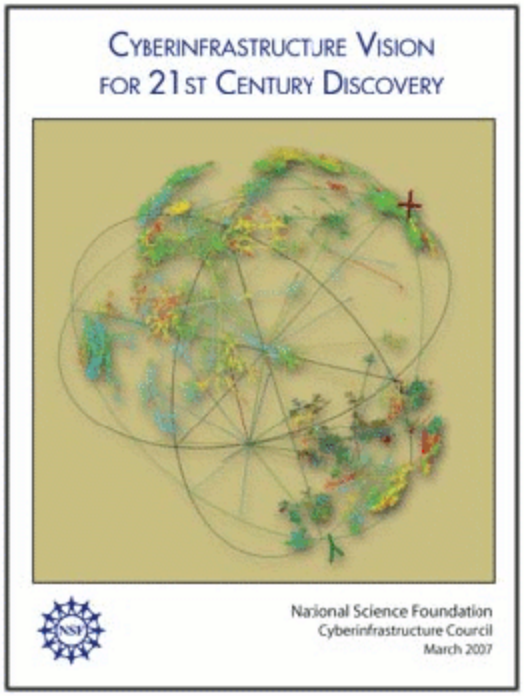
Science Driver 1: Correlated Electronic and Magnetic Materials

M. Jarrell and J. Perdew

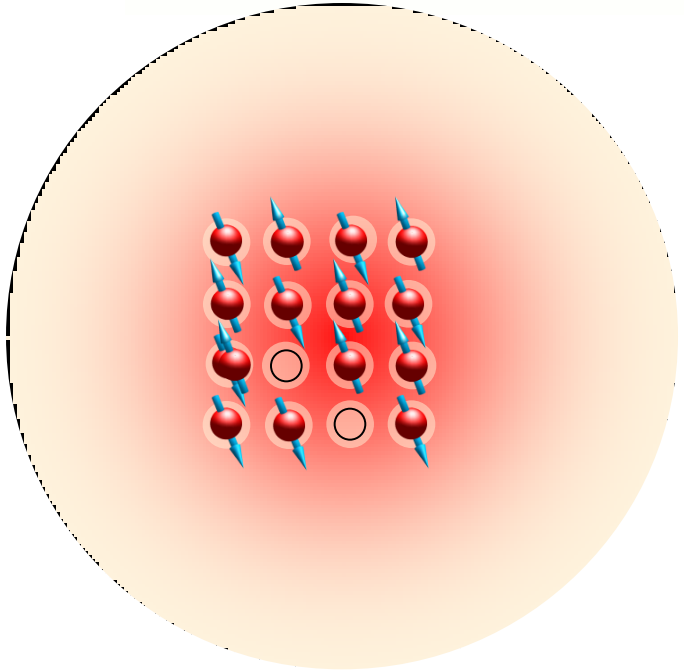
LA-SiGMA

Louisiana Alliance for Simulation-Guided Materials Applications

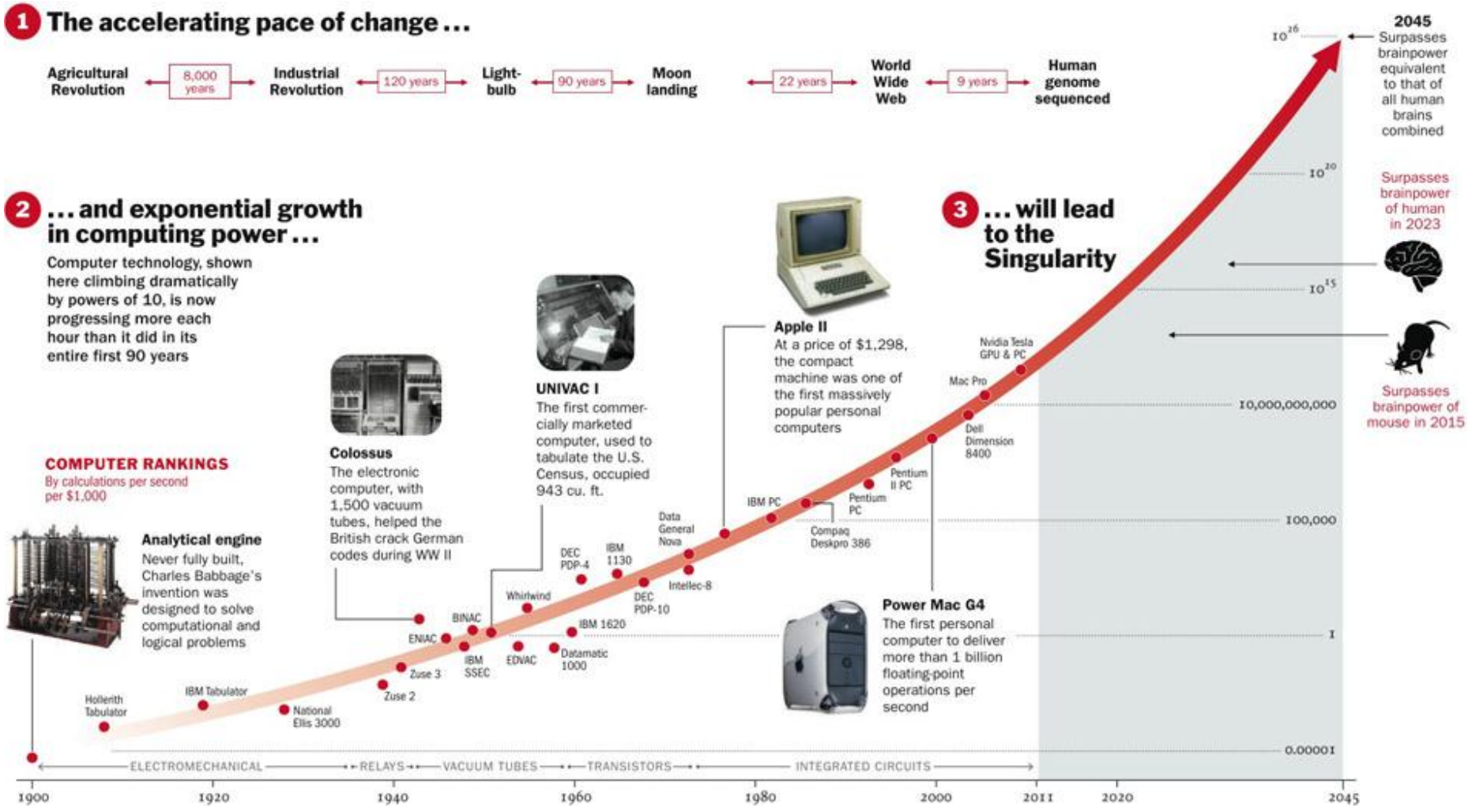
Computational Materials Science



- 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.



Exponential growth in computing power:

