Defect Physics of Structural Materials for Energy

Ab initio methods for simulating the structure, interactions and dynamics of defects in structural materials for extreme environments

G. Malcolm Stocks, Don M. Nicholson,

Markus Eisenbach, Paul Kent, Aurelian Rusanu,
Fernando Reboredo, Randy Hood (LLNL),

Jeongnim Kim (NCSA)

Energy Frontier Center for Defect Physics in Structural Materials (CDP)

Oak Ridge National Laboratory

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Center for Defect Physics in Structural Materials (CDP)

www.ms.ornl.gov/cdp/index.shtml

Goal: Provide a fundamental understanding of materials defects, defect interactions, and defect dynamics, thereby enabling atomistic control and manipulation of defects and the charting of new pathways to the development of improved materials with ultrahigh strength, toughness, and radiation resistance.

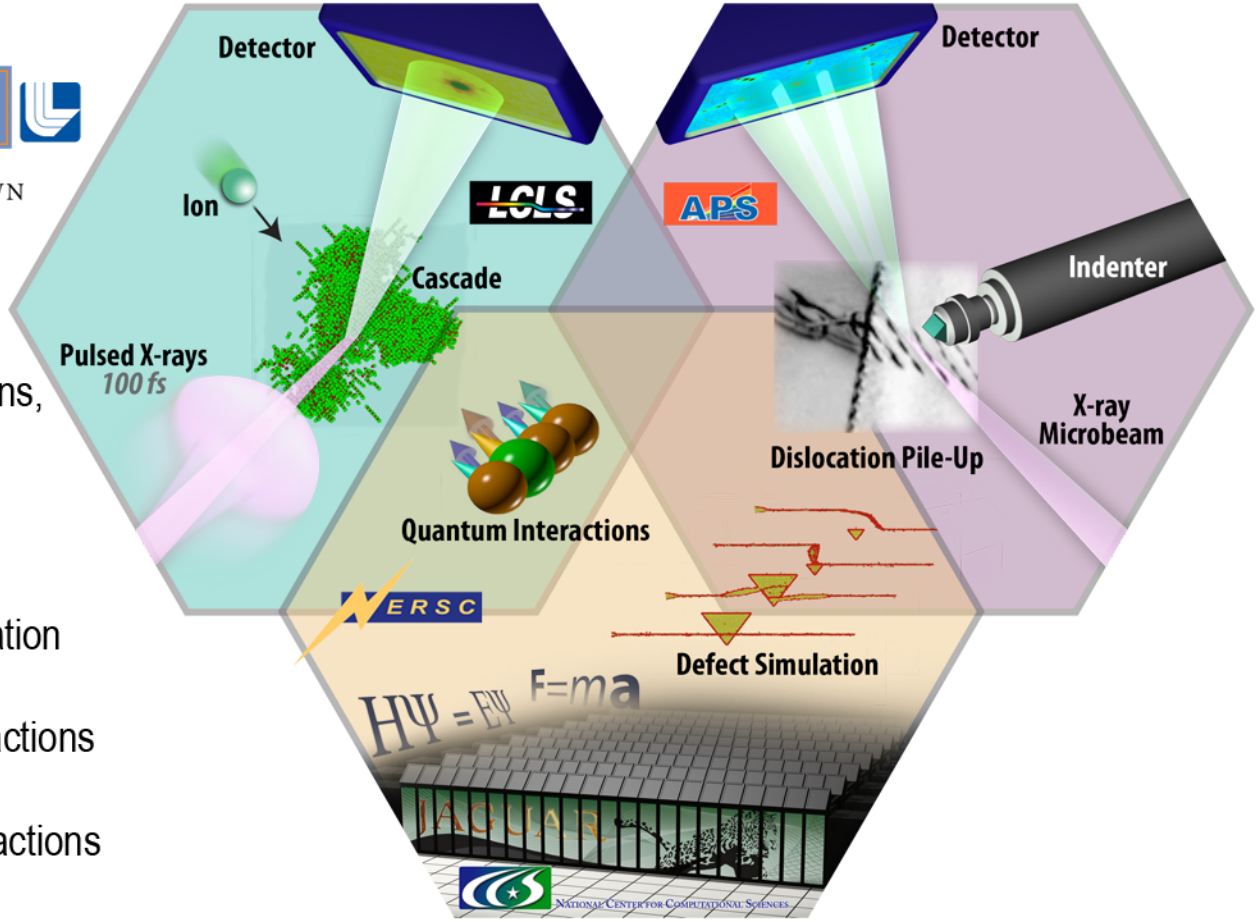
Partner Institutions:

Oak Ridge National Laboratory · UT · Berkeley University of California · Carnegie Mellon · The Ohio State University · Brown

