

Dynamical Scaling Implications of Ferrari, Prähofer, and Spohn's Remarkable Spatial Scaling Results for Facet-Edge Fluctuations

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Abstract Spurred by theoretical predictions from Ferrari et al. (Phys Rev E 69:035102(R), 2004), we rederived and extended their result heuristically. With experimental colleagues we used STM line scans to corroborate their prediction that the fluctuations of the step bounding a facet exhibit scaling properties distinct from those of isolated steps or steps on vicinal surfaces. The correlation functions was shown to go as $t^{0.15(3)}$, decidedly different from the $t^{0.26(2)}$ behavior for fluctuations of isolated steps.

Keywords Scaling · Fluctuations · Steps · Facets · STM

1 Introduction

Fluctuations of steps on surfaces play a central role in determining their impact on surface processes and the evolution of surface morphology. In the past nearly-two decades, the step continuum model has allowed several successful quantitative correlations of direct observations of step fluctuations with kinetic and thermodynamic descriptions of nanoscale structural evolution [1–6], bridging from the atomistic and nanoscale to the mesoscale. For steps on flat or vicinal (misoriented modestly from a facet orientation) surfaces, there are two well-defined scaling behaviors for temporal correlations, corresponding to cases B and A, conserved and non-conserved dynamics, respectively, in the framework of dynamic critical phenomena [7].

To Prof. Dr. Herbert Spohn, with appreciation.

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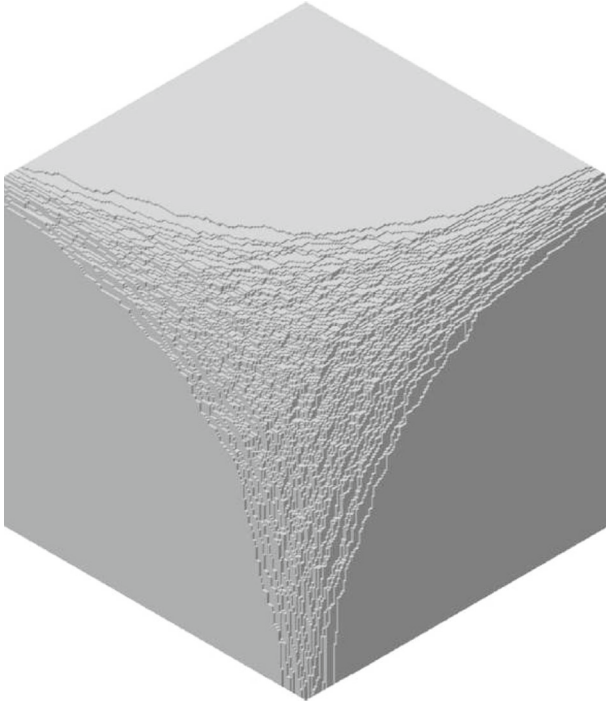


Fig. 1 Simple-cubic crystal corner viewed from the $\{111\}$ direction, from Refs. [15–18]

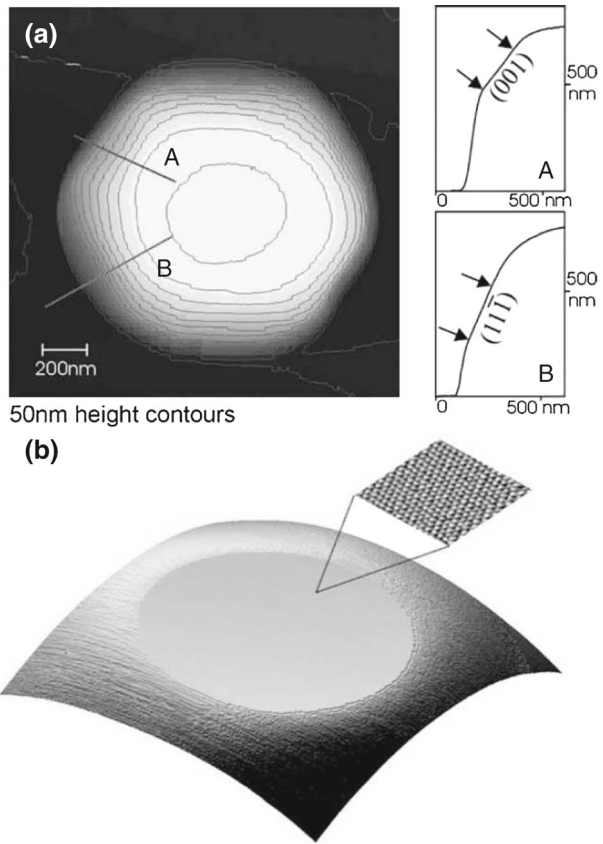
Several examples of both behaviors have been observed experimentally in physical systems [1,2,8] and numerically in Monte Carlo simulations [9–11].

