Recent Developments in Non-Equilibrium Statistical Physics

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Introduction

The statistical mechanics of a system at thermal equilibrium is encoded in the Boltzmann-Gibbs canonical law:

$$P_{\mathrm{eq}}(\mathcal{C}) = \frac{\mathrm{e}^{-E(\mathcal{C})/kT}}{Z}$$

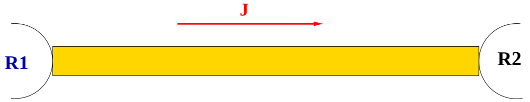
the Partition Function Z being related to the Thermodynamic Free Energy F:

$$F = -kT Log\, Z$$

This provides us with a well-defined prescription to analyze systems *at equilibrium*:
(i) Observables are mean values w.r.t. the canonical measure.
(ii) Statistical Mechanics predicts fluctuations (typically Gaussian) that are out of reach of Classical Thermodynamics (Brownian Motion).

Systems far from equilibrium

Consider a Stationary Driven System in contact with reservoirs at different potentials: no microscopic theory is yet available.

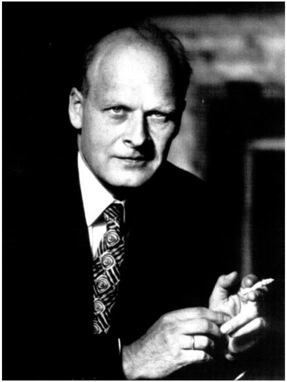


- What are the relevant macroscopic parameters?
- Which functions describe the state of a system?
- Do Universal Laws exist? Can one define Universality Classes?
- Can one postulate a general form for the microscopic measure?
- What do the fluctuations look like ('non-gaussianity')?

In the steady state, a non-vanishing macroscopic current J flows, thus breaking time-reversal invariance

EQUILIBRIUM

Lars Onsager (1903-1976)



‘As in other kinds of book-keeping, the trickiest questions that arise in the application of thermodynamics deal with the proper identification and classification of the entries; the arithmetics is straightforward’ (Onsager, 1967).

FIRST PRINCIPE

$$\boxed{\Delta U = W + Q}$$

THE ENERGY OF THE UNIVERSE IS CONSTANT.

IRREVERSIBILITY

Whenever dissipation and heat exchanges are involved, time reversibility seems to be lost
SOME EVENTS ARE ALLOWED BY NATURE BUT NOT THE OTHERS!

A criterion for separating allowed processes from impossible one is required (Clausius, Kelvin-Planck).

SECOND PRINCIPLE

A NEW physical concept (Clausius): ENTROPY.

$$\boxed{S_2 - S_1 \geq \int_{1\to 2} \frac{\partial Q}{T}}$$

Clausius Inequality (1851)

THE ENTROPY OF THE UNIVERSE INCREASES.

The Mistress of the World and Her Shadow

- A system wants to **minimize its energy**.
- A system wants to **maximize its entropy**.

This competition between energy and entropy is at the heart of most of everyday physical phenomena (such as phase transitions: ice → water).

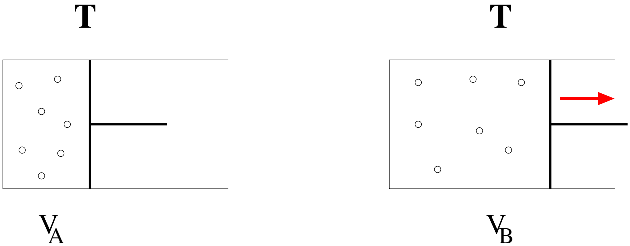
The two principles of thermodynamics can be embodied simultaneously by the **FREE ENERGY** F **:**

$$\boxed{F = U - TS}$$

The decrease of free energy represents the maximal work that one can extract from a system.

Free energy: Maximal Work Theorem

Consider a gas enclosed in a chamber with a moving piston. We suppose that the gas evolves from state A to B and that it can exchange heat only with it environment at fixed temperature T.



