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

MRSEC

Ted Einstein *Physics, U. of Maryland, College Park*

einstein@umd.edu http://www2.physics.umd.edu/~einstein

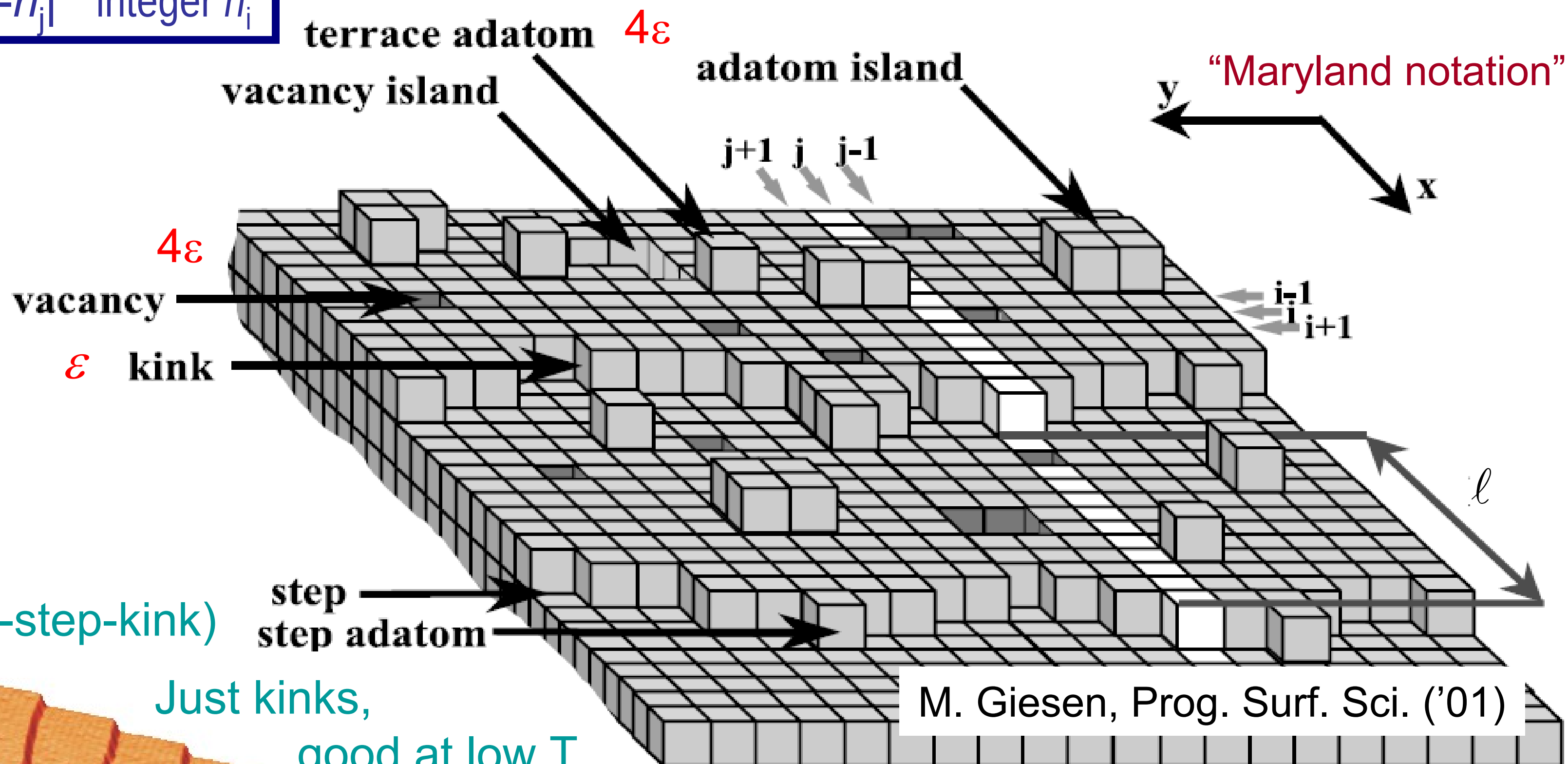
In collaboration with Alberto Pimpinelli, Hailu Gebremariam, Tim Stasevich, H.L. Richards, O. Pierre-Louis, S.D. Cohen, R.D. Schroll, N.C. Bartelt, and experimentalists Ellen D. Williams & J.E. Reutt-Robey at UM, M. Giesen & H. Ibach at FZ-Jülich, and J.-J. Métois at Marseilles

- 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 ϱ
- 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)

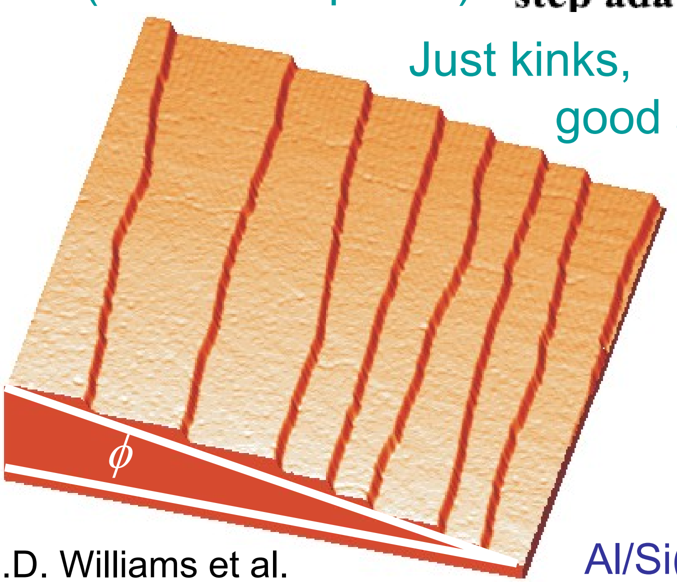
SOS (solid-on-solid) model of vicinals

$H = \varepsilon \sum_{\langle ij \rangle} |h_i - h_j|$ integer h_i



TSK (terrace-step-kink)

Just kinks,
good at low T



E.D. Williams et al.

Al/Si(111) - (√3×√3)R30°, STM

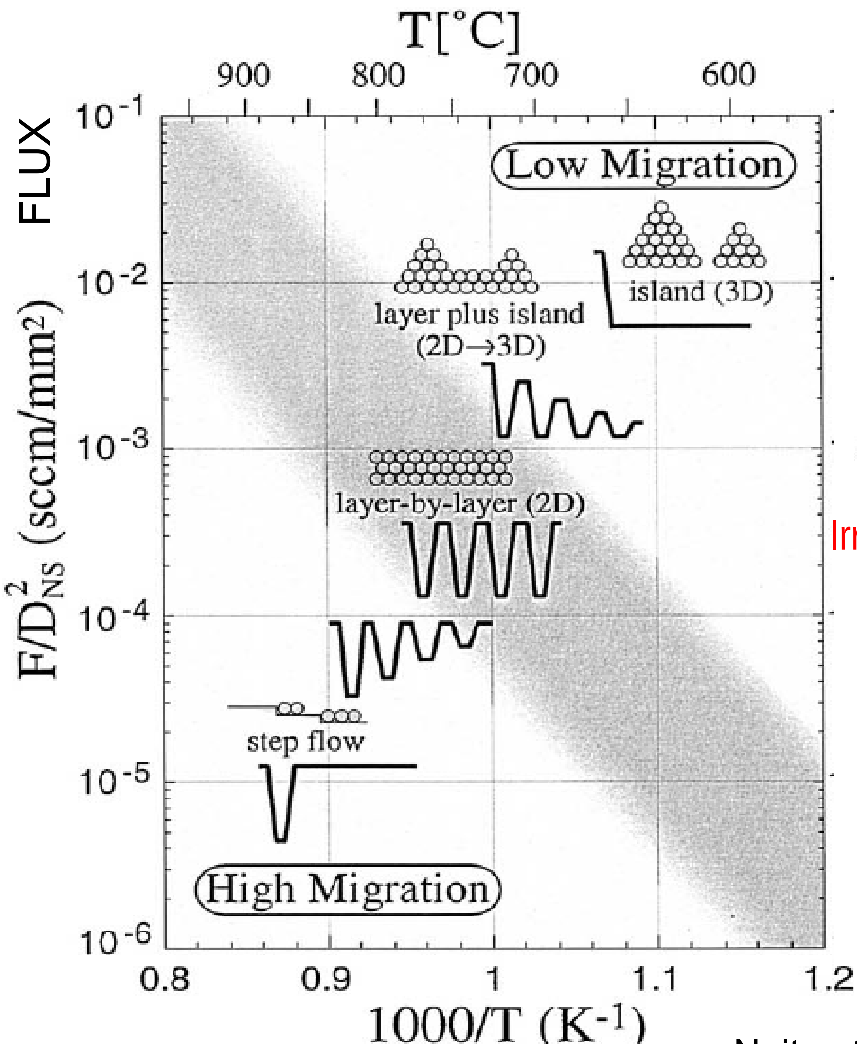
ϕ is the *misorientation angle*, fixed

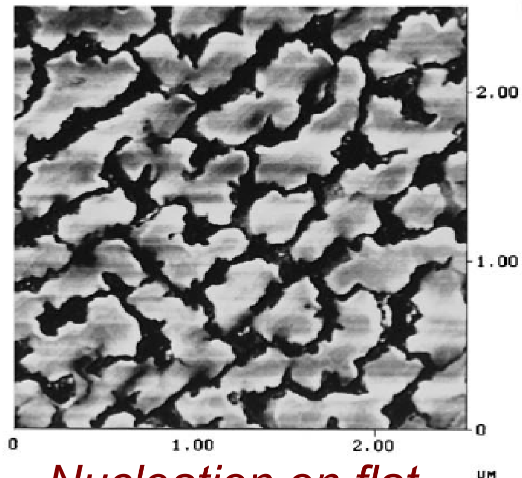
Mean step spacing $\langle \ell \rangle \propto 1/\tan(\phi)$

Slope $m = \tan(\phi)$ is a thermodynamic density

Vicinals as growth templates: controlled unidirectional defects in step-flow growth (vs. nucleation on flat)

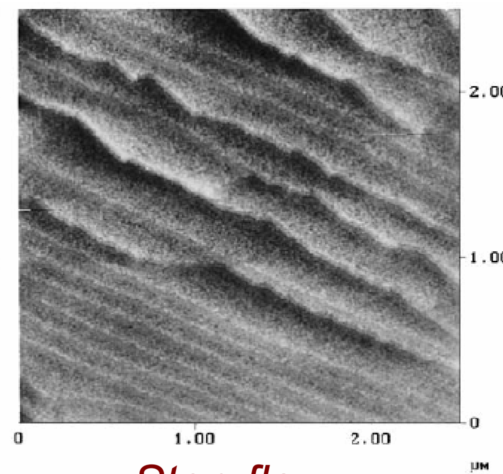
Competition of characteristic lengths for diffusion, nucleation, distance to step





Nucleation on flat

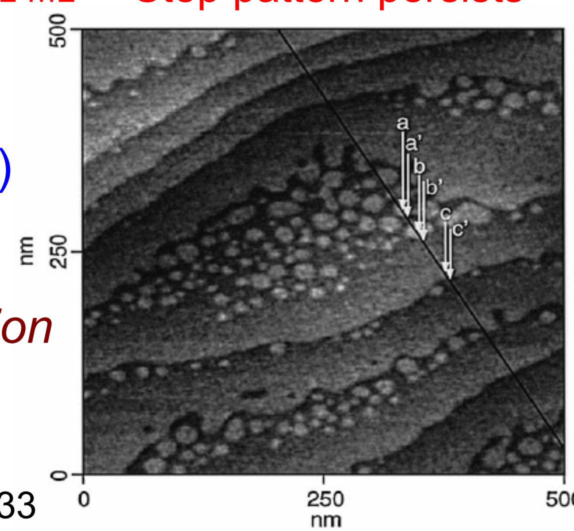
Irregular borders locked in by 1 ML



Step flow

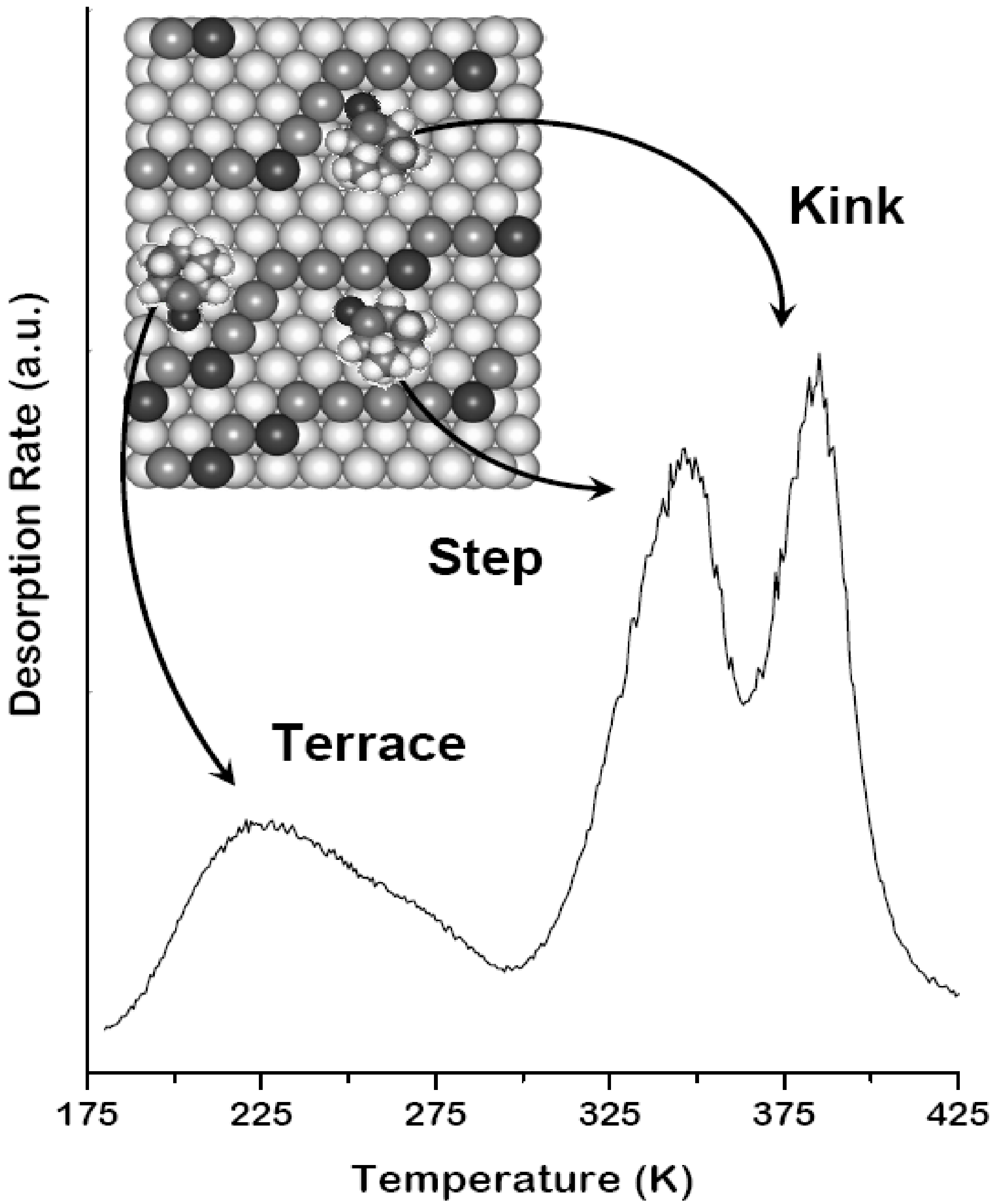
Step pattern persists

AFM of $SrTiO_3(001)$



Competition

Naito et al., Physica C 305 ('98) 233



Steps & kinks can alter chemical activity⇒ applications in *catalysis*

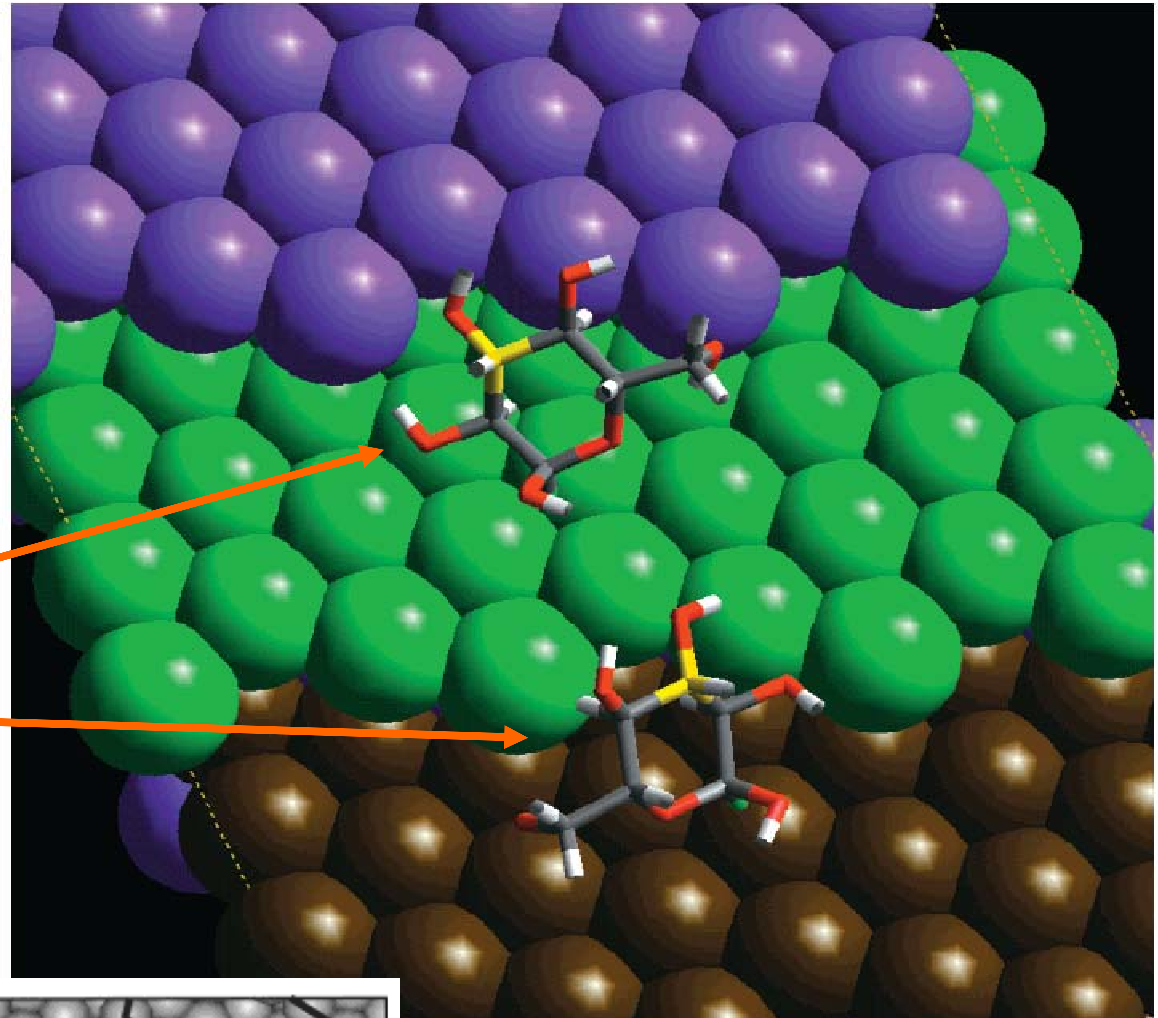
R-3-MCHO molecules on roughened Cu(643)

