Evolution of Terrace-width Distributions on Vicinal Surfaces: Fokker-Planck Derivation of the Generalized Wigner Surmise

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We use a Fokker-Planck equation to justify the generalization of the Wigner surmise for the energy-level spacing in quantum systems to the simple expression for the equilibrium terrace-width distribution of steps—with arbitrary-strength repulsions—on a vicinal surface, taking advantage of analogies to one-dimensional models of interacting, spinless fermions. This approach leads to an analytic description of the evolution toward equilibrium of steps from several experimentally relevant distributions: step bunches, perfect cleaved crystals, and pre-quench equilibrated distributions at different temperatures.

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Vicinal crystals [1], i.e. crystals misoriented from a high-symmetry plane, have great technological importance as growth templates with controlled step "defects" and as catalytic enhancers of some reactions due to the lower coordination of atoms on steps and of kinks thereon. Moreover, understanding the properties of steps provides a fruitful way to gain insight into nanostructures on surfaces. In particular, stepped surfaces differ considerably from perfect staircases, with thermal fluctuations playing a crucial role. The probability of finding neighboring steps at a specified separation, i.e. the terrace-width distribution (TWD), is denoted $P(s)$, where s is the step spacing w divided by its average value $\langle w\rangle$; $\langle w\rangle$ is the only characteristic length in the "downstairs" direction ($\hat{x}$). [Thus, $\langle s\rangle = 1$, and a perfect cleaved crystal has $P(s) = \delta(s-1)$.] Because steps extend unboundedly in the $\hat{y}$ direction (i.e., no islands on the terraces), they can be mapped onto the space-time trajectories of particles on a line. Because they cannot cross, these particles can be treated as spinless fermions with an entropic repulsion of the form $1/w^2$, where w is the terrace width. There generally also are elastic interactions with the same form, A/w^2, where in principle A should follow from the elastic constants of the crystal, but in practice it must be extracted from experimental data, especially the TWD. It is useful to deal with a dimensionless interaction strength, $\tilde{A} \equiv A\tilde{\beta}/(k_BT)^2$, where $\tilde{\beta}$ is the step stiffness.

Generalizing the so-called Wigner surmise [2], some of us have proposed that the TWDs have the simple form

$$P_\varrho(s) = a_\varrho s^\varrho \exp[-b_\varrho s^2], \qquad (1)$$

where the constants $b_\varrho = [\Gamma(\frac{\varrho+2}{2})/\Gamma(\frac{\varrho+1}{2})]^2$ and $a_\varrho = 2b_\varrho^{(\varrho+1)/2}/\Gamma(\frac{\varrho+1}{2})$ assure unit mean and normalization, respectively. The argument for Eq. (1) takes advantage of results for the Calogero-Sutherland (CS) Hamiltonian [3] describing fermions in one dimension (1D) with inverse-square repulsions; we thence find the identification

$$\varrho = 1 + \sqrt{1+4\tilde{A}} \qquad \Leftrightarrow \qquad \tilde{A} = \frac{\varrho}{2}\left(\frac{\varrho}{2} - 1\right) \qquad (2)$$

Historically, Eq. (1) came from efforts to describe the distribution of the energy separations of nuclear levels, obtained from the eigenvalues of Hamiltonians represented by random matrices [4] with orthogonal, unitary, or symplectic symmetry. Wigner's arguments based on these three symmetries lead to Eq. (1) for $\varrho = 1$, 2, or 4, respectively. Although not exact (as originally surmised), Eq. (1) provides a superb approximation [5] for these cases. There is a rather miraculous and fundamentally mysterious connection between the above distribution of energy spacings and the distribution of spatial separations of spinless fermions in 1D with mutual repulsions $\propto w^{-2}$ [3]. While $\tilde{A}$=0 (ϱ=2) notably corresponds to "free fermion," there is nothing intrinsically exceptional about $\tilde{A}$=2 (ϱ=4); furthermore, negative values of $\tilde{A}$ (esp. $-\frac{1}{4} \Leftrightarrow \varrho$=1) are unphysical.

