SAND2007-2488P

Theory & Simulation of Nanoscale Phenomena

CINT is focused on integrated nanomaterials.

How do the building blocks (electrons, atoms, molecules, ...) yield ***special****, especially collective and emergent, properties?*

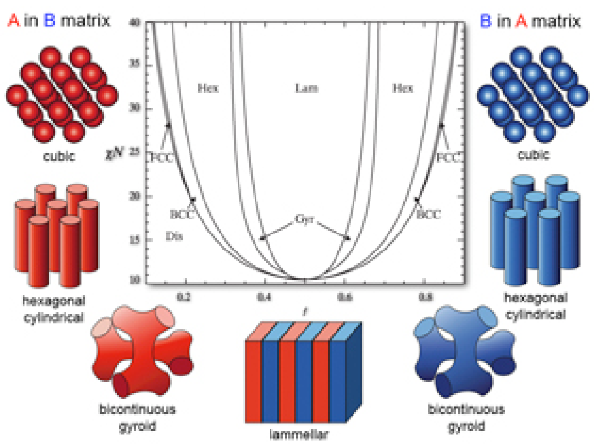
Competing Interactions ↔ Multiple Phases or States

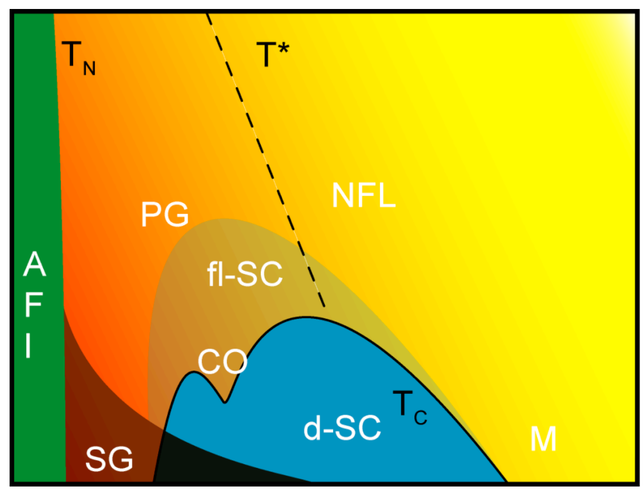
↘ ↙

Material Properties

Generic to Soft & Hard materials

block copolymer





High T_c

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Competing Interactions

- Classical interactions
  - Example: standard self-assembly of amphiphiles
    - hydrophobic, hydrophilic, packing, entropy
    - hydrogen bonds, van der Waals, electrostatic, entropy

soft materials

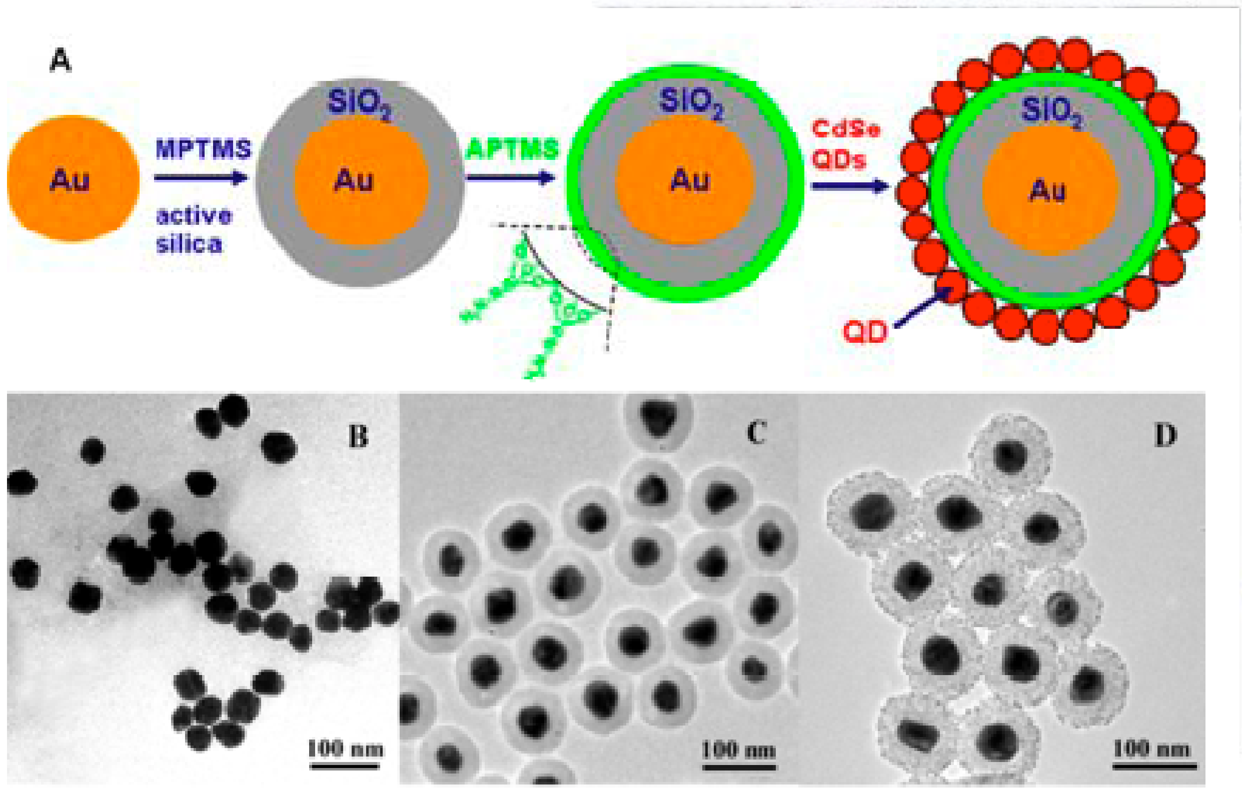
- Quantum interactions
  - Example: cuprate superconductors
    - antiferromagnetic, charge ordered, superconducting
    - Coulomb, spin, electron-phonon, pairing

hard materials

- Phase Competition (all cases)
  - balance of competing interactions sensitive to even small perturbation by an additional interaction
  - the complete interactions can yield new collective or emergent phenomena

