SAND2016-4253PE

Thrust Overview

Theory & Simulation of Nanoscale Phenomena (TSNP)

Gary S. Grest

SAND: xxxx

TSNP Personnel

Synthesis | Fabrication | Characterization | Theory

Gary S. Grest -- TL
0.75 FTE

Sergei Tretiak
0.5 FTE

Amalie L. Frischknecht
0.5 FTE

Stuart A. Trugman
0.5 FTE

Normand A. Modine
0.5 FTE

Jianxin Zhu – PSL
0.6 FTE

Mark J. Stevens
0.5 FTE

Post Docs, 1.0 FTE

K. Michael Salerno (SNL)
TBA (LANL)

Total: 4.85 FTE

Los Alamos NATIONAL LABORATORY

U.S. DEPARTMENT OF ENERGY | Office of Science

Sandia National Laboratories

Vision of Thrust

- **Understand and predict functionality through nanoscale integration**

- **What is the underlying mechanism of nanoscale integration for tailoring functionality in composite materials – nanocomposites and heterostructures?**
- **How to optimize nanoscale integration through the control of interactions?**
  - **weak enough to keep unique properties of nano component**
  - **strong enough to give new, collective behavior**

- **TSNP develops core capabilities to tackle these key questions and supports a wide CINT user community and CINT science**

TSNP Thrust Science Directions

- **Hierarchical Structure and Dynamics in Soft Matter**
- **Excitation and Transport in Nanostructured Systems**
- **Emergent Behavior at Surfaces and Interfaces**

>100 Users
143 CINT Publications
31 High Impact

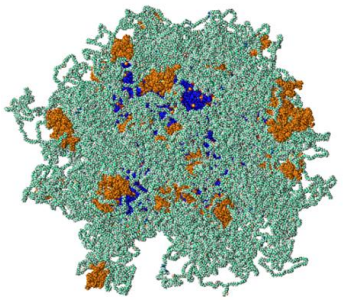
TSNP Integration Science Challenges

How does one control the interactions in integrated nanostructures to:

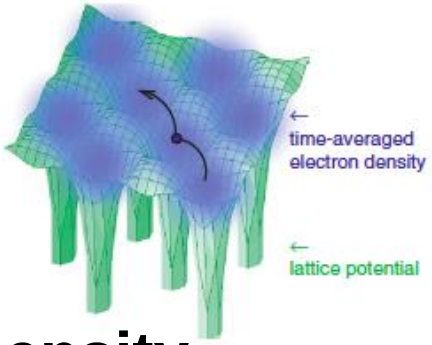
- **Enable hierarchical assembly of individual nano-constituents to harness their collective behaviors**
- **Control energy transfer across interfaces and on multiple length scales**
- **Generate new emergent phenomena from competing interactions**

Foundational Capabilities

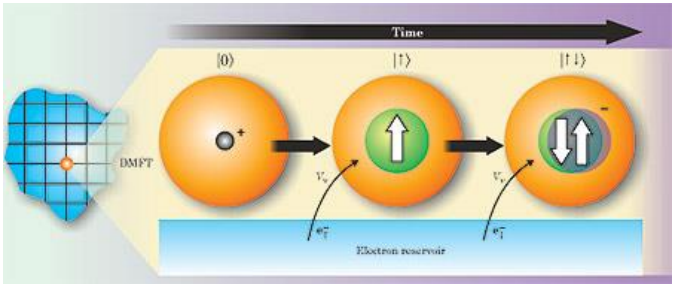
- Molecular Dynamics and Monte Carlo Simulations
- Classical and Quantum Density Functional Theory
- Dynamical Mean-Field Theory for Strongly Correlated Electronic Systems



Molecular Dynamics



Density Functional Theory

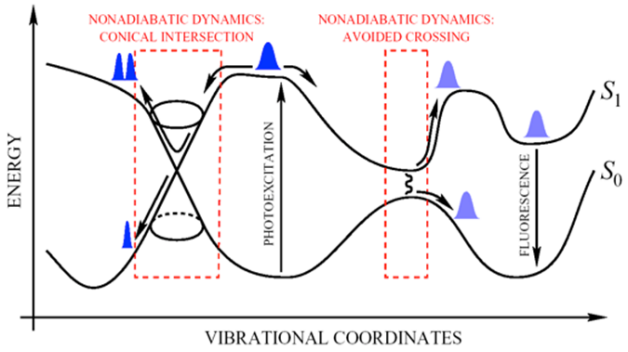


Dynamical Mean-Field Theory

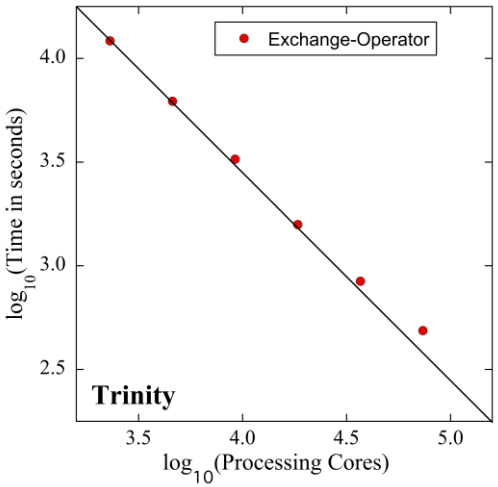
- Continuing No Cost, Institutional Support for Computation
> $1 million/year