For complex structures where mass transport is limited by geometry, the fundamental question of how fluctuations behave in a constrained environment becomes experimentally accessible. These issues become particularly important for smaller structures, especially nanostructures, where issues of finite volume (shape effects and volume conservation) become non-negligible [12,13]. Although the step can still be viewed as a 1D interface obeying a Langevin-type equation of motion, not only local deformation but global effects must be considered when calculating the step chemical potential. These considerations alter the equation of motion, including the noise term, resulting in different universality classes of dynamic scaling [14] (see Table II below).

Thus, it was especially enlightening and inspiring to read of a well-defined intermediate scaling regime in Ferrari, Prähofer, and Spohn’s stimulating paper [15] (hereafter FPS) (as well as related works [16–18]), in which they computed the scaling of equilibrium fluctuations of an atomic ledge bordering a crystalline facet surrounded by rough regions of the equilibrium crystal shape in their examination of a 3D Ising corner (Fig. 1). We refer to this boundary edge as the “shoreline” since it is the edge of an island-like region—the crystal facet-surrounded by a “sea” of steps, a situation which can now be observed (cf. Fig. 2) and studied quantitatively [19].

FPS derive an intriguing exact result, concerning how the width w of shoreline fluctuations scales as a function of the linear size of the facet. This length corresponds to the length of a step or the linear dimension of an island (or its circumference). This length is often called L [20,21] and other times ℓ [22] (while ℓ has a closely related but slightly different meaning in

Fig. 2 **a** Micron-size lead crystal (supported on Ru) imaged with a variable-temperature STM at $T = 95^\circ\text{C}$. Annealing at $T = 95^\circ\text{C}$ for 20h allowed it to obtain its stable, regular shape. Lines marked *a* and *b* indicate location of profiles. Profile A crosses a (0 0 1)-side facet, while profile B a (1 1 1)-side facet. **b** 770 nm \times 770 nm section of the top part of a Pb-crystal. The *insert* shows a 5.3 nm \times 5.3 nm area of the top facet, confirming its (1 1 1)-orientation. Both the main image and the insert were obtained at $T = 110^\circ\text{C}$; from Ref. [19]



Ref. [15]). To prevent any possible confusion, we henceforth denote this length by the Polish crossed L, \mathcal{L} , following the notation used in a celebratory presentation on this subject [23], on which this article is based.

We anticipate that

$$w \sim \mathcal{L}^\alpha, \tag{1}$$

where the value of roughness exponent α depends on the mode of mass transport and the geometry of the step. For the step that serves as the border a two-dimensional (2D) island on a high-symmetry crystal plane, one expects (and finds in physical and numerical experiments) that $w \sim \mathcal{L}^{1/2}$, i.e. $\alpha = 1/2$, since this step performs a random walk [24].

FPS show that, as we quipped in the title of our paper [22], “a crystal facet is not an island”. Indeed, they find that instead of the expected random-walk behavior,

$$w \sim \mathcal{L}^{1/3}, \tag{2}$$

i.e. $\alpha = 1/3$, for a crystal facet. They prove that the origin of the unusual $\mathcal{L}^{1/3}$ scaling lies in the step-step interactions between the facet ledge and the neighboring steps under conditions of conserved volume. Note that this value of α is intermediate between $\alpha = 1/2$ for isolated steps and $\alpha = 0$ ($w \sim \ln(\mathcal{L})$) [25] for a step on a vicinal surface, i.e. in a step train.

FPS’s formidable calculation is based on the use of free spinless fermions, transfer matrices, random-matrix properties, Airy functions, and specific models; as a purely static result,

it does not address the question of the time behavior of step fluctuations, which are easier to measure experimentally.

