Thermodynamic bound on the asymmetry of cross-correlations

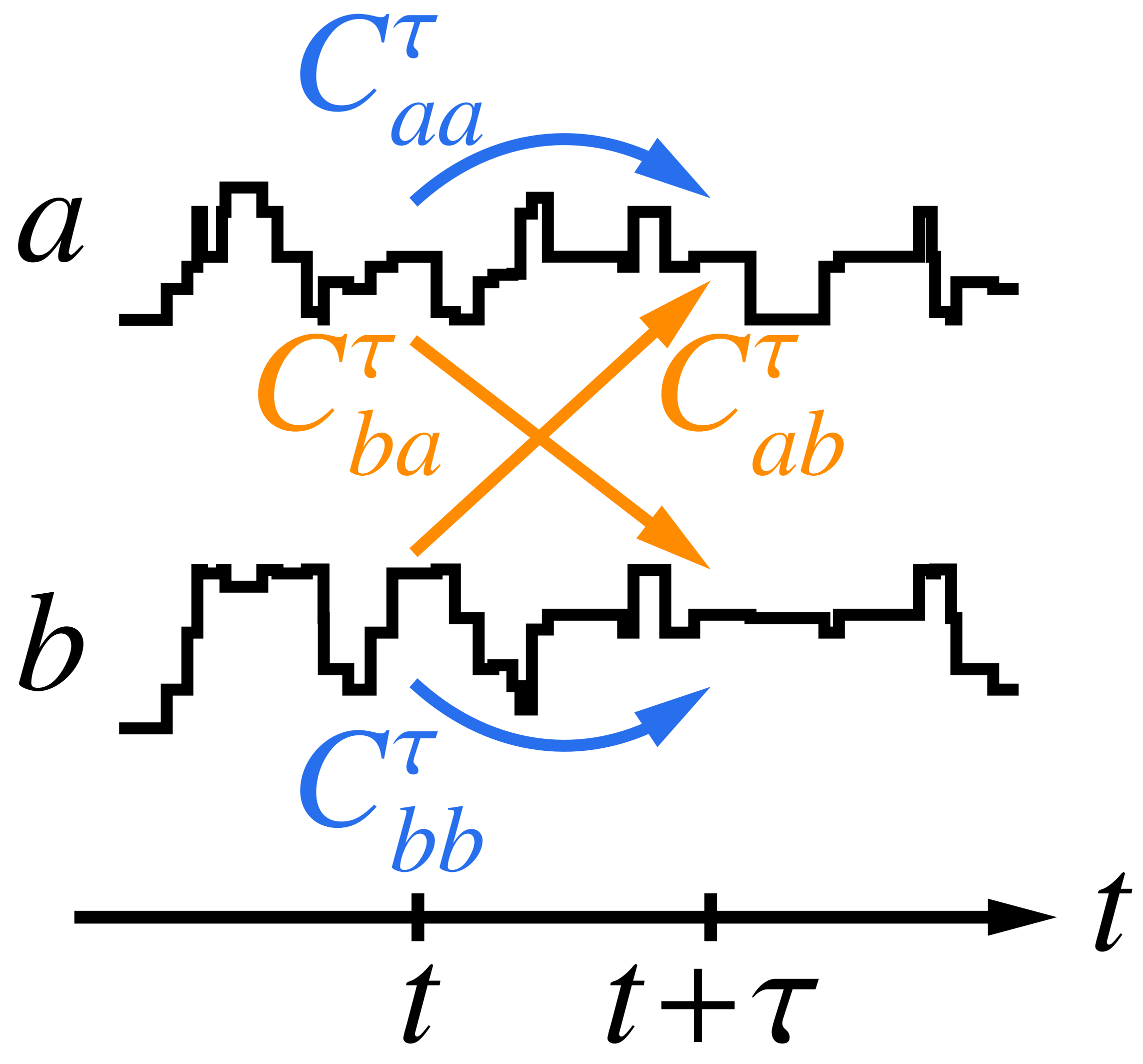
Naruo Ohga, Sosuke Ito, Artemy Kolchinsky, arXiv:2303.13116,
(Phys. Rev. Lett., in press.)

Ito group, The University of Tokyo
Naruo Ohga

Aug. 4, 2023 YITP-YSF symposium

Two-time correlations

Fluctuating system in steady state



Steady-state trajectory keeps fluctuating

a, b : Arbitrary two observables
(Even under time-reversal)

Two-time correlations

$$C^{\tau}_{ba} = \langle b(t+\tau)\, a(t) \rangle$$

C^{τ}_{aa}, C^{τ}_{bb} : Auto-correlation

C^{τ}_{ba}, C^{τ}_{ab} : Cross-correlation

- Captures the temporal structure at the trajectory level
- Experimentally accessible in various systems

Two noneq. features of two-time correlations

1 Asymmetry of cross-correlations

2 Oscillations

☞ 1 Asymmetry of cross-correlations

2 Oscillations (Eigenvalues)

Discussion & Summary

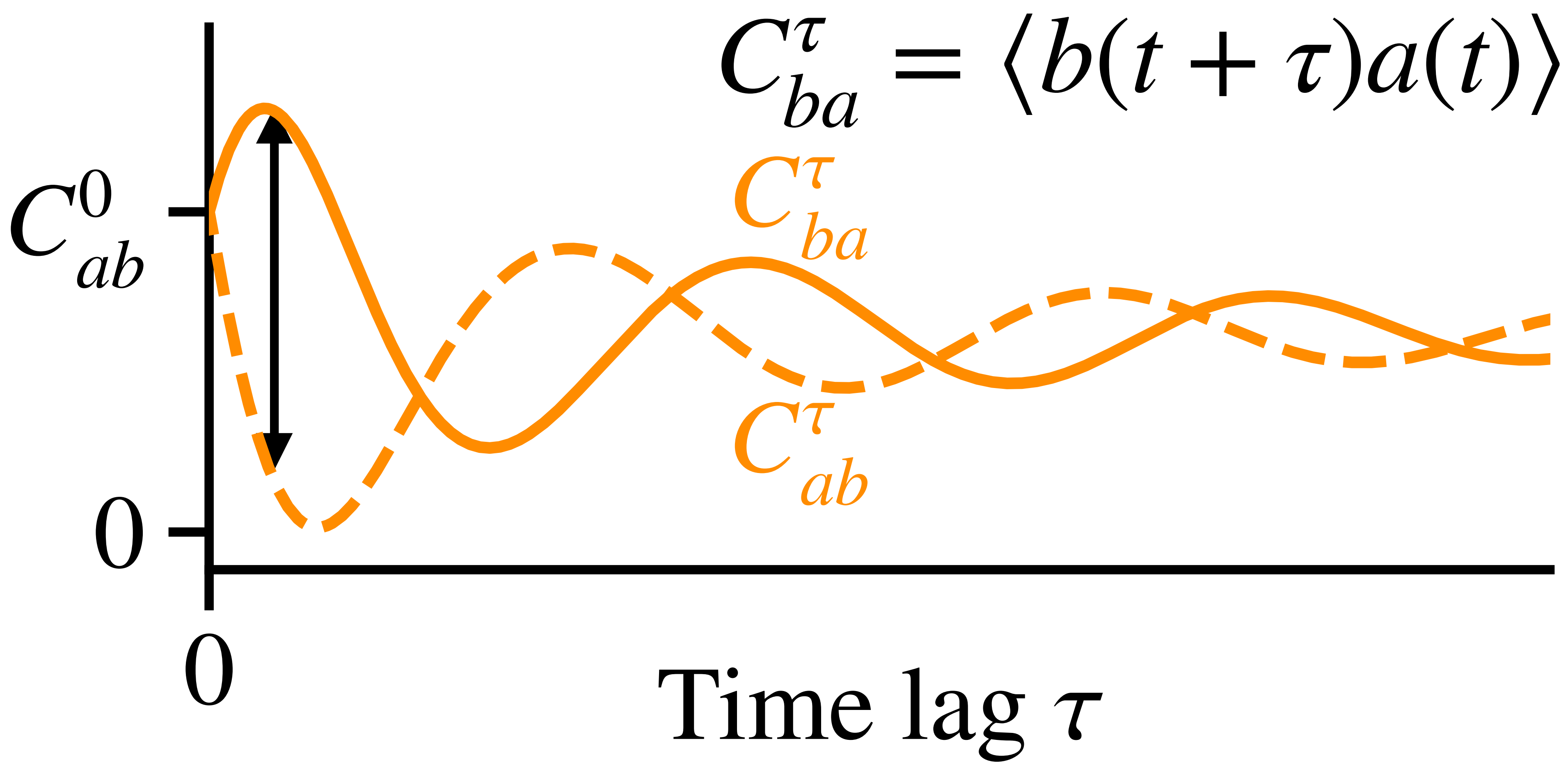
Asymmetry in cross-correlations

Equilibrium $C^\tau_{ba} = C^\tau_{ab}$ for any a, b

(**Microscopic reversibility**)

Noneq. Steady state $C^\tau_{ba} \neq C^\tau_{ab}$ for some a, b

(Breaking of micro. rev.)



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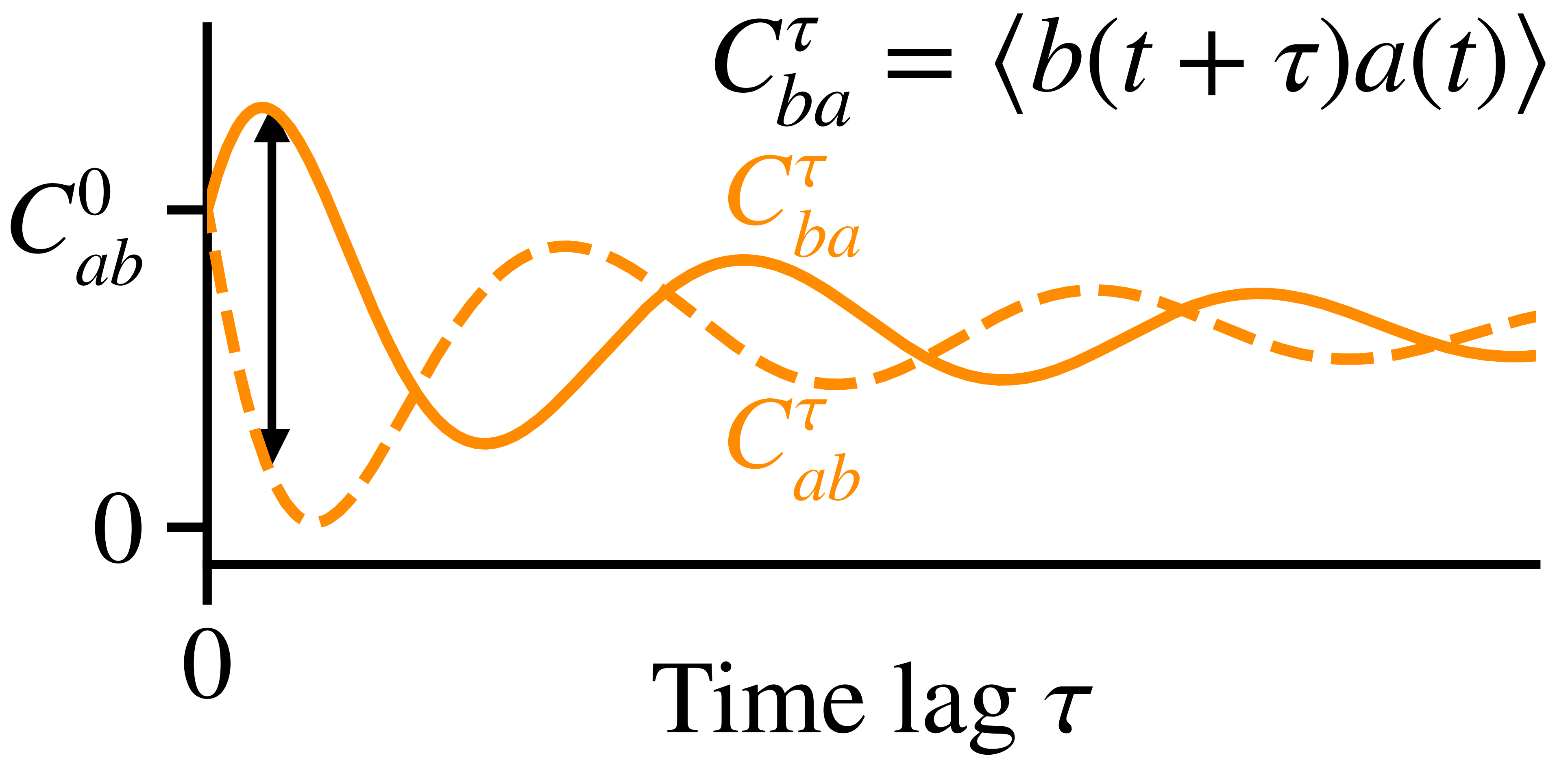
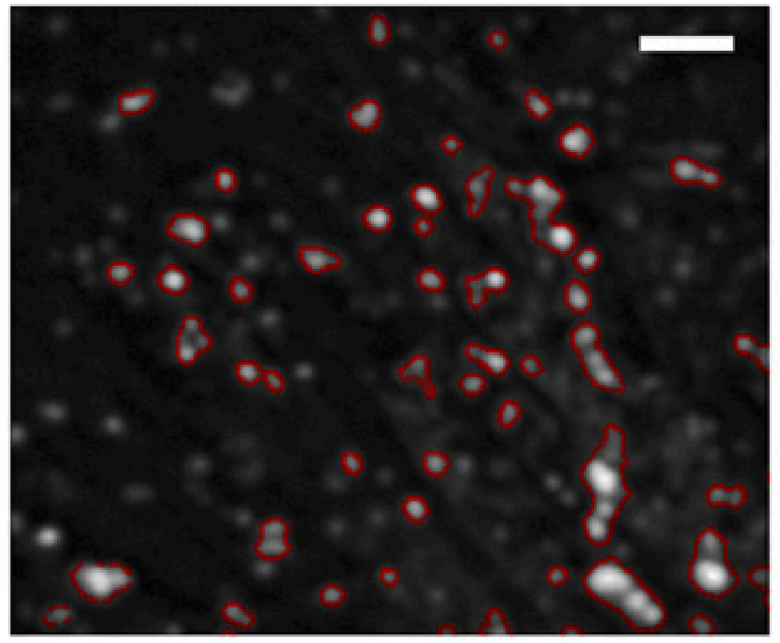


