LA-SiGMA

Louisiana Alliance for Simulation-Guided Materials Applications

SD1: Electronic and Magnetic Materials

Extend local approximations [Density Functional Theory (DFT) and Dynamical Mean Field Approximation (DMFA)] to multi-scale approaches

FOCUS 1

Multiscale Methods for Strongly Correlated Materials

FOCUS 2

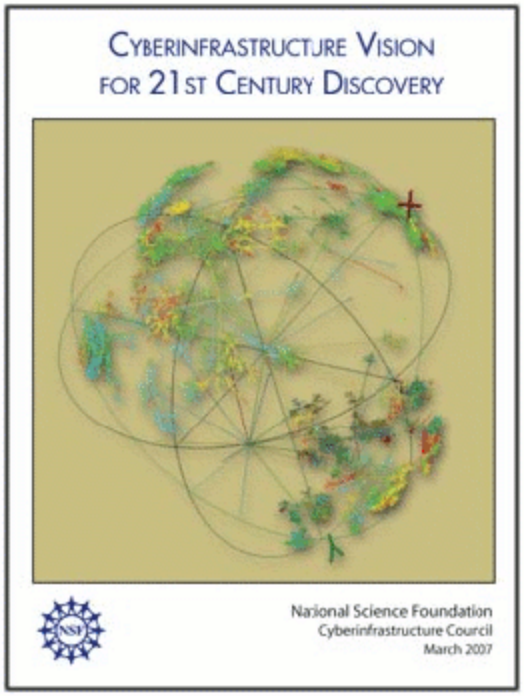
Correlated Organic & Ferroelectric Materials

FOCUS 3

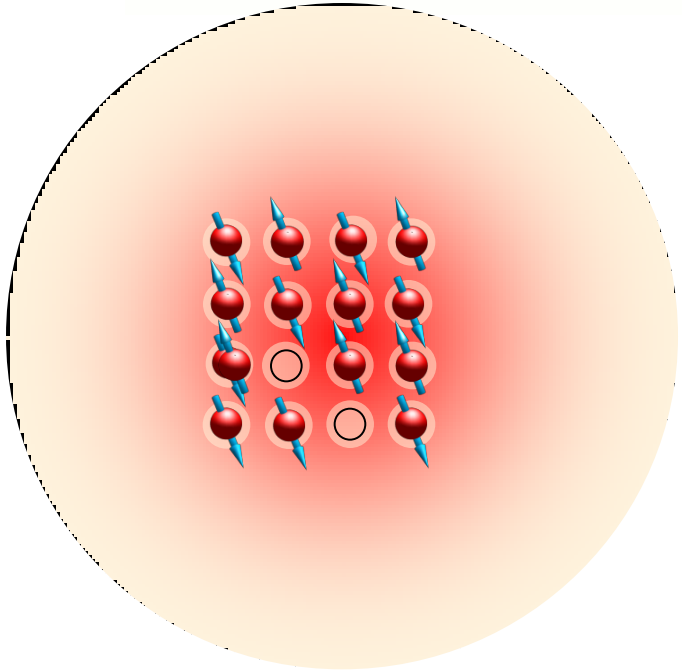
Superconducting Materials

Xavier University of Louisiana · Southern University · LSU Louisiana State University · Louisiana Tech · Grambling State University · UNO University of New Orleans · Tulane University

