Steps on Crystalline Surfaces: From Elementary Models to Universal Fluctuation Phenomena

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- Background, terminology, models
- Different roles of steps; applications in crystal growth, chemistry, nanowires, polymers
- Steps as Brownian strings; seeking signatures of mass transport modes
- Steps on vicinal surfaces as meandering fermions in (1+1)D...¿interactions?
- Terrace width distributions (TWDs), and what they reveal
- Simple models: mean field & 1D Schrödinger eqn...and their shortcomings
- Relevance of random matrix theory—universal features of fluctuations
- Generalizing the Wigner surmise from symmetry-based: meaning of \( \xi \)
- Fokker-Planck formulation: study of relaxation to equilibrium
- Growth: TWD narrowing; scaling of capture zones of islands

Prototype systems: Si (111), Cu (001) and (111)
SOS (solid-on-solid) model of vicinals

\[ H = \varepsilon \sum_{ij} |h_i - h_j| \quad \text{integer } h_i \]

- terrace adatom
- vacancy island
- adatom island
- vacancy
- kink
- step adatom
- step

\( \phi \) is the misorientation angle, fixed
Mean step spacing \( \langle \ell \rangle \propto 1/\tan(\phi) \)
Slope \( m = \tan(\phi) \) is a thermodynamic density

\[ H = \varepsilon \sum_{ij} |h_i - h_j| \quad \text{integer } h_i \]

Just kinks, good at low T

E.D. Williams et al.

Al/Si(111) - (\( \sqrt{3} \times \sqrt{3} \))R30°, STM
Vicinals as growth templates: controlled unidirectional defects in step-flow growth (vs. nucleation on flat)

Competition of characteristic lengths for diffusion, nucleation, distance to step

AFM of SrTiO$_3$(001)

Irregular borders locked in by 1 ML

Step pattern persists

Naito et al., Physica C 305 (’98) 233
Steps & kinks can alter chemical activity ⇒ applications in catalysis

R-3-MCHO molecules on roughened Cu(643)

Horvath & Gellman, Topics in Catalysis 25 (’03)
Enantioselectivity at chiral metal surfaces

D-glucose

L-glucose

Chiral Pt (643)

Attard, J. Phys Chem B 105 (’01) 3158

Sholl et al., J. Phys Chem B 105 (’01) 4771
Steps as Growth Template for Nanowires

Gd disilicide nanowires on Si(111)
11 nm wide
straight, limited by kinks on steps

Himpsel group, Nanotechnology 13 (’02) 545
Steps as polymers in 2D ⇒ non-crossing

Fig. 1. Model for a two-dimensional fiber structure.
Models & Key Energies
Discrete/atomistic $\rightarrow$ Step Continuum

$\varepsilon$  energy of unit height difference between NN sites + hopping barriers, attach/detach rates

$kink energy$

$\tilde{\beta}$  step stiffness $\beta(\theta) + \beta''(\theta)$: inertial “mass” of step

$A$  strength of step-step repulsion $A/\ell^2$

$\Gamma$  rate parameter, dependent on microscopic transport mechanism

Main test: Self-consistency of these 3 parameters to explain many phenomena
Coarse-grain: Relation of 3 nano/mesoscale parameters to atomistic energies??
Experimental Probes of Vicinals

- **Diffraction of electrons or atoms**
  \[ \textbf{k}-\text{space, sensitive to order} \]

- **STM (scanning tunneling microscope)**
  atomic resolution, but scanning

- **LEEM (low-energy electron microscope)**
  nanoscale resolution, real-time image, expensive

- **REM (reflection electron microscopy)**
  nanoscale resolution in 1 direction, real-time image

*All photos are of Si (111)*
Steps as Brownian strings: *Langevin* “capillary wave” approach

\[
\frac{\partial x(y, t)}{\partial t} = -\text{restoring “force” } + \text{ noise}(y, t)
\]
e.g. heal curvature

\[
x(y, t) = \sum_q e^{i q y} x_q(t) \quad \text{to deal with } \nabla y
\]

\[
\frac{\partial x_q(t)}{\partial t} = -\frac{x_q(t)}{\tau_q} + \text{noise}(q, t)
\]

\[
G_q(t-t') = \left\langle |x_q(t)-x_q(t')|^2 \right\rangle = \frac{2k_B T}{\beta q^2 L_y} \left( 1 - e^{-|t-t'|/\tau_q} \right)
\]

saturation \( G_q \Rightarrow \text{stiffness} \)