Because of irreversibility, the Work, $\mathcal{W}_{useful}$, that one can extract from this system is at most equal to to the decrease of free energy:

$$\boxed{\langle \mathcal{W}_{useful} \rangle \leq F_{\rm initial} - F_{\rm final} = -\Delta F}$$

STATISTICAL MECHANICS

J. C. Maxwell

L. Boltzmann

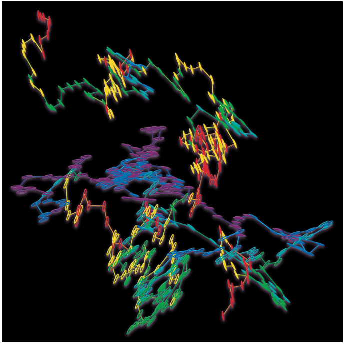
The connection with thermodynamics is given by Boltzmann's formula or, equivalently

$$F = -kT Log\ Z$$

Thermal Equilibrium: a dynamical state

Equilibrium is a dynamical concept. At the molecular scale things constantly change and a system keeps on evolving through various microscopic configurations:

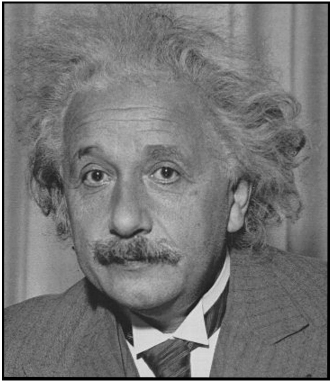
Thermodynamic observables are nothing but average values of fluctuating, probabilistic, microscopic quantities.



Robert Brown (1773-1858)

Physics of Brownian Motion: The Einstein Formula

The Brownian Particle is restlessly shaken by water molecules. It diffuses as a random-walker.



$$\boxed{D = \frac{RT}{6\pi\eta a\mathcal{N}}}$$

R: Perfect Gas Constant
T: Temperature
η : viscosity of water
a: diameter of the pollen
$\mathcal{N}$: Avogadro Number

Jean Perrin: ‘I have weighted the Hydrogen Atom’

Fluctuation-Dissipation Relation

Suppose that the Brownian Particle is subject to a small force $f_{\rm ext}$. Balancing with the viscous force $-(6\pi\eta a)v$ (Stokes) gives the limiting speed

$$v_\infty = \sigma f_{\rm ext} \quad \text{with } \sigma = \frac{1}{6\pi\eta a}$$

The response coefficient σ is called a susceptibility.

The Einstein Relation can be rewritten as

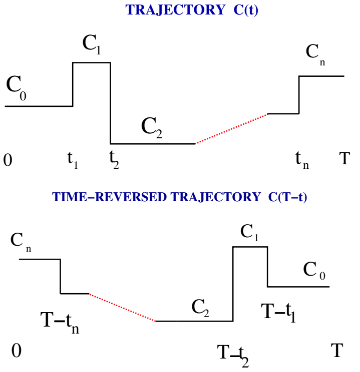
$$\sigma = \frac{D}{kT}$$

$$\boxed{\text{Susceptibility (Linear Response)} \equiv \text{Fluctuations at Equilibrium}}$$

(Kubo Formula)

Time-reversal Invariance and Detailed Balance

The microscopic equations are invariant by time-reversal: the probability of a given trajectory in phase-space is equal to the probability of the time reversed trajectory.



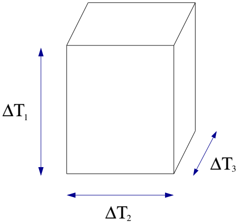
DETAILED BALANCE:

$$\boxed{\frac{e^{-\frac{E(\mathcal{C})}{kT}}\,W(\mathcal{C}\to\mathcal{C}')}{e^{-\frac{E(\mathcal{C}')}{kT}}\,W(\mathcal{C}'\to\mathcal{C})}=1}$$

Onsager (1931)

A system is at thermal equilibrium iff it satisfies detailed-balance.

Onsager's Reciprocity Relations (1931)



$$J_i = -\sum_{k=1}^{3} L_{ik} \frac{\partial T}{\partial x_k}$$

(Fourier Law)

The Conductivity Tensor **L** remains symmetric even though the crystal does not display any special symmetry

$$\boxed{L_{ik} = L_{ki}}$$

Crucial for Thermoelectric Effects.

Linear Response Theory

Brownian Fluctuations show that Equilibrium is a dynamical concept.

The fact that the dynamics converges towards **thermodynamic equilibrium** and **time-reversal invariance** (detailed-balance) are the key-properties behind Einstein and Onsager's Relations.

Thermodynamic equilibrium is characterized by the fact that the average values of all the **fluxes exchanged** between the system and its environment (matter, charge, energy, spin...) **identically vanish**.

