

Narrowing of Terrace-width Distributions During Growth on Vicinal Surfaces

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We present analytic and numerical results for the steady-state, non-equilibrium terrace-width distribution (TWD) of steps during growth on vicinal surfaces. Kinetic Monte Carlo shows that the TWD narrows progressively with increasing flux until the model breaks down. The narrowing corresponds to kinetic repulsion between moving steps, due to the intrinsic asymmetry of the adatom diffusion current on a growing surface. With a 1-dimensional (1D) model, from a Burton-Cabrera-Frank approach, we make contact with previous work, in which the attachment asymmetry can also be due to electromigration or to asymmetry in attachment rates; we deduce an expression for the narrowing via a Fokker-Planck analysis. We illustrate how Ehrlich-Schwoebel barriers (although inducing an instability in 2D) also lead to such asymmetry and narrowing.

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With equilibrium properties of vicinal surfaces [1, 2]—especially the form of the terrace width distribution (TWD) [3]—relatively well understood, much attention now focuses on non-equilibrium aspects, notably in long-fascinating field of growth [4–6]. In this paper, we apply a well-tested generic model to study how deposition modifies the TWD. We identify a deposition rate below which the flux does not measurably alter the TWD and show that higher flux produces a narrowing of the TWD equivalent to the creation of an effective repulsion between steps. This heretofore undocumented narrowing heralds the breakdown of the standard quasi-static approximation; the narrowing increases with flux until the step model loses meaning. We present a formal argument to account qualitatively for the narrowing in terms of flux-induced asymmetry in attachment probability p to upper and lower step edges. A more familiar source of such asymmetry is the Ehrlich-Schwoebel (ES) barrier [7] E_{ES} . In the limit of very slow growth, E_{ES} also produces an attachment asymmetry leading to such narrowing, but eventually leads to a Bales-Zangwill (BZ) meandering instability [8]. In contrast, an inverse Ehrlich-Schwoebel effect (in this context due to $E_{ES} < 0$) leads to a bunching instability [9] with an attendant bimodal TWD. Hence, it is of interest to investigate the stable case $E_{ES} = 0$, even if physical systems are not likely to precisely satisfy this condition [10]. (This limit has been examined, e.g., for spiral surface growth [11].) Other well-known sources of attachment asymmetry are electromigration [12, 13], [atomistically induced] differences in attachment rates [9, 14], and impurities [15–17].

In equilibrium the width of the TWD narrows with increasing strength A of the energetic (i.e. non-entropic) repulsion A/w^2 , where w is the separation between two steps (in the downstairs direction, \hat{x} in “Maryland notation” [1, 3, 18–20]). Invariably the analysis of the TWD is based on the transcription of the configuration of steps

in two spatial dimensions to the world lines of spinless fermions in one spatial dimension (\hat{x}) and a time-like dimension (\hat{y} , along the steps). It follows that the only dependence on A is through the dimensionless combination $\tilde{A} \equiv A\tilde{\beta}/(k_B T)^2$, where $\tilde{\beta}$ is the step stiffness. In the, alas, customary [1] fit of the TWD by a Gaussian, the standard deviation $\sigma \sim \tilde{A}^{-1/4}$. Whether this is a precise proportionality, and what the proportionality constant then is, depends on the approximation used [18]. A more sophisticated analysis [3, 18–20] uses a fit to the “generalized Wigner distribution” (GWD)

$$P_\varrho(s) = a_\varrho s^\varrho e^{-b_\varrho s^2}, \quad b_\varrho = \left[\frac{\Gamma(\frac{\varrho+2}{2})}{\Gamma(\frac{\varrho+1}{2})} \right]^2, \quad a_\varrho = \frac{2b_\varrho^{(\varrho+1)/2}}{\Gamma(\frac{\varrho+1}{2})}, \quad (1)$$

where the dimensionless $s \equiv w/\langle w \rangle$, with $\langle w \rangle$ the mean terrace width. The single adjustable parameter ϱ is related to \tilde{A} by $\tilde{A} = \frac{\varrho}{2} (\frac{\varrho}{2} - 1)$. This formalism also shows that a good estimate of \tilde{A} can be obtained from the standard deviation σ^2 of a Gaussian fit, albeit from a 4-term relation [3, 19, 21] rather than the conventional $\tilde{A} \propto \sigma^{-4}$.