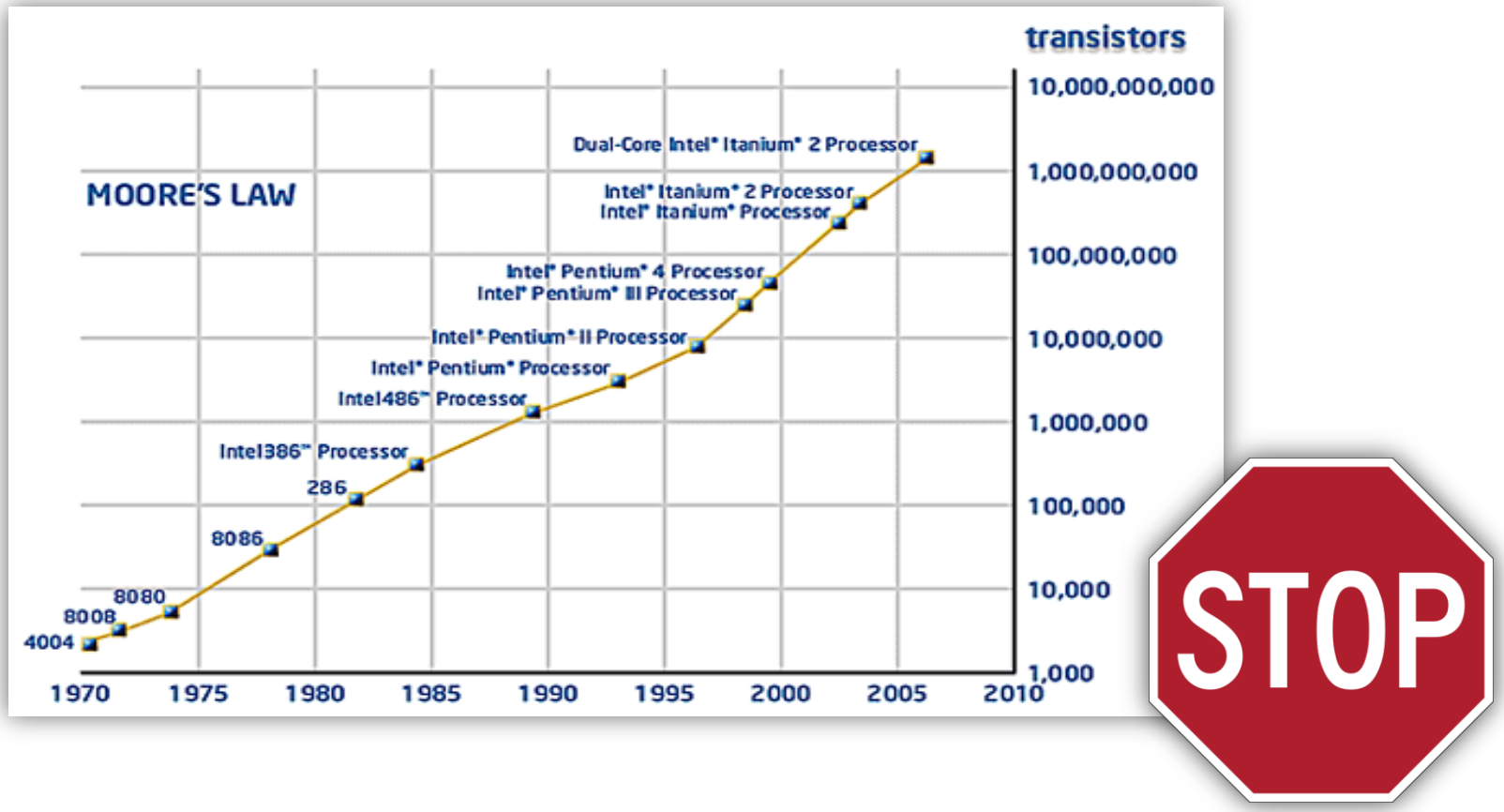
Computational Materials Science



- Petascale computing & the development of new formalism, algorithms and codes will allow the accurate modeling of materials.
- Calculations leverage new petascale and heterogeneous computing, bringing new problems to the tipping point of discovery.



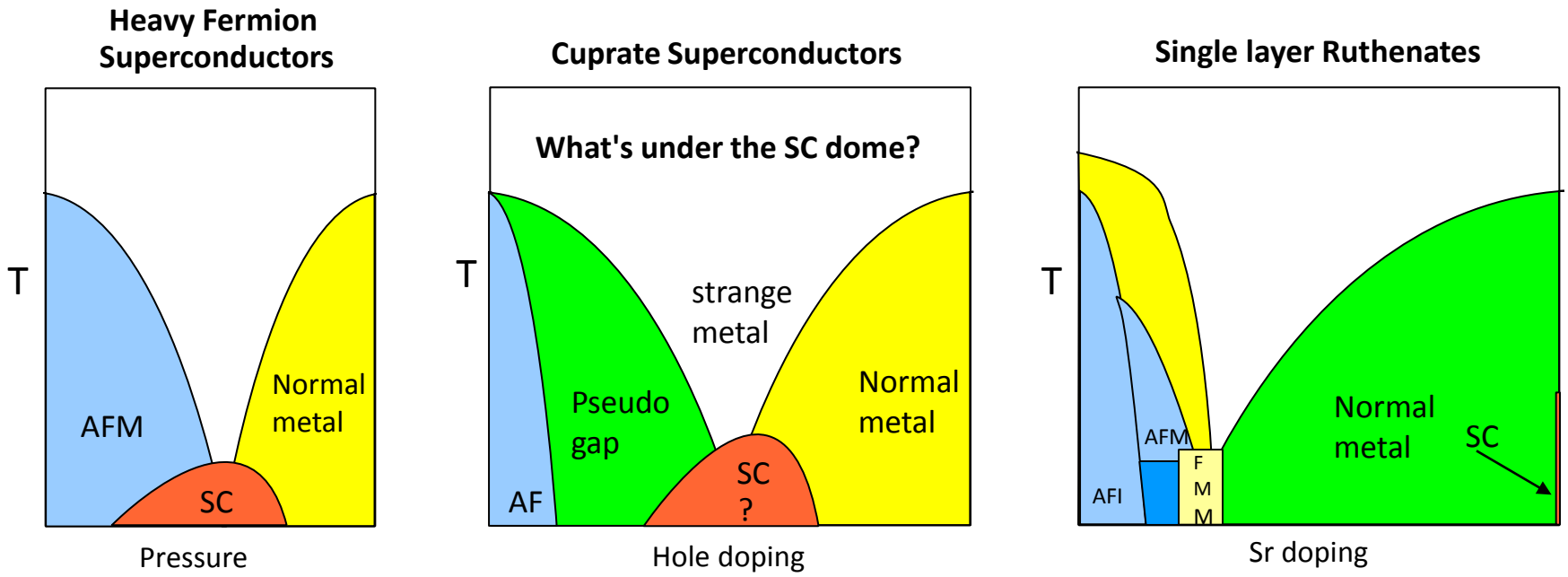
We need faster, smaller, more efficient chips



By 2020 a transistor in a chip may reach the size of a few atoms.

Devices based on a new paradigm are needed!