Approach: Deploy first-of-their-kind measurements and *ab initio* quantum calculations of the structure, interactions, and dynamics of defects in structural materials

Interrelated Thrust Areas:

- Fundamental physics of defect formation and evolution during irradiation
- Fundamental physics of defect interactions during deformation
- Quantum theory of defects and interactions



Project Overview

"Crystals are like people: it is the defects in them that make them interesting"

Sir Charles Frank

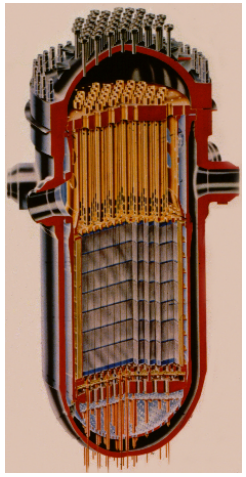
- Collective effects of defects determine real materials properties
  - Strength, toughness, resistance to degradation in extreme chemical and radiation environments
  - Underpinnings of our energy and transportation technologies.
- Performance limits are rarely the result of insurmountable physical principles
  - Structural alloys exhibit strengths that are typically only 5-10% of theoretical limits
  - Reactor vessel steels exposed to neutron irradiation become brittle.
- Increased understanding of defects will result in new materials with substantially improved properties

The Physics of Defects on Short Physical and Temporal Length Scales

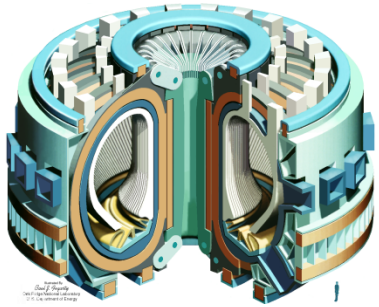
The Word of Unit Events

Fundamental Physics of Defect Formation and Evolution during Irradiation

- Radiation Damage Produces Substantial Changes in the Microstructure and Mechanical Properties of Structural Materials



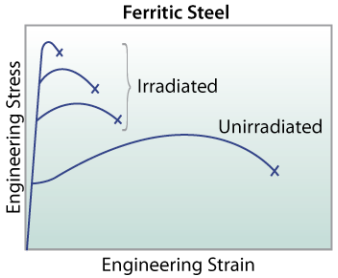
LWR Pressure Vessel

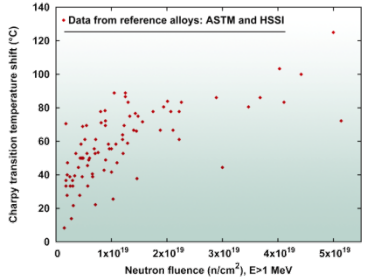


ITER Tokamak

Low temperatures (<0.4 T_M):

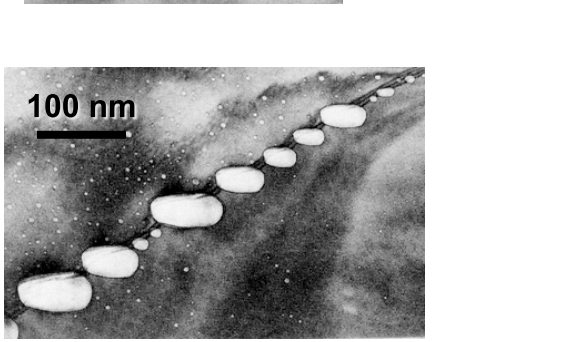
- Radiation-induced hardening and embrittlement

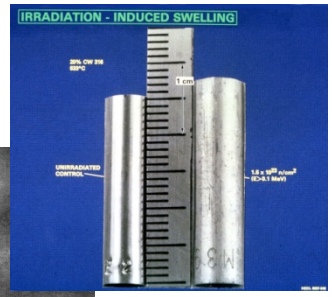




Intermediate temperatures (0.3-0.6 T_M):

- Phase instabilities from radiation-induced precipitation
- Irradiation creep
- Volumetric swelling from void formation