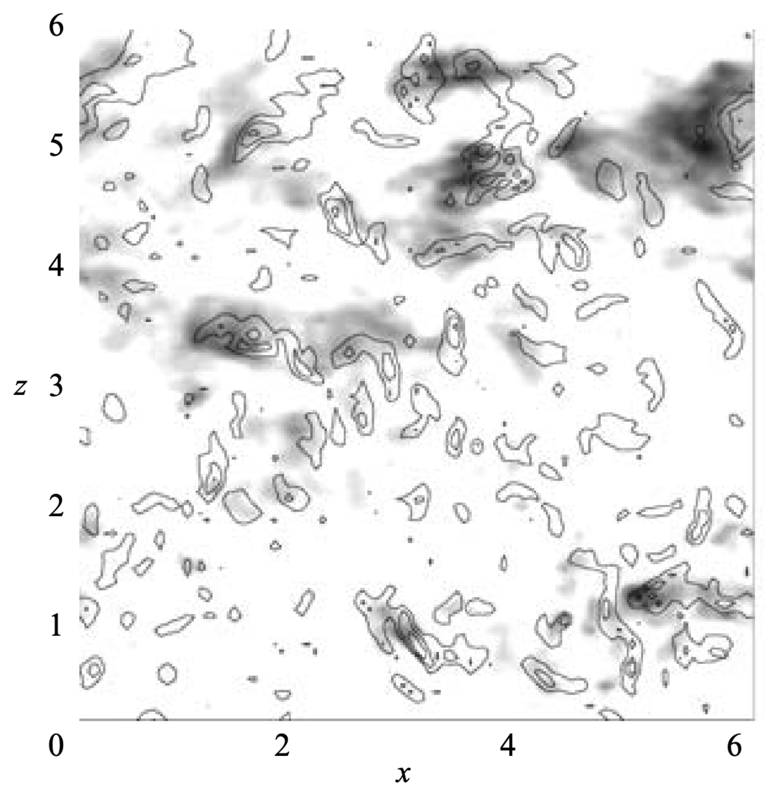
$C_{ba}^{\tau} \neq C_{ab}^{\tau}$: fundamental signature of nonequilibrium

Biochemical reaction



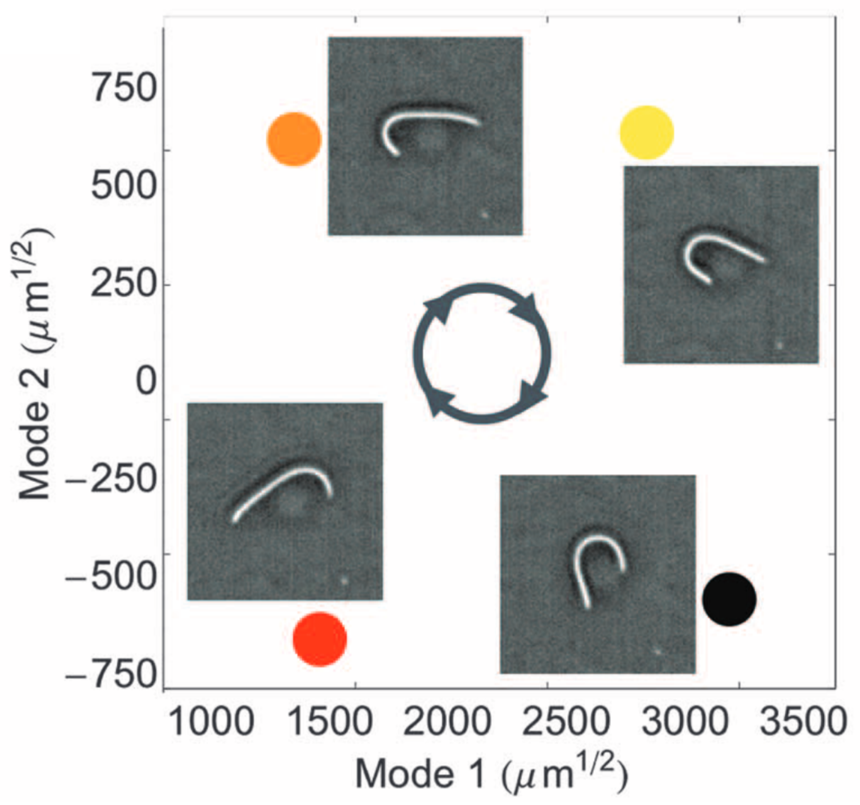
D. Sisan+ Biophys J. 2010

Turbulence



A. Jachens+, J. Fluid. Mech. 2006

Biological motion

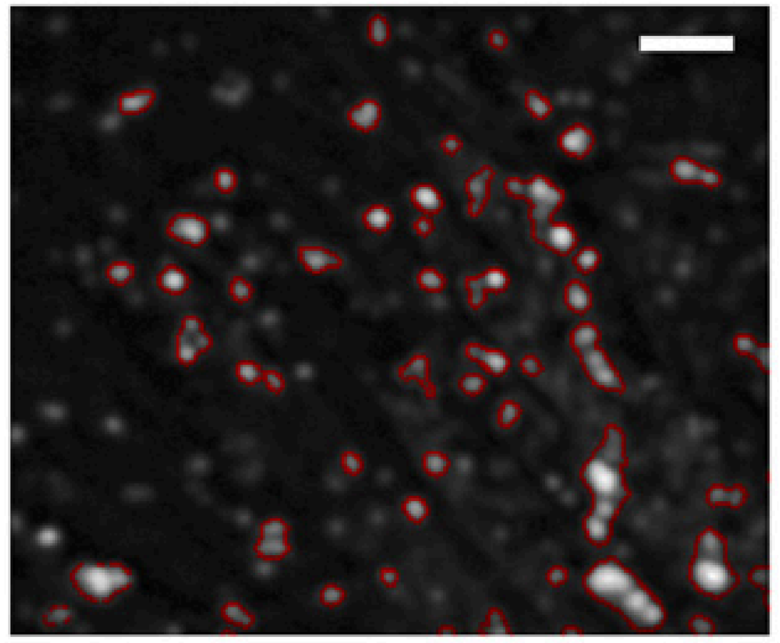


C. Battle+ Science 2016

Goal

Asymmetry $C^{\tau}_{ba} \neq C^{\tau}_{ab}$:

Biochemical reaction



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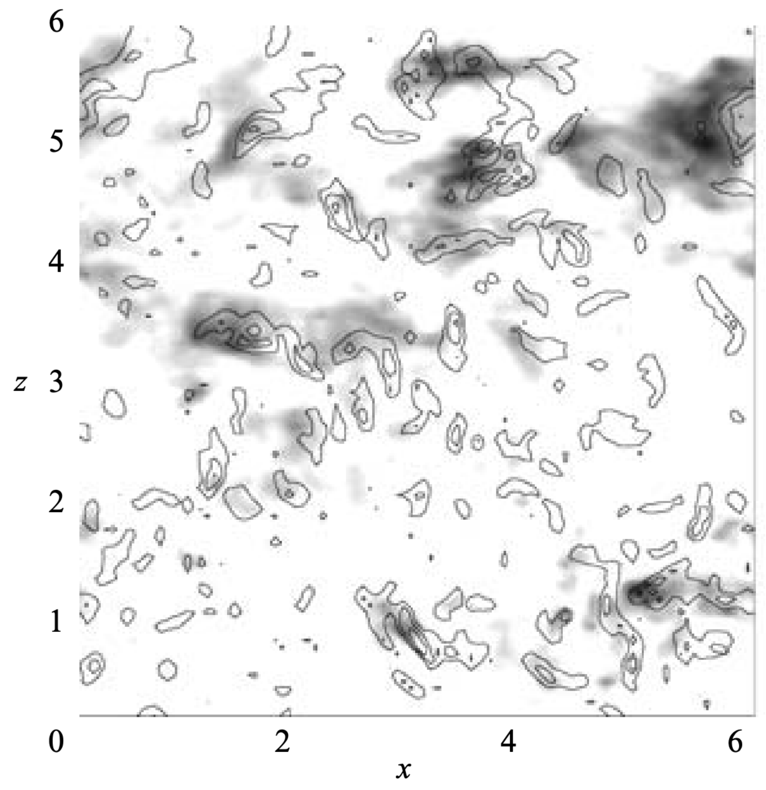

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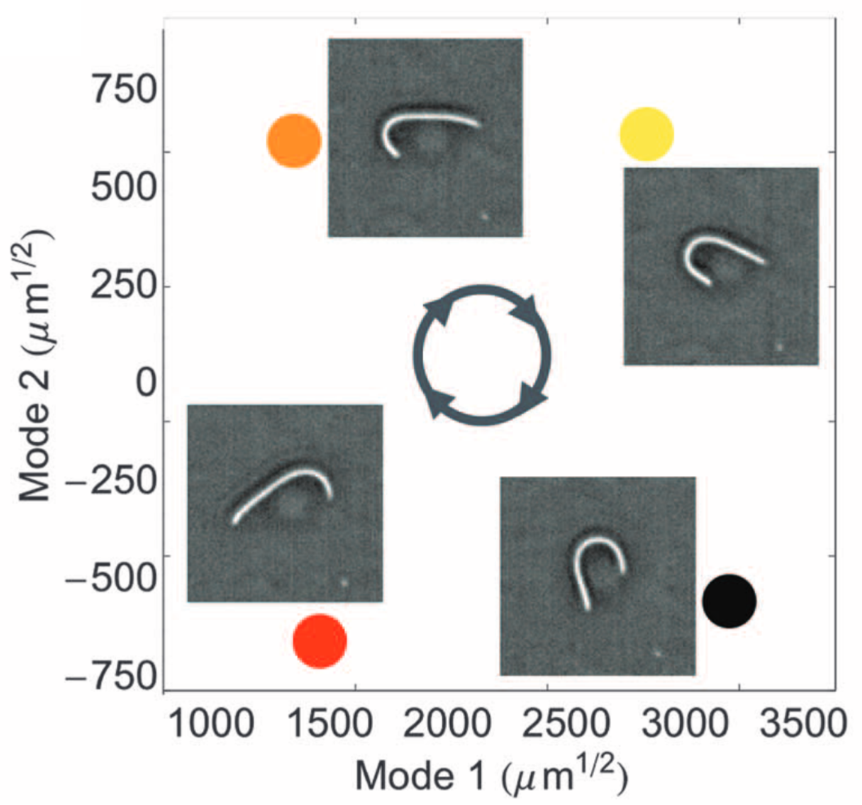

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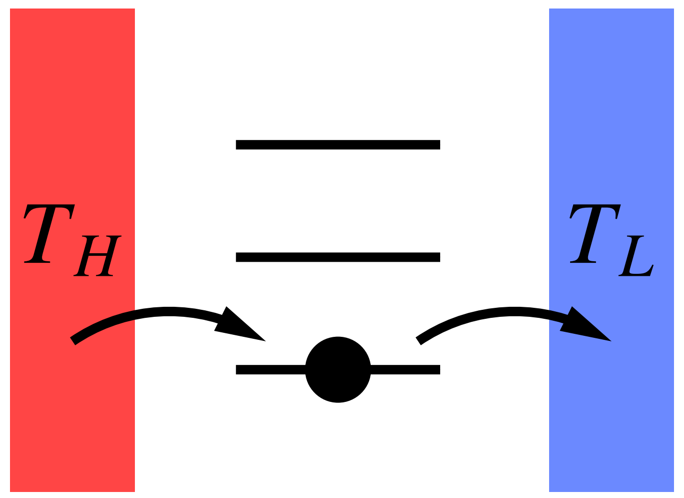
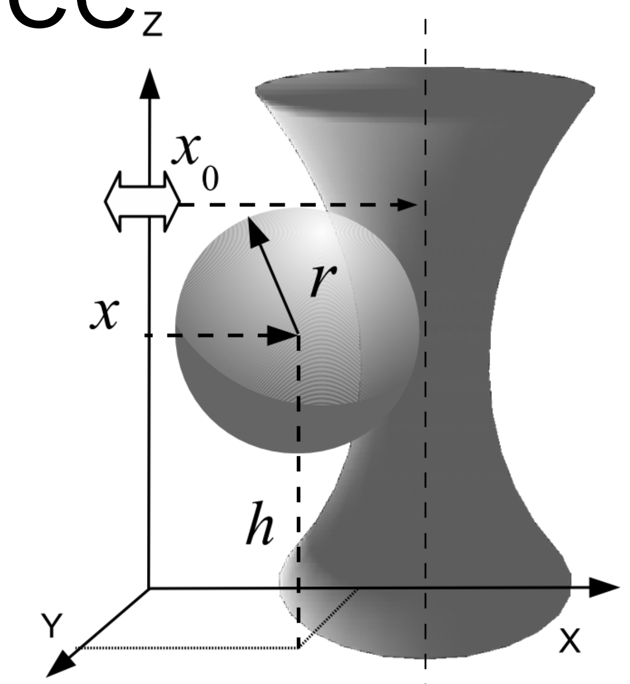
Any universal & **quantitative** relation?

