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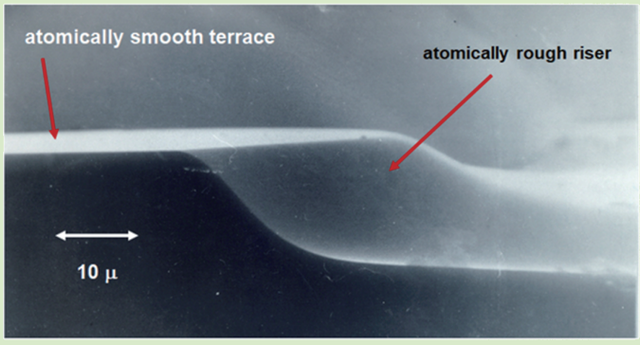

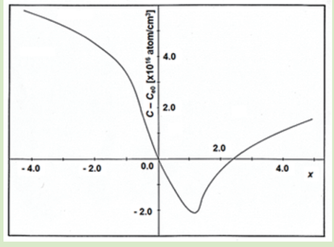

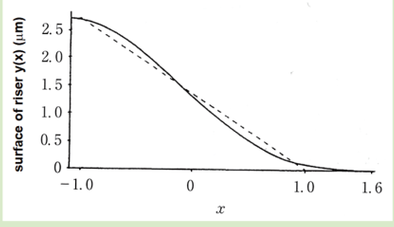

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Review Article

Distributions of Steps on Vicinal Surfaces: Renewed Interest

Theodore L. Einstein [1]

Recent experiments on curved crystals have created renewed interest in the properties of vicinal (misoriented from high-symmetry). We begin with a review of results for close-packed, straight steps. While the overall picture is rather familiar, we emphasize various noteworthy subtleties as well as use of a generalization of the Wigner surmise. The curved crystal experiments consider not just these steps but also steps that are fully kinked, zigzagged, half way in polar angle between two orientations with straight steps. The terrace-width distribution (TWD) does not satisfy the scaling relations routinely seen for straight steps. We discuss differences in the stiffness and the issues that arise in extending theoretical analysis of the TWD to these vicinal surfaces. There are several open issues that merit attention to address these questions.

1. Introduction, & Straight vs. Zigzag Steps

Vicinal surfaces are misoriented from facet planes of crystals by some fixed azimuthal angle $\theta = \tan^{-1}(h/\langle\ell\rangle)$, where h is the step height and $\langle\ell\rangle$ the mean separation between steps, a topic for which there were excellent classic reviews nearly two decades ago.[1,2] We begin by recapitulating the main theoretical underpinnings, standard results and less familiar aspects. Then we describe some recent results on curved crystals and discuss the challenges posed by these experimental observations.

While h is quantized by the interlayer spacing, $\langle\ell\rangle$ is not since vicinal surfaces are rough rather than facets (even though the terraces are). It is assumed that there is no polar misorientation from the close-packed direction. While computer simulations are based

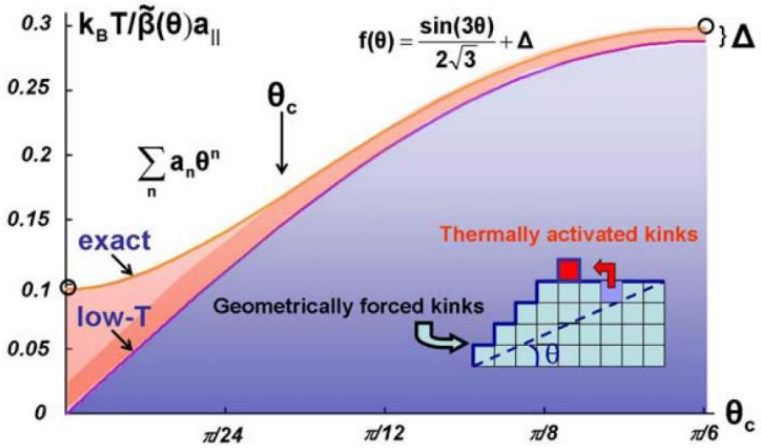

Fig. 1 The contributions to the step stiffness can be decomposed into parts originating from geometrically forced kinks (lower blue region bounded from above by the line labeled "low-T") and thermally activated kinks (the remaining red region, bounded from above by the line labeled "exact"). At relatively low temperatures, the {111} step stiffness is well approximated at angles greater than θ_c by a relatively simple function since the thermal part is evidently insensitive to angle. The method to account for smaller angles is detailed in ref. 7. The inset depicts a step edge from above. Each square represents an adatom which is part of the step edge. The uppermost square represents a thermally excited adatom, which forms four thermally-activated kinks. The remaining kinks are geometrically forced—they must be present to give the step edge an overall angle θ. From ref. 7.