Our expectations of the behavior of the TWD during growth are conditioned by intrinsically limited one-dimensional (1D), simplistic models, studied by Gossman *et al.* [22] and by Williams and Krishnamurthy [23]. In particular, the latter performed Monte Carlo simulations of a model in which atoms were deposited on terraces at a deposition rate F , and then attached to either ascending or descending steps with a probability p or $1-p$, respectively. The initial TWD was random, with a uniform probability between 0 and $2\langle w \rangle$. Gossman *et al.*'s model is couched in terms of the motion of steps in the presence of a diffusion bias, such as an electric field, rather than asymmetric attachment due to growth or asymmetry in attachment rates; the evolution equation for all these cases is the same in 1D.

So long as $p \leq 1/2$, the terrace widths evolved to-

wards a stationary state characterized by a Gaussian or Wigner distribution. For p close to 0 (corresponding to infinite ES barrier), the standard deviation of this distribution was found [22, 23] to behave as $(1 - 2p)^{-1/2}$, a result which is easily obtained from a simple mean-field argument, given below. On the other hand, in the seemingly simpler case of a symmetric attachment, $p = 1/2$, the standard deviation was observed to attain a finite value, as it should, instead of diverging as implied by $(1 - 2p)^{-1/2}$. The model seems superficially to be flux-independent until one recognizes that p depends on F . Williams and Krishnamurthy [23] argue that the long-time (saturation) standard deviation $\sigma = \sqrt{\langle w \rangle}$ (assuming, for now, unit lattice constant for clarity) and demonstrate it numerically for $p=0$; Krug and Schimschak [24] show rigorously that $\sigma = \sqrt{\langle w \rangle}$ for $p=0$, in which case the TWD is a Poisson distribution. In contrast, the equilibrium variance associated with 2D picture leading to Eq. (1) is $\sigma_{\text{GWD}} = [(\varrho + 1)/(2b_e) - 1]^{1/2} \langle w \rangle \rightarrow 0.422 \dots \langle w \rangle$ for steps with no energetic repulsion ($\tilde{A} = 0 \Rightarrow \varrho = 2$); the exact value for this special case is $\sigma_{\text{exact}} = 0.424 \dots \langle w \rangle$ [20, 25]. Thus, for $\langle w \rangle \geq 6$, the kinetic σ is less than σ_{GWD} ; thus, in this model, growth more strongly suppresses step fluctuations for more widely separated steps.

Analytic work on steps motion is based on the Burton, Cabrera and Frank (BCF) model [26], which assumes that adatoms on terraces obey a deposition-diffusion equation, with boundary conditions specified at the steps. In 2D, steps are lines, whose shape and position are dictated—through mass conservation—by the flux of adatoms to and from the steps themselves. Thus, the full 2D BCF model is highly nonlinear, and solving it is a formidable task, that can only be attacked with kinetic Monte Carlo (kMC) simulations. Analytic calculations can be performed when the time scales for an adatom and for a step to cross a terrace, $\langle w \rangle^2/D$, and $\langle w \rangle/v = 1/F$, respectively, are widely separated, namely when

$$1/F \gg \langle w \rangle^2/D \Rightarrow F/D \ll 1/\langle w \rangle^2, \quad (2)$$

where D is the surface diffusion constant. By this same reasoning, we estimate the temperature-dependent threshold value F_c for the flux, above which the effective interaction plays a significant role (and below which the equilibrium TWD obtains); equating rates in Eq. (2), we take

$$F_c = D/\langle w \rangle^2. \quad (3)$$

After solving the adatom deposition-diffusion equation assuming immobile steps, one computes the step velocity from the adatom flux. This *quasi-static approximation* resembles the Born-Oppenheimer approximation. Since steps are fixed when the adatom diffusion field is computed, step motion clearly cannot affect adatom diffusion

in the quasi-static regime. Decoupling step motion—and therefore, terrace-width fluctuations—from the diffusion field makes the TWD totally insensitive to step motion. It also implies that the adatom density on a terrace is symmetric with respect to the middle of that terrace. Hence, this situation is referred to as the *symmetric model* [27]. To make the adatom density asymmetric on a terrace within the quasi-static approximation, one must modify the boundary condition at one or both of the steps bordering that terrace (e.g. via E_{ES}) or introduce external drift.