**Single value of } y \)

\[
\tau_q^{-1} = \frac{\tilde{\beta}}{k_B T} \times \begin{cases} 
\Gamma_{\text{attach}} q^2 & \text{EC/AD : curvature-driven} \\
2\Gamma_{\text{diffu}} |q|^3 & \text{TD} \\
\Gamma_{\text{edge}} q^4 & \text{PD/SED : } -\nabla^2 \text{curvature} 
\end{cases}
\]

\( \tau_q^{-1} \Rightarrow \text{transport mode & associated } \Gamma \)

or early-time exponent \( \Rightarrow \phantom{t^{1/2}} \& \phantom{t^{1/3}} \phantom{t^{1/4}} \)

\[
G(t) = \left\langle [x(t_0+t)-x(t_0)]^2 \right\rangle_{y_0, t_0} \propto \begin{cases} 
t^{1/2} \\
t^{1/3} \\
t^{1/4}
\end{cases}
\]
Island — Adatom or Vacancy — Defined by Nearly Circular Step!
### Isolated Step Fluctuations: Signatures of Dominant Mass Transport Mechanism

<table>
<thead>
<tr>
<th></th>
<th>EC or AD (ADL)</th>
<th>TD (DL)</th>
<th>PD</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Limited by</strong></td>
<td>At/de/tach at step</td>
<td>Terrace diffu'n</td>
<td>Step-edge diffu'n</td>
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<tr>
<td><strong>Fluctuation healing</strong></td>
<td>(y^2)</td>
<td>(y^3)</td>
<td>(y^4)</td>
</tr>
<tr>
<td><strong>time--width</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Size dep. of island</strong></td>
<td>(R^{-1})</td>
<td>(R^{-2})</td>
<td>(R^{-3})</td>
</tr>
<tr>
<td><strong>diffu'n, (R \propto \sqrt{\text{area}})</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>(w^2(t))</strong></td>
<td>(t^{1/2})</td>
<td>(t^{1/3})</td>
<td>(t^{1/4})</td>
</tr>
<tr>
<td><strong>Island area decay</strong></td>
<td>(t)</td>
<td>(t^{2/3})</td>
<td>N/A</td>
</tr>
<tr>
<td><strong>Evolution of atom/vacancy island</strong></td>
<td>Shrink to round point (Grayson's Thm)</td>
<td></td>
<td>Wormlike, pinch-off</td>
</tr>
<tr>
<td><strong>Height decay of cone [&quot;facet&quot;]</strong></td>
<td>(t^{1/4})</td>
<td>(t^{1/4})</td>
<td>N/A</td>
</tr>
<tr>
<td><strong>Height decay of paraboloid [rough]</strong></td>
<td>(t^{1/3})</td>
<td>(t^{2/5})</td>
<td>N/A</td>
</tr>
</tbody>
</table>
Terrace-Width Distribution $P(s)$ for Special Cases

"Perfect Staircase" $\ell = \langle \ell \rangle \equiv 1/\tan \phi$ $s \equiv \ell / \langle \ell \rangle$ $\langle \ell \rangle$ is only characteristic length in $x$

Straight steps, randomly placed

Geometric distribution: $P(s) = e^{-s}$

Meandering steps

No step crossings

Scaled TWD: $P(s)$ indep. of $\langle \ell \rangle$

"entropic" (& energetic) repulsion

TSK
terrace-step-kink

kink energy $\varepsilon$

"static correlation" $\langle x_n(y) - x_{n-1}(y) - \langle \ell \rangle \rangle_{y,n}$

"Maryland notation"
Terrace-Width Distribution $P(s)$ for Special Cases

"Perfect Staircase" $\ell = \langle \ell \rangle \equiv 1/\tan \phi$  \hspace{1em} $s \equiv \ell / \langle \ell \rangle$

Straight steps, randomly placed
Geometric distribution: $P(s) = e^{-s}$

Step Continuum Model

Meandering steps
No step crossings

"Maryland notation"

Scaled TWD: $P(s)$ indep. of $\langle \ell \rangle$

"static correlation" $\langle x_n(y) - x_{n-1}(y) - \langle \ell \rangle \rangle_{y,n}$

TSK

stiffness $\tilde{\beta}$
Terrace-Width Distribution $P(s)$ for Special Cases

"Perfect Staircase" $\ell = \langle \ell \rangle \equiv 1/\tan \phi$  $s \equiv \ell / \langle \ell \rangle$

Straight steps, randomly placed
Geometric distribution: $P(s) = e^{-s}$

World lines of fermions evolving in 1D

No step crossings

Scaled TWD: $P(s)$ indep. of $\langle \ell \rangle$

"static correlation" $\langle x_n(y) - x_{n-1}(y) - \langle \ell \rangle \rangle_{y,n}$

