

Small fluctuations in epitaxial growth via conservative noise

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Received 5 May 2011, in final form 9 June 2011

Published 7 July 2011

Online at stacks.iop.org/JPhysA/44/315002

Abstract

We study the combined effect of growth (material deposition from above) and nearest-neighbor entropic and force-dipole interactions in a stochastically perturbed system of N line defects (steps) on a vicinal crystal surface in 1+1 dimensions. First, we formulate a general model of conservative white noise and derive simplified formulas for the terrace width distribution and terrace width correlations in the limit $N \rightarrow \infty$ for small step fluctuations. Our general result expresses terrace width correlations as an interplay of noise covariance and step interaction strength. Second, we apply our formalism to two specific noise models which stem, respectively, from (i) the fluctuation-dissipation theorem for diffusion of adsorbed atoms; and (ii) the phenomenological consideration of deposition-flux-induced *asymmetric* attachment and detachment of atoms at step edges. In both cases of noise, we find that terrace width correlations decay exponentially with the step number difference; this behavior leads to vanishing correlations in the macroscopic limit. Our analysis may be used to (i) determine the noise in quasi-one-dimensional surfaces and (ii) assess the validity of previous mean field approximations.

PACS numbers: 81.15.Aa, 05.10.Gg, 68.35.-p, 05.40.-a, 68.43.Jk

1. Introduction

A central question in nonequilibrium statistical mechanics is: how do *large-scale* evolution laws *emerge* from the deterministic or stochastic dynamics of *many-particle* systems [1]?

Descriptions of linkages of particle models to full continuum theories lie at the heart of computational physics [2]. Related themes of modeling and analysis date back to classic works for the Boltzmann equation; see, e.g., [3–6].

Epitaxial growth consists of out-of-equilibrium processes that occur on crystal surfaces, usually in the presence of material deposition from above. Such processes accompany the fabrication of nanoscale structures, e.g. quantum wires and dots [7]. In particular, *vicinal* surfaces are characterized by nanoscale terraces oriented in the high-symmetry direction; the terraces are separated by line defects (steps) of atomic size. For macroscale surface regions at not-too-high temperatures, the steps can be considered as monotone (of the same ‘sign’), with their number fixed by the miscut angle set by the experiment [8].

In this paper, we address the question: how do terraces fluctuate on a *macroscopic* vicinal surface in 1+1 dimensions? Our goal is to study the joint effect on the macroscopic behavior of noise and (microscopic) interactions of N steps in the presence of growth. The noise is white and conservative, and is subject to geometric-type constraints: the total length of the sample does not fluctuate and the covariance of terrace widths is finite. Although our formalism is reasonably general for 1+1 dimensions, we restrict attention to two specific models consistent with our constraints. One noise model comes from an application of the fluctuation-dissipation theorem to the density of adsorbed atoms (adatoms) [9]. Another model phenomenologically accounts for deposition-flux-induced fluctuations in the number of atoms that stick to steps [10]. For *small fluctuations*, the resulting stochastic differential equations (SDEs) are linearized around the average terrace width. We solve these equations for the terrace width distribution or density (TWD) and terrace width *pair* correlations as $N \rightarrow \infty$.

Our most general result is a formula that expresses terrace width correlations as an interplay between step interactions and noise covariance. We then apply our formula to the noise models mentioned above and show that in both cases terrace width correlations decay exponentially with the step number difference. The two noise models differ in that the first inhibits step bunching, whereas the second favors it. By taking the limit of vanishing step height, $a \rightarrow 0$, we show that terrace width correlations vanish, so that the details of noise correlations do not matter in the macroscopic limit. In principle, our results can be used to derive the covariance structure of noise in quasi-one-dimensional (1D) systems, given experimentally obtained (microscopic) terrace width correlations. We apply these findings to assess the validity of previous mean field approximations.

By the Burton–Cabrera–Frank (BCF) theory [11], the major kinetic processes incorporated into the deterministic model are (a) diffusion of adatoms on terraces; (b) attachment and detachment of atoms at step edges; and (c) external deposition with (given) flux, F . To simplify the analysis, we consider straight (kink-free) steps and impose diffusion-limited (DL) kinetics, in which the diffusion of adatoms on terraces is the slowest process. In this case, the adatom density at each step edge attains an equilibrium value [8]. In addition, the steps are allowed to interact entropically and as force dipoles [12, 13].

Our analysis forms part of a relatively long sequence of works in stochastic step dynamics. The setting was motivated by the experimental issue how to control the TWD by material deposition [14, 15]. Elements of our analysis were inspired by [9, 16]. In particular, reference [16] predicted the narrowing of the TWD during growth via ad hoc white noise, neglect of step interactions and application of a mean field approximation in 1+1 dimensions. This prediction compared favorably with kinetic Monte Carlo simulations in two space dimensions (2D) [16].

Earlier systematic theories, e.g. [9, 17, 18], make use of the fluctuation-dissipation theorem for the adatom density and flux in order to determine the noise, invoke richer step energetics and kinetics, and focus on effects of step meandering in 2+1 dimensions. In particular, in [9] the authors point out that sufficient increase of the deposition flux causes a morphological step

instability driven by adatom diffusion, which is distinct from effects of statistical fluctuations. Other studies include the phenomenological approaches of [10, 19] (see also [20]), where the noise models appear to have an empirical character.

Here, we aim to enrich the analysis of [16] in three ways. First, we include step interactions jointly with material deposition from above. Second, we use different models of noise. For instance, we account for a conservative noise consistent with the fluctuation-dissipation theorem in the spirit of [9]. As an alternative, we entertain the scenario of an F -dependent conservative white noise, following [10]. We show how these choices may tone down the F -dependence of the TWD predicted in [16]. Third, we describe terrace width pair correlations, thus not limiting our attention to the TWD. However, we leave out step meandering at this stage, in contrast to the more realistic (yet much more elaborate) treatment of [9, 17, 18]. One of our purposes is to quantify the connection of noise structure to terrace width correlations at both the microscale and the macroscale in a setting sufficiently simple, i.e. in 1D, to enable tractable computations.

Despite its 1D character, our linearized stochastic model with interactions can be viewed as prototypical for a class of *asymmetric* discrete processes in the limit of large N . The kinetic process for the motion of terraces is asymmetric because of a drift (average lateral step velocity) proportional to F (see section 2.2) [16, 21, 22]. This convective effect, although negligibly small in many material systems, is retained here for the sake of a more general treatment (in 1D). As a result, on every terrace the flux of deposited atoms to an upstep is different from the flux at a downstep. Similar kinetic effects arise from the Ehrlich–Schwoebel barrier [9, 23–27], electromigration currents [28–30], differences of atomistic origin in attachment rates [26, 31] and impurities [32, 33].

From a kinetic theory view, our work forms an extension of the treatment in [34–36] where the mean field approach of [16] is placed on the grounds of Bogoliubov–Born–Green–Kirkwood–Yvon (BBGKY) hierarchies for joint probability densities of terrace widths. In [34], the steps are energetically non-interacting, and the white noise is non-conservative. In [35], the steps are interacting, albeit in the absence of deposition (with $F = 0$). Both step interactions and nonzero flux F are discussed briefly in [36], which only reports (without derivations) the TWD from an ad hoc second-order conservative noise. Here, we consider different, physically inspired and more transparent models of noise and, in addition, describe terrace width pair correlations. Moreover, we discuss discrepancies of some of our results from a mean field outcome.

Assuming small fluctuations of each terrace, induced by ‘small’ noise terms, we linearize the SDEs. The ensuing terrace width stochastic process is Gaussian. The average terrace width is fixed at its initial value (consistent with the misorientation of the vicinal surface). We compute the terrace width covariance matrix analytically by allowing $N \rightarrow \infty$ while keeping the time t independent of N (see section 3). In particular, we derive relatively simple formulas for the terrace width covariance matrix in the steady state. By comparing our result for the terrace width variance to the mean field approach introduced in [16] and further discussed in [34], we indicate the role of the noise structure in terrace width pair correlations in the steady state; see sections 4.1 and 4.2. Plausible implications of our predictions are discussed in section 4.3.

Our model is limited in its applicability. One limitation is due to the assumed 1D geometry. Because steps are straight, meandering is suppressed, and the noise for terrace widths has a relatively simple form. This setting contrasts the 2D geometry invoked, e.g., in [9, 17, 18]. For instance, in [9, 18], Langevin forces are added to both the adatom diffusion equation and the boundary conditions for atom attachment–detachment at curved steps. In [17], the noise is white in both time and the space coordinate along the step edge. Here, we resort to a tradeoff.

On the one hand, we analyze a tractable geometry. On the other hand, we exclude some realistic effects such as step meandering.

Another limitation, which is a consequence of linearizing step interactions for small fluctuations, is that steps can cross. This feature is, of course, unphysical. However, it has a negligibly small likelihood provided the TWD variance is small (see section 4.4).

Throughout this paper, we assume familiarity of the reader with basic concepts of epitaxial systems. For reviews on the subject, the reader may consult, e.g., [7, 8, 37–39].

The remainder of our paper is organized as follows. In section 2, we formulate generally the SDEs for 1D step motion in the presence of growth, step interactions and conservative noise. We solve the linearized SDEs and give general formulas for the TWD and terrace width covariance matrix. In section 3, we compute the TWD and terrace width covariance matrix under two specific choices for the noise. In section 4, we discuss implications of our results: relation of noise structure to correlations, comparison to a mean field approach, validity of our model and plausible connections to experiment. The appendices provide technical details needed in the main text.

Notation and terminology. We apply the Einstein summation convention, in which summation is implied by repeated indices, unless we state otherwise. The symbol $\mathcal{B}(t)$ denotes Brownian motion, while $\eta(t) = d\mathcal{B}/dt$ is white noise (where the time derivative is interpreted in the sense of distributions). The symbol $\mathbb{E}[X]$ denotes the average of the random variable X ; the terms ‘mean’ and ‘expectation’ are also used for \mathbb{E} interchangeably. Matrices and vectors are boldface. The matrix \mathbf{C} is represented by $C_{k,l}$, indicating the entry at the k th row and l th column. The norm squared of the $N \times N$ circulant matrix \mathbf{C} is $|\mathbf{C}|^2 = \sum_{l=0}^{N-1} |C_{0,l}|^2 = \sum_{k=0}^{N-1} |C_{k,0}|^2$. By $f = \mathcal{O}(g)$, we imply that f/g is bounded as a parameter or variable approaches an extreme value. The term ‘deposition flux’ refers to F , i.e. material deposition from above; and ‘adatom flux’ refers to the current due to diffusion of adatoms on terraces.