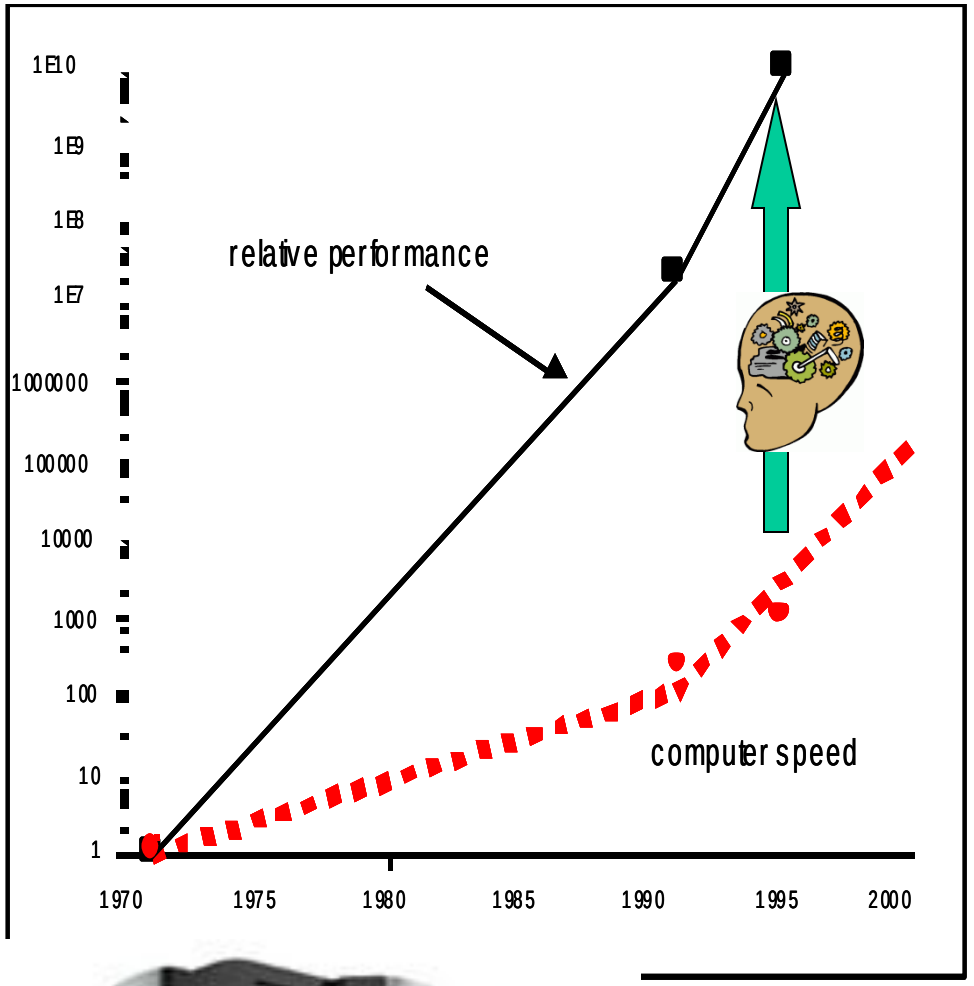


http://i.timeinc.net/time/daily/2011/1102/singularity_graphic.jpg

Bush Differential Analyzer -www.britannica.com

ENIAC - U.S. Army Photo

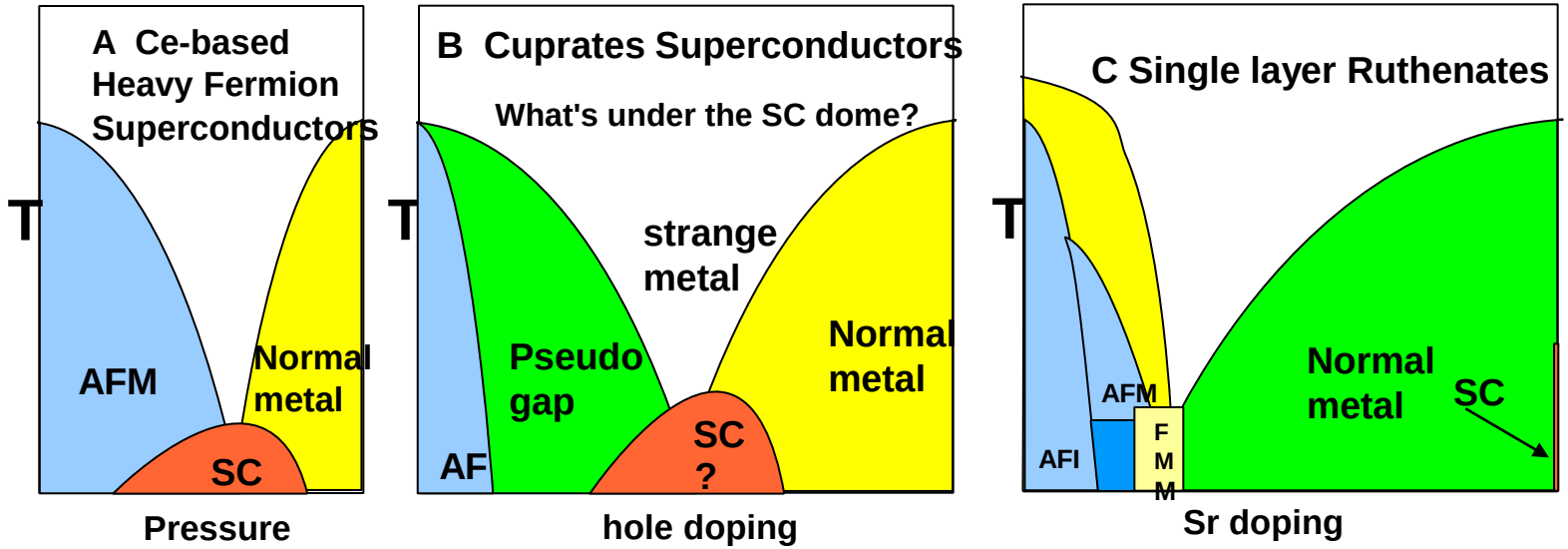
CRAY 1 - www.ucar.edu



(D. P. Landau, UGA)

