Application of the Wigner Distribution to Non-equilibrium Problems at Surfaces: Relaxation, Growth, and Scaling of Capture Zone

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- Summary of equilibrium terrace-width distributions, Wigner surmise (WS) from RMT $P_{\beta}(s) = a_{\beta}s^{\beta}\exp(-b_{\beta}s^2)$ ($\beta = 1,2,4$ for ensembles with orthogonal, unitary, symplectic symmetry) and applications
- Generalization to arbitrary positive $\beta \rightarrow \varrho$ (GWS), with no underlying symmetry; applications to surface problems, with ϱ related to step repulsion strength
- Fokker-Planck formulation: study of relaxation to equilibrium & way to get GWS
- Apparent narrowing during growth

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• Remarkable progress in characterization of island growth by focusing on capture-zone distribution – GWS with $\rho = i + 1$ [or 2(*i*+1) in 1D]







Deduced & drawn by Harald Ibach (¿while on sabbatical at UM, spring 1997?)



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•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 \Rightarrow added oscillatory term in U: (B/ ℓ^2) cos(2 $k_F \ell$ + ϕ)

•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

Physical Ideas Behind Application of Random Matrices

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

<u>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 <u>common symmetry property</u>, seeking generic properties of ensemble due to symmetry.</u>

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

1) time-reversal invariant with rotational symmetry:

 $H_{mn} = H_{nm} = H^*_{mn}$ (<u>orthogonal</u>)

2) time reversal violated (e.g. electron in fixed **B**)

 $H_{mn} = H^{\dagger}_{mn} (\underline{unitary})$

3) time-reversal invariant with 1/2-integer spin & broken rotational symmetry;

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

σj: Pauli spin matrices, j=1,2,3; I: 2 x 2 unit matrix; HO all real, H(0) sym, others asym

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

<u>Gaussian Orthogonal Ensemble</u>: $\beta=1$ <u>GUnitaryE</u>: $\beta=2$ <u>GSymplecticE</u>: $\beta=4$

<u>GRMT</u> 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

Wigner Surmise (WS)



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

U $P_2(s) = \frac{32}{\pi^2} s^2 \exp\left(-\frac{4}{\pi} s^2\right)$
S $P_4(s) = \left(\frac{64}{9\pi}\right)^3 s^4 \exp\left(-\frac{64}{9\pi} s^2\right)$

$$P_{\beta}(s) = a_{\beta}s^{\beta}\exp(-b_{\beta}s^2)$$

Wigner's argument for the surmise, for the orthogonal ensemble

 $p(\mathcal{H}) \sim \exp[-bN\operatorname{tr}(\mathcal{H}^2)]$ $N \rightarrow \infty$; using Gaussian weighting

 $H = \left(\begin{array}{cc} h_{11} & h_{12} \\ h_{12} & h_{22} \end{array}\right)$ Instead, consider *N*=2, orthogonal symmetry:

joint probability distribution $p(E_1, E_2)$ for adjacent eigenenergies E_1, E_2 to P(s)ds

$$\bar{h} \equiv (h_{11} + h_{22})/2 \qquad u \equiv h_{11} - h_{22} \qquad s \equiv \left(u^2 + 4h_{12}^2\right)^{1/2} = |E_2 - E_1| \qquad \text{so} \qquad E_{1,2} = \bar{h} \pm s/2$$

Consider all possible MEs h_{11}, h_{22}, h_{11}

$$h_{11}, h_{22}, h_{12}$$

$$\iiint p \, dh_{11} dh_{22} dh_{12} = \int ds \iint \exp\left[-2b(E_1^2 + E_2^2)\right] d\bar{h} du \left| \frac{dh_{12}}{ds} \right| \qquad \text{using} \qquad dh_{11} dh_{22} = d\bar{h} du$$

$$p(s)$$

$$h_{12} = \pm (1/2)(s^2 - u^2)^{1/2} \implies |dh_{12}/ds| = (s/2) \left(s^2 - u^2\right)^{-1/2}$$

$$(s/2) \int_{-s}^{s} \left(s^2 - u^2\right)^{-1/2} du = \pi s/2 \qquad 2(E_1^2 + E_2^2) = s^2 + 4\bar{h}^2$$

$$P(s) \sim s \exp(-bs^2)$$

Exact for N = 2 and excellent approximation as $N \to \infty$. For large N, the problem of level crossing $(s \to 0)$ still reduces to a 2×2 problem near the (usually avoided) degeneracy.

To get s = 0, u and h_{12} must vanish simultaneously.

$$p(s) = \int \mathrm{d}u \int \mathrm{d}h_{12} \, p(u, h_{12}) \, \delta\left(s - (u^2 + 4h_{12}^2)^{1/2}\right) \sim s, \ s \ll 1$$

GUE (Gaussian unitary ensemble): H is hermitean, so that h_{12} is complex, and *three* parameters must vanish simultaneously to get s = 0:

$$s = \left[(h_{11} - h_{22})^2 + 4(\Re e \ h_{12})^2 + 4(\Im m \ h_{12})^2 \right]^{1/2}$$

Hence, $p(s) \sim s^2$, corresponding to a spherical (rather than circular) shell of radius s in parameter space.