Strongly Correlated Systems: Complexity and Competing Orders

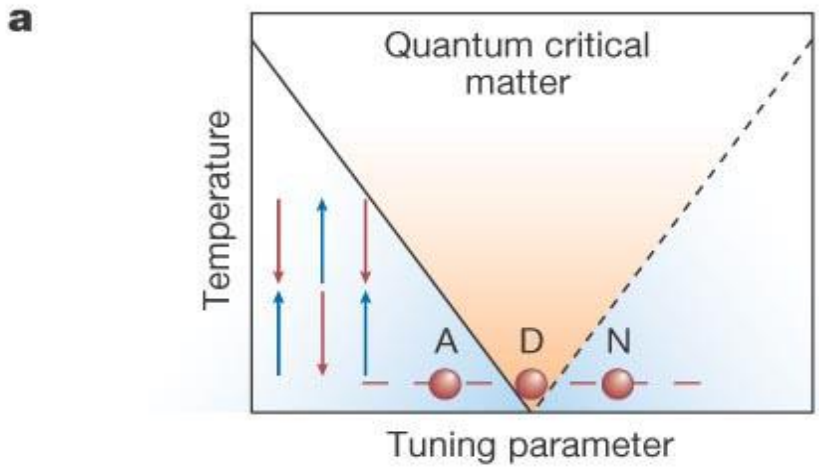


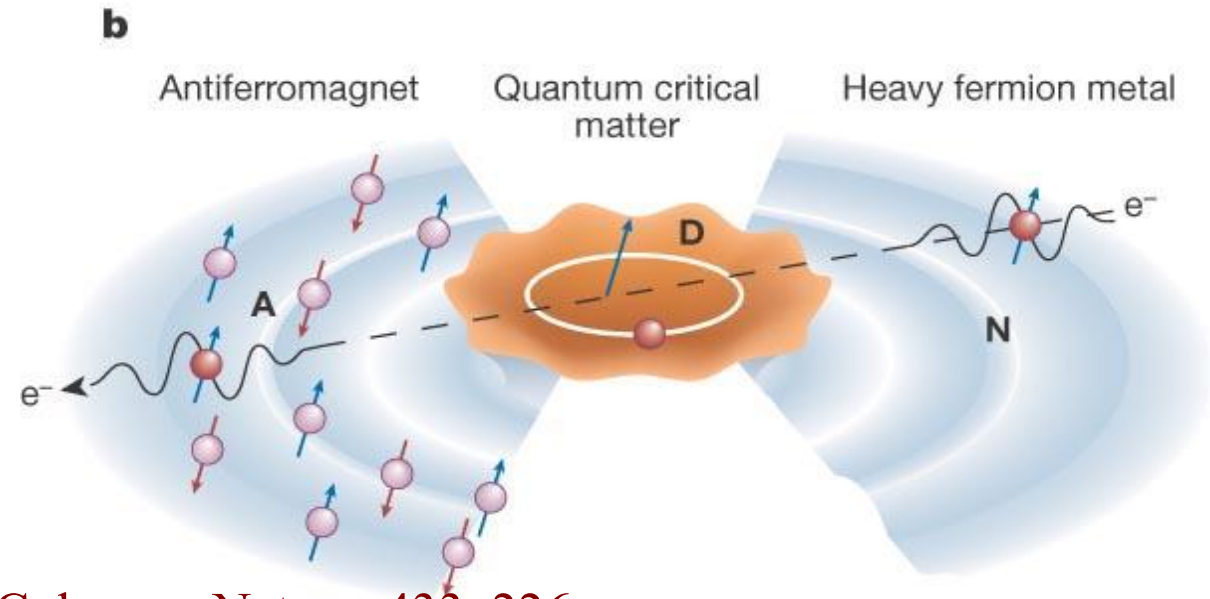
E. Dagotto, Science 2005

Complex metal oxides have potential for applications as emerging research memory and logic devices. A special set of the complex metal oxides, strongly correlated electron state materials, and their heterointerfaces may have potential to enable new logic devices with coupled spin and charge properties, ***ITRS 2007***

- Electron correlations lead to complex spin, charge, & orbital phases.
- Competing phases emerge as a function of control parameter.
- Tunability allows device applications.
- Paradigm shift: bandgap → spectral function engineering

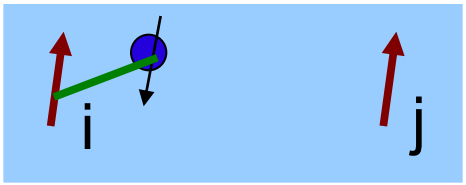
Simplest Case: Competition Between Exchange and Screening