Horvath & Gellman, Topics in Catalysis 25 ('03)

Enantio-selectivity at chiral metal surfaces

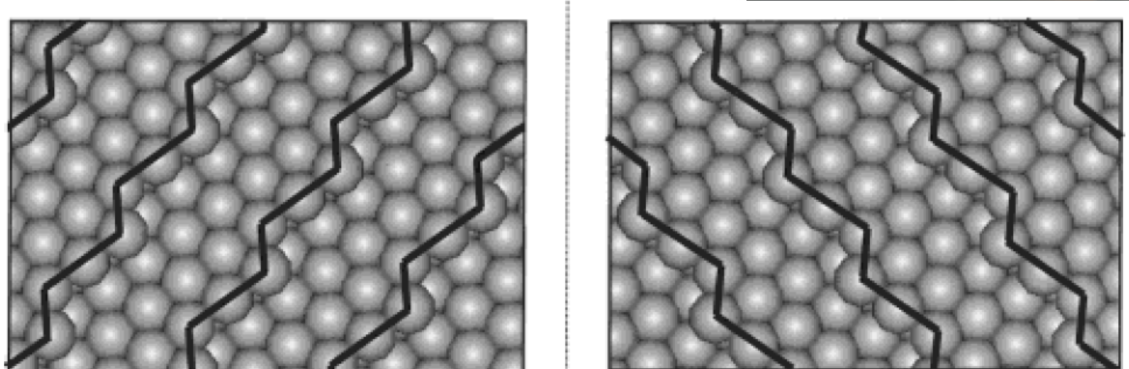
D-glucose

L-glucose



Attard, J. Phys Chem B 105 ('01) 3158

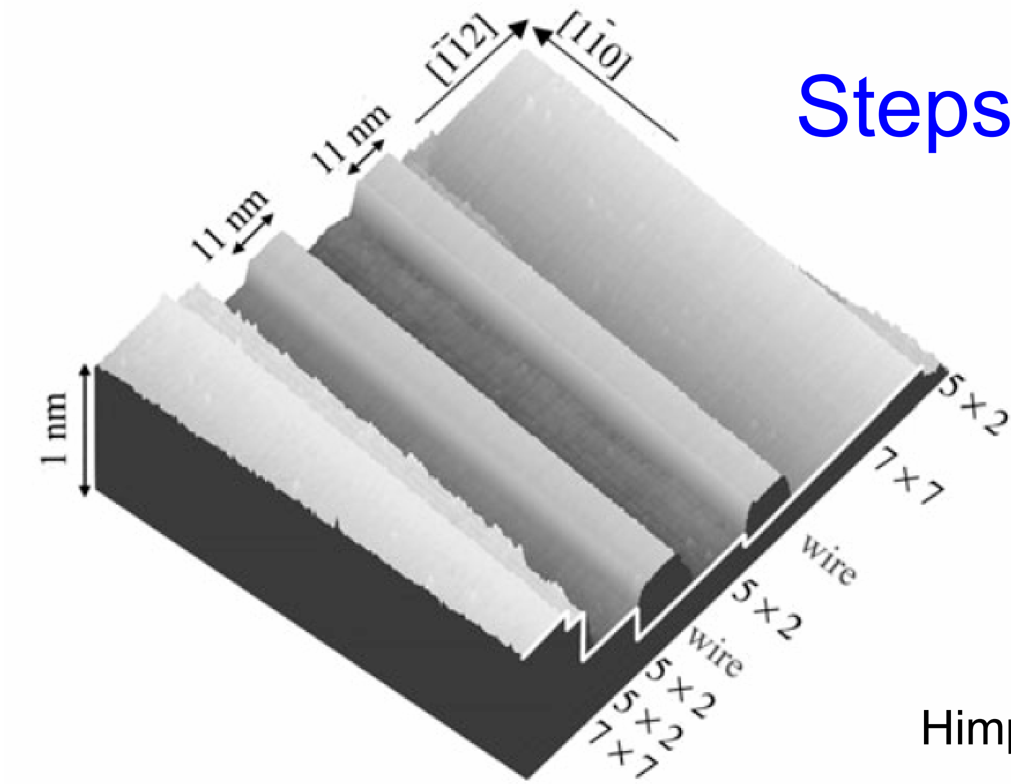
Chiral Pt (643)



fcc(643) fcc($\bar{6}\bar{4}\bar{3}$)

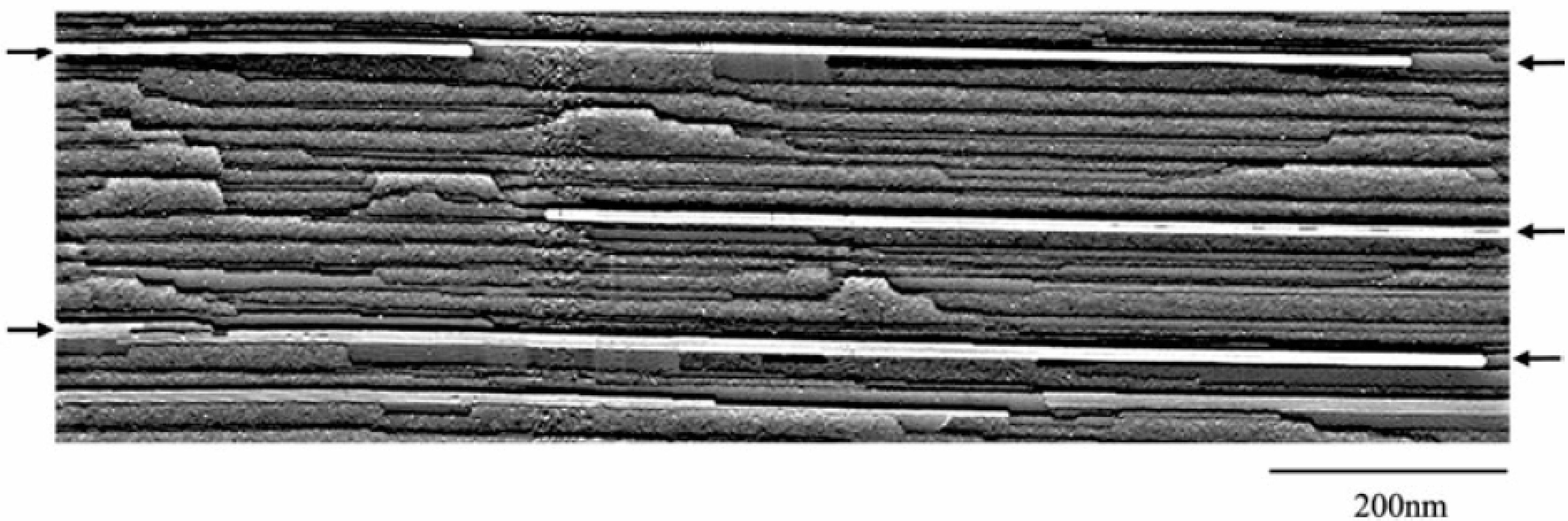
Sholl et al.,J. Phys Chem B 105 ('01) 4771

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

THE JOURNAL OF CHEMICAL PHYSICS VOLUME 48, NUMBER 5 1 MARCH 1968

Soluble Model for Fibrous Structures with Steric Constraints

P.-G. DE GENNES

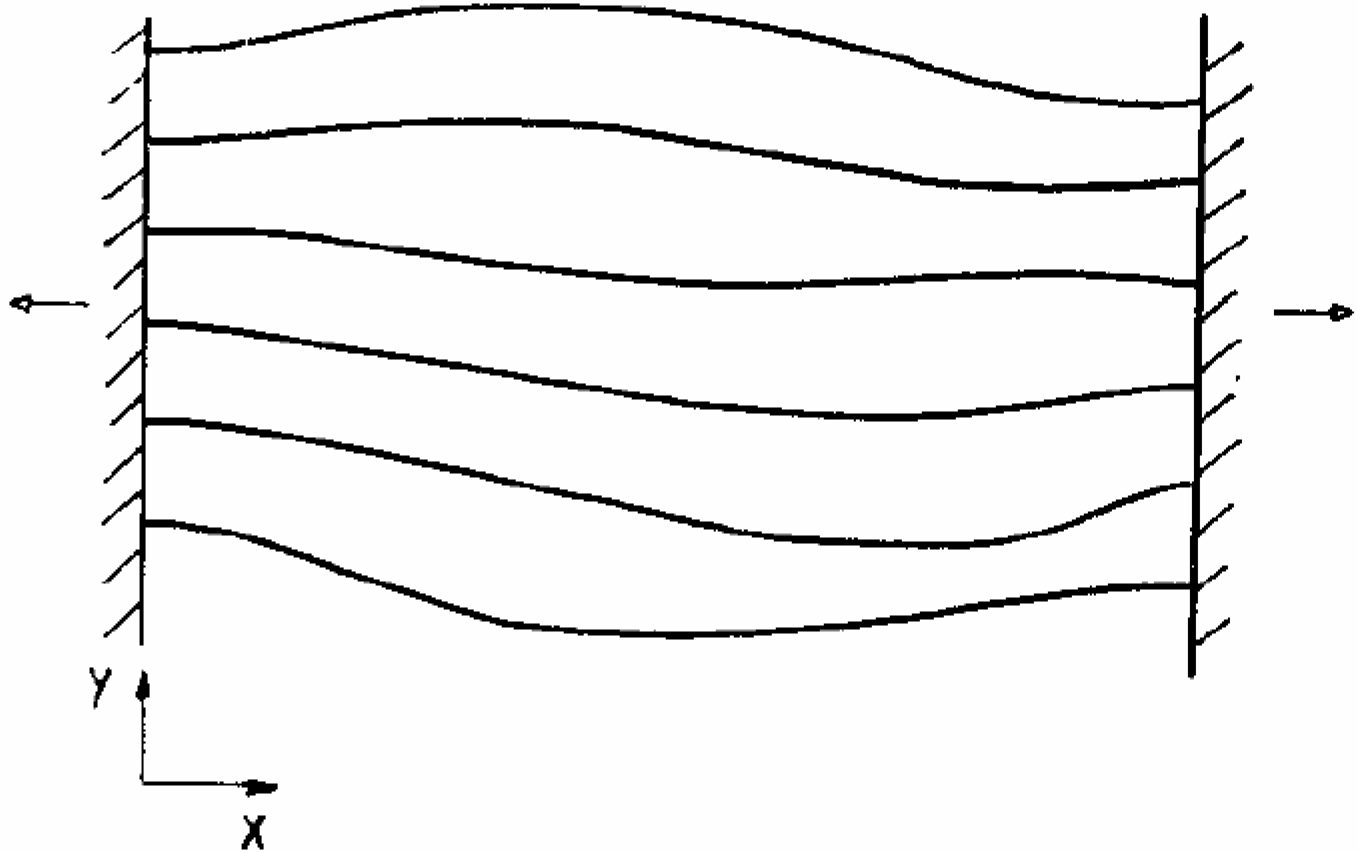
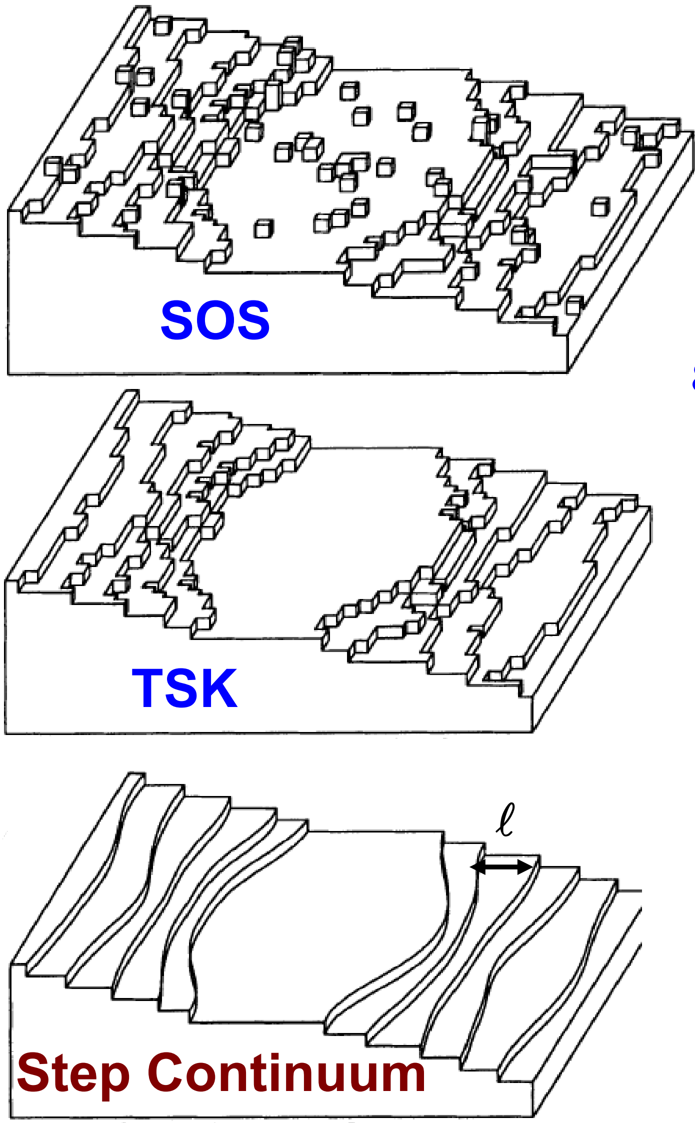


FIG. 1. Model for a two-dimensional fiber structure.

Models & Key Energies

Discrete/atomistic → Step Continuum



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

ε

kink energy

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

A *strength of step-step repulsion* A/ℓ^2

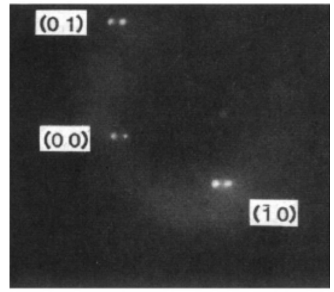
Γ *rate parameter, dependent on microscopic transport mechanism*

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

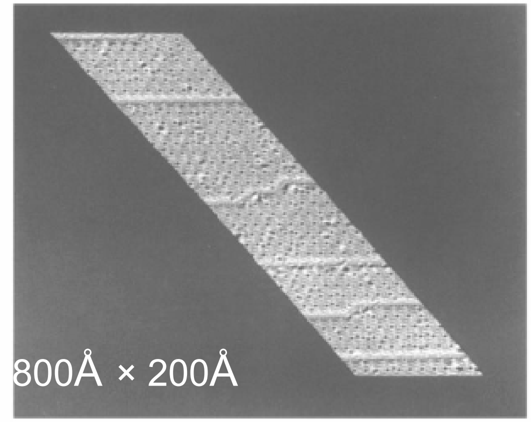
- Diffraction of electrons or atoms

  k-space, sensitive to order



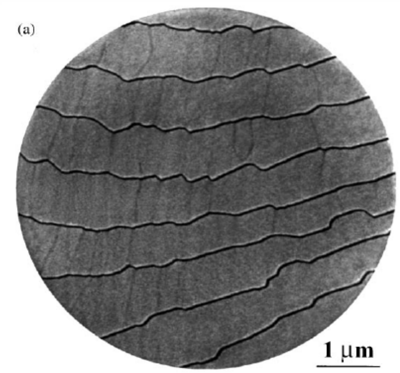
- STM (scanning tunneling microscope)

  atomic resolution, but scanning



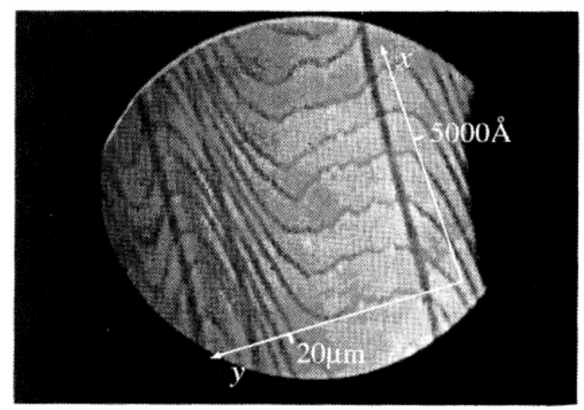
- LEEM (low-energy electron microscope)

  nanoscale resolution, real-time image, expensive



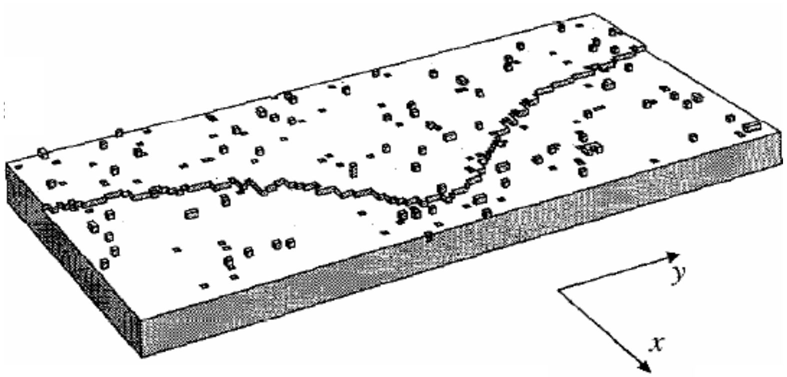
- REM (reflection electron microscopy)