Eventually the motion of the steps must affect the adatom density, since a fast-moving step will collect more adatoms from the terrace in front of it, as it sweeps through, than from the terrace behind it. To investigate this problem analytically, we restrict ourselves to the special case of straight steps.

Previously [28] we studied a vicinal surface relaxing to equilibrium by computing the behavior of the TWD as a function of time. Here we extend the numerical analysis in Ref. [28] by performing kMC simulations of the stationary TWD during deposition and growth on a vicinal surface, based on the standard, well-established [29] two-dimensional (2D) solid-on-solid (SOS) model with barrier energies E_b determined by lateral bond-counting: E_b is a diffusion barrier E_d plus a bond energy E_a times the number of lateral nearest neighbors in the initial state. This number is 1 for an edge atom leaving a straight segment of step edge for the terrace, 3 for a detaching atom that originally was part of this edge (leaving a notch or kink-antikink pair in the step), or 2 for a kink atom detaching, either to the step edge or the terrace. We adopt our oft-used [28, 30] generic values $E_d=1.0$ eV and $E_a=0.3$ eV, with $T=723$ K. (For larger E_a/E_d step-flow collapses at smaller F [31].) Sublimation is forbidden, and there is no interaction between steps besides the entropic repulsion. There are 1000 lattice sites in the \hat{y} direction along the steps, with periodic boundary conditions. In the \hat{x} direction, there are 200 lattice spacings a and $N=20$ steps, created by screw-periodic boundary conditions, so that $\langle w \rangle=10a$. To gauge F_c from Eq. (3), we estimate the hop rate as the generic value 10^{13}s^{-1} [32]. Then

$$F_c = \frac{4a^2 \times 10^{13} e^{-E_d/k_B T}}{\langle w \rangle^2 \text{s}} \rightarrow 4 \times 10^4 \frac{\text{atoms}}{\text{s}} = 0.2 \frac{\text{ML}}{\text{s}} \quad (4)$$

We focus on the effect of deposition with symmetric attachment ($E_{ES} = 0$), considering non-zero E_{ES} near the end. After trial runs at various deposition rates F , we carried out extensive studies for $F = 0.1, 1.0$, and 10 ML/s. These rates span the range from a barely perceptible effect to the verge of the meandering instabilities that herald the crossover from step-flow growth to nucleation-limited growth. Fig. 1 shows “snapshots” of the step configurations at these three deposition rates after 150 monolayers (ML), steady-state being reached by

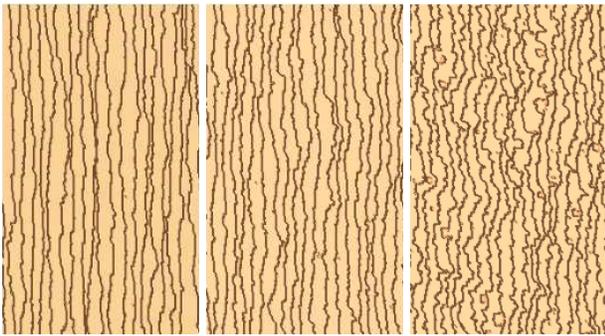


FIG. 1: Images (200×1000 sites) of the vicinal surface after deposition of 150 ML for 3 growth rates (left to right): near equilibrium ($F=0.1$ ML/s), moderate growth ($F=1$ ML/s), and rapid growth ($F=10$ ML/s) near the upper limit of step-flow growth.