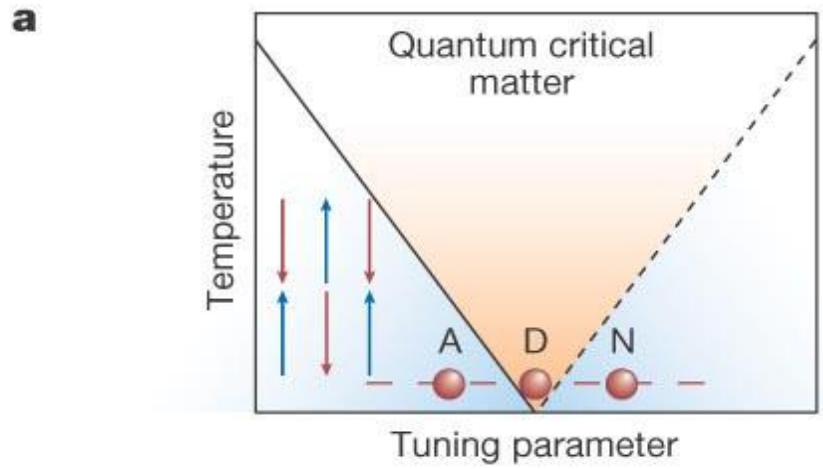
CRAY X1 – ORNL/CCS

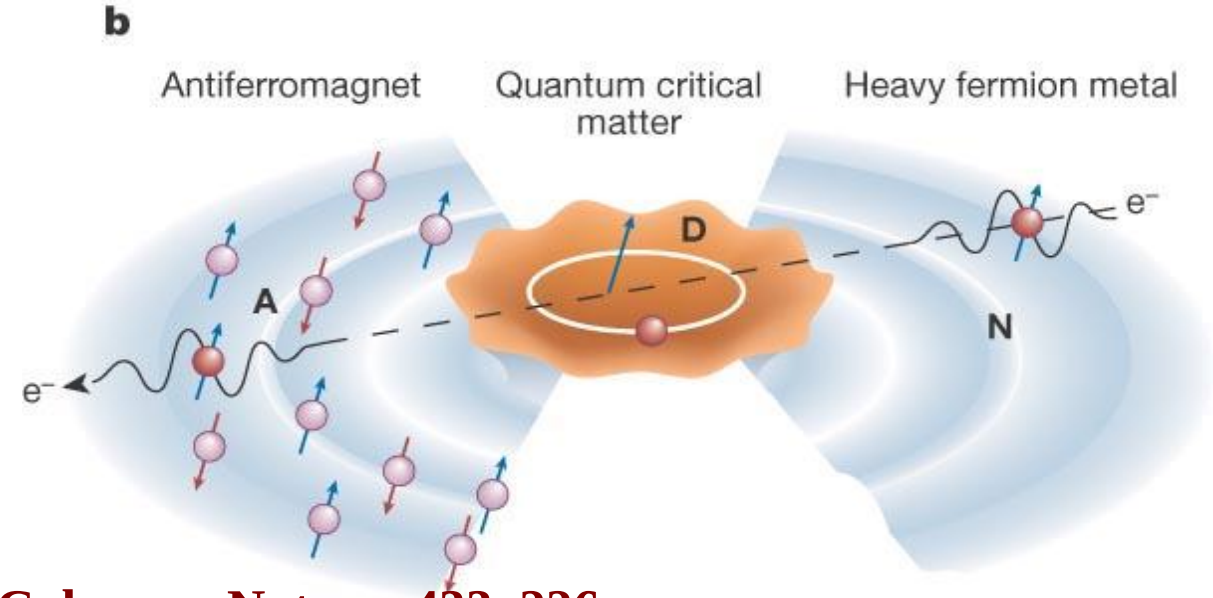
Strongly Correlated systems: Complexity and Competing Orders



- Correlations lead to formation of spin, charge, and orbital moments
- Competing phase emerge as a function of control parameter
  - HF superconductors
  - Cuprates
  - Single-layer Ruthenates (7 phases)
- Competition results in some transition temperatures vanishing as a function of a non-thermal control parameter

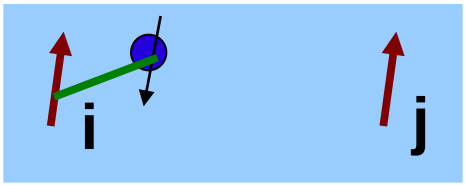
