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Implications of random-matrix theory for terrace-width distributions on vicinal surfaces: improved approximations and exact results

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Abstract

Quantitative measurement of the equilibrium terrace-width distribution of vicinal surfaces enables detailed investigation of step–step interactions. Using results from random-matrix theory, we point out simple analytical expressions that assist in this process, improving considerably over standard techniques and allowing assessment of weak repulsions, heretofore inaccessible except by indirect methods. This approach suggests new properties for experimentalists to measure and, by calibration with exact results, provides insights into controversies about assessing the interaction strength. © 1999 Elsevier Science B.V. All rights reserved.

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On vicinal (stepped) surfaces, terrace widths ℓ can now be measured quantitatively using several different surface-sensitive real-space imaging techniques [1,2]. The resulting equilibrium terrace-width distribution (TWD) $P(s)$ – where s is the (dimensionless) ratio of ℓ to its average value $\langle \ell \rangle$ – provides valuable information about the interaction between steps. These interactions, in addition to the step stiffness, are crucial to determining the morphology of these surfaces [1,2]. They are also vital to understanding phenomena involving kinetics and non-equilibrium responses of surfaces, such as step bunching [3,4]. Analyses typically rely on the mapping of the associated set of configurations in this two-dimensional problem onto the 'world lines' (plots of the evolution) of non-crossing particles, thus analogous to fermions in $1+1$ dimensions: the along-step $\hat{y}$ direction is taken to be time-like. (Alternatively, the problem in $1+1$D can be recast in terms of hard bosons rather than fermions [5].) A crucial, viable assumption in this mapping is that the interactions between the fermions are instantaneous, i.e. only for the same value of y on the interacting steps.

The non-crossing condition of the steps leads to a strongly temperature T-dependent entropic repulsion between them, since thermal fluctuations

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cause neighboring steps to approach each other. Since a fluctuation is more likely to lead to a crossing and so be forbidden when steps are closer, the interaction is repulsive, decaying like ℓ^{-2}, as is well known in many fields of physics, such as polymers [6] (or vortices) in two-dimensional systems or domain walls in magnetic (or ferroelectric) materials. In general, the step–step interaction energy per length – due to elastic or possibly dipolar coupling – has a similar decay, $U(\ell)=A/\ell^2$, but is relatively insensitive to T. (Note that A has dimensions of energy × length.) A key result of the mapping to fermions is that $P(s)$ depends only on the dimensionless ratio

$$\tilde{A} \equiv A\tilde{\beta}/(k_B T)^2, \tag{1}$$

where $\tilde{\beta}$ is the step stiffness (with units energy/length), and not on any of the three components separately.

Fits to data have relied on a simple 'single-particle' or mean-field-like analytic approximation [7] – which ipso facto forbids separations greater than $2\langle\ell\rangle$ – or, for more detail, trial-and-error Monte Carlo simulations. As recently as the last few months, two papers have lamented the lack of a simple analytic expression for $P(s)$, even in the simplest case of $A=0$ [8,9]. This Letter draws results from random-matrix theory (RMT) [10,11], which has been used with remarkable success to describe a broad variety of fluctuation phenomena. Although most of those cases involve the distribution of the spacings between adjacent energy levels, in several cases the fluctuations involved spatial variables, and here we consider the spacings between adjacent step positions. We find that a simple, single-parameter analytic expression accounts well for the shape of the TWD. Specifically, the so-called Wigner surmise provides a superb approximation to the TWD for the three values of $\tilde{A}$ for which the problem can be solved exactly. Furthermore, there is a natural, simple interpolation expression between these values that allows direct deduction of $\tilde{A}$ in the neighborhood of these values, i.e. for weak values of $\tilde{A}$ where neither single-particle approximation is viable. This generalized Wigner surmise (GWS) should provide an excellent ansatz for (approximate) analytic work. Although RMT has previously been used with remarkable success to unravel specific thorny questions [12], we show here the general applicability of the results of RMT. Other results from random-matrix theory also have implications for vicinal surfaces.

The simplest case of $A=0$ is the much-studied case of free fermions (FFs). In the venerable Gruber–Mullins (GM) approximation [7], one step is allowed to wander between two fixed straight steps at their mean positions. This situation is equivalent to the elementary quantum problem of a particle in a box of length $2\langle\ell\rangle$. In this approximation, the TWD is then given by the familiar ground-state density: $P(s)=\sin^2(\pi s/2)$. Sutherland [13–15] (see also Calogero [16–18]) showed that the full many-fermion problem can be solved exactly, and a sequence of analytic approximants – comprised of progressively lengthier combinations of elementary functions – were found [19] to provide an arbitrarily accurate explicit description. However, few experimentalists have made use of this result; most that did simply took numerical tabulations (or tracings) of $P(s)$ rather than regenerating it from the formal expressions.