2. Formulation and general results in 1+1 dimensions

In this section, we formulate SDEs for terrace widths in 1+1 dimensions. Adatom diffusion is the main process driving the motion of steps by mass conservation. Atoms that migrate to the terrace boundaries attach to or detach from step edges; as a result, steps advance or retreat. Force-dipole and entropic step interactions (between nearest-neighbor steps) determine the equilibrium adatom density at each step edge, which in turn influences the net mass flux toward a step.

Because we consider *continuous* adatom densities, we also introduce Gaussian noise to account for microscopic deviations from continuum motion [40]. For a sufficiently weak noise term, we linearize the SDEs and derive general formulas for the TWD and terrace width covariance matrix.

2.1. Deterministic model

Let us begin with elements of the deterministic motion of terraces and steps. The geometry consists of straight steps of height a at $x = x_i(t)$ (see figure 1). The i th terrace is the region $x_i < x < x_{i+1}$, where $w_i(t) := x_{i+1}(t) - x_i(t) > 0$ and $i = 0, \dots, N-1$. Apply screw periodic boundary conditions so that steps are mapped onto point particles on a ring. We set $w_i(0) = \varpi$, where ϖ is a constant.

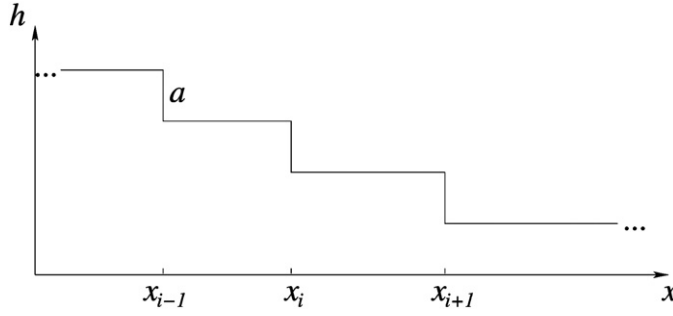


Figure 1. Schematic (cross section) of step geometry: $x = x_i(t)$ is the i th step position, a is the step height and h is the surface height.

The formulation of equations for $x_i(t)$ is outlined in [34] and summarized here with a more precise description of step interactions. In the presence of material deposition from above, steps have a typical (drift) velocity $v = Fa\varpi$, where F is the deposition rate. By a Galilean transformation to the comoving frame [16, 22], the adatom concentration $\varrho_i(x, t)$ on the i th terrace satisfies $(D\partial_{\tilde{x}}^2 + v\partial_{\tilde{x}})\varrho_i + F = \partial_{\tilde{t}}\varrho_i$, where D is the terrace diffusion constant and $(\tilde{x}, \tilde{t}) = (x - vt, t)$. By the quasi-steady approximation, we set $\partial_{\tilde{t}}\varrho_i \approx 0$, which holds if deviations of the actual step velocity from v are much smaller than the diffusive speed D/ϖ . Now remove the tildes for ease of notation ($\tilde{x} \rightarrow x$).

By linear kinetics, the atom attachment–detachment at the steps bounding the i th terrace is expressed by [8] $-J_i(x_i) = \kappa [\varrho_i(x_i) - \varrho_i^{\text{eq}}]$ and $J_i(x_{i+1}) = \kappa [\varrho_i(x_{i+1}) - \varrho_{i+1}^{\text{eq}}]$, where $J_i(x) = -D\partial_x\varrho_i - v\varrho_i$ is the adatom flux on the i th terrace, ϱ_i^{eq} is the equilibrium adatom concentration at the i th step edge and κ is a constant rate. The quantity ϱ_i^{eq} encapsulates energetics, e.g. force-dipole step interactions [8, 12, 13]. Distinct rates κ_u, κ_d for up- and down-step edges (Ehrlich–Schwoebel effect [23]) can also be included.

We enforce the conditions $v/\kappa \ll 1$ and $D/\kappa \ll \varpi$, which amount to DL kinetics⁵. This means that we formally let $\kappa \rightarrow \infty$ in the attachment–detachment conditions at step edges, so that $\varrho_i(x_i) \rightarrow \varrho_i^{\text{eq}}$ since the flux is finite [16].

By mass conservation, each step advances or retreats in response to the net mass flux incident on it. Thus, the step velocity reads $\dot{x}_i = dx_i/dt = (\Omega/a)[J_{i-1}(x_i) - J_i(x_i)]$, where Ω is the atomic area, $\Omega \approx a^2$. By solving the diffusion equation for ϱ_i (treating the positions x_i and densities ϱ_i^{eq} as fixed), and thus determining $J_i(x)$, we obtain a system of ordinary differential equations (ODEs) for $x_i(t)$, and in turn for $w_i(t)$ [34]:

$$\begin{aligned} \dot{w}_i = \frac{dw_i}{dt} = \frac{aF}{2} & \left\{ \frac{w_{i+1}e^{\frac{vw_{i+1}}{2D}}}{\sinh(\frac{vw_{i+1}}{2D})} - \frac{2w_i \cosh(\frac{vw_i}{2D})}{\sinh(\frac{vw_i}{2D})} + \frac{w_{i-1}e^{-\frac{vw_{i-1}}{2D}}}{\sinh(\frac{vw_{i-1}}{2D})} \right. \\ & + a\varpi \left[\varrho_{i+2}^{\text{eq}} \frac{e^{\frac{vw_{i+1}}{2D}}}{\sinh(\frac{vw_{i+1}}{2D})} - \varrho_{i+1}^{\text{eq}} \left(\frac{e^{-\frac{vw_{i+1}}{2D}}}{\sinh(\frac{vw_{i+1}}{2D})} + \frac{2e^{\frac{vw_i}{2D}}}{\sinh(\frac{vw_i}{2D})} \right) \right. \\ & \left. \left. + \varrho_i^{\text{eq}} \left(\frac{2e^{-\frac{vw_i}{2D}}}{\sinh(\frac{vw_i}{2D})} + \frac{e^{\frac{vw_{i-1}}{2D}}}{\sinh(\frac{vw_{i-1}}{2D})} \right) - \varrho_{i-1}^{\text{eq}} \frac{e^{-\frac{vw_{i-1}}{2D}}}{\sinh(\frac{vw_{i-1}}{2D})} \right] \right\}, \end{aligned} \quad (1)$$

where $i = 0, 1, \dots, N - 1$. It remains to express each ϱ_i^{eq} in terms of positions x_i .

⁵ For $v = \mathcal{O}(1) > 0$, a more precise condition on v reads $v/\kappa \ll \tanh[v\varpi/(2D)]$.

The step interactions are introduced explicitly in the i th-step chemical potential, μ_i , through the relation $\varrho_i^{\text{eq}} = \rho_0(1 + \mu_i/T)$, where ρ_0 is the equilibrium adatom density on a terrace and T is the Boltzmann energy [8]. If $E_N(\{x_i\})$ is the energy per unit length of the step train, we have $\mu_i = \Omega(\partial E_N/\partial x_i)$. For entropic and force-dipole interactions of strength \check{g} , this E_N reads [8, 12, 13]

$$E_N = \check{g} \sum_i \left(\frac{a}{w_i}\right)^2 \Rightarrow \mu_i = \check{g}a \left[\left(\frac{a}{w_i}\right)^3 - \left(\frac{a}{w_{i-1}}\right)^3 \right] \quad (\check{g} > 0). \quad (2)$$

Equation (1) is rewritten accordingly by eliminating all of the ϱ_i^{eq} in favor of w_i . We leave this task to the interested reader. If $w_i(0) = \varpi$, then $w_i(t) \equiv \varpi$ is a stable (equilibrium) solution for all $t > 0$.

2.2. Stochastically perturbed system and linearization

Next, we add noise to ODEs (1) by writing

$$\dot{w}_i(t) = \tilde{A}_i(w_{i-2}, w_{i-1}, w_i, w_{i+1}, w_{i+2}) + \varpi \tilde{Q}_{i,j} \tilde{\eta}_j, \quad w_i(0) = \varpi, \quad (3)$$

where $\tilde{A}_i(w_{i-2}, w_{i-1}, w_i, w_{i+1}, w_{i+2})$ is the right-hand side of (1) in view of (2); the matrix \tilde{Q} is circulant and has units of time raised to the power $-1/2$; and $\tilde{\eta}$ is a vector Gaussian white noise, i.e. $\tilde{\eta}_i = d\mathcal{B}_i/dt$. The matrix \tilde{Q} in principle depends on $\{w_i\}$ and $\{\tilde{\eta}_i\}$, but here we consider only constant \tilde{Q} . The factor ϖ multiplying \tilde{Q} has been extracted for later algebraic convenience. The components $\tilde{\eta}_j$ obey the relation $\mathbb{E}[\tilde{\eta}_i(t) \tilde{\eta}_j(t')] = \delta_{i,j} \delta(t - t')$, so that the matrix \tilde{Q} controls the covariance structure⁶ of the noise. In this section, we leave the noise matrix \tilde{Q} otherwise unspecified in order to derive general results. In section 3, we consider two special cases of noise and their implications for the TWD and terrace width pair correlations.

In the limit of a sufficiently small $\varpi^2 |\tilde{Q}|^2$, we treat the noise as a perturbation of (1). Because stable solutions of (1) respond linearly to small perturbations, we set $w_i(t) = \varpi[1 + \xi_i(t)]$, $|\xi_i| \ll 1$. By (1) and (2), the linearized version of (3) is

$$\dot{\xi}_i = aF\{(1-p)(\xi_{i+1} - \xi_i) + p(\xi_i - \xi_{i-1}) + g[-(1+\beta)(\xi_{i+2} - 3\xi_{i+1} + 3\xi_i - \xi_{i-1}) + \beta(\xi_{i+1} - 3\xi_i + 3\xi_{i-1} - \xi_{i-2})]\} + \tilde{Q}_{i,j} \tilde{\eta}_j, \quad (4)$$

where (abusing notation) we use the same symbol (ξ_i) for the approximation of ξ_i , i.e. the solution of the linearized equations. The parameters p , β and g are defined by

$$p = \frac{1}{2} \left(\frac{\nu}{\sinh^2 \nu} - \frac{e^{-\nu}}{\sinh \nu} \right), \quad \beta = \frac{1}{2} \frac{e^{-\nu}}{\sinh \nu}, \quad g = 3 \frac{\check{g}a^2}{T} \tilde{m}_0^3 \rho_0, \quad (5)$$

with $\nu = v\varpi/(2D)$, $\tilde{m}_0 = a/\varpi$ and $0 < p < 1/2$; cf (34) in [34] where $g = 0$. Note that $\xi_i = (w_i - \varpi)/\varpi$ is a stochastic process. We assume that fluctuations are small in probability: $1 - \Pr[\sup_{t>0} |\xi_i(t)| \ll 1] \ll 1$ for all i (\Pr denotes the probability).