As alluded to above, this article is intended to be an expansion [26] of a celebratory talk [23] which described the impact of FPS on our research, in particular the results found in three publications [21, 22, 27]. In Sect. 2 we summarize highlights of FPS that motivated and underpinned our subsequent work. In Sect. 3 we describe the relevant correlation functions. Next we present a heuristic derivation extending the reasoning of Pimpinelli et al. [20] that leads to the dynamic scaling of shoreline fluctuations, as well as the static result of FPS. Then we present a more formal analysis of scaling for curved steps in an asymmetric potential. In Sect. 4 we describe experiments using scanning tunneling microscopy (STM) that demonstrate the novel scaling behavior in a physical system. The Conclusion section offers some final remarks [28].¹

2 Summary of Highlights of FPS

FPS assume that there are no interactions between steps beyond entropic (i.e., the steric repulsions arising from the fact that steps cannot cross), so that the step configurations can be mapped to the world lines of free spinless fermions; the entropic repulsion is captured by the fermionic Pauli condition [29–31].² A key new feature compared to treatments of vicinal surfaces is that the volume of the crystallite is conserved. Their fermionic Hamiltonian \mathcal{H}_F is

$$\mathcal{H}_F = \sum_j \left(-a_j^\dagger a_{j+1} - a_{j+1}^\dagger a_j + 2a_j^\dagger a_j + \frac{j}{\lambda} a_j^\dagger a_j \right), \quad (3)$$

where λ^{-1} is a Lagrange multiplier associated with conserved volume. See Fig. 3. It is this final term that is new in their treatment. Its asymmetry is key to the novel behavior they find. They then derive an exact result for the step density in terms of J_j , the Bessel function of integer order j , and its derivative. Near the shoreline they find

$$\lim_{\lambda \rightarrow \infty} \lambda^{1/3} \rho_\lambda(\lambda^{1/3} x) = -x(\text{Ai}(x))^2 + (\text{Ai}'(x))^2, \quad (4)$$

where ρ_λ is the step density (for the particular value of λ).

The presence of the Airy function Ai results from the asymmetric potential implicit in \mathcal{H}_F and preordains exponents involving $1/3$. The variance of the wandering of the shoreline, the top fermionic world line in Fig. 3 and denoted by b , is given by

$$\text{Var}[b_\lambda(t) - b_\lambda(0)] \cong \lambda^{2/3} g(\lambda^{-2/3} t) \quad (5)$$

¹ This paper encountered unfortunate refereeing difficulties regarding expanded expositions of some material in the three refereed publications [21, 22, 27]. Since a great deal of time has already been spent on this project, with much more likely needed to resolve disagreements, and because our primary objective is to highlight in this celebratory volume—which will soon be sent to press—the importance of Prof. Spohn’s insights in our work, we have followed the editor’s directive to delete sections 3.2 and 5 from Ref. [26]. Section 5 describes a toy model; it was included in Ref. [26] because it had been mentioned in Refs. [21, 23, 27] rather than for its import. On the other hand, Section 3.2 contains scaling relations and critical exponents for curved geometries and asymmetric potentials that may be of interest, especially to those who do not dismiss results making use of [28]. Curious readers are invited to view the deleted material on the arXiv [26] and judge for themselves.

² Physically, steps—in contrast to fermions—actually can touch, just not cross, leading to a finite-size correction to the standard fermion results, see Ref. [29].

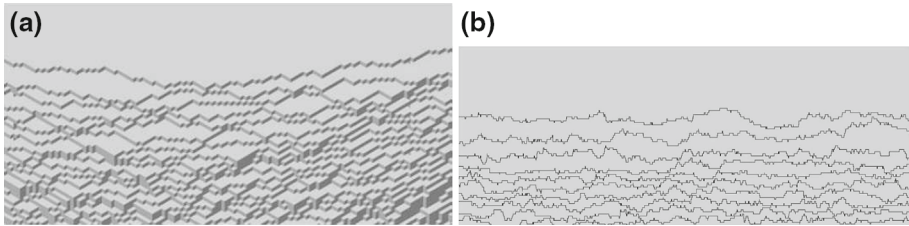


Fig. 3 **a** Magnified detail of the steps near the facet edge in Fig. 1, from Refs. [16, 17]; **b** Snapshot of computed configurations of the top steps (those near a facet at the flattened side portion of a cylinder) for a terrace-step-kink (TSK) model with volume constraint, from Refs. [15, 17]