Noting the initial quadratic rise and the eventual Gaussian decay of $P(s)$, Ibach [20,21] ingeniously devised an expression essentially equivalent to

$$P_2(s)=\frac{32}{\pi^2}\,s^2\exp\left(-\frac{4}{\pi}s^2\right). \tag{2}$$

(The numerical constants are readily determined from the two conditions on $P(s)$: that it is normalized and has unit mean.) As is evident from Fig. 1, this expression, in comparison with the GM expression, is a far superior approximation of the exact result, while still being a simple combination of elementary functions. The largest difference between it and the exact result is about 0.005 near the peak, and of order 1% over the region where it is significant. Eq. (2) is just the Wigner surmise for the distribution of levels of a Gaussian unitary ensemble (GUE) in random matrix theory. As listed in Table 1, its variance is barely 1% less than the exact value (versus over 27% for GM). It also peaks below the mean, close to the exact position,

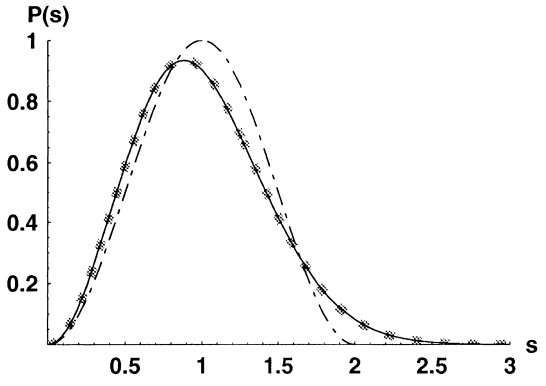

Fig. 1. $P(s)$ versus $s \equiv \ell/\langle \ell \rangle$ for the (sixth approximant [19] to the) exact result (solid curve), the Gruber–Mullins approximation (long–short dashed curve), and the Wigner surmise result (broad gray dashed curve; if plotted at normal thickness, it would barely be distinguishable from the exact result).

and has nearly the same skewness (which vanishes ipso facto for GM).

Sutherland [13–15] (see also Calogero [16–18]) also showed that the TWD could also be found exactly for two special cases of non-zero $\tilde{A}$, $\tilde{A}_4 = 2$ and $\tilde{A}_1 = -\frac{1}{4}$, corresponding to Gaussian symplectic and orthogonal ensembles respectively. Again one could generate a sequence of explicit analytic approximants, following Dyson's prescription [25]. However, for these cases there are also Wigner surmises:

$$P_4(s) = \left(\frac{64}{9\pi}\right)^3 s^4 \exp\left(-\frac{64}{9\pi} s^2\right);$$

$$P_1(s) = \frac{\pi}{2} s \exp\left(-\frac{\pi}{4} s^2\right). \tag{3}$$

Since steps on most vicinal surfaces have a repulsive interaction, the case $\tilde{A}_4 = 2$ is of much greater physical interest for vicinal surfaces (in contrast to most other applications of random matrix theory [11]). For strongly repulsive interactions, the GM approach leads to solving the problem of a particle in a 1D parabolic potential [26], with the outcome that

$$P(s) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{(s-1)^2}{2\sigma^2}\right]. \tag{4}$$

The potential must be strong enough, i.e. σ small enough, so that the tail of this Gaussian $P(s)$ outside $0 < s < 2$ is negligible. Moreover, $P(s)$ should be large only for $|s-1| \ll 1$, where the quadratic expansion of the potential of the neighboring steps is valid. If interactions with only adjacent (NN for nearest neighbor in Table 1) steps are considered, then $\sigma^{-2} = \sqrt{48\tilde{A}} \to 4\sqrt{6}$, where the specific value of σ is for $\tilde{A}_4 \equiv 2$. If one considers an infinite array of steps – all but the single active step straight and fixed at spacings of $\langle \ell \rangle$ – then the prefactor of 48 is replaced by $8\pi^4/15 \approx 52$; the corresponding row is labeled 'Gruber–Mullins (all)' in the Table 1. This latter approximation is more appropriate for comparison with the exact and the Wigner-surmise results, since those assume interactions with all fermions. Again, Table 1 shows that the Wigner surmise provides a better estimate of the variance (too small by 1/2%) than does GM (too small by about 7%) and of the kurtosis; it also includes skewness, which vanishes intrinsically without increasing algebraic complexity.

Several different expressions have been proposed to interpolate between the three cases of Eqs. (2) and (3) [11,27,28]. The simplest of these is [11]

$$P_\varrho(s) = a_\varrho s^\varrho \exp(-b_\varrho s^2), \tag{5}$$

where the constants b_ϱ and a_ϱ, determined by the unit-mean and normalization conditions [11] respectively, are given in terms of Γ functions:

$$b_\varrho = \left[\frac{\Gamma\left(\frac{\varrho+2}{2}\right)}{\Gamma\left(\frac{\varrho+1}{2}\right)}\right]^2; \quad a_\varrho = \frac{2b_\varrho^{(\varrho+1)/2}}{\Gamma\left(\frac{\varrho+1}{2}\right)}. \tag{6}$$

Thus, $P_\varrho(s)$ contains just the single adjustable parameter ϱ.[2] This variable can be simply related to the physical quantity of interest, the scaled

[2] We denote this parameter by ϱ rather than the conventional choice of β used in random-matrix theory to avoid possible confusion with the step energy per length and with $\tilde{\beta}$ the step stiffness. The analogy with inverse thermal energy [10,51,52], since a shift is involved, is not especially helpful in this problem.