Let us pause for a moment and take a closer look at (4). For $g = 0$ (no step interaction), this equation reduces to $\dot{w}_i \approx Fa[(1-p)(w_{i+1} - w_i) + p(w_i - w_{i-1})]$. We can consider p as the fraction of deposited atoms that attach to the downstep of terrace i , in the setting of figure 1; $1-p$ is the fraction of atoms that move to an upstep. Hence, the number of atoms per unit time that cause an increase of the i th terrace size is $pF(w_i - w_{i-1})$ by competition with the upper terrace and $(1-p)F(w_{i+1} - w_i)$ from the lower terrace. Here we ignore the

⁶ For a vector random variable, \mathbf{X} , the phrase ‘covariance structure’ is used to mean the structure of the matrix $\mathbb{E}[X_i X_j]$ (X_i : i th component of \mathbf{X}). We consider whether this matrix (i) is conservative (i.e. the sum of elements in any row or column is zero); (ii) is sparse; and/or (iii) has positive or negative off-diagonal elements. In this paper, we consider $X_i = \varpi \tilde{Q}_{i,j} \tilde{\eta}_j$ and $X_i = (w_i - \varpi)/\varpi$; cf (4).

possibility of nucleation. For a similar model, see [19] in regard to step motion in a diffusion bias (and [20] for an effect of impurities).

2.3. TWD and pair correlations: general formulas

In this section, we solve SDEs (4) with particular emphasis on the pair correlation for terrace widths. Equation (4) is fully non-dimensionalized via $t \mapsto \tau = tFa$ and $\tilde{\eta}_i(t) \mapsto \eta_i(\tau) = (1/\sqrt{Fa})\tilde{\eta}_i(t)$, $\tilde{Q} \mapsto Q = (1/\sqrt{Fa})\tilde{Q}$. SDEs (4) are then recast to the matrix form

$$\frac{d\xi}{d\tau} = -A \cdot \xi + Q \cdot \eta, \tag{6}$$

where $\xi = (\xi_0, \dots, \xi_{N-1})$, $\eta = (\eta_0, \dots, \eta_{N-1})$ and A is a sparse *circulant* matrix with first-row entries $[1 - 2p + 3g(1 + 2\beta), -1 + p - g(3 + 4\beta), g(1 + \beta), 0, \dots, 0, g\beta, p - g(1 + 4\beta)]$. The (non-dimensional) matrix Q is only required to be circulant and constant.

By integrating (6), we obtain

$$\xi(t) = \int_0^\tau e^{-A(\tau-s)} Q \cdot \eta(s) ds, \quad \tau = tFa. \tag{7}$$

Evidently, for every $t > 0$, (7) describes a vector Gaussian random variable with zero expectation. The joint probability density of any number of terrace widths is Gaussian. In light of the initial condition $\xi(0) = 0$, the covariance matrix is circulant, given by the expression (see appendix A)

$$\begin{aligned} \mathfrak{C}_m(t(\tau); N) &:= N^{-1} \mathbb{E}[\xi_j(t) \xi_{j+m}(t)] \\ &= \frac{1}{N} \int_0^\tau ds \int_0^\tau ds' \mathbb{E}\{\eta_j(s') [e^{-A^T(\tau-s')}]_{j,k} Q_{k,l}^T Q_{l,n} [e^{-A(\tau-s)}]_{n,p} [\eta(s)_m^c]_p\}, \end{aligned} \tag{8}$$

where $[\eta(s)_m^c]$ is the m th cyclic permutation of the vector η ,⁷ and the symbol A^T denotes the transpose of A . Recall that $\mathbb{E}[\eta_j(s) \eta_k(s')] = \delta_{j,k} \delta(s - s')$. In the case with $m = 0$, (8) reduces to a formula for the terrace width variance, namely

$$\mathfrak{C}_0(t; N) = \mathbb{E}[\xi_j^2(t)] = N^{-1} \mathbb{E}[\xi(t)^2] = \int_0^{tFa} |e^{-As} Q|^2 ds. \tag{9}$$

The probability density for a single (scaled) terrace width, ξ_i , is⁸

$$P(\xi, t; N) = \frac{1}{\sqrt{2\pi} \mathfrak{C}_0(t; N)} \exp\left[-\frac{\xi^2}{2\mathfrak{C}_0(t; N)}\right], \quad -\infty < \xi < \infty, \tag{10}$$

by which the TWD (for the dimensional terrace width, w_i) is obtained via $\xi = (w - \varpi)/\varpi$, $-\infty < w < \infty$. Within this approximation, $\Pr[w_i < 0] > 0$; there exists a nonzero probability of step crossing. This likelihood can be controlled by the step interaction strength, deposition rate and initial terrace width (see section 4.4). We henceforth consider $\Pr[w_i < 0]$ as negligibly small.

⁷ Note that ODEs (4) under the initial condition $\xi(0) = 0$ yield j -independent covariances, \mathfrak{C}_m .

⁸ By abusing notation, we use the same symbol, ξ , to denote both the independent real variable of the TWD and the stochastic process associated with each terrace width.

2.4. Covariance matrix: large- N limit

Next, we compute \mathfrak{C}_m by (8). First, we derive an equivalent expression valid for finite N and t . Second, we enforce the limit $N \rightarrow \infty$ and thereby extract a single-integral formula for $\mathfrak{C}_m(t) := \mathfrak{C}_m(t; \infty)$. We then simplify this formula by taking $\mathcal{O}(N) > tFa \gg 1$; tFa is treated as large yet independent of N .

By a property of circulant matrices (see appendix A), we write (8) as

$$\begin{aligned} \mathfrak{C}_m(t; N) &= \int_0^\tau N^{-1} \sum_{k=0}^{N-1} \vartheta_k e^{-\lambda_k s} \cos\left(\frac{2\pi km}{N}\right) ds \\ &= N^{-1} \sum_{k=0}^{N-1} \vartheta_k \lambda_k^{-1} (1 - e^{-\lambda_k \tau}) \cos\left(\frac{2\pi km}{N}\right), \end{aligned} \quad (11)$$

where λ_k and ϑ_k are the eigenvalues of $\mathbf{A} + \mathbf{A}^T$ and $\mathbf{Q}\mathbf{Q}^T$, respectively. The eigenvalues of (square) circulant matrices can be evaluated directly via the discrete Fourier transform [41]. Since both \mathbf{A} and \mathbf{Q} are circulant, $\mathbf{A} + \mathbf{A}^T$ and $\mathbf{Q}\mathbf{Q}^T$ are both circulant and symmetric. A simple calculation reveals that λ_k and ϑ_k share the same general form: they are given by⁹

$$\lambda_k = r_0 + 2 \sum_{j=1}^M r_j \cos\left(\frac{2\pi jk}{N}\right), \quad \vartheta_k = s_0 + 2 \sum_{j=1}^M s_j \cos\left(\frac{2\pi jk}{N}\right). \quad (12)$$

The coefficients r_j and s_j depend on the precise forms of \mathbf{A} and \mathbf{Q} , respectively. We specify these coefficients later. In (12), we define $M = (N - 1)/2$ for odd N , and $M = (N - 2)/2$ for even N .

By taking the limit $N \rightarrow \infty$, the sum in (11) becomes an integral:

$$\mathfrak{C}_m(t) = \mathfrak{C}_m(t; \infty) = \int_0^1 dy \vartheta(y) \lambda(y)^{-1} [1 - e^{-\lambda(y)\tau}] \cos(2\pi my), \quad (13)$$

where $\vartheta(y)$ and $\lambda(y)$ are the limits of ϑ_k and λ_k as $N \rightarrow \infty$, and y is the continuous variable replacing k/N ; $0 < y < 1$. The variance of the TWD is found by setting $m = 0$ in (13), which readily produces the result given in [35].

We comment on the interpretation of $\lambda(y)$ and $\vartheta(y)$. When N is finite, the sequences $\{\lambda_k\}$ and $\{\vartheta_k\}$ contain information about nearest-neighbor interactions and noise correlations, respectively. In particular, each nonzero coefficient r_j (s_j) in (12) arises from the existence of interactions (noise correlations) between a given terrace and its j th nearest neighbor on each side. In the limit $N \rightarrow \infty$, formulas (12) become continuous Fourier decompositions and $k/N \rightarrow y$ measures the surface height difference between two terraces. The continuous functions $\lambda(y)$ and $\vartheta(y)$ can yield information about the range of interactions and noise correlations as measured by the continuous variable y . If the largest index j' of the nonzero coefficients r_j (or s_j) becomes $j' = \mathcal{O}(N)$, the interactions (or noise correlations) extend to finite, $\mathcal{O}(1)$ values of y as $N \rightarrow \infty$.¹⁰

⁹ This property follows directly from the method of calculating the eigenvalues of a circulant matrix. Specifically, if \mathcal{C} is circulant, its eigenvalues c_k are given by $c_k = F_{k,l} \mathcal{C}_{l,0}$, where F is the matrix consisting of the discrete Fourier transform. Note that $F_{j,k} = \exp(-2\pi i jk/N)$, where $i^2 = -1$. If, in addition, \mathcal{C} is symmetric, the formula for c_k (where c_k is λ_k or ϑ_k) reduces to (12).

¹⁰ In the discrete setting (with finite N), the j th nearest neighbor is identified with the terrace that is j atomic heights below (or above) a given terrace. Consequently, the j th nearest neighbor amounts to a height difference given by the limiting value ja as $a \downarrow 0$, with Na kept fixed. If $j = \mathcal{O}(N)$, the limiting value of ja corresponds to a nonzero, finite height difference along the step train. If $j = \mathcal{O}(1)$, the limiting value of ja corresponds to a practically zero height difference.

In section 3, we consider two cases in which the maximum index j' appearing in (12) is fixed and $\mathcal{O}(1)$ in the limit of large N ; interactions and noise correlations are thus restricted to a few nearest neighbors for a given terrace site. This assumption leads to terrace width pair correlations that decay *exponentially* with respect to the nearest-neighbor index m : $\mathfrak{C}_m \propto c^{|m|}$, $0 < c < 1$ (where c is defined in section 3).

3. Particular cases of noise

In this section, we find the terrace width covariance matrix for two different forms of the noise matrix \mathbf{Q} . This procedure is motivated by the observation that the *origin* of noise should in principle depend on whether or not there is significant deposition.