Meandering steps

TSK

stiffness $\tilde{\beta} \rightarrow \text{“mass”}$

World lines of fermions evolving in 1D
Origin of elastic (dipolar) step repulsions

- Frustration of relaxation of terrace atoms between steps

\[ U(\ell) = \frac{A}{\ell^2} \]  
(Same \( y \) for points on two interacting steps separated by \( \ell \) along \( x \) \( \Rightarrow \) "instantaneous")

- Metallic surface states \( \Rightarrow \) additional oscillatory term in \( U \)

Importance of step repulsions

- 1 of 3 parameters of continuum step model of vicinals
- Determine 2D pressure
- Determine morphology: e.g. bunch or pair
- Drives kinetic evolution in decay
- Elastic and entropic repulsions \( \propto \ell^{-2} \)
  \( \Rightarrow \) universality of \( \langle \ell \rangle^{-1} P(\ell) \) vs. \( s = \ell \langle \ell \rangle \) so \( P(s;\langle \ell \rangle) \rightarrow P(s) \) scaling
Essence of Gruber-Mullins (MF)

Single active step meanders between 2 steps separated by twice mean spacing.

Fermion evolves in 1D between 2 fixed infinite barriers $2\langle \ell \rangle$ apart.
Particle in 1D Box vs. Exact

\[ E = \int \beta \sqrt{1 + x^2} \, dy \sim \text{const.} + \int \frac{\beta x^2}{2} \, dx^2 \]

1-D Schrödinger eqn

\[ \frac{\hbar^2}{2m} \to \frac{(k_B T)^2}{2\beta} \]

- Free fermions: repulsion just entropic

\[ \psi_0 = \frac{1}{\langle \ell \rangle} \sin \left( \frac{\pi x}{2\langle \ell \rangle} \right) \]

\[ E_0 = \frac{(k_B T)^2}{8\beta \langle \ell \rangle} \]

- \( U(\ell) = A/\ell^2 \), large \( A \)

\[ \psi_0 \propto e^{-x^2/4w^2} \]

\[ w^4 = \frac{(k_B T)^2}{8\beta U''(\langle \ell \rangle)} \]

\[ w = \text{const.} \tilde{A}^{-1/4}(\ell) \]

\[ A \text{ enters only as } \tilde{A}: \]

\[ \tilde{A} \equiv \frac{\tilde{\beta} A}{(k_B T)^2} \]

\[ \text{const. changes with approximation} \]
Steps in 2D $\rightarrow$ fermion worldlines in 1D

- Step non-crossing $\Rightarrow$ fermions or hard bosons
- Energy $\propto$ path-length $\times$ free energy/length $\beta$, expand $\Rightarrow$ 1D Schrödinger eqn., $m \rightarrow$ stiffness $\beta$
- Analogous to polymers in 2D (deGennes, JCP '68)
- Only dependence on $A$ via $\tilde{\Lambda} \equiv \beta A/(k_B T)^2$
- Mean-field (Gruber-Mullins): 1 active step, $0 \leq s \leq 2$
  - $\tilde{\Lambda} = 0$: particle in box, $P(s) = \psi_0^2 \propto \sin^2(\pi s/2)$, $\varepsilon_0 \propto T^2/\beta \langle \ell \rangle^2 \rightarrow$ entropic repulsion
  - $\tilde{\Lambda} \geq 1^{1/2}$: parabolic well, $P(s) \propto \exp[-(s-1)^2/2w_M^2]$, $w_M \propto \tilde{\Lambda}^{-1/4} \langle \ell \rangle$
- $\tilde{\Lambda} \rightarrow \infty$: “phonons”, variance of $P(s)$ is $2w_M^2$, not $w_M^2$
Wigner Surmise (WS) for TWD (terrace-width distribution)

\[
\begin{align*}
\tilde{A}_1 &= -1/4 : \quad P_1(s) = \frac{\pi}{2} s \exp \left(-\frac{\pi}{4} s^2\right) \\
\tilde{A}_2 &= 0 : \quad P_2(s) = \frac{3\pi}{4} s^2 \exp \left(-\frac{4}{\pi} s^2\right) \\
\tilde{A}_4 &= 2 : \quad P_4(s) = \left(\frac{64}{9\pi}\right)^3 s^4 \exp \left(-\frac{64}{9\pi} s^2\right)
\end{align*}
\]

\[U(\ell) = \frac{A}{\ell^2}\]

