Center for Nanophase Materials Sciences

U.S. DOE Nanoscale Science Research Center

An Office of Science User Facility

Collective Phenomena in Nanophases

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Center for Nanophase Materials Sciences

CNMS User Meeting

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U.S. DEPARTMENT OF ENERGY

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Nanoscale: Inherently Challenging

Atomic scale Nano-scale Macro-scale

Thermodynamics

Continuum description

Limit of large numbers

Mean field

$$\frac{\sigma_P}{\langle P \rangle} \propto \frac{1}{N^{1/2}}$$

$\approx \mu\mathrm{m}$

Importance of temporal and compositional fluctuations

no canonical "language"

Crossover from strong to weak correlations

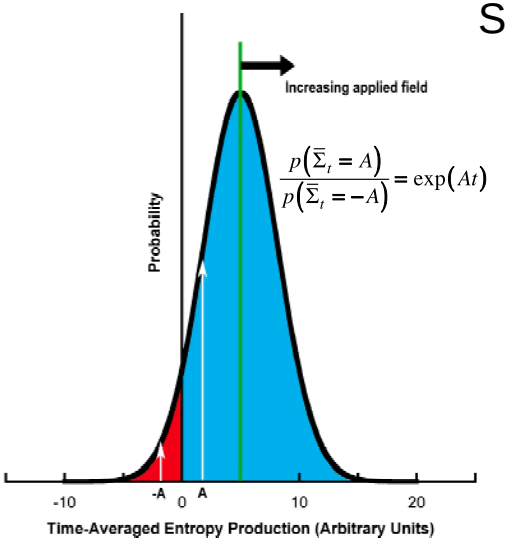
$\approx \mathrm{nm}$

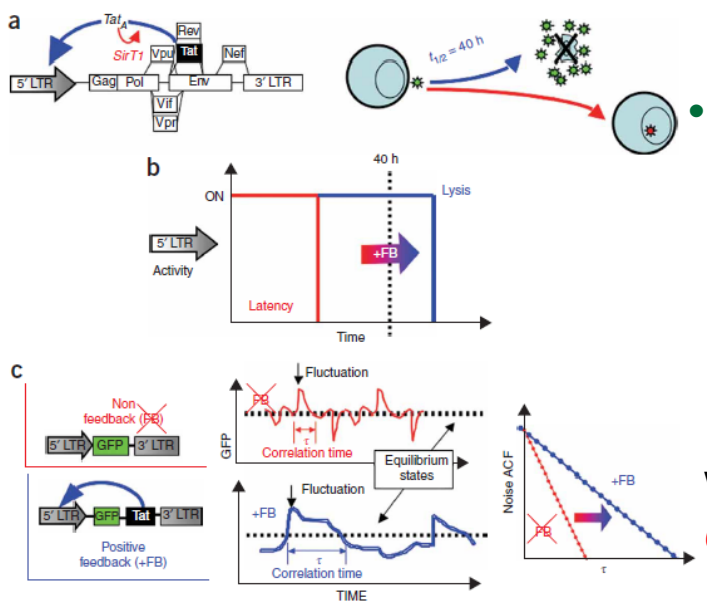
Quantum behavior and strong quantum correlations

Atomistic description

Fluctuations and Correlations in Physical and Biological Nanosystems: The Tale is in the Tails

Simpson and Cummings, *ACS Nano*, 2011





- Microscale revolution based on two fundamental concepts:
  - creating structures by top-down methods
  - *overcoming noise* by driving the systems at high voltages across components
- Typical of many man-made systems, but *inherently incompatible with the environment at the nanoscale*
- Stochasticity increases at nanoscale
  - understanding nature's approach to nanotechnology is key to creating efficient, functional structures that operate in predictable fashion with minimal energy consumption
- *Natural systems exploit stochasticity, rather than overwhelm it*, in order to create deterministic, yet highly adaptable, behavior
  - control of the swimming and tumbling periods of bacteria during chemotaxis
  - Probabilistic decision making in viral gene circuits

Weinberger, Dar, and Simpson *Nature Genetics*, 2008

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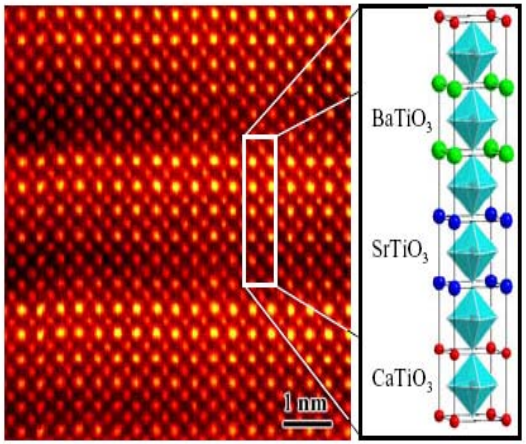
Theme organized around three thrusts

I. Inhomogeneities, dimensional confinement, and materials specificity in cuprate high-T_c superconductors

- Focus on cuprates as specific example of materials exhibiting complex emergent behavior resulting from electron correlations

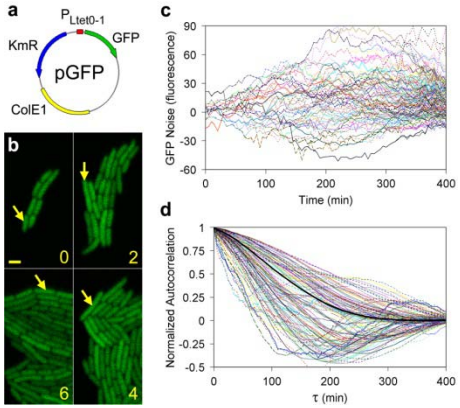


II. Quantum transport in nanostructures

- Focus on electron and spin transport with corresponding experiments

III. Nano-enabled synthetic biology

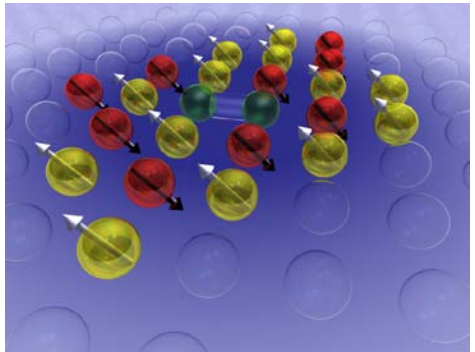
- **Focus on compositional and temporal fluctuations - learning from biology where function emerges from stochasticity**
- **"A bottom-up strategy for a minimal self-sustaining *synthetic* organism"**
- **Discovering simple and usable biological strategies for synthetic nanoscale systems**



Seeking unifying understanding of collective phenomenon

CPN Thrust Areas I & II (collective phenomenon in physical systems): *High-Tc superconductors & Electron Transport/Processes*