Thermodynamic driving

Temperature gradient

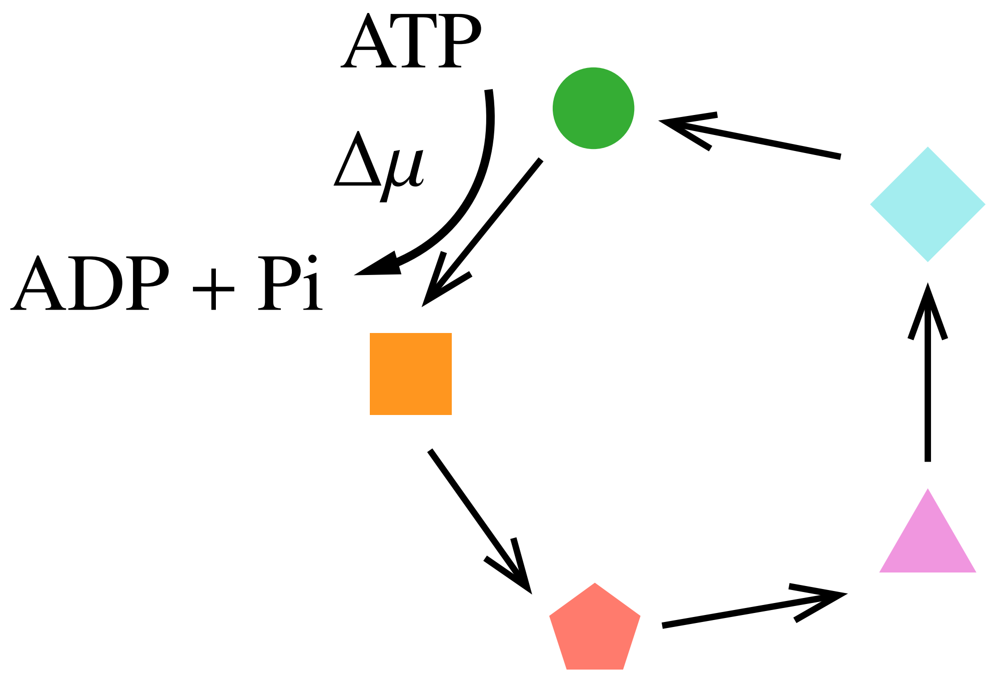


Mechanical force



J. R. Gomez-Solano+, EPL 2010

Chemical potential gradient



Thermodynamic bound on cross correlations

Dimensionless measure of the asymmetry of cross-correlation (New in this study)

Asymmetry of cross-corr.

$$\chi_{ba} = \lim_{\tau\to 0} \frac{C^\tau_{ba} - C^\tau_{ab}}{2\sqrt{(\Delta_\tau C_{aa})(\Delta_\tau C_{bb})}}$$

Decay of auto-corr.

$$C^\tau_{ba} = \langle b(t+\tau)\, a(t)\rangle$$

$$\Delta_\tau C_{aa} = C^0_{aa} - C^\tau_{aa} = \frac{1}{2}\langle [a(t+\tau) - a(t)]^2\rangle$$

(The change speed of a)

Invariant under rescaling of a, b and time. Experimentally accessible

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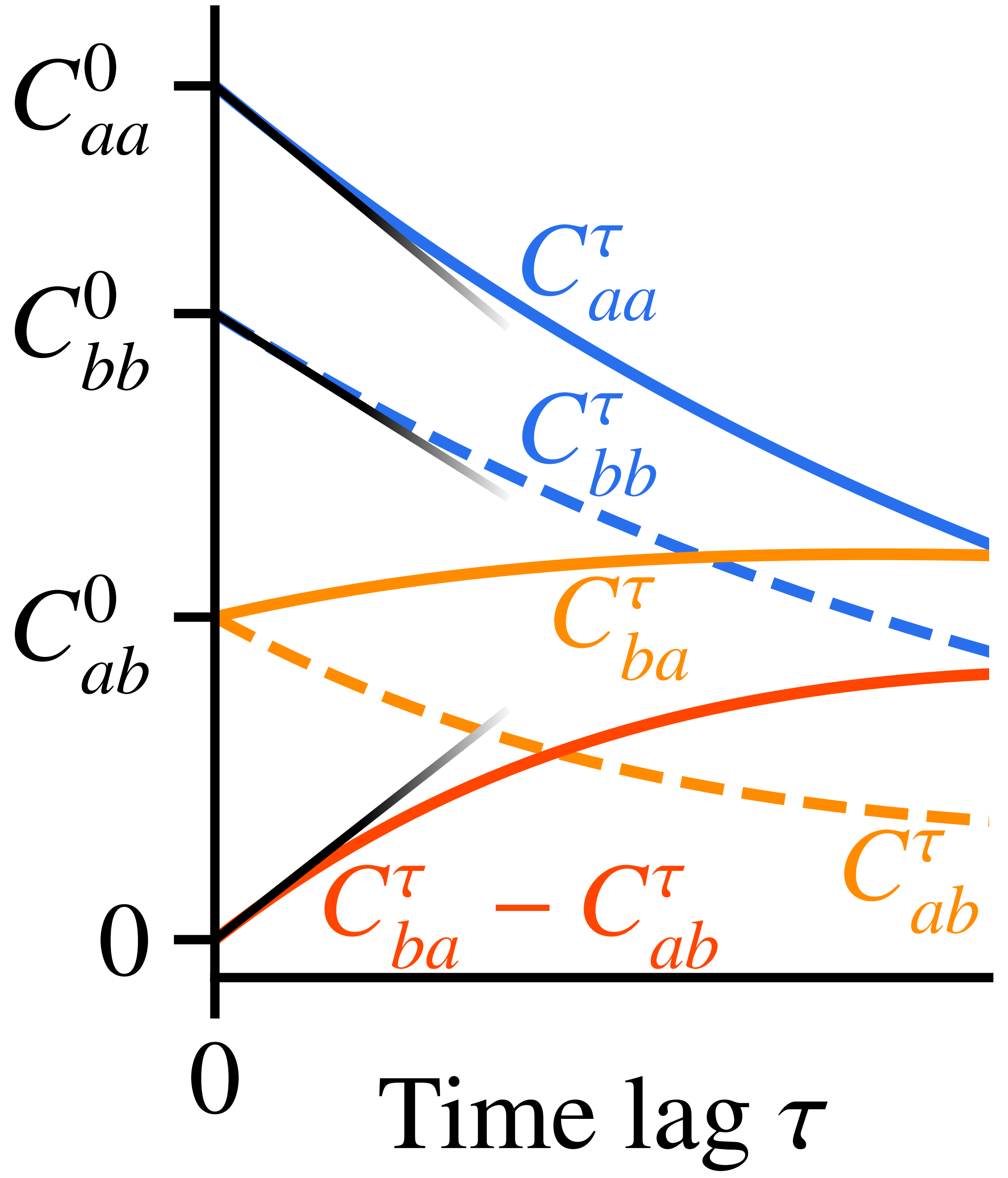

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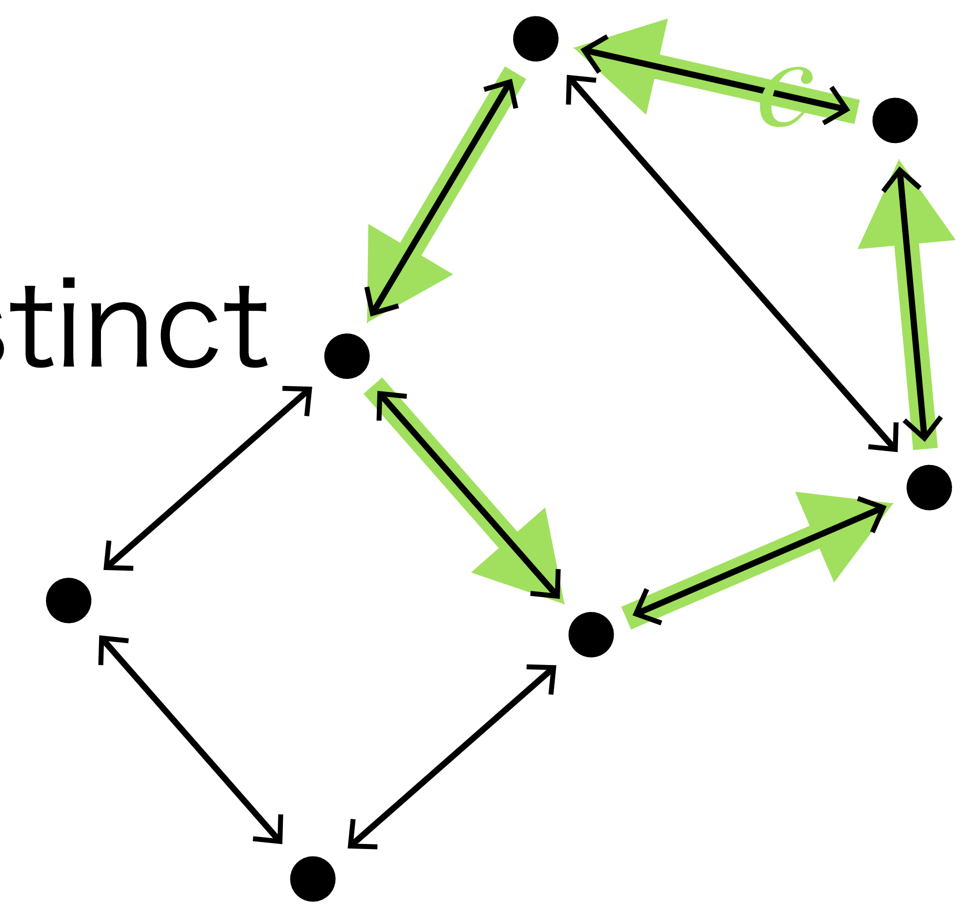
(The change speed of a)

Invariant under rescaling of a, b and time. Experimentally accessible

Strength of thermodynamic driving (Standard in discrete systems)

Cycle c

Cyclic sequence of distinct states connected with allowed transitions



Cycle affinity $\mathcal{F}_c$

= The sum of thermodynamic forces over one turn of the cycle

= Dissipation in the environment per one turn of the cycle

Determined by environmental parameters

Thermodynamic bound on cross correlations

Dimensionless measure of asymmetry

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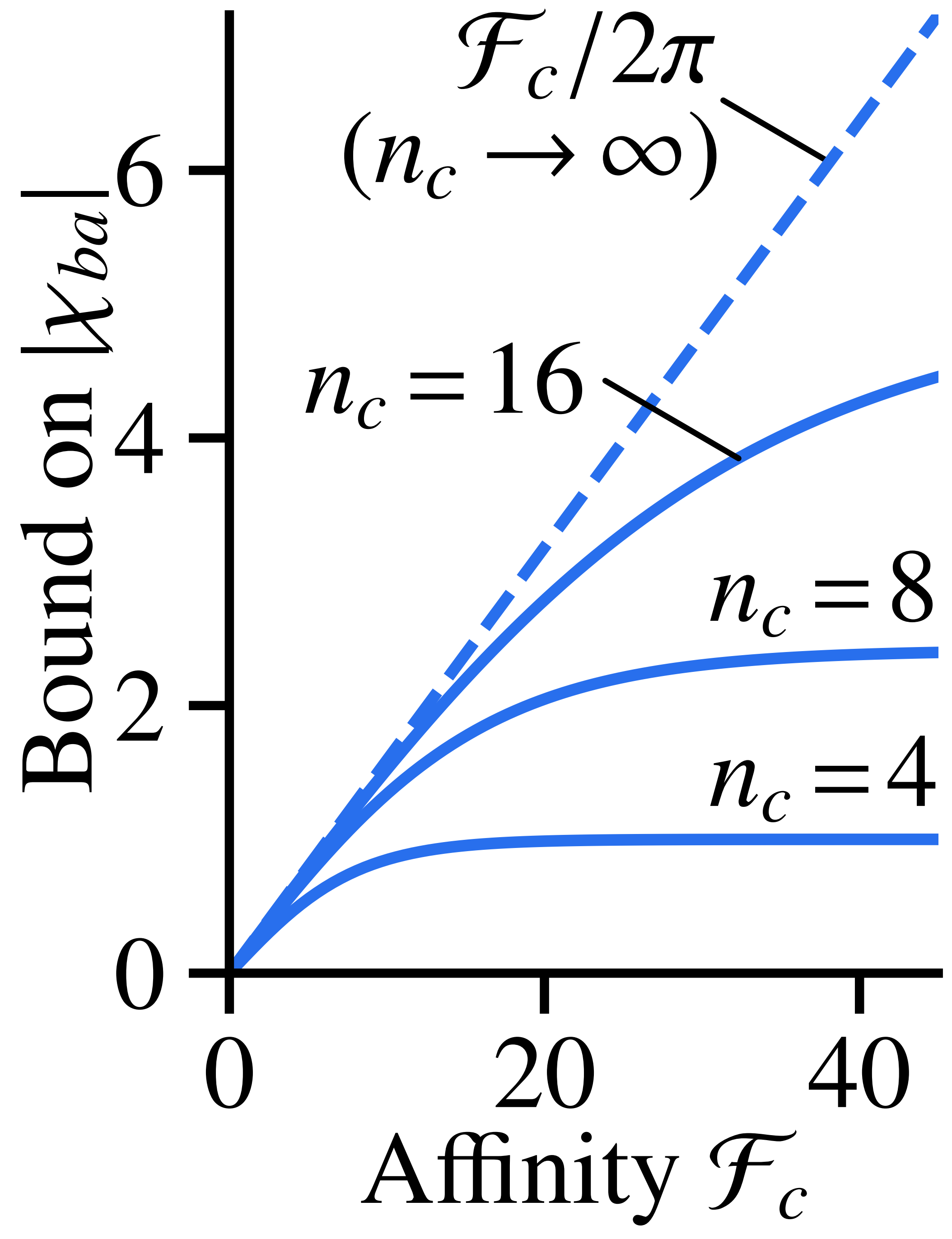
Main Result For any observable a, b

$$|\chi_{ba}| \le \max_c \frac{\tanh(\mathcal{F}_c/2n_c)}{\tan(\pi/n_c)} \le \max_c \frac{\mathcal{F}_c}{2\pi}$$