  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

$$x(y,t) = \sum_q e^{iqy} x_q(t)$$ to deal with ∇y

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

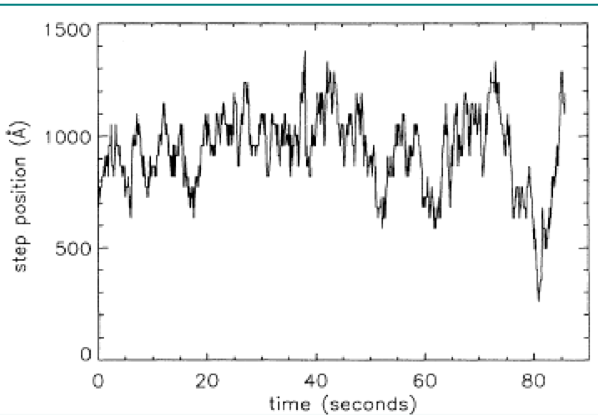
$$G_q(t-t') \equiv \left\langle \left| x_q(t) - x_q(t') \right|^2 \right\rangle = \frac{2k_B T}{\tilde{\beta} q^2 L_y} \left(1 - e^{|t-t'|/\tau_q}\right)$$

saturation $G_q \Rightarrow$ stiffness

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

$\tau_q^{-1} \Rightarrow$ transport mode & associated Γ

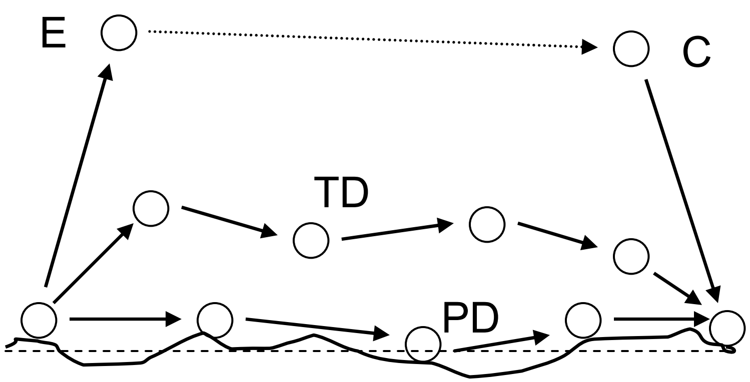
Single value of *y*



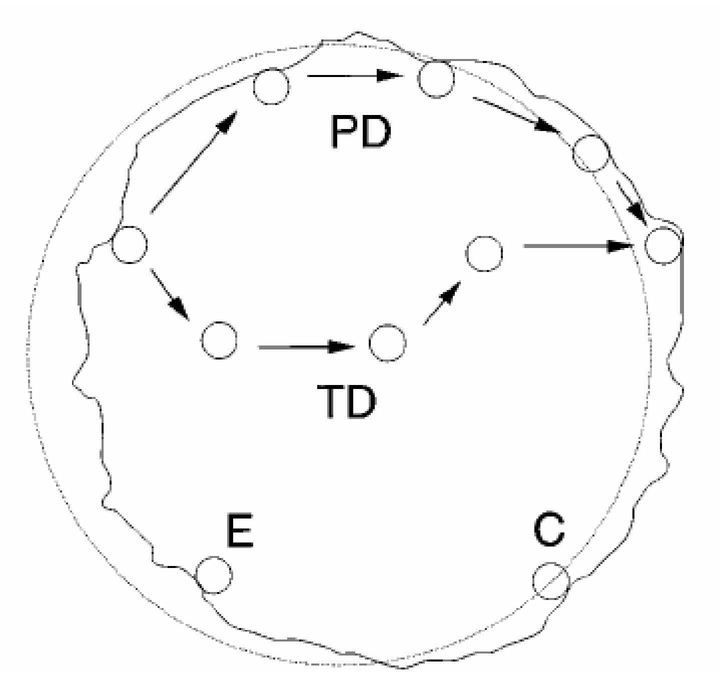
or early-time exponent $\Rightarrow$ " & "

$$G(t) \equiv \left\langle \left[x(t_0+t) - x(t_0) \right]^2 \right\rangle_{y_0,\, t_0} \propto \begin{cases} t^{1/2} \\ t^{1/3} \\ t^{1/4} \end{cases}$$

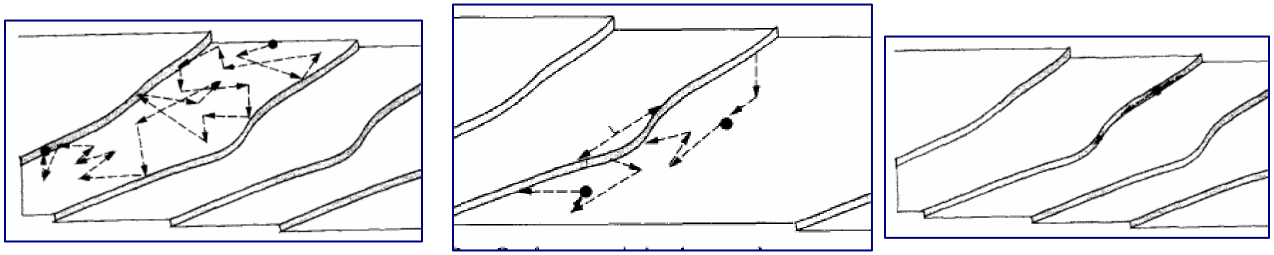
Island – Adatom or Vacancy – Defined by Nearly Circular Step!



vacancy island



Isolated Step Fluctuations: Signatures of Dominant Mass Transport Mechanism

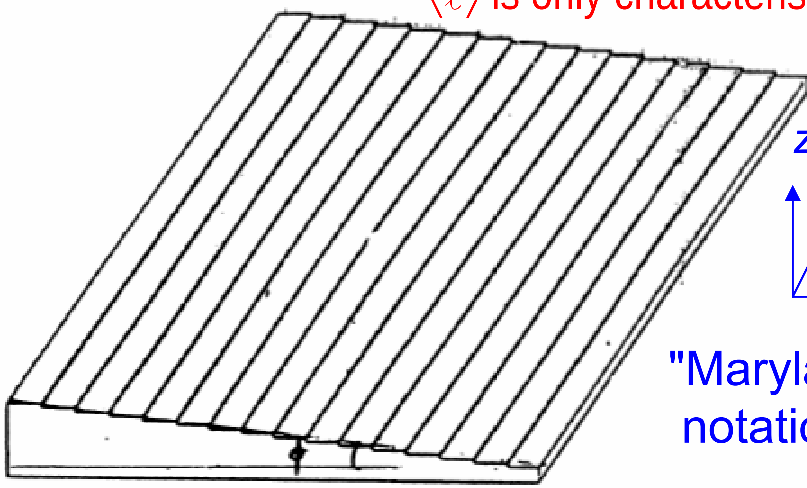


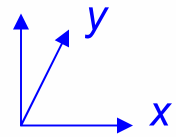
	EC or AD (ADL)	TD (DL)	PD
Limited by	At/de/tach at step	Terrace diffu'n	**Step-edge diffu'n**
Fluctuation healing time--width y	y^2	y^3	y^4
Size dep. of island diffu'n, $R \propto \sqrt{\text{area}}$	R^{-1}	R^{-2}	R^{-3}
$w^2(t)$	$t^{1/2}$	$t^{1/3}$	$t^{1/4}$
Island area decay	t^1	$t^{2/3}$	N/A
Evolution of atom/ vacancy island	Shrink to round point *(Grayson's Thm)*		Wormlike, pinch-off
Height decay of cone ["facet"]	$t^{1/4}$	$t^{1/4}$	N/A
Height decay of paraboloid [rough]	$t^{1/3}$	$t^{2/5}$	N/A

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

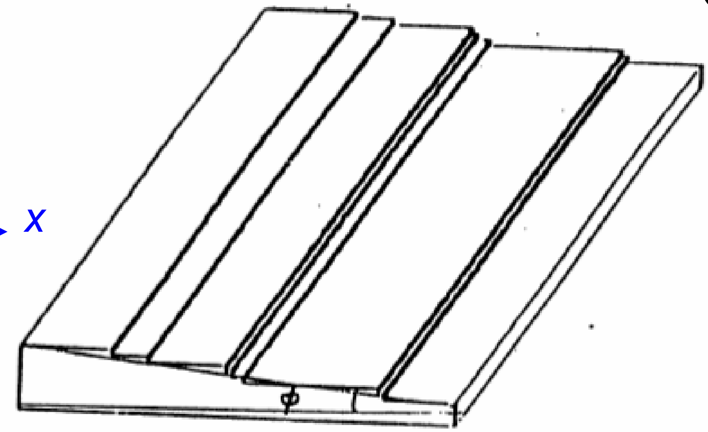




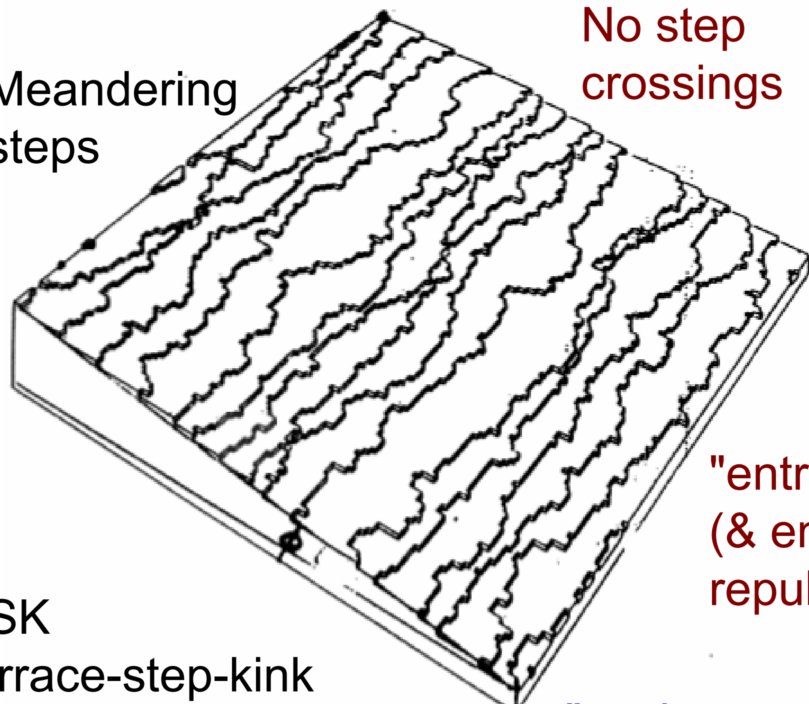
"Maryland notation"

Straight steps, randomly placed

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



Meandering steps



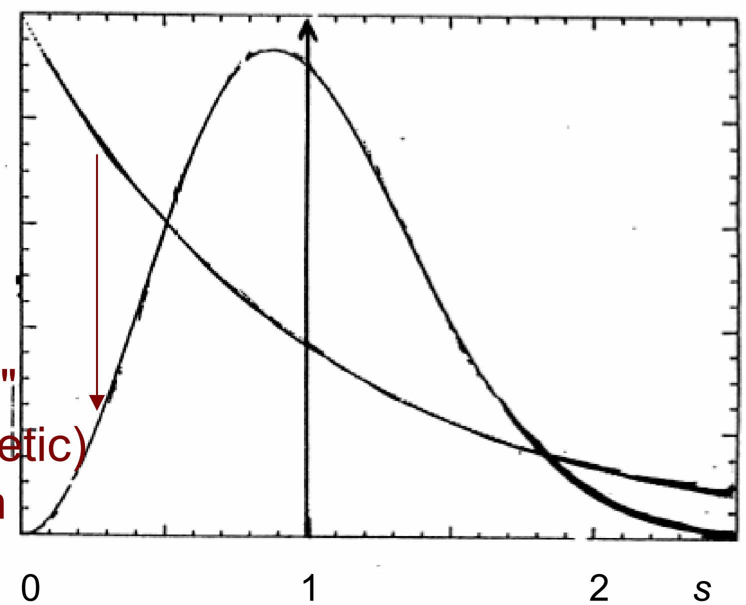
No step crossings

TSK

terrace-step-kink

kink energy ε

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

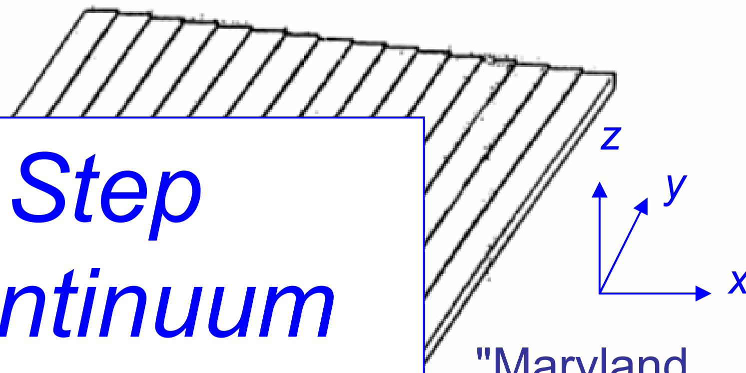


"entropic" (& energetic) repulsion

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

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

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

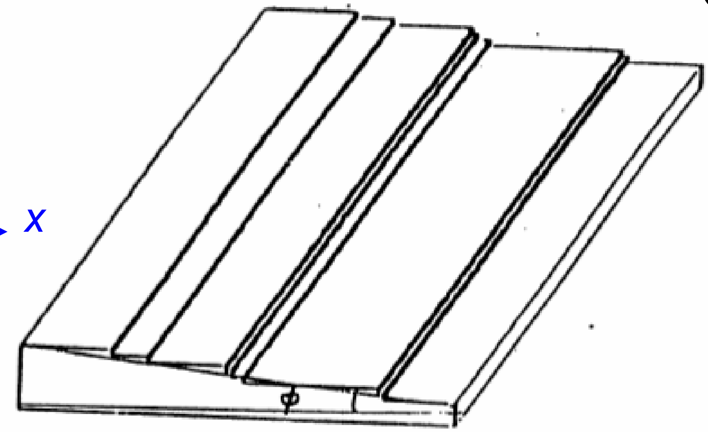


Step Continuum Model

"Maryland notation"

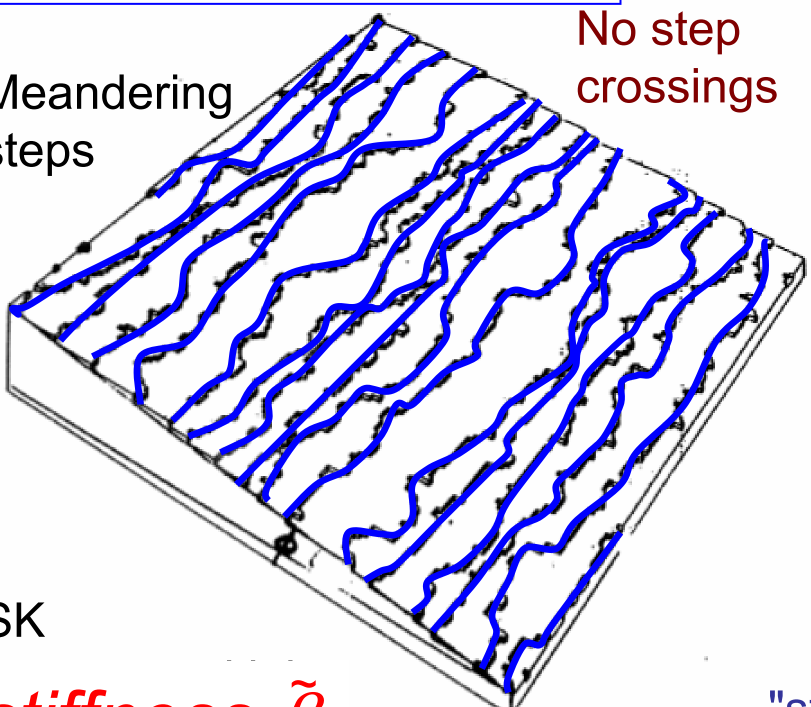
Straight steps, randomly placed

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



Meandering steps

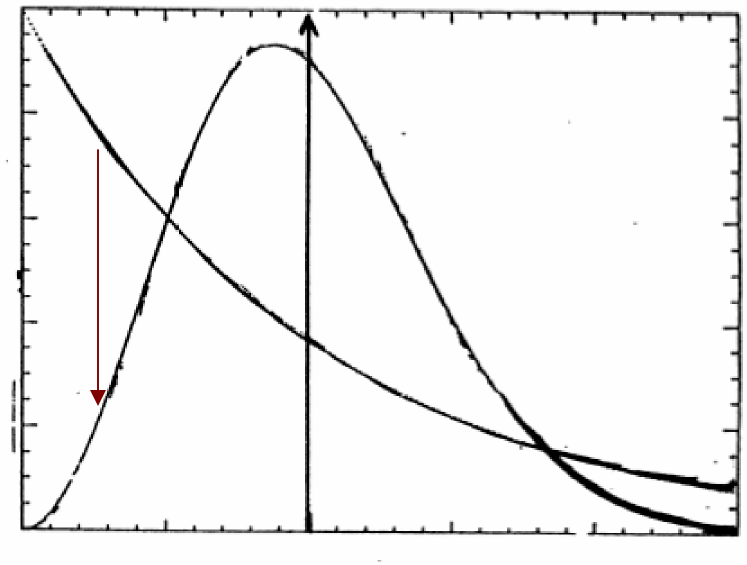
No step crossings



TSK

stiffness $\tilde{\beta}$

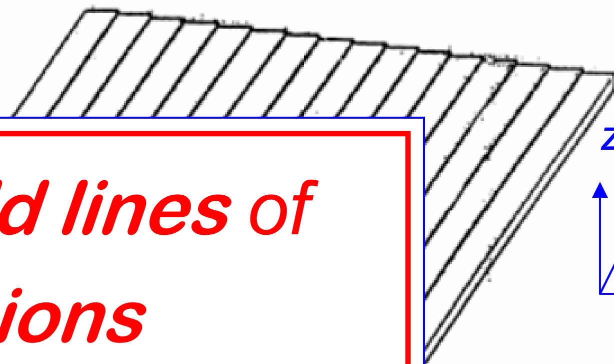
Scaled TWD: $P(s)$ indep. of $\langle \ell \rangle$



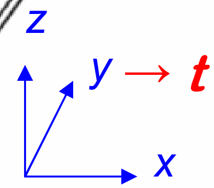
"static correlation" $\langle x_n(y) - x_{n-1}(y) - \langle \ell \rangle \rangle_{y,n}$

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

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

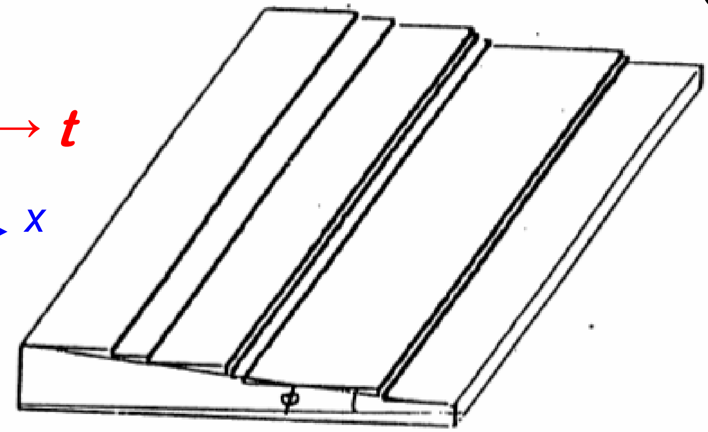


World lines of **fermions** evolving in **1D**



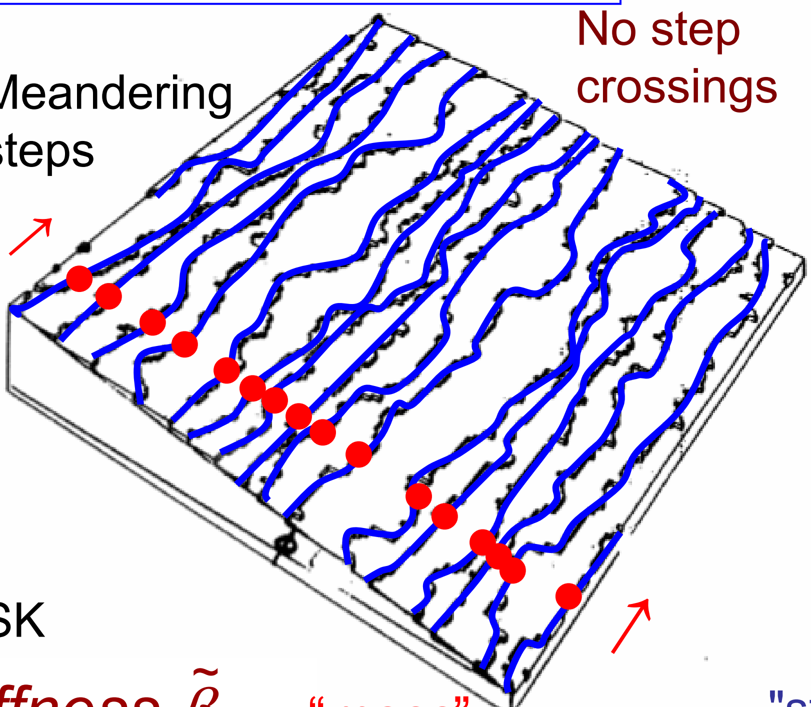
Straight steps, randomly placed

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



Meandering steps

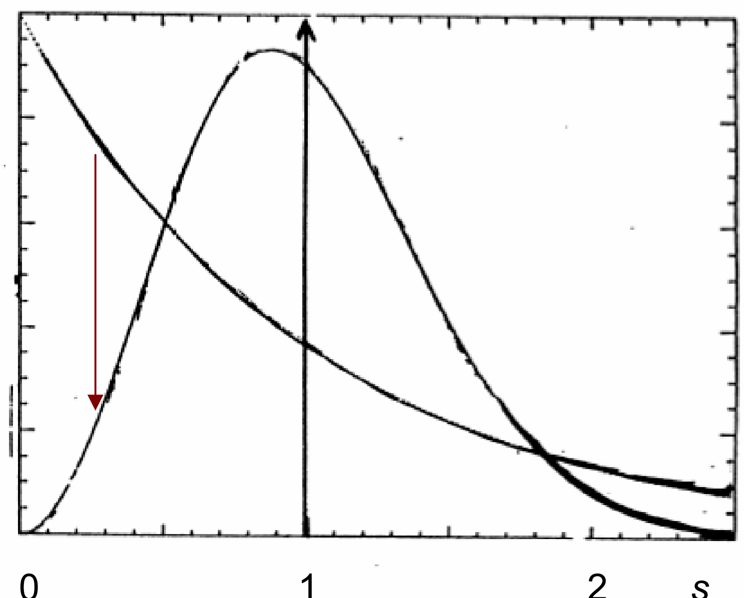
No step crossings



TSK

stiffness $\tilde{\beta} \rightarrow$ "mass"