where t is the fermionic “time” along the step; $g(s) \sim 2|s|$ for small s (diffusive meandering) and $\sim 1.6264 - 2/s^2$ for large s . They then set λ to a scaling parameter $\ell = (4N/1.202\dots)^{1/3}$, where 1.202... is Apéry’s constant and N is the number of atoms in the crystal, as in Fig. 1. They find

$$\text{Var}[b_\ell(\ell\tau + x) - b_\ell(\ell\tau)] \cong (\mathcal{A}\ell)^{2/3} g(\mathcal{A}^{1/3}\ell^{-2/3}x), \tag{6}$$

where $\mathcal{A} = (1/2)b''_\infty$.³ This leads to the central result that the width $w \sim \ell^{1/3}$. Furthermore, the fluctuations are non-Gaussian. They also show that near the shoreline, the deviation of the equilibrium crystal shape from the facet plane takes on the Gruber-Mullins-Pokrovsky-Talapov [30, 31] form $-(r - r_0)^{3/2}$, where r is the lateral distance from the facet center and r_0 is the radius of the facet.

3 Analytical Results and Heuristic Derivation

In this section, we discuss computation of the time scaling of step-edge fluctuations using two approaches. First, we adopt a simple scaling argument, starting from FPS’s exact result. Then we derive a continuum-equation description of the step bordering a crystal facet. Then, with simple power counting we rederive FPS’s result, as well as the temporal power-law scaling of edge fluctuations.

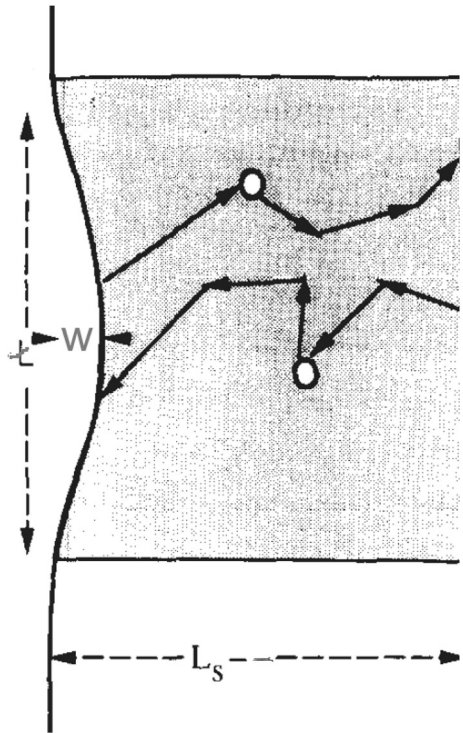
For straight steps, which underlie treatments of this problem, one adopts cartesian coordinates (x, y) , y being in the direction along the step edge, and $x(y)$ describing the step profile, in what has been called “Maryland notation” [32]. We focus attention on the step autocorrelation function

$$G(t) = \langle [x(y_0, t + t_0) - x(y_0, t_0)]^2 \rangle_{y_0, t_0} \underset{t \rightarrow 0}{\sim} t^{2\beta}, \tag{7}$$

which can readily be computed in a Monte Carlo simulation [9] and measured experimentally with a scanning probe like STM. It is less feasible to measure spatial correlation functions since such experiments do not take an instantaneous “snapshot.” Like a television screen, different parts of the micrograph correspond to different times, and it is problematic to deal with what transpires between successive visits by the STM tip to nearby positions. Furthermore, in such experiments one does not do a full average over y_0 but rather picks a single value; for that case we replace $G(t)$ by $G(y_0, t)$, for which there is no spatial average. The resulting plot of displacement x vs. time looks similar to scans of x along a step, and so are called “pseudo-images” [2]. (Cf. Fig. 6 below.) At short times $G(y_0, t)$ exhibits the

³ In Ref. [15] \mathcal{A} is defined as b''_∞ ; writing $\mathcal{A} = (1/2)\mathcal{A}$ simplifies Eq. (6).