- DOE Basic Energy Sciences five grand challenges[1] include
  - ”[T]o discover how remarkable properties of matter emerge from complex correlations of the atomic or electronic constituents and to control these properties."
- Thrust research is directly addressing this challenge
  - Overarching research goal is to understand, predict, design, control, and exploit complex behavior that emerges at nanoscale
    - Crossover from weak to strong electronic correlations: influence on nano-structured material‘s behavior, with specific focus on the role of inhomogeneities, fluctuations, dimensional confinement, and materials specificity in cuprate and unconventional high-Tc superconductors
    - Materials processes at the electron level: *how atomic scale structure and quantum mechanical effects impact electron transport and other electronic processes within nanostructures and across interfaces.*



[1]Fleming, G. and Ratner, M., “Directing Matter and Energy: Five Challenges for Science and the Imagination,” Basic Energy Sciences Advisory Committee, Office of Science, Department of Energy, 2007

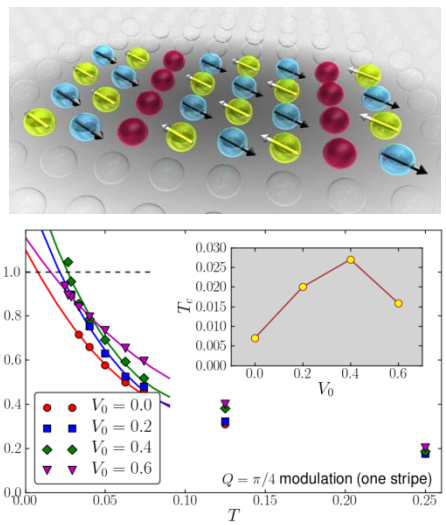
I. Inhomogeneities, dimensional confinement, and materials specificity in cuprate high-Tc superconductors

- Understand effect of inhomogeneities and dimensional confinement on superconductivity in cuprates
  - Use quantum cluster methods implemented in the DCA++ framework to study the physics of appropriate Hubbard-like models
  - Understand role of nano-scale stripe phases and other inhomogeneous states in Hubbard models with uni-directional charge modulations and random disorder
  - Understand superconductivity in cuprate multi-layers described by stack of Hubbard layers with layer dependent chemical potential
  - Address important question of optimal inhomogeneity
- Understand superconductivity in iron-pnictide high-temperature superconductors
  - Understand orbital ordering, pairing symmetry and mechanism in appropriate multi-orbital Hubbard models
- Potential benefit: Fundamental understanding of interplay between inhomogeneities and superconductivity could lead to design of new materials with optimized properties

I. Highlights

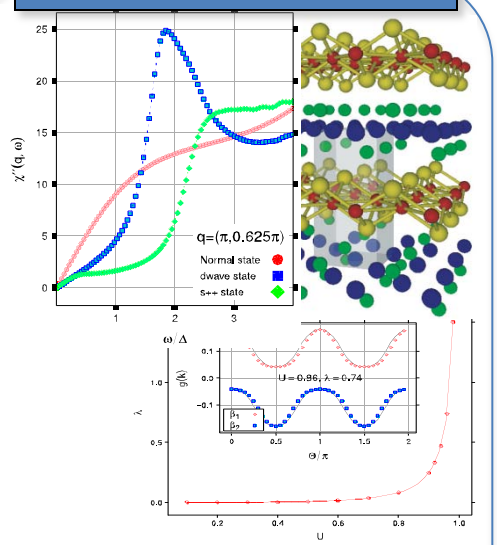
- Quantum Monte Carlo simulations of an inhomogeneous Hubbard model show that superconductivity is significantly enhanced by unidirectional charge stripes
- Random-phase approximation of multi-orbital Hubbard models used to study spin-fluctuation pairing mechanism for iron-based superconductors and its consequences with respect to the pairing symmetry and other experimentally observable properties.
- Discovered coexistence of ferromagnetism and pairing (superconductivity) in models for pnictides superconductors.

Cuprate superconductors



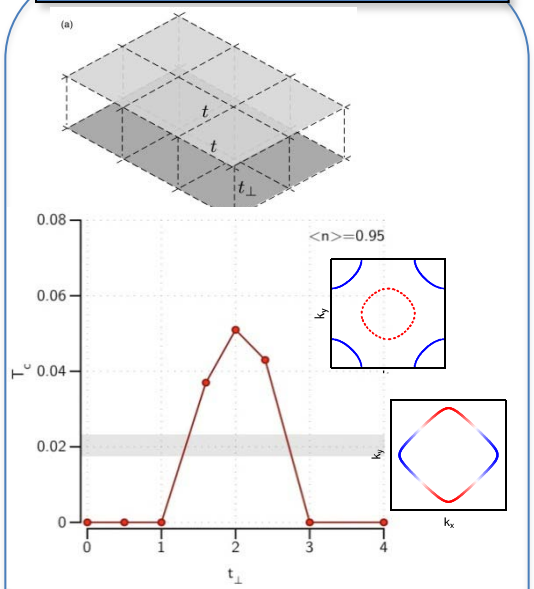
Nanoscale stripe inhomogeneity induced enhancement of superconductivity in QMC simulations of cuprate superconductors
[Phys. Rev. Lett. **104**, 247001 (2010), Phys. Rev. B **81**, 214525 (2010)]

Fe-based superconductors



Prediction of neutron resonance and superconducting gap structure in RPA calculations of iron pnictide superconductors
[Phys. Rev. B **83**, 100515(R) (2011), Phys. Rev. B **83**, 220505(R) (2011), New J. Phys. **12**, 073030 (2010), Phys. Rev. B **81**, 214503 (2010)]

Multi-Fermi-surface systems



Nature of pairing and enhancement of superconductivity in QMC simulations of multi-Fermi-surface bilayer Hubbard model
[submitted to Phys. Rev. Lett., 2011]

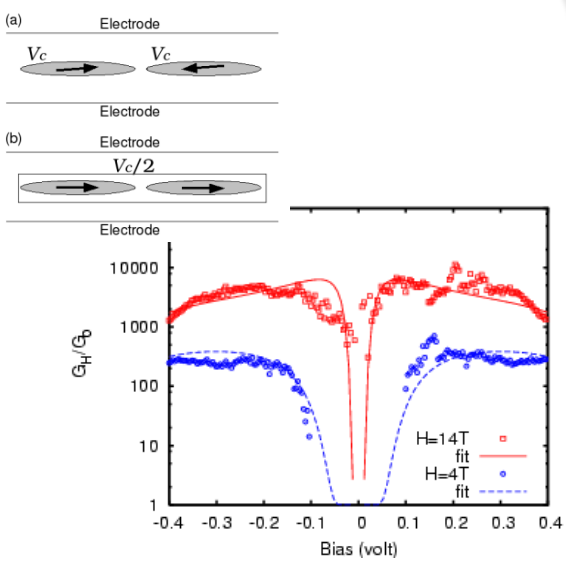
II. Quantum Transport in Nanostructures

Overall goal is to understand materials processes at the electron level: *how atomic scale structure and quantum mechanical effects impact electron transport and other electronic processes within nanostructures and across interfaces.*