Table 1
Tabulation of various measurable properties of terrace-width distributions $P(s)$ (where s is the terrace width normalized by its average value) based on exact results at the three soluble values of the dimensionless interaction strength $\tilde{A}$, the corresponding Wigner-surmise expression, and the 'single-fluctuating-step' (Gruber–Mullins) approximation[a]

				Range of elastic interactions Wigner-surmise interpolation range ⟶		
Property	Case	$\varrho=0$ 'Random'	$\varrho=1$ Attractive	$\varrho=2$ Non-interact	$\varrho=4$ Repulsive	$\varrho\to\infty$ Extreme rpl.
	$\tilde{A}=(\varrho-2)\varrho/4$	$\tilde{A}=0^-$	$\tilde{A}=-\frac{1}{4}$	$\tilde{A}=0$	$\tilde{A}=2$	$\tilde{A}=\varrho^2/4$
Symmetry assoc. with Sutherland $\mathscr{H}$		[Poisson]	orthogonal	unitary	symplectic	[SHO + phonons]
Special point character		NR free bosons	KT	free fermions	self-dual	
Variance $\sigma^2=\mu_2=\mu_2'-1$	Exact	1	0.286	0.180	0.105	0^+
	Wigner surmise	0.5708	0.2732	0.1781	0.1045	$0.500\varrho^{-1}$
	Gruber–Mullins (all)	–	–	0.1307	0.0981	$0.278\varrho^{-1}$
	Gruber–Mullins (NN)	–	–	0.1307	0.1021	$0.289\varrho^{-1}$
$\tilde{A}$ = Alternative estimate of σ^2	PIM (all) [8,19]	–	–	–	0.1747	$0.495\varrho^{-1}$
	PIM (NN) [8,19]	–	–	–	0.1838	$0.520\varrho^{-1}$
	Barbier et al.[23]	–	–	0.203	0.101	$0.405\varrho^{-1}$
Neighboring terraces	Exact cov[b] (s_1, s_2)	0	−0.27	−0.31	−0.34	
	Exact $\langle(s_1+s_2-2)^2\rangle$	2	0.416	0.248	0.138	0^+
Peak position $s_{\max}$	Exact	0	0.77	0.8840	0.94^-	1^-
	Wigner surmise	0	0.7979	0.8862	0.9400	$1-0.250\varrho^{-1}$
	Gruber–Mullins	–	–	1	1	1
Skewness $\mu_3/\sigma^3=(\mu_3'-1)/\sigma^3-3/\sigma$	Exact	2		0.4972		
	Wigner surmise	0.9953	0.6311	0.4857	0.3542	$0.707\varrho^{-1/2}$
	Gruber–Mullins (NN)	–	–	0	0	0
Kurtosis μ_4/σ^4	Approx. (GM, PIM)	9		3.1		
	Wigner surmise	3.8691	3.2451	3.1082	3.0370	$3+0.750\varrho^{-2}$
	Gruber–Mullins (NN)	–	–	2.4062	3	3

[a] As suggested by the arrow, the generalized Wigner $P_\varrho(s)$ should be viable somewhat – perhaps well beyond the interpolation range. For completeness, also included are: (1) the symmetries of the generic ensembles of random matrices, the Sutherland random matrices (and their underlying Hamiltonian matrices) [10,11], leading to the three non-trivial cases (interactions) for which the Sutherland Hamiltonian can consequently be solved exactly [13,14] – and for which the Wigner surmises are known to be excellent approximations; (2) the character of the solutions at the selected values of ϱ when viewed as special points along lines of Tomonaga–Luttinger-liquid critical points [12,24]. (Key to abbreviations: NR⇒non-relativistic; KT⇒Kosterlitz–Thouless; SHO⇒simple harmonic oscillator, i.e. uniformly spaced levels.) The extreme case $\varrho=0$, for which exact results are trivial, is included to dramatize trends in ϱ (although it lies on a different branch of solutions from the other tabulated values of ϱ [12]): as ϱ increases, the TWD becomes narrower, more symmetric, and more nearly Gaussian. Anticorrelations of neighboring terrace-width fluctuations increase. For the three exactly-solvable (non-trivial) cases, the Wigner surmise provides an excellent approximation, far better than any alternative.