In section 3.1, we introduce noise at the level of adatom diffusion, assuming that the system is sufficiently close to equilibrium. The noise coefficients are determined by the fluctuation-dissipation theorem [9]. By solving the free-boundary problem for step motion, we then obtain a second-order conservative noise for the motion of terraces. In section 3.2, we consider an asymmetric, first-order conservative noise whose amplitude depends on the deposition flux (F), based on a proposal in [10]: the noise coefficient is chosen to reflect (phenomenologically) the asymmetric attachment of atoms at step edges in correspondence to the drift velocity, $v = Fa\varpi$. Loosely speaking, this latter noise should apply to a system far enough from equilibrium so that near-equilibrium statistics do not hold for adatoms.

3.1. Adatom noise under fluctuation-dissipation theorem

In this section, we derive \mathbf{Q} by adding noise to the adatom equations of motion and solving the free boundary problem for terraces. Let us begin with a modified diffusion equation for adatoms in the step comoving frame, $(D\partial_x^2 + v\partial_x)q_i(x) + F - \partial_x q(x, t) = 0$ for terrace i , where we applied $\partial_t q_i \approx 0$ (quasistatic approximation). The noise source $q(x, t)$ is delta-correlated in time and space, and contributes to a random adatom flux [9]. Since we consider a (continuous) adatom density in the BCF model [11], q is intended to capture the effect of microscopic motions, which are usually left out of a deterministic continuum picture for terraces. The x -derivative acting on q ensures that the resulting noise is conservative (consistent with Fick's law for adatom diffusion).

The diffusion equation is supplemented with corresponding, modified boundary conditions: $-J_i(x_i) = \kappa[q_i(x_i) - \varrho_i^{\text{eq}} + \zeta_i^+]$ and $J_i(x_{i+1}) = \kappa[q_i(x_{i+1}) - \varrho_{i+1}^{\text{eq}} + \zeta_{i+1}^-]$, where $J_i(x) = -D\partial_x q_i - vq_i + q(x)$ is the modified adatom flux on the i th terrace. The terms ζ_i^\pm are Langevin forces accounting for random attachment and detachment events on the right (+) and left (−) sides of the i th step edge [9]. The terms ϱ_i^{eq} and κ are discussed in section 2.1.

In order to determine the covariance structure of ζ_i^\pm and q , we follow [9] in applying the fluctuation-dissipation theorem. First, suppose the system is in equilibrium (i.e. with $F = 0$, since we neglect desorption), where

$$\mathbb{E}[q(x, t)q(x', t')] = 2D\rho_0\delta(x - x')\delta(t - t'), \quad (14)$$

$$\mathbb{E}[\zeta_i^\alpha(t)\zeta_j^\beta(t')] = (2\rho_0/\kappa)\delta_{i,j}\delta(t - t')\delta_{\alpha,\beta}, \quad \alpha, \beta = \pm. \quad (15)$$

If the system is sufficiently close to equilibrium, one may replace ρ_0 by the average of $q_i(x)$. To simplify the analysis, we assume that corrections arising from this substitution are negligible and henceforth consider only the noise defined by (14) and (15).

By solving the diffusion equation in the limit of DL kinetics, one can find linearized SDEs for terrace motion that have the same form as (4), with the exception of the noise term. This term is now modified according to the replacement

$$\tilde{Q}_{i,j}\tilde{\eta}_j \rightarrow 2M_i(w_i) - M_{i-1}(w_{i-1}) - M_{i+1}(w_{i+1}), \quad (16)$$

where

$$M_i(w_i, t) = \frac{\tilde{m}_0 v e^{vw_i/2D}}{2D \sinh(vw_i/2D)} \int_{x_i}^{x_{i+1}} dz e^{-v(x_{i+1}-z)/D} q(z, t). \quad (17)$$

In the DL case, contributions from ζ^\pm do not appear in (17) since the DL limit, where $\kappa \rightarrow \infty$, implies $\mathbb{E}[\zeta_i^\pm \zeta_j^\pm] \rightarrow 0$ (cf (15)).

In the small fluctuation limit, we approximate $w_i \approx \varpi$ in the definition of $M_i(w_i, t)$.¹¹ Thus, the $M_i(w_i, t) \approx M_i(\varpi, t)$ can be treated as independent Gaussian random variables whose variances depend on the average terrace width. The noise is reduced to the form given in (4):

$$\tilde{Q}_{i,j}\tilde{\eta}_j = 2M_i - M_{i-1} - M_{i+1}, \quad (18)$$

where these M_i are independent Gaussian random variables satisfying

$$\mathbb{E}[M_i M_j] = \frac{\tilde{m}_0^2 v \rho_0 \sinh(2v)}{2(\sinh v)^2} \delta_{i,j} \delta(t - t'). \quad (19)$$

After some algebra, we obtain formulas for the eigenvalues of the matrices $\mathbf{A} + \mathbf{A}^T$ and $\mathbf{Q}\mathbf{Q}^T$ (defined in sections 2.2 and 2.3):

$$\lambda_k = 2 \left[1 - 2p + 2g(1 + 2\beta) \left(1 - \cos \frac{2\pi k}{N} \right) \right] \left(1 - \cos \frac{2\pi k}{N} \right), \quad (20)$$

$$\vartheta_k = \frac{2\tilde{m}_0^2 \varpi \rho_0 \sinh(2v)}{(\sinh v)^2} \left(1 - \cos \frac{2\pi k}{N} \right)^2. \quad (21)$$

With these definitions of ϑ_k and λ_k , we apply (13) to calculate the covariance matrix (for $|m| = 0, 1, \dots$):

$$\mathfrak{C}_m(t) = \frac{\tilde{m}_0^2 \varpi \rho_0 \sinh(2v)}{(\sinh v)^2} \int_0^1 dy \frac{[1 - \cos(2\pi y)] \cos(2\pi m y)}{(1 - 2p) + 2g(1 + 2\beta)(1 - \cos(2\pi y))} \times \{1 - e^{-\tau[2(1-2p)+4g(1+2\beta)(1-\cos(2\pi y))(1-\cos(2\pi y))]} \}. \quad (22)$$

We have not been able to compute this $\mathfrak{C}_m(t)$ in simple closed form for finite t . In the limit $t \rightarrow \infty$, the exponential term in the integrand of (22) can be dropped (see appendix B); thus, we obtain the steady-state formulas

$$\mathfrak{C}_{0,\text{st}} = \frac{\tilde{m}_0^2 \varpi \rho_0 \sinh(2v)/(\sinh v)^2}{[(1 - 2p) + 2g(1 + 2\beta)]} \frac{\alpha + \sqrt{1 - \alpha^2} - 1}{\alpha \sqrt{1 - \alpha^2}}, \quad (23)$$

$$\mathfrak{C}_{m,\text{st}} = -\frac{\tilde{m}_0^2 \varpi \rho_0 \sinh(2v)/(\sinh v)^2}{(1 - 2p) + 2g(1 + 2\beta)} \frac{1 - \alpha}{\alpha \sqrt{1 - \alpha^2}} \left[\frac{1 - \sqrt{1 - \alpha^2}}{\alpha} \right]^{|m|}, \quad (24)$$

where $|m| \geq 1$ and $\alpha = 2g(1 + 2\beta)/[(1 - 2p) + 2g(1 + 2\beta)]$ ($0 < \alpha < 1$). Evidently, covariances decay exponentially with m .

¹¹ A formal argument for this approximation can be made by adding to ODEs (1) the ‘small’ noise $\epsilon[2M_i(w_i) - M_{i-1}(w_{i-1}) - M_{i+1}(w_{i+1})]$, where $0 < \epsilon \ll 1$. Then, use the expansion $w_i(t) = \varpi[1 + \epsilon \xi_i(t) + \dots]$. Making the substitution $w_i, w_{i\pm 1} \rightarrow \varpi$ amounts to keeping only the leading-order terms in the resulting ϵ -expansion of the SDEs.

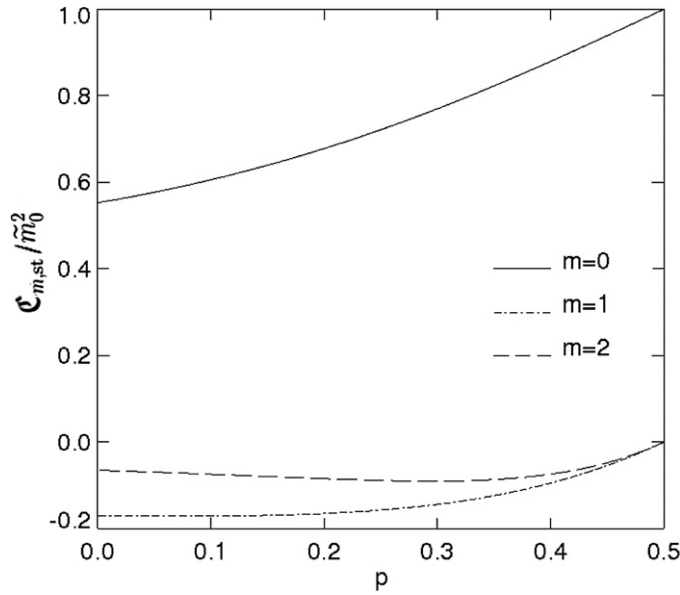


Figure 2. Steady-state, scaled covariance matrix, $\mathfrak{C}_{m,\text{st}}/\tilde{m}_0^2$, as a function of p for $m = 0$ (variance) and $m = 1, 2$ (covariance for first and second nearest neighbor). We employ $\varpi \varrho_0 = g = 1$.

Remark 1. The terrace width correlations given by (24) are all negative. This means that $\text{sgn}(\xi_i) = -\text{sgn}(\xi_{i\pm 1})$ with probability greater than $1/2$, where $\text{sgn}(x) = x/|x|$ for $x \neq 0$. Thus, short terraces are more likely to be surrounded by long terraces, and vice versa. Hence, the noise of (18) disfavors the formation of step bunches.