Generalizing from the special cases:
- The three special cases correspond to \( q = 1, 2, \) and 4.
- \( \tilde{A} \) and \( q \) are related by: \( \tilde{A} = (q - 2)\ell/4 \); \( q = 1 + \sqrt{1 + 4\tilde{A}} \)
- Simplest interpolation expression:
  \[
  P_q(s) = a_q s^q \exp(-b_q s^2)
  \]
- Two conditions on \( P_q(s) \): normalization & unit mean
  \[\Rightarrow\] values of \( a_q, b_q \) (in terms of \( \Gamma \) functions),

WS \rightarrow GWS
Physical Ideas Behind Application of Random Matrices


Standard stat mech: ensemble of identical physical systems with same Hamiltonian but different initial conditions; Wigner: ensemble of dynamical systems governed by different H's with some common symmetry property, seeking generic properties of ensemble due to symmetry.

Dyson, using group-theory results from Wigner, showed 3 generic ensembles:

1) time-reversal invariant with rotational symmetry:
   \( H_{mn} = H_{nm} = H^*_{mn} \) (orthogonal)

2) time reversal violated (e.g. electron in fixed B)
   \( H_{mn} = H^\dagger_{mn} \) (unitary)

3) time-reversal invariant with 1/2-integer spin & broken rotational symmetry:
   \( H(0)_{mn} I - i \Sigma_j H(i)_{mn} \sigma_j \) (symplectic)

\( \sigma_j \): Pauli spin matrices, \( j=1,2,3 \); \( I \): 2x2 unit matrix; \( H(0) \) all real, \( H(0) \) sym, others asym

Wigner: for convenience, Gaussian weights \( P(H) \propto \exp[-(\beta N/\lambda^2) \text{ tr } H^2] \)

- Gaussian Orthogonal Ensemble: \( \beta=1 \)
- GUnitaryE: \( \beta=2 \)
- GSymplecticE: \( \beta=4 \)

GRMT useless for average quantities, but fluctuations for large number of levels becomes independent of the form of the level spectrum and of the Gaussian weight factors, and attains
1957: Wigner surmise for $\beta=1$: $p_1(s) = a_1 s^1 \exp(-b_1 s^2)$, where $p$ is the distribution function of nearest-neighbor energy levels, with $s$ the real spacing over the [local] mean.

1960-62: Dyson: circular ensembles: CircularOE, CUE, CSE; NN unitary matrices, eigenvalues $\exp[i\theta_\mu]$, $\mu=1,\ldots,N$.

N-particle Coulomb gas on a circle (i.e. in 1D), with [shifted] inverse temperature $\beta$.

**Major ingredient:** von Neumann–Wigner level repulsion: 2 states connected by a non-vanishing matrix element repel each other—degree of repulsion is determined by symmetry of Hamiltonian—"A simple counting argument leads directly to the exponent $\beta = 1, 2, 4$ in the typical factor $|E_\mu - E_\nu|^\beta$ in the Vandermonde determinant."

Sutherland Hamiltonian for $N$ particles (spinless fermions) on a circle:

$$-\frac{\hbar^2}{2m} \sum_{i=1}^{N} \frac{\partial^2}{\partial \lambda_i^2} - \frac{\beta}{2} \left( \beta - 2 \right) \left( \frac{\pi}{N} \right)^2 \sum_{i<j} \frac{1}{\sin^2 \left\{ \pi \left( \lambda_i - \lambda_j \right) / N \right\} }$$

Application to specific step system: M. Lässig, "Vicinal Surfaces and the Calogero–Sutherland Model," Phys. Rev. Lett. 77 (96) 526, for Song & Mochrie's observation of tricritical behavior on vicinal Si (113).

**OTHER APPLICATIONS of RMT**

Localization theory—ensemble of impurity potentials

Clarifies various regimes in mesoscopic physics: clean, ballistic, ergodic, diffusive, critical, localized

Transport in quasi-1D wires

Fluctuations of persistent currents (esp. for non-interacting electrons)

Level spectra of small metallic particles & their response to EM field

Atomic nuclei, atoms and molecules

Classical chaos (e.g. Bunimovich stadium, Sinai billiard)

QCD, supersymmetry

2D quantum gravity
Examples of NN spacing distributions with GOE ($\varrho = 1$)

Fig. 1. Nearest-neighbor spacing distribution for the “Nuclear Data Ensemble” comprising 1726 spacings (histogram) versus $s = S/D$ with $D$ the mean level spacing and $S$ the actual spacing. For comparison, the RMT prediction labelled GOE and the result for a Poisson distribution are also shown as solid lines. Taken from Ref. [1].

Fig. 4. The nearest-neighbor spacing distribution versus $s$ (defined as in Fig. 1) for the Sinai billiard. The histogram comprises about 1000 consecutive eigenvalues. Taken from Ref. [5].