Differentiating Capabilities

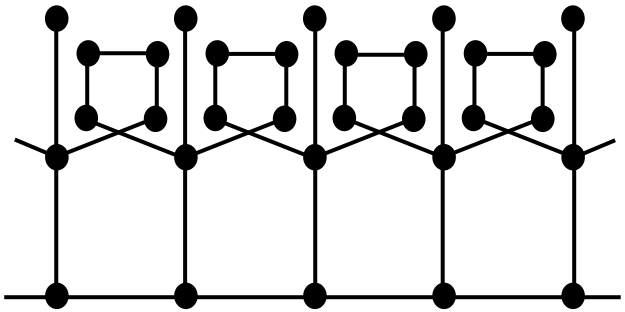
- Non-Adiabatic Excited State Molecular Dynamics for Molecules
- DFT calculations with hybrid functionals scaling to >100,000 cores
- Time-dependent Lanzcos using an efficient Hilbert space



NA-ESMD



Socorro DFT



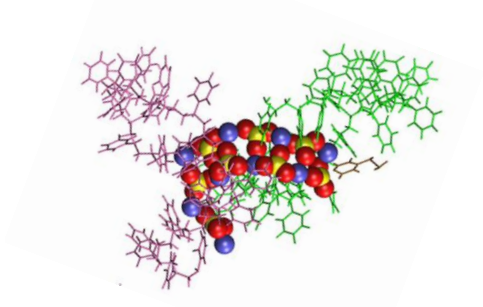
0

Time-dependent Lanzcos

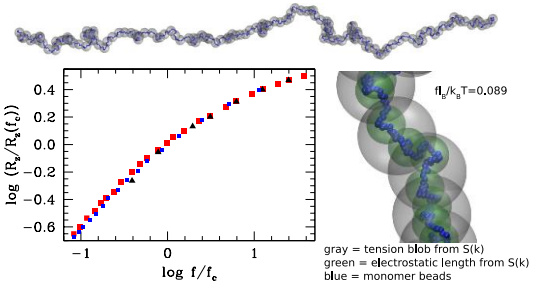
Hierarchical Structure and Dynamics in Soft Matter

- Propagate intrinsically unique behaviors of nanoscale materials into functional materials
- Internal structure on the nanoscale, strongly affects both equilibrium and dynamic material properties

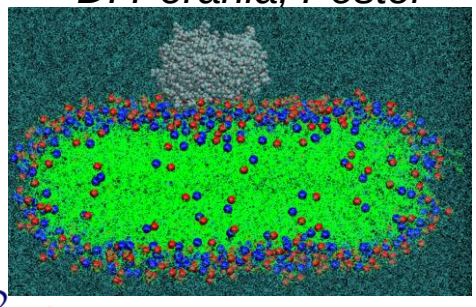
Ionomer Polymer Melts - Research Highlight



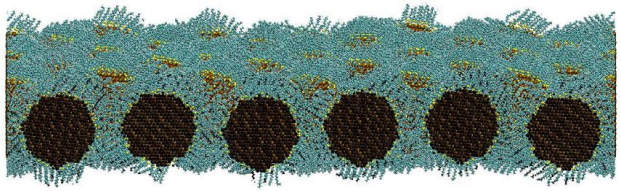
Single Molecule Biopolymer Mechanics - O.Saleh



Polydots At Lipid Membrane - D. Perahia, Poster



Nanoparticle Assembly K. M. Salerno, Poster



32 CINT Publications
11 with Experimentalists

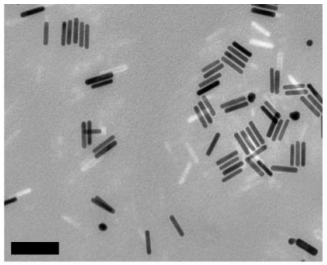
CINT Users: R. Composto (U Penn), A. Dobrynin (U Akron), G. Fredrickson (UCSB), S. Kumar (Columbia U), X.-M. Lin (Argonne), D. Perahia (Clemson U), M. Rubinstein (UNC), O. Saleh (UCSB), S. Rempe (SNL), J. Runt (Penn State), M. Tsige (U Akron), K. Winey (U Penn)