Competing Interactions: Nanoparticle Assemblies

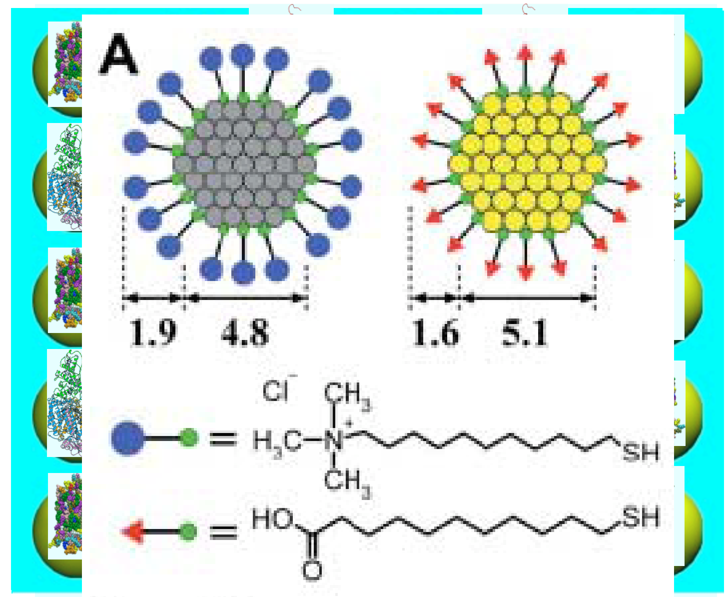
- multifunctional materials
  - multiple nanoparticle types
  - composite nanoparticles

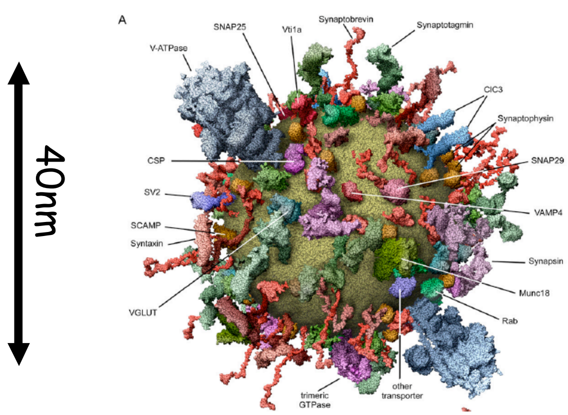


Quantum dots/metal nanoparticles

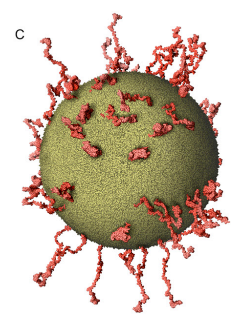
Competing Interactions: Nanoparticle Assemblies

- nanoparticle-nanoparticle interaction
  - core interactions
  - coating interactions
- solvent mediation of interactions
- nanoparticle assemblies
- proteins are nanoparticles
  - multifunctional
  - complex protein-protein interactions
- lots of possibilities
- plenty of challenges for theory