Remark 2. If $F \downarrow 0$ (i.e. $\nu = Fa\varpi^2/(2D) \downarrow 0$ or $p \uparrow 1/2$) and $g = \mathcal{O}(1) > 0$, by (23) the steady-state variance approaches a nonzero value: $\mathfrak{C}_{0,\text{st}} \rightarrow \tilde{m}_0^2 \varpi \rho_0 / g$; see figure 2 recalling that, by (5), $\beta = \beta(p)$ through ν . By (24), off-diagonal elements of the steady-state covariance (circulant) matrix approach zero; that is, $\mathfrak{C}_{m,\text{st}} \rightarrow 0$ as $F \downarrow 0$ for $m \neq 0$. The above result for $\mathfrak{C}_{0,\text{st}}$ is in agreement with the long-time limit of the TWD found in [35], where a similar model is invoked without deposition flux (for $F = 0$). In the limit $F \rightarrow \infty$, by (23) and (24) we compute

$$\mathfrak{C}_{0,\text{st}}|_{F \rightarrow \infty} = \frac{2\tilde{m}_0^2 \varpi \rho_0}{1 + 2g} \left[\frac{\alpha_* + \sqrt{1 - \alpha_*^2} - 1}{\alpha_* \sqrt{1 - \alpha_*^2}} \right], \quad (25)$$

$$\mathfrak{C}_{m,\text{st}}|_{F \rightarrow \infty} = \frac{-2\tilde{m}_0^2 \varpi \rho_0}{1 + 2g} \left[\frac{1 - \alpha_*}{\alpha_* \sqrt{1 - \alpha_*^2}} \right] \left[\frac{1 - \sqrt{1 - \alpha_*^2}}{\alpha_*} \right]^{|m|}, \quad |m| \geq 1, \quad (26)$$

where $\alpha_* = \lim_{F \rightarrow \infty} \alpha(F) = 2g/(1 + 2g)$; cf figure 2.

3.2. Deposition-flux-induced noise at steps

In this section, we consider a phenomenological noise that arises solely as a result of deposition, by choosing not to apply the fluctuation-dissipation theorem. Our approach is partly motivated by the fact that deposition is a non-equilibrium process. We propose a noise motivated by inspection of the linearized deterministic equations for $g = 0$, following the phenomenological

model of [10]. The idea is to consider the number of atoms arriving at each step edge as fluctuating in accord with the p -induced *asymmetry*, i.e. allow lateral fluxes that cause random noise to distinguish downsteps from upsteps.

Let $\mathcal{N}_i^\pm(t)$ be the (random) number of atoms attaching to an upstep (+) or a downstep (-) of the i th terrace. For macroscopic times and each i , we posit that the increments $\mathcal{N}_i^\pm(t_{n+1}) - \mathcal{N}_i^\pm(t_n)$ are independent, normally distributed and stationary random variables, with mean zero and variance proportional to $1 - p$ (+) or p (-) times $(t_{n+1} - t_n)Fw_i$, where $0 < t_n < t_{n+1}$.

Accordingly, we perturb (4), or (1), for the i th terrace motion by (i) the noise $\sqrt{(1-p)Fw_{i+1}\Omega}\tilde{\eta}_{i+1}$ (which has the dimension of speed) for fluctuations in the number of atoms attaching to the upstep bounding the $(i+1)$ th terrace; (ii) $\sqrt{pFw_{i-1}\Omega}\tilde{\eta}_{i-1}$ regarding the downstep of the $(i-1)$ th terrace; and (iii) $[\sqrt{pFw_i\Omega} - \sqrt{(1-p)Fw_i\Omega}]\tilde{\eta}_i$ for mass conservation purposes. See also the discussion following (4) and (5) for additional motivation. (Recall that $(\tilde{\eta}_0, \dots, \tilde{\eta}_{N-1})$ is a vector white noise, having independent, identically distributed components $\tilde{\eta}_i = d\mathcal{B}_i/dt$). We now propose the noise model¹²

$$\tilde{Q}_{i,j}\tilde{\eta}_j = \frac{a}{\varpi}\sqrt{F\varpi}[\sqrt{1-p}(\tilde{\eta}_{i+1} - \tilde{\eta}_i) + \sqrt{p}(\tilde{\eta}_i - \tilde{\eta}_{i-1})]. \quad (27)$$

With this choice, the eigenvalues of the noise matrix $\tilde{Q}\tilde{Q}^T = (1/Fa)Q\mathcal{Q}^T$ are

$$\vartheta_k = 2\tilde{m}_0 \left[1 + 2\sqrt{p(1-p)} \cos \frac{2\pi k}{N} \right] \left(1 - \cos \frac{2\pi k}{N} \right).$$

Accordingly, using (13), we derive the formula (for $m = 0, \pm 1, \pm 2, \dots$)

$$\mathfrak{E}_m(t) = \int_0^1 dy \frac{\tilde{m}_0 \cos(2\pi my) [1 + 2\sqrt{p(1-p)} \cos(2\pi y)]}{(1-2p) + 2g(1+2\beta)[1 - \cos(2\pi y)]} \times \{1 - e^{-\tau[2(1-2p)+4g(1+2\beta)(1-\cos(2\pi y))(1-\cos(2\pi y))]} \}. \quad (28)$$

As $tFa \rightarrow \infty$ with $g, \nu = \mathcal{O}(1) > 0$, the exponential term can be dropped; thus, the steady-state covariance matrix is comprised of the elements (see appendix B)

$$\mathfrak{E}_{0,\text{st}} = \frac{\tilde{m}_0}{2g(1+2\beta)\sqrt{1-\alpha^2}} [\alpha + 2\sqrt{p(1-p)}(1 - \sqrt{1-\alpha^2})], \quad (29)$$

$$\mathfrak{E}_{m,\text{st}} = \frac{\tilde{m}_0}{2g(1+2\beta)\sqrt{1-\alpha^2}} [\alpha + 2\sqrt{p(1-p)}] \left[\frac{1 - \sqrt{1-\alpha^2}}{\alpha} \right]^{|m|}, \quad (30)$$

where $|m| \geq 1$ and $\alpha = 2g(1+2\beta)/[(1-2p) + 2g(1+2\beta)]$; see figure 3.

We note in passing that, in the special (idealized) limit $g \downarrow 0$ (vanishing step interaction), the time-dependent TWD variance becomes (see appendix B)

$$\mathfrak{E}_0(t)|_{g \downarrow 0} = \frac{\tilde{m}_0}{1-2p} \{1 - e^{-\check{t}} [I_0(\check{t}) + 2\sqrt{p(1-p)} I_1(\check{t})]\}, \quad (31)$$

where $\check{t} = 2(1-2p)tFa$ and $I_n(t)$ is the modified Bessel function of n th order [42]. In the limit $\check{t} \rightarrow \infty$, we obtain $\mathfrak{E}_0(t)|_{g \downarrow 0} \rightarrow \tilde{m}_0(1-2p)^{-1}$.

Remark 3. If $\nu = Fa\varpi^2/(2D) \downarrow 0$ (i.e. $p \uparrow 1/2$) and $g = \mathcal{O}(1) > 0$, by (28) the covariance approaches *zero* for any time t and all m . This behavior is derived by use of $\beta = \mathcal{O}(\nu^{-1})$ as

¹² In (27), notice the replacement of w_i and $w_{i\pm 1}$ by the initial terrace width, ϖ , in the noise diffusion coefficients. A formal argument for this approximation can be made by adding to ODEs (1) the ‘small’ term $\epsilon\tilde{\eta}_i\sqrt{Fw_i a^2}(\sqrt{p} - \sqrt{1-p}) + \epsilon\tilde{\eta}_{i+1}\sqrt{1-p}\sqrt{Fw_{i+1} a^2} - \epsilon\tilde{\eta}_{i-1}\sqrt{p}\sqrt{Fw_{i-1} a^2}$ where $0 < \epsilon \ll 1$. Then, (27) are viewed as the lowest-order equations in ϵ .

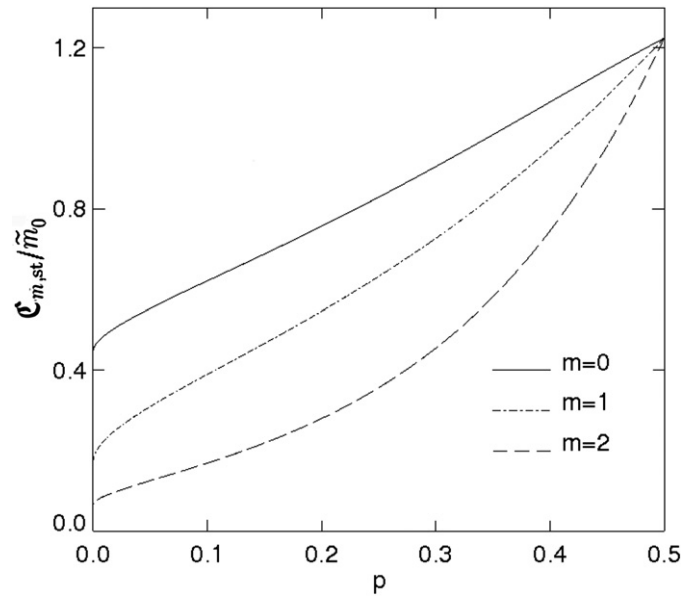


Figure 3. Steady-state, scaled covariance matrix, $\mathcal{C}_{m,st}/\tilde{m}_0$, as a function of p for $m = 0$ (variance) and $m = 1, 2$ (covariance for first and second nearest neighbor). We use $g = 1$. The narrowing of the TWD with F (decreasing p) is evident.

$\nu \downarrow 0$, and is consistent with SDEs (4), since these equations become (deterministic) ODEs in view of (27) as $F \downarrow 0$. Details for the precise role of time t in this limiting case are provided in remark 4 and appendix C; see also figure 4 for $m = 0$.

Remark 4. Interestingly, the limits $\nu \downarrow 0$ and $t \rightarrow \infty$ of (28) do *not* commute (for any m): the steady-state values (29) and (30) approach the finite value $\sqrt{3\tilde{m}_0^2/2g}$ as $\nu \downarrow 0$, whereas, by (28), $\mathcal{C}_m(t) \rightarrow 0$ as $p \uparrow 1/2$ for fixed $t > 0$ and g (cf figures 3, 4 and remark 3). To resolve this apparent paradox, we point out a transition in the asymptotics for \mathcal{C}_m if $tFa = \mathcal{O}(g\nu^{-3})$. If ν is small, \mathcal{C}_m given by (29) and (30) is recovered from (28) if aFt is large enough so that the vanishing exponential term does not suppress the emerging singularity in the integrand of (28). More precisely, for $\nu aFt \gg 1$, the major contribution to integration comes from the vicinity of $y = 0$. Thus, to extract the steady-state limits (29) and (30), we require that $1 - 2p > \mathcal{O}[g(1 + 2\beta)y^2]$ and $(1 - 2p)y^2 aFt = \mathcal{O}(1)$, by which $tFa \gg g\nu^{-3}$. By contrast, if $aFt \ll g\nu^{-3}$, then $\mathcal{C}_m(t)$ can be arbitrarily close to zero (see appendix C for further technical details).