OUT OF EQUILIBRIUM

In Nature, many systems are far from thermodynamic equilibrium and keep on exchanging matter, energy, information with their surroundings. There is no general conceptual framework to study such systems.

A Surprise: The Jarzynski Identity

Remember the maximal work inequality:

$$\boxed{\langle W \rangle \leq F_A - F_B = -\Delta F}$$

We put brackets to emphasize that we consider the average work:
Statistical Physics has taught us that physical observables fluctuate.

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Statistical Physics has taught us that physical observables fluctuate.

It was found very recently that there exists a remarkable equality that *underlies* this classical inequality.

$$\boxed{\left\langle e^{\frac{W}{kT}} \right\rangle = e^{-\frac{\Delta F}{kT}}}$$

The Jarzynski Identity

$$\boxed{\left\langle \mathrm{e}^{\frac{W}{kT}} \right\rangle = \mathrm{e}^{-\frac{\Delta F}{kT}}}$$

Jarzynski's Work Theorem (1997)

Consequences

1. Jarzynski's identity mathematically implies the good old maximal work inequality.

2. But, in order to have an EQUALITY, there must exist some occurrences in which

$$W > -\Delta F$$

There must be instances in which the classical inequality which results from the Entropy Principle is 'violated'.

3. Jarzynski's identity was checked experimentally on single RNA folding/unfolding experiments (Bustamante et al.): it has experimental applications in biophysics and at the nanoscale.

4. The relation of Crooks: a refinement Jarzynski's identity that allows us to quantify precisely the 'transient violations of the second principle'.

$$\frac{\mathrm{P}^{\mathrm{F}}(W)}{\mathrm{P}^{\mathrm{R}}(-W)} = \mathrm{e}^{\frac{W-\Delta F}{kT}} \qquad \text{(Crooks,1999)}$$

Vérification Expérimentale de l'identité de Crooks

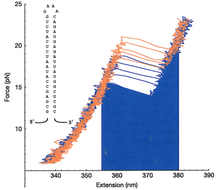

Figure 1 | Force–extension curves. The stochasticity of the unfolding and refolding process is characterized by a distribution of unfolding or refolding work trajectories. Five unfolding (orange) and refolding (blue) force–extension curves for the RNA hairpin are shown (loading rate of 7.5 pN s^{-1}). The blue area under the curve represents the work returned to the machine as the molecule switches from the unfolded to the folded state. The RNA sequence is shown as an inset.

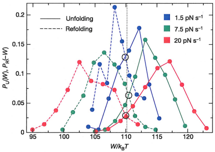

Figure 2 | Test of the CFT using an RNA hairpin. Work distributions for RNA unfolding (continuous lines) and refolding (dashed lines). We plot negative work, $P_R(-W)$, for refolding. Statistics: 130 pulls and three molecules ($r = 1.5\,\text{pN s}^{-1}$), 380 pulls and four molecules ($r = 7.5\,\text{pN s}^{-1}$), 700 pulls and three molecules ($r = 20.0\,\text{pN s}^{-1}$), for a total of ten separate experiments. Good reproducibility was obtained among molecules (see Supplementary Fig. S2). Work values were binned into about ten equally spaced intervals. Unfolding and refolding distributions at different speeds show a common crossing around $\Delta G = 110.3\,k_BT$.

Rare Events and Large Deviations

Let $\epsilon_1, \dots, \epsilon_N$ be N independent binary variables, $\epsilon_k = \pm 1$, with probability p (resp. $q = 1 - p$). Their sum is denoted by $S_N = \sum_1^N \epsilon_k$.

- The Law of Large Numbers implies that $S_N/N \to p - q$ a.s.
- The Central Limit Theorem implies that $[S_N - N(p-q)]/\sqrt{N}$ converges towards a Gaussian Law.

One can show that for $-1 < r < 1$, in the large N limit,

$$\mathrm{Pr}\left(\frac{S_N}{N} = r\right) \sim \mathrm{e}^{-N\,\Phi(r)}$$

where the positive function $\Phi(r)$ vanishes for $r = (p - q)$.