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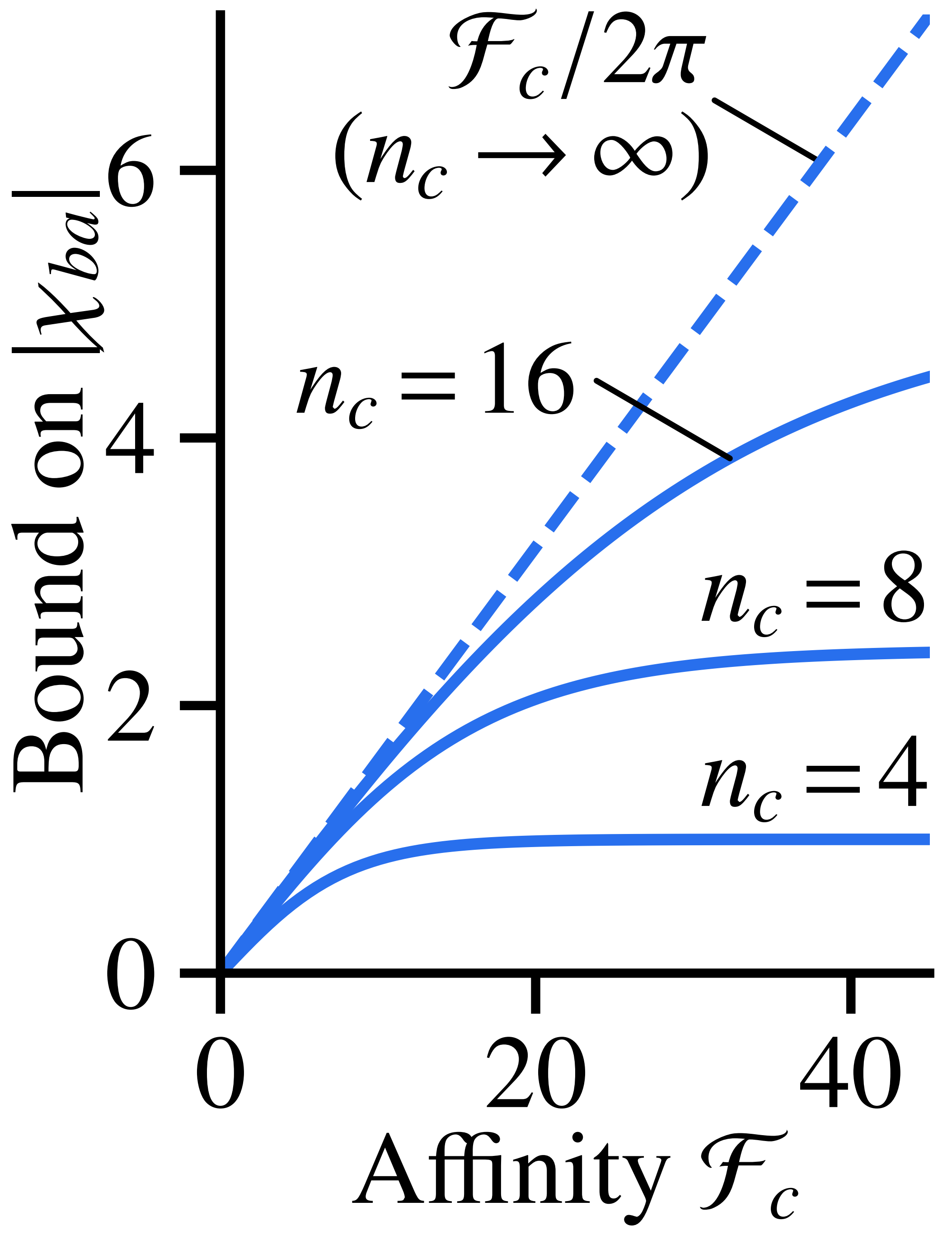
Maximum over all cycles in the system

Nonlinear function of

$\mathcal{F}_c$: Cycle affinity

n_c : # of states over the cycle



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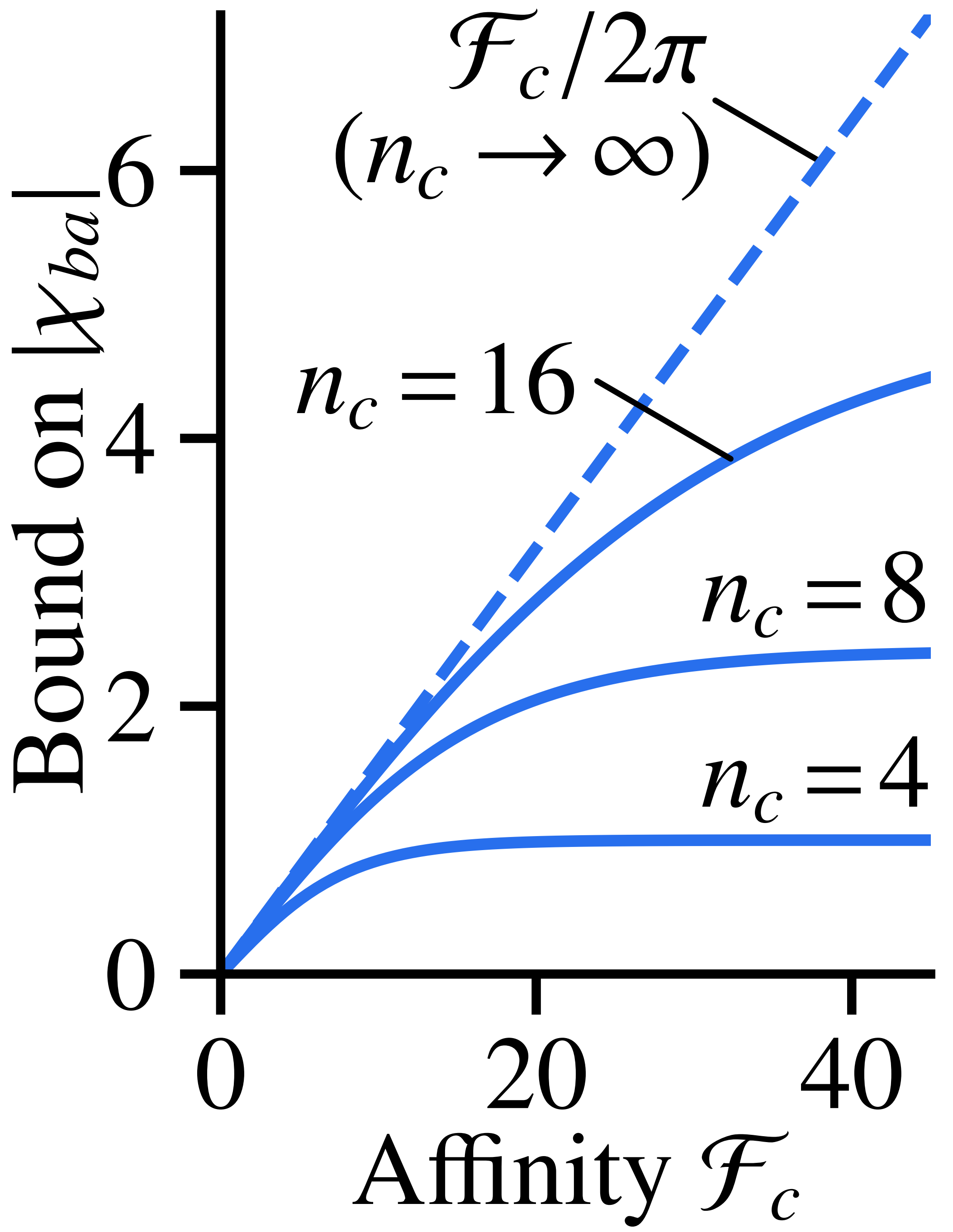
$\tanh x \le x$

$\tan x \ge x$

Linear function of

$\mathcal{F}_c$: cycle affinity



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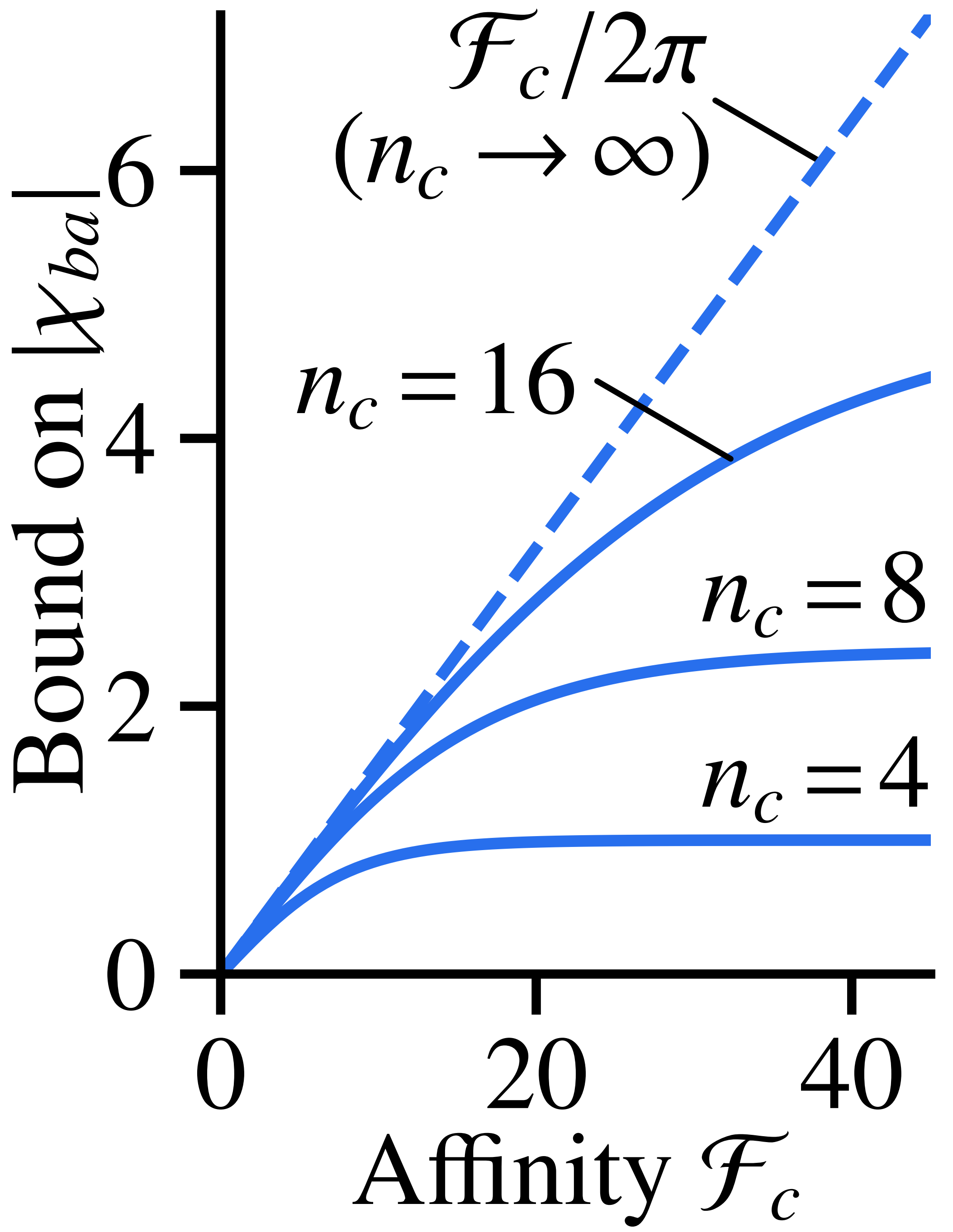
The magnitude of asymmetry is universally related to affinity!



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Implications

Dimensionless measure of asymmetry

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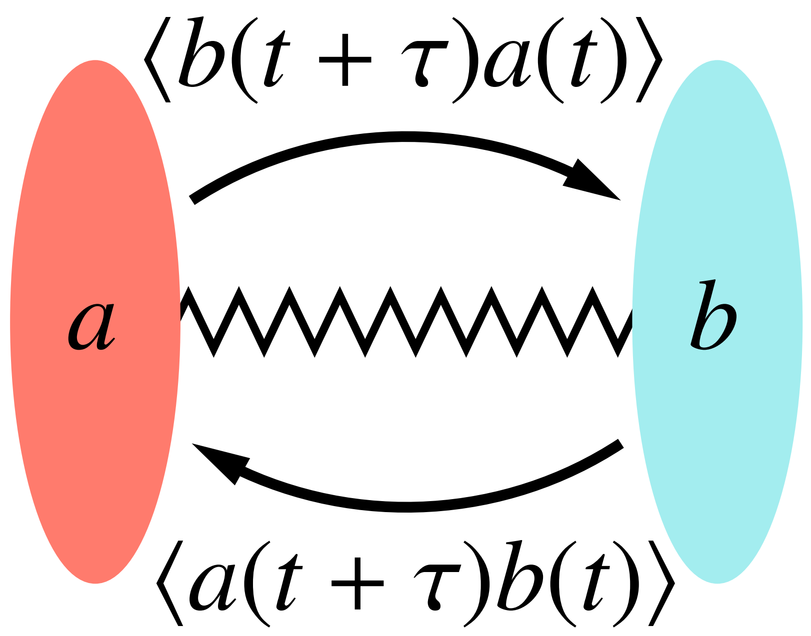
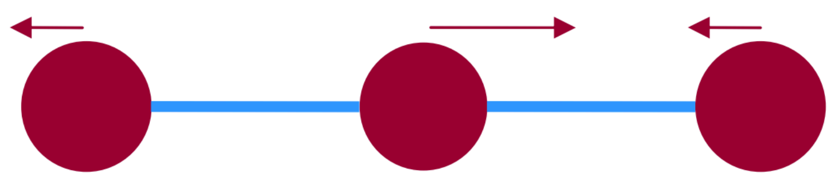
① Fundamental thermodynamic cost for various physical functions