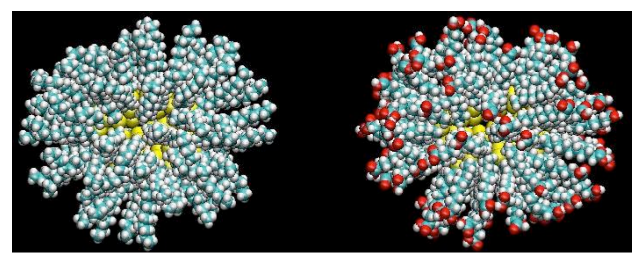




S. Takamori *et al.* Cell (2006)

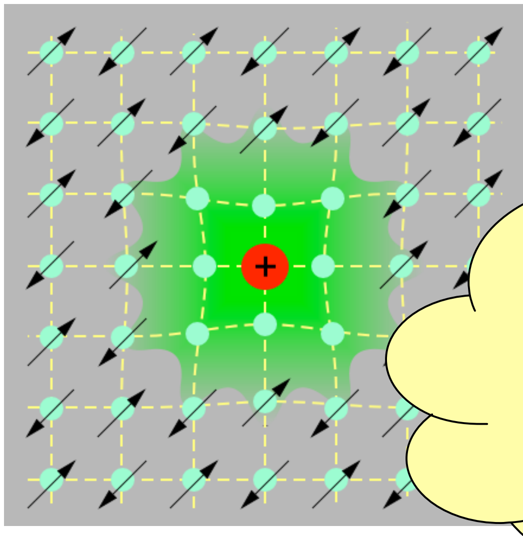


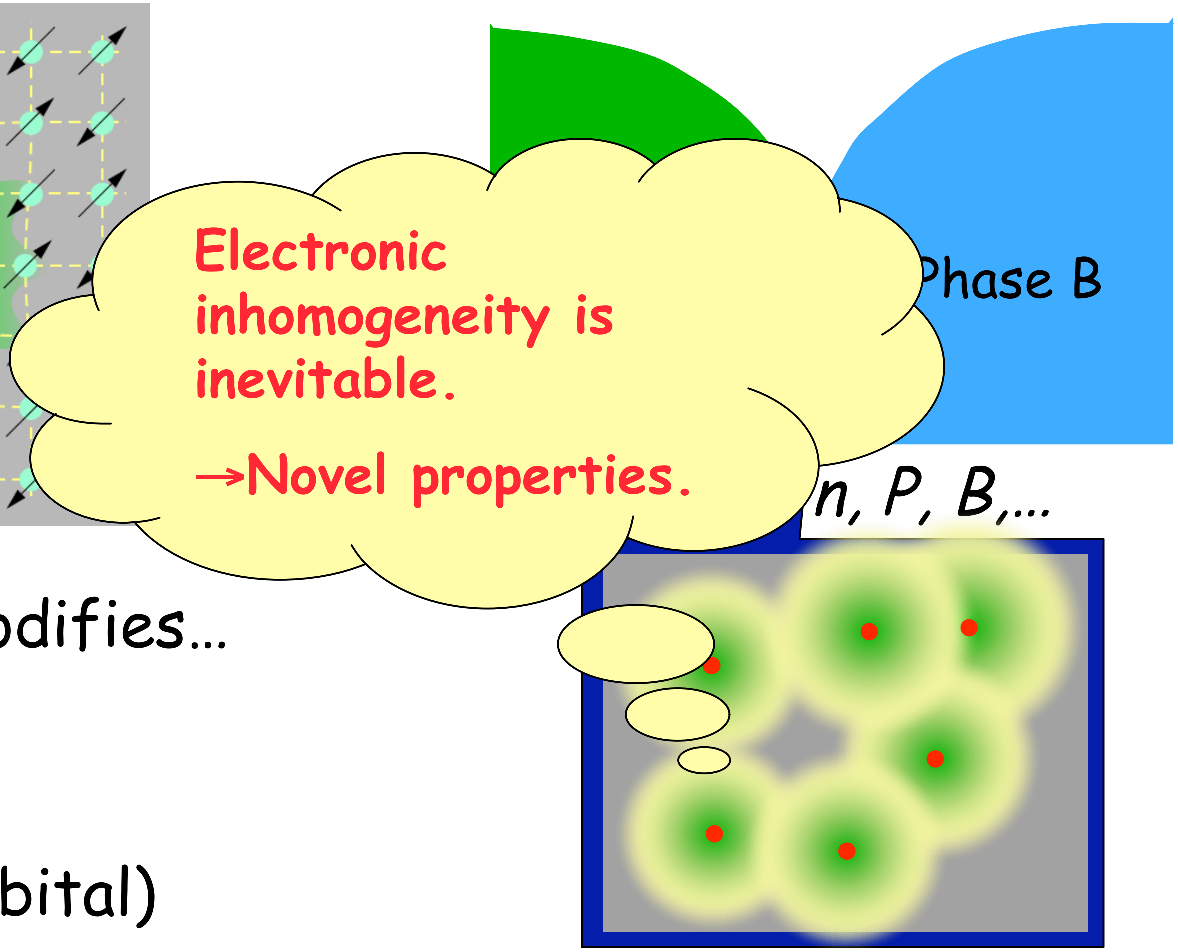
polymer on nanoparticle



alkanethiols on Au nanoclusters

Competing Interactions:
Electronic Degrees of Freedom





Impurity modifies...

- carrier
- spin
- lattice (orbital)

e.g. CMR...
High T_c SC, multiferroics
graphene (?)

Staff

quantum

Sasha Balatsky (LANL): Electronic properties at the nanoscale

Stuart Trugman (LANL): Many-body quantum techniques, probes of correlated systems, new numerical methods

Sergei Tretiak (LANL): (TD)DFT, excited state structure and dynamics, spectroscopy

Normand Modine (SNL): DFT, ab initio MD, organics + tunneling electronics, surface structure of semiconductors

classical

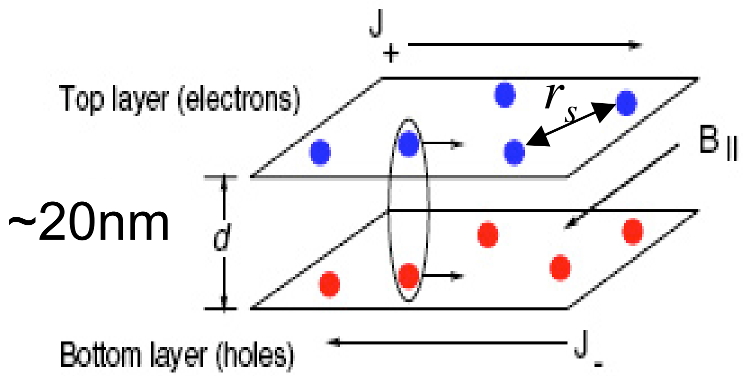
Mark Stevens (SNL): MD, charged & biomolecular systems, nanoparticle interactions, self & active assembly