Remark 5. Consider the steady-state variance $\mathcal{C}_{0,st}(p, g)$ of (29) for $1 - 2p > 0$. For fixed p , $\mathcal{C}_{0,st}$ decreases with g . For finite and fixed g , $\mathcal{C}_{0,st}$ increases with p and thus decreases with F . The parameter β decreases with F and becomes exponentially small (compared to unity) if $\nu = v\varpi/(2D) \gg 1$. Therefore, (30) predicts a narrowing of the TWD with increasing step interaction or deposition rate (see section 4.3).

Remark 6. The correlations given by (30) are positive, in contrast to the correlations associated with the adatom noise of section 3.1; cf (24). In the steady state, this means that $\text{sgn}(\xi_i) = \text{sgn}(\xi_{i\pm 1})$ with probability greater than $1/2$. Therefore, the noise given by (27)

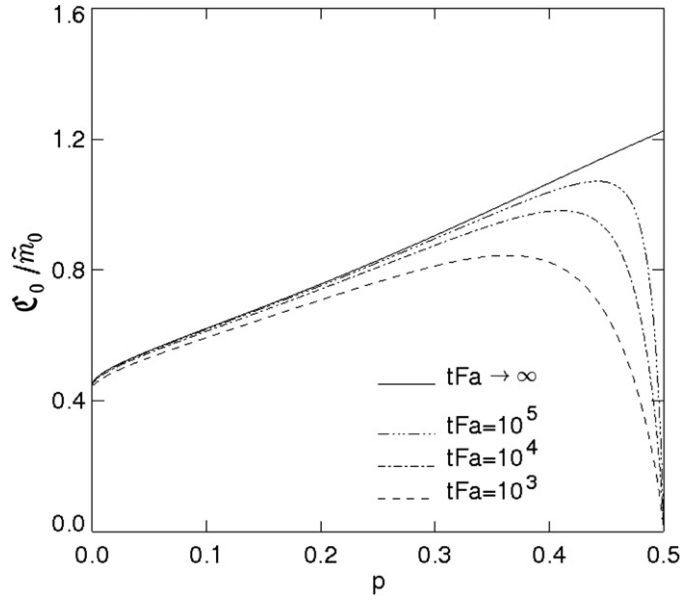


Figure 4. Time-dependent, scaled TWD variance, $c_0(t)/\tilde{m}_0$, computed by numerical integration of (28) (dashed curves) versus the steady-state variance of (29) (solid line); $g = 1$. For large tFa , the time-dependent variance converges to its steady-state value non-uniformly in p : the depicted behavior near $p = 1/2$ indicates that the limits $F \downarrow 0$ and $t \rightarrow \infty$ do not commute (cf remark 4).

gives rise to a stepped surface in which long trains of wide terraces are likely to be followed by step bunches. We emphasize that the structure of terrace width correlations does not come specifically from the flux dependence of the noise, but rather from the generic, asymmetric form of Q introduced by (27).

4. Discussion

In this section, we discuss the results of sections 2 and 3, with particular emphasis on the relationship between noise structure and terrace width correlations. We also compare the result of section 3.2 to a mean field approach [16, 34], outline a plausible connection to experiments and discuss the validity of our linearized model.

4.1. Noise structure and correlations

A major component of our work has been to determine the terrace width covariance structures that arise from specific choices of conservative noise (cf (13), (24) and (30)). For systems in equilibrium, the fluctuation-dissipation theorem allows one to calculate the structure of the noise in a system from known correlations of thermodynamic variables, and vice versa. We emphasize that application of this theorem is contingent upon knowledge of the steady state of the system, which acts as a constraint in determining the noise.

An alternate perspective, which we adopt here, is that the noise should be constrained by geometric properties of the system. First, the total system size should not fluctuate. This requirement is satisfied by choosing the noise to be conservative; that is, the sum of elements in any row or column of Q must be zero. This choice implies that summing equations (3)

over all i yields the deterministic equation $\sum_i \dot{\xi}_i = 0$, so that the total length of the system is constant. As an additional constraint, we require the covariance matrix to be bounded for long times. This requirement must be checked for each Q by taking the limit $t \rightarrow \infty$ in (13).¹³

For the two cases examined in sections 3.1 and 3.2, we find that our geometric constraints are sufficient to force the system to a steady state having a well-defined covariance matrix. Hence, our choices of noise are consistent with the fluctuation-dissipation theorem, insofar as there exist steady-state covariances that in turn imply our noises by the fluctuation-dissipation theorem. Such an application of the fluctuation-dissipation theorem does not require the system to attain or be near equilibrium.

The study of terrace width fluctuations in the BCF framework with noise provides a means of predicting how terrace width correlations should behave in a continuum description of surfaces. If we take $a = \mathcal{O}(N^{-1})$ as $N \rightarrow \infty$, then we can view the surface in terms of a continuous height profile (see figure 1 and section 2.4). The discrete solution, ξ_i , for terrace widths is mapped to a continuous variable, $\xi(h)$: $\xi_i \mapsto \xi(h)$; this $\xi(h)$ measures the relative deviation of the slope from its average value at height h (cf the discussion following (13), especially the footnote). In our formulation, $h = y$. Because the covariance matrices (24) and (30) depend on $m = h/a$, we see that values of $\xi(h)$ separated by *finite* h are *not* correlated.

This conclusion holds despite key differences in the structure and effects of the noise models used in sections 3.1 and 3.2. Specifically, the noise (16) results in terrace widths that are anti-correlated (no step bunching), while (27) leads to correlated terrace widths (step bunching). However, these detailed correlations are ‘washed out’ when ma is a macroscopic length (comparable to the size of the system), so that $|m| \gg 1$. This limit corresponds to a coarse-graining of the stepped surface.

4.2. Mean field approach and decorrelation hypothesis

Heuristically speaking, the main goal of a mean field formalism is to reduce the SDE system to a single Langevin-type equation that produces the *same* TWD as the starting, coupled system [34, 35]. For SDEs of the form

$$\dot{\xi}_i = G(\xi_{i-2}, \xi_{i-1}, \xi_i, \xi_{i+1}, \xi_{i+2}) + \sum_{k=0}^{N-1} Q_{i,k} \eta_k \quad (32)$$

(where η_k are independent white noises and $Q = [Q_{i,k}]$ is circulant), this task is pursued via the replacements $\xi_i \rightarrow \xi^{\text{mf}}$ and $\xi_{i\pm 1}, \xi_{i\pm 2} \rightarrow f(\xi^{\text{mf}}, t)$, where f is a (deterministic) field to be determined [34, 35]. In principle, this f depends on the joint probability density, p_5 , of five terraces. This p_5 satisfies a BBGKY hierarchy, which in principle involves the joint probability densities, p_n , of n terraces [35].

Since the η_k are independent, it can be argued that the term $\sum_{k=0}^{N-1} Q_{i,k} \eta_k$ should be replaced by $q \eta$ where $q^2 = \sum_{k=0}^{N-1} Q_{i,k}^2 = |Q|^2$. A formal justification comes from considering the first equation of the BBGKY hierarchy, which is an evolution equation for the TWD, $P(\xi, t)$, where the coefficient of $\partial_{\xi\xi} P(\xi, t)$ is $\frac{1}{2}|Q|^2$. Thus, (32) becomes

$$\dot{\xi}^{\text{mf}} = G(f(\xi^{\text{mf}}, t), f(\xi^{\text{mf}}, t), \xi^{\text{mf}}, f(\xi^{\text{mf}}, t), f(\xi^{\text{mf}}, t)) + q \eta. \quad (33)$$

By comparison of the Fokker–Planck equation for (33) with the first equation of the BBGKY hierarchy for (32), one obtains a (self-consistent) formula for f [35]. For a *linear*

¹³ The reader should not infer that the fixed system size constraint necessarily implies a bounded TWD variance; see [35] for an (unphysical) counter-example of a conservative noise yielding a TWD variance that is unbounded for long times. For finite (but large) N , this behavior [35] means that the TWD variance tends to become of the order of the system size at long times.

G , such a self-consistent f requires knowing the pair correlation. Specifically, one needs to know the conditional expectation for a terrace width, i.e. the average width of a terrace in a pair of terraces *given* the value of the width of the other terrace [34].

To circumvent the hurdle of solving the BBGKY hierarchy, it is tempting to apply the decorrelation ansatz $p_n(\vec{w}_n, t) = \prod_{j=1}^n P(w_j, t)$, where $\vec{w}_n = (w_0, \dots, w_n)$ [34, 35]. For a linear SDE system, i.e. when G is linear, the mean field is given by $f = \mathbb{E}\xi_i = 0$ [34, 35]. In [35], where the first-row entries for Q are set to $[2, -1, 0, \dots, 0, -1]$ (yielding a second-order conservative noise scheme similar to (18)), it was verified for the linearized SDEs that the long-time limit of the mean field variance *coincides* with that of the exact solution.

Motivated by these previous studies, we compute the mean field variance for SDEs (4) under the F -dependent noise (27). By taking $f = \mathbb{E}\xi_i = 0$, these equations are reduced to the *effective* Langevin equation

$$\frac{d\xi^{\text{dc}}}{d\tau} = -[1 - 2p + 3g(1 + 2\beta)]\xi^{\text{dc}} + q\tilde{\eta}(\tau), \quad q^2 = 2\tilde{m}_0[1 - \sqrt{p(1-p)}], \quad (34)$$

where ξ^{dc} approximates (in some sense) the mean field stochastic process ξ^{mf} under the decorrelation ansatz. Thus, ξ^{dc} is a Gaussian random variable with zero mean and variance:

$$\sigma^{\text{dc}}(t)^2 = \tilde{m}_0 \frac{1 - \sqrt{p(1-p)}}{1 - 2p + 3g(1 + 2\beta)} \{1 - e^{-2Fa[1 - 2p + 3g(1 + 2\beta)]t}\}, \quad (35)$$

which approaches $\sigma^{\text{dc}}(\infty)^2 = \tilde{m}_0[1 - \sqrt{p(1-p)}][1 - 2p + 3g(1 + 2\beta)]^{-1}$ as $t \rightarrow \infty$.

The long-time limit (35) differs from the variance calculated by (29). We attribute this discrepancy to the fact that terrace correlations persist at long time (cf (30)), in contradistinction to the decorrelation ansatz. For strong enough step interactions, i.e. $g(1 + 2\beta) \gg 1 - 2p$, we have $\sigma^{\text{dc}}(\infty)^2 = \mathcal{O}(g^{-1})$, while by (30) we have $\mathfrak{C}_{0,\text{st}} = \mathcal{O}(g^{-1/2})$. Thus, the decorrelation hypothesis exaggerates the narrowing of the TWD; the (positive) terrace width pair correlations favor broadening of the TWD.