Los Alamos NATIONAL LABORATORY

U.S. DEPARTMENT OF ENERGY | Office of Science

Sandia National Laboratories

Research Highlight: Tuning Nanorod Assembly

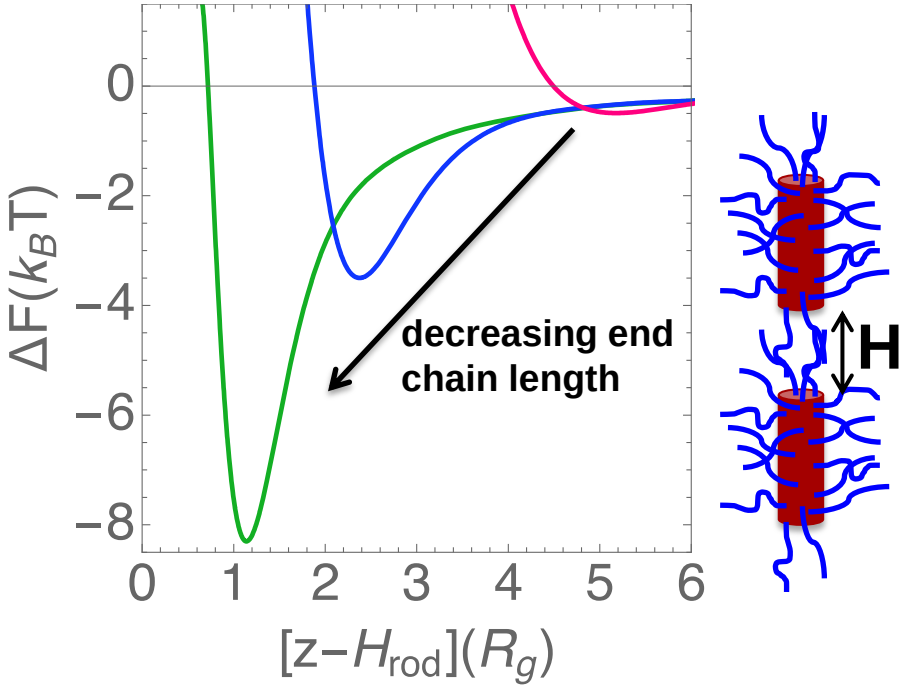


- Nanorods often align side-to-side
- Can we design end-to-end alignment in polymer-grafted nanorods in a polymer film
  - end-aligned gold NRs have large field enhancement between the rods, e.g. for sensing applications

SCFT calculations

- long polymer brush on sides
  - promotes wetting by matrix
  - dispersion side-to-side
- short polymer brush on ends
  - promotes matrix chains leaving brush
  - attraction end-to-end

-8 kT sufficient to form aggregates

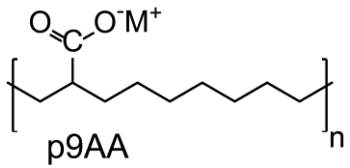


C. L. Ting* et al., Macromolecules 49, 1111 (2016)
CINT User R Composto (U Penn)
* Former Truman Fellow

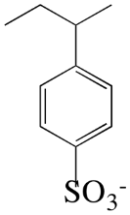
Research Highlight: Role of Ionic Clusters in Dynamics of Ionomer Melts

• Polymer substituted by ionizable groups whose properties are controlled by both the backbone and charges

- Selective transport
- Clean energy
- Water purification
- Sensors



Precise PE-acrylic acid

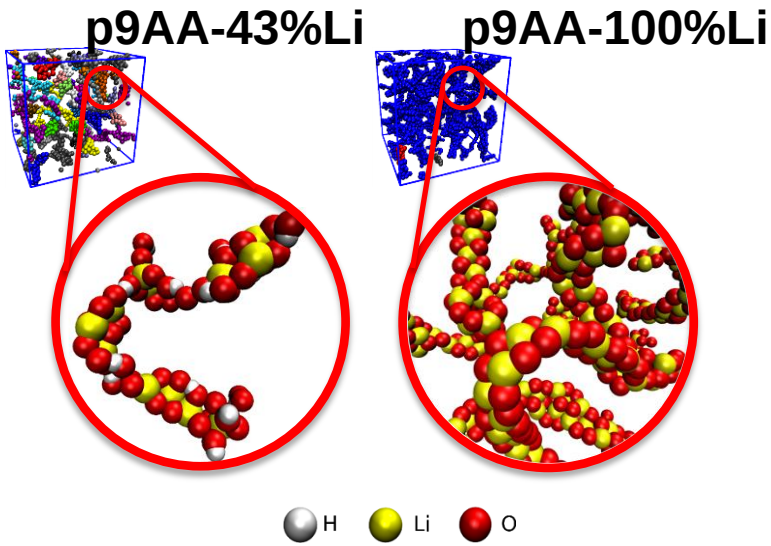


Polystyrene Sulfonate

Open questions:

- Factors that control cluster morphology
- Correlation between cluster morphology and dynamics in melts

Challenge: Under the conditions where transport is optimized, mechanical stability is often compromised

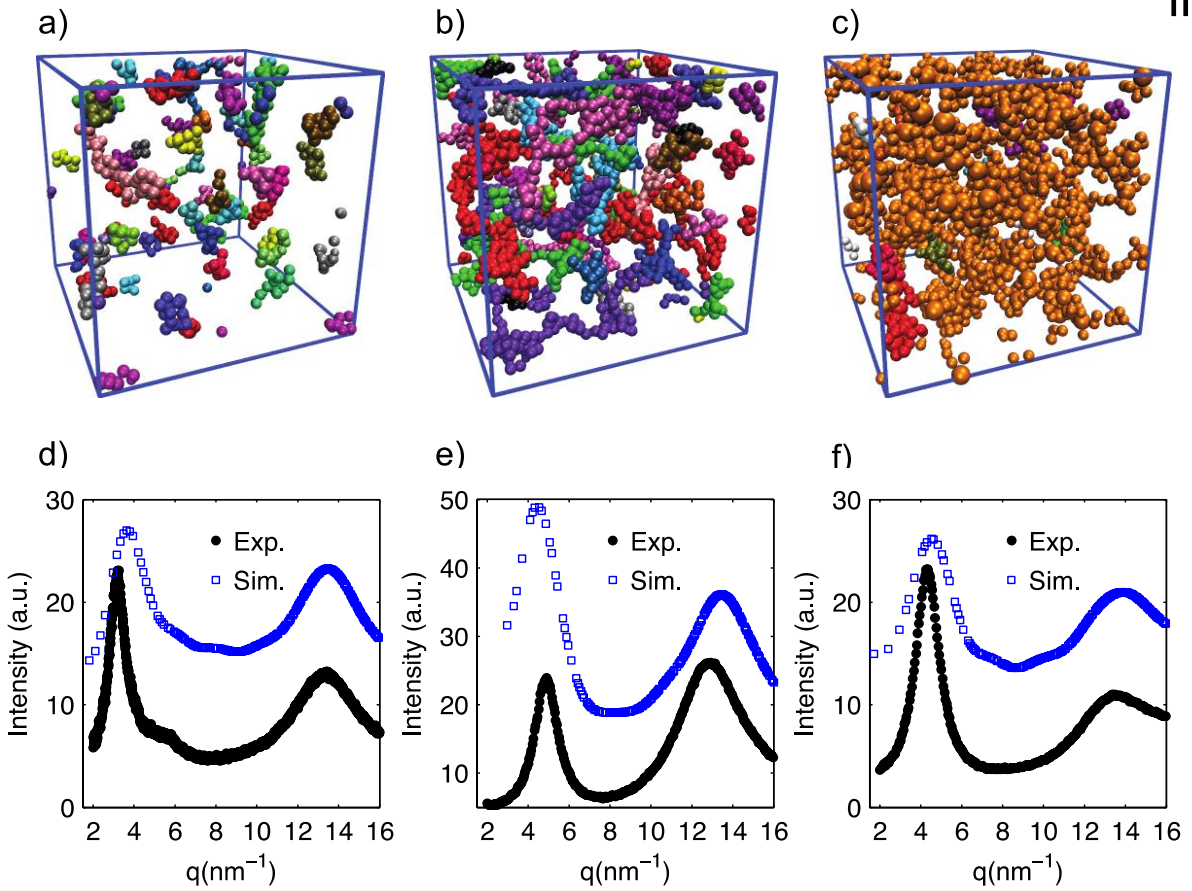


Research Highlight: Precise Ionomers

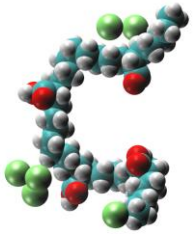
p21AA-56%Zn
Type 1

p9AA-43%Li
Type 2

p9AA-24%Cs
Type 3



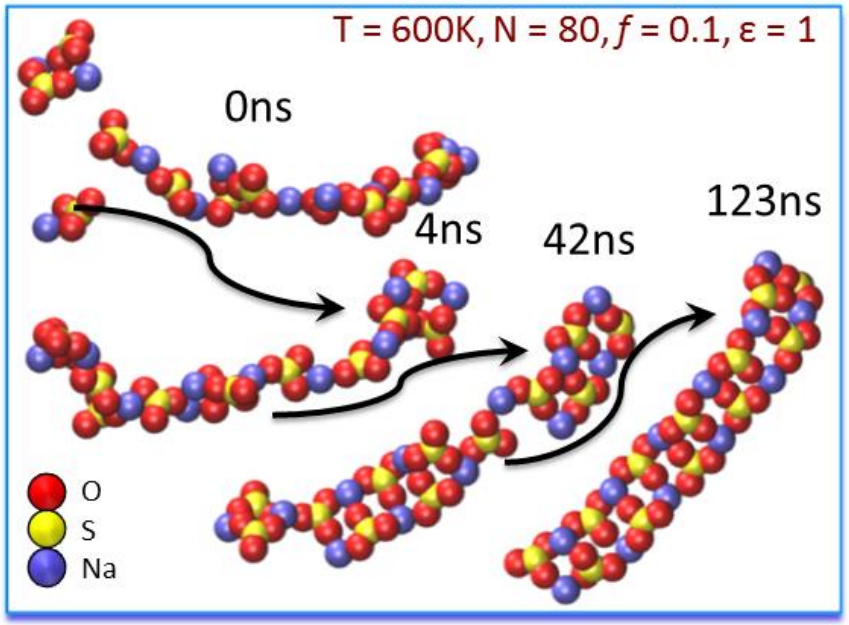
Ionic aggregates in atomistic MD

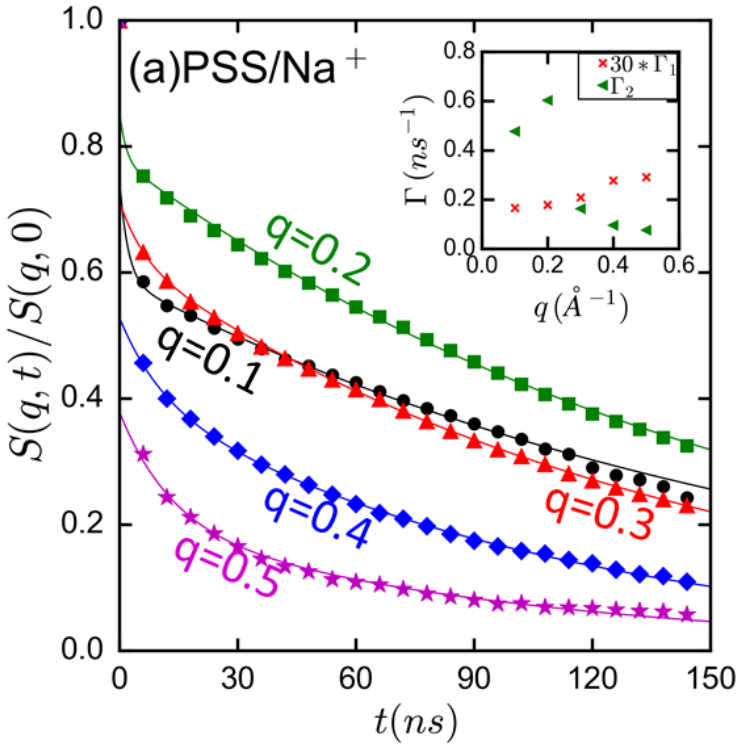


- Good agreement between X-ray & MD S(q)
- Simulations reveal variety of morphologies not discernable from X-ray data
- Modeling effect of morphology on dynamics underway

C. F. Buitrago *et al., Macromolecules* 48, 1210 (2015); CINT Users K. Winey (U Penn), J. Runt (Penn State)

Research Highlight: Polystyrene Sulfonate