Department of Physics & Condensed Matter Theory Center, University of Maryland, College Park, MD 20742-4111 USA
*Corresponding Author E-mail: einstein@umd.edu

on the full solid-on-solid model, much analytic progress has been made by coarse-graining along the steps to the step continuum model. In so-called "Maryland notation", the distance along the step is y, while the perpendicular displacement is x(y). For atomistic models, both x and y are discrete in lattice units while in the step continuum model, y becomes a continuous variable.

For a particular step, in the step continuum model, the correlation function

$$G_x(y) = \langle [x(y)-x(0)]^2 \rangle = (b^2(T)/a_\square)\,|y| = (k_B T/\tilde{\beta})\,|y|, \qquad (1)$$

where the diffusivity $b^2(T)$ is the mean-square perpendicular deviation with each pace forward along the step $a_\square$ and $\tilde{\beta}$ is the stiffness, the mass-like inertial term for steps. For square-lattice terraces with nearest and next-nearest neighbor interactions ϵ_1 and ϵ_2, respectively, this problem has been studied for over three decades,[3] and there is an extensive literature on the subject, a few examples of which are refs. 4-7.

This is the form of a simple random walk, a valid viewpoint until the meandering step "becomes aware" of adjacent steps. Assuming the simplest nearest-neighbor model with kink energy ε, the energy of a deviation (kink) that is n units long is $n\varepsilon$, then $b^2(T)/a_\perp^2$, where $a_\perp$ is the unit distance perpendicular to the step for kinks, is readily calculated in the SOS approximation (see e.g. ref. 5) to be

$$b^2(T)/a_\perp^2 = \tfrac{1}{2} \sinh^{-2} (\varepsilon/2k_B T), \qquad (2)$$

where ε is $-\tfrac{1}{2}\epsilon_1$ the corresponding nearest-neighbor energy on a square lattice.[4,5] For larger values of |y|, $G_x(y)$ grows less rapidly: eventually $G_x(y) \propto \ln |y|$.[5,8]

While Eq. (2) shows that $\tilde{\beta}$ for straight steps is dominated by thermal effects, steps which are sufficiently misoriented by a polar angle find their stiffness to be dominated by geometric effects. At $\theta = \pi/4$, many configurations have the same rectilinear L_1 ("Manhattan" or "city block") distance, steps can fluctuate with little or no (in ϵ_1–only models) cost in energy. In the low-temperature, geometric limit, the dimensionless inverse stiffness $k_B T/\tilde{\beta} a$ is $2^{-\frac{1}{2}} \exp(\epsilon_2/\epsilon_1) \to 2^{-\frac{1}{2}}$ when $\epsilon_2 \to 0$.[4,5] Similar behavior occurs on (111) surfaces, as illustrated in Fig. 1; both have been studied in exhaustive detail in ref. 7.

2. Step Spacings: Standard & Subtle Results

The distribution of spacings between adjacent neighboring steps is readily measured in real-space experiments (scanning probe, LEEM, etc.) and calculated in simulations; furthermore, it offers considerable physical insight. (See several earlier review by the author and co-workers[9-11] for more details of the following.) As realized half a century ago by de Gennes[12] in studies of polymers in two dimensions (2D), the configurations of the steps can be analyzed using a Schrödinger equation once one reinterprets the continuous y as a time-like variable. Specifically, if one assumes that the energy of a step is proportional to its length, then the integral over y is weighted by $[1 + (dx/dy)^2]^{1/2} \sim 1 + \tfrac{1}{2}(dx/dy)^2$. The first term is a constant. If we view the distance y along the step as a time-like variable, then the second is a kinetic energy. Thus, we view the path of a step in two spatial dimensions as the evolution of a particle in one spatial and one time-like dimension, and we seek the time- average of the step-step spacings. Since steps do not cross, the particles correspond to [spinless] fermions [or to hard-core bosons, used in some of the literature but not here]. The inertial coefficient or mass of these particles in 1+1 D is proportional to the stiffness $\tilde{\beta}$ of the step, and is one of the key parameters of the step continuum model.