Information transfer

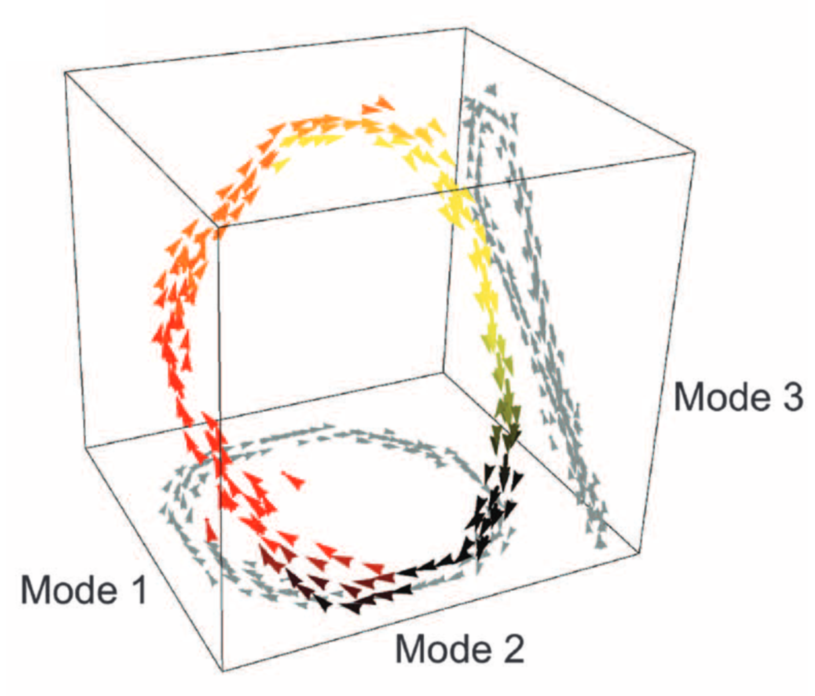


Nonreciprocal motion



R. Golestanian+
PRE 2008

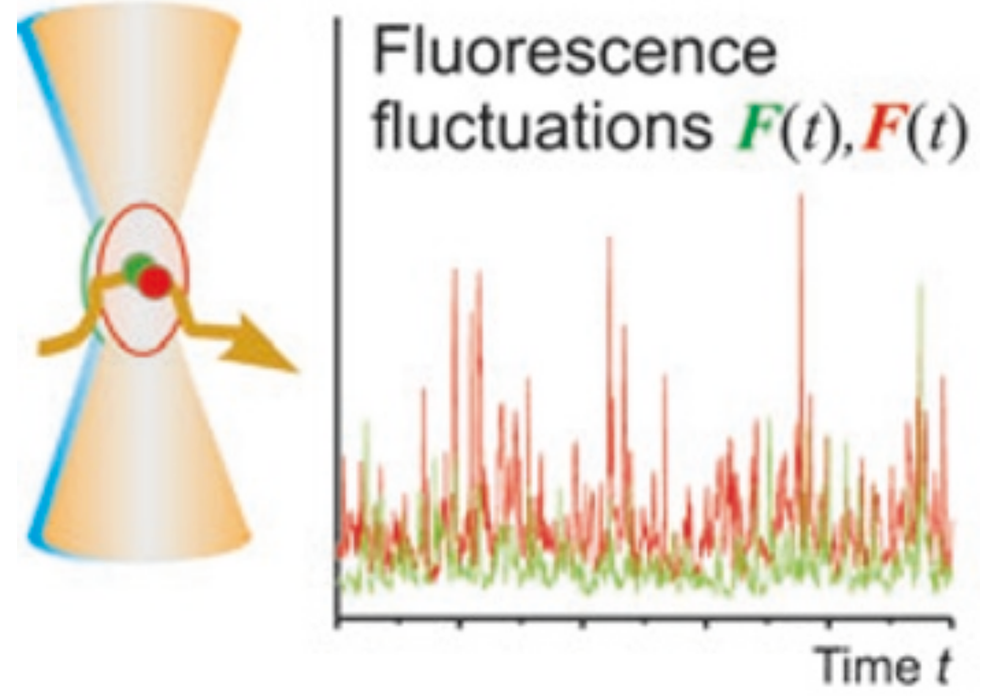
Circulation



C. Battle+
Science 2016

② A practical method to infer affinity from measured short-time correlations

e.g.)
Fluorescence cross-correlation spectroscopy



K. Bacia+, Nat. Methods 2011

Numerical example

5:30

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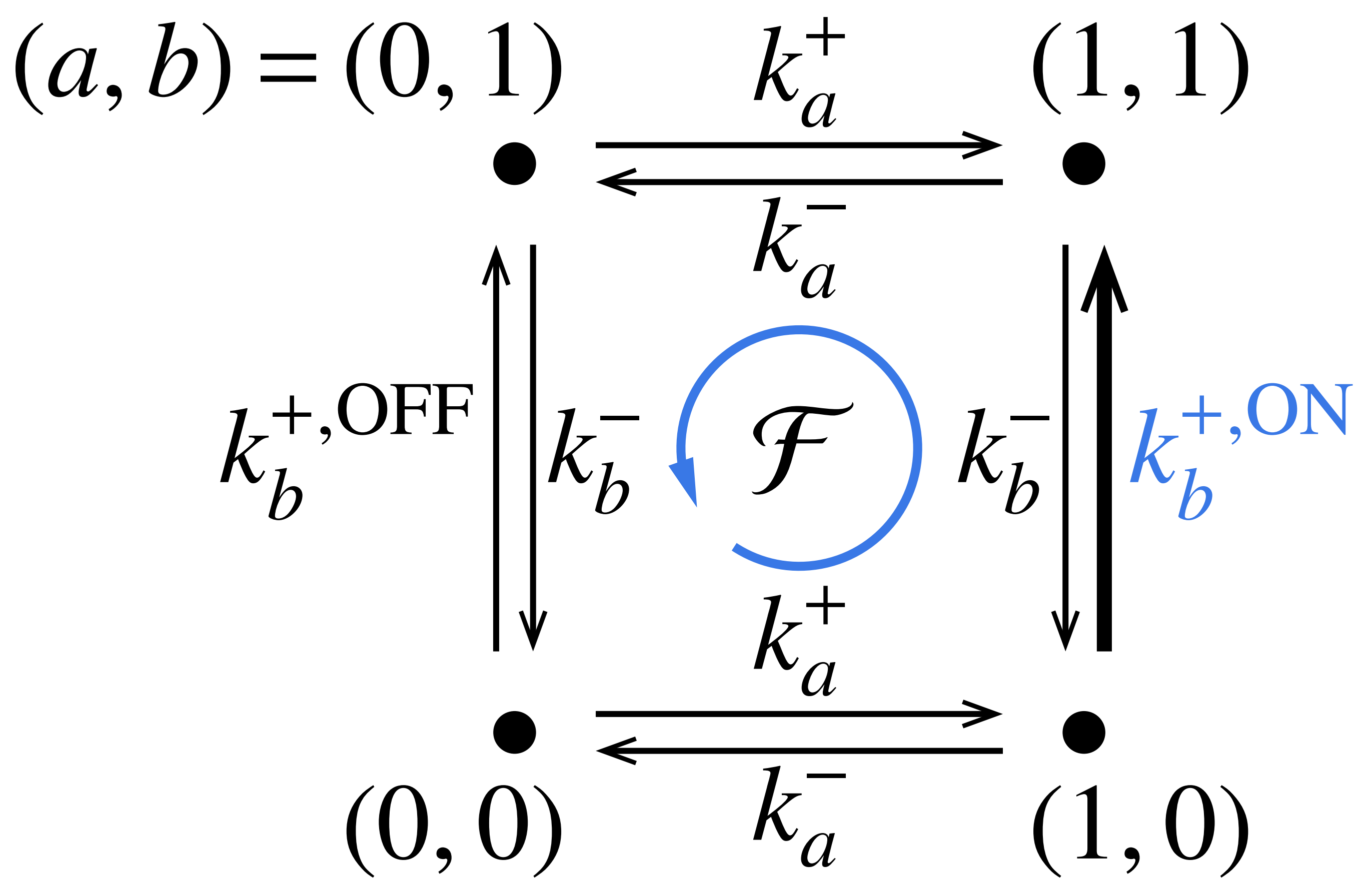
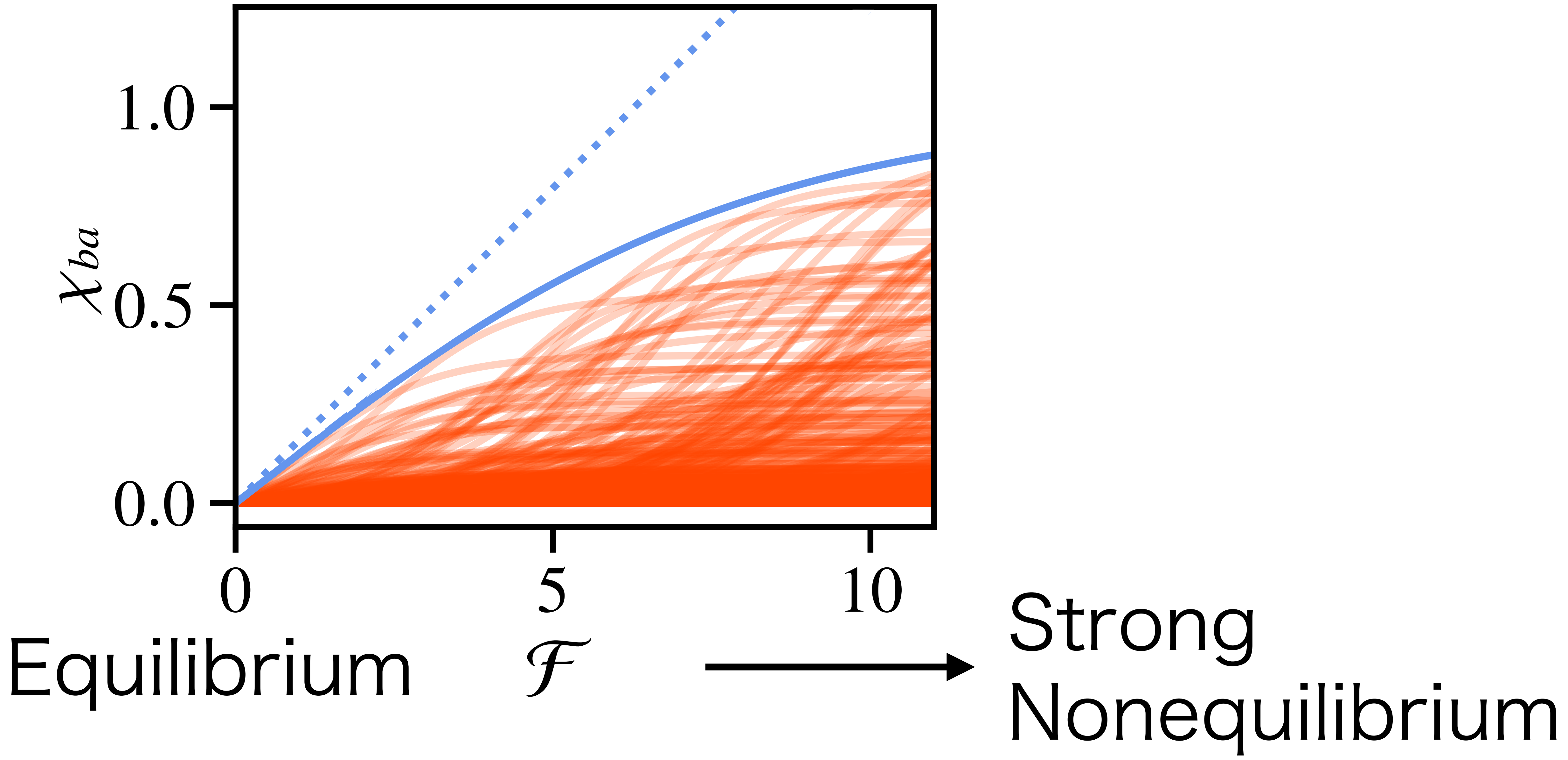
Model of biological information transduction

P. Mehta and D. J. Schwab, PNAS 2012.



Numerical example with random rates



Formulation and proof

Discrete-state Markov process

States $i = 1, \ldots, N$

Transition matrix R

$$\begin{cases} R_{ij} = \text{Transition rate from } j \text{ to } i \quad (i \neq j) \\ R_{ii} = -\sum_{j:j\neq i} R_{ji} \qquad \text{(Escape rate)} \end{cases}$$

Time evolution

$$d\boldsymbol{p}(t)/dt = \mathsf{R}\boldsymbol{p}(t) \implies \boldsymbol{p}(t) = e^{\mathsf{R}t}\boldsymbol{p}(0)$$

Steady-state distribution $\boldsymbol{q}$: $\mathsf{R}\boldsymbol{q} = 0$

Two-time correlation

$$C_{ba}^{\tau} = \langle b(\tau)a(0)\rangle = \sum_{ij} [e^{\mathsf{R}\tau}]_{ij} q_j b_i a_j$$

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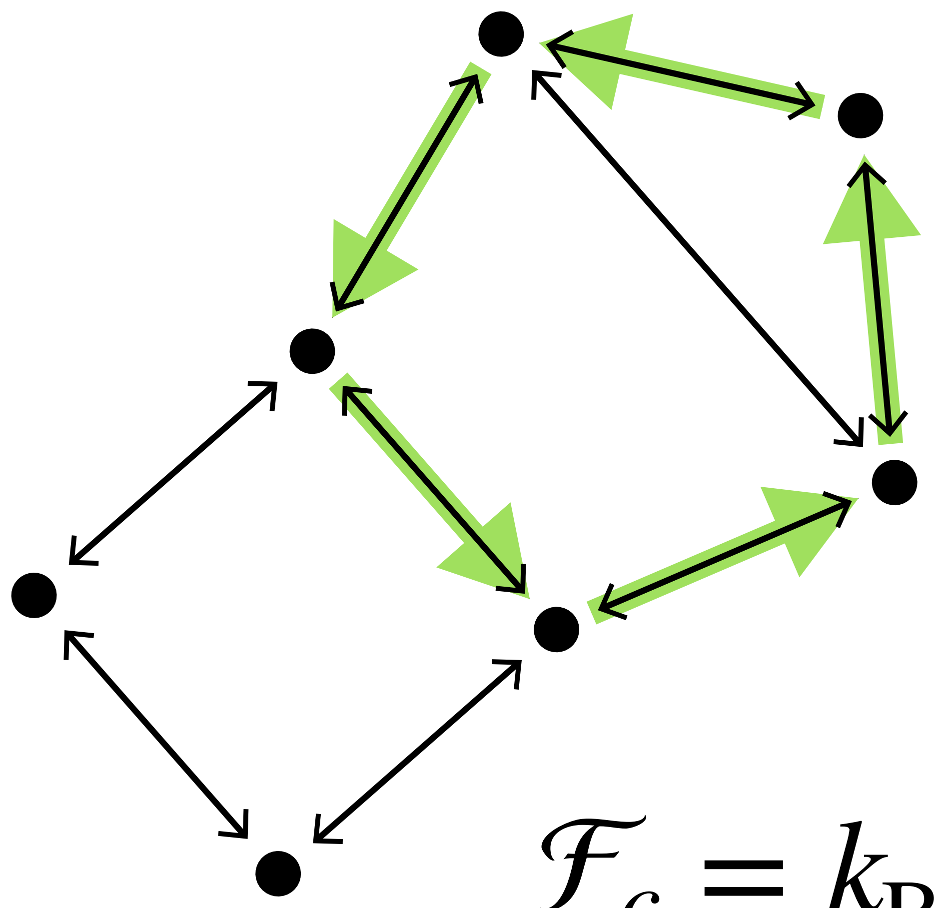
Cycle $c = (i_1 \to i_2 \to \cdots \to i_{n_c} \to i_1)$

with $R_{i_{k+1}\, i_k} \neq 0$



$$\mathcal{F}_c = k_\mathrm{B} \ln \frac{\prod \text{Forward rates}}{\prod \text{Backward rates}}$$

$$= k_\mathrm{B} \ln \frac{R_{i_2 i_1} R_{i_3 i_2} \ldots R_{i_1 i_{n_c}}}{R_{i_1 i_2} R_{i_2 i_3} \ldots R_{i_{n_c} i_1}}$$

Formulation and proof

Two-time correlation

$$C_{ba}^{\tau} = \langle b(\tau)a(0)\rangle = \sum_{ij} [e^{\mathsf{R}\tau}]_{ij} q_j\, b_i a_j$$