[b] $$\mathrm{cov}(s_1,s_2)\equiv\frac{\langle(s_1-\langle s_1\rangle)(s_2-\langle s_2\rangle)\rangle}{[\langle(s_1-\langle s_1\rangle)^2\rangle\langle(s_2-\langle s_2\rangle)^2\rangle]^{1/2}}=\sigma^{-2}(\langle s_1s_2\rangle-1)=-1+\langle(s_1+s_2-2)^2\rangle/2\sigma^2.$$

For Gruber-Mullins the covariance of neighboring terrace-width fluctuations is ipso facto -1. With the formalism of Pierre-Louis and coworkers [8,22] we find $-\frac{1}{3}$ (NN) or $-0.36\ldots$ (all); with that of Barbier et al. [23], we find -0.33. In all these cases, the covariance is independent of ϱ.

strength of the step–step interaction

$$\varrho = 1 + \sqrt{1 + 4\tilde{A}} \ (\equiv 2\tilde{A}_{\mathrm{eff}}^{1/2}); \quad \tilde{A} = (\varrho - 2)\varrho/4 \qquad (7)$$

by inspection of the Sutherland Hamiltonian [13–18]. (The idea of an effective interaction strength $\tilde{A}_{\mathrm{eff}}$ – identical to λ in Sutherland's notation [12–15] – which takes into account the entropic repulsion, at least in an average way, will prove useful below.) To state the obvious, Eq. (5) provides a convenient, nearly universal form for the scaled TWD that should prove useful for analytical modeling.

Although perhaps it is simplest and best to obtain ϱ as the value optimizing the fit of TWD data by Eq. (6) [29], one can also focus on several readily accessible properties of $P_\varrho(s)$. The second moment of $P_\varrho(s)$ about the origin is

$$\mu_2' \equiv 1 + \sigma^2 = \frac{\varrho + 1}{2b_\varrho}. \qquad (8)$$

The variance σ^2, of course, corresponds to the squared (half) Gaussian width of the TWD, typically measured feature. (This width is typically measured with respect to the position $s_{\max}$ of the maximum of the TWD. For $P_\varrho(s)$, $s_{\max}$ is $(\varrho/2b_\varrho)^{1/2} < 1$. In contrast, σ^2 is the second moment about the mean, which is unity. When the GM Gaussian approximation is reasonable, this problem does not appear to be significant [29].) For unreconstructed Si(111) surfaces at high temperatures, σ^2 is 0.11 [30], corresponding to $\tilde{A} \approx \frac{7}{4} < 2$, so that the GWS should be a better approximation than the GM Gaussian. For reconstructed or adsorbed Si(111) surfaces at lower temperatures, the variance is about 0.07 [31], corresponding to $\tilde{A} \approx 4$. For a wide range of vicinal noble metals, especially Cu(100) and Cu(111), Giesen and coworkers [21,29] have found variances in the range 0.04–0.1. There are examples of materials with narrower (dimensionless) widths. There are no reported Gaussian fits, to our knowledge, with broader (relative) widths. A likely reason is that these are obviously skewed (see just below) and so described in terms of FFs, since a Gaussian fit looks inappropriate. As illustrated manifestly in Fig. 2, we now have a way to gauge $\tilde{A}$ even for small (but non-zero) values, for which the GM approximations are not helpful. Recalling the definition of $\tilde{A}$ in Eq. (1), we expect that $\tilde{A}$ should decrease strongly with T, since the energetic parameter is relatively independent of T and $\tilde{\beta}$ decreases weakly with T. Thus, with the increased availability of high-T STMs, more measurements with small $\tilde{A}$ are likely to appear.

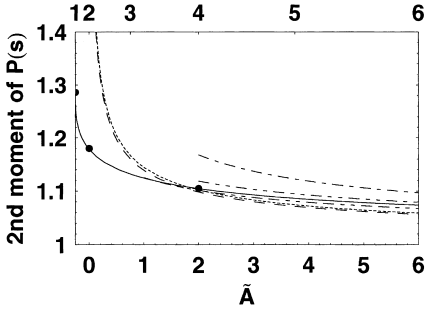

Fig. 2. Second moment (about the origin) μ_2' of $P(s)$ versus $\tilde{A}$ (solid curve), to illustrate how measurements of this moment or the variance $(\mu_2' - 1)$ of the TWD can be used to extract the strength of the step–step repulsion. In this and the subsequent figures, the tick marks across the top indicate the values of ϱ corresponding [see Eq. (7)] to the values of $\tilde{A}$ along the bottom. The three circles indicate the exact results at the three special values of $\tilde{A}$ for which they can be found. The dashed curves are the GM Gaussian approximations of Eq. (4), which clearly fail for small values of $\tilde{A}$. The short dashes give the 'NN' case in which interactions with nearest-neighbor steps are included; the long dashes show the 'all' case. The long-dash, short-dash curve gives the no-entropic-repulsion approximation of Ihle et al. [8] when (energetic) repulsions between all steps are included. The double long, double short curve shows the prediction of Barbier et al. [23] given in Eq. (9).