The other key parameter of the equilibrium properties of the step continuum model relates to the repulsion between steps. In most situations this has the form A/ℓ^2. There are two contributions to A. The first is the entropic or steric repulsion associated with the non-crossing of steps, as detailed elsewhere, and is evidently repulsive. The second is due to energetic repulsions associated with the frustrated relaxation of surface atoms between steps in a monotonic strain field associated with dipolar strain at each step. Note that

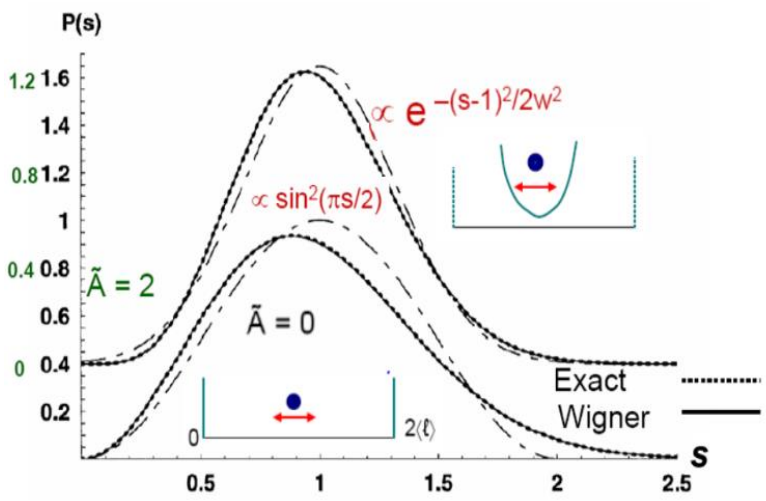

Fig. 2 P(s) vs. $s \equiv \ell/\langle\ell\rangle$ for essentially the exact "free fermion" $\tilde{A} = 0$ result (solid curve), the Gruber-Mullins (GM) approximation $\sin^2(\pi s/2)$ (long-short dashed curve), and the $\beta = 2$ Wigner surmise result (dotted curve), barely distinguishable from the exact result). Offset upward by 0.4 for clarity, a similar plot of an approximant of the exact result for $\tilde{A} = 2$ (solid curve), the GM Gaussian approximation $(24/\pi^2)^{1/4} \exp[-2\sqrt{6}\,(s-1)^2]$ (long-short dashed curve) and the $\beta = 4$ Wigner result (dotted curve). The insets depict the scenarios envisioned in the GM approximation viewed in 1+1 D, with the average over y becoming a time average. See text for explanations. Adapted from ref. 10.

the two contributions do not simply add: as the elastic repulsion weakens, neighboring steps encounter each other more often, increasing the entropic repulsion.[11] In the opposite limit, the steps behave like independent entities, with a separation variance twice that of a single active step.[13]

Pursuing de Gennes's picture for N steps, we find an N-particle Schrödinger equation with an explicit $A/(x_i - x_{i+1})^2$ repulsion. The kinetic energy is the translation of the steric repulsion. The simplest analysis is to invoke mean field, called the Gruber-Mullins approximation[14] in this case. All but one step is fixed at its equilibrium separation determined by the misorientation. We deal analytically with two situations, shown in Fig. 2. When A = 0 (lower inset panel), we have a particle in a 1D box that is $2\langle\ell\rangle$ wide. The ground state wavefunction ψ_0 is $\langle\ell\rangle^{-1} \sin(\pi x/2\langle\ell\rangle)$. Then the terrace-width distribution P(s) is $|\psi_0|^2 \propto \sin^2(\pi s/2)$, where s is the normalized dimensionless variable $x/\langle\ell\rangle$, and, remarkably, the ground-state energy is exactly the entropic repulsion.