Conventional TWD analyses [1] have been based on the "mean-field" Gruber-Mullins approximation [6]: a single step meanders between two straight neighboring steps fixed at the average step separation. In fermion language, the step is the 1D trajectory of a quantum particle confined to the segment $(0, 2\langle w\rangle)$ by an infinite potential. For a repulsion strong enough that the potential of the active step can be approximated by a parabolic well, the TWD is a Gaussian with a width $\propto \tilde{A}^{-1/4}$. More refined analyses [7–9] also yield Gaussian TWDs but with different proportionality constants between width and $\tilde{A}^{-1/4}$.

With ϱ as the adjustable parameter, Eq. (1) provides a better accounting of the TWD, both in numerical simulations and in actual experiments, than any of the preexisting Gaussian models [10]. However, it has not appeared in the literature on random matrix theory (RMT) [4] for general ϱ, and the only formal justification is based on a model single-particle Hamiltonian [11]. In this Letter, we show that Eq. (1) arises as the stationary solution of a Fokker-Planck equation (FPE) derived from a classic model of interacting particles in 1D. Furthermore and arguably more importantly, we obtain thereby a description of how experimentally relevant *non-equilibrium*

TWDs relax toward $P_\varrho(s)$.

Our work contrasts with efforts over decades to find distributions that describe how equilibrium fluctuations (in the spacings of eigenenergies) change as symmetry is gradually broken or as two subspaces of different symmetry are mixed [12, 13]. However, our time variable does chronicle evolution through an abstract parameter space. Nonetheless, we shall see that comparisons are possible with the physical approach to equilibrium. We discuss ways to connect our FP time with physical time.

We begin with the analogy drawn by Dyson between RMT and a Coulomb gas model [14]: N classical particles on a line, interacting with a logarithmic potential, and confined by an overall harmonic potential. The Coulomb gas model helps understanding the fluctuation properties of the spectrum of complex conserved systems. This model can be generalized to the *dynamic* Brownian motion model, in which the N particles are subject, besides the mutual repulsions, to dissipative forces [12]. The particle positions x_i then obey Langevin equations,

$$\dot{x}_i = -\gamma x_i + \sum_{i\neq j} \frac{\hat{\varrho}}{x_i - x_j} + \sqrt{\Gamma}\eta, \tag{3}$$

where η is a delta-correlated white noise and $\hat{\varrho}$ ($\propto \varrho$) is the "charge" of each particle. The probability of finding the particles at the positions $\{x_n\}$ at time t is the solution of the multi-dimensional FPE

$$\begin{aligned}\frac{\partial P(\{x_n\},t)}{\partial t} &= \sum_i \frac{\partial}{\partial x_i}\left[\frac{\partial}{\partial x_i}P(\{x_n\},t) + \gamma x_i P(\{x_n\},t)\right] \\ &- \sum_{i\neq j}\frac{\partial}{\partial x_i}\left[\frac{\hat{\varrho}}{x_i - x_j}P(\{x_n\},t)\right].\end{aligned} \tag{4}$$

In the 1D case, γ^{-1} would essentially be the variance of the stationary distribution. Narayan and Shastry [15] showed that the CS model is equivalent to Dyson's Brownian motion model, in the sense that the solution of the FPE (4) may be written as $P(\{x_n\},t) = \psi(\{x_n\},t)\psi_0(\{x_n\},t)$, where $\psi(\{x_n\},t)$ is the solution of a Schrödinger equation with imaginary time, derived from the CS Hamiltonian. The Langevin force of Eq. (3)

$$F(x_m) = -\gamma x_m - \sum_{k>m}\frac{\hat{\varrho}}{x_k - x_m} + \sum_{m>q}\frac{\hat{\varrho}}{x_m - x_q}, \tag{5}$$

so that

$$F(x_{m+1}) - F(x_m) = -\gamma(x_{m+1} - x_m) - \hat{\varrho}\left[\frac{-2}{x_{m+1}-x_m} + \right. \tag{6}$$

$$\left. \sum_{k>m+1}\frac{x_{m+1}-x_m}{(x_k - x_{m+1})(x_k - x_m)} + \sum_{m>q}\frac{x_{m+1}-x_m}{(x_{m+1}-x_q)(x_m - x_q)}\right].$$