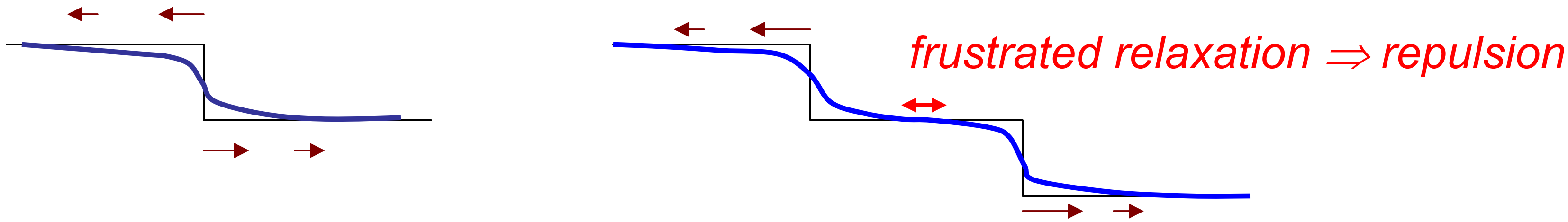
Scaled TWD: $P(s)$ indep. of $\langle \ell \rangle$



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

Origin of elastic (dipolar) step repulsions

- Frustration of relaxation of terrace atoms between steps



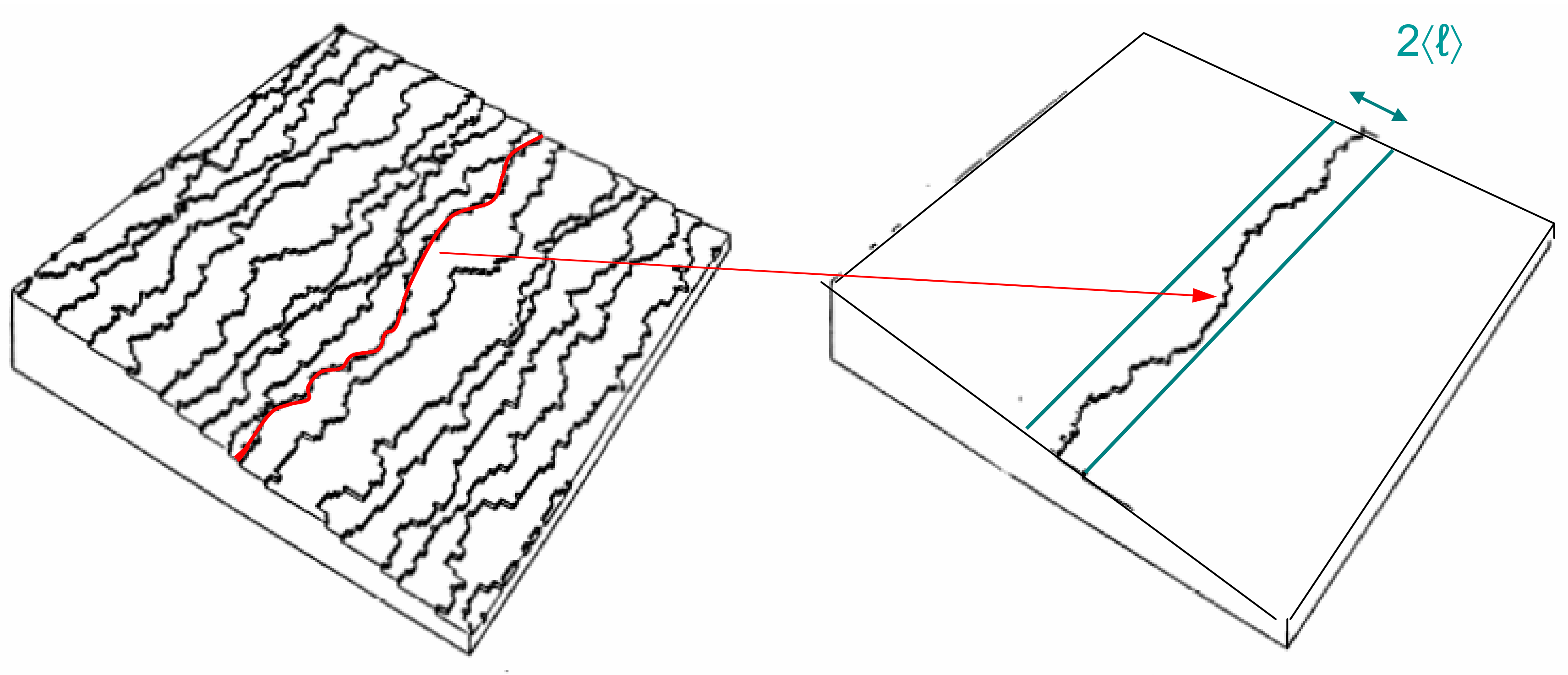
- Energy/length: $U(\ell) = A/\ell^2$ (Same y for points on two interacting steps separated by ℓ along $x \Rightarrow$ "instantaneous")
- Metallic surface states ⇒ additional oscillatory term in U

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. $s \equiv \ell / \langle \ell \rangle$ 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⟨ℓ⟩ apart.

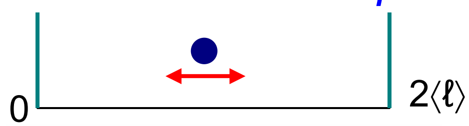
Particle in 1D Box vs. Exact

$$E = \int \beta \sqrt{1+x'^2}\, dy \sim \text{const.} + \int \frac{\tilde{\beta} x'^2}{2}$$

$x'^2 \to \dot{x}^2$

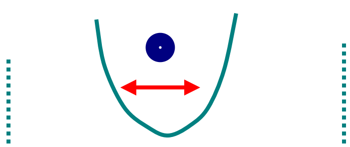
1-D Schrödinger eqn $\quad \frac{\hbar^2}{2m} \to \frac{(k_B T)^2}{2\tilde{\beta}}$

- *Free fermions: repulsion just entropic*



$$\psi_0 = \frac{1}{\langle \ell \rangle} \sin\left(\frac{\pi x}{2\langle \ell \rangle}\right) \qquad E_0 = \frac{(k_B T)^2 \pi^2}{8\tilde{\beta}\langle \ell \rangle^2}$$

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

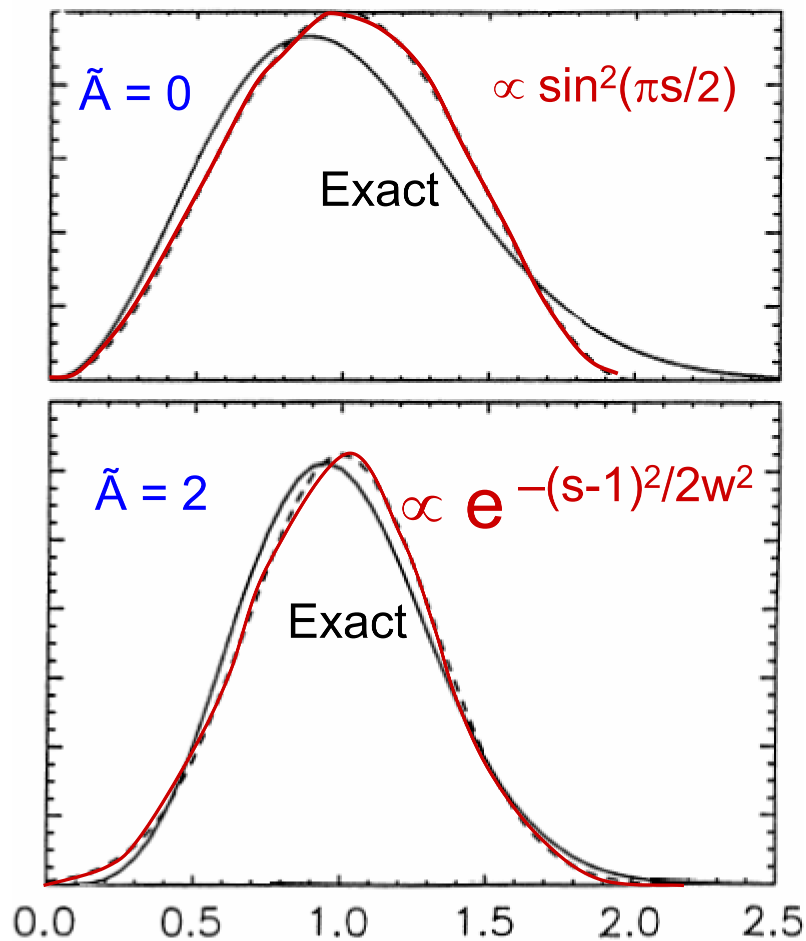


$$\psi_0 \propto e^{-x^2/4w^2} \qquad w^4 = \frac{(k_B T)^2}{8\tilde{\beta} U''(\langle \ell \rangle)}$$

$$w = \text{const.}\ \tilde{A}^{-1/4} \langle \ell \rangle$$

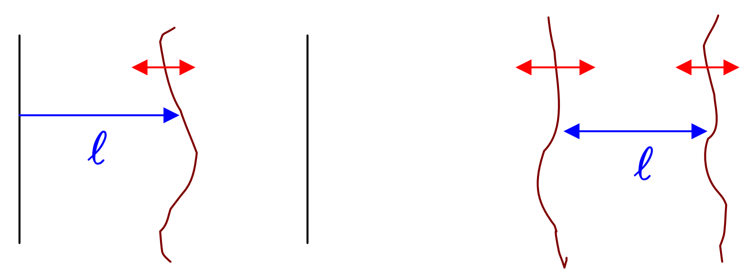
A enters only as Ã: $\quad \tilde{A} \equiv \frac{\tilde{\beta} A}{(k_B T)^2}$

const. *changes with approximation*

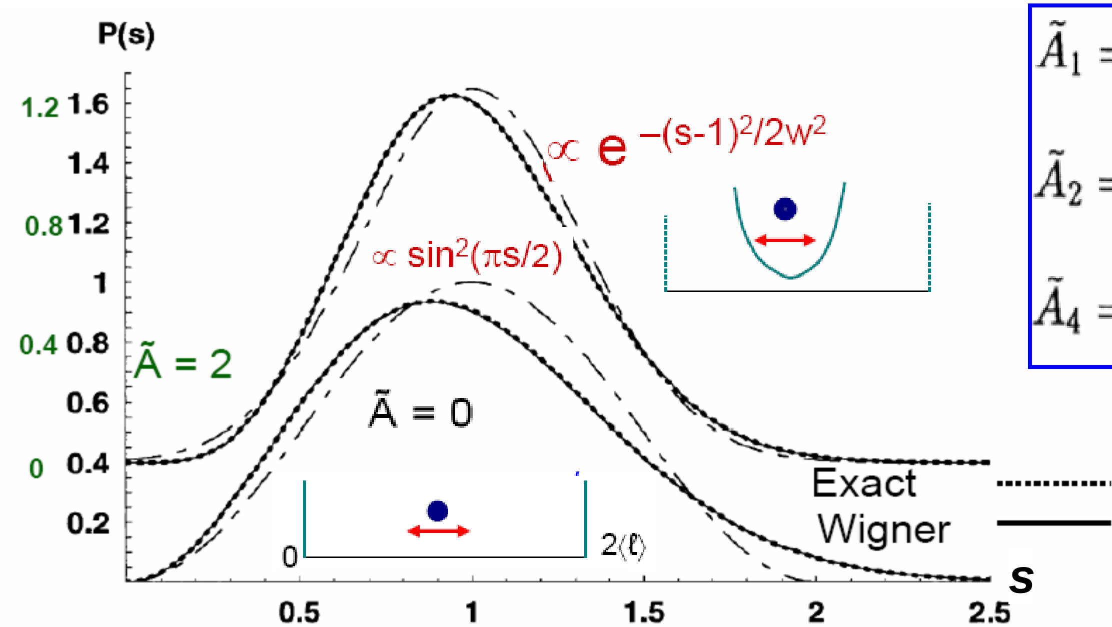


Steps in 2D → fermion worldlines in 1D

- Step non-crossing ⇒fermions or hard bosons
- Energy ∝ path-length × free energy/length β, expand ⇒ 1D Schrödinger eqn., $m\rightarrow$ stiffness β
- Analogous to polymers in 2D (deGennes, JCP '68)
- Only dependence on A via $\tilde{A} \equiv \tilde{\beta} A/(k_B T)^2$
- Mean-field (Gruber-Mullins): 1 active step, $0 \leq s \leq 2$ (← $\ell/\langle \ell \rangle$)
  - $\tilde{A}$ =0: particle in box, $P(s) = \psi_0^2 \propto \sin^2(\pi s/2)$, $\varepsilon_0 \propto T^2/\tilde{\beta}\langle \ell \rangle^2 \rightarrow$ *entropic repulsion*
  - $\tilde{A} \geq 1½$: parabolic well, $P(s) \propto \exp[-(s-1)^2/2w_M^2]$, $w_M \propto \tilde{A}^{-1/4}\langle \ell \rangle$
- $\tilde{A} \rightarrow \infty$: "phonons", variance of $P(s)$ is $2w_M^2$, not w_M^2



Wigner Surmise (WS) for TWD (terrace-width distribution)



$\tilde{A}_1 = -1/4: \quad P_1(s) = \frac{\pi}{2} s \exp\left(-\frac{\pi}{4}s^2\right)$

$\tilde{A}_2 = 0: \quad P_2(s) = \frac{32}{\pi^2} s^2 \exp\left(-\frac{4}{\pi}s^2\right)$

$\tilde{A}_4 = 2: \quad P_4(s) = \left(\frac{64}{9\pi}\right)^3 s^4 \exp\left(-\frac{64}{9\pi}s^2\right)$

$U(\ell) = A/\ell^2$

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

Generalizing from the special cases: **WS → GWS**

- The three special cases correspond to $\varrho = 1$, 2, and 4.
- $\tilde{A}$ and ϱ are related by: $\tilde{A} = (\varrho - 2)\varrho/4$; $\varrho = 1 + \sqrt{1 + 4\tilde{A}}$
- Simplest interpolation expression: $P_\varrho(s) = a_\varrho s^\varrho \exp(-b_\varrho s^2)$
- Two conditions on $P_\varrho(s)$: normalization & unit mean
  ⇒ values of a_ϱ, b_ϱ (in terms of Γ functions).

Physical Ideas Behind Application of Random Matrices

cf. T. Guhr, A. Müller-Groeling, H. A. Weidenmüller, Phys. Reports 299 ('98) 189 [cond-mat/97073]

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;

$H^{(0)}_{mn}\mathbf{I} - i\Sigma_j H^{(j)}_{mn}\sigma_j$ (symplectic)

σ_j: Pauli spin matrices, j=1,2,3; **I**: 2∞2 unit matrix; $H^{()}$ all real, $H^{(0)}$ sym, others asym

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

Gaussian **O**rthogonal **E**nsemble: β=1 **GU**nitary**E**: β=2 **GS**ymplectic**E**: β=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 β=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: Circular**OE**, **CUE**, **CSE;** NN unitary matrices, eigenvalues $\exp[i\theta_\mu]$, μ=1,...N

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

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 β = 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:

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

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

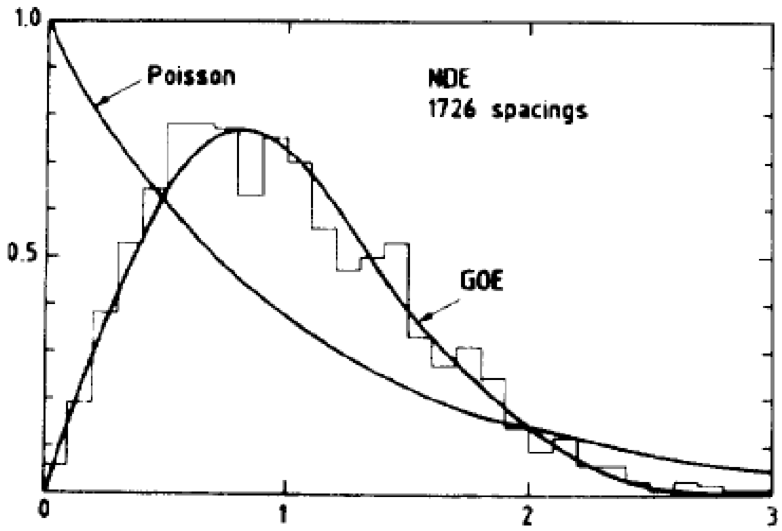
Classical chaos (e.g. Bunimovich stadium, Sinai billiard)

QCD, supersymmetry

2D quantum gravity

Examples of NN spacing distributions with GOE (ϱ =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].