4.3. Related past works and plausible connection to experiments

Next, we point out features of our predictions for the variances $\mathfrak{C}_{0,\text{st}}$ of (23) and (29) that may be experimentally testable, and discuss comparisons of these to results of [10, 16].

- (i) *Narrowing of TWD* [16]. In the case with a flux (F)-dependent noise and large $\nu = aF\varpi^2/(2D)$, by (29) the variance $\mathfrak{C}_{0,\text{st}}$ approaches the limit

$$\mathfrak{C}_{0,\text{st}} \rightarrow \tilde{m}_0(1 + 4g)^{-1/2} \quad \text{as } \nu \rightarrow \infty, \quad (36)$$

under the assumption that the quasi-steady approximation is meaningful. In the near-equilibrium case with large ν , the variance approaches a constant with complicated g -dependence; cf (25). These predictions should be contrasted with the corresponding variance in [16], where $\sigma^2 = \mathfrak{C}_{0,\text{st}} = \mathcal{O}(F^{-1})$ for large F .

For small deposition flux F ($p \uparrow 1/2$), (29) yields $\mathfrak{C}_{0,\text{st}} \rightarrow \mathcal{O}(g^{-1/2})$ as $\nu \downarrow 0$. In the near-equilibrium case, the variance (23) is $\mathcal{O}(g^{-1})$ as $F \downarrow 0$. These predictions stand in contrast to the behavior $\mathfrak{C}_{0,\text{st}} = \mathcal{O}(F^{-2})$ given in [16]. Hence, our noise models significantly tone down the narrowing of the TWD reported in [16], where deposition is the only source of TWD narrowing and the noise is F -independent. More precisely, the discrepancy of our result for $\mathfrak{C}_{0,\text{st}}$ with the variance in [16] is due to (i) the different scaling of the noise term with F and (ii) the application of the mean field approximation with a non-conservative white noise in [16]. Other models of noise for terraces can plausibly be constructed by mixing elements of (18) and (27).

- (ii) *Comparison to [10]*. In the model of [10], the deterministic equations account for the same kinetic (flux-induced) asymmetry in step motion but steps are *non-interacting*. The noise used in [10] for every terrace, w_i , appears to have the form $\eta_{i,1}\sqrt{Fpw_{i-1}} + \eta_{i,2}\sqrt{Fpw_i} + \eta_{i,3}\sqrt{F(1-p)w_i} + \eta_{i,4}\sqrt{F(1-p)w_{i+1}}$, where $\eta_{i,k}$ ($k = 1, \dots, 4$) are independent white noises and units with $a = 1$ are apparently used. In the mean field approximation (under the decorrelation ansatz for terrace widths), this model yields the (scaled by ϖ^2) steady-state variance $\hat{\sigma}(t \rightarrow \infty)^2 = (1 - 2p)^{-1}$ [10]. In [10] this prediction is found to appreciably overestimate the variance produced by kinetic Monte Carlo simulations for small rate F (see figure 4 in [10]). Since the simulations do *not* allow for step crossing, it is expected that these simulations *do* include *entropic repulsions* between steps. On the other hand, our analytical model does contain the effect of step repulsion explicitly (via \check{g}) and predicts a lower value of the variance for small F .
- (iii) *Experiment on Si(1 1 1)* [14, 43]. Scanning tunneling microscopy measurements on Si(1 1 1) can yield images of quasi-1D step trains [43]. These systems have been modeled previously by *deterministic* 1D evolution equations resembling (1) [8]; our stochastic model (4) accounts for the additional effects of noise. By measuring terrace width correlations under equilibrium and non-equilibrium conditions, it should be possible to test the validity of the noise models (18) and (27), respectively.

As mentioned in [16], experimental techniques that enable observation of equilibrium TWD can in principle also probe narrowing due to the combined influence of growth *and* step interactions. An example is the reflection electron microscopy applied in [14]. In this experiment [14], TWD narrowing is observed on vicinal Si(1 1 1) at 1100 °C and attributed solely to electromigration (which also causes a drift in the adatom flux) although a deposition flux from above and step interactions are present.

4.4. On the validity of the model

We repeat that a limitation of our model is due to its 1D character. Despite this feature, our analysis may be useful in describing quantitative aspects of quasi-1D step systems, e.g. those in [8, 43], as also noted in [35].

Another limitation is related to the perturbation expansion for ‘small’ noise (small terrace width fluctuations), which underlies our linearization. Within this approximation, an indication that the linearization may not be valid arises if the TWD variance satisfies $\mathfrak{C}_0(t) > 1$: by (10), the negative tail of the (approximate) TWD, $P(\xi, t; N)$, may have an appreciable effect on moments. To provide a condition necessary for the validity of our model, we require that $\mathfrak{C}_0(t)$ is sufficiently small for all t .

We also comment on the magnitude of the ratio $\nu = Fa\varpi^2/(2D)$ which first appears in (1). This constant is considered to be $\mathcal{O}(1)$, so that the convection (resulting from our transformation to the step comoving frame) is *not* necessarily negligible in our analysis. For many physical systems, ν can be estimated by [8]

$$\nu = \frac{Fa\tau_0}{e^{-E_D/T}} \frac{1}{2\tilde{m}_0^2}, \quad (37)$$

where E_D is the energy barrier (i.e. a diffusion barrier) that an adatom must overcome to move to an adjacent lattice site, and τ_0^{-1} is an attempt frequency; we use the relation $D = a^2\tau_0^{-1}\exp[-E_T/T]$ [8]. Typical values for E_T range from 0.04 eV for Al(1 1 1) to 0.97 eV \pm 0.07 eV for Si(1 1 1) [8]. The attempt frequency, τ_0^{-1} , is usually taken to be 10^{13} s^{-1} . For a deposition rate of 1 monolayer s^{-1} ($Fa = 1 \text{ s}^{-1}$) at room temperature, values of ν are found to range approximately from 10^{-11} to 10^8 . Although we find the upper bound

of these values to be exaggerated, we are in principle motivated to account for all $\nu \geq 0$ in our model. For many experimental systems of current interest, however, our model is expected to be in the small-deposition-flux regime, with small ν and $p \approx 1/2$. Notably, an advantage of the model for arbitrary ν is that it is generic to a variety of asymmetric kinetic processes (as noted in section 1). We repeat that care must be exercised in applying the long-time limit of (28) (see remark 4).

5. Conclusion

In this paper, we studied small stochastic fluctuations of line defects on a crystal surface when material is deposited from above in 1+1 dimensions. This work has been inspired by [9, 10, 16] and aims to complement recent kinetic studies in the steady-state distribution of terrace widths on vicinal crystal surfaces [34–36]. We started with a reasonably general formulation of terrace width fluctuations in 1+1 dimensions, allowing for a noise that stems from the fluctuation-dissipation theorem for adatoms [9] and, alternatively, from a phenomenological description of a deposition-dependent effect at step edges [10]. The ensuing models include a conservative noise, deposition-flux-induced kinetic asymmetry in the noise coefficients and step repulsion.

Our perturbation analysis led to a Gaussian TWD and a simple closed form for the associated variance. Similarly, we obtained closed-form expressions for the terrace width covariance matrices. The TWD, which is symmetric about the expectation of the terrace width, is plausibly valid for values of terrace widths near the peak of the actual TWD. On the basis of these results, we inferred that growth combined with step interaction sustain a reduced narrowing of the TWD with the deposition rate, F , in juxtaposition to the corresponding (more exaggerated) F -dependence noted in [16]. Furthermore, we applied a previous mean field approach [16, 34], and thereby indicated and discussed the role of terrace width pair correlations at long times. We found that these correlations decay exponentially in the height difference between two terraces and practically vanish in the macroscopic limit.

Our analysis points to several open questions. For example, the nonlinearities left out from our stochastic scheme should cause the TWD to be non-symmetric about the mean [35]. The derivation of such a modified TWD remains unresolved. Any effect of multiplicative noise (where the noise coefficients depend on terrace widths) is left for future work. Another open question concerns 2D geometries, which were not considered here. In real systems, edge atoms diffuse along steps (besides diffusing on terraces and attaching/detaching at step edges). In addition, kinks on steps influence the form of noise. Many-step interacting systems in 2D are the subject of work in progress.

Acknowledgments

The first author (PNP) acknowledges support under the National Institute of Standards and Technology American Recovery and Reinvestment Act Measurement Science and Engineering Fellowship Program Award 70NANB10H026 through the University of Maryland; and by the NSF under MRSEC grant DMR 0520471 at the University of Maryland. The third author's (DM) research was supported by NSF under grant DMS-0847587.

Appendix A. Derivations for covariant matrix

In this appendix, we derive (8) and use this result to show (11). In the following, we abandon the summation convention for repeated indices.

Lemma A.1. For arbitrary $N \times N$ circulant matrices \mathbf{A} and \mathbf{Q} , and a stochastic process $\boldsymbol{\xi}(t)$ given by (7) with $\boldsymbol{\xi}(0) = 0$, the covariance matrix $\mathfrak{C}_m(t)$ is given by (8).

Proof. Consider a given lattice site with index j . By the invariance of (4) under translations of j , the covariance matrix is

$$\begin{aligned} \mathfrak{C}_m(t) &= N^{-1} \sum_{j=0}^{N-1} \mathbb{E}[\xi_j(t) \xi_{j+m}(t)] = N^{-1} \mathbb{E}[\boldsymbol{\xi}(t)^T (\boldsymbol{\xi}(t))_m^c] \\ &= \frac{1}{N} \int_0^\tau ds \int_0^\tau ds' \mathbb{E}[(\mathbf{Q} e^{-\mathbf{A}(\tau-s)} \boldsymbol{\eta}(s))^T (\mathbf{Q} e^{-\mathbf{A}(\tau-s')} \boldsymbol{\eta}(s'))_m^c], \end{aligned} \quad (\text{A.1})$$

where the symbol $(\cdot)_m^c$ denotes the m th cyclic permutation of the vector inside the parenthesis. For any circulant matrix \mathbf{X} and vector \mathbf{y} , the i th component of $(\mathbf{X}\mathbf{y})_m^c$ is

$$\begin{aligned} [(\mathbf{X}\mathbf{y})_m^c]_i &= \sum_{j=0}^{N-1} X_{i+m,j} y_j = \sum_{j=0}^{N-1} X_{i+m,j+m} y_{j+m} \\ &= \sum_{j=0}^{N-1} X_{i,j} y_{j+m} = [\mathbf{X}(\mathbf{y})_m^c]_i, \end{aligned} \quad (\text{A.2})$$

where all indices are interpreted as modulo N , and the last line follows from the definition of a circulant matrix. Moreover, the product of any two circulant matrices is circulant. Hence, the term $\mathbf{Q} \exp[-\mathbf{A}(\tau - s')]$ appearing in (A.1) is circulant. Equation (8) is deduced from (A.2). \square

Next, we derive (11) on the basis of (8). This amounts to proving the following.