From W. Zwerger, "Theory of Coherent Transport," in T. Dittrich, ..., W. Zwerger, <u>Quantum Transport and Dissipation</u> (Wiley-VCH, Weinheim, 1998), chap. 1 1957: Wigner surmise for β=1: p₁(s) = a₁ s¹ exp(-b₁s²), 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[iθ_μ], 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_μ-E_ν|^β in the Vandermonde determinant."

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

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

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.



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.

What about ρ other than 1,2,4?

Mixed states of different symmetry; Brody distribution, etc.



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



 \Rightarrow values of a_{e} , b_{e} (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	e	Ã	$A_{\rm W}/A_{\rm G}$	Aw (eV Å)	Experimenters
Pt(110)-(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(111)	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(11,7,7)	306	0.085	5.1	4	1.37	0.004	Geisen [5,54]
Cu(111)	313	0.084	5.0	3.6	1.39	0.004	Geisen [5,54]
Cu(111)	301	0.073	6.0	6.0	1.58	0.006	Geisen [5,54]
Ag(100)	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(111)-(7 × 7)	1100	0.068	6.4	7.0	1.67	0.7	Williams [57]
Si(111)-(1 × 1)Br	853	0.068	6.4	7.0	1.67	0.1	XS. Wang, Williams [58]
Si(111)-Ga	823	0.068	6.6	7.6	1.67	1.8	Fujita Ichikawa [59]
Si(111)-A1 √3	1040	0.058	7.6	10.5	1.85	2.2	SchwennickeWilliams [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(111)	900	0.020	24	135	2.59	6	HahnKern [61]
Si(113) rotated	1200	0.004	124	3.8×10^{3}	2.92	(27 ± 5) ×	van Dijken, Zandvliet, Poel-
						10 ²	sema [9]

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 *Q* ≠ 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 à (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) $\tilde{s} = -\kappa s + \rho/s + noise$

 $t \equiv t$

Noise sets time scale.

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

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

 $\rightarrow P_{\varrho}(s)$



Fokker-Planck vs. Monte Carlo: Effect of Step-step Repulsions



Qualitative result: τ decreases as repulsion rises

Improved tests: Kinetic MC & SOS model



 $E_{barrier} = E_d + m E_a$ breaking m bonds $E_d = 0.9 - 1.1 \text{ eV}; E_a = 0.3 - 0.4 \text{ eV}$ T = 520 - 580 K $\langle \ell \rangle = 4 - 15, 5 \text{ steps}, 10000 \text{ x } L_x$



Fit:

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

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

Find $E_{\text{barrier}} \approx 1 E_{\text{d}} + 3 E_{\text{a}}$

Behavior of τ in SOS via KMC: Ramp E_d, E_a, T, $\langle \ell \rangle$

Unpublished; please write for preprint!

2 other situations of interest

Step Bunch: initially a delta function

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



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

$$P(s,\tilde{t}) = \underbrace{a_{\varrho}s^{\varrho}e^{-\tilde{b}_{\varrho}s^{2}}}_{(1-e^{-\tilde{t}}(1-b_{\varrho}/b_{\varrho_{\circ}}))^{\frac{\varrho_{\circ}+1}{2}}}_{1F_{1}}F_{1}\left(\frac{\varrho_{0}+1}{2},\frac{\varrho_{+}1}{2},\frac{\tilde{b}_{\varrho}s^{2}}{1+(b_{\varrho_{\circ}}/b_{\varrho})(e^{\tilde{t}}-1)}\right)$$

Final



10 ML/s

Does growth flux (step motion) alter TWD?

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

Unpublished; please write for preprint!

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

Evolution of Island Structures: Simulations of i=1

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



0.15 ML

0.20 ML

Island Size Scaling, stable config *i* Amar & Family, PRL 74 (95) 2066

Dynamic scaling assumption



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

Rate Eqns. Blackman & Mulheran, PRB 54 (96) 11681

P₄(*s*) fits numerical data at least as well as B&M's complicated theory expression (not expressible succinctly)

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



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



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





Island size distribution not so informative





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



AFM, 1.68 ML, 350x350nm², 500°C



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



Why it works: Phenomenological theory

CZ does "random walk" with 2 competing effects on *ds/dt*:

- 1] Neighboring CZs hinder growth \Rightarrow 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 + 1using 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]

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

- 4] Combine \Rightarrow 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_Q(s) cf. AP, HG, & TLE, Phys. Rev. Lett. 95 (05) 246101

Summary (see http://www2.physics.umd.edu/~einstein)

- TWD of vicinals provides physical entrée to intriguing 1D fermion models & RMT, can connections to many other current physics issues--universality in fluctuations
- Generalized Wigner surmise (GWS) relevant to problems in many fields, with *p* having physical meaning
- For TWD, $\rho = 1 + [1 + 4 A \tilde{\beta} / (k_{\rm B} T)^2]^{1/2}$
- With Fokker-Planck, study relaxation
- Narrowing of TWD due to growth
- Look at distribution of areas of capture zones, rather than island sizes
- CZ well described by GWS $P_{\varrho}(s)$, characteristic of universal fluctuations, with $\varrho = (2/d) (i + 1)$

References (download: http://www2.physics.umd.edu/~einstein)

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