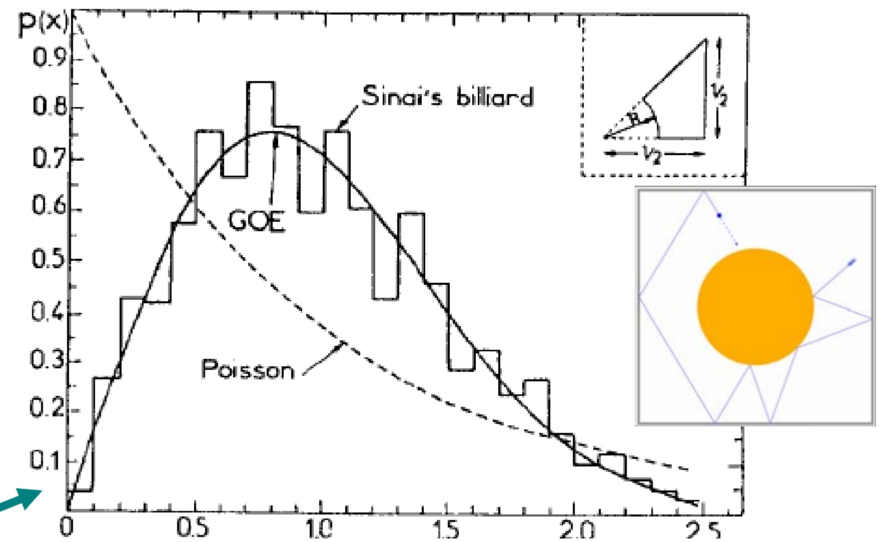


Fig. 4. The nearest-neighbor spacing distribution versus s (defined as in Fig. 1) for the Sinai billiard. The histogram comprises about 1000 consecutive eigenvalues. Taken from Ref. [5].

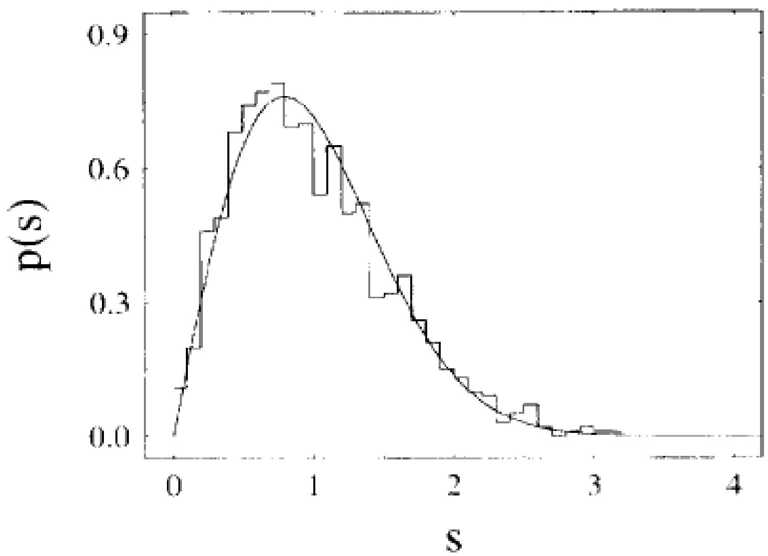
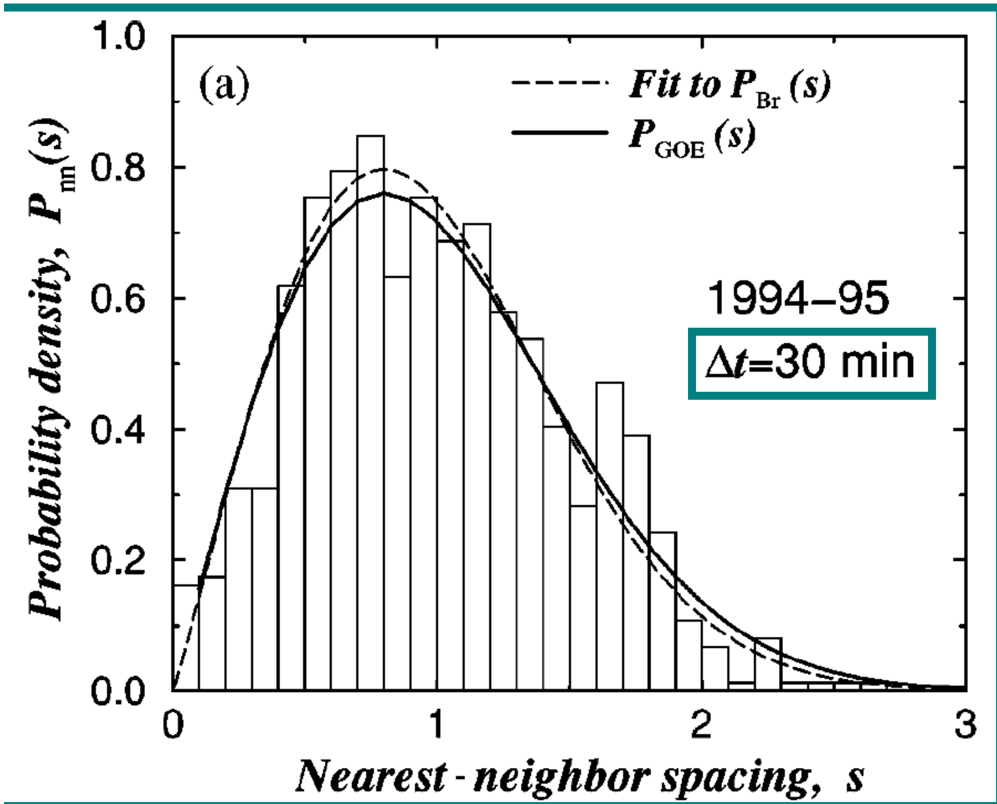


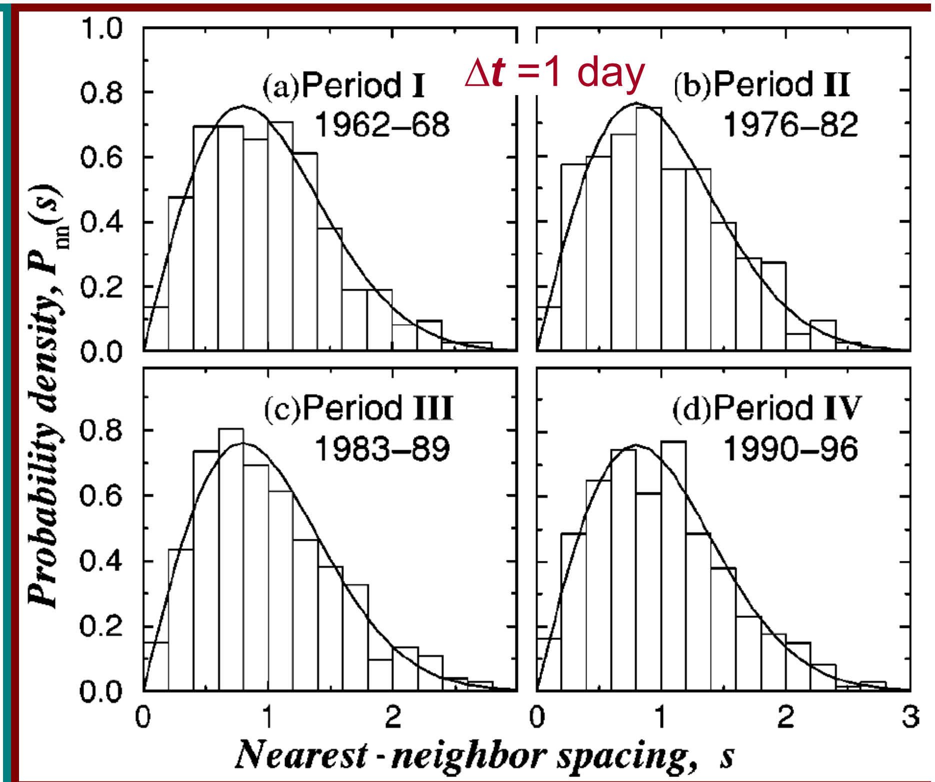
Fig. 6. Nearest-neighbor spacing distribution for elastomechanical modes in an irregularly shaped quartz crystal.

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

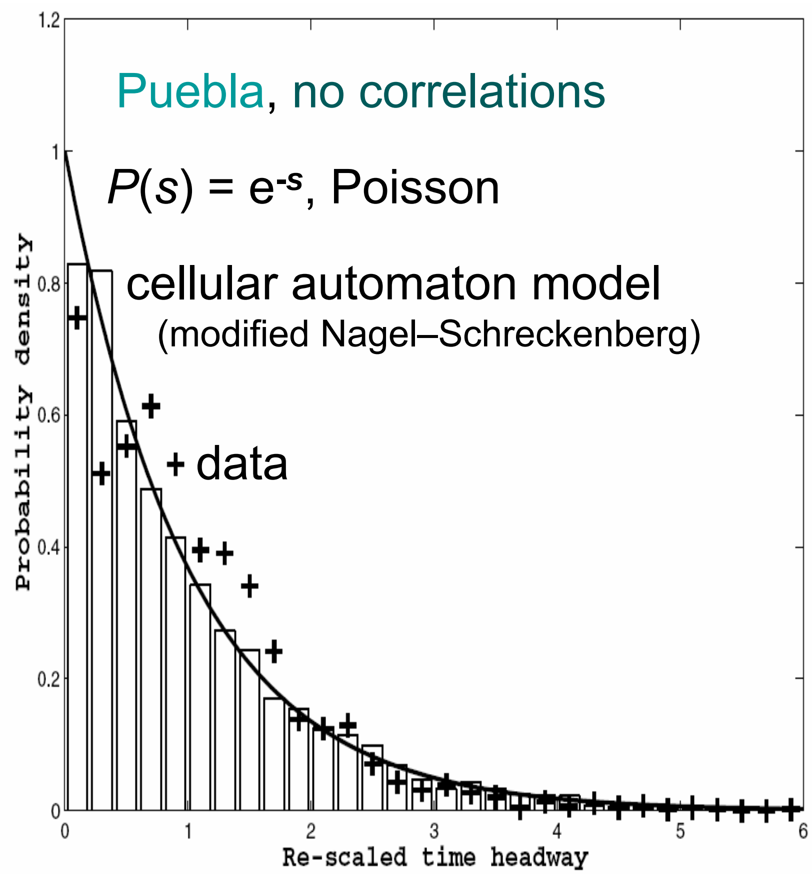


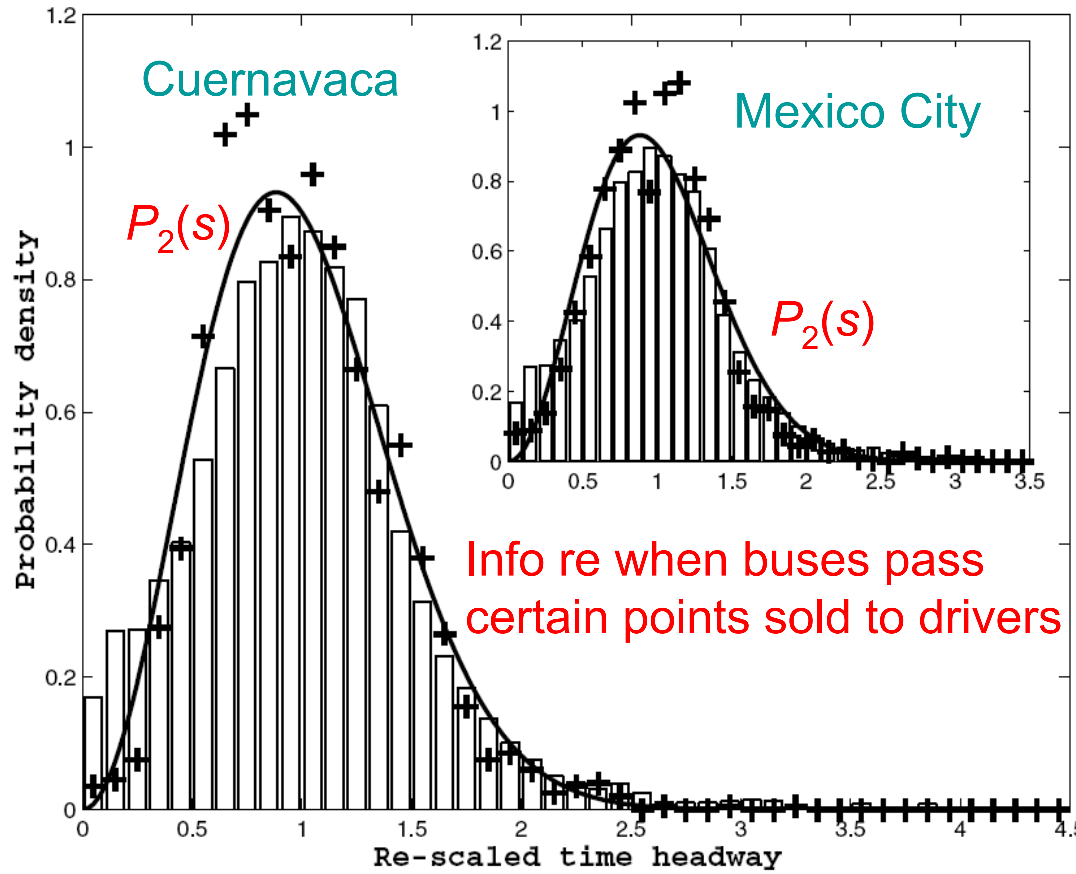


Headway statistics of buses in Mexican cities, using $P_2(s)$

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

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

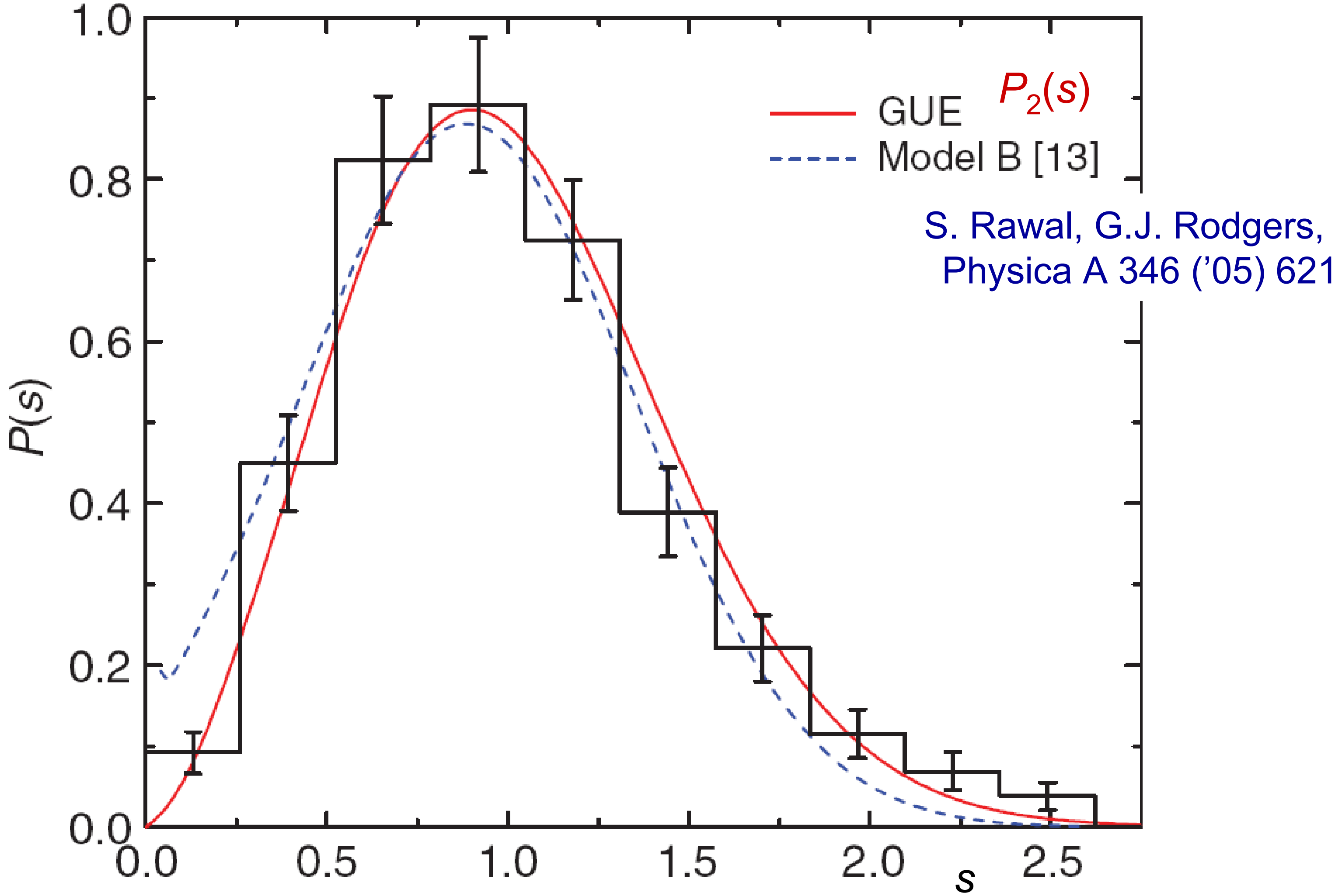




WS $P_2(s)$ better than CA because in CA, correlations only between NNs

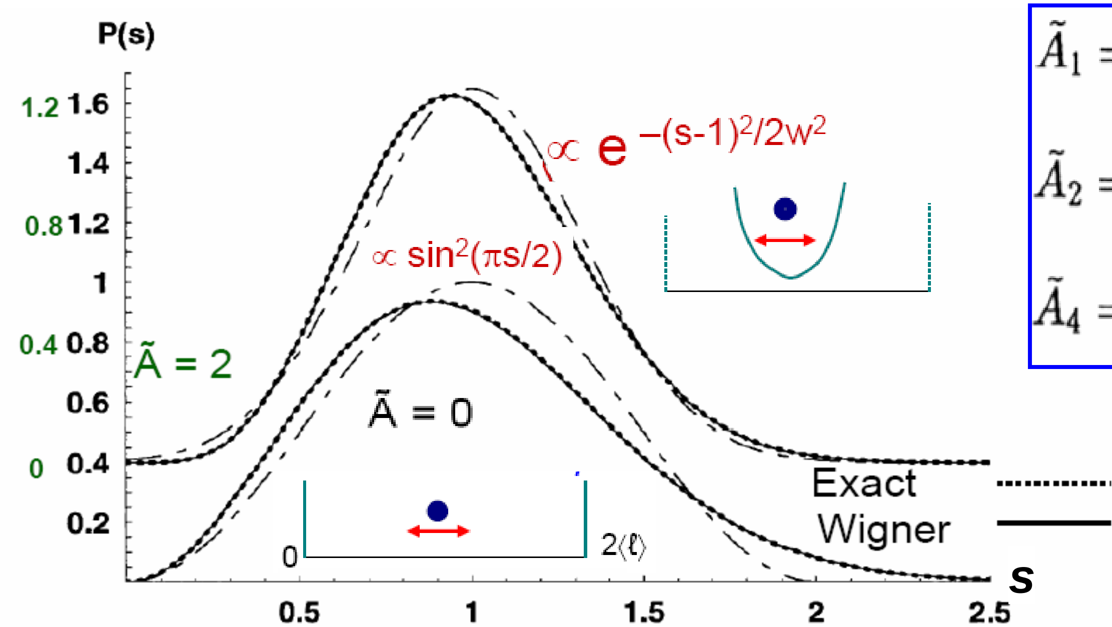
Modelling gap-size distribution of parked cars using RMT

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



Unlike random sequential process, Coulomb gas extends repulsion beyond geometric size.

