

Thermodynamics of Work in Open Quantum Systems (and some other stuff...)



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[S. Suomela *et al.,* Phys. Rev. B **90**, 094304 (2014); Phys. Rev. E **91**, 022126 (2015); S. Suomela, J. Pekola, T.A-N., unpublished]

Tapio's office

The campus is located in **Otaniemi Technology Hub** in Espoo city, 10 km from the center of Helsinki.

campus

Aalto University School of Science Designed by Alvar Aalto in 1960's. A unique combination of education, study and business densely packed into a small area by the sea.

Aalto University (HUT until 2010)

A merger of three leading Finnish universities in January 2010

University of Art and Design Helsinki founded 1871

Helsinki School of Economics founded 1911

Helsinki University of Technology founded 1849





















The first Foundation University in Finland, with an independent board Initial capital of the foundation 700 M€, now 1 000 M€



Aalto University School of Science

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MSP group summary







Miracles happen at nanoscale!



Arrow of time



Suppose the piece is a cube of aluminium, edge length x If x = 5 cm => $P_R/P_L = 10^{10^{10}}$ If x = 5 nm => $P_R/P_L = 4.5$



Stochastic thermodynamics



Thermodynamic quantities (heat, energy, entropy) become *fluctuating stochastic quantities,* which in many (*but not in all*) cases obey *fluctuation relations*

Jarzynski (1997):
$$\langle e^{-\beta W_d} \rangle = 1$$

Crooks FT (1998):
 $\frac{P_F(-W_d)}{P_R(W_d)} = e^{-\beta W_d}$
 x_0
 x_0
 $\lambda(t)$
Control parameter

Seifert (2005): $\langle e^{-\Delta S_T} \rangle = 1$ (generally true)

etc.

$$W_d = W - \Delta F$$

"dissipative work"









$$(*)\Delta S_T[X] = \ln \frac{P[x_0]P[X|x_0]}{P[\tilde{x}_0]P[\tilde{X}|\tilde{x}_0]}$$

School of Science





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This can be split as

$$\Delta S_T[X] = \Delta S_S[X] + \Delta S_R[X] = \ln \frac{P[x_0]}{P[\tilde{x}_0]} + \ln \frac{P[X|x_0]}{P[\tilde{X}|\tilde{x}_0]}$$



$$(*)\Delta S_T[X] = \ln \frac{P[x_0]P[X|x_0]}{P[\tilde{x}_0]P[\tilde{X}|\tilde{x}_0]}$$

$$(**)\langle e^{-\Delta S_T[X]}\rangle = 1$$

Mathematical identity Conservation of probability

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Assuming "local" detailed balance and Boltzmann distribution

$$\frac{W_{x_j, x_{j-1}}(\lambda_j)}{W_{x_{j-1}, x_j}(\lambda_j)} = e^{-\beta (E_{x_j} - E_{x_{j-1}})(\lambda_j)}$$





Using (local) detailed balance & canonical equilibrium:

$$\begin{split} \Delta S_T[X] &= \beta \Delta E[X] - \beta \Delta F - \beta Q[X] = \beta (W[X] - \Delta F) \\ \text{The total entropy expression } (*) \Delta S_T[X] = \ln \frac{P[x_0] P[X|x_0]}{P[\tilde{x}_0] P[\tilde{X}|\tilde{x}_0]} \end{split}$$

$$\frac{P_F(-W_d)}{P_R(W_d)} = e^{-\beta W_d}$$

The Seifert integral FT $(**)\langle e^{-\Delta S_T[X]}\rangle = 1$ becomes the Jarzynski eqn.

$$\langle e^{-\beta W_d} \rangle = 1$$







Problem: There is no (unique) Hermitian "work" operator \hat{W} (there is only the energy operator \hat{H})

Solution(?): Several possible definitions of quantum work have been proposed:

M. Esposito, U. Harbola, and S. Mukamel, Rev. Mod. Phys. 81, 1665 (2009)
P. Solinas, D.V. Averin, and J.P. Pekola, Phys. Rev. B 87, 060508 (2013)
J.M. Horowitz and J.M.R. Parrondo, New J. Phys. 15, 085028 (2013)
S. Suomela, P. Solinas, J.P. Pekola, J. Ankerhold and T. Ala-Nissila, Phys. Rev. B 90, 094304 (2014)







The probability to be in state $|\psi_n^{(A)}
angle$

$$p_n^{(A)} = \frac{e^{-\beta E_n^{(A)}}}{\mathcal{Z}_A} = e^{-\beta (F_A - E_n^{(A)})}$$

$$F_A = -k_B T \ln \mathcal{Z}_A$$







$$\langle e^{-\beta W} \rangle = e^{-\beta \Delta F}$$

- Jarzynski equation is recovered in the *two-point* measurement protocol (TMP) in the case where the dynamics is unitary
- Similar to the classical case, if the system is not isolated during the drive, there are problems with the Jarzynski equation...