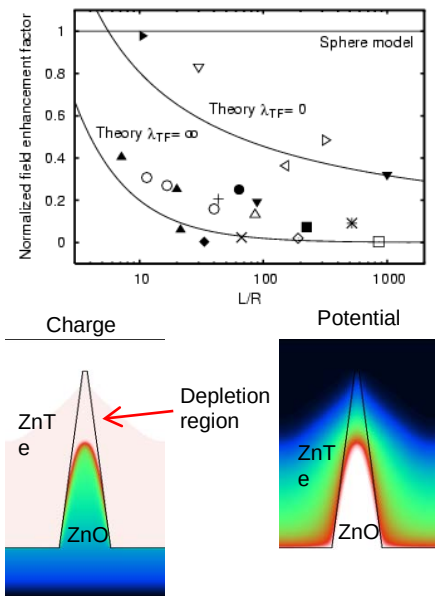
- Beyond elastic electron transport
  - Explore *inelastic quantum transport*, including the effect of dissipative scattering of electrons by phonons and magnons, interaction between an electric current and molecular motion, and the time-dependent dynamics of nanoscale structures under an alternating current.
  - Combine the DFT-based Keldysh-Green function method with the theory of electron-intramolecular coupling in order to account for the effect of elastic-inelastic channel coupling, high order phonon processes, and the effect of surface states, including evanescent to compare with measurements in a new in-house variable-temperature STM
- Theoretical understanding of electron transport in state-of-the-art STM experiments
  - Study theoretically the energy, strength and lifetime of the vibrational modes of single molecules of acetylene (C2H2) and carbon monoxide (CO) adsorbed on different faces of Ni and Cu in close collaboration with CNMS experimentalist Peter Maksymovych.
- Electron transport in complex networks
  - Study switched complex networks where the switching is determined by the nanoscale process corresponding to the electron transport through these junctions, with application to characterizing heterogeneous nanowire and nanotube films.

II. Highlights

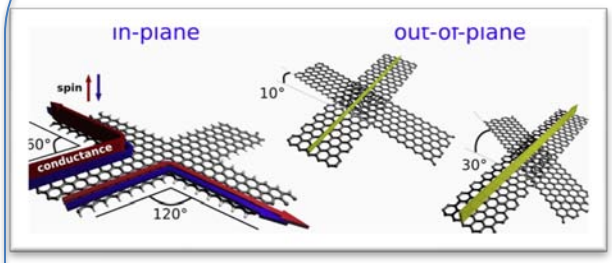
- Theoretical prediction of giant Coulomb blockade magnetoresistance
- Prediction of superior carrier transport properties in nanocones over nanorods for photovoltaics applications
- Prediction of transport property dependance on the intersection angles between in-plane and out-of-plane GNRs with certain intersections exhibiting different transmission probability for spin up and spin down electrons – network design for guiding electrons along desired and predetermined paths

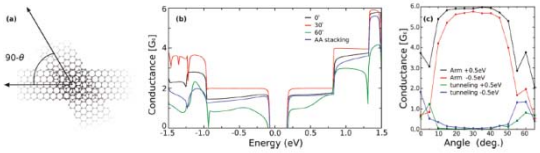


Prediction of giant Coulomb blockade magnetoresistance [Phys. Rev. B 81, 064438 (2010)]



Screening in nanowires and nanocones [Nano Lett. 9, 4306-4310 (2009); Appl. Phys. Lett. 96, 193116 (2010)]





Transport in complex networks

[*Nano Lett.* 11, 3058 (2011); *Adv. Func. Mater.* **19,** 1193 (2009); *ACS Nano* 3, 3603(2009); *Phys. Rev. Lett.* 105, 045501 (2010)]

III. Nano-Enabled Synthetic Biology

- *<u>The</u> Grand Challenge for Nanoscience is to master energy and information on the nanoscale to create new technologies <u>with capabilities rivaling those of living systems</u>**

Collective Phenomenon Theme

- Scale → inherently multiscale: functionality emerges from nanoscale elements, but plays out at the micro and macroscales
  - Multiscale Functionality & Functional Polymer Architecture Themes
- Materials → heterogeneous mixture of materials
  - Multiscale Functionality & Functional Polymer Architecture Themes
- Synthesis → top-down and bottom-up
  - Multiscale Functionality & Functional Polymer Architecture Themes

**Matter and Energy: Five Challenges for Science and the Imagination, A Report from the Basic Energy Sciences Advisory Committee Chapter 5,* REALIZING THE *<u>DREAM OF NANOSCIENCE</u>*: ENERGY AND INFORMATION ON THE NANOSCALE

III. Nano-Enabled Synthetic Biology

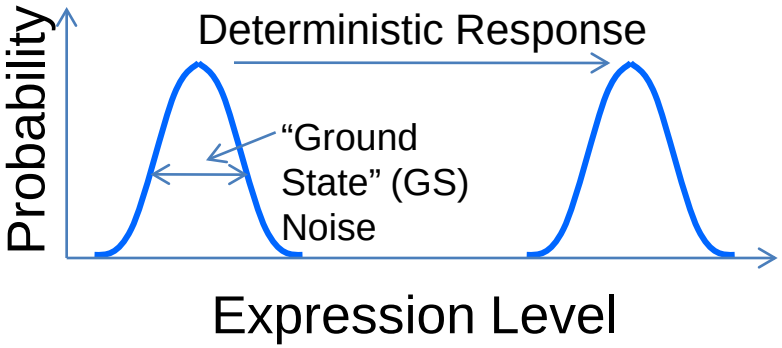
- BES Grand Challenges Report Envisions
  - A bottom-up strategy for a minimal self-sustaining *synthetic* organism
  - Discovering simple and usable biological strategies for synthetic nanoscale systems
  - Elaborate functions, such as self-repair of the photosynthetic machinery
- CNMS vision first detailed in Doktycz and Simpson "Nano-enabled synthetic biology," *Mol. Sys. Bio*. 2007
  - Design is an indispensable tool for unraveling complexity
  - Drastically different organizational approaches
    - Exploit weak interactions
    - Provide specificity
    - Lead from simply high spatial density to high functional density
  - Ability to control the synthesis and direct the assembly of synthetic nanoscale elements → bottom-up design and construction of nanosystems with cell-like complexity
    - Encoding of information within small populations
    - Function emerging from an environment with large stochastic fluctuations

