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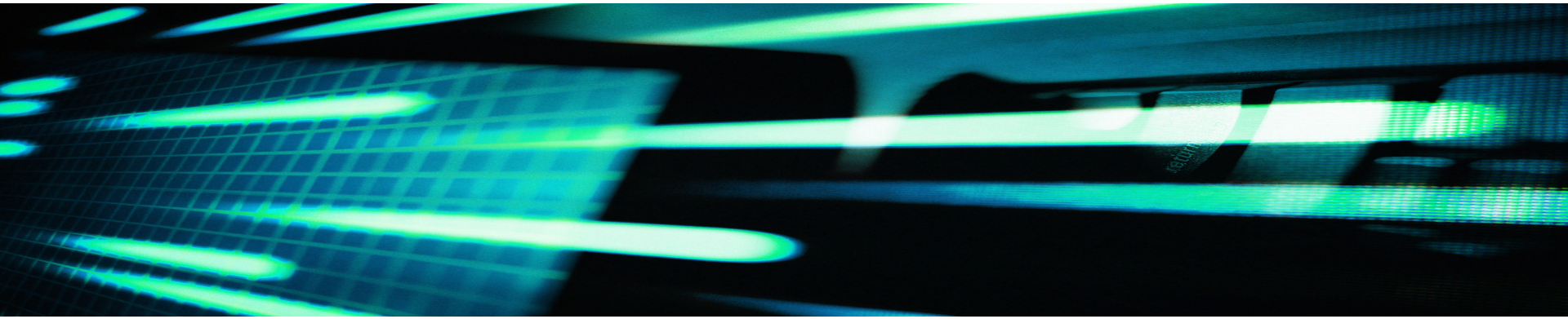