Simplest Case: Competition Between Exchange and Screening





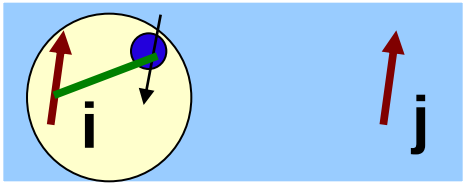
Coleman, Nature, 433, 226

- **RKKY exchange**



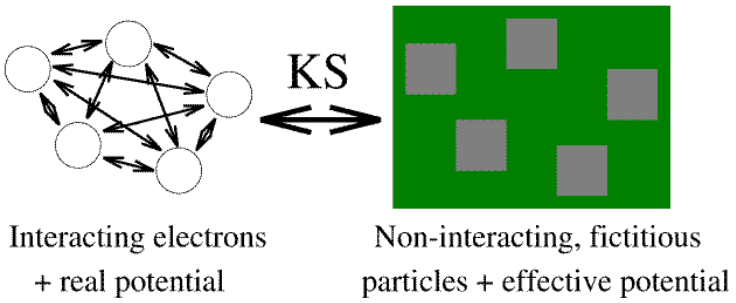
$J_{ij} \sim J^2$

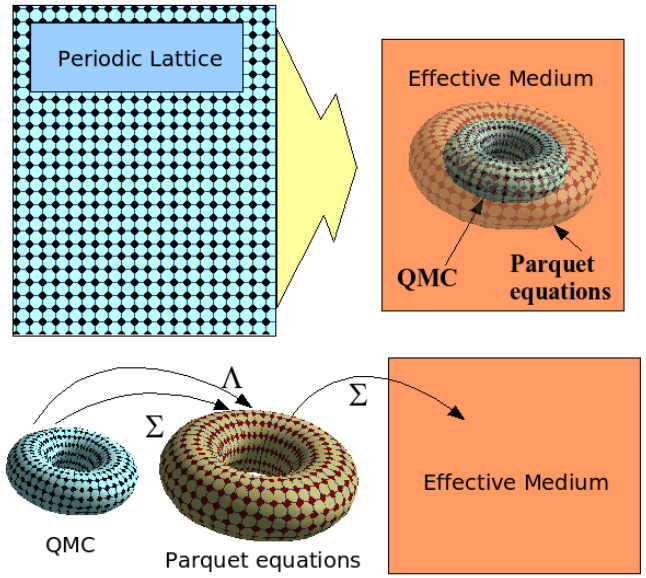
- **Kondo effect**



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

Methods Being Developed





- Non-local Approximations for Density Functional Theory
- Multi-scale Many Body Methods
- Combinations of Both
- Validation
  - Iron Based Superconductors
  - Organic Magnetic
  - Porphorines
  - Inverse LEED method