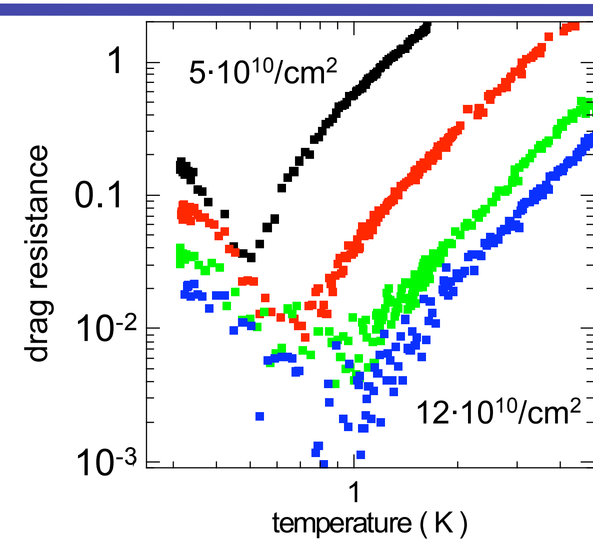
Gary Grest (SNL): MD simulations, nanoparticle rheology, self-assembly, polymer membranes

Amalie Frischknecht: (SNL) molecular theory and simulation of complex fluids, nanoparticle self-assembly, nanoparticle/polymer composites

Excitons in electron-hole bilayers

- Samples are grown at Sandia (Lilly, BES core)
- Theory (Joglekar, Balatsky) developed in CINT

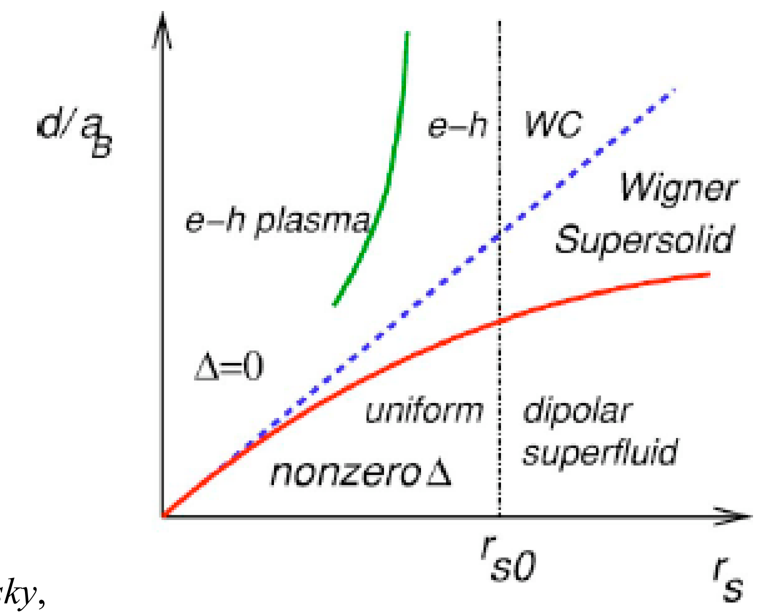




Uncoupled bilayers: $d > r_s$

Phase-coherent bilayers: $d < r_s$

Wigner crystal: $r_s > O(10)$



Y. Joglekar, *M. Lilly, A.V. Balatsky*, S. Das Sarma, PRB **74**, p 233302 (2007).

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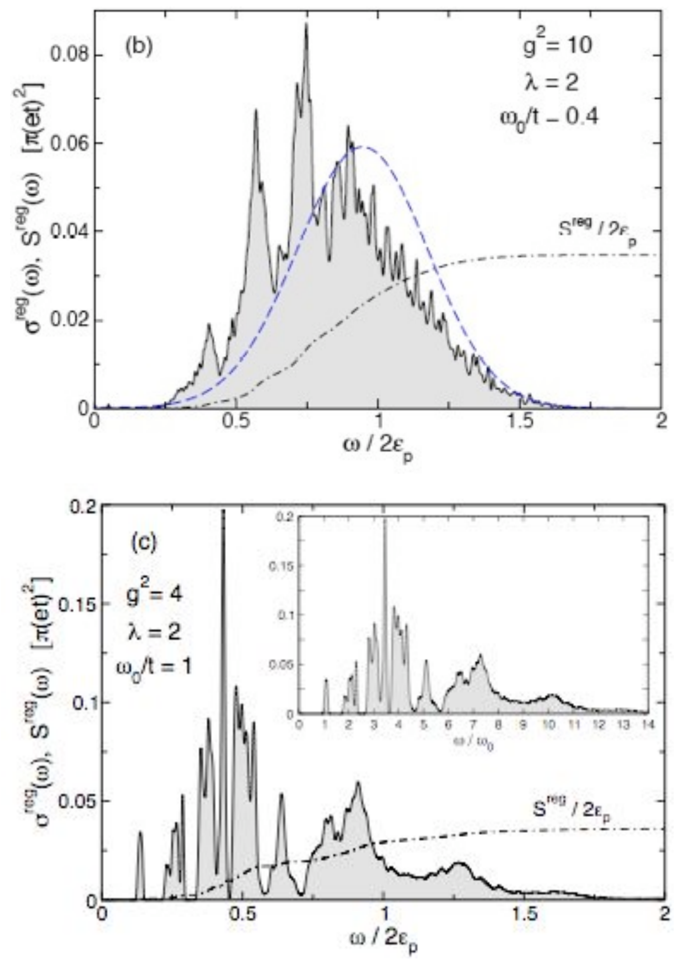
Electron-Phonon Coupling

- semiconductors, organic molecular crystals, high-Tc cuprates, CMR manganites
- nanoscale inhomogeneity
- full quantum treatment
  - both electron & phonon (polaron)

$$H = -t \sum_{\langle i,j \rangle} c_i^\dagger c_j - \bar{g} \sum_i (b_i^\dagger + b_i) n_i + \omega_0 \sum_i b_i^\dagger b_i$$