- Ionic groups assemble into ladder like structures
- Ionic clusters control mobility

- Decay for q=0.2 Å^{-1} corresponding to the peak in S(q), slower than other q values

A. Agrawal et al. PRL 116, 158001 (2016), PRE 92, 022601 (2015). CINT User Perahia (Clemson U)

Future Science Vision – Soft Materials

- Behavior of systems of nanoparticles embedded in complex environments
  - Polymer Nanocomposites
  - Nanoparticle Assembly at Liquid/Vapor and Liquid/Liquid Interfaces
  - Field Directed Nanoparticle Assembly

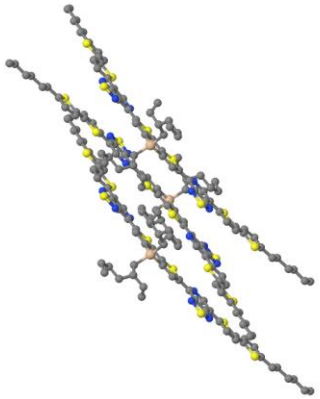
- Structure & Dynamics at the Nanoscale
  - Nanoscale Clusters in Ionic Polymers
  - Conductivity of Ionic Clusters in an Electric Field
  - Polymeric Ionic Liquids

- Coarse grained models from underlying atomistic models to expand spatial and temporal range while maintaining chemistry