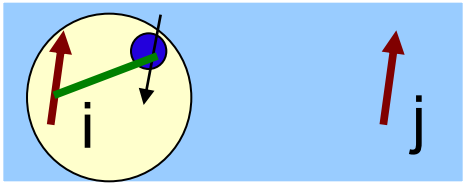
Coleman, Nature, 433, 226

- RKKY exchange



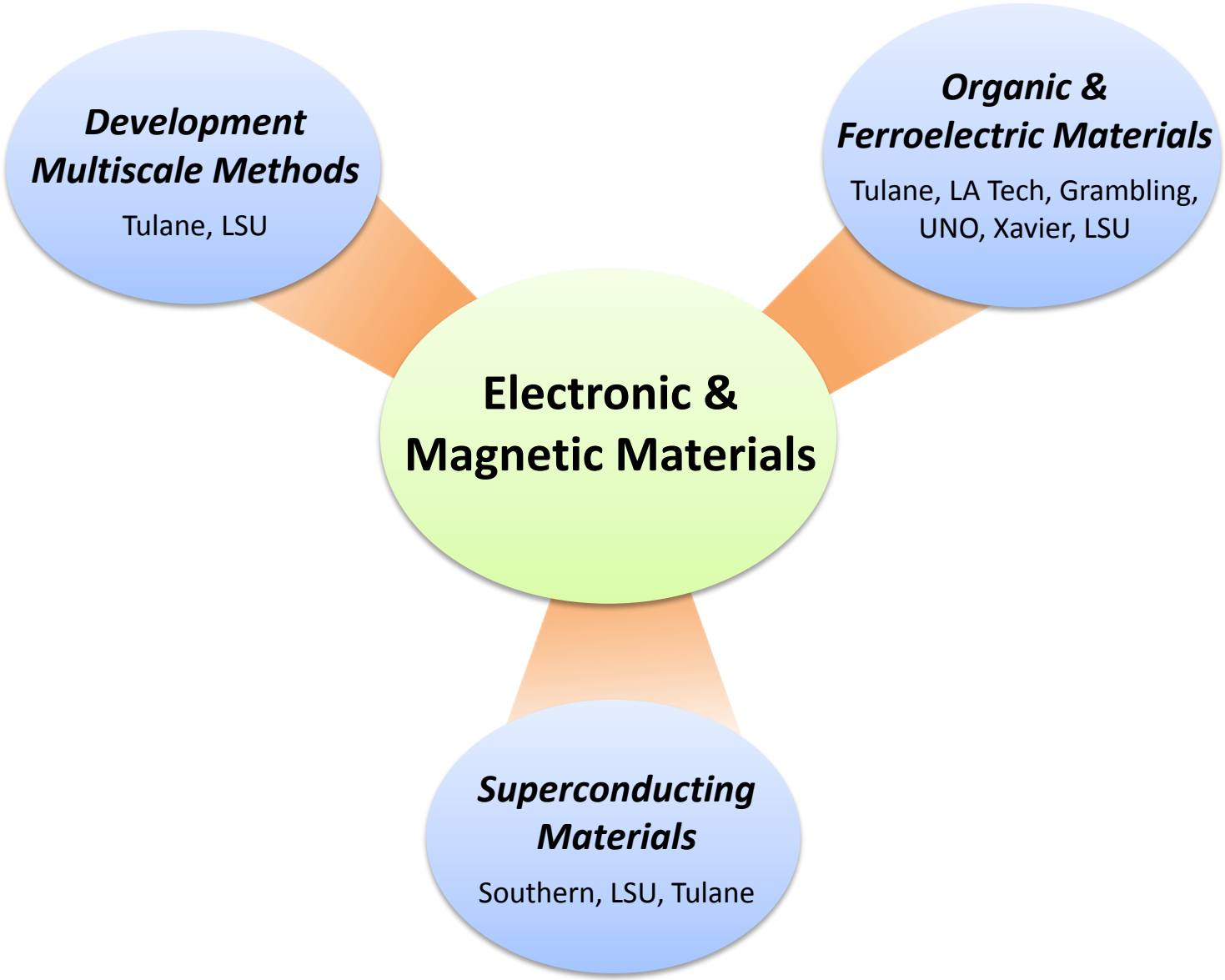
$J_{ij} \sim J^2$

- Kondo effect



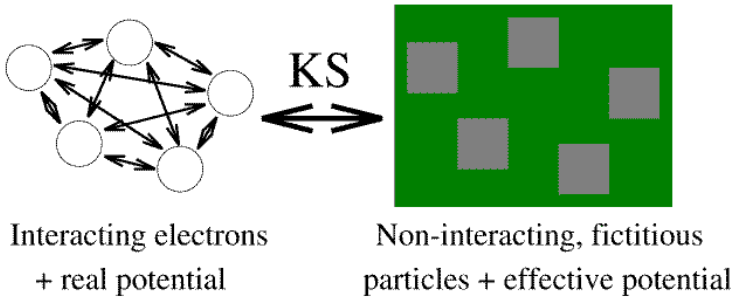
$T_K \sim e^{-1/\cdot(0)J}$

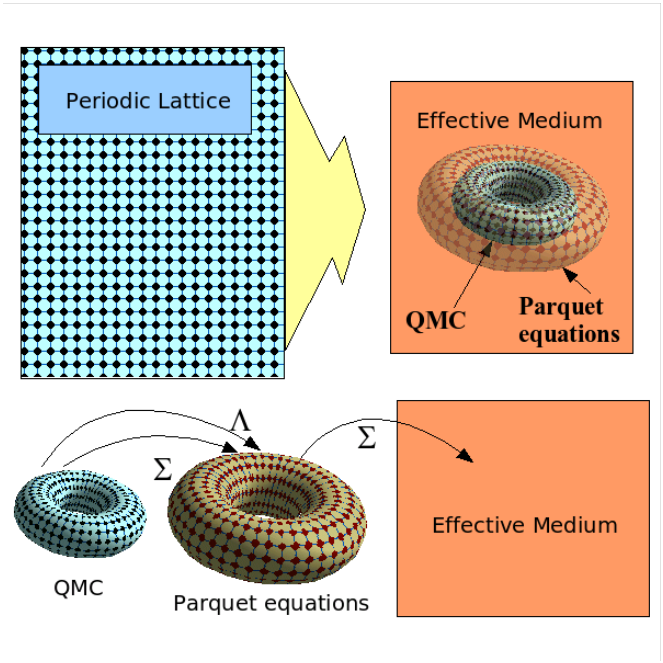
SD1 Research Themes



Focus 1

Development of Multiscale Methods for Strongly Correlated Materials





- Non-local Approximations for Density Functional Theory
- Multi-scale Many Body Methods
- Combinations of Both
- Validation
  - Iron Based Superconductors
  - Organic Magnets
  - Porphyrins
  - Inverse LEED method for structural investigations (LI/LA-SiGMA)

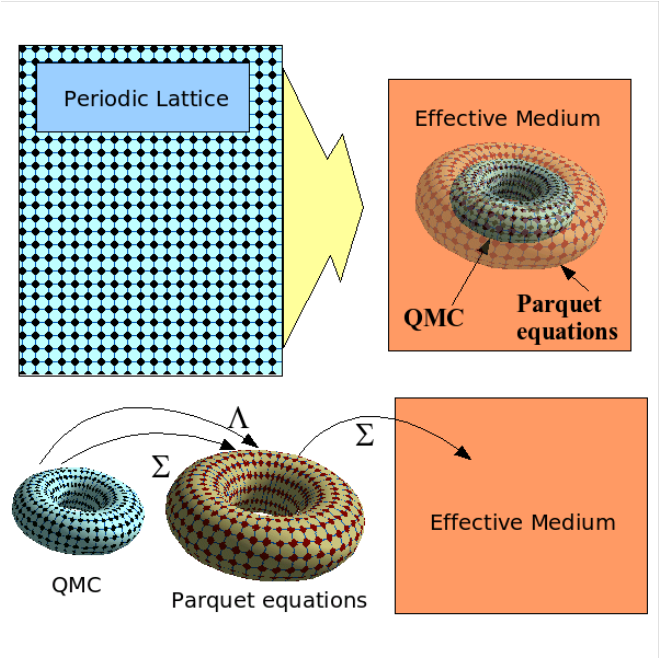
SD1; F1

Computing at the Petascale: Multiscale Many Body

Dual Fermion Dynamical Cluster Approach for Strongly Correlated Systems, S.-X. Yang, H. Fotso, H. Hafermann, K.-M. Tam, J. Moreno, T. Pruschke, M. Jarrell, arXiv:1104.1739.

Solving the Parquet Equations for the Hubbard Model beyond Weak Coupling, K.M. Tang, S.-X. Yang, H. Fotso, J. Moreno, J. Ramanujam, M. Jarrell. In preparation.

- QMC scales exponentially with problem size.
- Multi-Scale Many-Body scales algebraically.
  - QMC for short length scales
  - Dual-Fermion diagrammatics for intermediate length scales
  - Mean-field approximation for long length scales



SD1; F1

Density Functional Theory: Improved Approximations For the Exchange-Correlation Energy

John P. Perdew and Adrienn Ruzsinszky, Tulane University; Mark Jarrell, LSU

Kohn-Sham density functional theory predicts in principle the exact ground-state energy E, electron spin densities $n_\uparrow(\vec{r}), n_\downarrow(\vec{r}),$ and Born-Oppenheimer forces on the nuclei, for any many-electron system, by selfconsistent solution of a one-electron Schroedinger equation. In practice, the exchange-correlation energy $E_{xc}[n_\uparrow, n_\downarrow]$ must be approximated. The Kohn-Sham orbitals and orbital energies are also a starting point for more explicit many-body methods (GW, RPA, DMFT, QMC, etc.).