Proposition A.1. For arbitrary $N \times N$, constant circulant matrices \mathbf{A} and \mathbf{Q} , and delta-correlated noise $\eta(s)$, where $\mathbb{E}[\eta(s)\eta(s')] = \delta(s - s')$, the following relation holds:

$$\mathbb{E}[\boldsymbol{\eta}(s')^T e^{-\mathbf{A}^T(\tau-s')} \mathbf{Q}^T \mathbf{Q} e^{-\mathbf{A}(\tau-s)} (\boldsymbol{\eta}(s))_m^c] = \delta(s - s') \sum_{k=0}^{N-1} \vartheta_k e^{-\lambda_k(\tau-s)} \cos\left[\frac{2\pi km}{N}\right], \quad (\text{A.3})$$

where ϑ_k are the eigenvalues of $\mathbf{Q}^T \mathbf{Q}$ and λ_k are the eigenvalues of $\mathbf{A} + \mathbf{A}^T$.

Proof. Since \mathbf{A} and \mathbf{Q} are circulant, they are diagonalized by the discrete Fourier transform matrix \mathbf{F} , where $F_{k,l} = e^{-2\pi i(kl/N)}$ ($i^2 = -1$). Specifically,

$$e^{-\mathbf{A}^T(\tau-s')} \mathbf{Q}^T \mathbf{Q} e^{-\mathbf{A}(\tau-s)} = \mathbf{F}^{-1} \mathcal{D} \mathbf{F}, \quad \mathcal{D}_{j,k} = \delta_{j,k} \vartheta_k e^{-\lambda_k(\tau-s)}, \quad (\text{A.4})$$

which follows from the relation $\mathbf{F}^{-1} \exp(\mathbf{C}) \mathbf{F} = \exp(\mathbf{F}^{-1} \mathbf{C} \mathbf{F})$ for any matrix \mathbf{C} . Hence, the expectation of the left-hand side of (A.3) simplifies to

$$\begin{aligned} \mathbb{E}[\boldsymbol{\eta}(s')^T \mathbf{F}^{-1} \mathcal{D} \mathbf{F} (\boldsymbol{\eta}(s))_m^c] &= \frac{1}{N} \sum_{j,k,n=0}^{N-1} \mathbb{E}[\eta_j(s') e^{2\pi ijk/N} \mathcal{D}_{k,k} e^{-2\pi ikn/N} [(\boldsymbol{\eta}(s))_m^c]_n] \\ &= \frac{\delta(s - s')}{N} \sum_{j,k,n=0}^{N-1} \mathcal{D}_{k,k} e^{2\pi ik(j-n)/N} \delta_{j+m,n} \\ &= \delta(s - s') \sum_{k=0}^{N-1} \mathcal{D}_{k,k} e^{-2\pi ikm/N}, \end{aligned} \quad (\text{A.5})$$

where we have written sums explicitly to avoid any ambiguity when summing over k .

Furthermore, note that the eigenvalues of a circulant matrix C are given by the product $F_{j,k} C_{k,0}$. Since $A+A^T$ and QQ^T are both symmetric and circulant, they satisfy $C_{k,0} = C_{N-k,0}$. This last property implies (A.3). \square

Proposition A.1 offers an extension of a result given in [35]. More precisely, in the case of the TWD variance, i.e. with $m = 0$, we recover formula (16) given in [35].

Appendix B. Useful relations

This appendix provides some relations that help simplify results of section 2.

B.1. Long-time behavior of integral (13)

We show that the exponential term in (13) vanishes when $\tau \rightarrow \infty$. This property holds for both forms of $\vartheta(x)$ given in section 3, and the λ_k given by (20) when $F \neq 0$. Specifically, the following inequality holds:

$$\begin{aligned} \left| \int_0^1 dy \vartheta(y) \lambda^{-1}(y) e^{-\tau \lambda(y)} \cos(2\pi m y) \right| &\leq c_1 \int_0^1 dy e^{-\tau c_2 (1 - \cos(2\pi y))} \\ &< c_1 \int_{-\infty}^{\infty} e^{-\tau c_2 y^2} dy \\ &= \mathcal{O}(\tau^{-1/2}) \quad \text{as } \tau \rightarrow \infty, \end{aligned} \tag{B.1}$$

where $c_1, c_2 > 0$ are constants independent of y .

B.2. Two formulas

In certain cases, the time dependence of the covariance matrix, e.g. in (31), can be resolved via the identity [42]

$$e^{z \cos(x)} = I_0(z) + 2 \sum_{n=1}^{\infty} I_n(z) \cos(nx), \tag{B.2}$$

where $I_n(z)$ is the modified Bessel function of order n .

The following integral, when combined with the appropriate trigonometric identities, is useful for calculating steady-state covariances by (8):

$$\int_0^\pi dx \frac{\cos(nx)}{1 - \alpha \cos(x)} = \frac{\pi}{\sqrt{1 - \alpha^2}} \left[\frac{1 - \sqrt{1 - \alpha^2}}{\alpha} \right]^{|n|}, \tag{B.3}$$

where $|n| = 0, 1, 2, \dots$ and $0 \leq \alpha < 1$. This formula can be shown by contour integration or, alternatively, it can be found in integral tables, e.g. [44].

Appendix C. Time-dependent covariance for $\nu \ll 1$

In this appendix, we asymptotically evaluate integral (28) for small $\nu = Fa\varpi^2/(2D)$, $g = \mathcal{O}(1)$ and $m = \mathcal{O}(1)$. For algebraic convenience, set $\tilde{m}_0 = 1$ (only in this appendix).

By the change of the integration variable to $z = \sin(\phi/2)$, the integral becomes

$$\mathfrak{C}_m(t) = \frac{1}{2\pi} \frac{1}{g(1+2\beta)} \int_0^1 \frac{dz T_m(1-2z^2)}{\sqrt{1-z^2}} [1 + 2\sqrt{p(1-p)}(1-2z^2)] \times \frac{1 - e^{-(\alpha_1^2 + z^2)\alpha_2^2 z^2}}{z^2 + \alpha_1^2}, \tag{C.1}$$

where $T_m(x)$ is the m th-order Chebyshev polynomial of the first kind, $\alpha_1^2 = (1 - 2p)[4g(1 + 2\beta)]^{-1}$ and $\alpha_2^2 = 16tgFa(1+2\beta)$. As $\nu \downarrow 0$ with fixed g and $tFa \geq \mathcal{O}(1)$, we have $\alpha_1^2 = \mathcal{O}(\nu^2)$ while $\alpha_2^2 \geq \mathcal{O}(\nu^{-1})$. Thus, the task is to evaluate $\mathfrak{C}_m(t)$ by (C.1) for *small* α_1 and *large* α_2 (where $\alpha_1, \alpha_2 > 0$).

By inspection of the integrand in (C.1), we distinguish the following cases.

(i) $\alpha_2^{-1} \gg \alpha_1^2$, i.e. $tFa \ll g(1 + 2\beta)(1 - 2p)^{-2}$. The major contribution to integration comes from a neighborhood of width $\mathcal{O}(\alpha_2^{-1/2})$ around $z = 0$, and α_1^2 is neglected compared to z^2 . Specifically, we have

$$\begin{aligned} \mathfrak{C}_m(t) &\approx \frac{1}{\pi} \frac{1}{g(1 + 2\beta)} \int_0^1 \sqrt{1 - z^2} \frac{1 - e^{-\alpha_2^2 z^4}}{z^2} dz \\ &= \frac{1}{\pi} \frac{1}{g(1 + 2\beta)} \left(\int_0^A + \int_A^{\alpha_2^2} \right) \int_0^1 \sqrt{1 - z^2} z^2 e^{-yz^4} dz dy, \end{aligned} \quad (C.2)$$

where A is any fixed yet large positive number and $T_m(1 - 2z^2) \approx 1$ to leading order. Thus, the y in $\int_A^{\alpha_2^2}$ is large and the respective integral in z is evaluated by expanding the integrand near $z = 0$. Finally, we obtain

$$\mathfrak{C}_m(t) \approx \frac{2}{\pi} \Gamma\left(\frac{3}{4}\right) [g(1 + 2\beta)]^{-3/4} (tFa)^{1/4}, \quad (C.3)$$

where $\Gamma(\zeta)$ is the Gamma function [45]. So, if tFa is fixed as $\nu \downarrow 0$, then $\mathfrak{C}_m(t)$ vanishes as $\mathcal{O}(\nu^{3/4})$. However, if instead $2Dt/\varpi^2$ is kept fixed as $\nu \downarrow 0$, then $\mathfrak{C}_m(t) = \mathcal{O}(\nu) = \mathcal{O}(F)$.

(ii) $\alpha_2^{-1} \ll \alpha_1^2$, i.e. $tFa \gg g(1 + 2\beta)(1 - 2p)^{-2}$. The major contribution to integration in the time-dependent term of (C.1) arises from a vicinity of width $\mathcal{O}((\alpha_1\alpha_2)^{-1}) = \mathcal{O}(\nu tFa)^{-1/2}$ around $z = 0$. Approximating $T_m(1 - 2z^2) \approx 1$, we obtain

$$\mathfrak{C}_m(t) \approx \frac{1}{2\pi} \frac{1}{g(1 + 2\beta)} \int_{-1}^1 \frac{\sqrt{1 - z^2}}{z^2 + \alpha_1^2} dz. \quad (C.4)$$

To evaluate this integral to leading order in ν , we apply analytic continuation of the integrand to complex z . So, deform the path of integration to the upper half of the complex z -plane so as to pick up the residue from the simple pole at $z = i\alpha_1$ ($i^2 = -1$). Thus, compute

$$\mathfrak{C}_m(t) = (2g/3)^{-1/2} + \mathcal{O}(\nu) = \mathfrak{C}_m(t \rightarrow \infty; p \uparrow 1/2, g), \quad (C.5)$$

which is the limit of steady-state formulas (29) and (30) for small $\nu = Fa\varpi^2/(2D)$.

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