Fig. 6. Nearest-neighbor spacing distribution for elastomechanical modes in an irregularly shaped quartz crystal.

T. Guhr et al. / Physics Reports 299 (1998) 189
RMT & financial data: Cross-correlations of price fluctuations of different stocks, using $P_1(s)$

V. Plerou, ..., T. Guhr, and H. E. Stanley, PRE 66 (’02) 066126
Headway statistics of buses in Mexican cities, using $P_2(s)$

M. Krbálek & P. Šeba, J. Phys. A 36 ('03) L7; 33 ('00) L229

Headway: time interval $\Delta t$ between bus and next bus passing the same point
No timetable for buses in Mexico; independent drivers seek to optimize # riders/fares

WS $P_2(s)$ better than CA because in CA, correlations only between NNs
Modelling gap-size distribution of parked cars using RMT

A.Y. Abul-Magd, Physica A 368 (’06) 536

S. Rawal, G.J. Rodgers, Physica A 346 (’05) 621

Unlike random sequential process, Coulomb gas extends repulsion beyond geometric size.
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\[ U(\ell) = \frac{A}{\ell^2} \]

Generalizing from the special cases:

- The three special cases correspond to \( q = 1, 2, \) and 4.

- \( \tilde{A} \) and \( \varrho \) are related by:
  \[ \tilde{A} = (\varrho - 2)\varrho / 4; \quad \varrho = 1 + \sqrt{1 + 4\tilde{A}} \]

- Simplest interpolation expression:
  \[ P_\varrho(s) = a_\varrho s^\varrho \exp(-b_\varrho s^2) \]

- Two conditions on \( P_\varrho(s) \): normalization & unit mean
  \( \Rightarrow \) values of \( a_\varrho, b_\varrho \) (in terms of \( \Gamma \) functions),

\[ \tilde{A}_1 = -1/4: \quad P_1(s) = \frac{\pi}{2} s \exp\left(-\frac{\pi}{4} s^2\right) \]
\[ \tilde{A}_2 = 0: \quad P_2(s) = \frac{\varpi}{\pi^2} s^2 \exp\left(-\frac{4}{\pi} s^2\right) \]
\[ \tilde{A}_4 = 2: \quad P_4(s) = \left(\frac{64}{9\pi}\right)^{1/2} s^4 \exp\left(-\frac{64}{9\pi} s^2\right) \]
Experiments measuring variances of TWDs