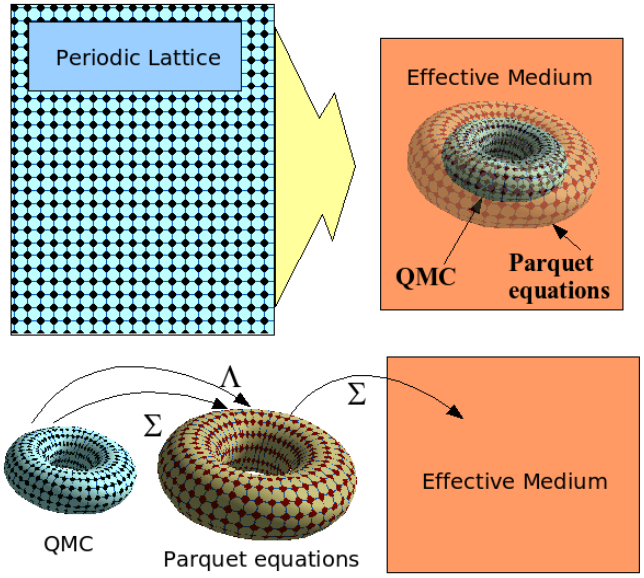
Computing at the petascale: MSMB

- **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**
- **Solving the Parquet Equations for the Hubbard Model beyond Weak Coupling, K.M. Tang, S. 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**



M

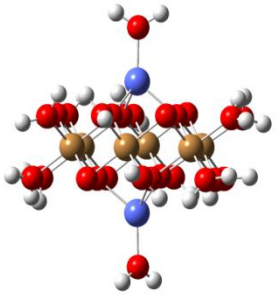
Iron Oxide Molecular Clusters as Building Blocks of Non-Volatile Memory

A joint research project between Xavier, Tulane and UNO teams

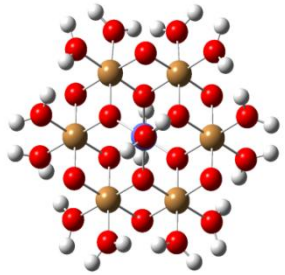
Aims of the project:

- synthesis of novel polynuclear coordination complexes containing spin-coupled paramagnetic ions (single-molecule magnets) (V. Kolesnichenko, G. Goloverda)
- Structural and spectroscopic characterization of new compounds (C. Stevens)
- Magnetic properties studies (L. Spinu)
- Computational studies: energy of spin states (A. Burin, J. Perdew)

The targets: molecular species with structure similar to structural motifs found in ferrimagnetic ferrites:



The smallest atomic assembly representing structural motif of ferrites.



Part One: Synthesis (ongoing)

Strategy: metal ion condensation promoted by a base and tuned by complexing agent L:

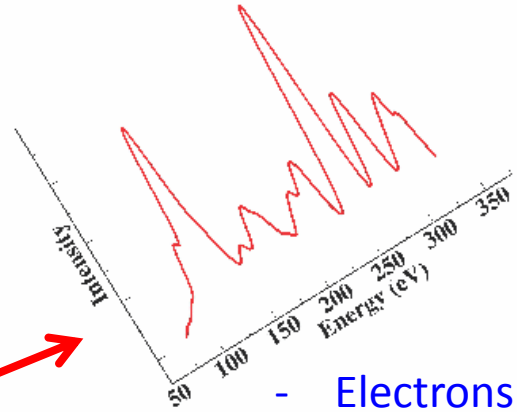
$$5Fe^{3+} + 3Fe^{2+} + 24RO^{-} + 12\ H_2O + 14L \rightarrow Fe_8O_{12}L_{14}^{3-} + 24ROH$$

Variables: L (polydentate bridging/chelating ligand); Mn^{2+}, Co^{2+} or metal(III) instead of Fe^{2+};
reaction stoichiometry leading to larger clusters like $Fe_{17}O_{22}(\mu\text{-}L)_6L_{16}^{+}$, $Fe_{26}O_{26}(\mu\text{-}OR)_{18}L_{18}^{-}$

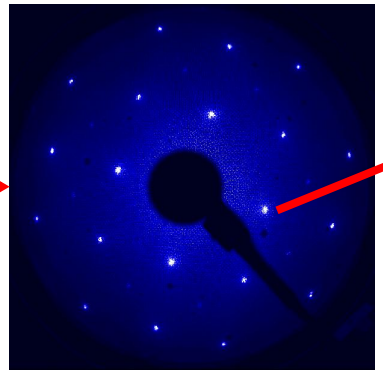
Parts Two, Three, …

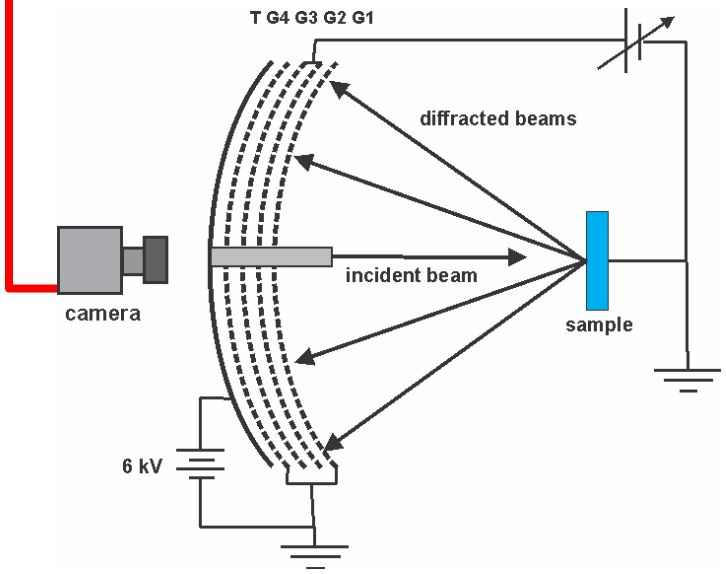
Magnetic properties of new structurally characterized compounds will be studied
Computational methods will be used to determine their electronic structure