- new methods development
- fast optics (Nanophotonics)
  - correlated systems
  - exciton-phonon is similar

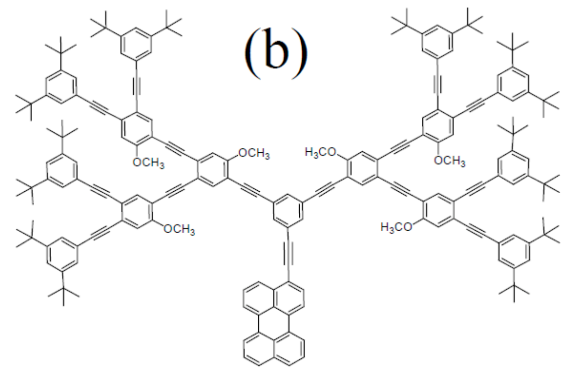
- CINT: S. Trugman
- User: Holger Fehske (U. Greifswald)

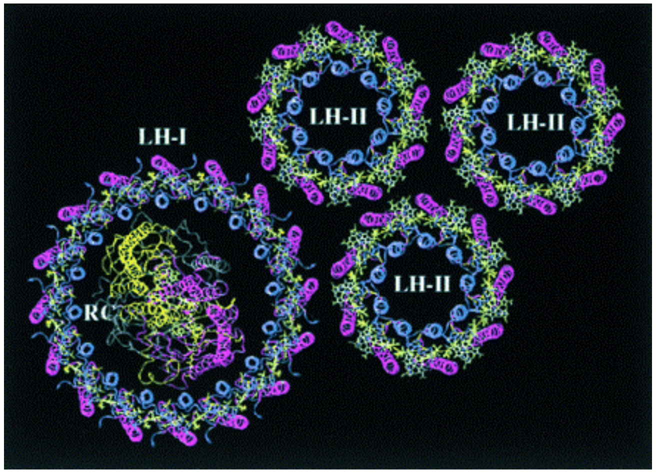


Optical response of Polaron

Local Properties: the building blocks

- Ex. Carbon nanotube, DNA
- exciton scattering in molecular networks
  - Delocalized excitonic excited states in large tree-like quasi-one-dimensional structure can be represented as standing waves
  - These result from exciton scattering at the molecular ends, joints, and branching vertices
  - This model is asymptotically exact in the limit of the exciton size being short compared to the linear segment lengths
  - Any branching center can be treated as a scattering center with an appropriate unitary frequency-dependent scattering matrix.





Light Harvesting Complex

C. Wu, S. Malinin, *S. Tretiak*, and *V. Chernyak*, Nature Phys., 2, 631 (2006).

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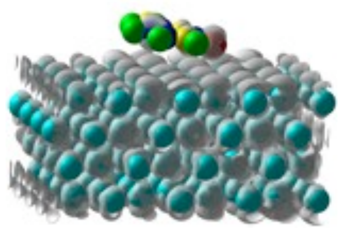
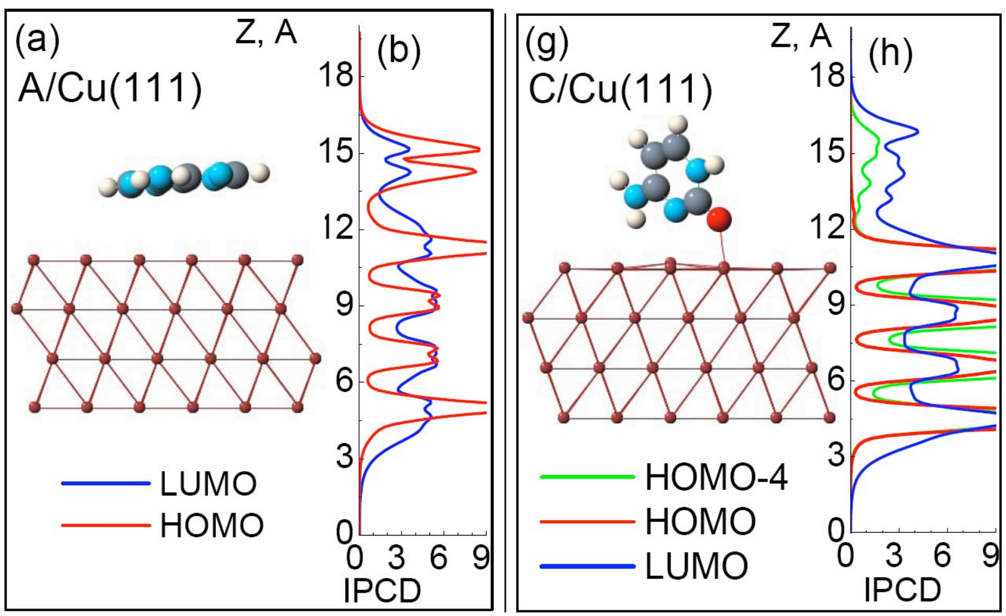
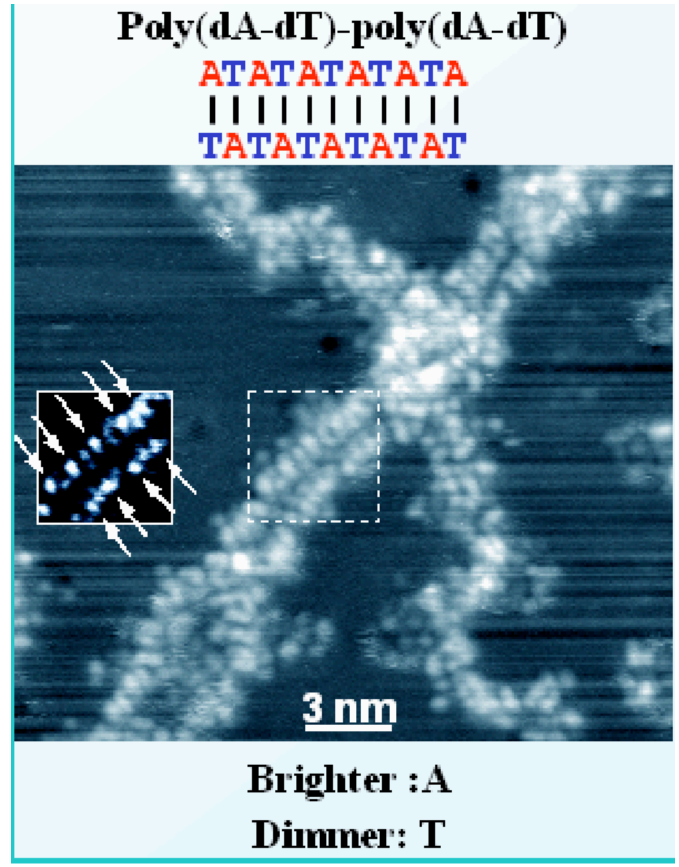
Modeling STM Probes of DNA