Since we seek the distribution of widths w, we construct a one-"particle" theory [6] where the dynamical variable is the nearest-neighbor distance $w_m \equiv x_{m+1} - x_m$. To decouple the force on w_m from the other particles, we assume—in a Gruber-Mullins spirit—that the denominators $(x_k - x_{m+1})(x_k - x_m)$ in Eq. (6) are replaced by their mean values, the average being taken in the stationary state:

$$\langle (x_k - x_{m+1})(x_k - x_m)\rangle_{st} = \langle w^2\rangle_{st}(k-m-1)(k-m), \tag{7}$$

Each of the two sums in Eq. (6) then simplifies greatly, taking the form $[(x_{m+1} - x_m)/\langle w^2\rangle_{st}]\sum_{p>0}[(p+1)p]^{-1}$, where the sum is unity for $N \to \infty$ In summary, the interaction of a particle pair with all other particles acts on average as a harmonic potential, increasing the "spring constant" of the external one. We arrive at a one-particle Langevin equation for the terrace width w:

$$\dot{w} = -2\left[\left(\frac{\gamma}{2} + \frac{\hat{\varrho}}{\langle w^2\rangle_{st}}\right)w - \frac{\hat{\varrho}}{w}\right] + \sqrt{2\Gamma}\eta. \tag{8}$$

Eq. (8) is called a Rayleigh process [16–19] if $\sqrt{2\Gamma} = \hat{\varrho}$.

Our goal is to convert Eq. (8) into a Fokker-Planck equation for which Eq. (1) is a steady-state solution. We change to dimensionless variables $s \equiv w/\langle w\rangle_{st}$ and $\tilde{t} \equiv \Gamma t/\langle w\rangle_{st}^2$. Treating γ as a self-consistency parameter and recognizing $\hat{\varrho} = \varrho/\Gamma$, we set $\gamma = 2/\Gamma\langle w^2\rangle_{st}$. Then the coefficient in parentheses in Eq. (8) becomes $(1+\varrho)/\Gamma\langle w^2\rangle_{st}$; since $\langle s^2\rangle$ of Eq. (1) is $(\varrho+1)/2b_\varrho$ [9, 12], the coefficient can be rewritten as $2b_\varrho/\Gamma\langle w\rangle_{st}^2$. Thus, we have the sought-after FPE:

$$\frac{\partial P(s,\tilde{t})}{\partial \tilde{t}} = \frac{\partial}{\partial s}\left[\left(2b_\varrho s - \frac{\varrho}{s}\right)P(s,\tilde{t})\right] + \frac{\partial^2}{\partial s^2}[P(s,\tilde{t})]. \tag{9}$$

Before solving Eq. (9) we make a few remarks. First, Eq. (9) may be interpreted as a mean-field version of Eq. (4). Also, defining $U(s) = b_\varrho s^2 - \varrho\ln s$ and using the transformation $\Psi(s,\tilde{t}) = P(s,\tilde{t})\exp[U(s)/2]$, we can transform the Fokker-Planck equation into a self-adjoint equation (or Schrödinger equation with imaginary time)

$$\frac{\partial\Psi(s,\tilde{t})}{\partial\tilde{t}} = \left[b_\varrho^2(s^2-1) + \frac{\frac{\varrho}{2}\left(\frac{\varrho}{2}-1\right)}{s^2}\right]\Psi + \frac{\partial^2\Psi}{\partial s^2}, \tag{10}$$