Computational Materials Science for Materials under Extremes

Alfredo Caro Los Alamos National Laboratory USA

Primer encuentro virtual de la red CompuMat

May 23 2014

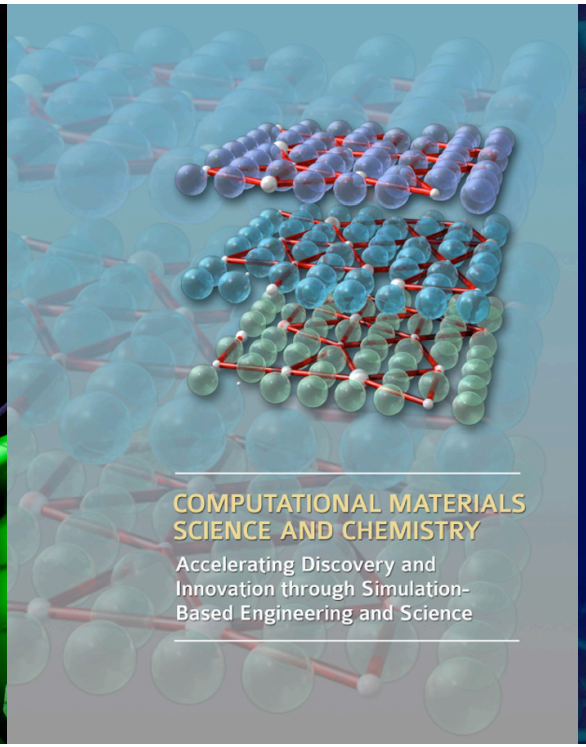
Overview

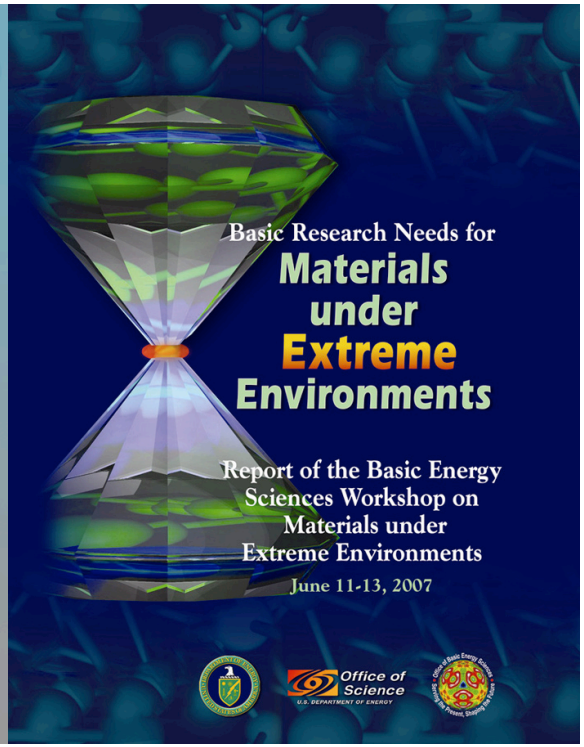
- This talk is about some of our work on materials science modeling and simulation at the atomic scale
- We'll present cases in which we develop new models, computational algorithms, or tools; in some other cases, we use existing tools on supercomputers to advance our knowledge on materials sciences
- A diversity of agencies pay for the work presented here, in particular: NEAMS, CASL (DOE's NE), EFRC (DOE's BES), LDRD (LANL's)
- How it supports the mission and programs
- This research aligns with the Materials for the Future Pillar of the Labs Materials Strategy (Defects and interfaces, Extreme environments, Emergent phenomena)
- The work presented here involves many people form different Organizations within LANL

How science directions are chosen:

http://science.energy.gov/bes/news-and-resources/reports/

Computational materials science, particularly at the mesoscale, is a focus

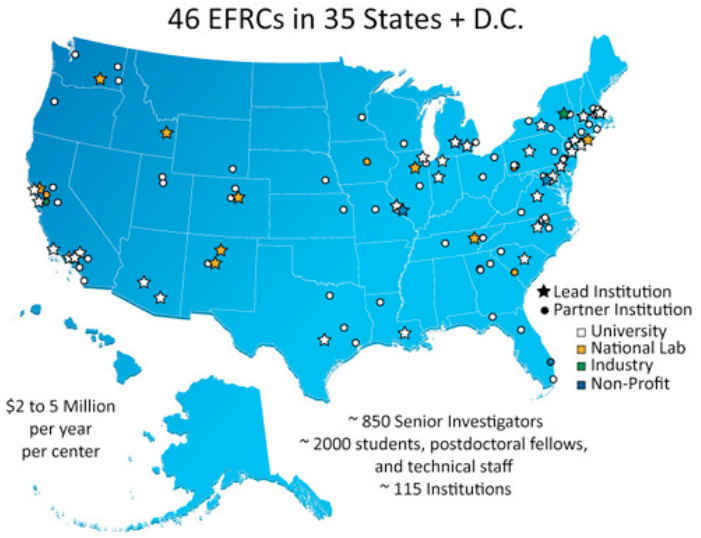




Energy Frontier Research Center

for Materials under Irradiation and Mechanical Extremes

http://science.energy.gov/bes/efrc/



Energy Frontier Research Center
CMIME
Center for Materials at Irradiation and Mechanical Extremes

Institutions:
LANL, MIT, UIUC, CMU, UNL

CMIME Org Chart
http://cmime.lanl.gov

Amit Misra, *Director*
Irene J. Beyerlein, *Co-Director*
Linda Chavez (admin)

Scientific Advisory Committee
Michael I. Baskes (Chair); William D. Nix; John P. Hirth; G. Robert Odette; Adrian Sutton; Francois Willaime.