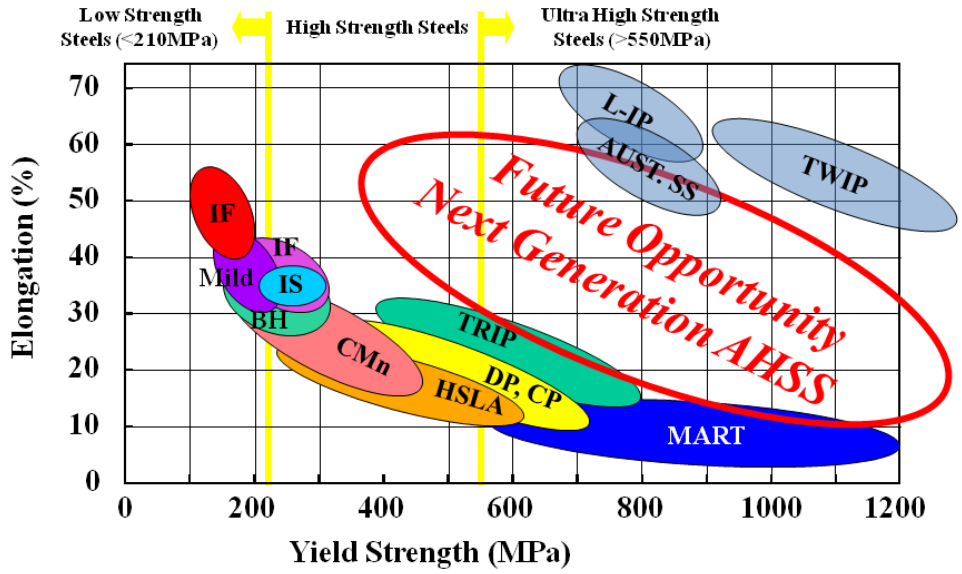


High temperature(>0.5 T_M):

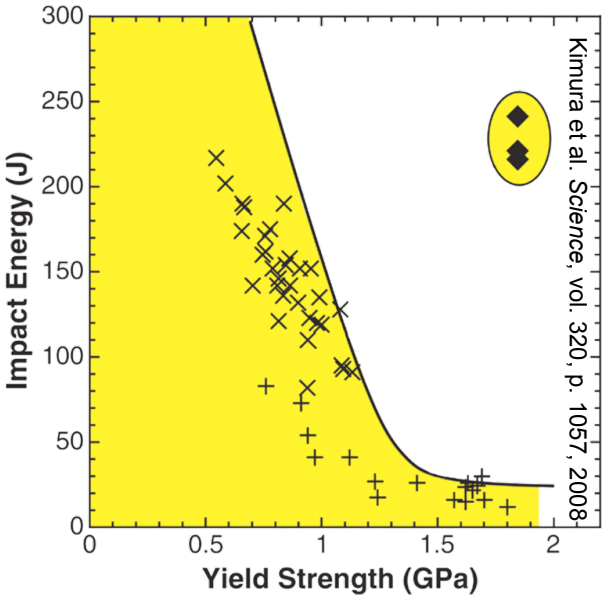
- He embrittlement

Fundamental Physics of Defect Interactions during Deformation

"Banana curve" showing typical inverse relationship between strength and ductility

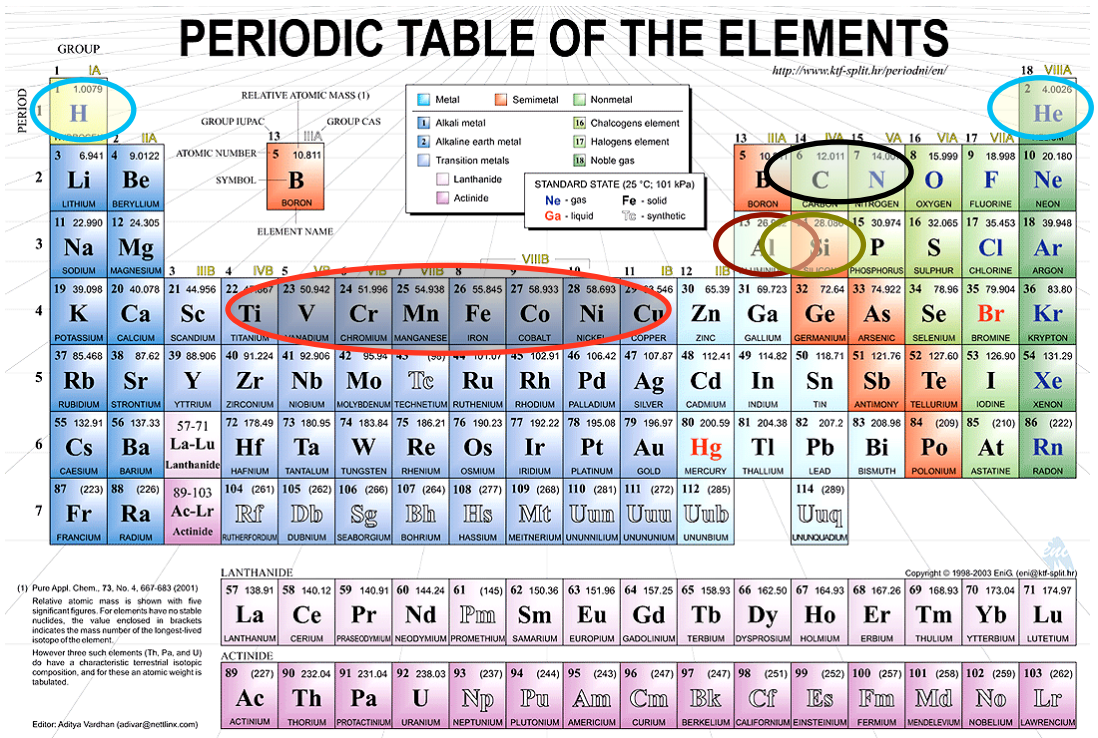


Recent results show that microstructural refinement combined with interface control can decouple strength-toughness