that reproduces the effective one-body approximation [11] of the CS model. Second, if we multiply Eq. (8) by $2s$, and let $v = s^2$, we obtain a stochastic differential equation for v in what in quantitative finance is called the Heston model [20]; the equivalent of Eq. (9) describes the evolution of the distribution function for the stochastic variance v in a second stochastic equation for stock returns. Third, there is no explicit rate parameter in Eq. (9); the scale of evolution is determined by the definition of $\tilde{t}$, so the characteristic time τ is $\langle w\rangle_{st}^2/\Gamma$. Thus, any dynamic information must enter through Γ, which we postulated in Eq. (3) and calibrated using standard fluctuation-dissipation arguments.

To solve Eq. (9) we must specify the initial distribution in s_0. Stratonovich [16] proceeds by separation of variables, finding the spatial eigenfunctions in terms of Laguerre polynomials. After some manipulation, we then find [21] the distribution in s at rescaled time $\tilde{t}$, given an initial sharp distribution $\delta(s-s_0)$, to be

$$P(s,\tilde{t}|s_0)=2\tilde{b}_\varrho\frac{s^{\alpha+1}}{\widetilde{s_0}^{\,\alpha}}I_\alpha\left(2\tilde{b}_\varrho s\widetilde{s_0}\right)\exp[-\tilde{b}_\varrho(s^2+\widetilde{s_0}^{\,2})] \quad (11)$$

where $\alpha=(\varrho-1)/2$, $\tilde{b}_\varrho\equiv b_\varrho/(1-\mathrm{e}^{-\tilde{t}})$, $\widetilde{s_0}\equiv s_0\exp(-\tilde{t}/2)$, and I_α is the modified Bessel function of the first kind.

Using $I_\alpha(x)\underset{x\to 0}{\sim}(x/2)^\alpha/\Gamma(\alpha+1)$, we find

$$P(s,\tilde{t})\sim\frac{a_\varrho s^\varrho}{(1-\mathrm{e}^{-\tilde{t}})^{(\varrho+1)/2}}\exp[-s^2b_\varrho/(1-\mathrm{e}^{-\tilde{t}})] \quad (12)$$

confirming that at long enough times $\tilde{t}$, any dependence on s_0 disappears and that Eq. (11) ultimately approaches Eq. (1). Three initial distributions are of special physical interest: "perfect" vicinals, step bunches, and equilibrium TWDs before thermal quenches or upquenches.

Perfectly cleaved crystals: If the initial distribution is a delta function at the mean spacing, i.e. $\delta(s_0-1)$, then the [normalized] solution Eq. (11) reduces to [22]

$$P(s,\tilde{t})=2\tilde{b}_\varrho s^{\alpha+1}\mathrm{e}^{\frac{\alpha\tilde{t}}{2}}I_\alpha\left(2\tilde{b}_\varrho s\mathrm{e}^{-\frac{\tilde{t}}{2}}\right)\exp[-\tilde{b}_\varrho(s^2+\mathrm{e}^{-\tilde{t}})]. \quad (13)$$

In experiments, $P(s)$ is generally characterized just by its variance $\sigma^2\equiv\mu_2-\mu_1^2$, which can be calculated from its first and second moments, μ_1 and μ_2, respectively:

$$\mu_1=\frac{\Gamma(\frac{\varrho+2}{2})_1F_1\left(\frac{\varrho+2}{2},\frac{\varrho+1}{2},\tilde{b}_\varrho\mathrm{e}^{-\tilde{t}}\right)}{\Gamma(\frac{\varrho+1}{2})\,\tilde{b}_\varrho^{1/2}\exp(\tilde{b}_\varrho\mathrm{e}^{-\tilde{t}})} \qquad \mu_2=\frac{\varrho+1}{2\tilde{b}_\varrho}+\mathrm{e}^{-\tilde{t}} \quad (14)$$

with ${}_1F_1$ (sometimes written M) the Kummer confluent hypergeometric function [23]. To the extent that $\mu_1=1$ [24] [and in any case for qualitative purposes],

$$\sigma^2(\tilde{t})=\sigma_W^2(1-\mathrm{e}^{-\tilde{t}}), \quad (15)$$

where [cf. Eq. (1)] the Wigner variance $\sigma_W^2=\langle s^2\rangle-1=[(\varrho+1)/2b_\varrho]-1$.