The function $\Phi(r)$ is a Large Deviation Function: it encodes the probability of rare events.

$$\Phi(r) = \frac{1+r}{2}\ln\left(\frac{1+r}{2p}\right) + \frac{1-r}{2}\ln\left(\frac{1-r}{2q}\right)$$

Density Fluctuations

Consider a gas in a room, at thermal equilibrium. The probability of observing a density profile $\rho(x)$ takes the form:

$$\mathrm{Pr}\{\rho(x)\} \sim \mathrm{e}^{-\beta V\, \mathcal{F}(\{\rho(x)\}}$$

What is $\mathcal{F}(\{\rho(x)\}$?

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What is $\mathcal{F}(\{\rho(x)\}$?

$$\mathcal{F}(\{\rho(x)\}) = \int_0^1 \left(f(\rho(x), T) - f(\bar{\rho}, T)\right) d^3x$$

Free Energy can be viewed as a Large Deviation Function.

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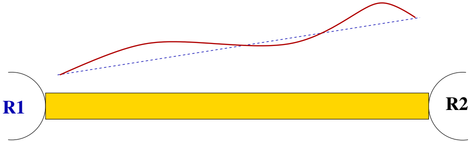
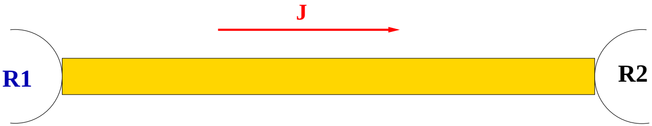


What is the probability of observing an atypical density profile in the steady state? What does the functional $\mathcal{F}(\{\rho(x)\})$ look like for such a non-equilibrium system?

Large Deviations of the Total Current



Let Y_t be the total charge transported through the system (total current) between time 0 and time t.

In the stationary state: a non-vanishing mean-current $\frac{Y_t}{t} \to J$

The fluctuations of Y_t obey a Large Deviation Principle:

$$P\left(\frac{Y_t}{t} = j\right) \sim e^{-t\Phi(j)}$$

$\Phi(j)$ being the *large deviation function* of the total current.

Note that $\Phi(j)$ is positive, vanishes at $j = J$ and is convex (in general).

The Gallavotti and Cohen Symmetry

Large deviation functions obey remarkable identities that remain valid far from equilibrium: *The Fluctuation Theorem of Gallavotti and Cohen.*

Large deviation functions obey a symmetry that remains valid far from equilibrium:

$$\boxed{\Phi(j) - \Phi(-j) = \alpha j}$$

Equivalently,

$$\frac{Prob\,(\,j\,)}{Prob\,(-j)} \sim e^{-t\alpha j}$$

This Fluctuation Theorem of Gallavotti and Cohen is deep and general: it reflects covariance properties under time-reversal.

In the vicinity of equilibrium the Fluctuation Theorem yields the fluctuation-dissipation relation (Einstein), Onsager's relations and linear response theory (Kubo).

The General Large Deviations Problem

More generally, the probability to observe an atypical current $j(x,t)$ and the corresponding density profile $\rho(x,t)$ during $0 \leq s \leq L^2\,T$ (L being the size of the system) is given by

$$\mathrm{Pr}\{j(x,t), \rho(x,t)\} \sim \mathrm{e}^{-L\,\mathcal{I}(j,\rho)}$$

Is there a Principle which gives this large deviation functional for systems out of equilibrium?

Why study Large Deviations?

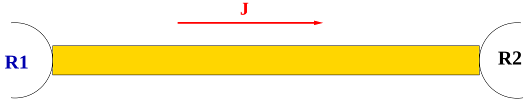
- Equilibrium Thermodynamic potentials (Entropy, Free Energy) can be defined as large deviation functions.
- Large deviations are well defined far from equilibrium: they are good candidates for being non-equilibrium potentials.
- Large deviation functions obey remarkable identities, valid far from equilibrium (Gallavotti-Cohen Fluctuation Theorem; Jarzynski and Crooks Relations).
- These identities imply, in the vicinity of equilibrium, the fluctuation dissipation relation (Einstein), Onsager's relations and linear response theory (Kubo).

EXACT RESULTS

FAR FROM EQUILIBRIUM

Study Non-Equilibrium via Model Solving

The fundamental non-equilibrium system



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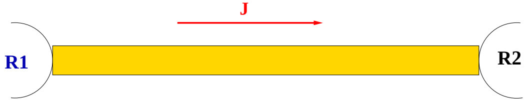
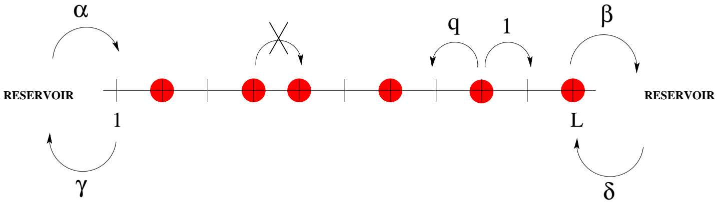


The asymmetric exclusion model with open boundaries (ASEP)



Thousands of articles devoted to this model in the last 20 years:
Paradigm for non-equilibrium behaviour

Statistics of the current and of the density profile?