Fig. 4 Schematic of a stepped surface seen from above, to illustrate the “pipe” concept of Ref. [20]. The fluctuating step, depicted as a *thick wavy line*, shows the definitions of w and L . For the two particular cases considered in the text, the thickness L_s of the pipe-like reservoir to the right of the step, in which atom exchange occurs, is of order the lattice spacing a . Adapted from Ref. [20]



same $t^{2\beta}$ behavior as $G(t)$. The value of exponent β depends on the atomistic processes responsible for the fluctuations of the step, but also on the position of the step with respect to a crystal facet, as we proved in Ref. [22] and recount shortly below. In the other limit, $G(t \rightarrow \infty)$ saturates to $2w^2$, where w is width of the fluctuations.

When doing simulations [25] (or if one had a probe that could take instantaneous snapshots), one can probe

$$G(y, t_0) = \langle [x(y_0 + y, t_0) - x(y_0, t_0)]^2 \rangle_{y_0} \sim w^2, \quad (8)$$

for large values of y . Then the roughness exponent α can be extracted from the saturation value of the width w of the fluctuating step by using Eq. (1) and identifying L with the size of the system in the y -direction.

Starting from FPS's result that the roughness exponent $\alpha = 1/3$ in Eq. (1), we apply Pimpinelli et al.'s argument [20] for finding the temporal scaling: Referring to Fig. 4, consider a protruding portion of step of length L and width w . At equilibrium, the protrusion is due to fluctuations in the number of atoms to and from a reservoir. The reservoir is assumed to be, depending on the limiting atom transport process, either another part of the same step, or a region on the crystal surface. Before considering a specific kinetic process, let us derive a few general relations. On average, a number $N(t)$ of atoms continually move between the step and the reservoir during a time interval t . The reservoir is by definition situated one diffusion length L_s away from the protrusion. While the net flux to and from the step vanishes, the number $N(t)$ fluctuates around its vanishing mean; assuming that atom fluxes in different time intervals are uncorrelated, we can compute the typical size of the fluctuation δN , which is of order $\sqrt{N(t)}$. Denoting by Ω the atomic area, we estimate the size w of the

protrusion along the step edge (defined, as said above, as the amplitude or width of a typical step fluctuation of length L) from $w \times L \approx \Omega \sqrt{N(t)}$. To estimate $N(t)$, we note that the size (surface area) of the reservoir feeding the fluctuation is $L L_s$. Then the number of atoms moving to and from the step edge during time t is proportional to the number of diffusing atoms in the region feeding the step, $c_{eq} L L_s$ (where c_{eq} is the equilibrium particle density) and to the fraction of time the atoms spend in this region, t/τ^* . The characteristic time τ^* depends on the specific transport process (see below). Then, as in Ref. [20]:

$$N(t) \approx \frac{c_{eq}}{\tau^*} L L_s t. \tag{9}$$

Furthermore, the squared area of the fluctuating bulge is

$$w^2 L^2 \approx (\delta N)^2 \approx N, \tag{10}$$

where we assume $\delta N \approx \sqrt{N}$.

While Ref. [20] considers several different cases, we focus here on the two primary scenarios: i) non-conservative mass transport by attachment-detachment to/from the step edge, with fast terrace diffusion (A/D) and ii) conservative mass transport by step-edge diffusion (SED).

- i) In the A/D case, $1/\tau^* \approx k$, where k is an appropriate kinetic coefficient. For fast surface diffusion, the step effectively exchanges atoms with a “2D adatom vapor” on the surface. Then, L_s is of order the lattice spacing a , and Eq. (9) yields

$$w^2 L^2 \approx k c_{eq} L a t. \tag{11}$$

Using Eq. (2) leads to $L^{5/3} \sim t$, and eventually to

$$w \sim t^{1/5} \rightarrow G(t) \sim t^{2/5}. \tag{12}$$

In comparison, $G(t) \sim t^{1/2}$ for a straight step or an isolated 2D island [10,20].