For numerical comparisons, we chose the simplest model, the terrace-step-kink model [10] with $A=0$ (just entropic interactions), setting k_BT to half the kink energy, with 4 steps, 200 spacings wide, and $\langle w\rangle=6$. In this model, the motion of individual atoms across a terrace is not tracked, so we use the Metropolis algorithm, with a single rate for moves. To compare simulations with the FPE prediction, we manually set the time scale of the simulation to "match" that of the dimensionless $\tilde{t}$: 10^3 MCS corresponds to 1.4 units of $\tilde{t}$. As shown in Fig. 1, the agreement is remarkably good.

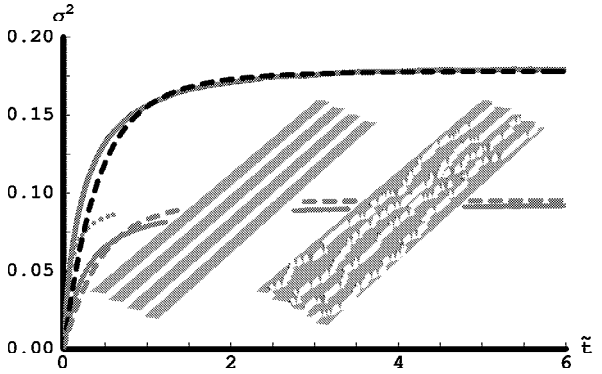

FIG. 1: [Color online] Comparison of the TWD variance predicted using Eq. (14) [blue, dashed] and the variance [red, solid] computed using conventional Metropolis Monte Carlo for a terrace-step-kink model for "free fermions" ($\tilde{A}=0$, $\varrho=2$). The left and right inset panels show the initial and a typical late-time configuration, respectively. Both curves approach equilibrium, $\sigma_W^2\approx 0.18$ [5, 9]. The lower pair of curves show the predicted [magenta, dashed] and computed [green, solid] variances for $\tilde{A}\approx 2.762$, $\varrho\approx 4.47$, with a different scale factor between $\tilde{t}$ and MC time, indicative of a smaller characteristic time. Both lower curves approach $\sigma_W^2\approx 0.095$. The dotted green curve shows the beginning of the green solid curve using the same scale factor as for $A=0$. See text for discussion.

Keeping everything else the same, we next took $A=0.5$, leading to $\tilde{A}\approx 2.762$. To now get good agreement between simulation and prediction, we find that 10^3 MCS corresponds to 4.5 units of $\tilde{t}$, indicative of a smaller characteristic time $\tau\propto\Gamma^{-1}$. There are indeed reasons to expect Γ to increase with ϱ [25].

While our analytic work assumes a continuum for w, at short times our model's discrete nature becomes important. Initially (i.e. at $\tilde{t}\ll 1$) step units are barely aware of the neighboring steps; the activation energy for the elementary excitation is the energy of a kink-antikink pair. For physical values of A and $\langle w\rangle\geq 4$, the contribution of the repulsion to this energy is negligible, so we expect (and find in MC) that $\sigma^2(\tilde{t}\ll 1)$ is independent of ϱ: the dotted line, replotting the beginning of the solid green curve with the same $\tilde{t}$ scaling as for $A=0$, initially coincides with the red line.