- Collaboration between Balatsky, Tretiak, Taylor (LANL) & Modine (SNL)
- calculations of
  - Density of States for each base
  - STM images
  - DNA on Cu







H. Tanaka, 2003

S. Kilina, S. Tretiak, D. Yarotskii, J.X. Zhu, N. Modine, A. Taylor and A.V. Balatsky (submitted)

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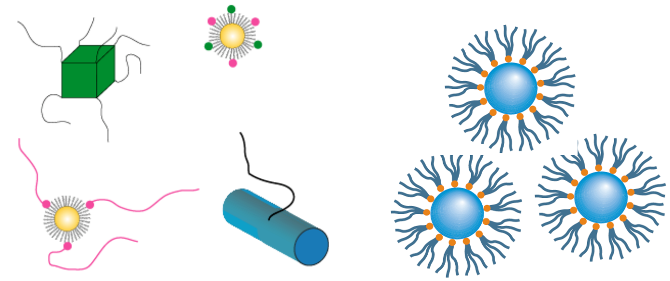
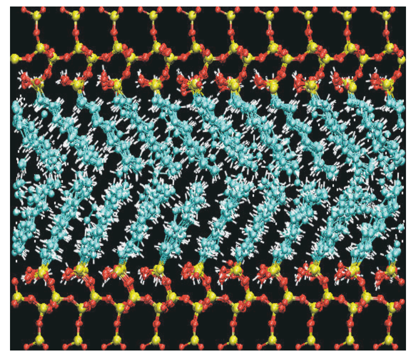
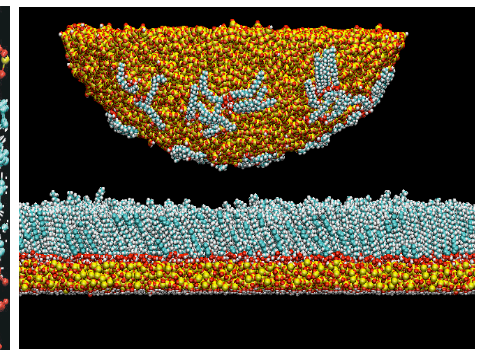
Nanoparticle interactions

- Self-assembled monolayers
  - •MD simulations
  - •atomic scale interactions
  - •adhesion, friction
- flat → curved surfaces → nanoparticles
  - •shape
- coatings
  - • short molecules (atomistic)
  - • polymers (coarse-grained)
- solvent mediated interactions
  - •hydrodynamic interactions
- self-assembly of system