Irradiation extremes thrust

Metals ***Oxides***

Staff scientists / faculty

Michael J. Demkowicz (MIT), lead
J. Alfredo Caro, PI
Amit Misra, PI
$**Stuart Maloy**, Co-I
$**Mike Nastasi** (UNL), Co-I
Blas P. Uberuaga, lead
Steve M. Valone, Co-I
$**Arthur F. Voter**, Co-I
$**Quanxi Jia**, Co-I

Post-docs

Nan Li, 50%, (Misra)
Osman Anderoglu, 50%, (Maloy/Misra)
Enrique Martinez-Seaz, (Caro)
Engang Fu, (Nastasi/ Y. Wang)
****Samrat Choudhary**, *fellow* (Uberuaga/Stanek)
Kedarnath Kolluri, 50% (Uberuaga/Valone)
Louis Vernon, 50%, (Voter/Uberuaga)
Zhenxing Bi, (Jia)
Jeff Aguiar, (Uberuaga/Misra)

Graduate Students

Liang Zhang, MIT (Demkowicz)
Abishek Kashinath, MIT (Demkowicz)
***Yun Xu**, NMSU (Luo/Uberuaga)
***Helen Telila**, UNM (Atlas/Valone)

Mechanical extremes thrust

Severe Plastic Deformation ***High Strain Rates***

Staff scientists / faculty

Irene J. Beyerlein, lead (theory)
Nathan Mara, lead (experiments)
Pascal Bellon (UIUC), PI
Robert S. Averback (UIUC), Co-I
$**Tony Rollett** (CMU), Co-I
****Jian Wang**, Co-I
Timothy C. Germann, lead
Ellen Cerreta, PI
Rusty Gray, Co-I
Richard G. Hoagland, consultant (Lab associate)

Post-docs

**** Keonwook Kang** (Beyerlein)
****John Carpenter** (Mara)
**** Shijian Zheng** (Mara)
Weizhong Han (Mara/Misra/Cerreta)
Christian Brandl, 50%, (Germann)
****Ruifeng Zhang**, *fellow* (Germann/ Beyerlein)
Alex Perez-Bergquist, 50%, (Cerreta)

Graduate Students

Elvan Ekiz, UIUC (Bellon)
Tim Lach, UIUC (Averback)
Jon LeDonne, CMU (Rollett/Mara)
***Thomas Nizolek**, UCSB (Pollock/Mara)
***Katrina Uychaco, UCSD** (Meyers/Germann)
****Benjamin Eftink, UIUC** (Robertson/Mara)

LANL Technical Staff:
Rob and Pat Dickerson (EML);
Yong Wang (IBML); J. Kevin Baldwin (PVD);
Faculty collaborators thru' *LANL Institutes*:
Tresa Pollock, UC-SB
Susan Atlas, UNM
Hongmei Luo, NMSU
Marc Meyers, UCSD

Dec 25th, 2011

*GRA supported through *LANL Institutes*
$ (post-doc or student support only)
** LANL/LDRD Leveraged

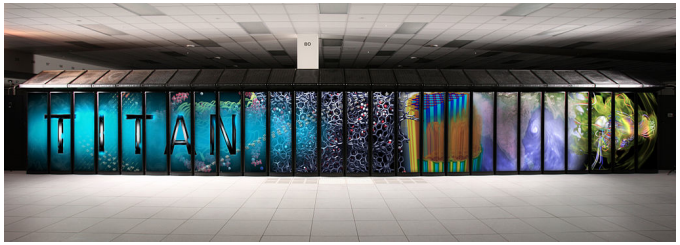
Former post-docs:
A. Vattre; X-M. Bai; A. Karakuscu; Mujin Zhuo.

18 LANL staff ≈ 4 FTE
5 faculty
16 post-docs (≈ 13.5 FTE)
10 students, including 5 supported by LANL institutional resources