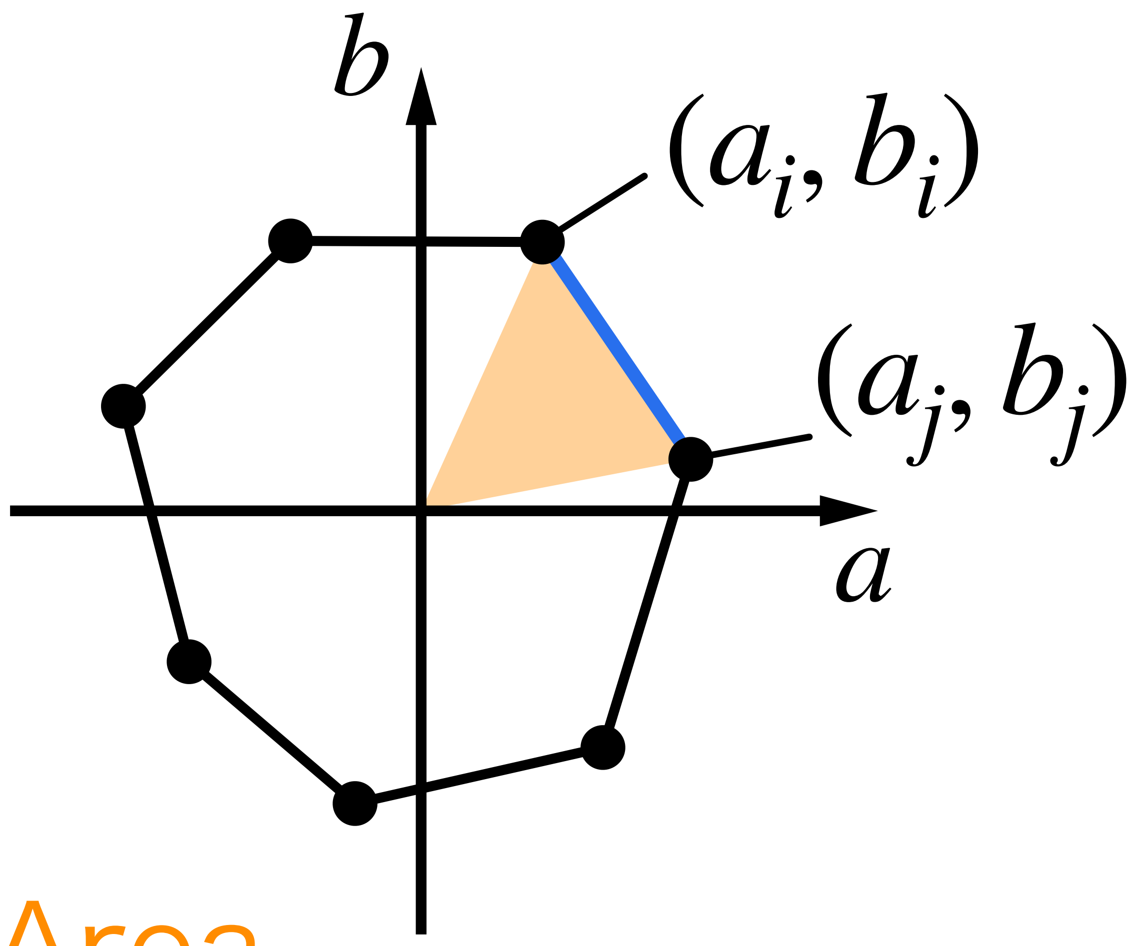
$\boldsymbol{q}$: Steady-state distribution

Proof

Use $[e^{\mathsf{R}\tau}]_{ij} \approx 1 + \tau R_{ij}$

$$\chi_{ba} = \lim_{\tau\to 0} \frac{C_{ba}^{\tau} - C_{ab}^{\tau}}{2\sqrt{\Delta_\tau C_{aa}\, \Delta_\tau C_{bb}}}$$

$$= \frac{2\sum_{ij} R_{ij} q_j \overbrace{(b_i a_j - a_i b_j)}^{\text{Area}}}{\sum_{ij} R_{ij} q_j \underbrace{\left[(a_i - a_j)^2 + (b_i - b_j)^2\right]}_{\text{Length}}}$$



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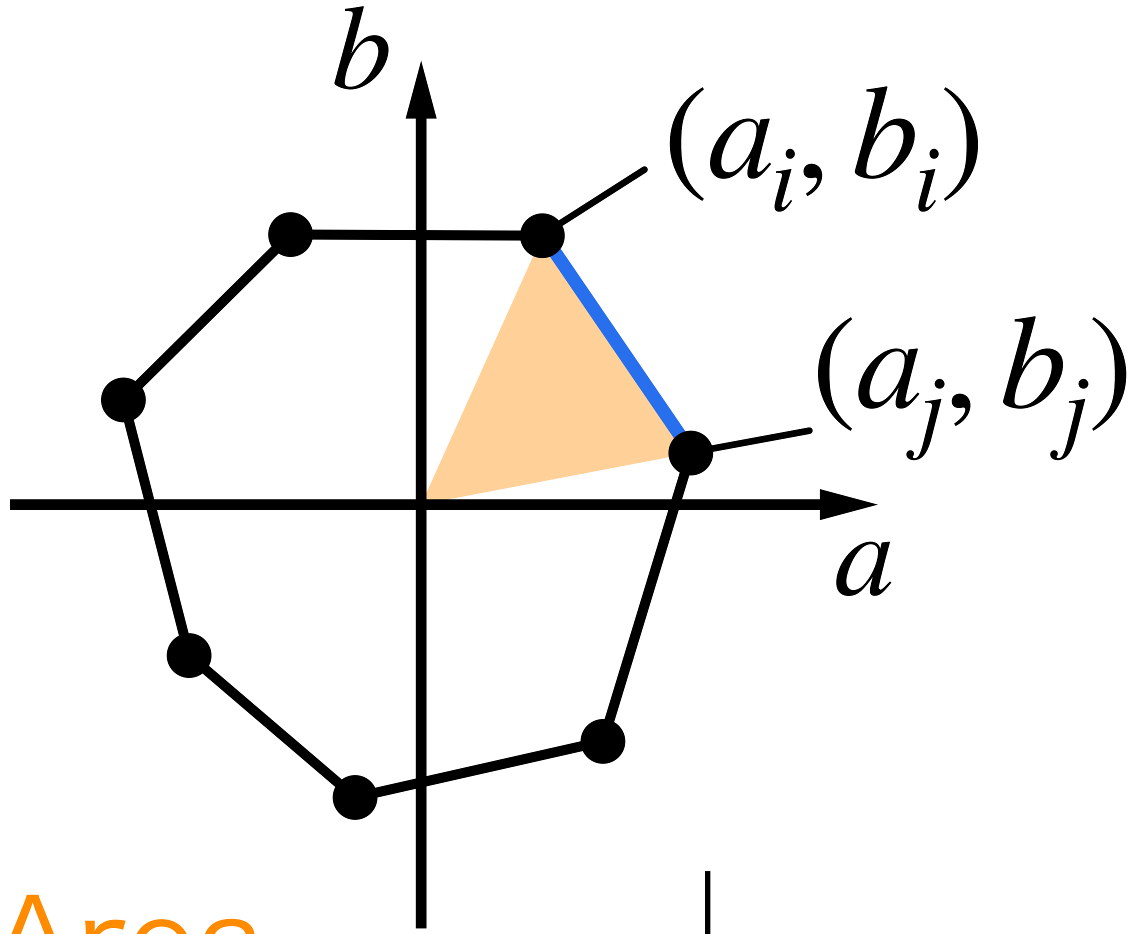
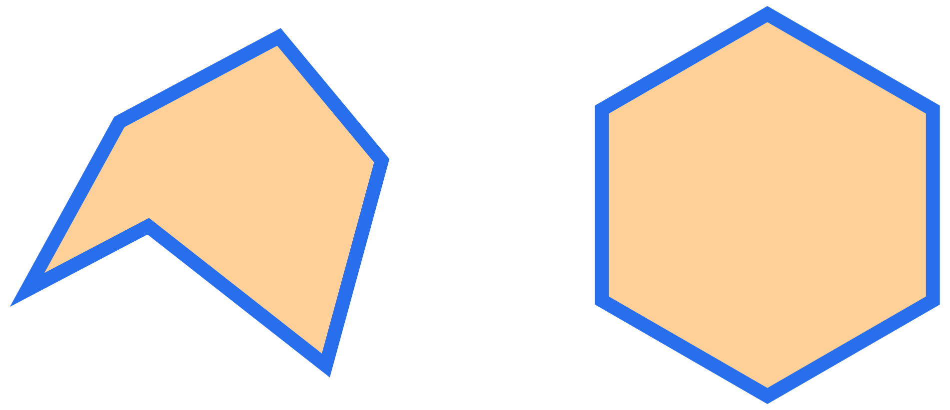


① Isoperimetric inequality
(from planer geometry)

For any n-sided polygon,



$$(\text{Area}) \le \underbrace{\frac{(\text{Perimeter})^2}{4n\tan(\pi/n)}}$$

Area of the regular n-sided polygon with the same perimeter.

② Cycle-wise affinity TUR

$$\Longrightarrow \boxed{|\chi_{ba}| \le \max_c \frac{\tanh(\mathcal{F}_c/2n_c)}{\tan(\pi/n_c)} \le \max_c \frac{\mathcal{F}_c}{2\pi}}$$

1 Asymmetry of cross-correlations

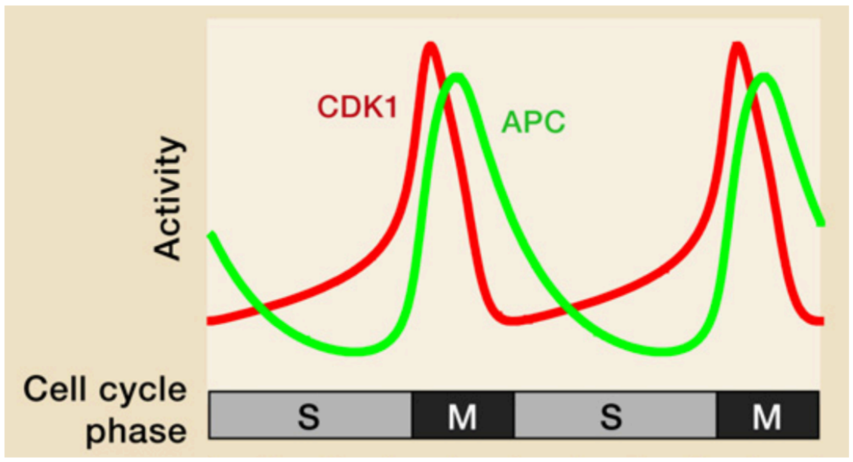
☞ 2 Oscillations (Eigenvalues)