Low Energy Electron Diffraction(LEED)



LSU: Von Braun Nascimento, Ward Plummer, and Hannah Manuel (math undergraduate):
Brazil: Professsors de Carvalho & Avelar Soares; GS Duarte dos Reis

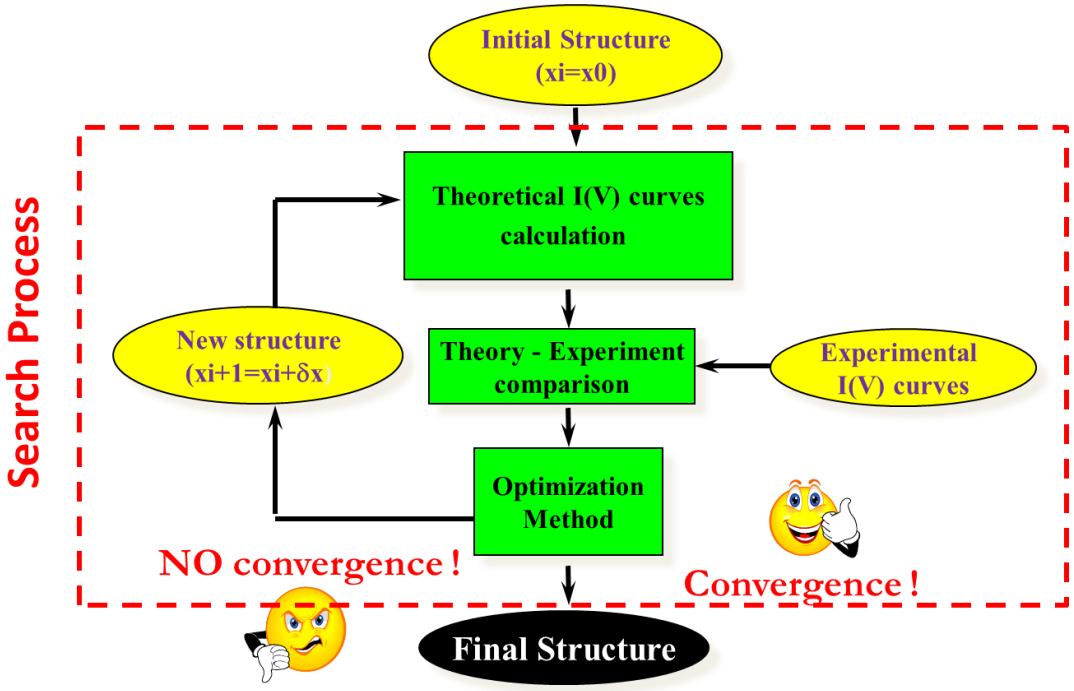




- Electrons with energy in the 20 ~ 500 eV used to determine surface structure.
- **Surface Probe:** Penetration depth is very short due to strong electron-electron interaction. I vs. V curves (above) contain structural information.
- **Problems:** Requires multiple scattering theory and global searching procedures. New complex materials have MANY atoms in the unit cell.
- **The Future with LA-SiGMA:** extend to complex systems
- 1) Improve Multiple scattering codes-non spherical potentials.
- 2) Better searching procedures
- 3) Parallel processing

Structure Determination by LEED

- Multiple scattering forces LEED analysis to be indirect
- Quantitative comparison theory-experiment
- Search for the best fitted structural model : Search Problem
- Hard task: locate the global minimum in an N-dimensional parameters space;



Complex Transition Metal Oxides :
complex structure –> many structural parameters to be optimized;

Necessary Improvements
1) Global Search Methods;
2) Faster Multiple Scattering calculations
3) **Direct Methods (inverse problem),** surface structure directly from experimental data -> **Holy Grail of LEED !**

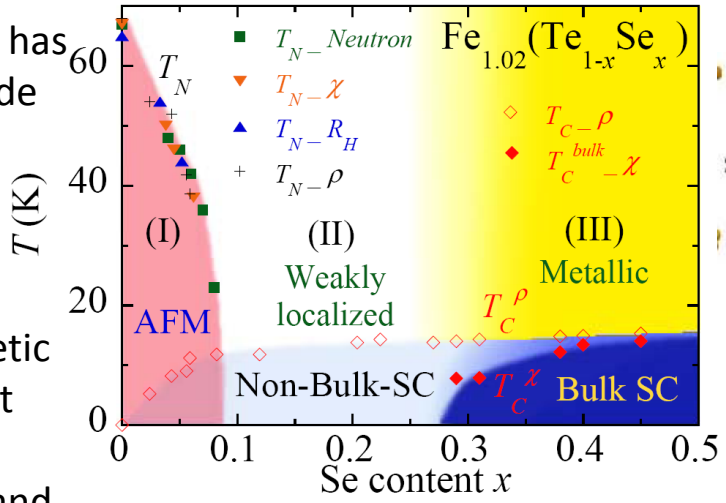
Unusual interplay between magnetism and superconductivity in iron chalcogenide $Fe_{1.02}(Te_{1-x}Se_x)$

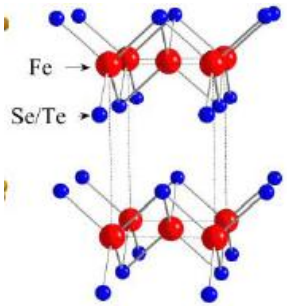
The discovery of superconductivity in iron pnictides has generated tremendous excitement .Iron chalcogenide $Fe_{1.02}(Te_{1-x}Se_x)$ is the simplified version of Fe-based superconductors.