Semilocal approximations (e.g., revTPSS meta-GGA)

$$E_{xc}^{s/}[n_\uparrow, n_\downarrow] = \int d^3 r e_{xc}(n_\uparrow(\vec{r}), n_\downarrow(\vec{r}), \nabla n_\uparrow(\vec{r}), \nabla n_\downarrow(\vec{r}), \tau_\uparrow(\vec{r}), \tau_\downarrow(\vec{r}))$$

SD1; F1

Nonlocal approximations

Needed to describe, e.g., stretched bonds over which electrons are shared, and for many strongly-correlated systems at equilibrium.

Global hybrid functionals

$$E_{xc}^{gh} = aE_x^{exact} + (1-a)E_x^{sl} + E_c^{sl} \qquad (0 \le a \le 1),$$

with $a = 0.1$ for the revTPSS hybrid, are widely used. But extra exact exchange (bigger a) is needed when bonds are more stretched.

Local hybrid functionals

$$E_{xc}^{lh} = \int d^3 r \left\{ a(\vec{r}) e_x^{exact}(\vec{r}) + \left[1 - a(\vec{r})\right] e_x^{sl}(\vec{r}) + e_c^{sl}(\vec{r}) \right\}$$

Are promising, because they can satisfy nearly all known exact constraints on E_{xc}. We are trying to find improved density functionals for $a(\vec{r})$, to be used with improved semilocal approximations.

SD1; F1

RANDOM PHASE APPROXIMATION (RPA)

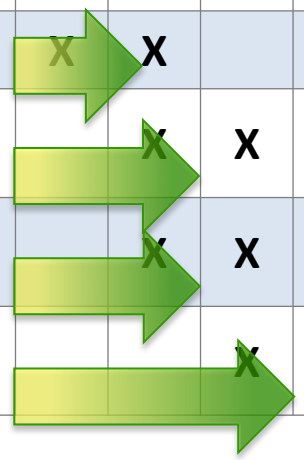
At the same time, a low-level many-body method and a high-level density functional method (since the Kohn-Sham orbitals and orbital energies are functionals of the density). It is now available in VASP. We will test RPA and corrected RPA for solid state problems.

WE WILL BUILD OUR NEW FUNCTIONALS INTO VASP AND GAUSSIAN.

SD1; F1

SD1 Focus 1 Milestones and Score Card

Milestones	Y1	Y2	Y3	Y4	Y5	
Develop QMC solver for GPU accelerated supercomputers.	X	X				*On Track*
Incorporate Hyper-GGA functionals into common DFT codes including VASP.		X	X			*Ahead of Schedule*
Develop MSMB solver able to treat multiple correlated orbitals.		X	X	X		*Ahead of Schedule*
Port hyperparallel codes to NSF national leadership class machines (Blue Waters).			X	X	X	*Ahead of Schedule*



SD1; F1

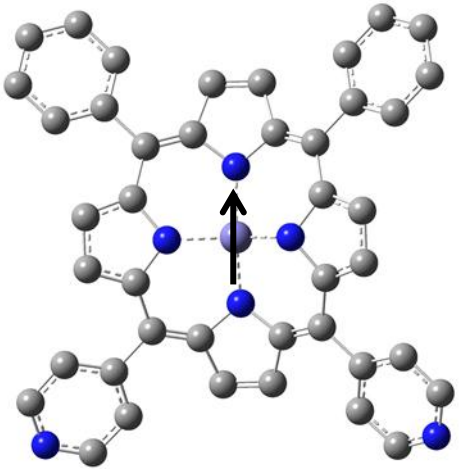
Focus 2

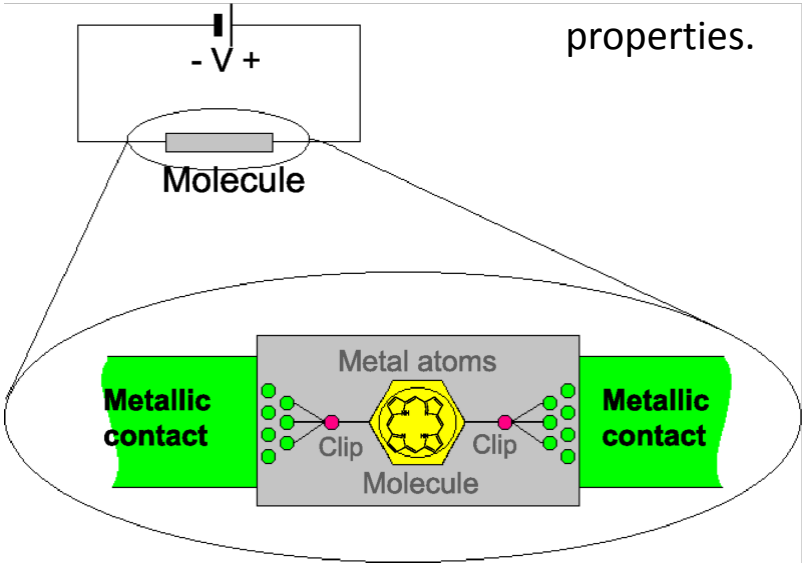
Correlated Organic & Ferroelectric Materials

Magnetic and Electronic Properties of Organic Semiconductors

Spins + Electronics = Spintronics might replace current electronics.

Some molecules, such as porphyrins, can be combined with a number of different atomic ions resulting in very different electronic and magnetic properties.





Objective: Test of existing and implementation of new methods to accurately predict magnetic, electronic, and transport properties of organic semiconductors.