Quantitative understanding of defect interactions is key to replicating this scientifically rather than by trial and error

Structural Materials and Defect Physics



- The structural materials' "periodic table"
  - Fe-based alloys: Steels
    - Fe+(TM)+(C,N)+(O…)

      316-Stainless

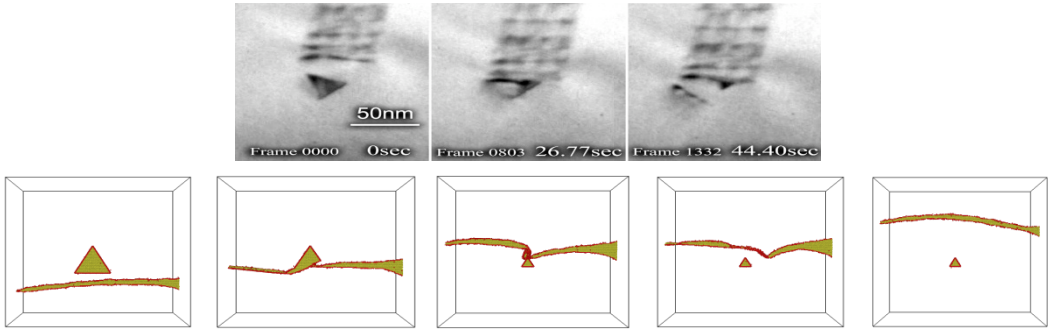
      $Fe_xCr_{(12-18)}\ Ni_{(10-14)}Mn_2Mo_{(2-3)}(C,Si,P,S)$

      In Fe_x x-comprises the balance of 100%
  - Radiation effects
    - He, H, Vacancies, Decay products,..
  - Fusion:
    - Fe, V, Zr, SiC, Be, Li..
  - Research Reactors
    - Al (largely historical)

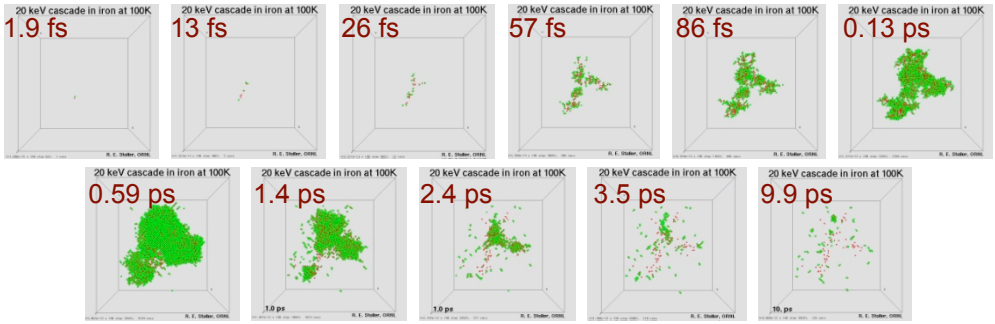
- Computational Materials Science Issues
  - Complex Materials: multi-component, disordered alloys, extended defects require methods capable of dealing with large numbers of atoms
  - Importance of magnetism: Fe and transition metal based systems require methods capable of dealing with atomic and spin degrees of freedom on an equal *ab initio* footing
  - Importance of metallurgical accuracy (~0.01mRy/atom*)

** For reference alloy heats of solution are typically a few kJ/mol (1 kJ/mol=0.7447mRyd/atom)*

Structural Materials and Defect Physics



Dislocation/Stacking-fault-tetrahedra Interactions



Radiation damage cascade dynamics

- Defects structure, interactions and dynamics
  - Point defects and point defect-clusters
    - Interstials, vacancies, impurities
      - He, H, Decay products,..
  - Extended defects
    - Interstitial loops, voids, stacking-fault-tetrahedra, grain boundaries
    - Dislocations
  - Radiation damage dynamics

- Computational Materials Science Issues
  - Extended defects require methods capable of dealing with large numbers of atoms
  - Core of low symmetry defect structures requires methods capable of dealing with atomic and spin degrees of freedom on an equal *ab initio* footing
  - Importance of metallurgical accuracy