These Centers involve significant resources and many people

Our Center at LANL focuses into interfaces

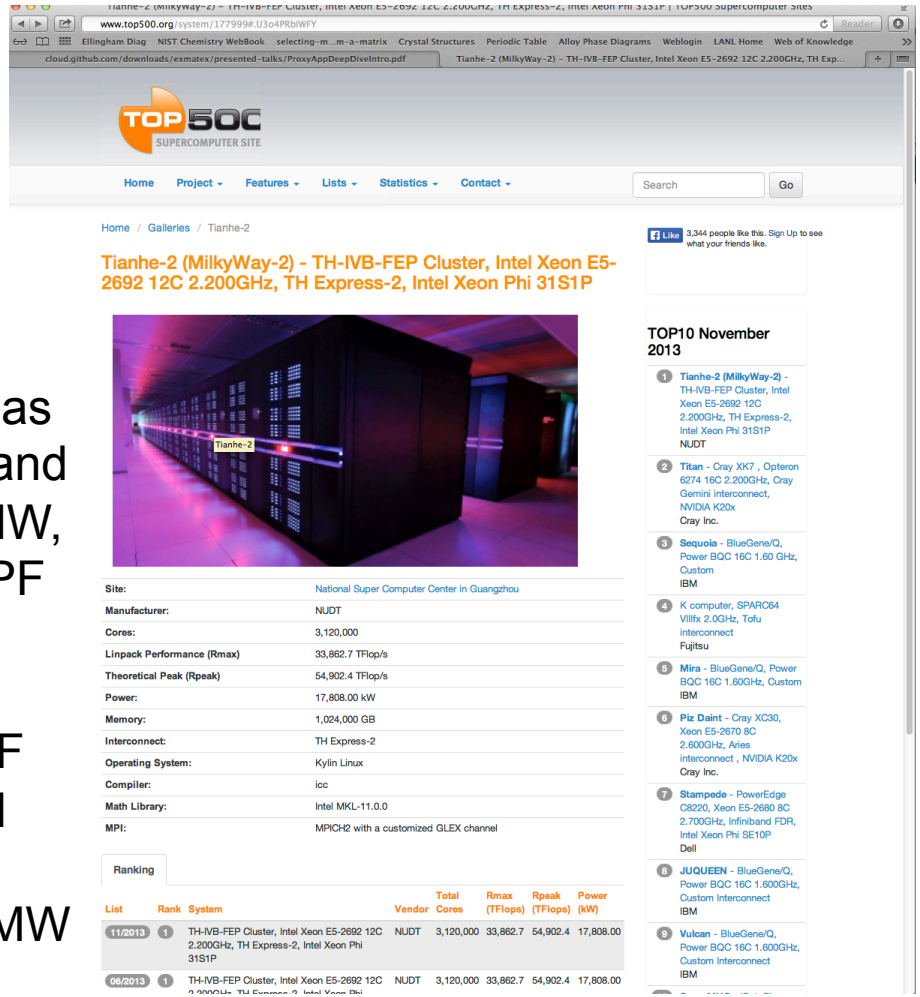
Government and industry are looking into exascale computer power



10^6 mega M
10^9 giga G
10^{12} tera T
10^{15} peta P
10^{18} exa E

The # 1 today has 3 million cores and consumes 17 MW, to deliver ~ 50 PF

With this technology, 1 EF computer would need 60 million cores and 340 MW



ExMatEx Extreme M Extreme Materials at Extreme Scale

NEWS | ABOUT + | PROXY APPS + | RESEARCH AREAS + | PUBLICATIONS | ENGAGEMENT | LINKS

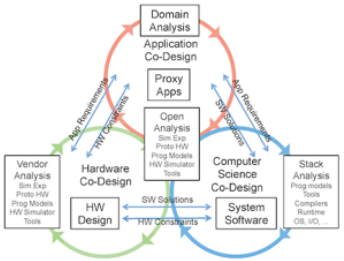
ExMatEx Overview

DoE Exascale Co-Design Center for Materials in Extreme Environments.

The objective of the Exascale Co-design Center for Materials in Extreme Environments (ExMatEx) is to establish the interrelationship among algorithms, system software, and hardware required to develop a multiphysics exascale simulation framework for modeling materials subjected to extreme mechanical and radiation environments. Such a simulation capability will play a key role in solving many of today's most pressing problems, including producing clean energy, extending nuclear reactor lifetimes, and certifying the aging nuclear stockpile.

Our goal is to establish the interrelationships between hardware, middleware (software stack), programming models and algorithms to enable a productive exascale environment for multiphysics simulations of materials in extreme mechanical and radiation environments.