In the other limit, depicted in the upper inset of Fig. 2, we expand the interaction term for the repulsions from the two adjacent particles [evolving steps $x(y) \to x(t)$] about the mean position of the active step: $A\,[\ell^{-2} + (2\langle\ell\rangle - \ell)^{-2}] \approx A\,\langle\ell\rangle^{-2}\{1 + (s-1)\}^{-2} + \{1 - (s-1)\}^{-2}] \approx A\,\langle\ell\rangle^{-2}\,[2 + (s-1)^2 + \ldots]$. Hence, for s near 1 (i.e., near the middle), the potential is quadratic; when the interaction is not weak, the particle rarely collides with a neighbor. The strength of the repulsion enters only in the dimensionless form $\tilde{A} = A\tilde{\beta}/(k_B T)^2$. In this case, we find the familiar harmonic oscillator result, with a Gaussian ground state ψ_0 centered at the mean spacing, so $P \propto \exp[(s-1)^2/2w^2]$, where w is $(48\tilde{A})^{-1/4}$. (The 48 takes into account expansions for all—not just nearest—pairs of neighboring fixed particles.) In addition to the faltering of the expansion at small $\tilde{A}$, for large $\tilde{A}$ the predicted variance is half what it should be since only the fluctuations of the active step and not its neighbor are taken into account.

Notice that these distributions depend only on *s* and not on $\langle\ell\rangle$ (or x and $\langle\ell\rangle$ separately). This is a consequence of both the kinetic and the potential energy in the Schrödinger equation scaling like ℓ^{-2}. This scaling has been seen in many experiments, starting nearly 3 decades ago.[15] However, when the step interactions contain more complicated interactions, this result fails so that P(s) depends also on $\langle\ell\rangle$. Arguably the nicest illustration of this idea is the non-scaling of the TWD on vicinal Ag(110),[16] on which the steps (in addition to the dipolar repulsion) interact via a metallic surface state with the consequent RKKY-like oscillatory interaction and with a new length scale introduced, the Fermi wavelength of this Shockley state.

There are two fundamental difficulties with this Gaussian procedure. For small $\tilde{A}$ it fails because replacing the interaction by just a quadratic is a poor approximation. For large $\tilde{A}$ the variance of the fluctuations of the step spacings is underestimated because

the fluctuations of only the active step and ipso facto not the rigid neighbor steps are not included.[13] An improved approximation is provided by a generalization of the so-called Wigner surmise of random matrix theory[17]

$$P_\beta(s) = a_\beta s^\beta \exp(-b_\beta s^2), \text{ where } a_\beta = \frac{2\left[\Gamma\left(\frac{\beta}{2}+1\right)\right]^{\beta+1}}{\left[\Gamma\left(\frac{\beta}{2}+\frac{1}{2}\right)\right]^{\beta+2}} \quad \text{and} \quad b_\beta = \left[\frac{\Gamma\left(\frac{\beta}{2}+1\right)}{\Gamma\left(\frac{\beta}{2}+\frac{1}{2}\right)}\right]^2, \qquad (3)$$

hereafter called GWD (generalized Wigner distribution). The coefficients a_β and b_β are β-dependent combinations of gamma functions that assure normalization and unit mean, respectively, in this single-parameter expression. The exponent β (not to be confused with the step free energy per length) is related to the dimensionless repulsion by $\beta = 1 + (1 + 4\tilde{A})^{1/2}$, or $\tilde{A} = (\beta/2)[(\beta/2) - 1]$. The relevant properties of $P_\beta(s)$ have been tabulated.[11,18] In particular, the variance σ^2 is $[(\beta + 1)/2b_\beta] - 1$, and the peak occurs at[11]

$$s_{mode} = \left(\beta \big/ 2b_\beta\right)^{1/2} \approx 1 - \tfrac{1}{4}\left(\beta^{-1} + e^{-\beta}\right) \qquad (4)$$

Both σ and s_{mode} are rather insensitive to Ã, so conversely the value of Ã extracted from σ or s_{mode} as determined by experiment is very sensitive to uncertainties in them. Values of Ã from fitting the peak width (actually the standard deviation about 1) and the peak position should consistent, as should the values obtained from the skewness and kurtosis. Hence, one should fit to the full TWD (in particular, the range $0.5 < s < 2.0$ where there are enough counts for the experimental data to be reliable) rather than just one characteristic of it. Thus, if the experimental TWD is not in good agreement with Eq. (3), then using its variance or peak position to extract Ã will be decidedly suspect.