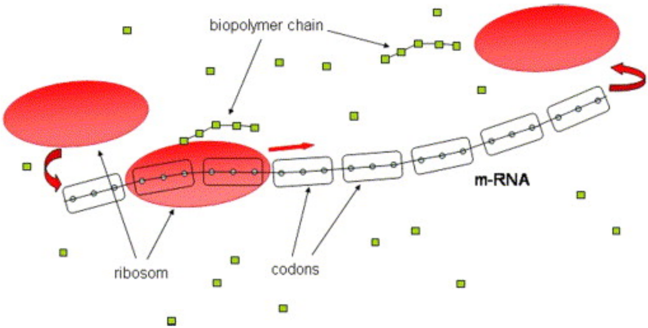
ORIGINS

- Interacting Brownian Processes (Spitzer, Harris, Liggett).
- Driven diffusive systems (Katz, Lebowitz and Spohn).
- Transport of Macromolecules through thin vessels.
  Motion of RNA templates.
- Hopping conductivity in solid electrolytes.
- Directed Polymers in random media. Reptation models.
- Interface dynamics. KPZ equation

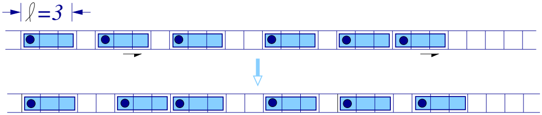
APPLICATIONS

- Traffic flow.
- Sequence matching.
- Brownian motors.

An Elementary Model for Protein Synthesis



C. T. MacDonald, J. H. Gibbs and A.C. Pipkin, Kinetics of biopolymerization on nucleic acid templates, *Biopolymers* (1968).



The Matrix Ansatz for ASEP (DEHP, 1993)

The stationary probability of a configuration $\mathcal{C}$ is given by

$$P(\mathcal{C}) = \frac{1}{Z_L} \langle W | \prod_{i=1}^{L} \left(\tau_i D + (1 - \tau_i) E \right) | V \rangle$$

where $\tau_i = 1$ (or 0) if the site i is occupied (or empty).
The normalization constant $Z_L = \langle W | (D + E)^L | V \rangle$.

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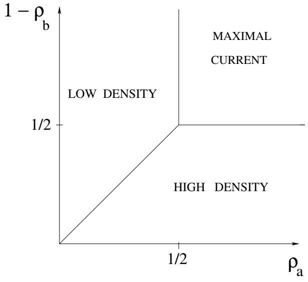
The operators D and E, the vectors $\langle W|$ and $|V\rangle$ satisfy

$$\begin{aligned} D\,E - qED &= (1-q)\,(D+E) \\ (\beta\,D - \delta\,E)\,|V\rangle &= |V\rangle \\ \langle W|(\alpha\,E - \gamma\,D) &= \langle W| \end{aligned}$$

This algebra, related to q-deformed oscillators, encodes the stationary properties of the system and allows us to derive the exact phase diagram of the model.

The Phase Diagram of the open ASEP



$\rho_a = \frac{1}{a_+ + 1}$: effective left reservoir density.

$\rho_b = \frac{b_+}{b_+ + 1}$: effective right reservoir density.

$$a_\pm = \frac{(1-q-\alpha+\gamma) \pm \sqrt{(1-q-\alpha+\gamma)^2 + 4\alpha\gamma}}{2\alpha}$$

$$b_\pm = \frac{(1-q-\beta+\delta) \pm \sqrt{(1-q-\beta+\delta)^2 + 4\beta\delta}}{2\beta}$$

Large Deviations of the Density Profile in ASEP

The probability of observing an **atypical density profile in the steady state of the ASEP** was calculated starting from Matrix Ansatz for the exact microscopic solution (B. Derrida, J. Lebowitz E. Speer, 2002).
In the symmetric case $q = 1$:

$$\mathcal{F}(\{\rho(x)\}) = \int_0^1 dx \left(B(\rho(x), F(x)) + \log \frac{F'(x)}{\rho_2 - \rho_1} \right)$$

where $B(u, v) = (1 - u) \log \frac{1-u}{1-v} + u \log \frac{u}{v}$ and $F(x)$ satisfies

$$F\left(F'^2 + (1 - F)F''\right) = F'^2 \rho \quad \text{with} \quad F(0) = \rho_1 \text{ and } F(1) = \rho_2 \,.$$

This functional is non-local as soon as $\rho_1 \neq \rho_2$.