Electronic Structure Theory: "The Jewels in the Crown"

- Properties of condensed mater contained in solution of many electron Schrödinger equation for the solid

$$\{-\sum_i \frac{1}{2}\nabla_i^2 + \sum_{i,j} \frac{1}{|r_i - r_j|} + \sum_{i,R_k} \frac{Z}{|r_i - R_k|}\}\Psi(r_1,..r_N) = E\Psi(r_1,..r_N)$$

"Direct" approaches to Solution
Quantum Chemistry
Quantum Monte Carlo

- Density functional theory (DFT): Walter Kohn 1998 Nobel Prize in Chemistry
  - By focusing on ground state density $n(\mathbf{r})$ proved a series of powerful theorems
    - Ground state energy of system is a unique functional of the density
    - Density can be obtained from solution of single particle like SCF equations

Total Energy:

$$E[n] = T_s[n] + V_{ext} + U[n] + E_{xc}[n]$$

Euler - Lagrange equations:

$$\{-\nabla^2 + V^{eff}(\mathbf{r})\}\Psi_i(\mathbf{r}) = \varepsilon_i\Psi_i(\mathbf{r});$$

Density:

$$n(\mathbf{r}) = \sum_i |\Psi_i(\mathbf{r})|^2$$

Effective Potential

$$V^{eff}[n] = V^{ext} + \int d\mathbf{r}' \frac{n(\mathbf{r})}{|\mathbf{r} - \mathbf{r}'|} + \frac{\delta E_{xc}}{\delta n(\mathbf{r})}$$

$E_{xc}^{Exact}[n] \Rightarrow Unknown$

$E_{xc}^{Exact}[n] \Rightarrow E_{xc}^{App}[n]$

$App \Rightarrow LDA, GGA, ...$

$E_{xc}^{App}[n] \Rightarrow Industry$

DFT Electronic Structure Codes

Industry

All electron, Pseudo-potential

:

Plane-wave Pseudo-potential
(VASP, PWSCF, …)

:

Multiple Scattering Green's Function
(LSMS,…)

Major Computational Physics Issues Addressed by HPC

- Materials and Defect Complexity
  - Need approaches for dealing with large numbers of atoms
    - ~10^3: Standard LDA codes for structural optimization (VASP or other standard LDA code)
    - 10^3 to 10^5 atoms: Optimization of complex magnetic ground states and non-equilibrium magnetic states [Locally self-consistent multiple scattering (LSMS) method]
- Importance of magnetism
  - Need to deal with positional and spin degrees of freedom on an equal *ab initio* footing
    - Finite temperature spin statistics (Wang Landau Monte Carlo based on *ab initio* LSMS energies of spin configurations (WL-LSMS)
    - Combined Molecular and Spin Dynamics [TBD]
- Importance of metallurgical accuracy
  - Require high quantum accuracy: Beyond LDA-DFT
    - Self-healing Diffusion Quantum Monte Carlo (SH-DQMC)

Current HPC Requirements

- Machines used
  - SH-DQMC: NERSC Franklin, NCCS Jaguar
  - VASP: ORNL Clusters; NERSC Franklin;
  - LSMS: ORNL Clusters; NERSC Franklin; NCCS Jaguar
  - WL-LSMS: NCCS Jaguar
- # cores, amount of memory, input/output, disk storage typically used
  - SH-DQMC: Cores: 10^3 to ~10^5: Memory 1-2GB/core; I/O and disk no severe limitations
  - VASP: Cores: 10^2 to ~10^3: Memory ~2GB/core; I/O and disk no severe limitations
  - LSMS: Cores:10^3 to N*10^4 (N~1-3); Memory: ~1-2GB/core; I/O and disk no severe limitations
  - WL-LSMS: Cores: N*10^5 (N~2) [so far!]; Memory: <1GB/core; I/O and disk no severe limitations [~200 Kbytes restart file!]
- Necessary software, services or infrastructure
  - Standard libraries optimized by platform (ScaLAPACK, LAPACK, BLAS, MPI, …)
- Data transfer requirements (within NERSC or to/from NERSC)
  - Nothing special is required

Current HPC Requirements (Cont.)