While it is beyond the scope and length of this paper to explore theoretical underpinnings of Eq. (3) in much detail, we offer some succinct comments: Wigner wrote down this expression as conjecture for the distribution of the spacings between N energy levels of nuclei with random couplings that have some common symmetry, based on an analytic study of the case N=2. The 3 symmetries of such problems are orthogonal, unitary, and symplectic, corresponding to β = 1, 2, and 4, respectively. The thinking behind this pursuit is Wigner's insight that ensembles of dynamical systems with different Hamiltonians but some common symmetry should exhibit some generic common properties. The exact solutions for these three special cases are known; Eq. (3) turns out not to be an exact solution, but it is a fine approximation, especially for β = 2 and 4. See Fig. 2.

The connection to vicinal surfaces comes via a family of problems called Calogero-Sutherland models.[17,19,20] They describe spinless fermions on a chain or ring that interact with each other by a $(\text{separation})^{-2}$ repulsion. Rather miraculously, the many-particle ground state of this problem corresponds to that of the random Hamiltonian matrix problem, so that the many step-spacing distribution (the joint probability distribution) corresponds to that of the differences of eigenenergies of the matrix problem. For steps, there is no symmetry that says that β should have one of the 3 special values, and in fact, β = 1 corresponds to an attractive step interaction. Also, β = 2 corresponds to "free fermions", steps which have no energetic repulsion (viz. Ã = 0). The case β =4 has very few physical realizations in Hamiltonian systems. Alas, it is not clear how to extract the distribution of spacings between pairs of steps from the full multistep wavefunction in general, and there is no exact solution (for arbitrary Ã) against which to calibrate the accuracy of Eq. (3), although some unsophisticated numerical simulations show that it is a better approximation than any of the other analytic forms that have been used to describe TWD's.[21] (Le Caër et al.[22] proposed an interesting analytic framework for this issue, and high-grade numerical simulations should now be possible, so there is hope that the viability of the GWD for arbitrary positive β will be elucidated.) Attempts to confront actual data yield positive values of β up to about 10.[10,11]

Many cautions were given in refs. 9-11. We recount a few as well as offering some more recent results.

1) In general the spacing ℓ between steps is not a simple integer multiple of the spacing in that direction on the terrace plane. There is an offset corresponding to the offset between adjacent 2D nets of successive layers below the terrace. The affine offset is half the lattice spacing for close-packed steps on a (100) bcc surface and 1/3 or 2/3 for such steps on a (111) fcc surface. These effects are particularly important if one focuses experiments principally on s_{mode}.

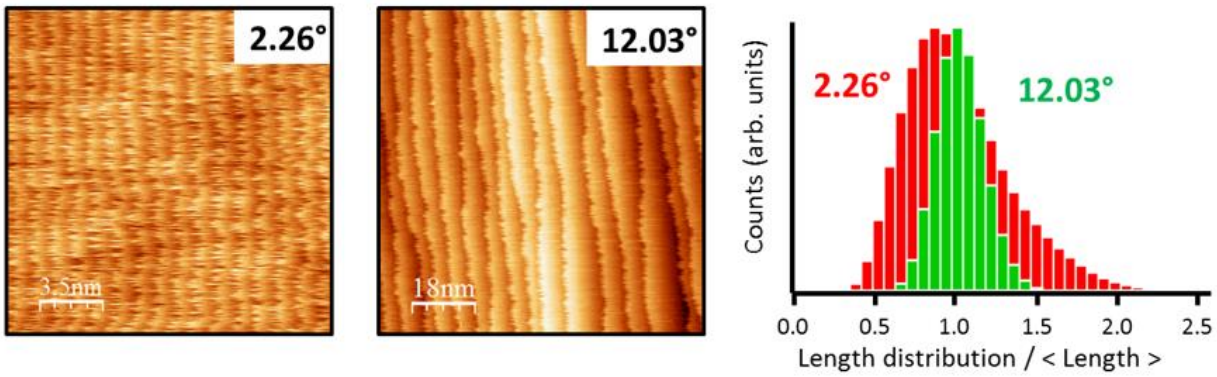

Fig. 3 STM images of high and shallow misorientations on a curved Ag (654) crystal with zigzag, fully kinked steps. The TWD's of the two clearly do not scale (are not just a function of the abscissa coordinate *s* and even have different skewness, in stark distinction from the properties of straight steps such as A steps ((100) microfacet) on curved Ag (111), not shown. From ref. 34.