<table>
<thead>
<tr>
<th>Vicinal</th>
<th>T (K)</th>
<th>$\sigma^2$</th>
<th>$q$</th>
<th>$\tilde{A}$</th>
<th>$A_W/A_G$</th>
<th>$A_W$ (eV $\tilde{A}$)</th>
<th>Experimenters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pt(1 1 0)-(1 x 2)</td>
<td>298</td>
<td>0.122</td>
<td>4.1</td>
<td>2.2</td>
<td>0.77</td>
<td>0.005</td>
<td>Swamy, Bertel [36]</td>
</tr>
<tr>
<td>Cu(19, 17, 17)</td>
<td>353</td>
<td>0.091</td>
<td>4.8</td>
<td>4</td>
<td>1.37</td>
<td>0.004</td>
<td>Geisen [5,54]</td>
</tr>
<tr>
<td>Si(1 1 1)</td>
<td>1173</td>
<td>0.16</td>
<td>5.5</td>
<td>6.9</td>
<td>0.64</td>
<td>0.012</td>
<td>Bermond, Métois [55]</td>
</tr>
<tr>
<td>Cu(1,1,13)</td>
<td>348</td>
<td>0.085</td>
<td>6.0</td>
<td>7.9</td>
<td>1.64</td>
<td>0.012</td>
<td>Geisen [5,56]</td>
</tr>
<tr>
<td>Cu(11,7,7)</td>
<td>306</td>
<td>0.068</td>
<td>6.4</td>
<td>6</td>
<td>0.67</td>
<td>0.012</td>
<td>Geisen [5,54]</td>
</tr>
<tr>
<td>Cu(1 1 1)</td>
<td>313</td>
<td>0.044</td>
<td>8.7</td>
<td>7.0</td>
<td>0.17</td>
<td>0.02</td>
<td>Geisen [5,54]</td>
</tr>
<tr>
<td>Cu(1 1 1)</td>
<td>313</td>
<td>0.073</td>
<td>6.0</td>
<td>7.0</td>
<td>0.17</td>
<td>0.02</td>
<td>Williams [57]</td>
</tr>
<tr>
<td>Ag(1 0 0)</td>
<td>300</td>
<td>0.073</td>
<td>8.7</td>
<td>7.0</td>
<td>0.17</td>
<td>0.02</td>
<td>Williams [57]</td>
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<tr>
<td>Cu(1 1 1)</td>
<td>320</td>
<td>0.070</td>
<td>6.7</td>
<td>7.0</td>
<td>0.17</td>
<td>0.02</td>
<td>Williams [57]</td>
</tr>
<tr>
<td>Si(1 1 1)-(7 x 7)</td>
<td>1100</td>
<td>0.068</td>
<td>6.4</td>
<td>7.0</td>
<td>0.16</td>
<td>0.01</td>
<td>Fujita...Ichikawa [59]</td>
</tr>
<tr>
<td>Si(1 1 1)-(1 x 1)Br</td>
<td>853</td>
<td>0.068</td>
<td>6.6</td>
<td>6.4</td>
<td>1</td>
<td>1.8</td>
<td>Schwennicke...Williams [60]</td>
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<tr>
<td>Si(1 1 1)-Ga</td>
<td>823</td>
<td>0.068</td>
<td>6.6</td>
<td>7.6</td>
<td>0.16</td>
<td>0.02</td>
<td>Barbier et al. [21]</td>
</tr>
<tr>
<td>Si(1 1 1)-Al $\sqrt{3}$</td>
<td>1040</td>
<td>0.058</td>
<td>7.6</td>
<td>10.5</td>
<td>1.85</td>
<td>2.2</td>
<td>Geisen [5,56]</td>
</tr>
<tr>
<td>Cu(1, 1, 11)</td>
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<td>0.053</td>
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<td>7.0</td>
<td>1.67</td>
<td>0.1</td>
<td>Geisen [5,56]</td>
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<tr>
<td>Cu(1, 1, 13)</td>
<td>285</td>
<td>0.044</td>
<td>10</td>
<td>15</td>
<td>1.95</td>
<td>0.02</td>
<td>Geisen [5,56]</td>
</tr>
<tr>
<td>Pt(1 1 1)</td>
<td>900</td>
<td>0.020</td>
<td>24</td>
<td>3.8 x 10^3</td>
<td>2.92</td>
<td>(27 ± 5) x 10^2</td>
<td>Hahn...Kern [61]</td>
</tr>
<tr>
<td>Si(1 1 3) rotated</td>
<td>1200</td>
<td>0.004</td>
<td>124</td>
<td>2.92</td>
<td>10^2</td>
<td>6</td>
<td>van Dijken, Zandvliet, Poelsema [9]</td>
</tr>
</tbody>
</table>
Monte Carlo data confronts approximations

Dots: MC data
Line: Wigner
Dashes: Gruber-Mullins (mean field)
Long-short [-short]: Grenoble
  (no entropic int’n, EA)
Long-long-short-short: Saclay
  (continuum roughening, R)

Lower plot highlights differences:
remove $\rho^{-1}$ asymptotic decay
Wigner is best, quantitatively and conceptually

Hailu Gebremariam et al.,
Phys. Rev. B 69 ('04)125404
Why Look for Fokker-Planck Equation for TWD?

• Justification/derivation of generalized continuum Wigner surmise (beyond $H_{\text{eff}}$ of Richards et al.) since no symmetry basis for $\rho \neq 1, 2, \text{ or } 4$

• Dynamics: how non-equilibrium TWD (e.g. step bunch) evolves toward equilibrium

• Quench or upquench: sudden change of $T$ does not change $A$ much but changes $\tilde{A}$ (and so $\rho$) considerably

• Connections with other problems, e.g. capture zone distribution (& Heston model of econophysics)
Derivation of Fokker-Planck for TWD

- Start with Dyson Coulomb gas/Brownian motion model: repulsions $\propto 1/($separation$)$ & parabolic well
  \[ \dot{x}_i = -\gamma x_i + \sum_{i \neq j} \frac{\hat{q}}{x_i - x_j} + \sqrt{\Gamma}\eta \]

- Assume steps beyond nearest neighbors are at integer times mean spacing (cf. Gruber-Mullins)
  \[ \dot{s} = -\kappa s + \rho/s + \text{noise} \]

  \textit{Noise sets time scale.}

- Demand self-consistency for width of parabolic confining well: $\kappa \to 2b_\rho$

  \[ \frac{\partial P(s, \tilde{t})}{\partial \tilde{t}} = \frac{\partial}{\partial s} \left[ \left( 2b_\rho s - \frac{\rho}{s} \right) P(s, \tilde{t}) \right] + \frac{\partial^2}{\partial s^2} [P(s, \tilde{t})] \quad \to \quad P_\varphi(s) \]
Check of Fokker-Planck with Monte Carlo

cleaved $\rightarrow$ equilibrium

TSK model (no adatom carriers)