Since the third moment of $P_\varrho(s)$ about the origin $\mu_3' = (\varrho + 2)/2b_\varrho$, the skewness μ_3/σ^3 [viz. $(\mu_3' - 1)/\sigma^3 - 3/\sigma$, since μ_3 is the third moment about the mean of unity] of the GWS can likewise be written as a function of $\tilde{A}$. (See Fig. 3.) The comparable skewness of experimental data emerges only after moderate effort. A more direct way [32] to measure the asymmetry, with comparable magnitude, is the following: denote by $s_{\max}$ the value of s for which the TWD – or a smooth fit to it in an actual experiment – has its maximum; locate the two points $s_>$ and $s_<$ of the (smoothed) TWD, above and below $s_{\max}$ respectively, at half the height $P(s_{\max})$ of its maximum. Then it is a quick matter to find the ratio of the *difference* of the distances of these two points from $s_{\max}$ to

the *average* of these distances, i.e. $[(s_> - s_{max}) - (s_{max} - s_<)]/[(s_> - s_<)/2]$. For the GWS approximation $P_\varrho(s)$, this ratio must be determined numerically from Eq. (5). As illustrated in Fig. 3, this computed ratio – multiplied by $\frac{5}{2}$ – provides an excellent, i.e. to within about 1%, approximation of the skewness for $1 \le \tilde{A} \le 6$. For larger values of $\tilde{A}$, the skewness is small and provides little useful information. For $\tilde{A} \le 1$ the asymmetry ratio can be approximated to within 1% by empirically generated expressions,[3] also included (with the rescaling of $\frac{5}{2}$) in Fig. 3. Finally, there are other analytical expressions derivable from GWS that may serve as approximate consistency checks in fits of data.[4]

Higher moments can also be predicted analytically from the GWS, but are unlikely to be of much help in experimental analyses. It is noteworthy that the kurtosis of the GWS and of the exact solutions are slowly decreasing functions of ϱ, slightly above the value of 3 characteristic of Gaussian approximations (including the alternatives to Gruber–Mullins discussed below). For the 'FF' case, the kurtosis is decidedly smaller than 3.

There are some alternative approaches to extracting ϱ. Lässig [12] proposed that ϱ be obtained as the exponent in a fit of the initial (small s) rise of experimental data to a power-law form. Since $P(s)$ is small for small s, the relative uncertainty in the data in this regime is large; thus, this tactic is unlikely to produce satisfactory results. (Furthermore, the exact value of the prefactor differs somewhat from a_ϱ [19].) Instead one can fit the position s_{max} of the peak of (smoothed) data – or the point of inflection of integrated data. Then the result in the GWS result that $s_{max} = (\varrho/2b_\varrho)^{1/2}$ can be invoked. At first glance this approach seems to eliminate the need to find the normalization constant, but a_ϱ is at least implicitly needed to find $\langle \ell \rangle$ (to convert the independent variable from ℓ to s).

[3] Explicitly, $0.3 - 0.135(\tilde{A} + \frac{1}{4})^{1/4}$ for $-0.1 \le \tilde{A} \le 1$ and $0.302 - 0.154(\tilde{A} + \frac{1}{4})^{1/3}$ for $-0.24 \le \tilde{A} \le 0.1$.

[4] From inspection of GWS moments about the origin, we see $\mu_3' = 2\mu_2' - s_{max}^2$. From Eq. (5) we get the remarkable result $P_\varrho'''(s_{max}) = -P_\varrho''(s_{max})/s_{max}$; comparable experimental numbers can be obtained from a polynomial fit to data near the peak.

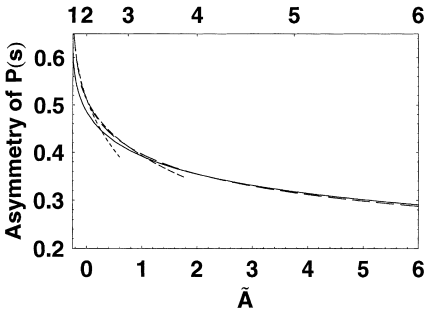

Fig. 3. Plots of the asymmetry of the TWD, estimated from $P_\varrho(s)$. Solid curve: the skewness μ_3/σ^3, computed from the analytical results for the third and second moments of $P_\varrho(s)$ about the origin, μ_3' and μ_2' respectively. Long-dashed curve: the normalized asymmetry; the difference of the 'half'-widths at half maximum divided by their average, as described in the text, multiplied by $\frac{5}{2}$ so as to be comparable with the skewness in size. For small values of $\tilde{A}$ the skewness does not provide an analytical way to describe this experimentally useful curve, so two empirically produced analytical expressions, described in the text, are included (moderately short-dashed curve applicable for $-0.1 \le \tilde{A} \le 1$ and shortest-dashed curve for $-0.24 \le \tilde{A} \le 0.1$), both of which are extended beyond their range of validity for improved visibility. Note well that both the Gruber–Mullins approximations and the newer alternative approximations [8,22,23,35,38] have zero asymmetry.