To improve our understanding of τ by investigating a system with better-defined dynamics, we have undertaken kinetic Monte Carlo simulations using an SOS model that incorporates adatom attachment/detachment events at the step along with terrace hops [26]. For attachment/detachment-limited kinetics, one expects that τ is set by the detachment rate of an atom from a kink site onto the terrace. In a simple bond-counting scheme as implemented in our simulations, such an event requires breaking 2 in-plane bonds (each of energy E_n), as well as hopping once on the surface (over diffusion barrier E_D). Our results (discussed in detail elsewhere [22]) show that the TWD variance behaves as a generaliza-

tion of Eq. (15), $\sigma^2(t)=\sigma_W^2 S(t/\tau)$, where τ is thermally activated, with the expected activation energy $2E_n+E_D$.

Step bunch: Eq. (12) is also obtained as the full time-dependent solution when the initial distribution is a delta function centered around a vanishing terrace size, $P(s_0,0) = \delta(s_0)$. Experimentally, this situation may be even easier to realize, for instance by inducing the formation of a bunch of steps (step bunching), and then watching the steps spread out to their equilibrium arrangement. In this case, the experimental TWD should be found to coincide at all times with Eq. (12); $\mu_1 = \sqrt{1-\mathrm{e}^{-\tilde{t}}}$ increases rapidly from its initial value to unity, and Eq. (15) holds.

Quenches: Of greatest interest physically is what happens during a quench (or upquench), in which a system which has attained equilibrium at the initial temperature is suddenly subjected to a different temperature. In this problem that translates to a sudden change from ϱ_0 to ϱ. Integrating Eq. (11) weighted by $P_{\varrho_0}(s_0)$ yields, after some manipulation [21]

$$P(s,\tilde{t}) = a_\varrho s^\varrho \mathrm{e}^{-\tilde{b}_\varrho s^2} \frac{(1-\mathrm{e}^{-\tilde{t}})^{\frac{\varrho_\circ-\varrho}{2}}}{(1-\mathrm{e}^{-\tilde{t}}(1-b_\varrho/b_{\varrho_\circ}))^{\frac{\varrho_\circ+1}{2}}} \times {}_1F_1\left(\frac{\varrho_0+1}{2}, \frac{\varrho+1}{2}, \frac{\tilde{b}_\varrho s^2}{1+(b_{\varrho_\circ}/b_\varrho)(\mathrm{e}^{\tilde{t}}-1)}\right). \quad (16)$$

Eq. (16) satisfies the following necessary conditions. For $\varrho_0 = \varrho$, it reduces to Eq. (1), since ${}_1F_1(a,a,z)=\exp(z)$. For arbitrary ϱ_0 and ϱ, it is initially $P_{\varrho_\circ}(s)$ and approaches $P_\varrho(s)$ at long times [23]. For asymptotically large ϱ_0, Eq. (16) reduces to Eq. (13). Analytic but more complicated expressions [21] generalize Eq. (14).

We have also formulated [21] a FPE description of the evolution of a train of steps with attractive effective interactions of kinetic origin, such as during step bunching. Two kinds of simple models of step evolution lead to step bunching: i) a model that produces an inverse Schwoebel effect; ii) a model including two diffusing and reacting species of particles [27].

Our formulation not only places the generalized Wigner surmise on firmer theoretical footing, it allows the exploration of the evolution of TWDs as a whole rather than just positions of individual steps. The results invite quantitative experimental studies to check the predictions and numerical simulations to clarify the dependencies of the characteristic time.

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[24] Note μ_1 starts at 1, dips quickly to about 0.975 before $\tilde{t}$=1, then gradually returns to 1. It has a non-negligible, though not qualitative, effect on $\sigma^2(\tilde{t})$. The deviation is presumably due to the approximations in using Eq. (7).

[25] For a Rayleigh process, $\Gamma \propto \varrho^{2/3}$; in this case, $\tau_{\tilde{A}=2.762} \approx 0.6\tau_{\tilde{A}=0}$. The decrease we observe in MC is nearly twice as great, but qualitatively similar.

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