Wigner Surmise (WS) for TWD (terrace-width distribution)



$\tilde{A}_1 = -1/4: \quad P_1(s) = \frac{\pi}{2} s \exp\left(-\frac{\pi}{4} s^2\right)$

$\tilde{A}_2 = 0: \quad P_2(s) = \frac{32}{\pi^2} s^2 \exp\left(-\frac{4}{\pi} s^2\right)$

$\tilde{A}_4 = 2: \quad P_4(s) = \left(\frac{64}{9\pi}\right)^3 s^4 \exp\left(-\frac{64}{9\pi} s^2\right)$

$U(\ell) = A/\ell^2$

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

Generalizing from the special cases: WS → GWS

- The three special cases correspond to $\varrho = 1, 2$, and 4.
- $\tilde{A}$ and ϱ are related by: $\tilde{A} = (\varrho - 2)\varrho/4$; $\varrho = 1 + \sqrt{1 + 4\tilde{A}}$
- 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_ϱ, b_ϱ (in terms of Γ functions).

Comparison of variance of $P(s)$ vs. Ã computed with Monte Carlo:

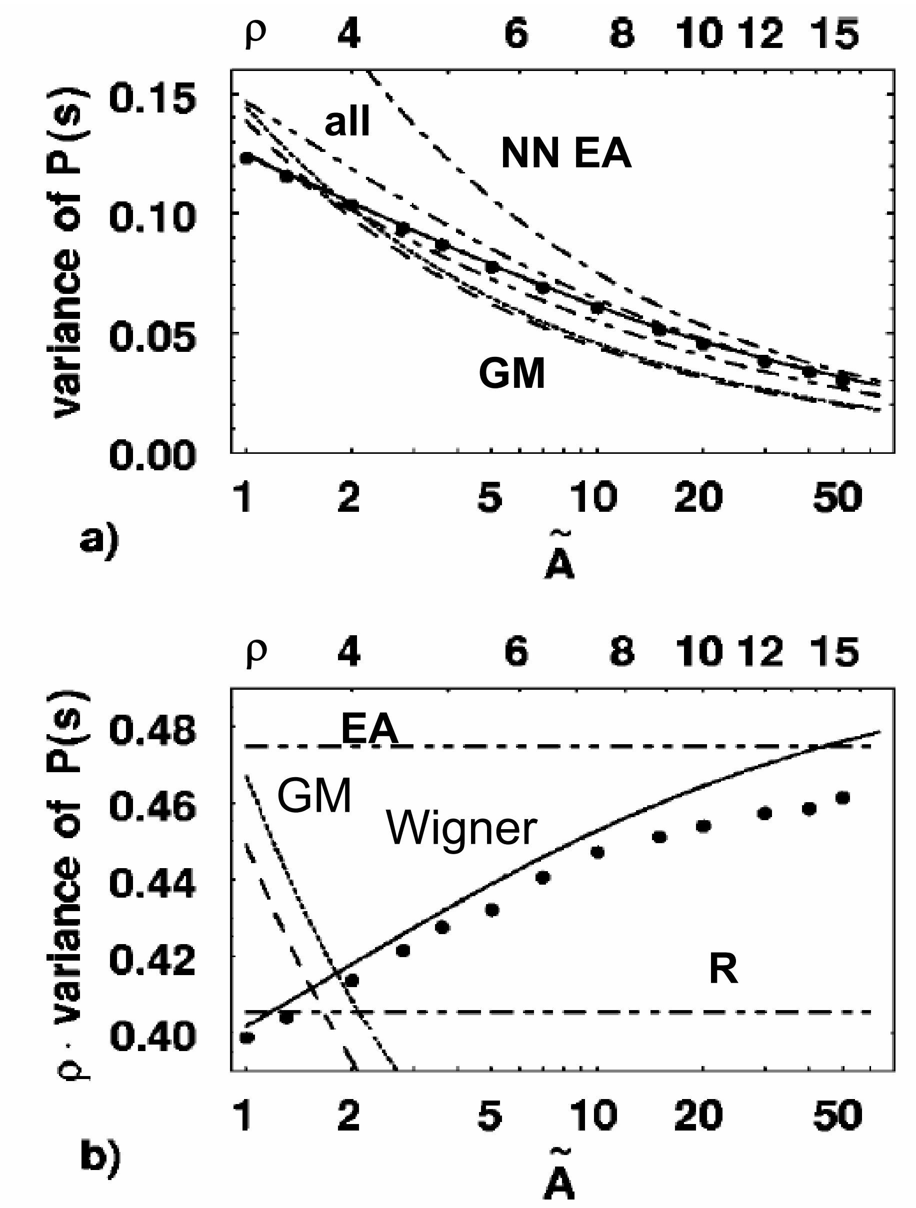
GWS does better, quantitatively & conceptually, than any other approximation

Hailu Gebremariam et al., Phys. Rev. B 69 ('04)125404

Experiments measuring variances of TWDs

Vicinal	T (K)	σ^2	ϱ	$\tilde{A}$	A_W/A_G	A_W (eV Å)	Experimenters
Pt(1 1 0)-(1 × 2)	298		2.2	0.13	–	$\tilde{\beta}=?$	Swamy, Bertel [36]
Cu(19, 17, 17)	353	0.122	4.1	2.2	0.77	0.005	Geisen [5,54]
Si(1 1 1)	1173	0.11	3.8	1.7	0.96	0.4	Bermond, Métois [55]
Cu(1,1,13)	348	0.091	4.8	3.0	1.27	0.007	Giesen [5,56]
Cu(1 1,7,7)	306	0.085	5.1	4	1.37	0.004	Geisen [5,54]
Cu(1 1 1)	313	0.084	5.0	3.6	1.39	0.004	Geisen [5,54]
Cu(1 1 1)	301	0.073	6.0	6.0	1.58	0.006	Geisen [5,54]
Ag(1 0 0)	300	0.073	6.4	6.9	1.58	$\tilde{\beta}=?$	P. Wang...Williams
Cu(1, 1, 19)	320	0.070	6.7	7.9	1.64	0.012	Geisen [5,56]
Si(1 1 1)-(7 × 7)	1100	0.068	6.4	7.0	1.67	0.7	Williams [57]
Si(1 1 1)-(1 × 1)Br	853	0.068	6.4	7.0	1.67	0.1	X.-S. Wang, Williams [58]
Si(1 1 1)-Ga	823	0.068	6.6	7.6	1.67	1.8	Fujita...Ichikawa [59]
Si(1 1 1)-Al $\sqrt{3}$	1040	0.058	7.6	10.5	1.85	2.2	Schwennicke...Williams [60]
Cu(1, 1, 11)	300	0.053	8.7	15	1.95	0.02	Barbier et al. [21]
Cu(1, 1, 13)	285	0.044	10	20	2.12	0.02	Geisen [5,56]
Pt(1 1 1)	900	0.020	24	135	2.59	6	Hahn...Kern [61]
Si(1 1 3) rotated	1200	0.004	124	3.8×10^3	2.92	$(27\pm5)\times10^2$	van Dijken, Zandvliet, Poelsema [9]

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 ρ^{-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_{eff} of Richards et al.) since no symmetry basis for $\varrho \neq 1, 2,$ 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 Ã (and so ϱ) 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 ∝ 1/(separation) & parabolic well

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

- Assume steps beyond nearest neighbors are at integer times mean spacing (cf. Gruber-Mullins)

  $\ddot{s} = -\kappa s + \rho/s$ + noise

  $\tilde{t} \equiv t\Gamma / \langle \ell \rangle^2$ 1/τ

  Noise sets time scale.

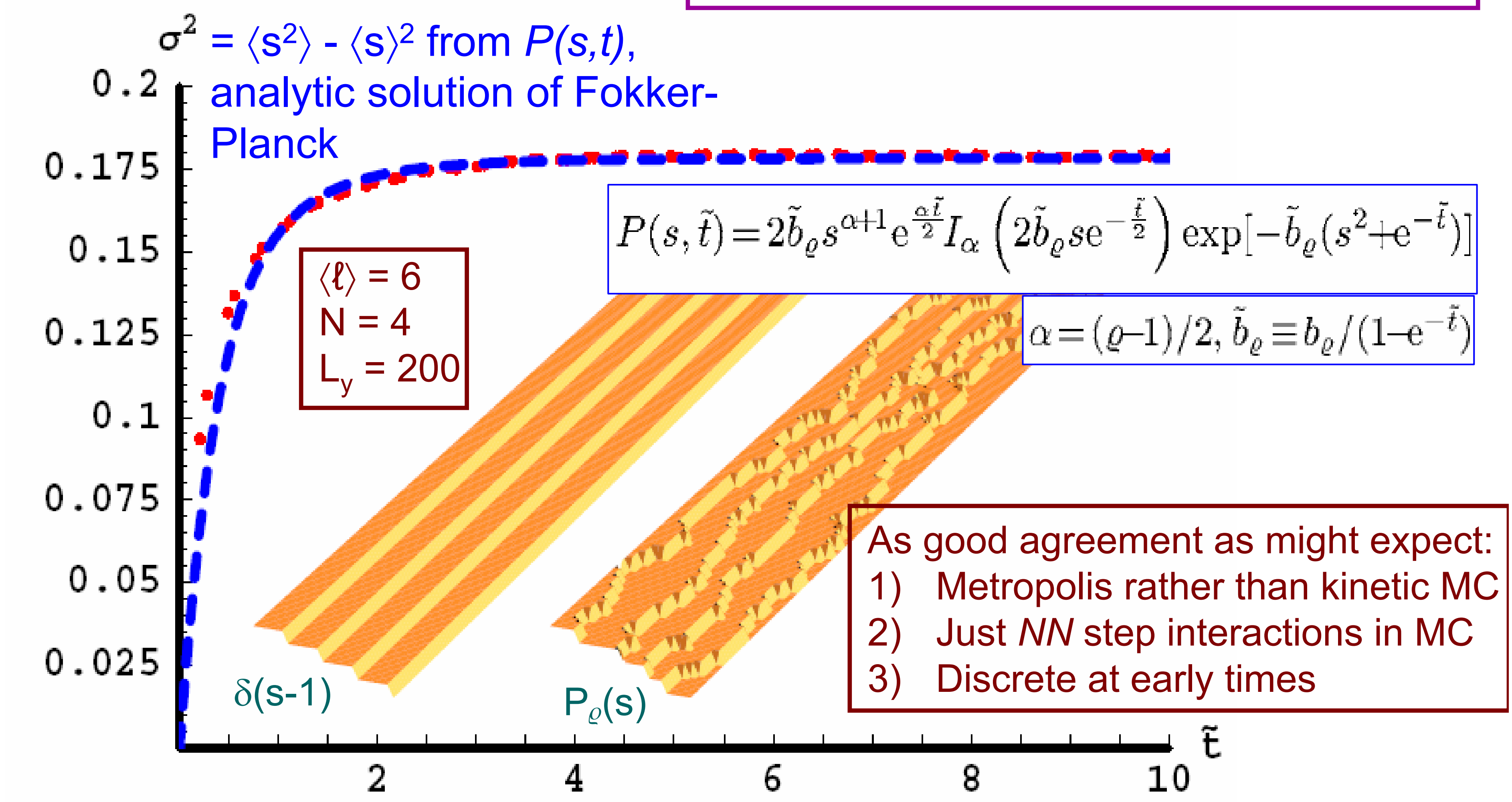
- Demand self-consistency for width of parabolic confining well: $\kappa \rightarrow 2b_\rho$

$$\frac{\partial P(s,\tilde{t})}{\partial \tilde{t}} = 0 = \frac{\partial}{\partial s}\left[\left(2b_\varrho s - \frac{\varrho}{s}\right) P(s,\tilde{t})\right] + \frac{\partial^2}{\partial s^2}[P(s,\tilde{t})] \rightarrow P_\varrho(s)$$

Check of Fokker-Planck with Monte Carlo

cleaved → equilibrium TSK model (no adatom carriers)

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



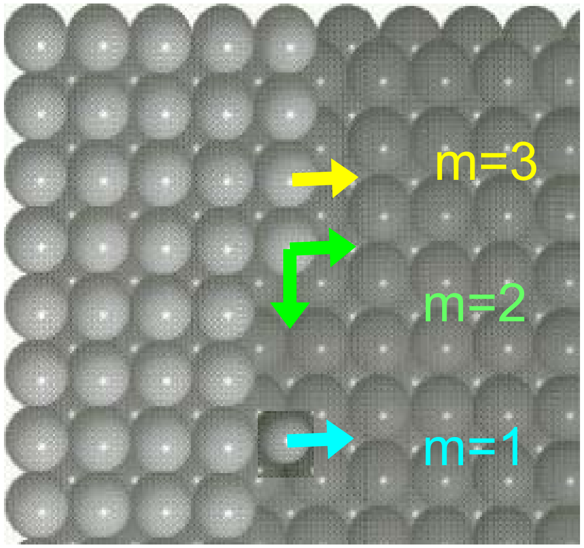
$$P(s,\tilde{t}) = 2\tilde{b}_\varrho s^{\alpha+1} e^{\frac{\alpha\tilde{t}}{2}} I_\alpha\left(2\tilde{b}_\varrho s e^{-\frac{\tilde{t}}{2}}\right) \exp[-\tilde{b}_\varrho(s^2 + e^{-\tilde{t}})]$$

$$\alpha = (\varrho - 1)/2,\ \tilde{b}_\varrho \equiv b_\varrho/(1 - e^{-\tilde{t}})$$

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

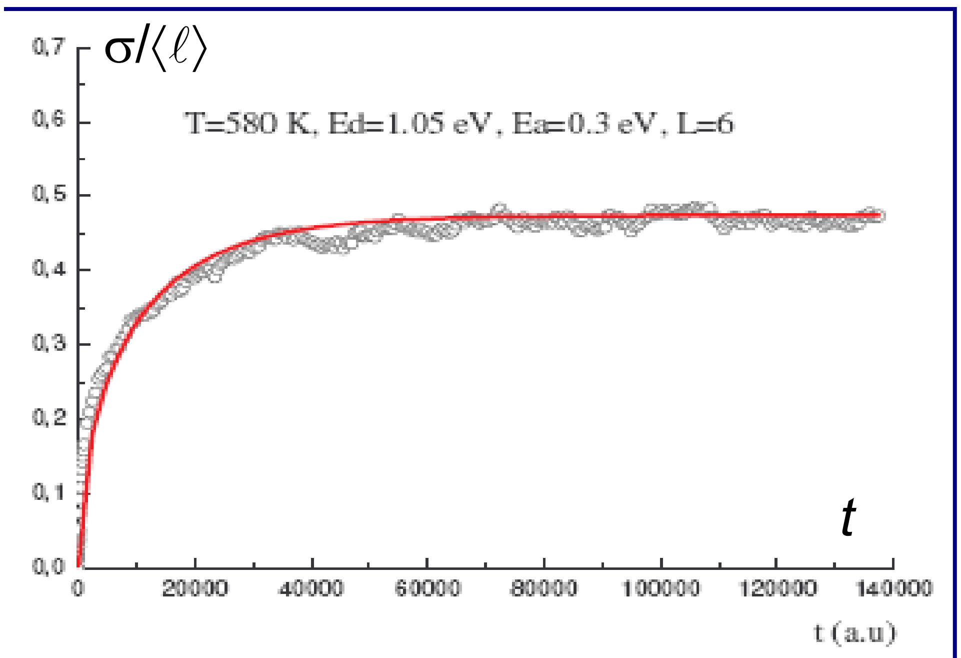


$E_{barrier} = E_d + m\,E_a$ breaking m bonds

E_d = 0.9 -1.1 eV; E_a = 0.3 - 0.4 eV

T = 520 - 580 K

$\langle \ell \rangle$ = 4-15, 5 steps, 10000 x L_x



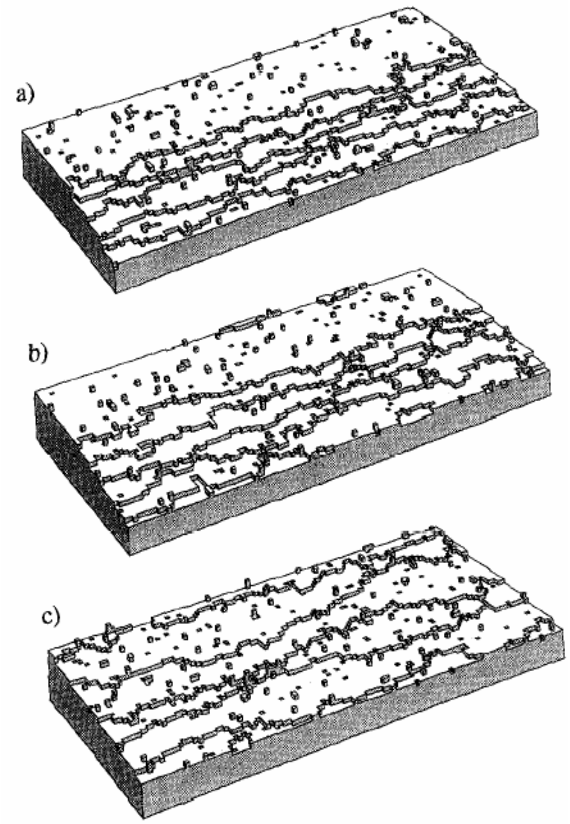
Fit:

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

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

Find $E_{barrier} \approx 1\,E_d + 3\,E_a$

2 other situations of interest



Step Bunch: initially a delta function

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

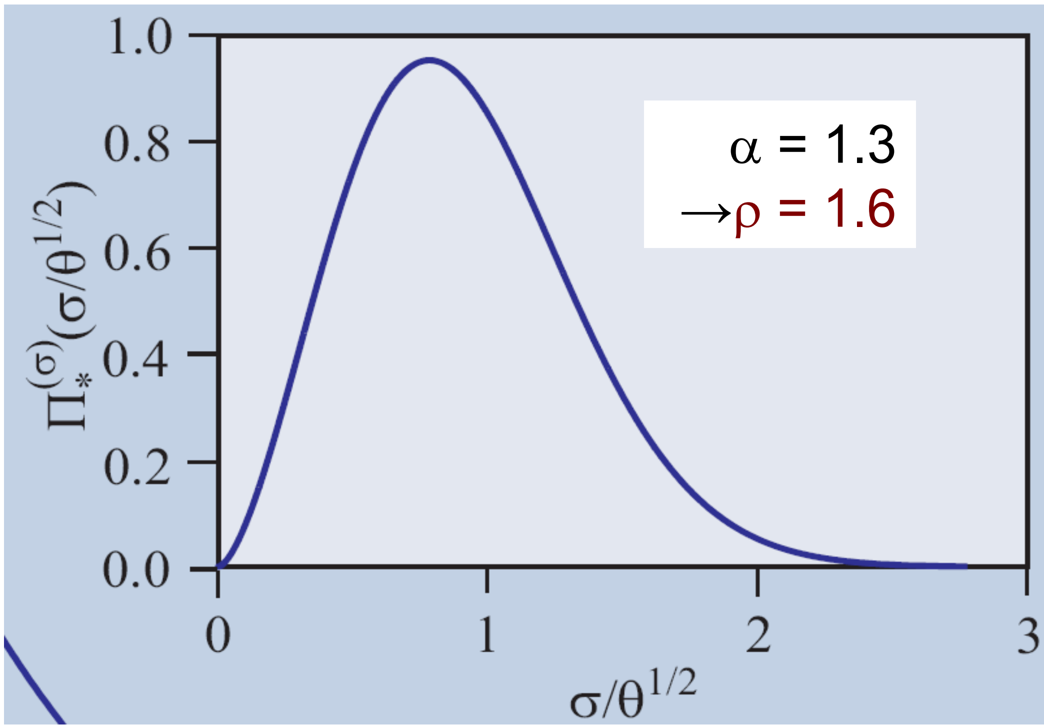
Quench or upquench: change from initial ρ_0 to ρ, e.g. change in temperature

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

Final

Analogies in Econophysics

A Dragulescu & V M Yakovenko: "Probability distribution of returns in the Heston model with stochastic volatility", Quant. Finance 2, 443 ('02)



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

Consider a stock whose price S_t obeys the stochastic differential equation of Brownian motion.