The key remaining question is how good an approximation the GWS is when it is used to extrapolate beyond the regime of weak $\tilde{A}$ (of either sign) to moderately repulsive $\tilde{A}$. From Eq. (7) we see that the exactly soluble case $\tilde{A} = -\frac{1}{4}$ is in fact the smallest value of $\tilde{A}$ for which a real solution for ϱ exists. As ϱ decreases below unity, the associated value of $\tilde{A}$ increases. The case $\varrho = 0$, which corresponds to straight steps placed at random so as to achieve the correct average misorientation, can be solved trivially and has the 'Poisson' distribution $P_{[\varrho=0]}(s) = \exp(-s)$. This case is included in Table 1 for completeness [33] and because of its simplicity rather than for any physical significance. It does highlight that, in this limit, the GWS is a poor approximation, even qualitatively.

What happens at $\varrho > 4$ is more controversial (and important). Recently Pierre-Louis and Misbah [22] and Ihle et al. [8] (hereafter referred to collectively as PIM) suggested that the GM Gaussian expression of Eq. (4) underestimates the variance by a factor $4\sqrt{2}/\pi \approx 1.801$ for the NN

case. (The proportionality constant is ~5% smaller for the 'all' case: $(4\sqrt{3}/\pi)[\gamma + \ln(2\pi) - \mathrm{ci}(2\pi)] \approx 1.711$.) The essence of their approach is the neglect of entropic repulsion: the steps are kept apart strictly by the energetic repulsion, and the entropic contribution to the free energy is due to the (Gaussian) wandering of the steps, viewed as isolated entities. Then the variance of the terrace width should be notably larger than that of one of its bounding steps (which is in essence how the variance is computed in the Gruber–Mullins picture). Since the fraction of the step repulsion that is entropic becomes vanishingly small with increasing $\tilde{A}$, this viewpoint might be expected to become appropriate as $\tilde{A} \to \infty$.[5] Similar objections to the GM Gaussian approach had been lodged earlier [23,35]; in particular, Barbier et al. [23] argued that

$$\langle (s_1 + s_2 + \ldots + s_m - m)^2 \rangle = \frac{2}{\pi^2 \varrho}[\ln(m) + 2] \quad \Rightarrow \quad \sigma^2 = \frac{4}{\pi^2 \varrho}, \tag{9}$$

where $s_1 + s_2 + \ldots + s_m$ denotes the total width of m adjacent terraces. This result follows from the general form they present for the correlation function of a rough phase, in the limit that step wandering is small compared with mean spacing, invoking a coarse-grained (along $\hat{x}$ as well as $\hat{y}$) free energy used fruitfully in earlier work on roughening [36].

The actual limit of $\varrho \to \infty$ has been well characterized: the TWD approaches the functional form $\delta(s-1)$ characteristic of a perfect staircase (with uniform spacings, analogous to the energy spacings of a harmonic oscillator [33]). However, we are not aware of any exact information on how this limit is approached. As documented in Table 1, the lowest-order limiting behavior (in ϱ^{-1}) of the variance of the PIM analysis is comparable with that of the GWS, both of which are larger than that of the GM Gaussian approximation by the predicted factor of about 1.8. However, there is no concrete evidence that the GWS is a good approximation in this limit.

Further evidence favoring the alternative pictures of PIM and of Barbier et al. in this limit is provided by the covariance of the steps the expectation of the product of their differences from their mean values (unity) normalized by their variance. As listed in Table 1, the covariance is negative: a fluctuation in the width of a terrace above $\langle \ell \rangle$ tends to be compensated by a decrease in the width of its neighbor. Such a phenomenon is tacitly assumed in the GM approximations, which ipso facto take the covariance to be -1, independent of the value of ϱ. Although perhaps the negative covariance accounts partially for the relative success of the GM viewpoint, the great overestimation of this effect sounds a note of caution in weighing its quantitative validity. At the other extreme is the case of independently fluctuating (straight) steps: the variance of the sum of the widths of neighboring terraces is twice that of a single step, making the covariance vanish. This does occur for the special 'Poisson' case $\varrho = 0$. The PIM model [8, 9] also leads to a ϱ-independent covariance, but with the more plausible value of $-0.36\ldots$ (or $-\frac{1}{3}$ when only NN steps are considered). Inspection of Table 1 shows that this value is close to the apparent limiting value of the covariances of the exactly soluble cases, extrapolated to infinite repulsion. On the other hand, these three covariances are smaller in magnitude than this limiting value, consistent with the poor estimates of the variance for those interactions. Consequent to Eq. (9), the model of Barbier et al. gives a covariance of $[(\ln 2)/4] - \frac{1}{2} \approx -0.327$, *also* ϱ-independent and in not quite so good agreement with the seeming asymptotic exact value as the PIM value.