Work in open quantum systems



• Consider an open quantum system driven by a classical protocol $\lambda(t)$. The total Hamiltonian

$$\begin{split} \hat{H}(t) &= \hat{H}_S(t) + \hat{H}_B + \hat{H}_C \\ \text{system bath system-bath coupling} \\ \hline \mathbf{DRIVE} \quad \underbrace{\mathbf{U} \quad \mathbf{Q}}_{\mathbf{QUANTUM}} \quad \underbrace{\mathbf{U} \quad \mathbf{Q}}_{\mathbf{QUANTUM}} \\ \hat{H}_S(t) &= \hat{H}_0 + \hat{V}(t) = \hat{H}_0 + \lambda(t)\hat{V}_0 \end{split} \\ \end{split}$$

[Quantum drive considered in J. Salmilehto et al., PRE 89, 052128 (2014)]



<u>Work in open quantum</u> <u>systems</u>



 To consider work on an O.Q.S., define the *Power Operator* [Solinas *et al.* PRB (2013)]:

$$\hat{P}(t) := \frac{\partial \hat{H}}{\partial \lambda} \dot{\lambda} = \frac{\partial \hat{H}}{\partial t}$$

• The average power is given by

$$\langle \hat{P}(t) \rangle = \text{Tr}[\hat{\rho}(t)\hat{P}(t)]$$

and the work
$$\langle W \rangle := \int_{t_A}^{t_B} \langle \hat{P}(t) \rangle dt$$







• If the dynamics can be described by a master equation with dissipation operator $\hat{\mathcal{L}}(\rho)$, then

$$\langle W \rangle = \langle \hat{H}(t_B) \rangle - \langle \hat{H}(t_A) \rangle - \int_{t_A}^{t_B} dt \operatorname{Tr}[\hat{\mathcal{L}}(\rho)\hat{H}(t)]$$

vanishes for isolated systems ("heat" term)

Power operator approach gives work different from TMP even for closed systems when at coherent superposition of eigenstates



Stochastic quantum dynamics



- The classical stochastic dynamics can be generalized to the quantum case by considering instantaneous transitions induced by (weak) coupling to the environment
- For open systems, the Lindblad master equation contains coupling terms with the environment
- For single realizations: Stochastic evolution [Horowitz & Parrondo, New J. Phys. (2013)]





Stochastic quantum dynamics



• The Lindblad master equation:

$$\frac{d\hat{\rho}(t)}{dt} = -\frac{i}{\hbar} \left[\hat{H}_S(t), \hat{\rho}(t) \right] + \sum_m \hat{C}_m(t)\hat{\rho}(t)\hat{C}_m^{\dagger}(t) - \frac{1}{2} \sum_m \left(\hat{C}_m^{\dagger}(t)\hat{C}_m(t)\hat{\rho}(t) + \hat{\rho}(t)\hat{C}_m^{\dagger}(t)\hat{C}_m(t) \right)$$

Main assumptions: Weak coupling to the heat bath, memory-free heat bath, and neglecting fast oscillations



Stochastic quantum dynamics



• The Lindblad master equation:

$$\begin{aligned} \frac{d\hat{\rho}(t)}{dt} &= -\frac{i}{\hbar} \left[\hat{H}_S(t), \hat{\rho}(t) \right] + \sum_m \hat{C}_m(t) \hat{\rho}(t) \hat{C}_m^{\dagger}(t) \\ &- \frac{1}{2} \sum_m \left(\hat{C}_m^{\dagger}(t) \hat{C}_m(t) \hat{\rho}(t) + \hat{\rho}(t) \hat{C}_m^{\dagger}(t) \hat{C}_m(t) \right) \end{aligned}$$

Main assumptions: Weak coupling to the heat bath, memory-free heat bath, and neglecting fast oscillations



<u>Stochastic quantum</u> <u>dynamics</u>



• The Lindblad master equation:



Main assumptions: Weak coupling to the heat bath, memory-free heat bath, and neglecting fast oscillations



<u>Stochastic quantum</u> <u>dynamics</u>



- Quantum jump (Monte Carlo wave function) method: Stochastic unraveling of the Lindblad equation
- Semi-Classical: Bath acts as a classical measurer



[Mølmer, Castin & Dalibard, JOSA B (1993); Badescu, Ying & TA-N, PRL (2001); Hekking & Pekola, PRL (2013)]





1. Derivation of Integral Fluctuation Theorem for stochastic quantum dynamics

[S. Suomela, J. Salmilehto, I.G. Savenko, T.A-N & M. Möttönen, Phys. Rev. E **91**, 022126 (2015)]