- # of runs, types, length, reasons
  - SH-DQMC: N*10^6-processor hours/project (N~10); is required with multiple (~5) systems/year. Demands are beyond standard ERCAP proposals
  - VASP: many (~10^2/year) runs using 64 to 10^3 cores (within ERCAP type requests)
  - LSMS: N*10 (N~5) runs/year using10^3 -10^4cores
  - WL-LSMS: Nx10^6-processor hours/project (N~5) is required for demonstration projects; codes can clearly scale to 50x current # of cores with concomitant increase in demand for CPU-hours.
    - NB the runs that won the 2009 Gordon Bell Prize (1.8 peta-flops) were more proof of principle than production, production will require 10x to 100x increase
- Known limitations / obstacles / bottlenecks
  - SH-DQMC: Scaling of algorithms to large numbers of electrons, metals and magnetism remains to be demonstrated
    - Possible inadequacy of pseudo-potential approximation for treating core electrons can be overcome by treating more electrons
  - VASP: poor scaling beyond ~10^3 atoms and 10^3 cores (ultimately N^3)
  - LSMS: LSMS_1.x codes make spherical approximation to LDA potential LSMS_2.x codes are full potential but currently less robust and much slower - the prefactor of O[N] significantly increases (currently by >10x)
  - WL-LSMS: Need to do a fully SCF calculation for instantaneous magnetic state [The 2009 GB-prize calculations used the frozen potential approximation] this will greatly increase computational effort (~10x).

HPC Requirements 5-Years Hence

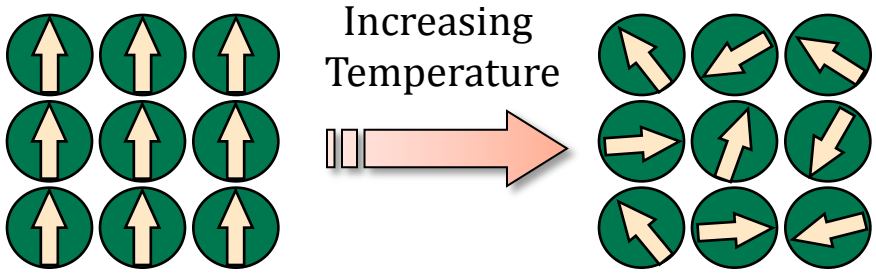
- Upcoming changes to codes/methods/approaches
  - SH-DQMC: Extension to metals and defects
  - LSMS: Full potential- relativistic
  - WL-LSMS: Continued scaling and implementation of methods for calculating joint density of states
    - Multiple observables: Energy, Magnetization, Site-Magnetization…: Currently only Energy is binned
  - General: Approaches to exploit next generation architectures (GPUs etc)
- Estimate of MPP hours needed to achieve science goals
  - SH-DQMC, LSMS, WL-LSMS: 10 to 100x
- # cores, amount of memory, input/output, disk storage typically used
  - SH-DQMC, LSMS, WL-LSMS: 10x-100x-cores
- Changes to necessary software, services or infrastructure
  - Base assumption: standard libraries will be optimized by platform and will scale.
- Anticipated limitations/obstacles/bottlenecks on 10K-1,000K core system.
  - SH-DQMC, WL-LSMS: should scale to very large core counts
  - SH-DQMC, WL-LSMS: increased CPU count keeps time to solution constant while increasing realism of simulation (best use of people!).
  - LSMS: scaling to ~10^5-cores should be fine, beyond that we will be in unknown territory
- Preparations for, or use of, emerging HPC Architectures and Programming Models
  - LSMS: Current MPI program paradigm will require modification to exploit GPUS – initial test using a hybrid scheme are underway

Additional Comments

- Significant increase (10-100x) in computational demands to solve outstanding issues in defect physics of structural materials
  - WL-LSMS
    - Not feasible @ NERSC: requires O(million) core-hours for convergence not possible within our allocation [same is true for *ab initio* QMC!]
    - Move to multi-dimensional Statistical Density of States (S-DOS) (or more intelligent scheme to be developed!) will result in increase of MC steps required for convergence – which converts to longer run times
    - Will need O(10 million) core-hours for converged two dimensional S-DOS for a single system.
      - However, we can use all the cores we can get! (Assuming memory and storage/ core remain at current levels)
  - Full potential LSMS
    - Shift of computational balance to Integral/Differential equation solver, away from pure linear algebra (matrix inversion), increased memory requirements
    - How will full potential perform for large systems? Can we exploit new computing paradigms?
- Need to think about National Computing Environment
  - Capability (One 100x-Franklin) versus Capacity (100-Franklins)
    - Need both depending on problem – favor 100-Franklins
  - Balance between support for hardware usage and software development
    - We need grey matter support
      - This is a game best played by multi-disciplinary teams

Finite Temperature Statistical Physics

- Statistical Physics of Moment Orientations



Ab initio Spin-dynamics
V. P. Antropov, et al., PRL (1995). PRB (1996)
Q. Niu, et al., PRL (1999) K. Capelle, PRL, 87(2001).
Ab initio Monte Carlo
Here!!