- ii) For mass transport by step-edge diffusion along a portion of step of size L , $1/\tau^* \approx D_e/L^2$, where D_e is the edge diffusion coefficient. Again in this case $L_s \approx a$, so that Eq. (9) becomes

$$N(t) \approx t c_{eq} D_e a / L. \tag{13}$$

From Eq. (10)

$$w^2 \approx t c_{eq} D_e a / L^3. \tag{14}$$

With Eq. (2) we now find $t \sim L^{11/3}$, so that

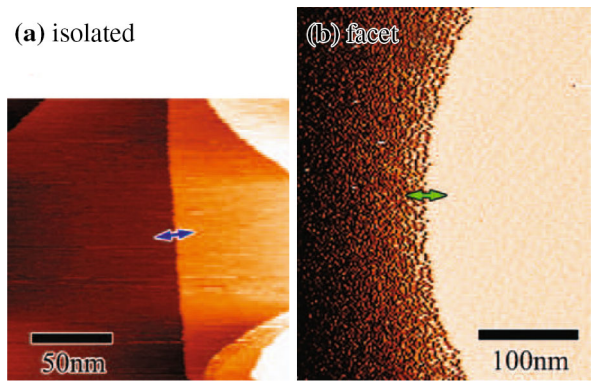
$$w \approx t^{1/11} \rightarrow G(t) \sim t^{2/11} \tag{15}$$

($\beta = 1/11$) for a crystal facet fluctuating through step edge diffusion. In comparison, $G(t) \sim t^{1/4}$ ($\beta = 1/2$) for a straight step or an isolated (large) 2D island [10,20].

4 STM Experiments

The first experimental observations of the novel scaling predicted for facet-edge fluctuations on crystallites were performed at the University of Maryland by Masashi Degawa supervised by Williams [21]. Crystallites were formed by depositing a 20–30 nm Pb film at room temperature on a Ru(0001) substrate in UHV [34–36], and subsequently dewetting at 620 K. The liquid Pb droplets solidified upon slow cooling and were left to equilibrate to a stable state

Fig. 5 An STM image of **a** an isolated step on a crystallite facet (room temperature) and **b** a crystal facet edge (350 K). The small superimposed *double-arrow* indicates the tip path that leads to line-scan images as in Fig. 6. From Refs. [21,33]



at the T of the experiment [19,34,35,37]. The crystallites were observed with a variable-temperature scanning tunneling microscope (VT-STM) after equilibration. Figure 5 depicts an STM image of (a) an isolated step (at room temperature) and (b) facet-edge (at 350 K). A crystallite in a stable state as shown in (b) has a flat, close-to-circular (111) facet and a smoothly-connecting vicinal region.

Since STM images are scans rather than instantaneous “snapshots”, the data for dynamic scaling can be more compelling than that for static scaling. By repeatedly scanning perpendicularly to a single position (y_0) along the facet-edge or step (cf. Fig. 5), we obtain a line-scan STM image [2] $x(t)$, as shown in Fig. 6 for (a) an isolated step (step originating from a screw dislocation) and (b) a facet-edge, both at 350 K. Digitized step-displacement positions $x(t)$ extracted from these “pseudoimages” are used for statistical analysis. To evaluate the growth exponent β , we calculated the early behavior of the time correlation function $G(t)$ given in Eq. (7). To evaluate the roughness exponent α , we can calculate either the saturation value of the width w of the fluctuating step as in Eq. (1) or the spatial correlation function $G(y, t_0) \sim y^{2\alpha}$ for y less than the correlation length [25].

Figure 7 shows $G(t)$ determined for (a) facet-edges and (b) isolated step-edges. Squares, circles, and triangles represent measurements at 300, 350 and 400 K, respectively. Each curve