SD1; F2

MetalloPorphyrins, a test system

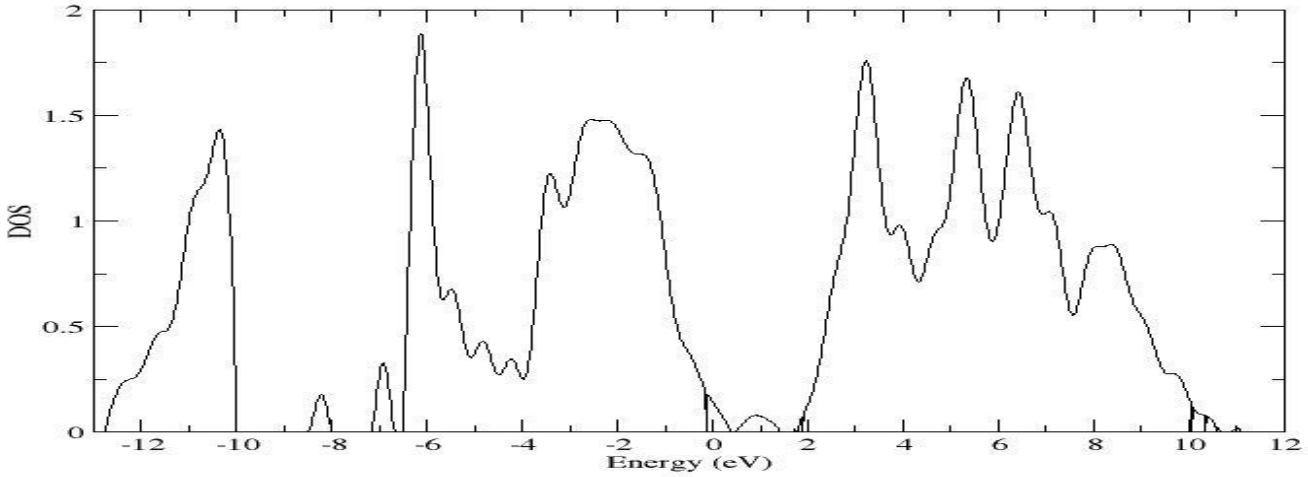
BROOKHAVEN NATIONAL LABORATORY

- A number of DFT functionals will be tested to determine their accuracy.
- Using down-folding methods we will extract appropriate parameters from the DFT calculation to build effective Hamiltonians.
- Those Hamiltonians will be used to predict magnetic and electronic properties using the Dynamical Mean Field Approximation.

Collaboration with Brookhaven National Lab

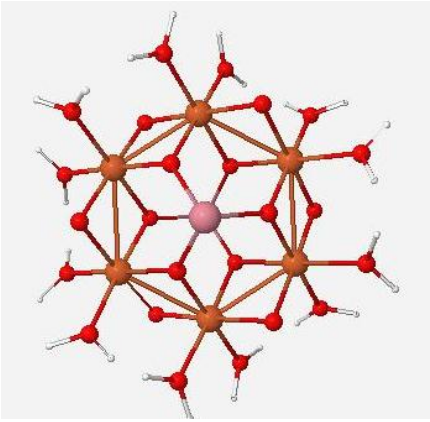
- Testing down-folding methods in dilute magnetic semiconductors.

LDA+DMFT for Magnetic Semiconductors and Metalorganics (Nelson et al.)



Iron Oxide Molecular Clusters as Building Blocks of Non-Volatile Memory (Xavier, Tulane)

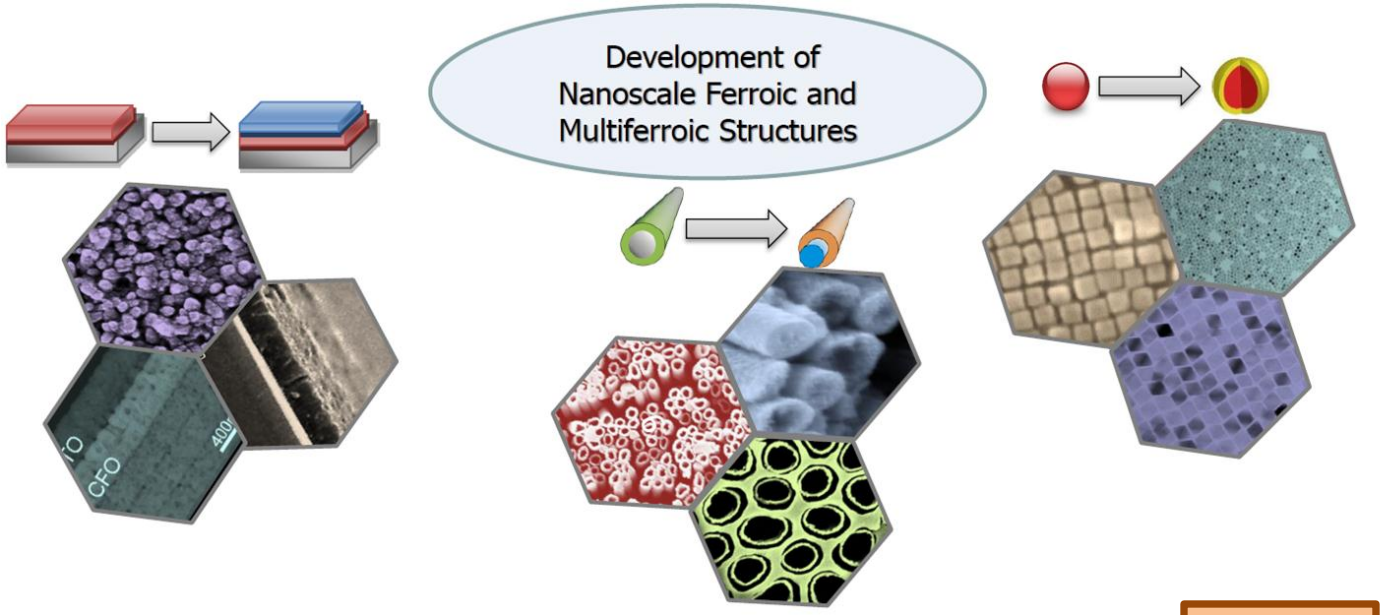
Elementary clusters derived from spinel-type structures (magnetite, ferrites etc) are candidates for molecular magnets behavior.



$Co_2Fe_6H_{24}O_{24}$ cluster. Spinel-type structure with water ligands.