This functional is NOT identical to the one given by local equilibrium.

Note that in the case of equilibrium, for $\rho_1 = \rho_2 = \bar{\rho}$, we recover

$$\mathcal{F}(\{\rho(x)\}) = \int_0^1 dx \left\{ (1 - \rho(x)) \log \frac{1 - \rho(x)}{1 - \bar{\rho}} + \rho(x) \log \frac{\rho(x)}{\bar{\rho}} \right\}$$

Current Statistics

A parametric representation of the cumulant generating function $E(\mu)$ is obtained using integrability techniques (Bethe Ansatz).
For $\alpha = \beta = 1$:

$$\begin{aligned} \mu &= -\sum_{k=1}^{\infty} \frac{(2k)!}{k!} \frac{[2k(L+1)]!}{[k(L+1)]!\,[k(L+2)]!} \frac{B^k}{2k}\,, \\ E &= -\sum_{k=1}^{\infty} \frac{(2k)!}{k!} \frac{[2k(L+1)-2]!}{[k(L+1)-1]!\,[k(L+2)-1]!} \frac{B^k}{2k}\,. \end{aligned}$$

First cumulants of the current

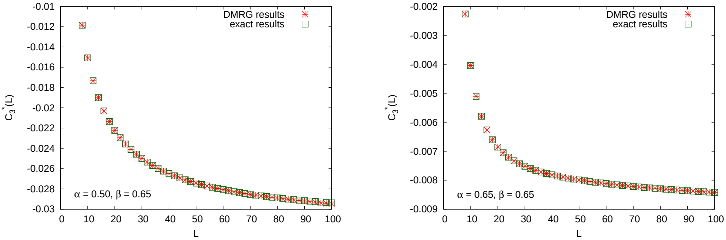
- Mean Value : $J = \frac{L+2}{2(2L+1)}$
- Variance : $\Delta = \frac{3}{2} \frac{(4L+1)![L!(L+2)!]^2}{[(2L+1)!]^3(2L+3)!}$
- Skewness :
  $E_3 = 12 \frac{[(L+1)!]^2[(L+2)!]^4}{(2L+1)[(2L+2)!]^3} \left\{ 9 \frac{(L+1)!(L+2)!(4L+2)!(4L+4)!}{(2L+1)![(2L+2)!]^2[(2L+4)!]^2} - 20 \frac{(6L+4)!}{(3L+2)!(3L+6)!} \right\}$
  For large systems: $E_3 \to \frac{2187-1280\sqrt{3}}{10368}\pi \sim -0.0090978\ldots$

Large Deviation Function of the Current

In the limit of large size systems, the following exact expression is found for the Large Deviation Function of the current:

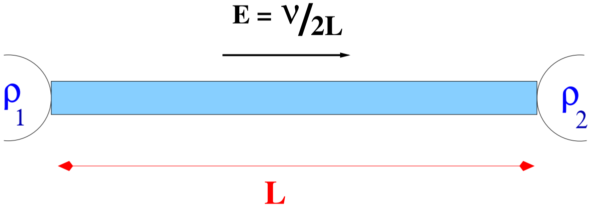
$$\boxed{\Phi(j) = (1-q)\left\{\rho_a - r + r(1-r)\ln\left(\frac{1-\rho_a}{\rho_a}\frac{r}{1-r}\right)\right\}}$$

where the current j is parametrized as $j = (1-q)r(1-r)$.



SKEWNESS

The Hydrodynamic Limit: Diffusive case



Starting from the microscopic level, define local density $\rho(x,t)$ and current $j(x,t)$ with macroscopic space-time variables $x = i/L, t = s/L^2$ (diffusive scaling).