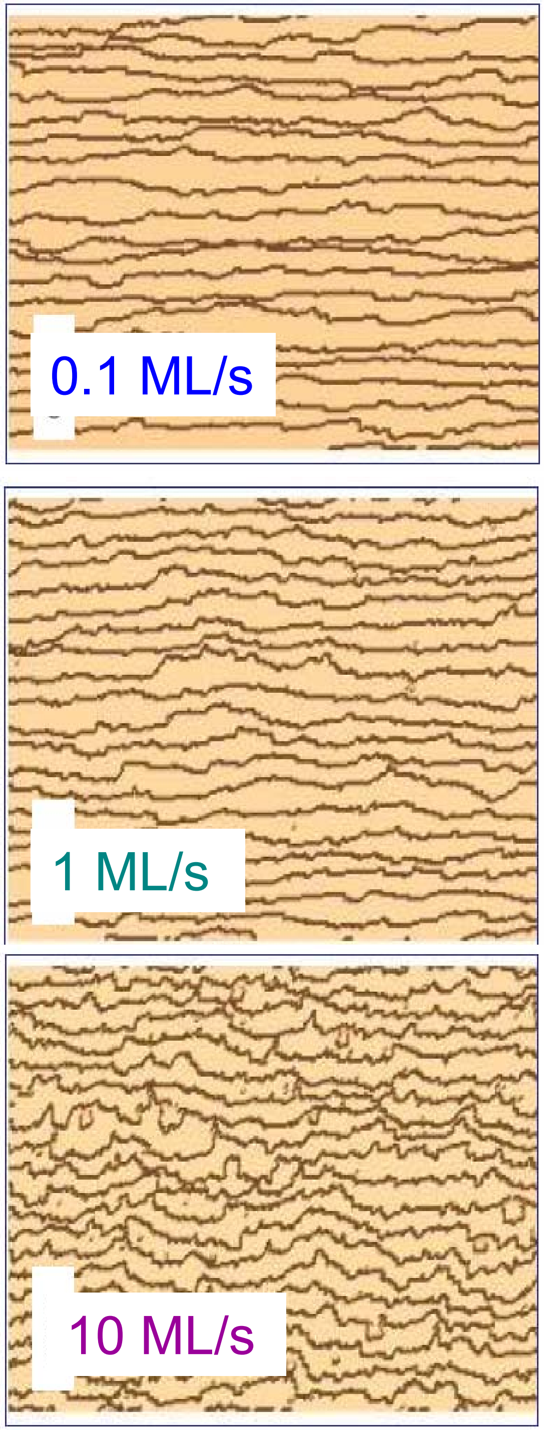
Volatility → stochastic variance obeys a mean-reverting stochastic DE.

The stationary PDF of volatility σ is

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

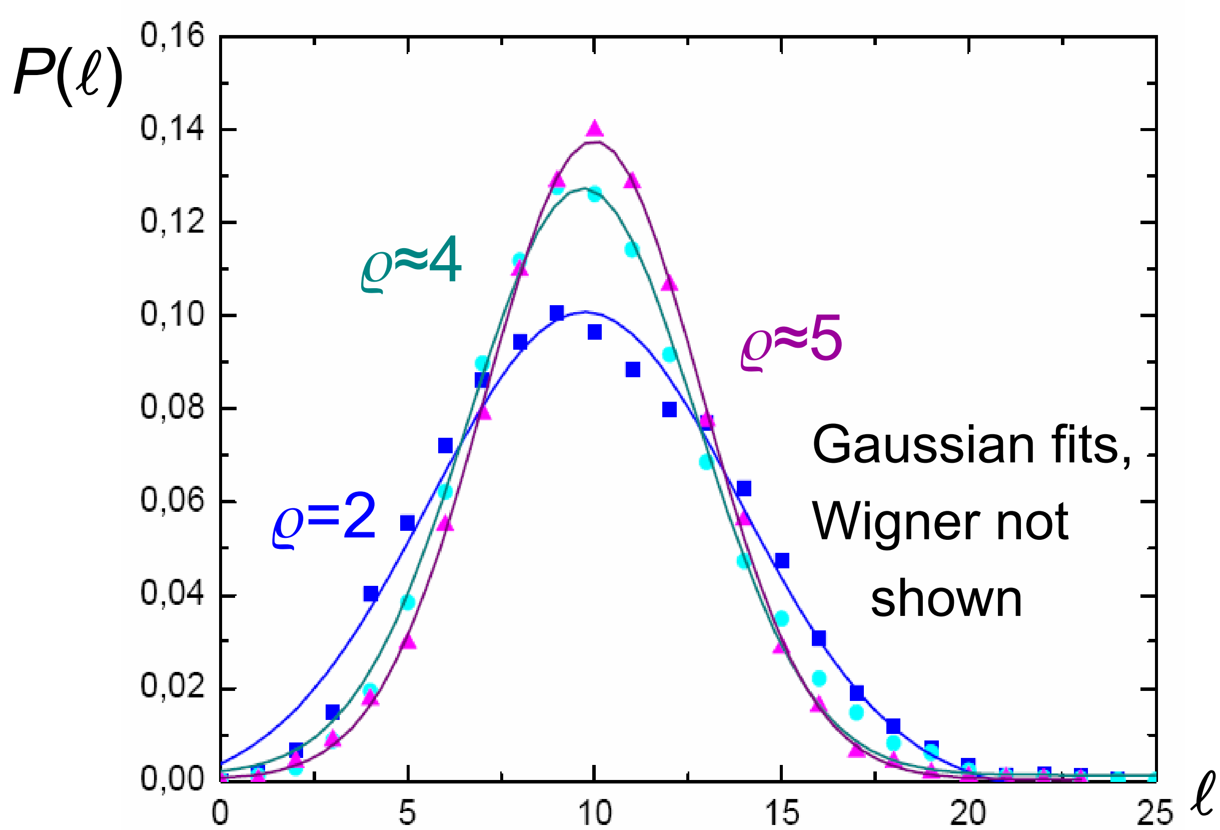
$2\alpha\text{-}1 \rightarrow \varrho$

$\theta \rightarrow \langle \ell^2 \rangle$



Does growth flux (step motion) alter TWD?

Test: *no* energetic interaction (ϱ=2), 150 ML

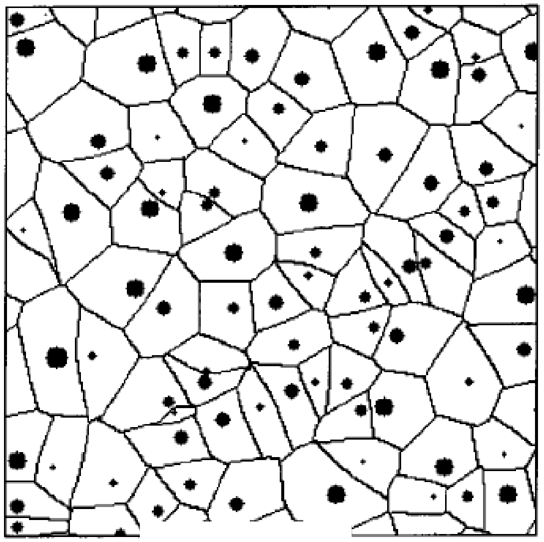


- Narrower ⇒ *effective* repulsion that rises with flux, higher ϱ, more Gaussian-like
- Decreased apparent stiffness $\tilde{\beta}$

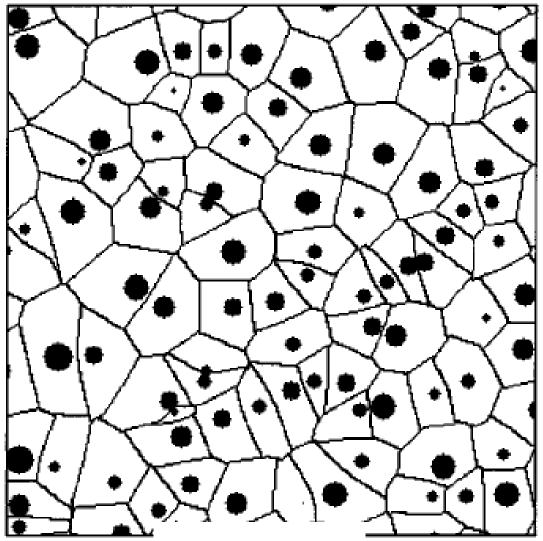
20 steps, 1000x200, T=723K, E_d=1.0eV, E_a=0.3eV

Evolution of Island Structures: Simulations of i=1

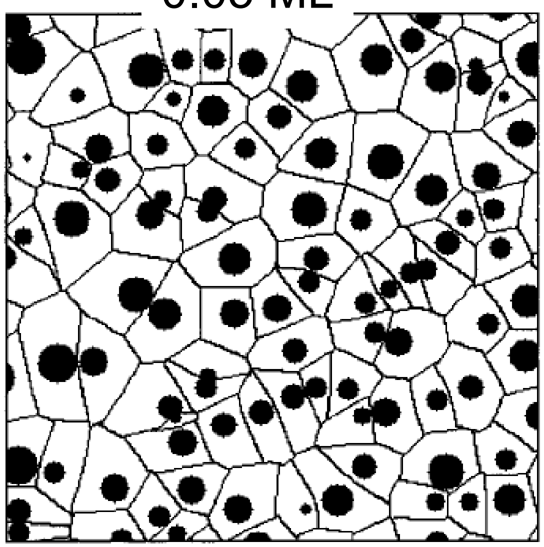
Circular Islands Mulheran & Blackman, PRB 53 (96) 10261



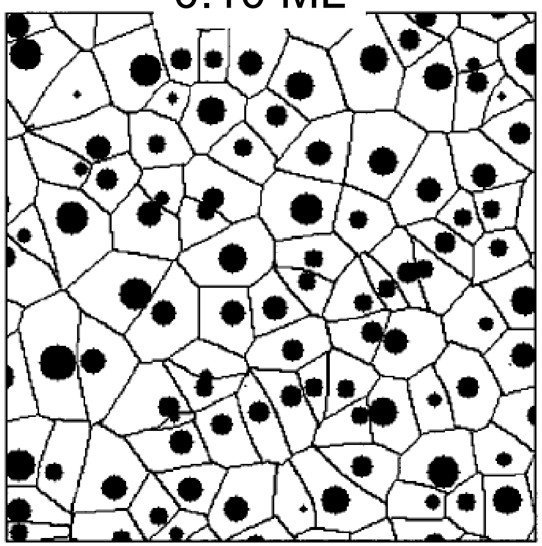
0.05 ML



0.10 ML

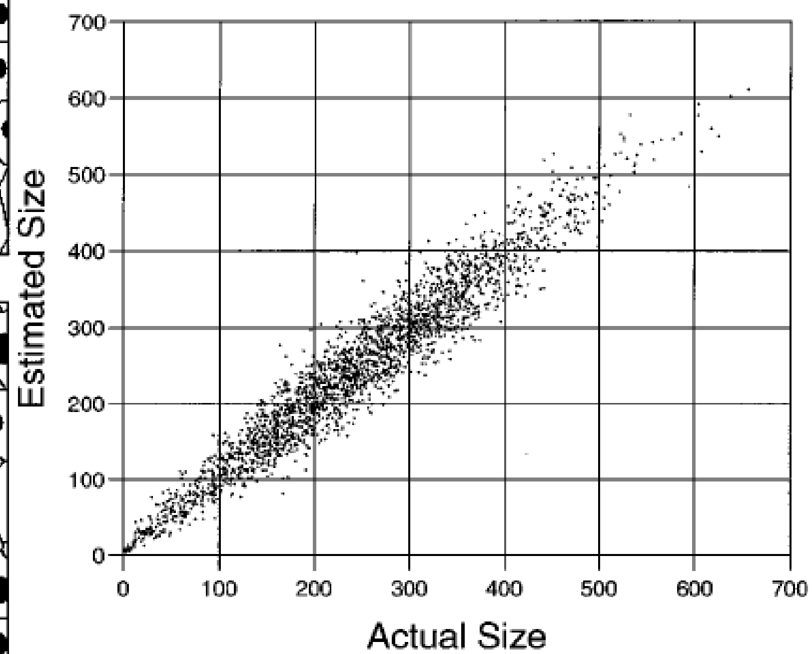


0.15 ML



0.20 ML

Estimated size of island based on Voronoi polygon CZ $\propto$ actual size of island



Island Size Scaling, stable config *i*

Amar & Family, PRL 74 (95) 2066

Dynamic scaling assumption

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

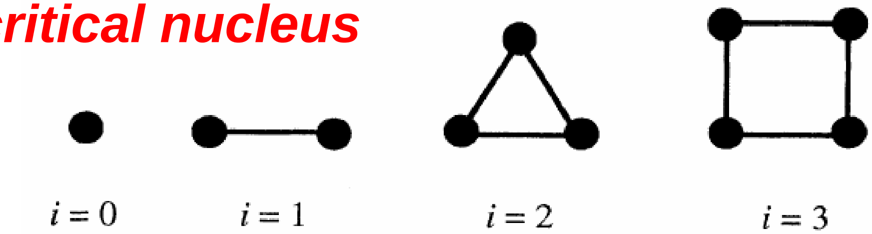
Bartelt & Evans

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

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

***i*+1 atoms: smallest stable island**

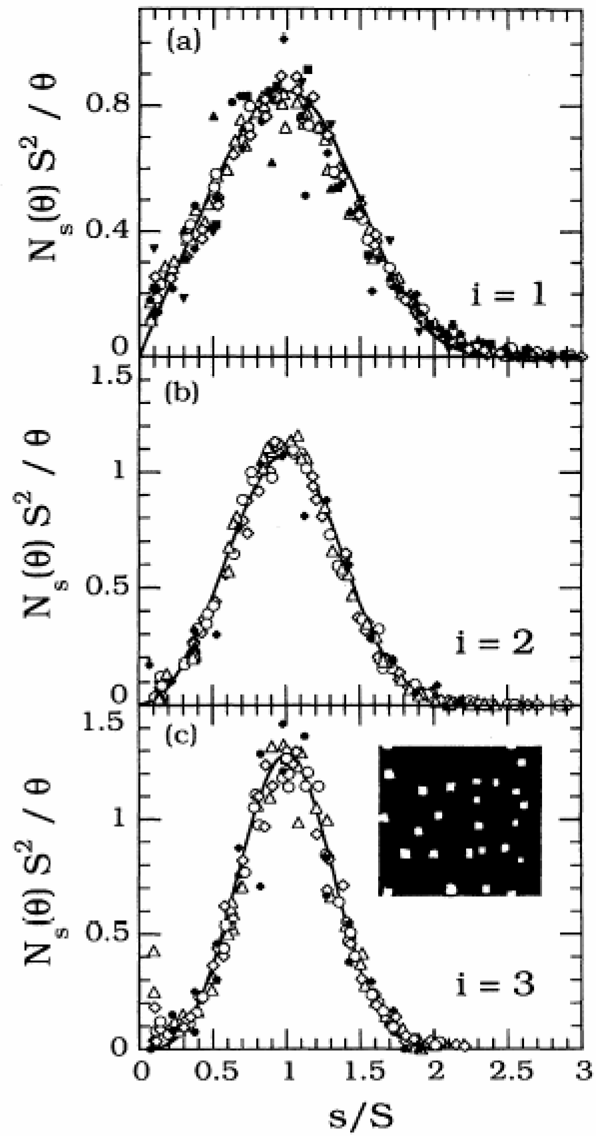
critical nucleus



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

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

$$f_i(u) = 0; \quad u > \frac{i+2}{i+1}$$



Scaling During Growth in 1D: Going Beyond Mean-Field Rate Eqns.

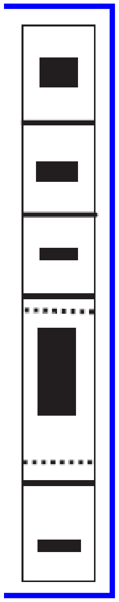
Blackman & Mulheran, PRB 54 (96) 11681

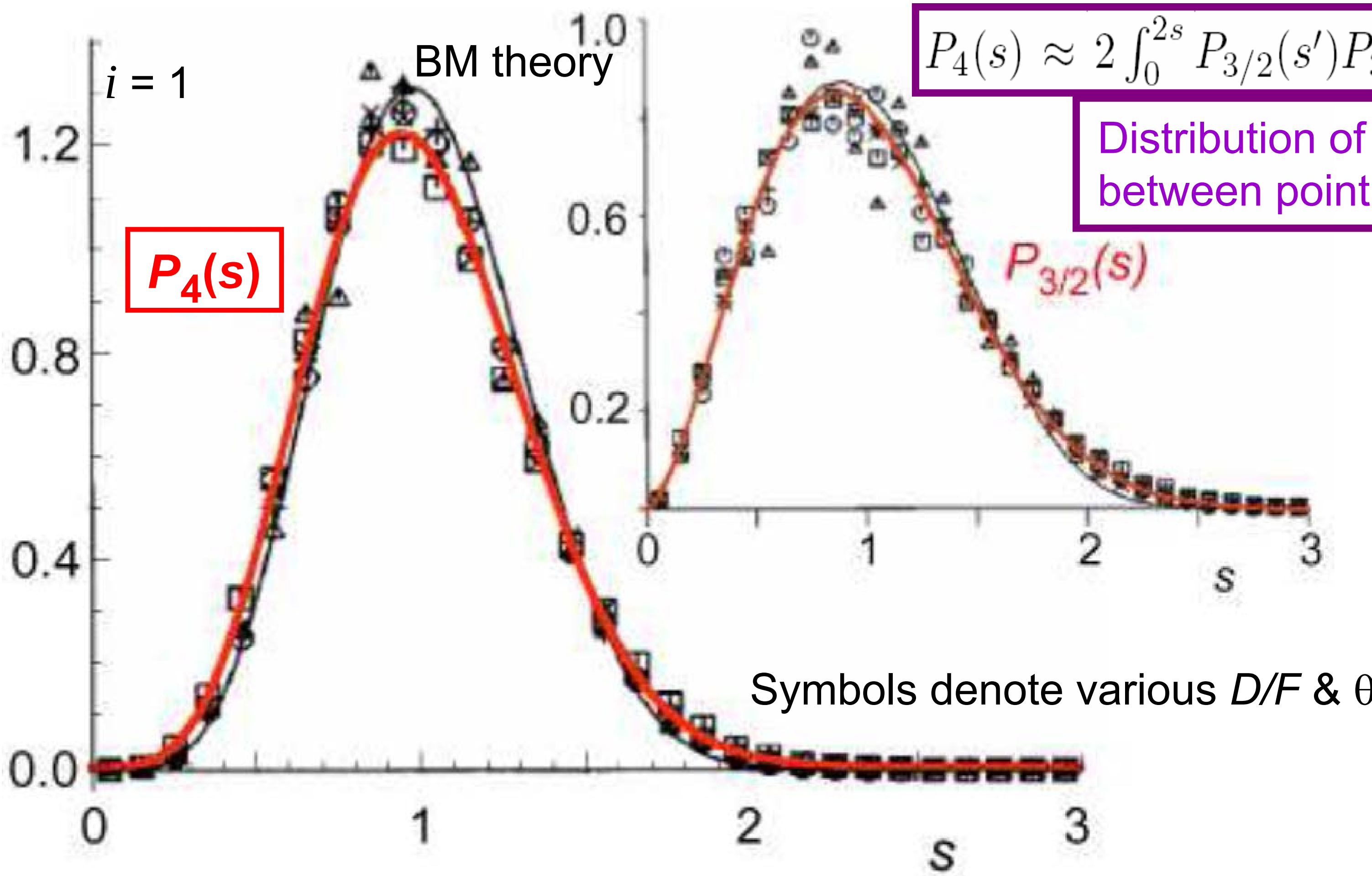
$P_4(s)$ fits numerical data at least as well as B&M's complicated theory expression (not expressible succinctly)