Fluctuations in Mesoscale Systems

III. Nano-enabled synthetic biology

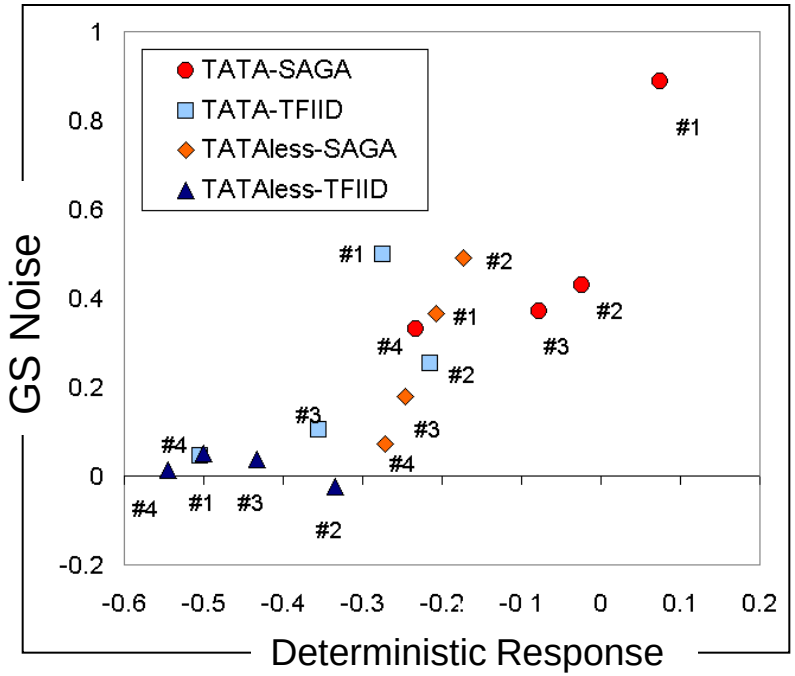
- Model system 1: *Saccharomyces cerevisiae*
  - System-wide study of noise
  - Relationship between stochasticity and deterministic response
- Model system 2: Retrovirus
  - Understand the role of noise in producing more function in less space
- Model system 3: Cell Mimic
  - "Can we make the equivalent of artificial cells?"
  - Abiotic systems that test and advance function-structure relationships
  - Near term: sustained reaction systems with "simple" genetic regulatory networks (e.g. repressilator and toggle switch)
  - Longer-term: development of defined reaction systems with minimal components and simple feedstocks