The typical evolution of the system is given by the hydrodynamic behaviour (Burgers-type equation):

$$\partial_t \rho = \nabla \left(D(\rho) \nabla \rho \right) - \nu \nabla \sigma(\rho) \quad \text{with} \quad D(\rho) = 1 \text{ and } \sigma(\rho) = 2\rho(1-\rho)$$

(Lebowitz, Spohn, Varadhan)

How can Fluctuations be taken into account?

Fluctuating Hydrodynamics

Consider Y_t the total number of particles transfered from the left reservoir to the right reservoir during time t.

- $\lim_{t\to\infty} \frac{\langle Y_t \rangle}{t} = D(\rho)\frac{\rho_1-\rho_2}{L} + \sigma(\rho)\frac{\nu}{L}$ for $(\rho_1-\rho_2)$ small
- $\lim_{t\to\infty} \frac{\langle Y_t^2 \rangle}{t} = \frac{\sigma(\rho)}{L}$ for $\rho_1 = \rho_2 = \rho$ and $\nu = 0$.

Then, the equation of motion is obtained as:

$$\partial_t \rho = -\partial_x j \quad \text{with} \quad j = -D(\rho)\nabla\rho + \nu\sigma(\rho) + \sqrt{\sigma(\rho)}\xi(x,t)$$

where $\xi(x,t)$ is a Gaussian white noise with variance

$$\langle \xi(x',t')\xi(x,t)\rangle = \frac{1}{L}\delta(x-x')\delta(t-t')$$

For the symmetric exclusion process, the ‘phenomenological’ coefficients are given by

$$D(\rho) = 1 \quad \text{and} \quad \sigma(\rho) = 2\rho(1-\rho)$$

A General Principle for Large Deviations?

The probability to observe an atypical current $j(x,t)$ and the corresponding density profile $\rho(x,t)$ during a time $L^2 T$ (L being the size of the system) is given by

$$\mathrm{Pr}\{j(x,t),\rho(x,t)\} \sim \mathrm{e}^{-L\,\mathcal{I}(j,\rho)}$$

A general principle has been found (G. Jona-Lasinio et al.), to express this large deviation functional $\mathcal{I}(j,\rho)$ as an optimal path problem:

$$\mathcal{I}(j,\rho) = \min_{\rho,j}\left\{\int_0^T dt \int_0^1 dx \frac{(j - \nu\sigma(\rho) + D(\rho)\nabla\rho)^2}{2\sigma(\rho)}\right\}$$

with the constraint: $\partial_t \rho = -\nabla . j$

Knowing $\mathcal{I}(j,\rho)$, one could derive the large deviations of the current and of the density profile. For instance, $\Phi(j) = \min_\rho\{\mathcal{I}(j,\rho)\}$

However, at present, the available results for this variational theory are precisely the ones given by exact solutions of the ASEP.

Macroscopic Fluctuation Theory

Mathematically, one has to solve the corresponding Euler-Lagrange equations. The Hamiltonian structure is expressed by a pair of conjugate variables (p, q).

After some transformations, one obtains a set of coupled PDE's (here, we take $\nu = 0$):

$$\begin{aligned} \partial_t q &= \partial_x [D(q) \partial_x q] - \partial_x [\sigma(q) \partial_x p] \\ \partial_t p &= -D(q) \partial_{xx} p - \frac{1}{2} \sigma'(q) (\partial_x p)^2 \end{aligned}$$

where $q(x,t)$ is the density-field and $p(x,t)$ is a conjugate field.

The 'transport coefficients' $D(q)(= 1)$ and $\sigma(q)(= 2q(1-q))$ contain the information of the microscopic dynamics relevant at the macroscopic scale.

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- A general framework but the MFT equations are very difficult to solve in general. By using them one can in principle calculate large deviation functions directly at the macroscopic level.
- The analysis of this new set of 'hydrodynamic equations' has just begun!

Conclusions

Non-Equilibrium Statistical Physics has undergone remarkable developments in the last two decades and a unified framework is emerging.

Large deviation functions (LDF) appear as a generalization of the thermodynamic potentials for non-equilibrium systems. They satisfy remarquable identities (Gallavotti-Cohen, Jarzynski-Crooks) valid far from equilibrium.

The LDF's are very likely to play a key-role in the future of non-equilibrium statistical mechanics.

Current fluctuations are a signature of non-equilibrium behaviour. The exact results derived for the Exclusion Process can be used to calibrate the more general framework of fluctuating hydrodynamics (MFT), which is currently being developed.