Table 1 provides decisive evidence that PIM's objections are not globally significant. For moderate $\tilde{A}$, specifically at $\tilde{A} = 2$, the variance of the GM Gaussian approximation is much closer to the exact result than the alternative estimates. As $\varrho \to \infty$ the variances predicted by the alternative views are about 1.8 times those of the GM Gaussian approximations. They are also nearly identical to that of the GWS, although there is no

[5] In other words, since the strong repulsion keeps the steps apart mostly, the entropic interaction rarely comes into play. By the same token, the entropic repulsion plays a progressively larger role as the interaction grows weaker and eventually becomes negative, so that the effective interaction $\tilde{A}_{\mathrm{eff}}$ only vanishes in the unphysical, limiting Poisson case [34].

reason to believe that Eq. (5) does well in that extreme limit, especially since it does so poorly in the opposite limit ($\varrho=0$). However, it is noteworthy that for the four tabulated cases for which exact results exist, the Wigner surmise *underestimates* the variance. If it is still a lower bound as $\varrho\to\infty$, then the PIM estimate should be better than the GM Gaussian one. The ratio of the variance of the GWS to that of the GM Gaussian (all) value rises smoothly from $\sim$1.2 to about $\sim$1.4 as $\tilde{A}$ increases from 4 to 10, reaching the asymptotic value of $\sim$1.8 only very gradually.

If one accepts that the PIM picture is correct for ultrastrong repulsions, the crucial question becomes how good an approximation it is for finite but strong $\tilde{A}$. Another key issue regards the quality of the GWS as an *extrapolation* formula. Although, most likely, only numerical (Monte Carlo) studies can definitively answer these questions, we present in Fig. 4 some information which may be suggestive. This figure essentially replots the information of Fig. 2 on a logarithmic scale for the interaction strength. We see the PIM variance becomes a good approximation around $\tilde{A}\approx 30$ and an excellent one by $\tilde{A}\approx 100$. The variance of the GWS and that of the PIM 'all' case do not cross until $\sigma^2=3.44\times10^{-3}$ at $\varrho\approx144$ [$\tilde{A}\approx5100$], well above any reported value. (A 10% difference in the two variances occurs for $\tilde{A}\approx52$. For comparison, the variance of the GM Gaussian

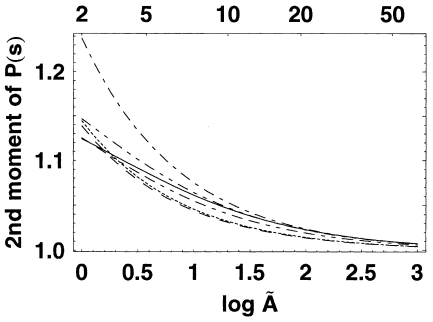

Fig. 4. Replot of Fig. 2 using a common (base-10) logarithmic scale for $\tilde{A}$. The various curves are as in Fig. 2, with the addition of the long-dash, double short-dash curve trace showing the approximation (for all-step repulsions) of Ihle et al. [8], modified to take entropic interactions partially into account by replacing with $\tilde{A}_{\rm eff}$ in the formula for the Gaussian width. See text for details.

approximation is within 10% of that of the GWS for $1.23\le\tilde{A}\le2.96$.)

Fig. 4 also includes a heuristic, phenomenological attempt to improve the estimate of the variance given by PIM's formalism. According to the expansion of the projected free energy of vicinal surfaces with slope (tangent of misorientation), the effective repulsion between steps, including both entropic and A/ℓ^2 energetic repulsions, replaces $\tilde{A}$ by $\tilde{A}_{\rm eff}$ defined in Eq. (7).[6] When $\tilde{A}$ is replaced by $\tilde{A}_{\rm eff}$ in evaluating the Gaussian width σ, so that the variance becomes $1.711/(51.95\tilde{A}_{\rm eff})^{1/2}$, we find that the GWS variance is well approximated down to $\tilde{A}\approx10$. This expression for the variance can be rewritten as $0.475/\varrho$; Eq. (9) shows that the corresponding prefactor in the analysis of Barbier and coworkers [23,35,38,39] is $4/\pi^2\approx0.405$, thus smaller in the asymptotic limit and below the variance of the GWS, but closer to it for moderate $\tilde{A}$. The physical meaning behind these relative behaviors is not yet clear. Calculations of leading-order behavior as $\varrho\to\infty$ could resolve many issues, but there has been little interest in this limit among random-matrix theorists; most of the systems concerning them occur in the range $0\le\varrho\le2$.

Obviously the problem of extracting $\tilde{A}$ from the TWD, especially from its width, is exacerbated by the extreme sensitivity of σ to $\tilde{A}$: fractional errors in the latter are magnified by a factor of four in the former. For many applications, the relative size of the step–step repulsions between different systems is more important than their absolute sizes; thus, it is crucial that the analysis of $\tilde{A}$ be done using the same approach. [Likewise, experimentalists should state clearly the raw (dimensionless) width, σ – or the value of ϱ in a fit to the GWS – before beginning any analysis!] Often the extracted value is scrutinized using (or misusing) the celebrated result of Marchenko and Parshin [40] relating the step repulsions to surface stress. This formula applies to the asymptotic regime of isotropic substrates; often one or both these conditions do not apply, and there is no well-prescribed procedure to make appropriate corrections.