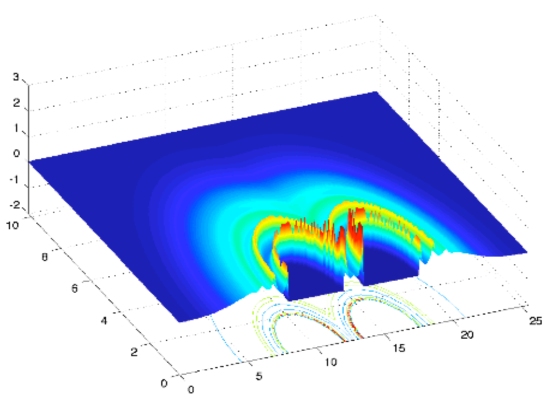






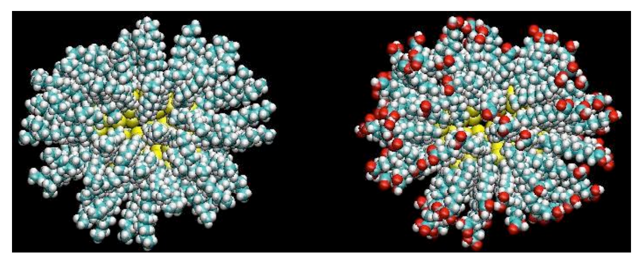
Grest, Stevens

AFM tip-surface interactions



Molecular Theory of nanorods with grafted polymer

Frischknecht



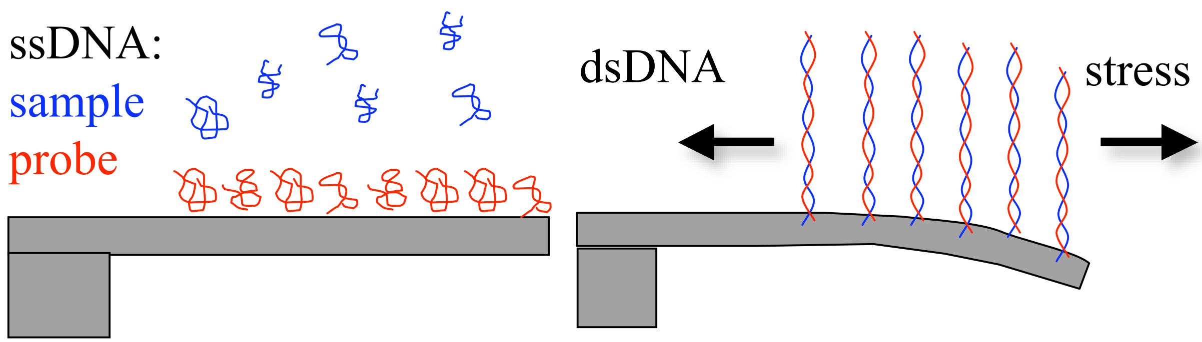
alkanethiols on Au nanoclusters

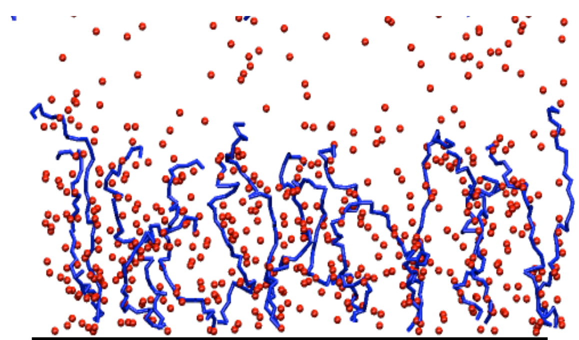
Coupling Bio & Silico Systems

- DNA & antibody cantilever sensors
  - What is the mechanism of stress transfer?
  - Bidirectional communication between bio & silica

CINT: Stevens

User: Jan Hoh, Johns Hopkins Med School





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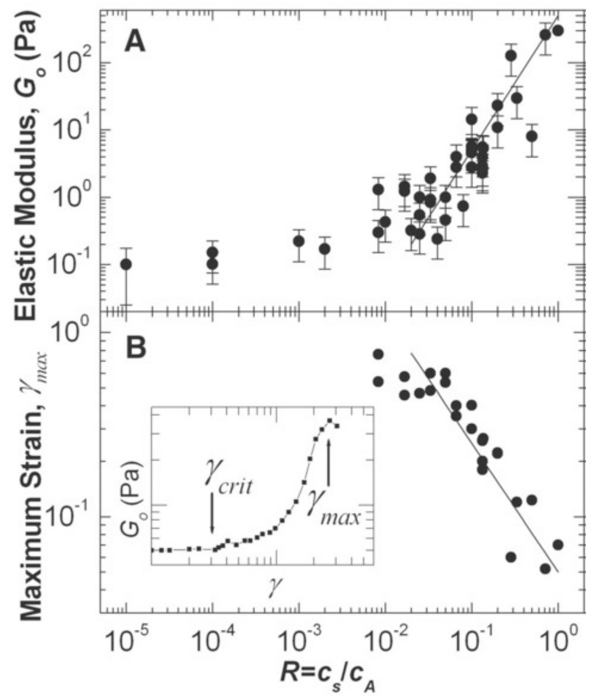
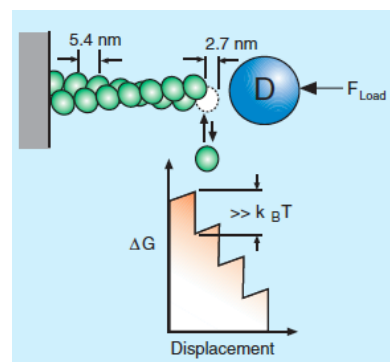
Nonequilibrium Dynamics & Assembly

- nonequilibrium assembly is important & a challenge
  - biomolecular processes tend to consume energy
  - part of SBCN work
  - energy transfer challenge
  - development of new methods in simulation
- actin networks
  - cytoskeleton
  - special network mechanical properties
    - semiflexible polymer
  - dynamical polymerization using GTP
    - monomer (G-actin) is protein (i.e. nanoparticle)
  - rachet force & membrane deformations
  - microtubules have same features
    - G. Bachand, CINT, SBCN





Mogilner & Oster, Current Biology 2003

CINT: Grest, Stevens
user: A. Levine, UCLA

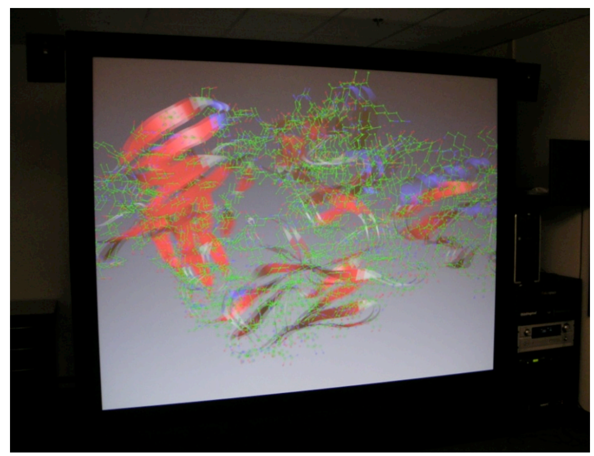
Semiconductor Crystal Growth

- Device properties depend on nanostructure within material
- Bulk structure depends on surface reconstruction during growth
  - layer structure 'frozen in' during growth process
  - surface has nanoscale domains
- powerful tools (STM) can probe surface nanostructure
- interpretation and understanding require theory
  - semiconductor crystal growth as competing interactions
- Example of the Theory user process
  - Project of Modine (CINT) with J. Mirecki-Millunchick's experimental group at U. Michigan
  - Jessica Bickel (Student) visits CINT
  - get up to speed in performing simulation
  - return to home university and run jobs remotely