Stochasticity and deterministic response

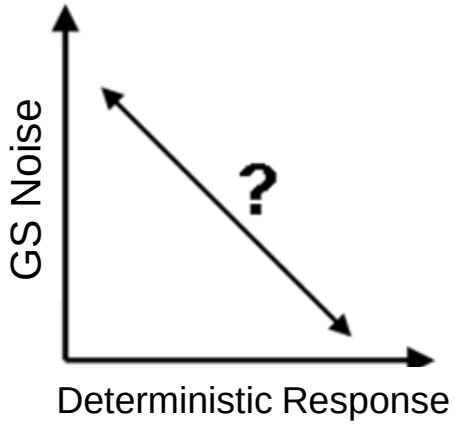


Result

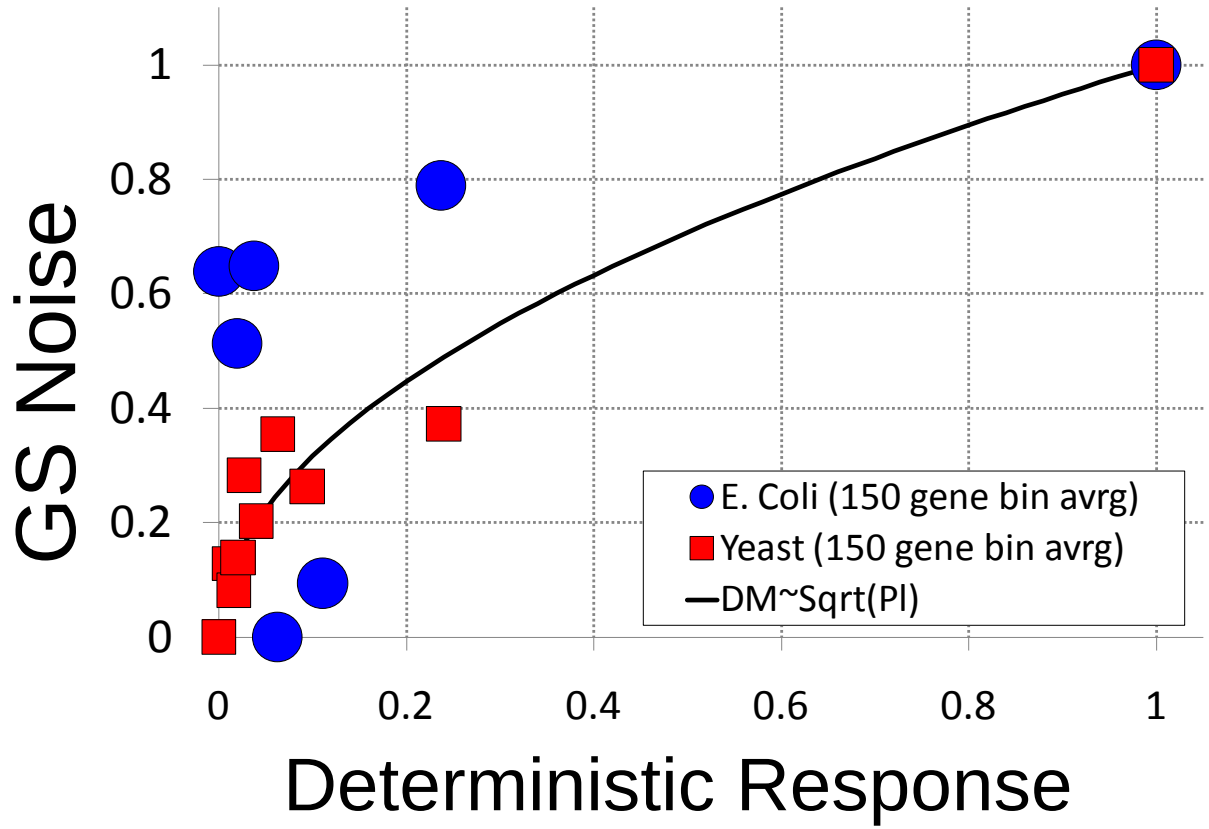
Dar *et al.*, *Chaos*, 2010

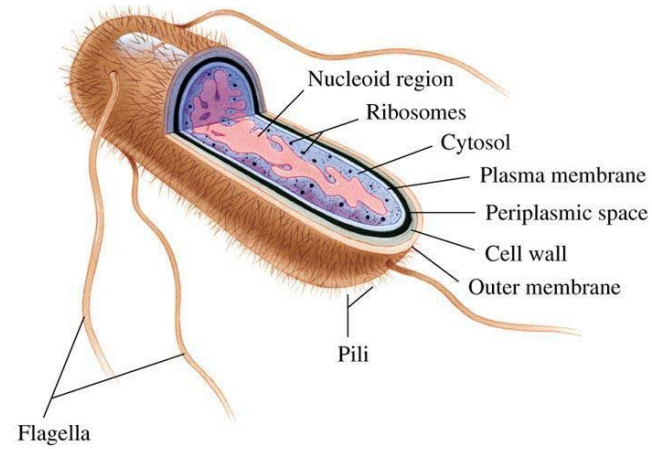


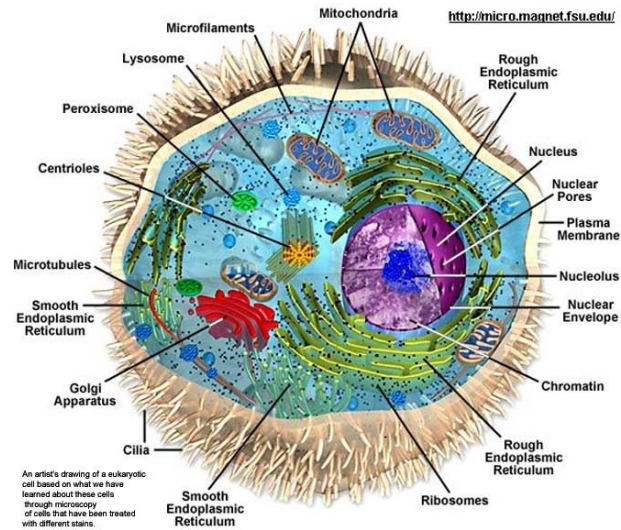
Hypothesis



Is stochastic-deterministic coupling fundamental to adaptive systems?



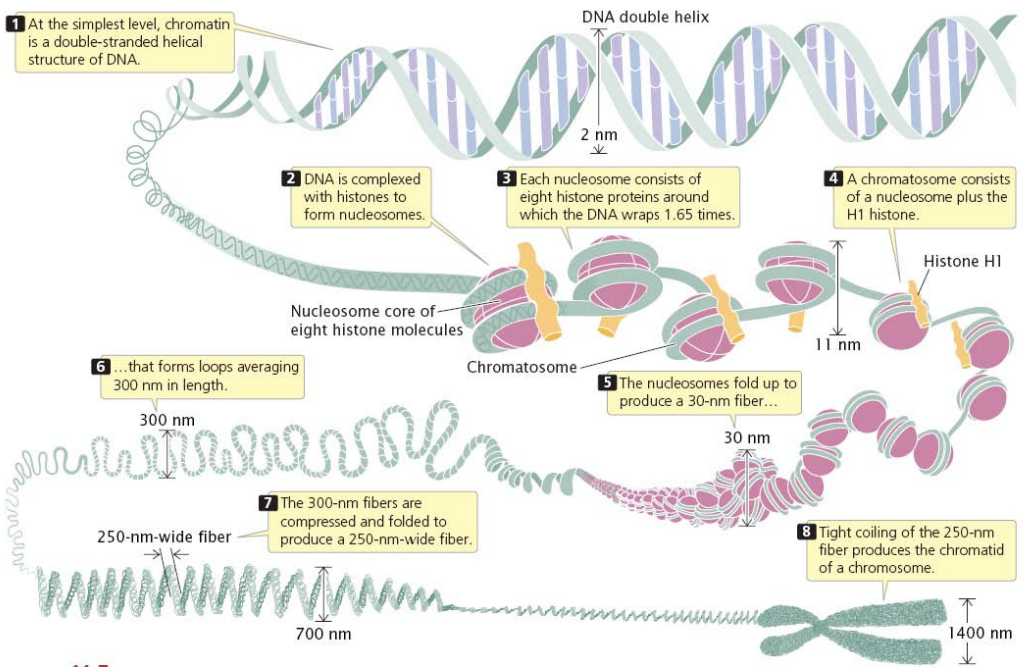




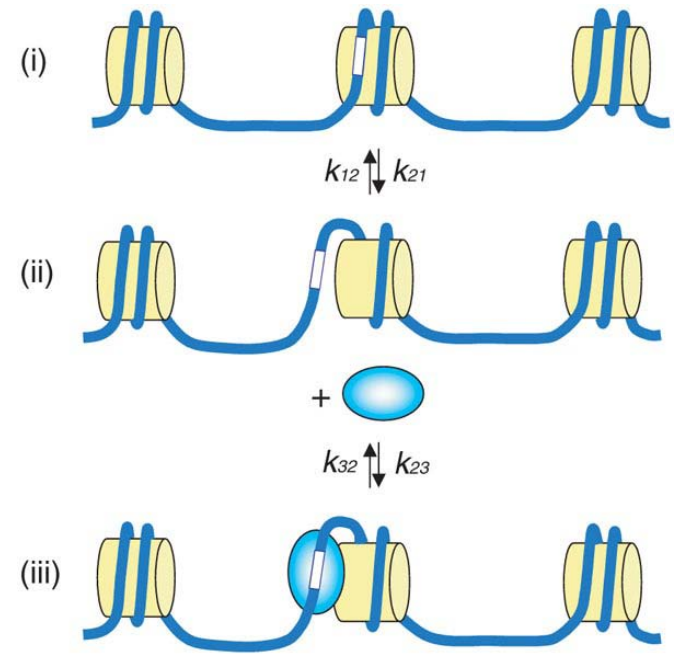
What couples stochastic and deterministic responses?

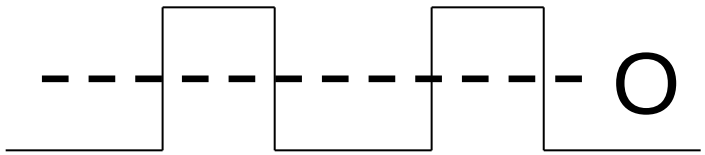
"Each of us has enough DNA to reach from here to the sun and back, more than 300 times. How is all of that DNA packaged so tightly into chromosomes and squeezed into a tiny nucleus?"
Annunziato, A. (2008) DNA packaging: Nucleosomes and chromatin. Nature Education 1(1)