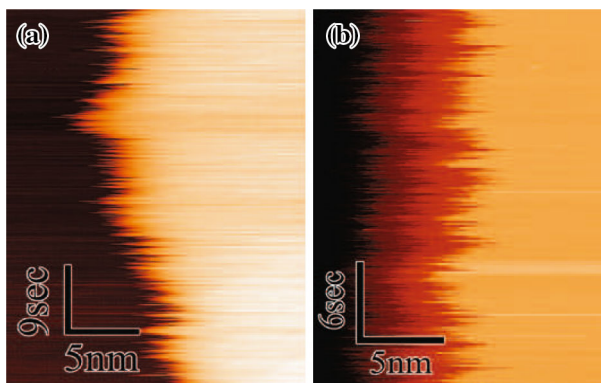
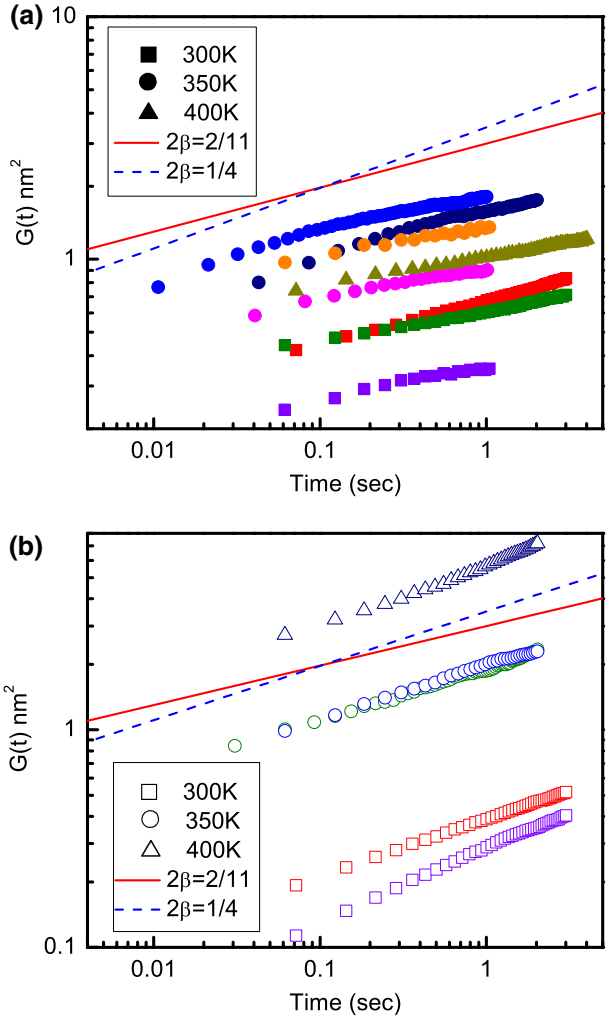


Fig. 6 Segment of a line-scan pseudoimage of **a** an isolated step (step from screw dislocation) and **b** a facet-edge at 350 K, showing also the correlated fluctuations of the neighboring steps. The time interval between lines is 0.02 s, and 2,000 lines are measured per image. From Refs. [21,33]

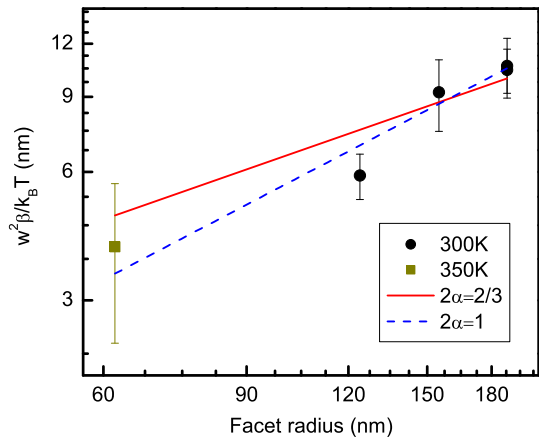
Fig. 7 Log-log plot of $G(y_0, t)$ (cf. Eq. (7)) for **a** facet edges and **b** isolated steps with facet radii from 60 to 190 nm. The symbols represent: 300 K (squares), 350 K (circles), and 400 K (triangles). For guidance, solid and dashed lines show slopes $2\beta = 2/11 = 0.18$ and $1/4$, respectively. Individual fits to each of the data sets yield slopes of **a** facet edges: 300 K: 0.18(1), 0.13(6), 0.13(2); 350 K: 0.17(4), 0.17(4), 0.12(3), 0.11(5); 400 K: 0.12(12), and **b** isolated steps: 300 K: 0.32(3), 0.26(1); 350 K: 0.24(3), 0.24(4); 400 K: 0.30(4). From Refs. [21,33]



displays the average over the correlation functions for 10–30 measurements of $x(y_0, t)$. The exponent 2β for each temperature is obtained from the slope of the curve on the log-log plot; the values of these slopes are listed in the figure caption. As expected the exponents show no systematic thermal dependence: from all data sets, the σ^{-2} -weighted average exponent is $2\beta = 0.149 \pm 0.032$ for facet-edges and $2\beta = 0.262 \pm 0.021$ for isolated steps. With over 99.9% confidence (using Student’s t test), these values come from different parent populations. Each of the two results is within one standard deviation, σ , of their respective predicted values of $2/11$ and $1/4$.