Best match for 1.4 FP time units = $10^3$ MCS

\[ \langle \ell \rangle = 6 \]
\[ N = 4 \]
\[ L_y = 200 \]

\[
\sigma^2 = \langle s^2 \rangle - \langle s \rangle^2 \text{ from } P(s,t), \text{ analytic solution of Fokker-Planck}
\]

\[
P(s, \tilde{t}) = 2\tilde{b}_q s^{\alpha+1} e^{\frac{\alpha \tilde{t}}{2}} I_\alpha \left( 2\tilde{b}_q se^{-\frac{\tilde{t}}{2}} \right) \exp\left[ -\tilde{b}_q (s^2 + e^{-\tilde{t}}) \right]
\]

\[ \alpha = (q-1)/2, \tilde{b}_q \equiv b_q / (1-e^{-\tilde{t}}) \]

As good agreement as might expect:
1) Metropolis rather than kinetic MC
2) Just NN step interactions in MC
3) Discrete at early times
Improved tests: Kinetic MC & SOS model

$$E_{\text{barrier}} = E_d + m E_a$$ \quad \text{breaking m bonds}

$$E_d = 0.9 - 1.1 \text{ eV}; \ E_a = 0.3 - 0.4 \text{ eV}$$

$$T = 520 - 580 \text{ K}$$

$$\langle \ell \rangle = 4-15, \ 5 \text{ steps, } 10000 \times L_x$$

Fit:

$$\sigma(t) = \sigma_{sat} \sqrt{1 - \exp(-t / \tau)}$$

Expect $\tau \propto \exp(E_{\text{barrier}}/k_B T)$

Find $E_{\text{barrier}} \approx 1 E_d + 3 E_a$
2 other situations of interest

**Step Bunch:** initially a delta function

\[
P(s, \tilde{t}) \rightarrow \frac{a_0 s^q e^{-\tilde{t}^2}}{(1 - e^{-\tilde{t}})(e^{\tilde{t}+1}/2)^{1/2}} \exp[-s^2 b_\sigma/(1 - e^{-\tilde{t}})]
\]

**Quench or upquench:** change from initial \( \rho_0 \) to \( \rho \), e.g. change in temperature

\[
P(s, \tilde{t}) = a_0 s^q e^{-\tilde{t}^2} \frac{(1 - e^{-\tilde{t}})^{\frac{\rho_0 - \rho}{2}}}{(1 - e^{-\tilde{t}}(1 - b_\sigma/b_{\sigma_0}))^{\frac{\rho_0 + 1}{2}}} \ _1F_1 \left( \frac{\rho_0 + 1}{2}, \frac{\rho + 1}{2}, \frac{\tilde{b}_\sigma s^2}{1 + (b_{\sigma_0}/b_\sigma)(e^{\tilde{t}} - 1)} \right)
\]
Consider a stock whose price $S_t$ obeys the stochastic differential equation of Brownian motion.

Volatility $\rightarrow$ stochastic variance obeys a mean-reverting stochastic DE.

The stationary PDF of volatility $\sigma$ is

$$
\Pi_*(\sigma) = \frac{2\alpha^{\alpha}}{\Gamma(\alpha)} \frac{\sigma^{2\alpha-1}}{\theta^{\alpha}} e^{-\alpha \sigma^2 / \theta}
$$

$\rightarrow s \sqrt{\frac{b_\rho}{\rho^{2-1}}}$

$2\alpha - 1 \rightarrow \varrho$

$\theta \rightarrow \langle \ell^2 \rangle$
Does growth flux (step motion) alter TWD?

Test: no energetic interaction ($\varphi=2$), 150 ML

- Narrower $\Rightarrow$ effective repulsion that rises with flux, higher $\varphi$, more Gaussian-like
- Decreased apparent stiffness $\tilde{\beta}$

20 steps, 1000x200, $T=723K$, $E_d=1.0eV$, $E_a=0.3eV$
Evolution of Island Structures: Simulations of $i=1$

*Circular Islands* Mulheran & Blackman, PRB 53 (96) 10261

Estimated size of island based on Voronoi polygon CZ $\propto$ actual size of island
Island Size Scaling, stable config $i$