Synthesis and Processing of Nanoscale Multiferroic Structures (UNO)

Maximize the coupling between the ferroelectric and magnetostrictive phases and enhance the ME response at room temperature



SD1; F2

SD1 Focus 2 Milestones and Score Card

Milestones	Y1	Y2	Y3	Y4	Y5	
Test array of DFT functionals for prediction of metalloporphyrin and ferroelectric properties.	X	X				*On Track*
Prepare and measure electrical/magnetic properties of metalloporphyrin nanostructures.	X	X				*On Track*
Prepare organic magnets and ferroelectrics.	X	X	X			*On Track*
Develop experimentally validated computational models for porphyrin systems using magnetoresistance and electrical conductance measurements as guides		X	X	X	X	
Develop multiscale models of metalloporphyrin systems using DFT parameters			X	X	X	
Predict charge transport in metalloporphyrins and compare with experiments			X	X	X	
Predict properties of ferroelectrics using new nonlocal meta-GGA DFT functionals			X	X	X	
Develop experimentally validated models of organic magnets and ferroelectrics			X	X	X	

SD1; F2

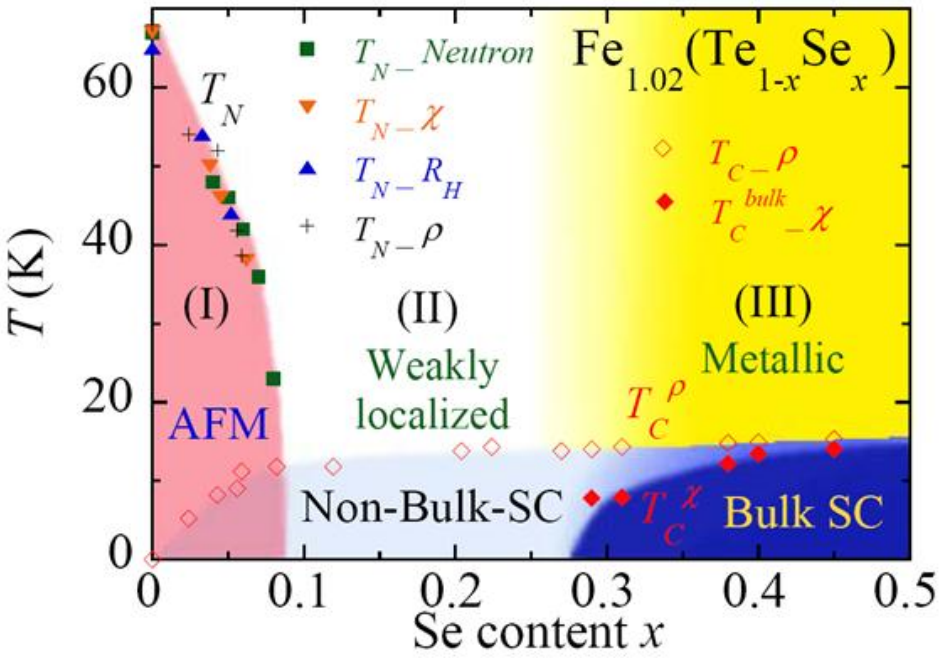
Focus 3

Superconducting Materials

What is the pairing mechanism in pnictides?
What is under the superconducting dome in the cuprates?

Mao (Tulane): Unusual interplay between magnetism and superconductivity in iron chalcogenide $Fe_{1.02}(Te_{1-x}Se_x)$

Bulk superconductivity occurs only when the (π, 0) magnetic correlations are strongly suppressed and spin fluctuations near (π, π) become dominant.

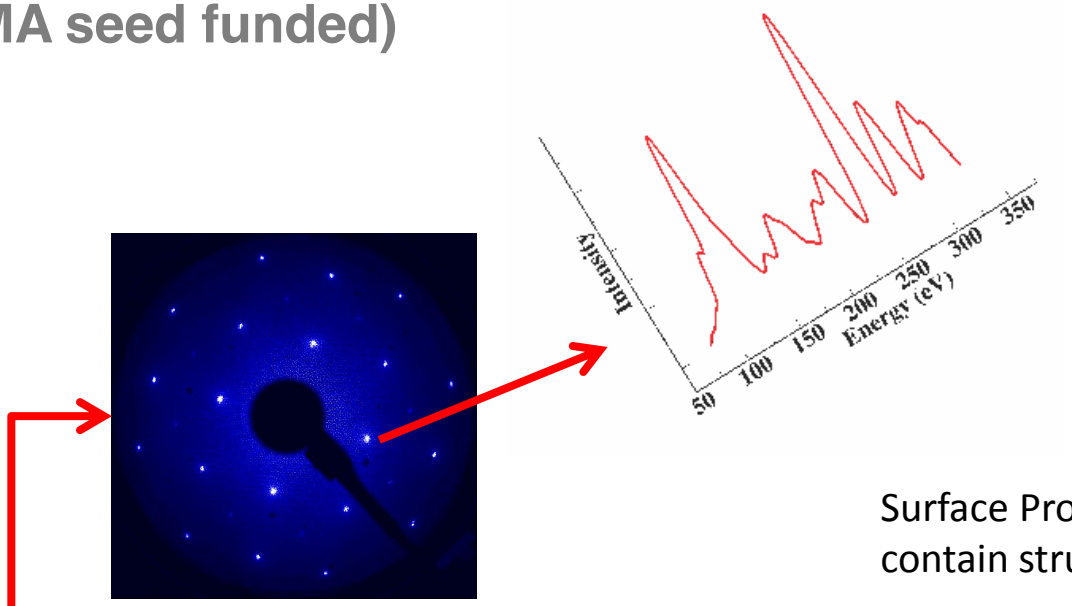


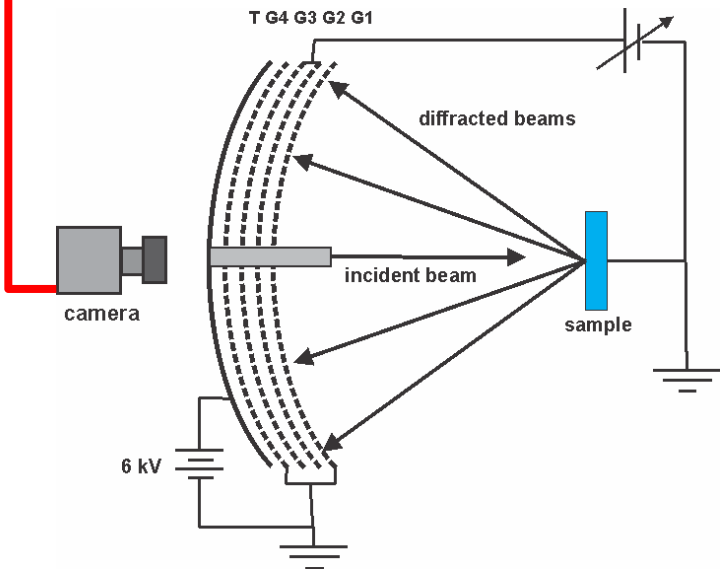
Liu et al., Nature Materials 2010

SD1; F3

Low Energy Electron Diffraction(LEED)