Structure at the nanoscale



Two state model

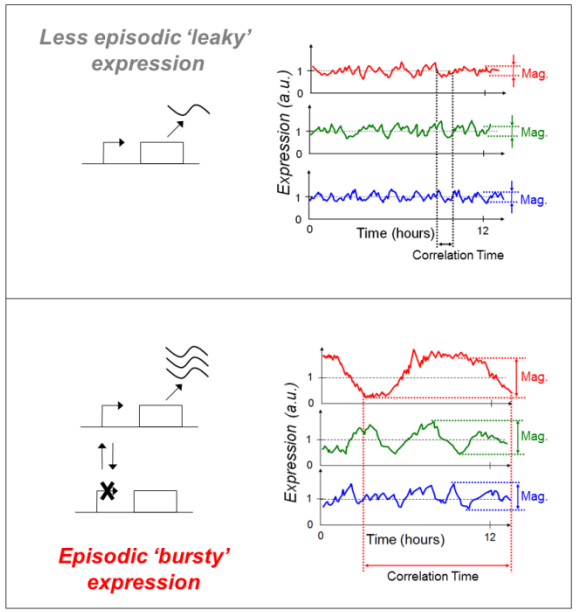


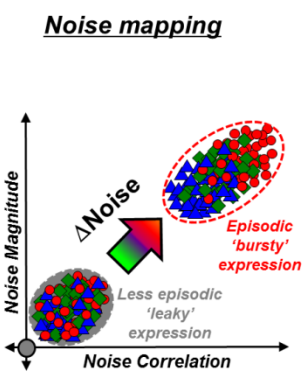


$$R \propto \frac{1}{O_{GS}}$$

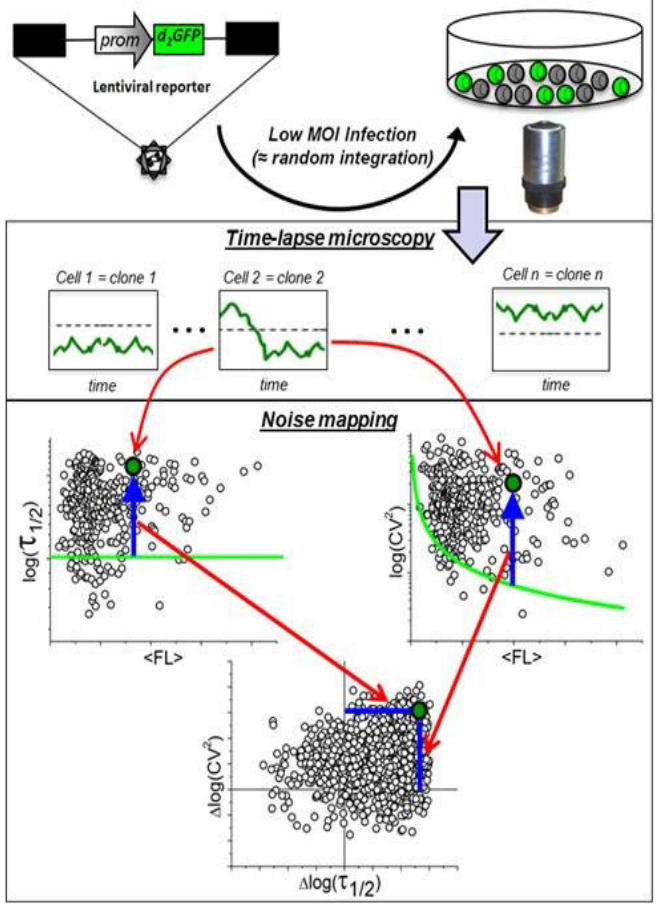
$$Noise \propto \sqrt{\frac{(1-O_{GS})}{O_{GS}}} \propto \sqrt{R}$$