Determination of the roughness exponent α requires evaluation of the system-size dependence. A detailed examination is a challenge beyond the capability of the STM experiments being used. However, we can demonstrate that size does affect fluctuations. Under the assumption $L \sim R$ (the facet radius) for confined steps (Fig. 7a), we expect $w^2 \sim R^{2\alpha}$. (For the unconfined steps, the system size is larger than the limitations imposed by the finite measurement time [4]). The effect of the facet size is apparent in Fig. 7a since the three upper sets

Fig. 8 Product of squared saturation width and reduced stiffness as a function of facet radius (facet-edge only). Circles and squares are room temperature and 350 K, respectively. Solid and dashed lines are a fit to the 350 K data with $\alpha = 1/3$ and $\alpha = 1/2$, respectively. From Refs. [21,33]



of data at 350 K were taken on larger crystallites, having radii > 100 nm. More quantitatively, Fig. 8 plots the characteristic length $w^2 \tilde{\beta} / k_B T$ versus facet radius at 300 K and 350 K, using for the step stiffness $\tilde{\beta}$ the values 0.339 and 0.327 eV/nm [38,39], respectively. Fits to the data yield exponents within the predicted range of $\alpha = 1/3$ (solid) to $\alpha = 1/2$ (dash). Although there are insufficient data to distinguish between these two values,⁴ the results do show that R influences the fluctuations, providing further evidence that effects of crystal confinement govern the behavior of $G(t)$.

The fluctuations of facet edges evidently belong to a different universality class of dynamic scaling from that of an isolated step on a surface. In contrast to previous predictions for step exponents [1,2,40–45], this difference does not stem from the type of kinetics. Instead, the effect is predicted to result from the coupling of the step chemical potential to the fluctuations: For facet-edge fluctuations the step confinement is produced by an increase in local step chemical potential $\mu(x)$ when the step is displaced from equilibrium. The functional behavior of $\mu(x)$ results from a competition between the step-repulsions from the vicinal region and the 2D pressure of the adatom density on the facet, which in turn stems from the constraints governing the crystallite shape [13,46]. For a step symmetrically confined on a vicinal surface, the confinement corresponds to a potential that is quadratic in displacement [47]. However, for the facet-edge step, the asymmetry in the $\mu(x)$ corresponds to an asymmetric confining potential that includes a cubic term in displacement [48] and, consequently, leads to non-linear terms in the equation of motion discussed above.

5 Conclusions

The work presented here is a very good example of the interplay in statistical physics between exact results, scaling arguments, numerical simulations, and experiments. Spohn and coworkers have produced a novel, exact static result. This has motivated us to apply old scaling arguments to derive novel dynamical behaviors. In turn, the latter have opened new avenues for experimentalists to explore. And the results of experiments have motivated numerical simulations of model systems. In particular, for the first time it has been possible to observe experimental evidence for a nonlinear term in equilibrium fluctuations. The result agrees with our predictions for the case of geometrically confined fluctuations. When power-law temporal

⁴ Direct experimental observation, e.g. of the spatial correlation function on a quenched crystallite, would be needed to obtain α for facet-edge fluctuations.

correlations are measured, the measured value of the power β is significantly smaller than the unconfined exponent of $\beta = 1/8$, and is within 1σ of the predicted value of $\beta = 1/11$ for a universality class of dynamical scaling with $\alpha = 1/3$ and $z = 11/3$. Thanks to the extensions by Spohn and coworkers of earlier links between KPZ [49] behavior and the behavior of facet edges [50, 51], we were able, for the first time (to the best of our knowledge), to provide an example in which a KPZ-type equation of motion accounted for equilibrium fluctuations [26]. We were also able to experimentally verify the predictions of the theory. The experiments spurred a more detailed numerical study of the problem. As a result, it was discovered that the fluctuations and equation of motion of steps at equilibrium are very sensitive to the step environment [52], a discovery that may introduce new opportunities for controlling the fabrication of nanostructures and for understanding new aspects of their dynamic properties.

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