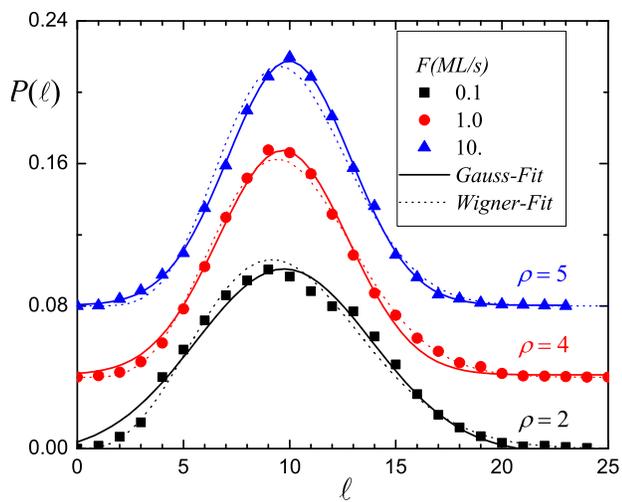


FIG. 2: [Color online] Simulated terrace-width distribution (TWD) for the 3 cases of Fig. 1. The $F=1.0$ set is displaced upward by 0.4, the $F=10$ case by 0.8, for clarity. The solid curves show fits to, a Gaussian while the dotted curves are GWDs, Eq. (1), for the indicated values of ϱ .

100ML. [30] In Fig. 2 there are corresponding plots of the TWD. [30] Unlike in our equilibrium studies, we do not average over several runs. Noteworthy results include:

- 1) For $F = 0.1$ ML/s the TWD is nearly unchanged from the equilibrium TWD. The curve is well described by the Wigner distribution for $\varrho=2$ (best fit value = 2.07), corresponding to only entropic interactions (“free fermions”). This behavior is consistent with our estimate in Eq. (4): $F = 0.1 \text{ ML/s} \lesssim F_c$ is a “small” deposition rate, at which the quasi-static approximation is expected to apply. The higher deposition rates, above F_c , lead to best fits with $\varrho \approx 4.39$ and 5.24 (which values, at equilibrium, would translate into effective interaction strengths $\tilde{A} \approx 2.6$ and 4.2) for $F = 1$ and 10 ML/s, respectively. In other words, the deposition leads to a TWD progres-

sively narrower than in equilibrium; this can be expressed as an effective repulsion between the steps, and points to a breakdown of the quasi-static approximation.

- 2) The breakdown of the quasi-static approximation takes place well into the step flow regime, before any islands nucleate on the terraces. As such, it is experimentally relevant, and may complicate the estimates of the ES barrier, which is known to also produce narrowing of the TWD. (See quantitative analysis below.)
- 3) For the highest two deposition rates the TWD can be adequately described by a Gaussian, although it is then less straightforward [18] to extract the effective interaction strength. [21] This TWD behavior is reminiscent of that found by Videcoq *et al.* [33].
- 4) The meandering of the individual steps increases with F . In equilibrium this would correspond to a decrease in the step stiffness $\tilde{\beta}$. Correspondingly, at fixed A we would expect $\tilde{A} \equiv A\tilde{\beta}/(k_B T)^2$, and so ϱ , to decrease as well. The observed increase in ϱ clearly highlights the kinetic origin of the repulsions (all the more so because $A=0$ in our simulations).
- 5) For the largest value of F , we see that a key assumption, that the step position is a single-valued function of the position y along the step, is about to break down.

This analysis was done for $\langle w \rangle = 10$ on a 200×1000 lattice. The large size in the \hat{y} direction minimizes finite-size effects. In some early runs [30], we considered several values of $\langle w \rangle$ —2, 4, 8, 10, 20— but on a 400×400 lattice. Saturation of σ was achieved after of order 100 ML. These saturation values were semiquantitatively consistent with the $\sqrt{\langle w \rangle}$ behavior found in Ref. [23], but better fit by a logarithmic rise with $\langle w \rangle$. Further investigation is warranted.

We can gain some understanding of the origin of the effective repulsion, in the previously mentioned 1D perspective [22, 23] by considering the equations of motion of a train of [descending] steps (cf. Fig. 3) in the co-moving frame [4], from the BCF perspective [26]. The deposition-diffusion equation satisfied by the adatom density $c_{m+1}(x)$ on the terrace between down-steps at x_m and x_{m+1} , and thus of width $w_{m+1} = x_{m+1} - x_m$ is

$$Dc'' + vc' + F = 0, \quad (5)$$

where $v = F\langle w \rangle$ is the velocity of the *comoving* frame. Eq. (5) is the limit of zero desorption (infinite desorption time τ) of a long-known [34, 35] and often applied [9, 11, 27, 36] result. In this limit one loses the concept of equilibrium flux and finds an infinite surface diffusion length $x_s = \sqrt{D\tau_s}$ and so an infinite Péclet number vx_s/D [35], rendering the step motion “slow”.