(LI/LA-SiGMA seed funded)





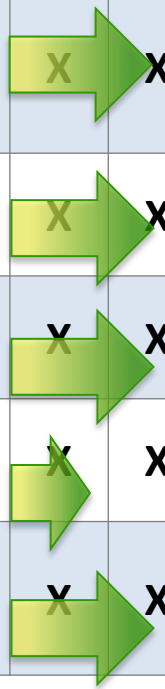
Surface Probe: I vs. V curves contain structural information.

New complex materials have MANY atoms in the unit cell: Computational challenge.

SD1; F2

SD1 Focus 3 Milestones and Score Card

Milestones	Y1	Y2	Y3	Y4	Y5	
Address the bottlenecks and numerical instabilities in the parquet equations by employing better parallel linear systems solvers and develop multiband parquet codes.	X	X	X	X	X	*On Track*
Incorporate latency hiding methods into parquet codes.	X	X				*On Track*
Use hybrid QMC to address the origin of the QCP and competing order in cuprate models.	X	X				*On Track*
Study overscreening in pnictide models using new Hyper-GGA functionals .	X	X	X			*Behind*
Use methods that combine LDA models obtained from downfolding and DCA/MSMB to study correlation and phonon effects in the pnictides.	X	X	X	X	X	*On Track*



SD1; F3

Outreach

- Nanodays. LA-SiGMA faculty members gave four public lectures and graduate students led demonstrations at the BREC's Highland Road Observatory and the Louisiana Arts and Science Museum: over 300 visitors.
- CCT and LA-SiGMA REU programs

Graduate Students and Postdoctoral Education

Density Functional Workshop, Summer 2011
(40 registrants, 5 speakers, LA Tech, LSU, SUBR, Tulane, Xavier)

John Perdew
Tulane

Shobhana Narasimhan
Jawaharlal Nehru Centre

Kieron Burke
UC Irvine

Mel Levy
Tulane & N.C. A&T

Weitao Yang
Duke

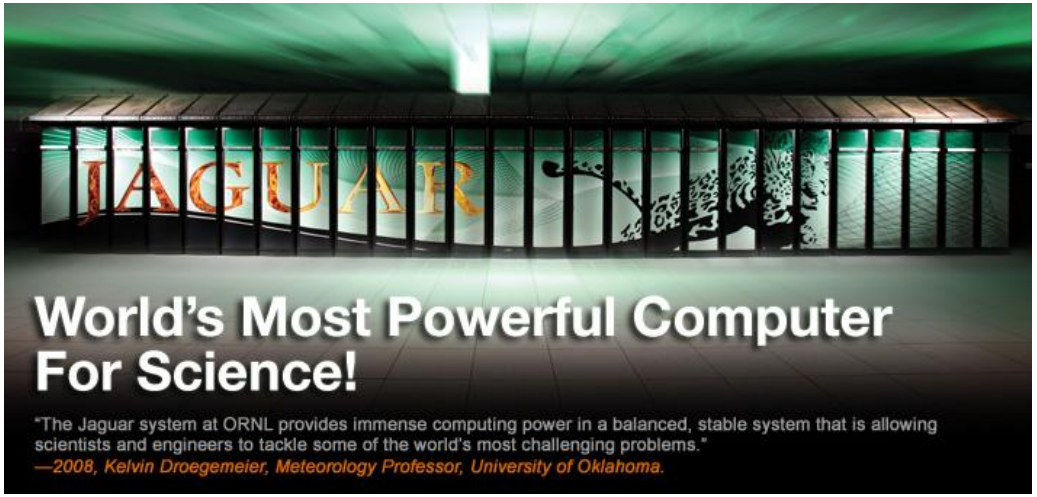
Tanusri Saha-Dasgupta
S.N. Bose National Centre

Graduate Student Education

- Distance Learning Courses:
  - Computational Solid State Physics
  - Advanced Solid State Physics with Computation
  - Computational Physics
  - Simulations of Quantum Many-Body Systems
- SD1 Seminars (EVO)
  - Jianwei Sun (Tulane), *RPA within the adiabatic connection fluctuation dissipation theory (ACFDT)*
  - Shuxiang Yang (LSU) *Hierarchy of approximate methods within a unified framework: the parquet formalism*
  - Mark Jarrell (LSU) *Grassmann algebra*
  - Mark Jarrell, *Fermion Path Integrals*
  - Mark Jarrell, *Feynman-Dyson perturbation theory.*
- GPU Team Meetings
  - Thursdays at 2:00

Partnership with Pacific Northwest National Lab

- Environmental Molecular Sciences Laboratory (EMSL)
  - Home of NWChem!
- INCITE request for 20,000,000 over 3 years on Jaguar and Titan supercomputers (20+ PFLOPs, Kepler GPUs)
- Internship program for LA-SiGMA students
  - 3-6 month visits
  - Working with EMSL Open Source Code Developers
  - undergraduate to PhD
- Indo-US SCES Centre



EMSL Associate Lab Director Bill Shelton participating in LA-SiGMA REU panel

Pacific Northwest NATIONAL LABORATORY

Funding

- Teragrid award for 11M Sus
- Two Computational Materials Science Network awards (Brookhaven, Ames, SLAC, Argonne, Pacific Northwest National Laboratory)
- INCITE proposal (in collaboration with PNNL) for 200,000,000 compute hours on Jaguar and Titan
- NSF SISI proposal for GPU code development
- Indo-US Center proposal (IUSSTF)
- SCiDAC proposal (under development)

Summary

- Meeting Goals and Milestones
- Strong Overlap with CTCI
- Outreach and Education
  - Courses
  - DFT Workshop
  - GPU Team
- Partnerships with National Labs (BNL+PNNL)
- Seeking Other Sources of Funding