[6] The extra factor of $\pi^2/6\equiv\zeta(2)$ in the free-energy expansion [1,2,37] comes from a sum over the interaction all, not just NN, steps.

Furthermore, the in-plane component of the stress dipole is not measurable and is often neglected. On another front, the assumption that the step interactions are in the $1+1$D perspective (i.e. occur only between points on steps at the same coordinate along the mean step direction $\hat{y}$) becomes particularly questionable when steps are close together and have large wandering fluctuations. In other words, efforts to make quantitative connections between measured TWDs and surface stresses are fraught with dangers more severe than the issues related to extracting $\tilde{A}$.

RMT provides other results of physical significance for vicinal surfaces [10]. From exact results and from scaling procedures, one can compute the width distributions of two, three, or more adjacent steps. These would allow greater information to be gleaned from real-space images. Exact results [10,11] are also available for the variance $\Sigma_\varrho^2 = 1,2,4\ (L_x)$ of the number of steps over an interval L_x (perpendicular to the steps, along x),[7] i.e. of the local slope, which exhibit a logarithmic divergence reminiscent of the roughness of vicinal surfaces.

In summary, we have illustrated how results from random matrix theory offer a simple analytical approximation, the generalized Wigner surmise, that accounts well for terrace width distributions and other properties of stepped surfaces for small to moderate dimensionless interactions $\tilde{A}$. This will be useful both in analyzing data and in pursuing model calculations. The regime of weak (dimensionless) step interactions was heretofore inaccessible by direct means; it will become increasingly important as more data is taken with high-temperature STMs. At the other end of the scale, there is great physical interest in large values of ϱ, in contrast to other applications of RMT, and significant open questions.

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Note added in proof

We have recently learned that some exact results have been determined for the Sutherland model for other values than those listed in Table 1. Most notably, Forrester [41] proved for even-integer values of ϱ that when s is (asymptotically) small

$$P(s) \approx \frac{[(\varrho/2)!]^3}{\varrho!(3\varrho/2)!}\,(\pi\varrho s)^\varrho . \tag{10}$$

Eq. (10) has the same leading behaviour as the Wigner surmise. The ratio of the prefactor of s^ϱ to α_ϱ is about one for small values of ϱ (viz. 1.015 and 0.995 for $\varrho = 2$ and 4, respectively) but decreases gradually (to 0.971, 0.939, 0.909 for $\varrho =$ 10, 20, 30, respectively). Thus, for large ϱ one might expect the Wigner surmise to be somewhat broader than the exact TWD and so to provide an *upper* bound of the true variance in this regime, and so an upper bound (overestimate) of the physical $\tilde{A}$.

Forrester [41–43] also presented analytic expressions for the 'pair correlation function', i.e. the probability of finding a step a distance ℓ from another, regardless of whether other steps lie between them. This two-body function, which naturally is much easier to calculate than the many-body $P(s)$, does not contain enough information (viz. many-step correlations) to enable calculation of $P(s)$ with the quantitative accuracy needed for reliable extraction of moments (although it does reduce to $P(s)$ for small separations) [19,41,44]. While experiments to date have reported just the TWD, there does not appear to be any intrinsic problem in also extracting the pair correlation function, at least for step separations up to a few times $\langle\ell\rangle$.

Based on Jack symmetric polynomials (as in Ref. [43]), there has been considerable activity

[7] For the cases of interest, to order $1/L_x$, $\Sigma_4^2(L_x) = (2\pi^2)^{-1}[\ln(4\pi L_x)+\gamma+1+\pi^2/8]$, $\Sigma_2^2(L_x) = \pi^{-2}[\ln(2\pi L_x)+\gamma+1]$, where $\gamma \approx 0.577$ is Euler's constant.

[45,46] devoted to deriving exact results for the Sutherland model for arbitrary rational (not just even-integer) ϱ, although most of this work has focused on the dynamical correlation functions [47] (requiring calculation of the excited states of the Sutherland Hamiltonian $\mathscr{H}$) rather than just the static correlations (associated with the ground state of this $\mathscr{H}$) that characterise the 'instantaneous' distribution of steps.

Some analytical results have also been derived for the strong-coupling (large-ϱ) limit of the Sutherland model, e.g. Refs. [47–50] (cf. comments shortly after Eq. (9)). In this limit, the leading correction to the (one-dimensional, 'Haldane–Shastry') lattice model (i.e. with uniform terrace widths) are acoustic phonons (including zero-point motion) with a 'sound velocity' proportional to ϱ [47–50].

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