Assuming the boundary condition $c(x_m) = c(x_{m+1}) = 0$, we find the adatom density to be

$$c_{m+1}(x) = \frac{F}{v}(x_m - x) + \frac{F}{v}w_{m+1} \frac{1 - \exp[-v(x - x_m)/D]}{1 - \exp[-vw_{m+1}/D]}. \quad (6)$$

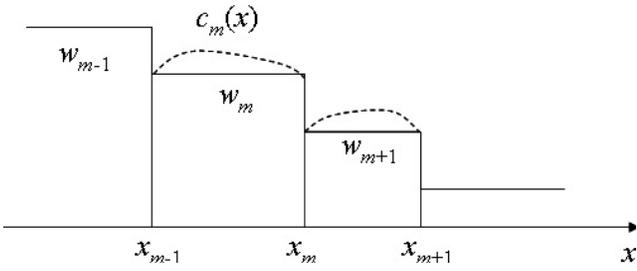


FIG. 3: Schematic of 1D model of a vicinal surface. The position is step m is x_m . Terrace m has width $w_m = x_m - x_{m-1}$ and adatom concentration $c_m(x)$.

From Eq. (6) it is straightforward to find the velocity $D \left[(\partial c_{m+1} / \partial x)|_{x=x_m^+} - (\partial c_m / \partial x)|_{x=x_m^-} \right]$ of the m^{th} step:

$$\dot{x}_m = \frac{F}{2} \left[\frac{w_{m+1} e^{vw_{m+1}/2D}}{\sinh(vw_{m+1}/2D)} - \frac{w_m e^{-vw_m/2D}}{\sinh(vw_m/2D)} \right]. \quad (7)$$

By subtracting from Eq. (7) the corresponding equation for \dot{x}_{m-1} , we obtain the equation for \dot{w}_m . For comparison with previous work, [22, 23] we linearize this equation of motion, expanding to lowest order in each $w_m - \langle w \rangle$. After straightforward algebra, we find

$$\dot{w}_m = F \left[(1-p) \{w_{m+1} - w_m\} + p \{w_m - w_{m-1}\} \right], \quad (8)$$

$$p \equiv \frac{1}{2} \left[\frac{\Phi}{\sinh^2 \Phi} - \frac{e^{-\Phi}}{\sinh \Phi} \right], \quad \Phi \equiv \frac{v \langle w \rangle}{2D} = \frac{F \langle w \rangle^2}{2D}. \quad (9)$$

From Eq. (3) we have $\Phi = F/2F_c$. Expanding in F , $p \sim 1/2 - \Phi/3 + (2/45)\Phi^2 + \mathcal{O}(\Phi^5)$; also $1-2p \approx \tanh(2\Phi/3) = \tanh(F/3F_c)$ (where the approximation is accurate to better than 0.5%). Thus, for nonvanishing F there is an apparent asymmetric attachment of diffusing atoms to steps, with preferential attachment to ascending steps ($p < 1/2$), even with no ES barrier.

To proceed, we make the mean-field assumption that $w_{m+1} = w_{m-1} = \langle w \rangle$. Since these neighboring terrace widths are anticorrelated with w_m , the quadratic potential about $\langle w \rangle$ in this approximation underestimates the restoring force. Introducing white [37] delta-correlated noise η , we find the Langevin equation

$$\dot{s} = -F(1-2p)(s-1) + \eta. \quad (10)$$

The corresponding Fokker-Planck equation is

$$\frac{\partial P(s, t)}{\partial t} = \frac{\partial}{\partial s} [F(1-2p)(s-1)P(s, t)] + \frac{\partial^2}{\partial s^2} [P(s, t)], \quad (11)$$

which, assuming initially uniform spacing and setting $\tilde{t} \equiv F(1-2p)t$, has the solution [38]

$$P(s, \tilde{t}) = \left[\frac{F(1-2p)}{2\pi(1-e^{-2\tilde{t}})} \right]^{1/2} \exp \left[-\frac{F(1-2p)(s-1)^2}{2(1-e^{-2\tilde{t}})} \right]. \quad (12)$$