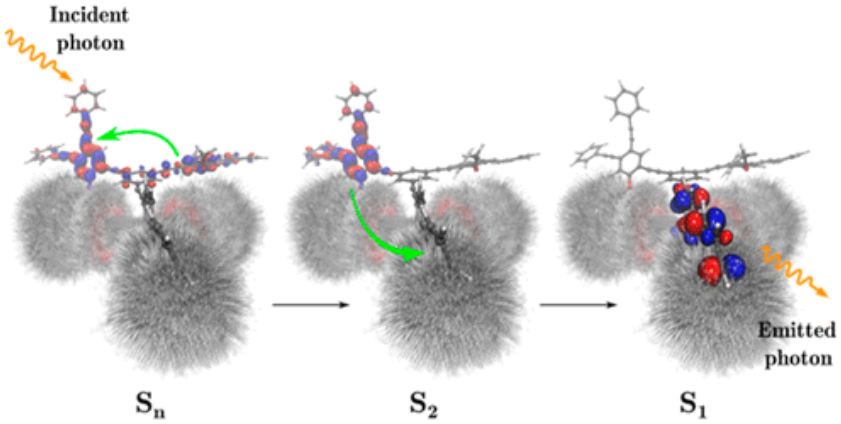
Excitation and Transport in Nanostructured Systems

- **Interactions enable and control transport**
- **Nanostructure controls interactions**

Excitation & Transport in Nanosystems Research Highlight



Photoexcited Dynamics in Conjugated Macromolecules Adrian Roitberg, User Talk

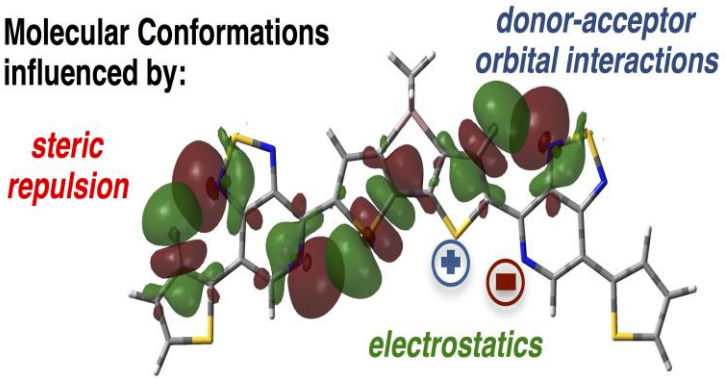


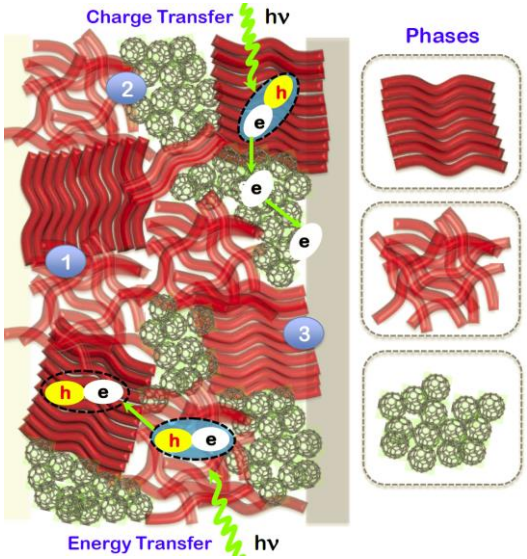
58 CINT Publications
23 w/ Experimentalists

CINT Users: J. Bonca (Ljubjana), V. Chernyak (Wayne State), X. Lin (Boston), S. Dexheimer (Washington State), A. Zhugayevych (Skolkovo IST), H. Lischka (Texas tech), H. Fehske and A. Alvermann (Greifswald), S. Kilina (N Dakota), C. Lo (Washington @ St Louis), Y. Zhao (NTU), S. Fernandez-Alberti (Nacional de Quilmes), A. Roitberg (Florida), J. Gierschner (Madrid IAS), O. Prezhdo (Rochester), C. Katan (CNRS), D. Kilin (S Dakota), E. Benassi (Scuola Normale Superiore)

Research Highlight: Excitation and Transport in Nanostructured Systems

- Determine flux of energy and charges in amorphous systems
- Multiscale theoretical methods developed to predict molecular morphology and calculate carrier transport on the device scale from first principles (NA-ESMD used)





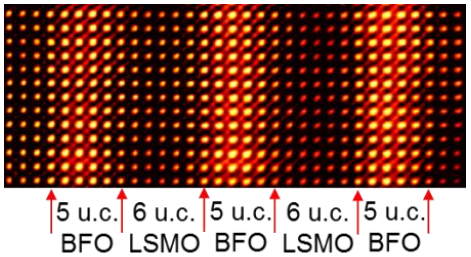
- Developed predictive capability to model carrier transport on multiple time- and length- scales