2) The average spacing between steps $\langle \ell \rangle$ need not be one of these ℓ's but can be arbitrary since the vicinal orientation is not a facet, but rather thermodynamically rough.)

3) For small ℓ, the ℓ^{-3} corrections[23] to the dipolar repulsion and the differences between discrete and continuum x(y) can become significant.

4) Unlike fermions, steps can touch, just not cross. When they touch there can be an additional energy ε_t. For the fermion condition, $\varepsilon_t = \infty$; for $\varepsilon_t < 0$, step doubling is favored at low temperatures. For any finite ε_t the effective repulsion between steps decreases (from ∞), as manifested by a drop in the value of the characteristic exponent β. This behavior can be analyzed as a finite-size effect in $\varepsilon_t / k_B T$, diminishing as $\langle \ell \rangle \to \infty$. See ref. 24 for details. Note that this breaks, to some degree, the vaunted independence of $P_\beta(s)$ from $\langle \ell \rangle$.

5) Eq. (3) can also be derived in a Fokker-Planck framework as the stationary-state solution of an effective Hamiltonian.[25] This approach offers some idea of how equilibration of steps occurs. In particular, the standard deviation of the TWD approaches its saturation value as $[1 - \exp(-t/\tau)]^{1/2}$, as confirmed with kinetic Monte Carlo simulations.[26] (Here $\ln \tau$ is expected to be the barrier energy over $k_B T$, so study of τ gives information about the dominant kinetic process.) It also provides a route to apply this analysis to other problems ostensibly unrelated, most notably to show that GWD could be used to characterize the capture-zone distribution of submonolayer islands growing on a surface.[27] That is, rather than looking at the size distribution of these islands, one can instead look at the distribution of areas of Voronoi (proximity) cells determined by the islands. In this problem there is no intrinsic connection corresponding to the Calogero-Sutherland models, and the GWD works well only near the center portion of the distribution, where there is significant experimental data (in contrast to computer simulations, where adequate data in the tails can be generated). In this case, the characteristic exponent β can be related to the critical nucleus size, the largest size of an unstable island. On a macro scale, this distribution also can be used to describe the area distribution of secondary administrative units (counties in the southeast USA, French arrondissements (districts), but not ken in Japan) and of the areas of Voronoi cells determined by subway stations in major cities. See ref. 28 for further analysis and refs. 29,30 for short reviews.

3. Curved Crystals and Zigzag Steps

These questions have experienced a renaissance due to exciting experiments on curved crystals primarily by Ortega's group,[31-34] also by Juurlink's group.[35,36] In such experiments, one can study a range of misorientations under the same experimental conditions, a "dream come true" for testing scaling predictions. There are a large variety of intriguing results for curved crystals of late transition metals around (111) planes with straight steps, including step doubling) and around (654) planes with fully kinked steps. Arguably most striking is the failure of scaling of the TWD for the latter, as seen in Fig. 3.

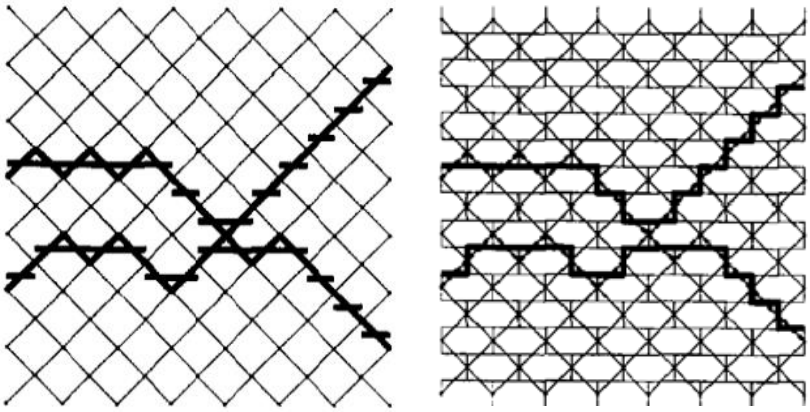

Fig. 4 Mapping by Abraham et al.[42] from zigzag steps on a rotated square lattice to restricted straight steps. In the left panel, horizontal lines of length $2^{-1/2}$ the lattice constant are constructed through the midpoints of each diagonal link. Where needed, the endpoints are connected by a vertical line of the same length. Note that after each pace the step can stay the same or move up or down by 1 unit. However, a move in one direction cannot be immediately followed by a move in the other direction (but requires at least one non-displacive move in between).