Between jumps the time evolution is given by

$$\hat{U}(t_{j+1}, t_j) = \mathcal{T}_{\leftarrow} e^{-\frac{i}{\hbar} \int_{t_j}^{t_{j+1}} \left(\hat{H}_S(t) - \frac{i\hbar}{2} \sum_m \hat{C}_m^{\dagger} \hat{C}_m \right) dt}$$









<u>Stochastic quantum</u> <u>dynamics: IFT</u>



$$\left\langle \frac{Pr'(E(\tau) = E_{\tau})}{Pr(E(t_0) = E_0)} \prod_{j=1}^{N} \frac{\Gamma'_{m_j}(t_j)}{\Gamma_{m_j}(t_j)} \right\rangle = 1$$

Assuming Boltzmann distribution *and* that the noise source follows detailed balance s.t. the ratio of the transition rate gives dissipated heat, gives the Jarzynski eq.

$$\langle e^{-\beta W} \rangle = e^{-\beta \Delta F}$$





2. Derivation of generating functional and moments of work

[S. Suomela, P. Solinas, J.P. Pekola, J. Ankerhold & T.A-N, Phys. Rev. B **90**, 094304 (2014)]



where $\hat{C}_1(\tau) = \hat{P}(\tau) = \partial_{\tau} \hat{H}(\tau),$ $\hat{C}_2(\tau) = \left[\hat{H}(\tau), \partial_{\tau} \hat{H}(\tau)\right],$

etc.

[Generalization of Esposito, Harbola, Mukamel, RMP (2009)]



<u>Generating functional within</u> <u>TMP: Moments of work</u>



$$\langle W \rangle = \int_0^\tau dt_1 \langle \hat{P}^H(t_1) \rangle,$$

$$\langle W^2 \rangle = 2 \int_0^\tau dt_1 \int_0^{t_1} dt_2 \operatorname{Re} \left\{ \langle \hat{P}^H(t_1) \hat{P}^H(t_2) \rangle \right\},$$



. . .

<u>Generating functional within</u> <u>TMP: Moments of work</u>



$$\begin{split} \langle W^{3} \rangle_{0} &= 3 \int_{0}^{\tau} dt_{1} \int_{0}^{t_{1}} dt_{2} \int_{0}^{t_{2}} dt_{3} \operatorname{Re} \left\{ \langle \hat{P}^{H}(t_{1}) \hat{P}^{H}(t_{2}) \hat{P}^{H}(t_{3}) \\ &+ \langle \hat{P}^{H}(t_{3}) \hat{P}^{H}(t_{1}) \hat{P}^{H}(t_{2}) \rangle \right\}, \quad \text{if } [\hat{H}(t), \partial_{t} \hat{H}(t)] = 0 \\ \langle W^{3} \rangle &= \langle W^{3} \rangle_{0} + \frac{1}{4} \int_{0}^{\tau} dt \langle \hat{C}_{3}^{H}(t) \rangle \\ &+ \frac{3}{2} \int_{0}^{\tau} dt_{1} \int_{0}^{t_{1}} dt_{2} \operatorname{Re} \left\{ \langle \hat{C}_{1}^{H}(t_{1}) \hat{C}_{2}^{H}(t_{2}) \rangle \right\}, \end{split}$$



<u>Moments of work: Weakly</u> driven & coupled 2-level OQS





[F.W.J. Hekking and J.P. Pekola, PRL **111**, 093602 (2013)]



<u>Moments of work: Weakly</u> driven & coupled 2-level OQS









3. Nearly adiabatically driven OQS

[S. Suomela, J. Salmilehto, I.G. Savenko, T.A-N & M. Möttönen, Phys. Rev. E **91**, 022126 (2015)]



 Adiabatic renormalization of the time-dependent basis gives the Lindblad equation (Samu Suomela's talk on Thu.) (

$$\dot{\hat{\rho}}_{S} = -\frac{i}{\hbar} \left[\hat{H}_{S}, \hat{\rho}_{S} \right] \\ + \sum_{i=0}^{2} \left(\hat{L}_{(n,i)} \hat{\rho}_{S} \hat{L}_{(n,i)}^{\dagger} - \frac{1}{2} \left\{ \hat{L}_{(n,i)}^{\dagger} \hat{L}_{(n,i)}, \hat{\rho}_{S} \right\} \right)$$

$$\hat{L}_{(n,0)} = \sqrt{\Gamma_{(n,0)}} |g^{(n)}\rangle \langle e^{(n)}|, \quad \hat{L}_{(n,1)} = \sqrt{\Gamma_{(n,1)}} |e^{(n)}\rangle \langle g^{(n)}|,$$
$$\hat{L}_{(n,2)} = \sqrt{\Gamma_{(n,2)}} (|e^{(n)}\rangle \langle e^{(n)}| - |g^{(n)}\rangle \langle g^{(n)}|)$$