**Dynamic scaling assumption**

$$N_s(\theta) = \theta S^{-2} f_i(s/S)$$

$$f_i(u) = C_i u^i e^{-ia_i u^{1/a_i}}$$

$$\Gamma[(i + 2)a_i] / \Gamma[(i + 1)a_i] = (ia_i)^{a_i}, \quad C_i = \frac{(ia_i)^{(i+1)a_i}}{a_i \Gamma[(i + 1)a_i]}$$

$i+1$ atoms: smallest stable island

**critical nucleus**

In contrast to Point-Island Rate Eqn for large D/F

$$f_i(u) = \frac{1}{i + 2} \left(1 - \frac{i + 1}{i + 2} u\right)^{-\frac{i}{i+1}}; \quad 0 \leq u \leq \frac{i + 2}{i + 1}$$

$$f_i(u) = 0; \quad u > \frac{i + 2}{i + 1}$$
Scaling During Growth in 1D: Going Beyond Mean-Field Rate Eqns.  Blackman & Mulheran, PRB 54 (96) 11681

$P_4(s)$ fits numerical data at least as well as B&M’s complicated theory expression (not expressible succinctly)

$d = 1 \Rightarrow \varrho = 2(i + 1)$

$P_4(s) \approx 2 \int_0^{2s} P_{3/2}(s')P_{3/2}(2s - s') \, ds'$

Distribution of gaps between point islands

Symbols denote various $D/F$ & $\theta$
Theory of CZ size distributions in growth, Mulheran & Robbie, EPL 49(00)617

\[ d = 2 \Rightarrow \varrho = i + 1 \]

Wigner distribution \( P_\varphi(s) \) fits much better than M&R theory

Island size distribution not so informative
Exp’t: Pentacene/SiO$_2$ Pratontop et al., PRB 69 (04) 165201

Gamma func’n

$$\Pi_\alpha(x) = \frac{\alpha^\alpha}{\Gamma(\alpha)} x^{\alpha-1} \exp(-\alpha x)$$

Why Gamma, not Wigner?

CZ behaves better

but $\Pi$ more skewed

Values of $p$ and $p_{cz}$

Deposition rate (nm/min)
Scale invariance in thin film growth: InAs quantum dots on GaAs(001)

AFM, 1.68 ML, 350x350nm², 500°C

Q dot volume distribution

Voronoi areas

\[ \Pi_{4,1}(s) \]

\[ P_2(s) \]

\[
\begin{array}{cccccccc}
\Theta (\text{ML}) & 1.65 & 1.68 & 1.70 & 1.73 & 1.79 & 1.85 \\
\alpha & 4.1 & 4.6 & 4.5 & 4.7 & 4.5 & 4.6 \\
\end{array}
\]
Why it works: Phenomenological theory

CZ does “random walk” with 2 competing effects on $ds/dt$:

1] Neighboring CZs hinder growth ⇒ external pressure, repulsion $B$
   leads to force $-KBs$  Also noise $\eta$

2] Non-symmetric confining potential, new island nucleated with large size so force stops fluctuations of CZ to tiny values
   In Dyson model, logarithmic interaction, so $+K (\ ) /s$

3] Can argue in 2D that $(\ )$ is $i + 1$
   using critical density $\propto s^i$, # sites visited in lifetime $\propto s^1$
   entropy $\propto$ - product $s^{i+1}$, & force $-\partial$ (entropy) / $\partial s$
   [Also $i + 1$ in 3D & 4D; but $2(i + 1)$ in 1D]

4] Combine ⇒ Langevin eq. $ds/dt = K [(2/d)(i +1)/s - Bs ] + \eta$  [d=1,2]

5] Leads to Fokker-Planck eq. with stationary sol’n $P_\varnothing(s)$
   cf. AP, HG, & TLE, Phys. Rev. Lett. 95 (05) 246101
Summary (see http://www2.physics.umd.edu/~einstein)

- Steps are useful for many applications, bear on many problems of current interest, and embody fascinating physics
- Sophisticated experiments, with powerful theoretical and computational calculations, allow for quantitative measurements that yield numerical assessment of key parameters and allow prediction of associated phenomena
- TWD of vicinals provides physical entrée to intriguing 1D fermion models & RMT, can connect to many other current physics issues --- universality in fluctuations --- Wigner surmise for 3 special cases based on explicit or implicit symmetry
- Generalized Wigner surmise $P_e(s) = a s^e e^{-bs^2}$ easy to use & describes universal fluctuations $\Rightarrow$ broad applications
- For TWD width $e \Rightarrow$ strength of elastic repulsion
- Fokker-Planck “derivation” & application to relaxation of steps from arbitrary initial configurations
- Focus on distribution of areas of capture zones, rather than island sizes; $e \Rightarrow$ critical nucleus size $i$ and spatial dimension of host lattice