Discussion & Summary

Fluctuating oscillations

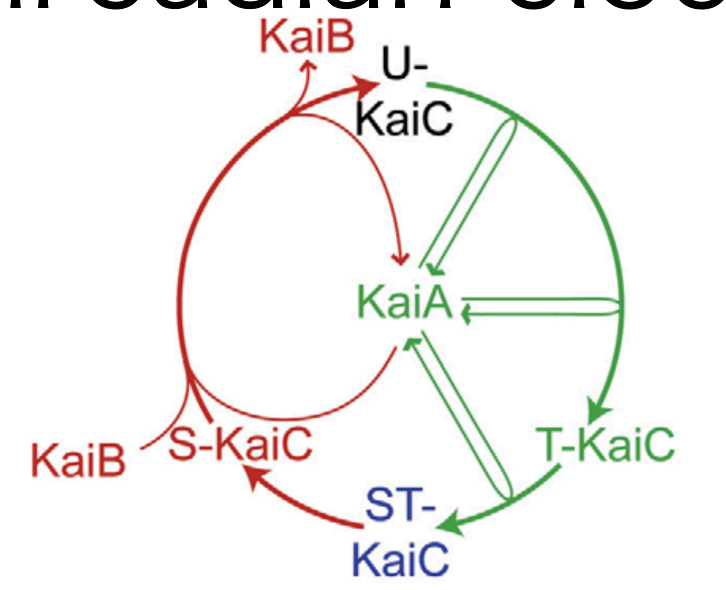
e.g.) Biochemical oscillations

Cell cycle

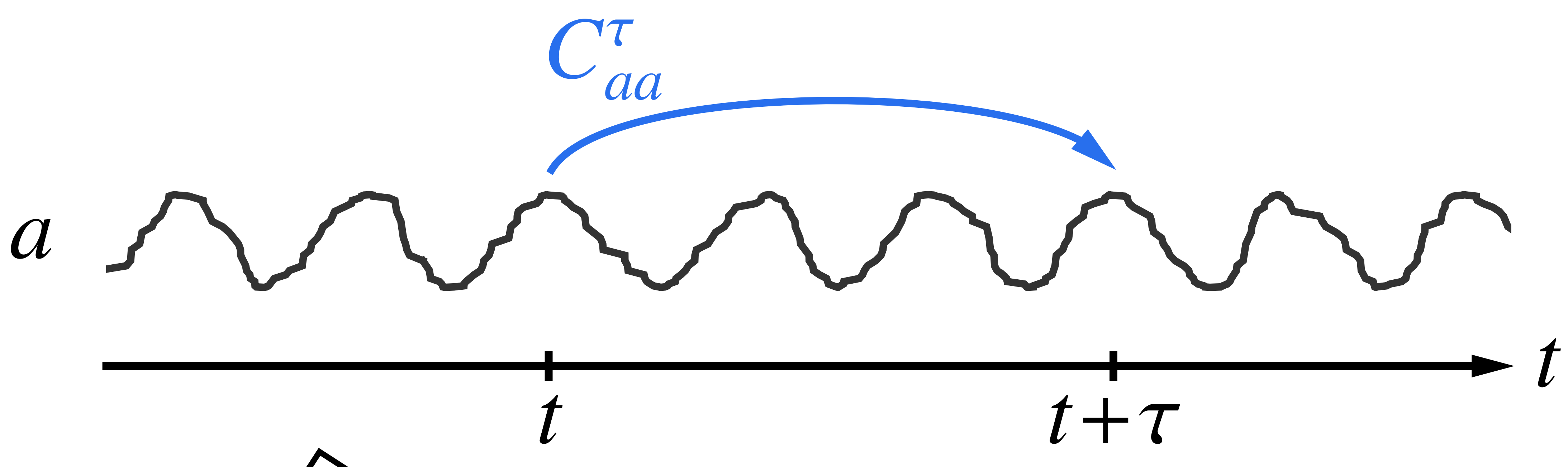


Ferrell+ Cell 2011

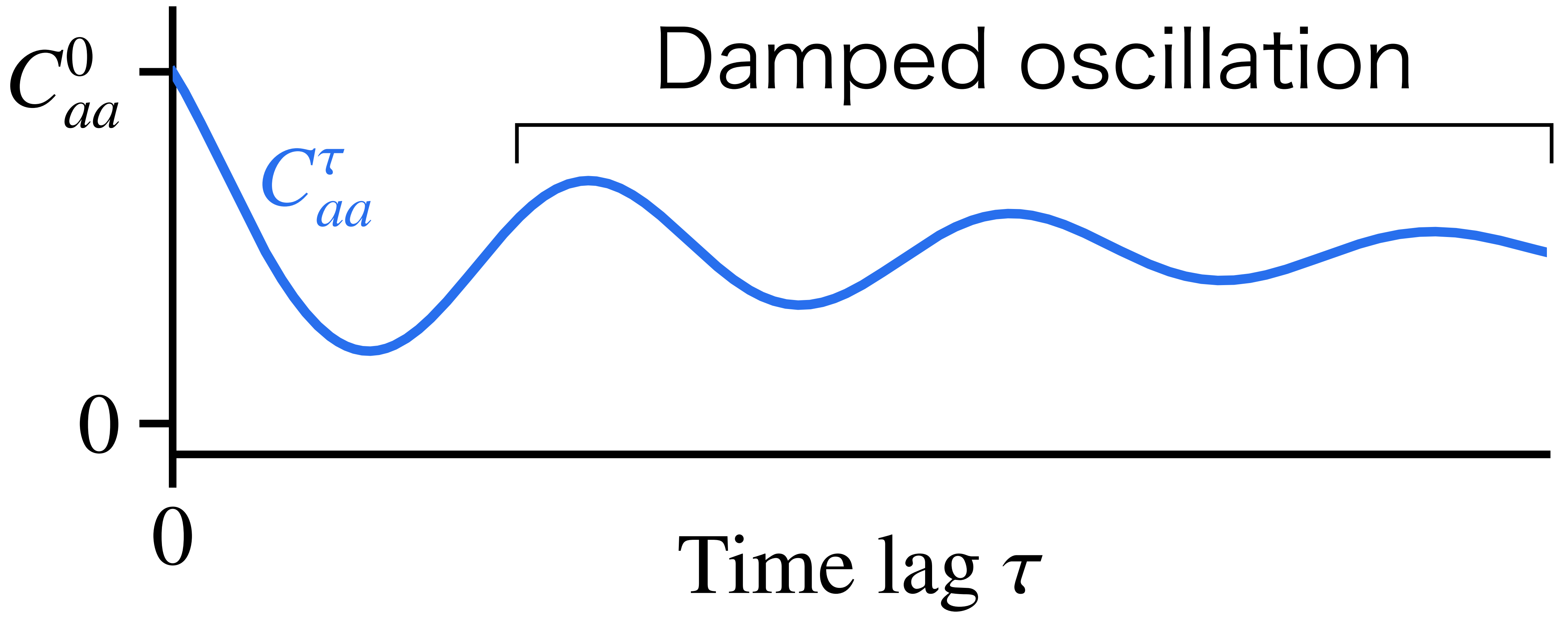
Circadian clock



Dong+ Curr. Opin. Microbiol. 2008



Oscillations should be coherent in time for reliable biochemical functionality.



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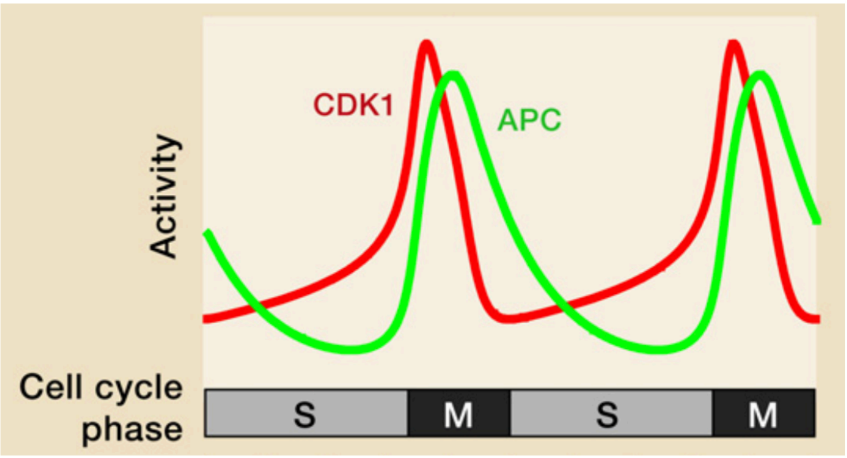

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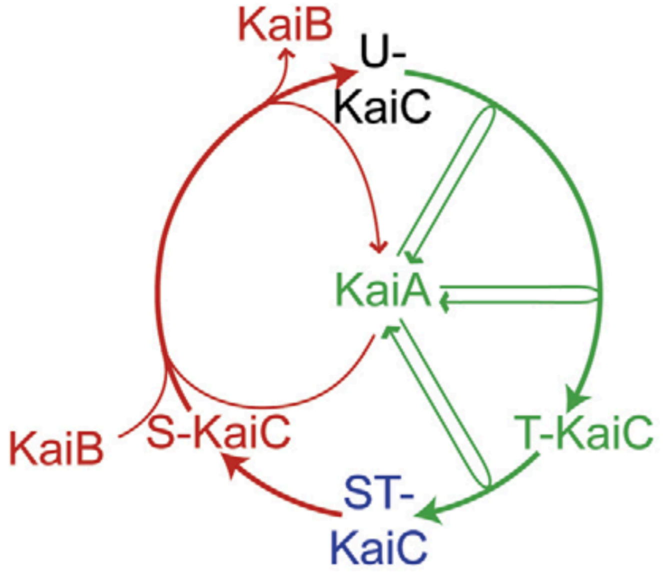

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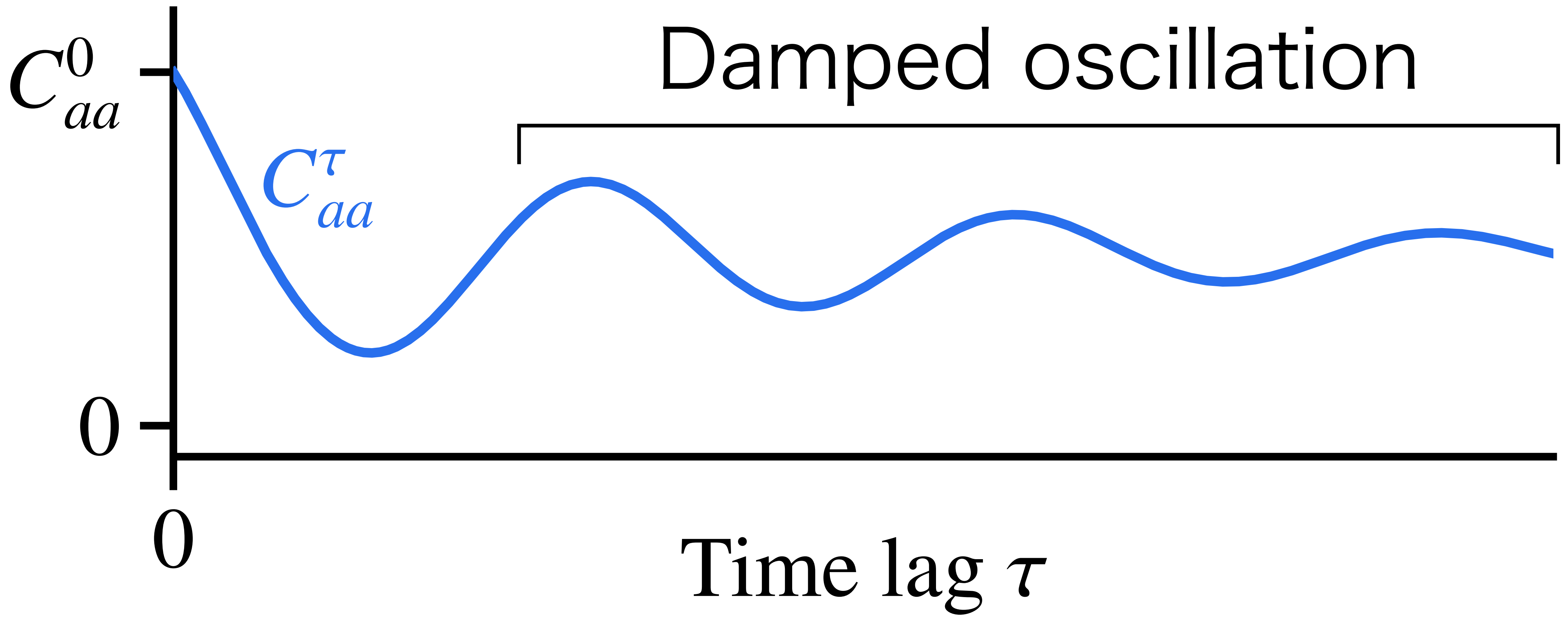


Finite-time corr. $C_{ba}^{\tau} = \sum_{ij} [e^{\mathrm{R}\tau}]_{ij}\, q_j\, b_i a_j$

$\lambda_\alpha = -\mu_\alpha + \mathrm{i}\omega_\alpha$: α-th eigenvalue of R

$\rightarrow$ C_{ba}^{τ} is a superposition of

$$\exp(\lambda_\alpha \tau) = \underbrace{\exp(-\mu_\alpha \tau)}_{\text{Relaxation}}\ \underbrace{\exp(i\omega_\alpha \tau)}_{\text{Oscillation}}$$

Assume that one of the modes α is dominant.

Quantitative measure of coherence

$$\left(\begin{array}{l}\text{\# of oscillations before the auto-}\\ \text{correlation decays}\end{array}\right)$$

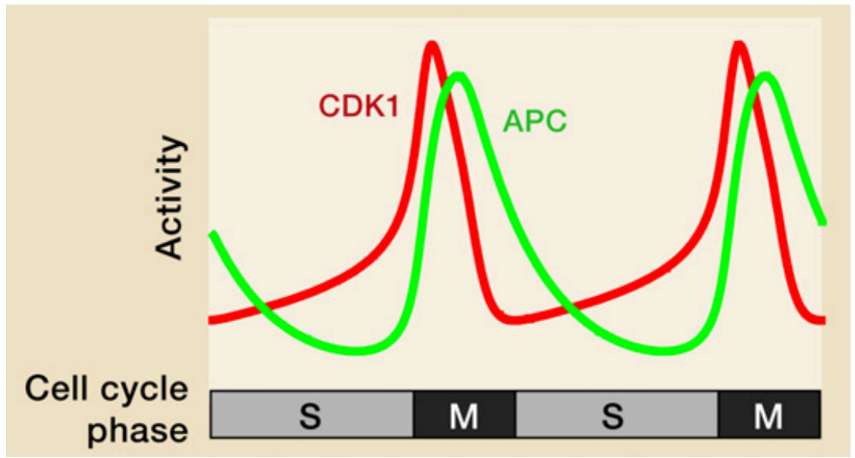
$$= \frac{\text{Decay time}}{\text{Period}} = \frac{(\mu_\alpha)^{-1}}{2\pi|\omega_\alpha|^{-1}} = \frac{|\omega_\alpha|}{2\pi\mu_\alpha}$$

Fluctuating oscillations

Equilibrium $\Longrightarrow$ $\omega_\alpha = 0$ for any α (No oscillation)