Diabatic basis (a) $|e\rangle$ **MSP** Group D lg> E $\hat{D}_1(t)$ -Adiabatic basis (b) ₹ $\Gamma_{(1,i)}(t)$ $g^{(1)}(t)$ E First super- $\hat{D}_2(t)$ adiabatic basis (c) $e^{(2)}(t)$ $T_{(2,i)}(t)$ $g^{(2)}$ E $\hat{D}_3(t)$ Tehran April 15th, 2015





The adiabatic renormalization gives the following master equation:

$$\begin{aligned} \dot{\hat{\rho}}_S &= -\frac{\imath}{\hbar} \left[\hat{H}_S, \hat{\rho}_S \right] \\ &+ \sum_{i=0}^2 \left(\hat{L}_{(n,i)} \hat{\rho}_S \hat{L}_{(n,i)}^{\dagger} - \frac{1}{2} \left\{ \hat{L}_{(n,i)}^{\dagger} \hat{L}_{(n,i)}, \hat{\rho}_S \right\} \right) \end{aligned}$$

$$\hat{L}_{(n,0)} = \sqrt{\Gamma_{(n,0)}} |g^{(n)}\rangle \langle e^{(n)}|, \quad \hat{L}_{(n,1)} = \sqrt{\Gamma_{(n,1)}} |e^{(n)}\rangle \langle g^{(n)}|,$$
$$\hat{L}_{(n,2)} = \sqrt{\Gamma_{(n,2)}} (|e^{(n)}\rangle \langle e^{(n)}| - |g^{(n)}\rangle \langle g^{(n)}|)$$





• Recall the IFT:
$$\left\langle \frac{Pr'(E(\tau) = E_{\tau})}{Pr(E(t_0) = E_0)} \prod_{j=1}^{N} \frac{\Gamma'_{m_j}(t_j)}{\Gamma_{m_j}(t_j)} \right\rangle = 1$$





• Recall the IFT:
$$\left\langle \frac{Pr'(E(\tau) = E_{\tau})}{Pr(E(t_0) = E_0)} \prod_{j=1}^{N} \frac{\Gamma'_{m_j}(t_j)}{\Gamma_{m_j}(t_j)} \right\rangle = 1$$

$$\langle e^{-\beta W^{(n)}} \rangle_{(n)} = e^{-\beta \Delta F}$$









Single Electron Box (SEB)





Electrons are driven one by one from lead to island and *stochastically* undergo transitions



SEB

ENERGY / E_C

n = 1

O

n

 $n_g = C_g V_g / e$

 $H = E_C (n - n_q)^2$

Single Electron Box (SEB)



Dissipation in single-electron transitions

Heat generated in a tunneling event *i*:

 $Q_i = \pm 2E_C(n_{g,i} - 1/2)$

Total heat generated in a process:

$$Q = \sum_{i} Q_{i}$$

Work in a process:

$$W = Q + \Delta U$$

 \uparrow
Change in internal
(charging) energy

[D. Averin and J. Pekola, EPL 96, 67004 (2011)]

n = 0



SEB in Equilibrium: Experimental Results



[O.-P. Saira et al., PRL 109, 180601 (2012); J.V. Koski et al., Nat. Phys. 9, 644 (2013)]







Analytic and Numerical Results for Overheated SEB



 $n_g(t)$ β + $\Delta\beta(C,Q)$ Analytically assume single (double) jump trajectory, expand tunneling rates in $\Delta\beta$: $\frac{k_B}{4C}\beta^2 \langle Q \rangle^2 + \mathcal{O}[(\Delta\beta)^2]$ $\langle e^{-\beta(W - \Delta F)} \rangle = 1 -$ 30 1.00 20(b) $\langle e^{-\beta(W-\Delta F)} \rangle$ 10 0.98 -0.4₹ -0.35Monte Carlo: 10^{8} ∡ 0.96 • 10¹¹ repetitions ₹ ⋠ 10^{4} (a) • 8000 time steps 0.94 • 95 % single 10⁰ 0.50 jumps 0.92 0.04 0.06 0.08 0.10 0.120.00 0.02 $\frac{\Delta T}{T}$







For driven, open quantum systems it's possible to derive fluctuation relations for work in the TMP (Lindblad) setting

The power operator naturally appears in the (exact) expressions for the moments of the work distribution

Main collaborators here:

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Paolo Solinas (Genova) Joachim Ankerhold (Ulm) Ivan Savenko (Aalto) Mikko Möttönen (Aalto)

Juha Salmilehto (Yale) Frank Hekking (Lorraine)