Undoped parent compound $Fe_{1+y}Te$ exhibits a antiferromagnetic (AFM) order with in-plane magnetic wave-vector (π,0). This contrasts the pnictide parent compounds where the AFM order has an in-plane magnetic wave-vector (π, π). Yet both the pnictide and chalcogenide Fe-superconductors exhibit superconducting spin resonances around (π, π).

A central question in this burgeoning field is how (π, π) superconductivity can emerge from a (π,0) magnetic instability for iron chalcogenides.

Zhiqiang Mao' group has addressed this challenging issue through systematic investigation of the phase diagram of $Fe_{1.02}(Te_{1-x}Se_x)$ in collaboration with several other research groups.





Liu *et al.*, Nature Materials 9, 716(2010)

Conclusions and Significance

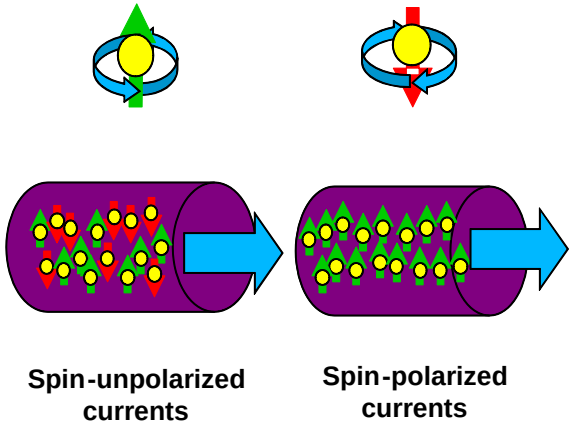
The magnetic soft mode evolving from the (π,0)-type order causes diffusive magnetic scattering to charge carriers, thus suppressing bulk superconductivity and leading to weak charge carrier localization in under-doped samples. Bulk superconductivity occurs only when the (π, 0) magnetic correlations are strongly suppressed and spin fluctuations near (π, π) become dominant, suggesting a common magnetic origin for superconductivity in iron chalcogenide and pnictide superconductors. This result significantly advances our emerging understanding of iron based superconductivity.

Magnetic and Electronic Properties of Organic Semiconductors

G. Joseph (Grambling), R. Nelson (LSU), A. Paudyal, N. Ranjitkar (LaTech),
D. Browne (LSU), P. Derosa (Grambling, LaTech), J. Garno, M. Jarrell, J. Moreno (LSU),
B. Ramachandran (LaTech)

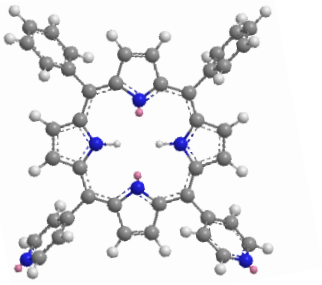
Motivation

- By 2020 the size of a transistor in a chip will be just a few atoms
- A new kind of transistors which employ the charge and spin of the carriers to convey information may be the solution for this situation: **Spins + Electronics = Spintronics**
- Organic conductors are currently of great interest in applications such as flexible electronic and solar cells
- Molecules, such as porphyrins, can be combined with a number of different atomic species 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.



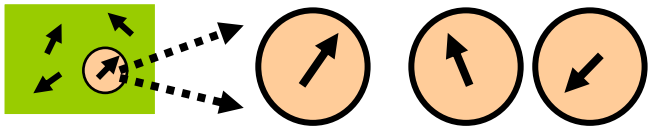
5,10-diphenyl-15,20-di-pyridin-4-porphyrin

MetalloPorphyrins, a test system

- Magnetic atoms can be inserted on the central cavity of metalloporphyrins resulting on distinct magnetic and electronic properties.
- Existing methods will be tested and new methods implemented via a synergistic simulation-experiment collaboration.

Model & Methods

- 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.



- Transport properties will be calculated using non-equilibrium Green function methods, and Keldysh and Wagner formalisms.
- Experiments will combine scanning probe lithography, imaging and current measurements in the presence of an AC field.

Molecular Magnets. Investigation of Spin of Iron Oxide Clusters

LA Sigma

J. M. Leveritt*, A. Kurnosov*, S. L. Tesar*, V. Kolesnichenko, G. Goloverda**, A. L. Burin***

***Tulane University, **Xavier University**

The elementary clusters derived from spinel-type structures (magnetite, ferrites etc) are candidates for molecular magnets behavior . As a first step in our investigation we considered $Co_2Fe_6H_{24}O_{24}$ cluster.