A. Zhugayevych and S. Tretiak, Annu. Rev. Phys. Chem. **66**, 305 (2015). 10 CINT publications with additional users.

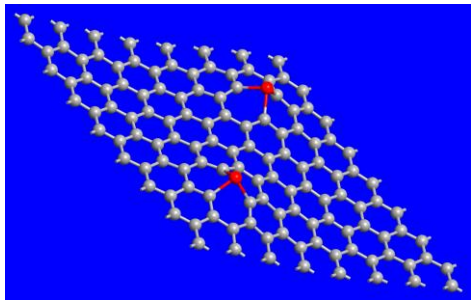
Research Highlight: Emergent Behavior at Surfaces and Interfaces

- Coexistence and competition between domains or phases lead to remarkable behavior
  - Transition-Metal Oxides (TMOs), 2D Materials

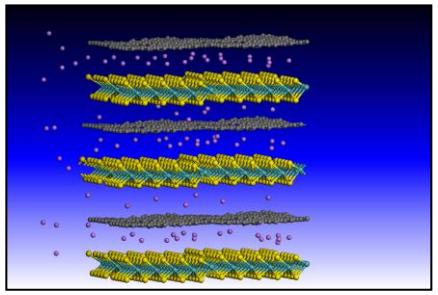
Induced Magnetism at Interfaces
- Research Highlight



Emergent Behavior at surface/interface
– J. Haraldsen Poster



Electronic Properties of 2D van der Waals Heterostructures
– T. Ahmed Poster

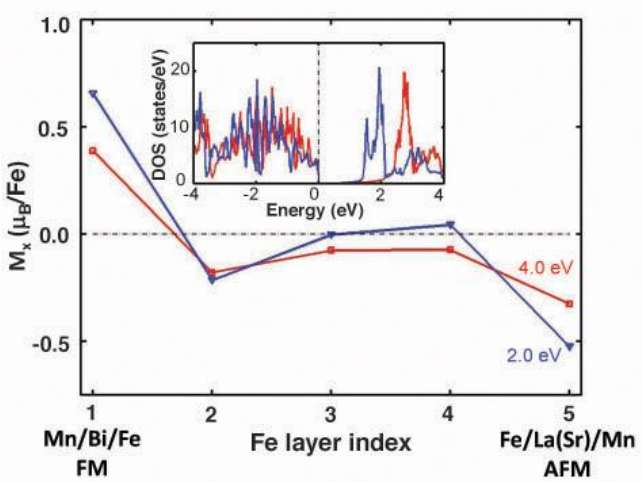


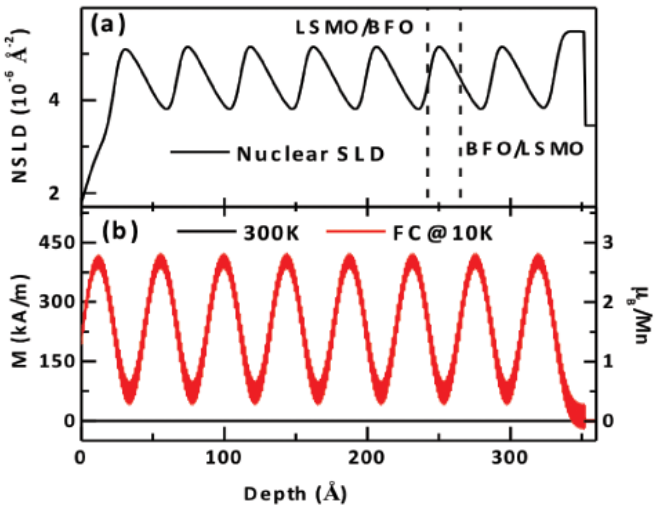
49 CINT Publications
24 w/ Experimentalists

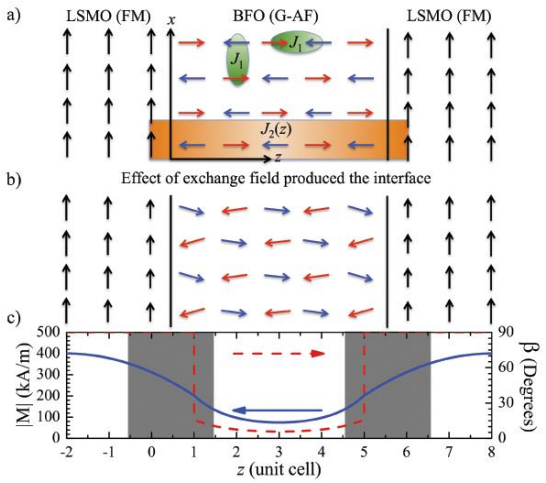
CINT Users: A. Kocharian (CalStat LA), R. Hatcher (Lockheed Martin and Samsung), T. Ahmed (LANL), J. Haraldsen (North Florida), E. Chia (NTU), C.-S. Ting (Houston), J. Millunchick (Michigan), S. Krishna (UNM), B. Malone and M. Stopa (Harvard), Q. Si (Rice), J.-P. Julien (Neel Inst), J. Fransson (Uppsala), G. Zhang (Indiana), F. Mancarella (KTH)