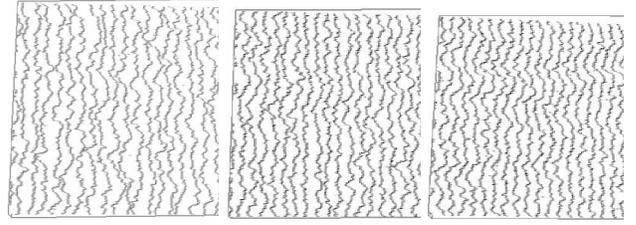


FIG. 4: Images of the simulated 200×1000 vicinal surface after deposition of 20 ML for Ehrlich-Schwoebel barriers E_{ES} , from left to right: 0.1eV, 0.3eV, infinite. Other parameters are $L=10$, $T=723\text{K}$, $F=0.01\text{ML/s}$, $E_d=1.0\text{eV}$, and $E_a=0.3\text{eV}$.

The variance σ^2 of this Gaussian distribution is

$$\sigma^2 = \frac{1 - \exp(-2\tilde{t})}{F(1-2p)} \xrightarrow{\tilde{t} \rightarrow \infty} \frac{1}{F(1-2p)} \approx \frac{1}{F \tanh\left(\frac{F}{3F_c}\right)}, \quad (13)$$

the characteristic 1D dependence found by Gossman *et al.* [22]; the distribution starts as the delta function $\delta(s-1)$ and broadens monotonically to the long-time limit in Eq. (13), with width $\sigma \propto F^{-1}$ for small F and $\sigma \propto F^{-1/2}$ for large F . Since the Gaussian is centered about $s=1$ and since $P(s < 0) > 0$ is unphysical, the analysis must fail once $\sigma \approx 1$, setting a lower limit on F for the description to be viable.

Since this analysis neglects both entropic or energetic repulsions, the behavior for small flux, $F \rightarrow 0^+$, $p \rightarrow \frac{1}{2}^-$, should have the form of a Poisson distribution, $P(s) = \exp(-s)$, associated with creation of a vicinal surface by the random deposition of straight stiff steps. (Note also Ref. [24].) Furthermore, the dependence on F is expected to be exaggerated in the model since only F is the only source of TWD narrowing. Indeed, it would predict that, in our kMC simulations, the TWD for $F=10\text{ML/s}$ should be narrower than that for $F=1\text{ML/s}$ by a factor $\geq \sqrt{10} \approx 3.2$, much greater than observed, highlighting the risk of using such simple 1D models (also unfaithful at early times) for more than qualitative purposes.

Another concern is the effect of the mean-field nature of our calculation. In equilibrium, the venerable Gruber-Mullins approximation [39] is known to underestimate the variance as a function of step-step repulsion [40]. Margetis [41] has recently obtained the exact variance of the 1D linear model, Eq. (8), going beyond the mean-field limit given in Eq. (13). While the power series of his result agrees with that of the first result in Eq. (13) until the third-order term, $\propto \tilde{t}^3/(1-2p)^2$, the exact variance actually diverges for long time like $\sqrt{\tilde{t}/(1-2p)}$ rather than saturating, in contrast to the numerical results in 2D; however, this long-time behavior is sensitive to the assumptions about the noise term added to Eq. (8) [42].

