

**References**


Fig. 1. (a) Schematic view of vicinal stepped surface close to the bottom of a spherical dimple. For right-to-left applied electric field (and conventional current) direction, the left side (right side) has step-up (step-down) current. (b) and (c): Derivative-mode AFM images (which appear as if illuminated from the left) of the surface annealed for 3.0 h at 940 °C under free sublimation conditions ($R_{net} \approx -0.002 \text{ Å/s}$) for (b) step-up and (c) step-down current. (d) and (e): surfaces annealed at 940 °C under net deposition of 0.01 Å/s for (d) step-up and (e) step-down current conditions. Bunching is only seen for step-down current for both net-sublimation and net-deposition conditions.
Fig. 2. Derivative mode AFM images for Si(111) samples DC annealed for 3.0 h at 1090 °C. The DC is oriented from right to left in all images as in Fig. 1. (a) and (b): annealed under free sublimation conditions (with $R_{net} \cong -0.16$ Å/S), (c) and (d): annealed under near-balanced deposition conditions ($R_{net} \cong 0$ Å/s), and (e) and (f): annealed under net deposition conditions ($R_{net} \cong +0.09$ Å/S). In all cases the step train remains uniform for step-down current (right side), but bunch under step-up current (left side).
Figure 3: (a) Typical large area scan of bunched surface (annealed at $T=1090$ °C) with a high Si deposition flux ($R_{\text{dep}} \cong 0.17$ Å/S $\rightarrow R_{\text{net}} \cong 0.01$ Å/S). Close up views of pinning site under slight (b) net sublimation and (c) net deposition conditions. In net sublimation (net deposition) the steps retract from (advance past) the pinning site.
Fig. 4. (a) Schematic of a bunched surface, assuming that the applied field causes a higher adatom concentration on the down-hill edge of a wide terrace between bunches. (b)-(d) the corresponding adatom concentration across the terrace (solid line) relative to the equilibrium concentration (dashed line) for (b) net sublimation, (c) balanced, and (d) net deposition conditions. For balanced conditions, the bottom (top) step of the bunch on the left (right) side of the terrace will both be stable against breaking away from the bunch.
Fig. 5. Plot of the average bunch height after a 3.0 anneal as a function of sample miscut for Regimes I and II. (a) 1090 °C anneal for net conditions: (●) $R_{\text{net}} \approx -0.16 \, \text{Å/s}$, (♦) -0.045 Å/s, (■) -0.015 Å/s, (▲) -0.005 Å/s, (○) 0.005 Å/s, (□) 0.025 Å/s, and (Δ) 0.085 Å/s. (b) 940 °C anneal for net conditions (■)$R_{\text{net}} \approx -0.002 \, \text{Å/s}$ and (○) 0.01 Å/s. For reference, the solid lines are power law fits, with best-fit exponents 0.92 and 0.93 for (a) and (b) respectively. There exists a clear near-linear increase in bunch height with initial surface miscut angle, but with little dependence on net deposition/sublimation conditions.