- Wang-Landau Monte Carlo algorithm and high performance computing facilitate *ab initio* studies of finite temperature magnetic response
  - Calculate statistical density of states
  - Thermodynamics at all temperatures

$$Z = \int g(E) e^{-E/k_B T} dE$$

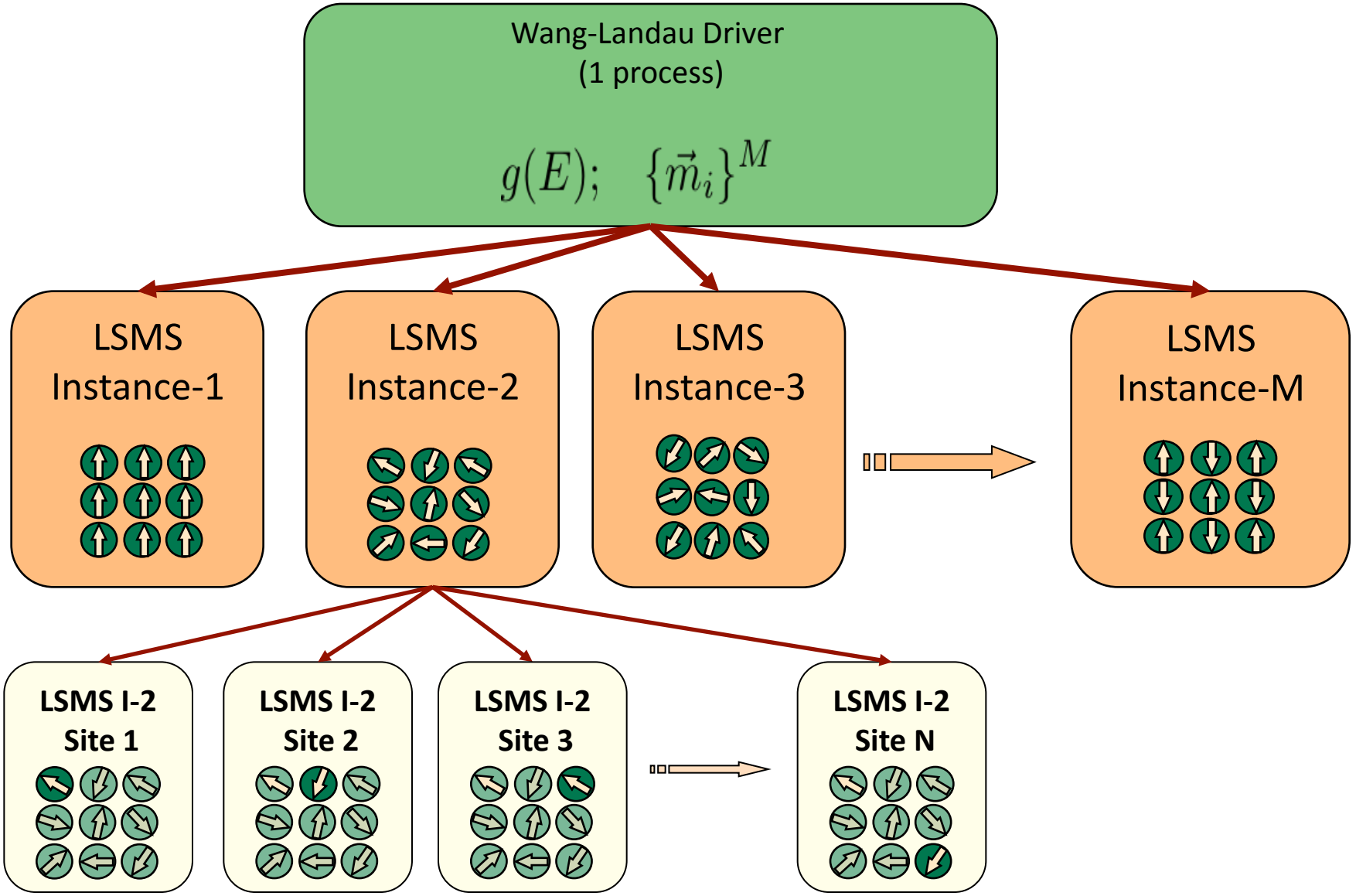
- First-principles Wang-Landau treatment of thermodynamic fluctuations

First-principles Wang-Landau Treatment of Thermodynamic Fluctuations

What are the challenges and how do we address them?