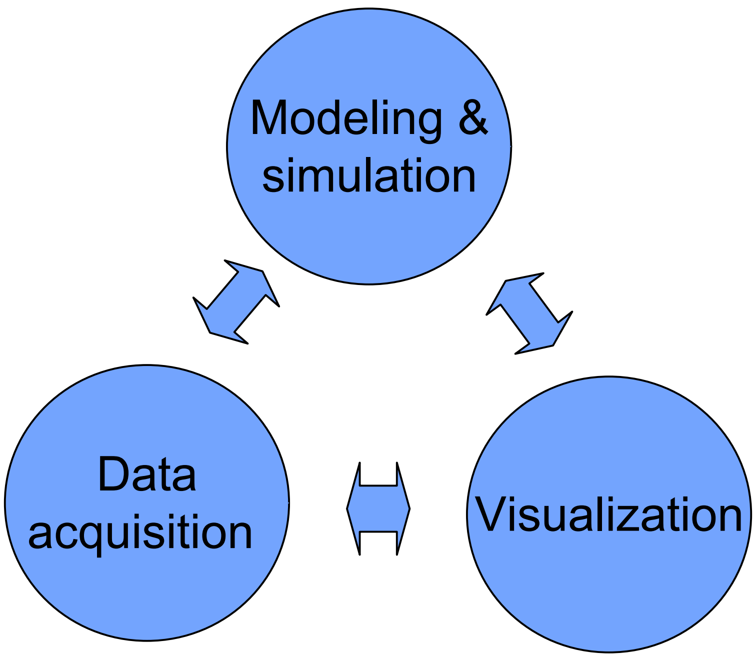
Capabilities

- High performance computing
  - parallel computers & codes
  - treat complex systems
  - quantum to classical methods
- Visualization
  – tour

PixelVizion cluster
(2006 R&D 100)

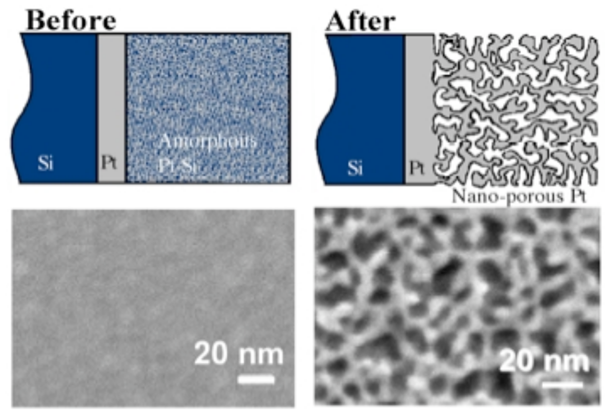


Fakespace rear-projection system

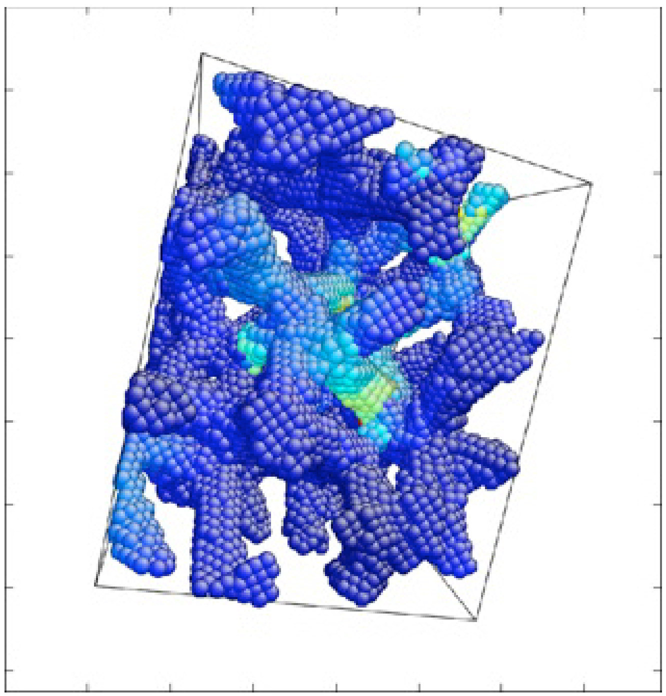


Visualization of Nano-foam

- ultra-high strength nano-foam
- atomistic modeling bases on foam nodes and struts
- new deformation mechanism:
  - plastic deformation nucleates at free surfaces



Picraux, Misra, Demkowicz



Light-colored areas show atoms with large displacements during the plastic deformation event

Thrust Overview

SBCN
NPON ⟷ **Soft Materials**

Classical interactions

–nanoparticle assemblies
–energy driven dynamics and assembly
–surface & interfacial interactions

Hard Materials ⟷ NEMS
NPON

Quantum interactions

–nanoscale inhomogeneity
–quasiparticles in nanostructures
–low dimensional quantum states
–excited states & dynamics
–nanodomains