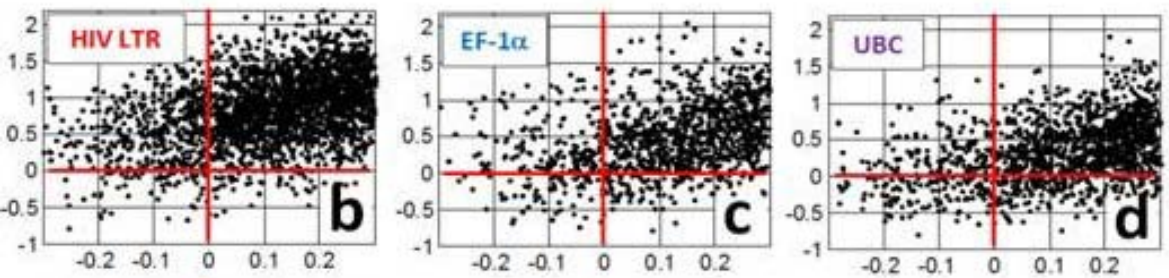
System-wide burstiness





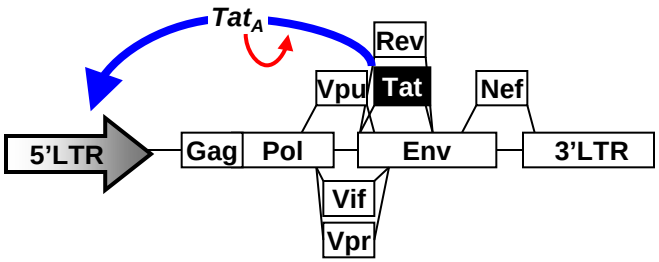
Dar *et al.*, *Nat. Meth.*, in review

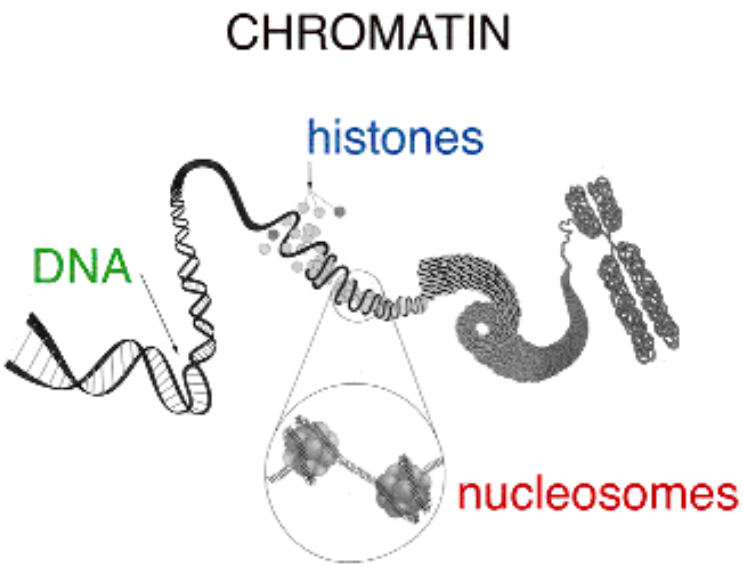


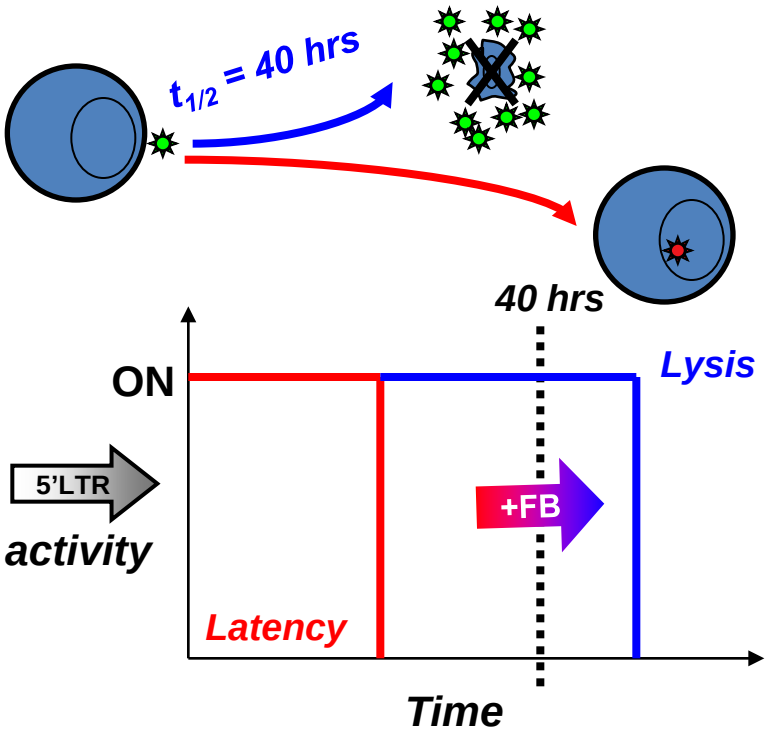


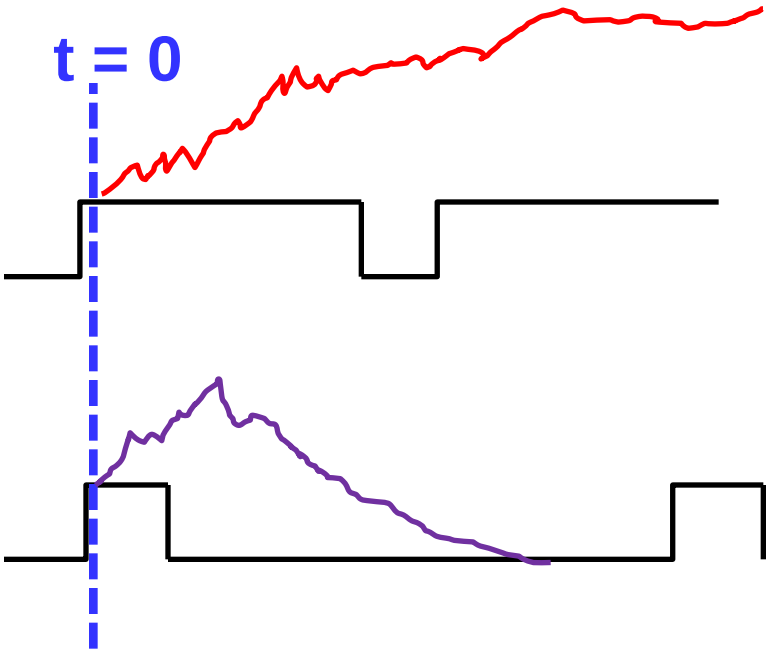
- Episodic (bursty) expression is the rule
- Two-state burst mechanism allows genes to share the space & resources…but it creates noise
- Noise may be used to functional advantage