An Ehrlich-Schwoebel barrier [7] offers a different, well-known way to break the upstairs-downstairs symmetry

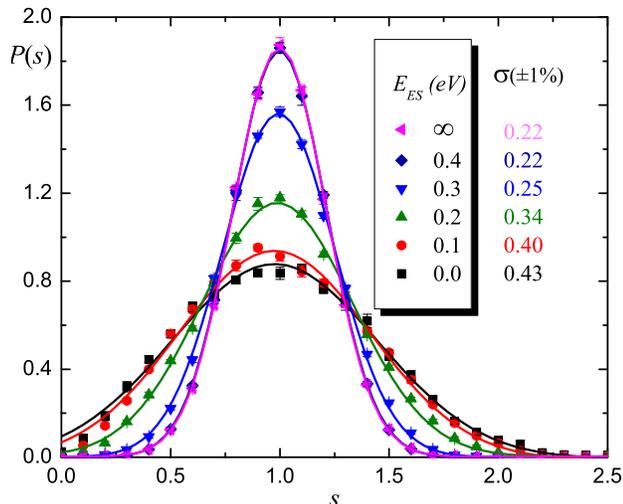


FIG. 5: [Color online] TWDs from simulated configurations (cf. Fig. 4) with $T=723\text{K}$ and very small $F=0.01$ ML/s. The smooth curves are Gaussian fits, with the indicated standard deviations σ . With increasing E_{ES} the TWD evidently narrows (σ decreases).

[4–6, 43, 44]. The resulting anisotropy is controlled through E_{ES} , which ranges from 0 to ∞ ; we consider the intermediate values 0.1, 0.2, 0.3, and 0.4 eV. At $T=723\text{K}$, the TWD has nearly converged to the infinite-barrier limit by $E_{\text{ES}}=0.4$, and the runs become prohibitively slow. To distinguish ES-induced narrowing from flux-induced narrowing, we simulate at low enough flux that there is no observable narrowing when $E_{\text{ES}}=0$; we use $F=0.01$ ML/s. (Even at $F=0.1$ ML/s, by 100 ML there is a Bales-Zangwill (BZ) [8] instability—absent when $E_{\text{ES}}=0$.)

Fig. 4 shows snapshots of the step configurations for a range of values of E_{ES} . Initially sharp, the TWD reaches its saturation width after a short transient regime of a few ML growth. The configurations are recorded after 20 ML, before the onset of periodic unstable BZ meandering. The resulting TWDs, for all values of E_{ES} that we studied, are displayed in Fig. 5, along with Gaussian fits. The standard deviation of the TWD decreases from $\sigma \sim 0.43$ (NB: $\sigma=0.42$ for $\tilde{A}=0$ at equilibrium [20, 28]) to $\sigma \sim 0.22$ for an infinite ES barrier. Correspondingly, in Fig. 4 we see behavior reminiscent of Bullet 4 above, with increasing E_{ES} rather than F ; the number of close-approaches decreases [45], and the number of kinks increases, though now as a prelude to the BZ instability. When F is too small to produce significant attachment asymmetry, the asymmetry due to E_{ES} satisfies $p/(1-p) = \exp(-E_{\text{ES}}/k_B T)$. With Eq. 9 we obtain an *effective* barrier—when actually $E_{\text{ES}}=0$ —due to the

flux:

$$\frac{E_{\text{ES}}^{\text{eff}}}{k_B T} = \ln \left(\frac{2 \sinh^2 \Phi}{\Phi - e^{-\Phi} \sinh \Phi} - 1 \right) \sim \frac{4}{3} \Phi + \frac{4}{3} \Phi^3 + \mathcal{O}(\Phi^5). \quad (14)$$

Similarly we could deduce an effective electromigration force [36] due to the flux.

In summary, we have gauged the flux ($F\langle w \rangle^2/D < 2 \times 10^{-7} \exp(E_d/k_B T)$ in our model, 0.1 ML/s for our choice of parameters) below which there is negligible change from the equilibrium TWD. With stronger flux, the quasi-static approximation fails, and the TWD narrows progressively, consistent with an effective step-step repulsion. Most experimental techniques used to probe equilibrium TWDs could observe the TWD narrowing during growth, though efforts have focused instead on step bunching and other instabilities [46]. In simulations of etching, the TWD can have the same GWD-like shape when the etchant is stirred (i.e. surface diffusion is unimportant) [47]; however, σ should be rate-independent since etched atoms do not diffuse before detaching [48]. Flux effects might well contribute to the TWD narrowing (by $\sim 1/4$) observed with reflection electron microscopy on vicinal Si(111) at 1100°C (compared to 900°C), where an incident flux compensated for desorption [49].

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