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

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

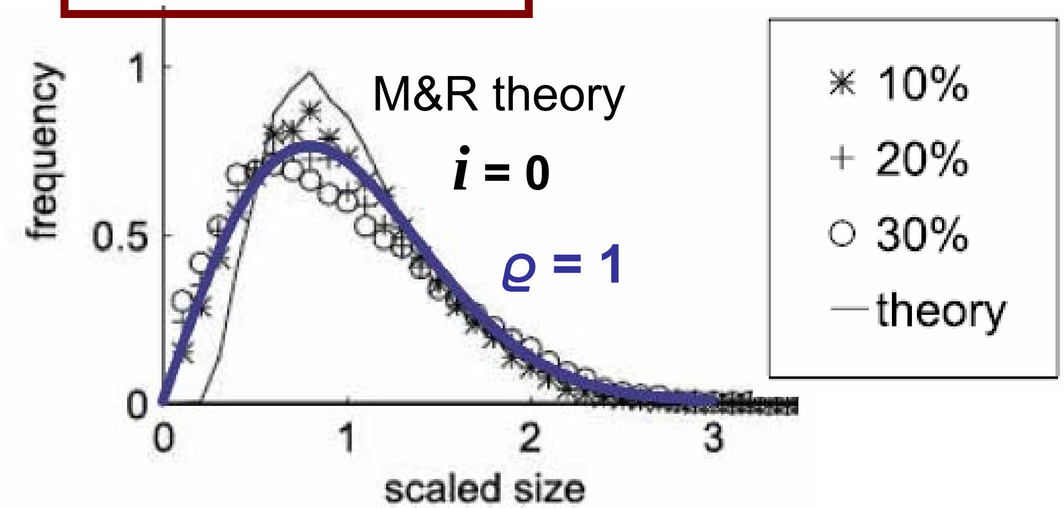
Distribution of gaps between point islands



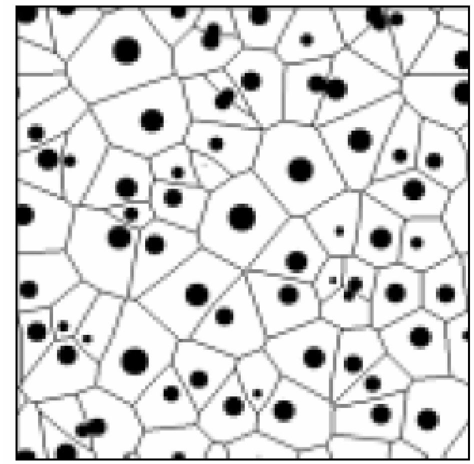


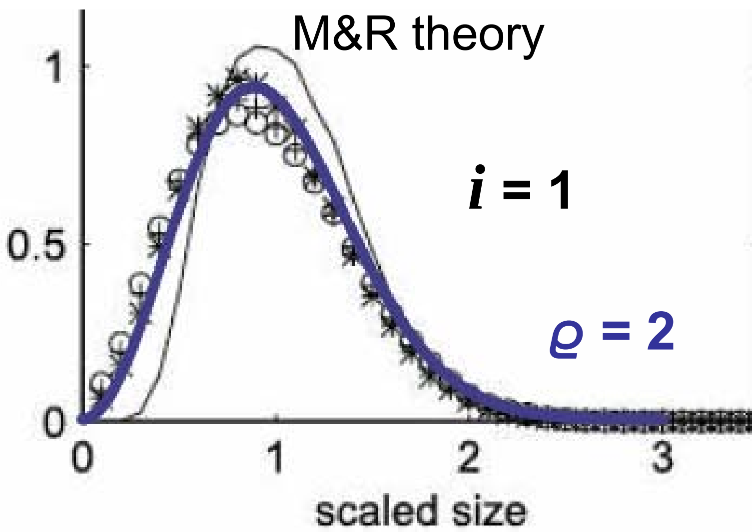
Theory of CZ size distributions in growth, *Mulheran & Robbie*, EPL 49(00)617

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

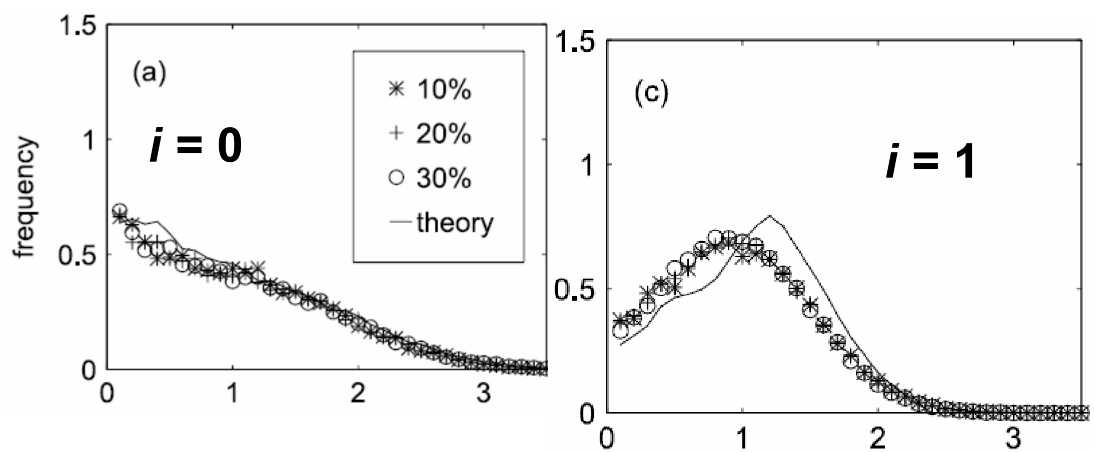


Wigner distribution $P_{\varrho}(s)$ fits much better than M&R theory



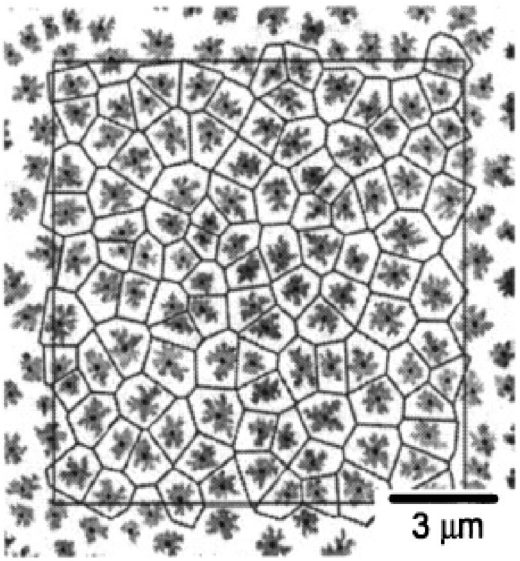


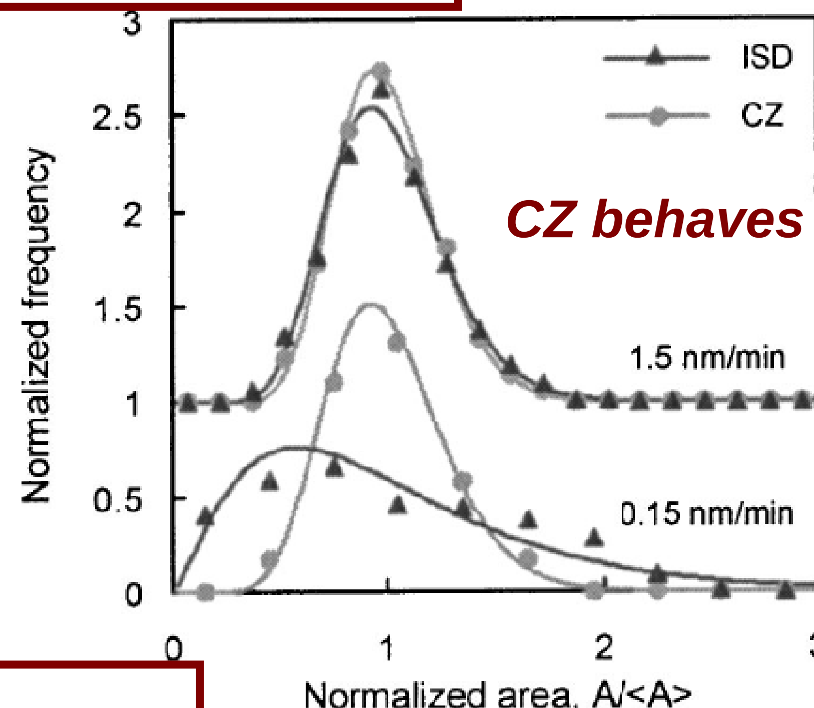
Island size distribution not so informative



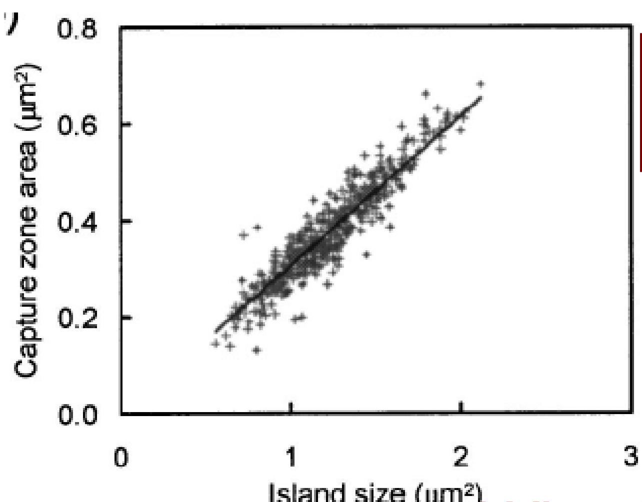
Exp't: Pentacene/SiO_2 Pratontop et al.,PRB 69 (04) 165201

Gamma func'n $\Pi_\alpha(x) = [\alpha^\alpha/\Gamma(\alpha)]\ x^{\alpha-1}\exp(-\alpha x)$



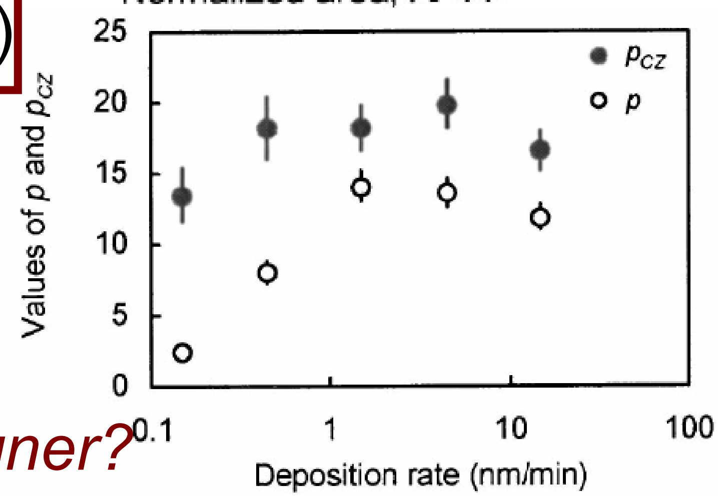


CZ behaves better



$\Pi_{2\varrho+\alpha_0}(s) \approx P_\varrho(s)$

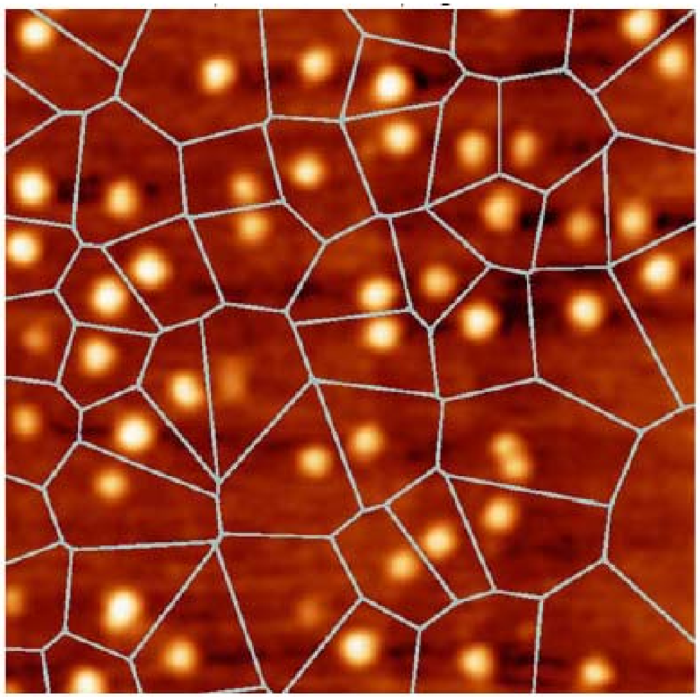
but Π more skewed



Why Gamma, not Wigner?

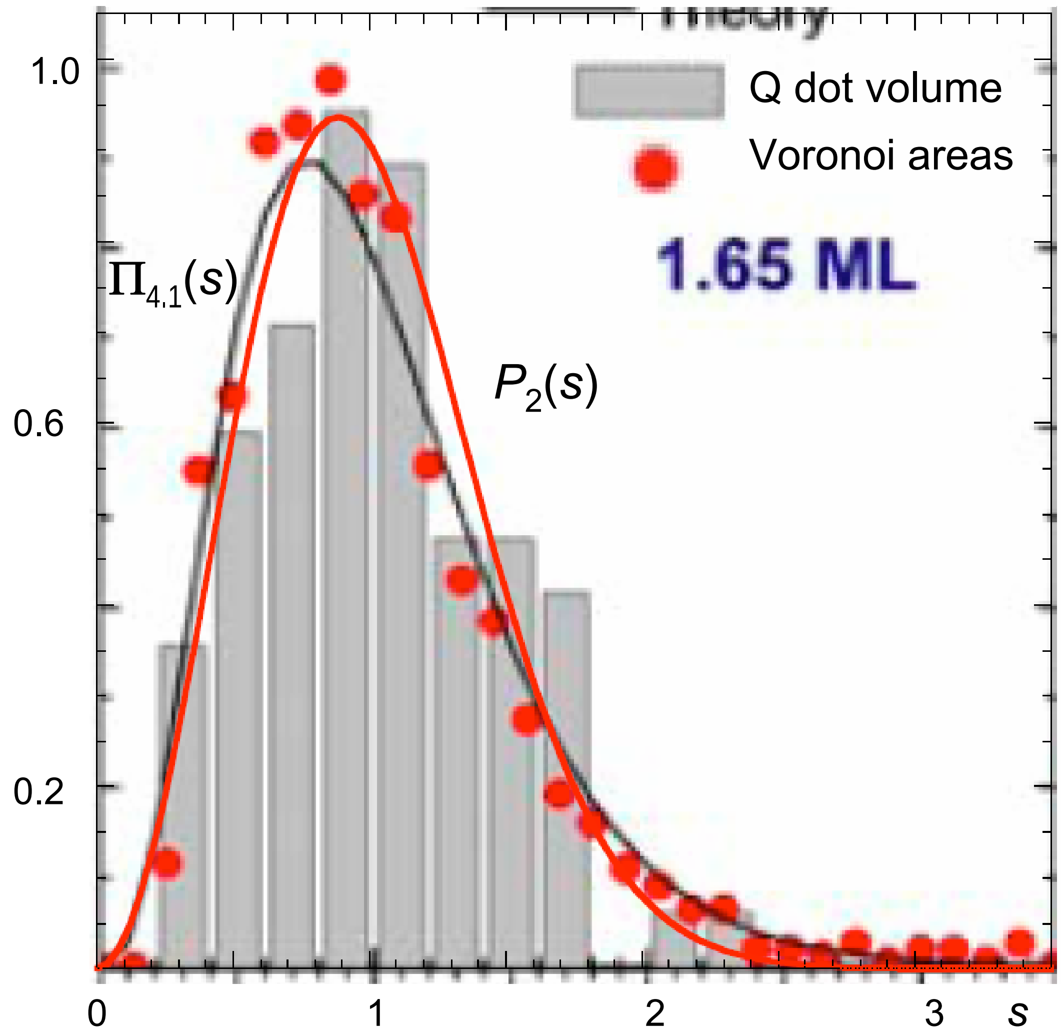
Scale invariance in thin film growth: InAs *quantum dots* on GaAs(001)

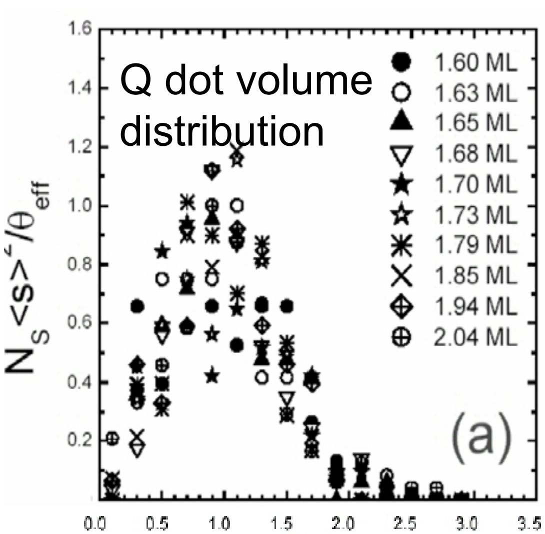
M. Fanfoni *et al.*, PRB **75** (' 07) xxx



AFM, 1.68 ML, 350x350nm^2, 500°C

Θ (ML)	1.65	1.68	1.70	1.73	1.79	1.85
α	4.1	4.6	4.5	4.7	4.5	4.6





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 η

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) / ∂s
[Also i + 1 in 3D & 4D; but $2(i + 1)$ in 1D]

$\acute{N} = \sigma n N_i = \sigma n^{i+1}$
$\sigma = D/\ell^{2-d}$ $s \equiv \ell^d$
$n \propto \ell^2 \approx s^{2/d}$
prod $\propto s^{(2/d)(i+1)}$

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 $P_\varrho(s)$
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 $P_\varrho(s) = a\, s^\varrho\, e^{-bs^2}$ easy to use & describes universal fluctuations $\Rightarrow$ broad applications
- For TWD width $\varrho \Rightarrow$ 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; $\varrho \Rightarrow$ critical nucleus size i and spatial dimension of host lattice