Noise-driven function in HIV



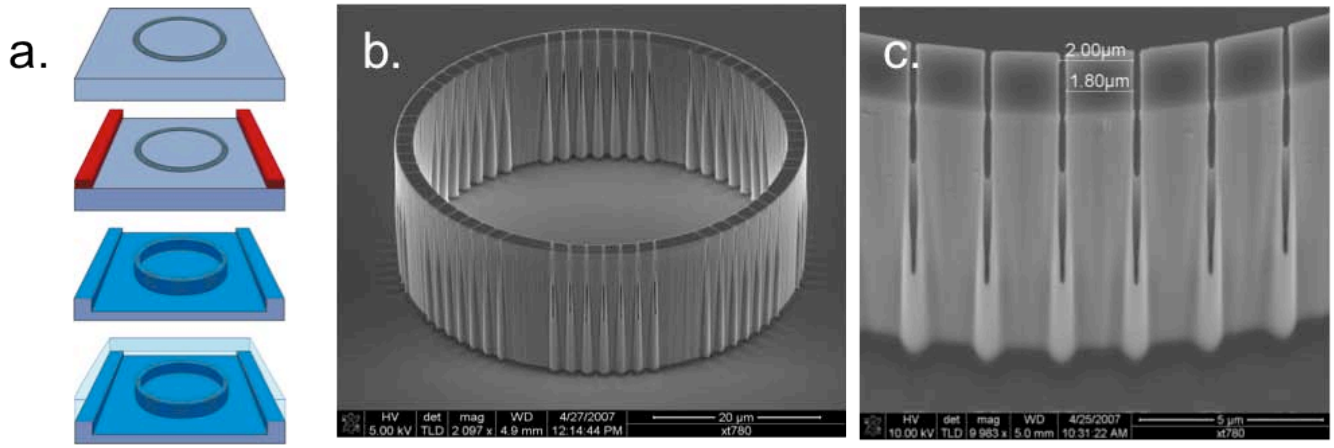




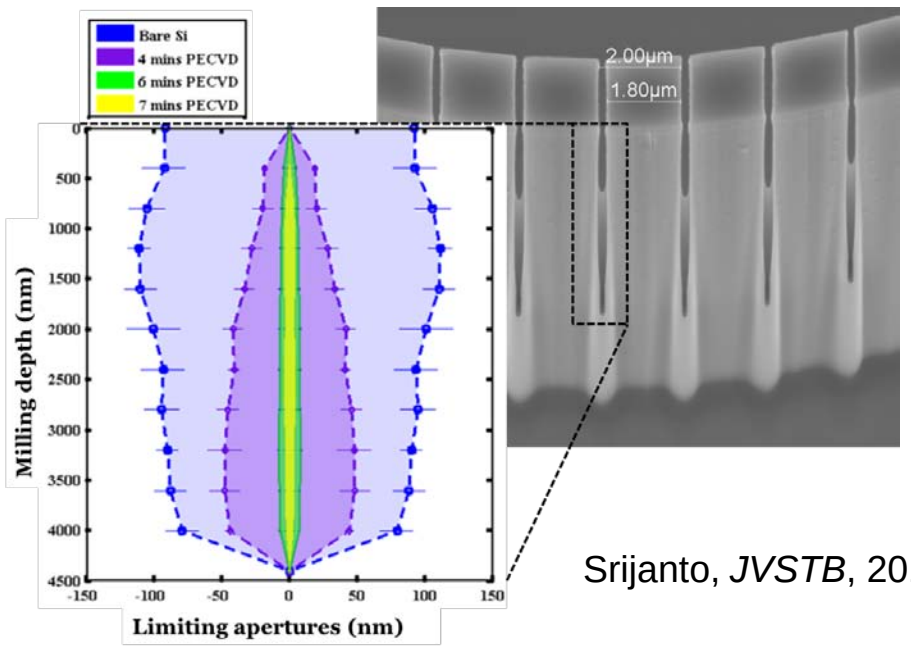


Weinberger, Dar, & Simpson, Nature Genetics 40(4), 466-470, March 2008

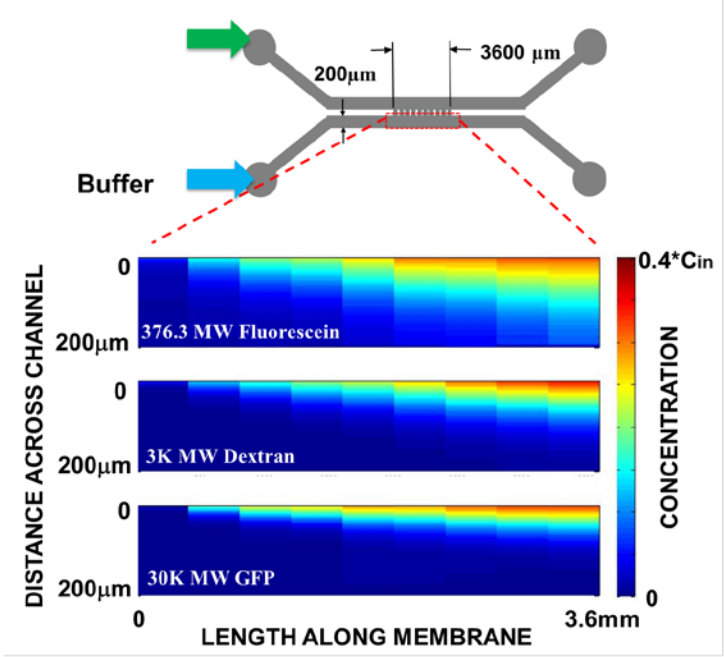
A bottom-up strategy for a minimal self-sustaining *synthetic* organism



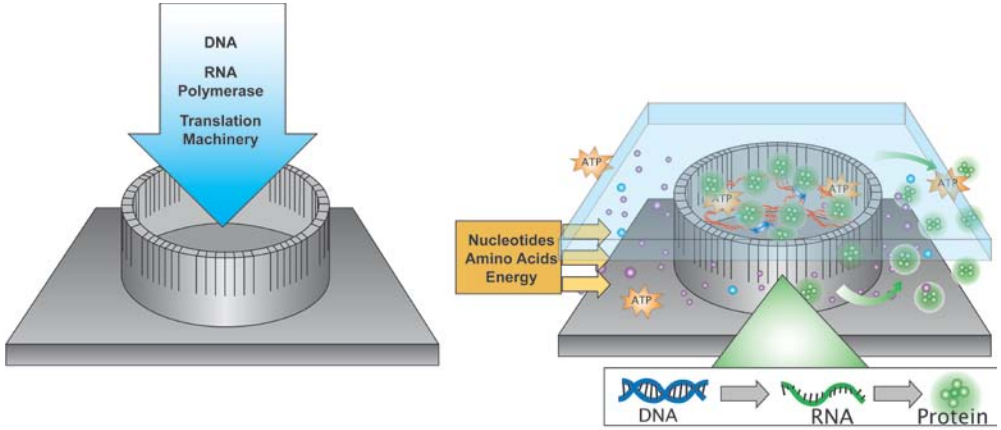
Retterer *et al.* *Lab on a Chip*, 2010



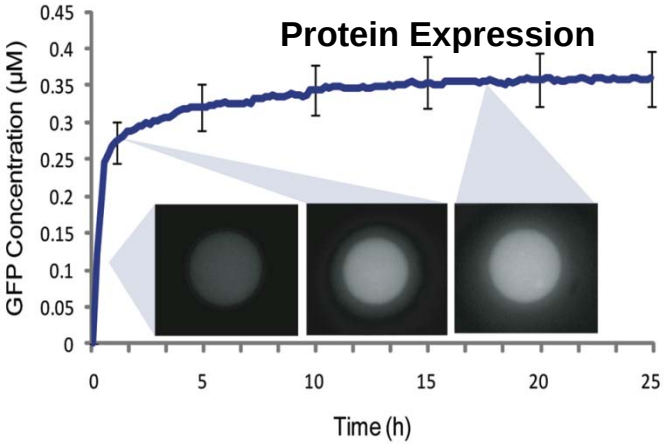
Srijanto, *JVSTB*, 2010



Collective phenomenon



Siuti et al., *Lab on Chip*, in press

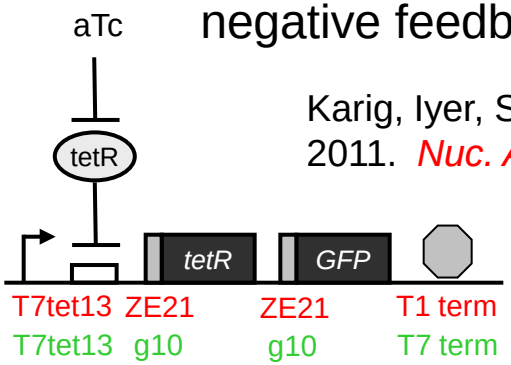


Collier and Simpson, *Current Opinion in Biotechnology*, 2011
Jung, Retterer, Collier *Lab on a Chip*, 2010
Lavrik et al. *Lab on a Chip* 2010
Choi et al., *ACS Nano*, 2010

Characterize genetic parts (repressible T7 promoters)

Tune system parameters (ribosome binding site, transcriptional terminators, etc.)

Assemble and test systems (T7 negative feedback)



Karig, Iyer, Simpson, Doktycz. 2011. *Nuc. Acids Res*. (in review)

Summary

- Emergent Behavior Theme is now the Collective Phenomenon in Nanophases Theme
- Three major thrusts
  - Inhomogeneities, dimensional confinement, and materials specificity in cuprate high-T_c superconductors (see poster)
  - Quantum transport in nanostructures (see poster)
  - Nano-enabled synthetic biology
- *Grand Challenge for Nanoscience is to master energy and information on the nanoscale to create new technologies* ***<u>with capabilities rivaling those of living systems</u>***
  - Interplay between deterministic and stochastic responses to create "more function in less space"
  - "Can we make the equivalent of artificial cells?"

Acknowledgements

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- Ivan Kravchenko
- Nick Lavrik
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- Darrell Thomas
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