The essence of the approach – to optimize geometry of the cluster with different spins. The molecular magnet behavior is expected if the ground state is realized with high spin. For the first step the cluster is assumed to be neutral. The initial geometry is presented in fig. 1.

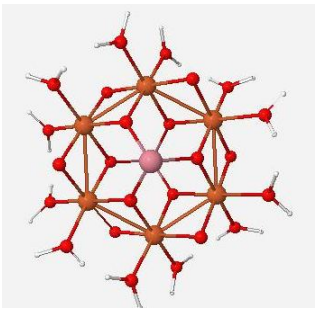


Fig. 1. $Co_2Fe_6H_{24}O_{24}$ cluster. The spinel-type structure with water ligands.

Calculations.

The pure DFT methods did not gain the result because of ill-convergence. MO:MM method was applied: ONIOM(Mpwpw91:UFF)[1]. This computational technique models large molecules by defining two layers within the structure that are treated at different levels of accuracy. We applied high-accuracy treatment for metal and medium level for oxygen and hydrogen atoms.

Results.

The ground state was found for spin = 4, the energy E_4 = -278368.44 eV. The geometry of the singlet state could not be optimized, because any convergence criteria were failed. The reasonable explanation is instability of this system for singlet state.

Energy–E_4 (eV)	Spin
11.79	1
0.22	2
0.02	3
0	4
2.16	5

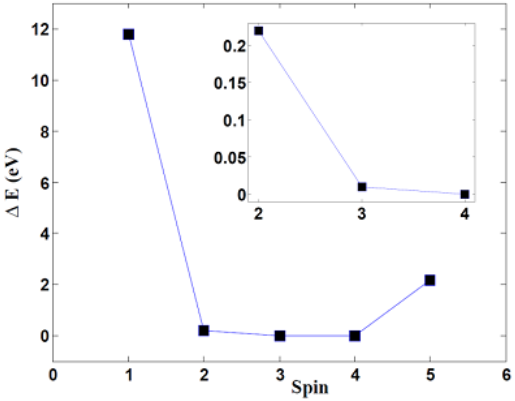


Fig. 2. The energy (with respect to the ground state E_4) as function of the spin. The absolute value E_4 = -278368.44 eV.

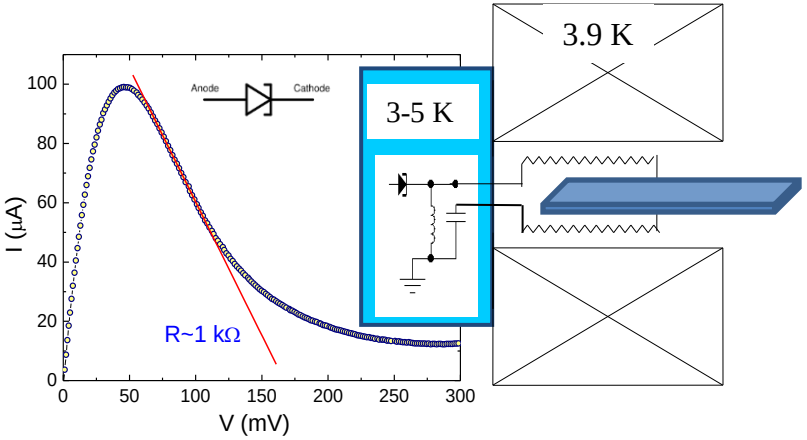
References

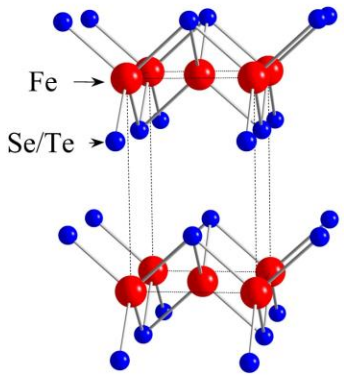
1. Carles Bo and Feliu Maseras, *Dalton Trans.*, 2008, 2911–2919

Penetration depth measurements in $Fe_{1.02}Te_{1-x}Se_x$ an Iron-based Superconductor

Andrei Diaconu, Zhiqiang Mao and Leonard Spinu
Advanced Materials Research Institute (AMRI) and UNO

Precise measurements of the penetration depth (λ_L)as a function of temperature, magnetic field and crystal orientation can provide detailed information about the pairing state and are among the most useful tools to probe low energy quasiparticles in superconductors, also temperature dependence can give information about the pairing state, symmetry of the energy gap; its zero value is directly related to the superfluid density in the ground state.





Measuring Penetration Depth using Tunnel Diode Oscillator – essentially a cavity perturbation technique, albeit at rf frequencies. Easy to implement at low temperatures and high applied magnetic fields with often higher precision than other methods. $\Delta f/fo=(-Vs/Vc)(1-(\lambda_L/R)\tanh(R/\lambda_L))$

In February 2008, the group of Hideo Hosono discovered superconductivity in LaFeAsO with a Tc of 26 K. In this study we will examine $Fe_{1.02}Te_{1-x}Se_{x.}$

Graduate 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.***