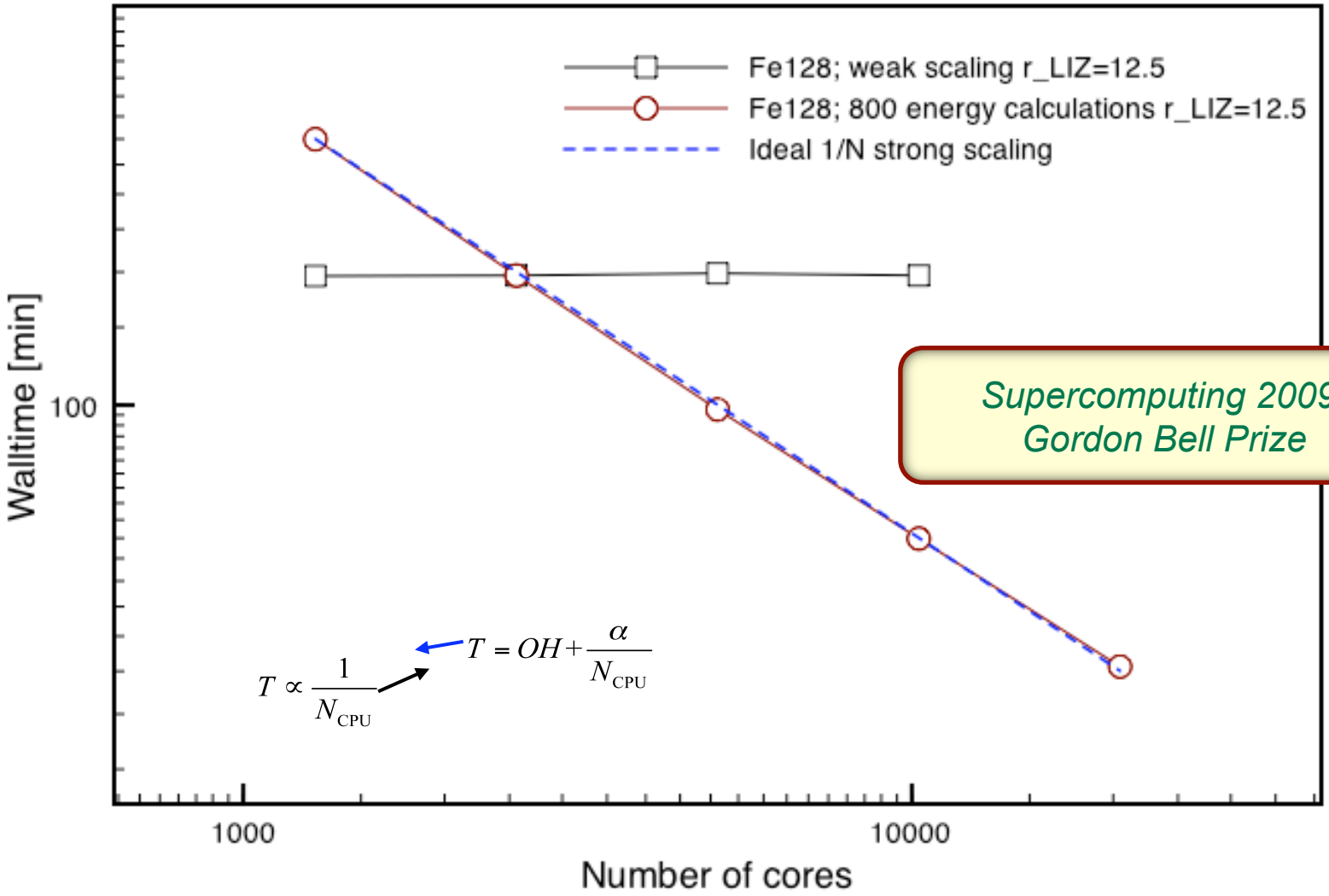
- Efficient Approach to Treating Magnetic Fluctuations
  - Wang-Landau Monte-Carlo algorithm
    - *F. Wang & D. P. Landau PRL 86, 2050, (2001); C-G. Zhou et al. PRL 96, 120201 (2006)*
- Method for returning energies of general spin configurations
  - Constrained density functional theory for local moments
    - *P.H. Dederichs et al., PRL 53, 2512 (1984); G.M. Stocks et al., Phil. Mag. B 78, 665 (1998); B. Újfalussy et al., JAP 85, 4824 (1999)*
- Electronic structure code to calculate energies of large systems
  - Order-N Locally Self-consistent Multiple Scattering (LSMS) method
    - *Y. Wang et al., PRL 75, 2867 (1995)*
- Computational capabilities (software/hardware) to address real systems
  - Implementation that exploits intrinsic parallelism (WL and LSMS)
  - High performance (petatflop) computing environment to run codes

Wang-Landau-LSMS allows multi-level parallelism



WL-LSMS Fully Exploits Leadership Class Computers

Strong scaling of a 128 atom system using up to 224,000 cores
Peak performance: 1.8 PetaFlop/s on JanguarPF at ORNL CCS



Supercomputing 2009
Gordon Bell Prize