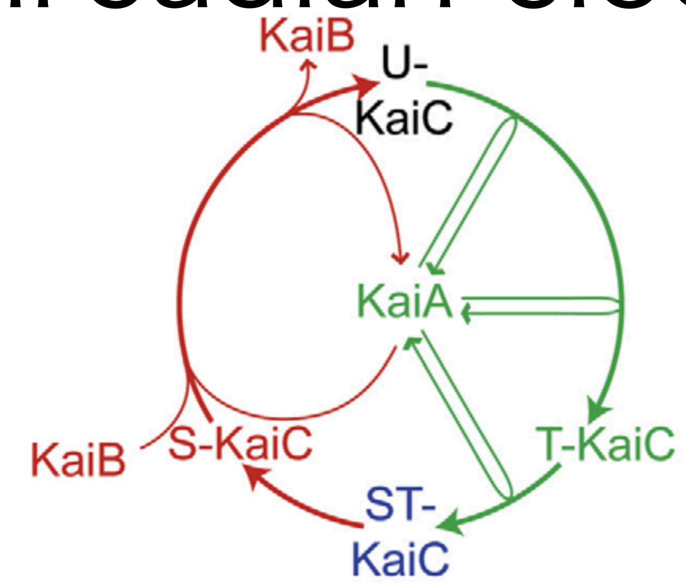
Coherence of oscillation

Cell cycle

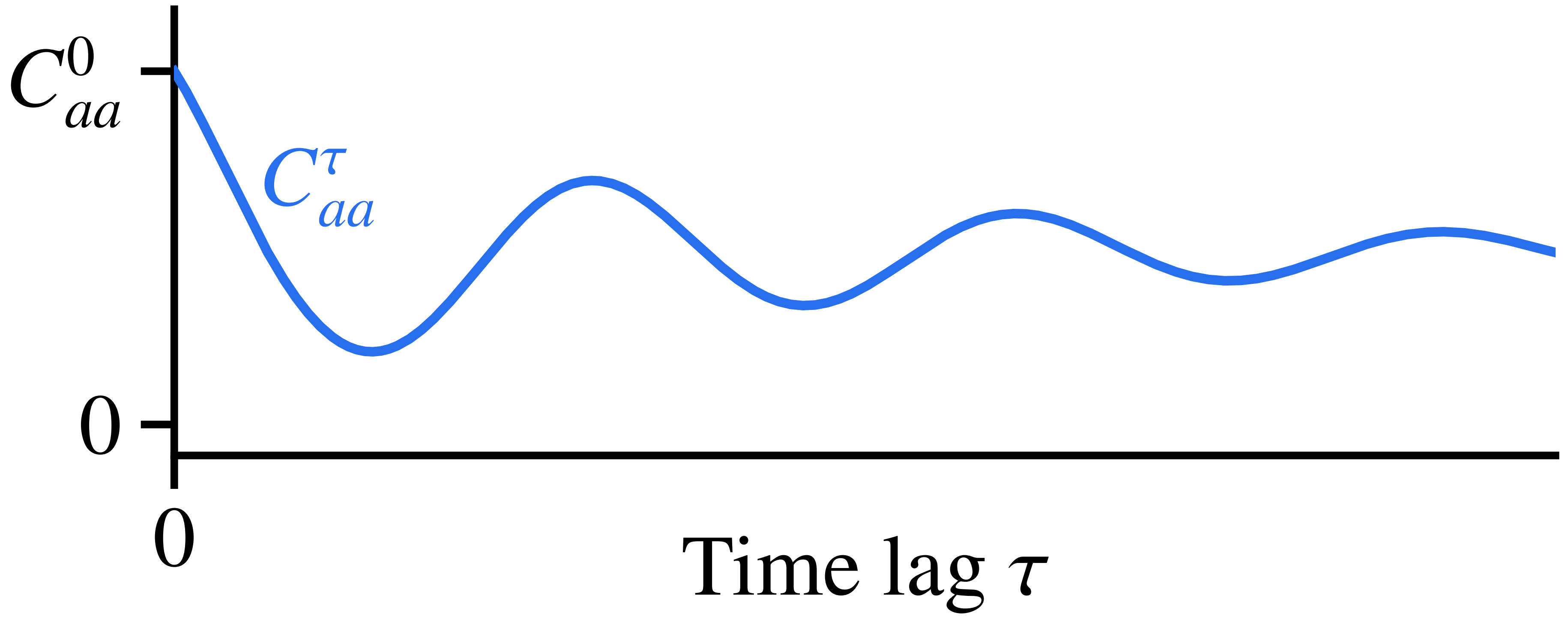


Ferrell+ Cell 2011

Circadian clock



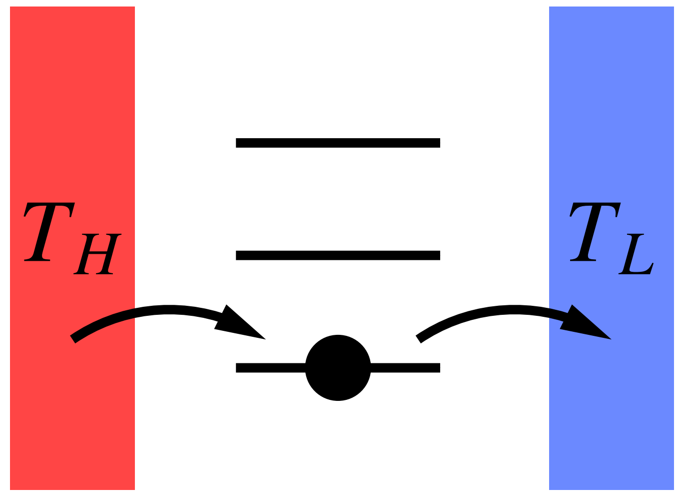
Dong+ Curr. Opin. Microbiol. 2008



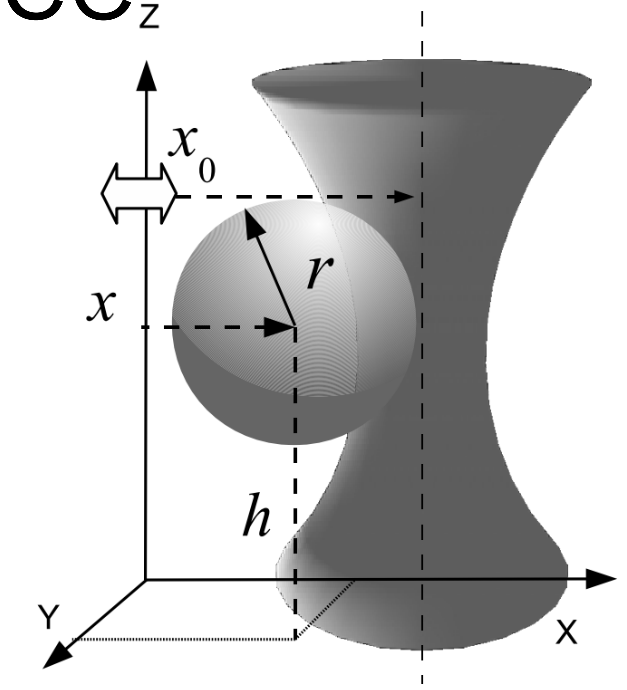
Any universal
& **quantitative**
relation?

Thermodynamic driving

Temperature gradient

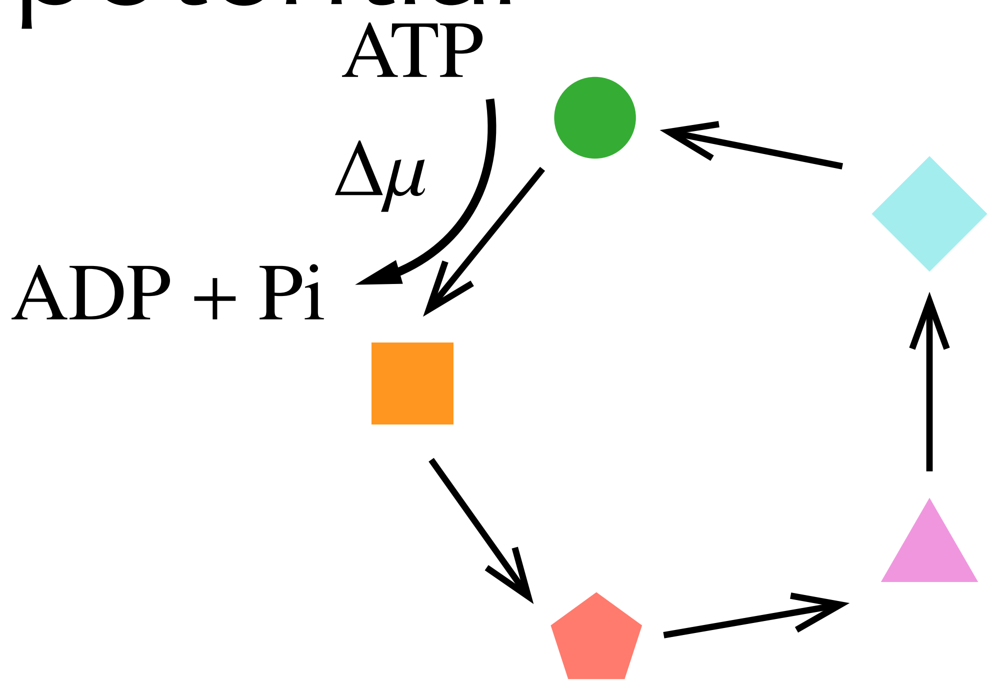


Mechanical force



J. R. Gomez-Solano+, EPL 2010

Chemical potential gradient



Bounds on eigenvalues

Measure of coherence

$$\frac{\text{Decay time}}{\text{Period}} = \frac{|\omega_\alpha|}{2\pi\mu_\alpha}$$

$\lambda_\alpha = -\mu_\alpha + i\omega_\alpha$: eigenvalue

Conjecture

$$\frac{|\omega_\alpha|}{\mu_\alpha} \leq \max_c \frac{\tanh(\mathcal{F}_c/2n_c)}{\tan(\pi/n_c)} \leq \max_c \frac{\mathcal{F}_c}{2\pi}$$

The coherence of oscillation is universally bounded by affinity

Conjectured by Barato and Seifert (2017) based on numerical evidence.

Not rigorously proven before.

Bounds on eigenvalues

Measure of coherence

$$\frac{\text{Decay time}}{\text{Period}} = \frac{|\omega_\alpha|}{2\pi\mu_\alpha}$$

$\lambda_\alpha = -\mu_\alpha + i\omega_\alpha$: eigenvalue

We prove the conjecture as a corollary!

Corollary For all modes α

$$\frac{|\omega_\alpha|}{\mu_\alpha} \leq \max_c \frac{\tanh(\mathcal{F}_c/2n_c)}{\tan(\pi/n_c)} \leq \max_c \frac{\mathcal{F}_c}{2\pi}$$

$\Leftarrow$

Main Result for any a, b

$$|\chi_{ba}| \leq \max_c \frac{\tanh(\mathcal{F}_c/2n_c)}{\tan(\pi/n_c)} \leq \max_c \frac{\mathcal{F}_c}{2\pi}$$

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Main Result for any a, b

$$|\chi_{ba}| \le \max_c \frac{\tanh(\mathcal{F}_c/2n_c)}{\tan(\pi/n_c)} \le \max_c \frac{\mathcal{F}_c}{2\pi}$$

Proof

$$-\mu_\alpha + \mathrm{i}\omega_\alpha = \frac{\partial}{\partial\tau}\exp(\lambda_\alpha\tau)\Big|_{\tau=0} = \underbrace{\frac{\partial}{\partial\tau}C^\tau_{z^*z}\Big|_{\tau=0}}_{\text{Complex-valued "Observable"}} = \underbrace{\frac{\partial}{\partial\tau}\left[(C^\tau_{aa}+C^\tau_{bb}) + \mathrm{i}\,(C^\tau_{ba}-C^\tau_{ab})\right]\Big|_{\tau=0}}_{\text{Real-valued "Observable"}}$$

Complex-valued "Observable"

$z_i = u_i^{(\alpha)}/q_i$

eigenvector

Real-valued "Observable"

$b_i = \mathrm{Im}\, z_i$

$a_i = \mathrm{Re}\, z_i$

Bounds on eigenvalues

$\lambda_\alpha = -\mu_\alpha + i\omega_\alpha$: eigenvalue

We prove the conjecture as a corollary!

Corollary For all modes α

$$\frac{|\omega_\alpha|}{\mu_\alpha} \leq \max_c \frac{\tanh(\mathcal{F}_c/2n_c)}{\tan(\pi/n_c)} \leq \max_c \frac{\mathcal{F}_c}{2\pi}$$

$\Leftarrow$

Main Result for any a, b

$$|\chi_{ba}| \leq \max_c \frac{\tanh(\mathcal{F}_c/2n_c)}{\tan(\pi/n_c)} \leq \max_c \frac{\mathcal{F}_c}{2\pi}$$

Proof

$$-\mu_\alpha + \mathrm{i}\omega_\alpha = \frac{\partial}{\partial\tau}\exp(\lambda_\alpha\tau)\Big|_{\tau=0} = \frac{\partial}{\partial\tau}C^\tau_{z^*z}\Big|_{\tau=0} = \frac{\partial}{\partial\tau}\left[(C^\tau_{aa}+C^\tau_{bb}) + \mathrm{i}\,(C^\tau_{ba}-C^\tau_{ab})\right]\Big|_{\tau=0}$$

$$\frac{|\omega_\alpha|}{\mu_\alpha} = \lim_{\tau\to0}\frac{|C^\tau_{ba}-C^\tau_{ab}|}{\Delta_\tau C_{aa}+\Delta_\tau C_{bb}} \leq \lim_{\tau\to0}\frac{|C^\tau_{ba}-C^\tau_{ab}|}{2\sqrt{(\Delta_\tau C_{aa})(\Delta_\tau C_{bb})}} = |\chi_{ba}| \leq \text{(affinity bound)}$$

$$x + y \geq 2\sqrt{xy}$$

1 Asymmetry of cross-correlations

2 Oscillations (Eigenvalues)

☞ Discussion & Summary

Affinity bounds vs. entropy bounds

	This talk	**Thermo. uncertainty relations**
	$\lim_{\tau\to 0} \frac{C^\tau_{ba} - C^\tau_{ab}}{2\sqrt{\Delta_\tau C_{aa}\,\Delta_\tau C_{bb}}} \leq \max_c \frac{\mathcal{F}_c}{2\pi}$	$\frac{\langle J^\tau_d \rangle^2}{\mathrm{Var}\, J^\tau_d} \leq \frac{\Sigma^\tau}{2}$ J^τ_d : accumulated current
Statistics	Correlations of state observables	Precision of a current observable
Thermodynamic signature	Cycle affinity $\mathcal{F}_c$	Entropy production Σ^τ

Complementary ways to relate statistical and thermodynamic signatures

Affinity bounds vs. entropy bounds

	Cycle affinity $\mathcal{F}_c$	Entropy production Σ^τ
Determined by	Strength of driving Environmental parameters (e.g., chemical potentials)	Resulting dissipation rate
Dependence on the steady state	Independent	Dependent
Response to microscopic kinetic changes	Robust	Sensitive

⇩ **Macroscopic setup**

⇩ **Microscopic behavior**

→ Complementary characterizations of the thermodynamic cost

Summary

$\mathcal{F}_c$: Cycle affinity (Strength of thermodynamic driving)
n_c : # of states on cycle c

Result

The asymmetry of cross-correlation is universally related to affinity

$$|\chi_{ba}| \leq \max_c \frac{\tanh(\mathcal{F}_c/2n_c)}{\tan(\pi/n_c)} \leq \max_c \frac{\mathcal{F}_c}{2\pi}$$

$$\chi_{ba} = \lim_{\tau\to 0} \frac{C^\tau_{ba} - C^\tau_{ab}}{2\sqrt{\Delta_\tau C_{aa}\,\Delta_\tau C_{bb}}} \left[\begin{array}{l} C^\tau_{ba} = \langle b(t+\tau)\,a(t)\rangle \\ \Delta_\tau C_{aa} = C^0_{aa} - C^\tau_{aa} \end{array}\right]$$

Proof: Use the isoperimetric inequality between area and length

Corollary

The coherence of oscillation is universally bounded by affinity

$$\frac{|\omega_\alpha|}{\mu_\alpha} \leq \max_c \frac{\tanh(\mathcal{F}_c/2n_c)}{\tan(\pi/n_c)} \leq \max_c \frac{\mathcal{F}_c}{2\pi}$$

Conjectured by Barato & Seifert (2017)

$\lambda_\alpha = -\mu_\alpha + \mathrm{i}\omega_\alpha$: Eigenvalue of R

Proof: Extract λ_α from a complex-valued correlations

Details → N. Ohga, S. Ito, A. Kolchinsky, arXiv:2303.13116
See also N. Shiraishi, arXiv:2304.12775
T. Van Vu, V. T. Vo, K. Saito, arXiv:2305.18000