Research Highlight: Emergent Behavior at Surfaces and Interfaces

- Induced magnetism of TMO superlattice explained with first-principles theory and effective modeling
- Framework to address the key challenges in understanding of emergent behaviors at oxide interfaces
- Simulation/modeling leads to interpretation/prediction
- Integrated effort (TSNP, NEM, external user)



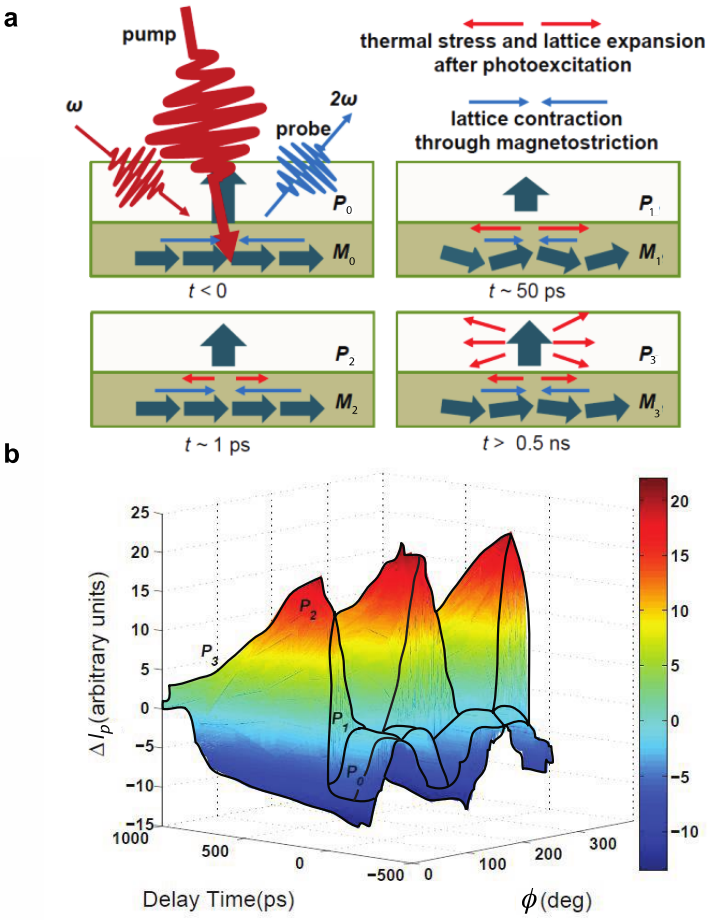




S. Singh, J.-X. Zhu, Q. Jia et al., *PRL***113**, 047204 (2014); CINT user J. H. Haraldsen

Research Highlight: Emergent Behavior at Surfaces and Interfaces

- Ultrafast dynamics phenomenological theory guides experiment to control magnetoelectric coupling in a FE/FM TMO heterostructure
- Theoretically identifying the role of spin-lattice relaxation for the timescale of ME coupling
- Integration effort (TSNP, NPON, NEM)



Y. M. Sheu, S. A. Trugman, R. Prasankumar et al. *Nat. Comm*. 5, 5832 (2014)

Future Science Vision – Hard Materials

- Dynamics and energy transport in hybrid nanostructured systems
  - Photoinduced dynamics, energy and carrier transport in hybrid nanostructured systems
  - Dynamical optical and magnetic properties with hybrid material interactions

- Emerging phenomena in integrated intrinsic and artificial nanoscale architectures
  - Competing phases at interfaces
  - Relationship between interfacial structure and emergent phenomena at upscale
  - Dynamic response out of equilibrium

TSNP Thrust Summary

- TSNP thrust is developing the essential knowledge and simulation capabilities needed to understand and design integrated nanotechnology

- TSNP thrust supports the other CINT thrusts and a diverse group of Users (experimental and theoretical)

User Talks/ Posters

Talks:

• Omar Saleh (UCSB) - Single Molecule Biopolymer Mechanics Across the Force Regimes

• Adrian Roitberg (U Florida) - Non-Adiabatic Excited States Molecular Dynamics: Photoexcited Dynamics in Conjugated Macromolecules

Posters:

• Towfiq Ahmed (LANL) - Electronic Properties of 2D Van der Waals Heterostructures

• Jason Haraldsen (U North Florida) - Emergent magnetism from transition-metal interactions at interfaces and surfaces

• Dvora Perahia (Clemson U) - Internal Correlations and Stability of Polydots, Soft Conjugated Polymeric Nanoparticles

• K. Michael Salerno (Sandia) - Influence of Nanostructure and Environment on Nanoparticle Membrane and Superlattice Mechanical Properties