The fully kinked orientation of steps on a square lattice (the p-RSOS, point-contact restricted SOS model) is most natural for analysis with the PWFRG (product-wave-function renormalization group) approach, fruitfully and often applied by N. Akutsu and co-workers.[37-41] In this model, A = 0, and the analog of ε_t (see item 4 in the previous section) is negative (sticky steps) but involves touching only at a point (corner) rather than along a whole unit link. Akutsu et al. can calculate slopes of equilibrium crystal shapes, phase diagrams, and two-step TWDs, but not [yet] more general TWDs.

In contrast, most of the theory described in section 2 is rooted in straight steps with thermally excited kinks. There are some serious concerns in porting that framework to this orientation, where thermal effects are subsidiary to geometric ones, which also severely restrict the deviation (perpendicular to the meander direction) for each pace along the step. Here are some issues:

1) It seems likely that the dipolar A/ℓ^2 repulsion persists, but it is less clear how to make contact with terrace-plane elastic constants, particularly when they are anisotropic, as is typical. There could be offsets in the new [rotated] x direction, and corrections to ℓ^{-2} could enter already for larger ℓ.

2) More problematic is the path from lattice models to the step continuum model, the Schrödinger equation, Calogero- Sutherland models, and the Wigner surmise of Eq. (3). So it is by no means clear that the GWD is still viable for these steps. To solve exactly the 6-vertex model, Abraham et al.[42] devised the mapping shown in Fig. 4. (Note the difference between this figure and the SOS interface analyzed by Burkhardt,[3] in which there is no restriction on the number of perpendicular deviations as one paces along the step.) From this, one can read off the value of ℓ for each pace. The analytic accounting of paths is not independent at each gridpoint along the horizontal (see Fig. 4 caption), so it is not clear how helpful this mapping will be for producing convenient formulas, although they have proved convenient for computer simulations.[43,44]

Indeed, to investigate relaxation dynamics and steady-state configurations for fully-kinked—compared to straight steps on the (100) surface of a simple cubic lattice, Hamouda and co-workers[44] have performed kinetic Monte Carlo calculations. Among their findings are that processes breaking 2 bonds dominate over those breaking 3 bonds (which dominate the case of straight steps). Steps relax more quickly to steady state, with kink generation-annihilation processes assuming a significant role; also, the standard deviation of the TWD is larger and has two characteristic times for the approach to steady state $[1 - \exp(-t/\tau_1) - \exp(-t/\tau_2)]^{1/2}$ (cf. item 5 in the previous section for straight steps): τ_1 arises from the rapid decrease of kinks in transient regimes at early times while the much larger τ_2 comes from relaxation to steady state, so is the actual relaxation time.

4. Conclusions and Prospects

Further analytic developments, approximations, simulations were obtained by Jarmillo et al. [45] for TWDs as well as for step pair correlation functions,[11,46] as might be measured in scattering experiments.

There are many intriguing phenomena on vicinal surfaces beyond the scope of this short review. A striking example is the network of steps formed near, e.g. Au and Pt, (110) missing-row reconstructed surfaces. In the model calculation[47] of the orientation dependence of the equilibrium free energy, there is evidence of what amounts to a ℓ^{-1} interaction between steps (based on the presence of a quadratic [rather than cubic] term in the expansion of this energy in terms of misorientation slope); however, this pattern is metastable or unstable. Similar patterns can be generated using the p-RSOS model,[40] as can step bunching,[38] a widely observed and discussed nonequilibrium occurrence, as reviewed relatively recently by Misbah et al.[48] Except for brief comments on relaxation, we have said little about the vast literature on step dynamics.

In summary, the exciting prospect of experiments on curved crystals as revitalized interest in vicinal surfaces. The theoretical machinery for dealing with close-packed steps, in particular the use of the GWD (Eq. (3)) to study TWDs is well charted. However, there are open questions regarding how to analyze data from misorientations with zigzag (fully kinked) steps which were not resolved in the heyday of vicinal surfaces. It is crucial that they now be addressed (and answered), so that one can confidently interpret the data from curved crystals with fully-kinked steps.

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