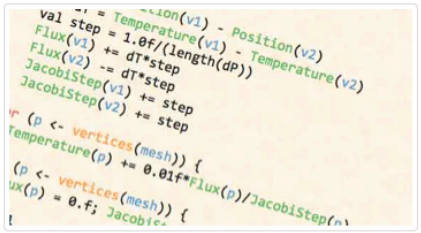
We will exploit, rather than avoid, the greatly increased levels of concurrency, heterogeneity, and flop/byte ratios expected on the upcoming extreme scale platforms.



Co-design of the exascale ecosystem involves ExMatEx and other application co-design centers working in concert with hardware vendors and other computer science research activities as illustrated in the image to the right. The computer system architecture is shown to contain both the emerging hardware as well as all of the software stack required to operate the computer. The role of the application co-design center is to both introduce our evolving application requirements and workflow into the exascale ecosystem through proxy application and to evaluate these applications in the context of emerging hardware and software solutions.

Research Areas

Our research spans a broad set of topics including computer science, algorithms, and applications.



PROGRAMMING MODELS

We're engaged in the assessment and evaluation of both traditional programming languages, emering programming paradigms, and domain specific languages.

RUNTIME SYSTEMS

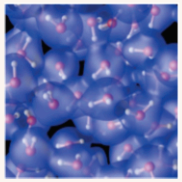
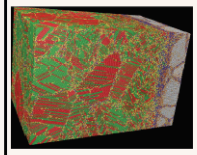
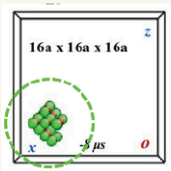
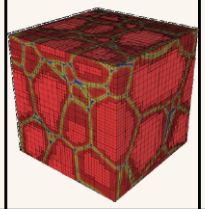
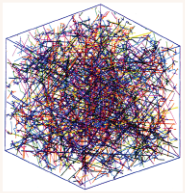
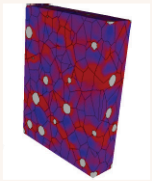
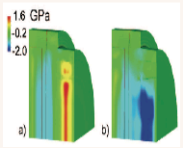
Runtime system services can support "programming in the large"—coupling multiple diverse components of a dynamic multi-scale computation and orchestrating its execution on the system.

ANALYSIS, MODELING & SIMULATION

Hardware-interfacing tools combining analytical models, architectural simulation, system emulation and empirical measurements drive the co-design process.

Subjects chosen to become the equivalent to LINPACK benchmarks for the exascale era

The Motifs of Materials Application Codes

Ab-initio	MD	Long-time	Phase Field	Dislocation	Crystal	Continuum
Inter-atomic forces, EOS	**Defects and interfaces, nucleation**	**defects and defect structures**	**Meso-scale multi-phase evolution**	**Meso-scale strength**	**Macro-scale material response**	**Macro-scale material response**
						
Code: Qbox/ LATTE Motif: Particles and wavefunctions, plane wave DFT, ScaLAPACK, BLACS, and custom parallel 3D FFTs Prog. Model: MPI + CUBLAS/ CUDA	Code: SPaSM/ ddcMD/CoMD Motif: Particles, explicit time integration, neighbor and linked lists, dynamic load balancing, parity error recovery, and *in situ* visualization Prog. Model: MPI + Threads	Code: SEAKMC Motif: Particles and defects, explicit time integration, neighbor and linked lists, and *in situ* visualization Prog. Model: MPI + Threads	Code: AMPE/GL Motif: Regular and adaptive grids, implicit time integration, real-space and spectral methods, complex order parameter Prog. Model: MPI	Code: ParaDis Motif: "segments" Regular mesh, implicit time integration, fast multipole method Prog. Model: MPI	Code: VP-FFT Motif: Regular grids, tensor arithmatic, meshless image processing, implicit time integration, 3D FFTs. Prog. Model: MPI + Threads	Code: ALE3D/ LULESH Motif: Regular and irregular grids, explicit and implicit time integration. Prog. Model: MPI + Threads

ExMatEx

U.S. DEPARTMENT OF ENERGY | Office of Science

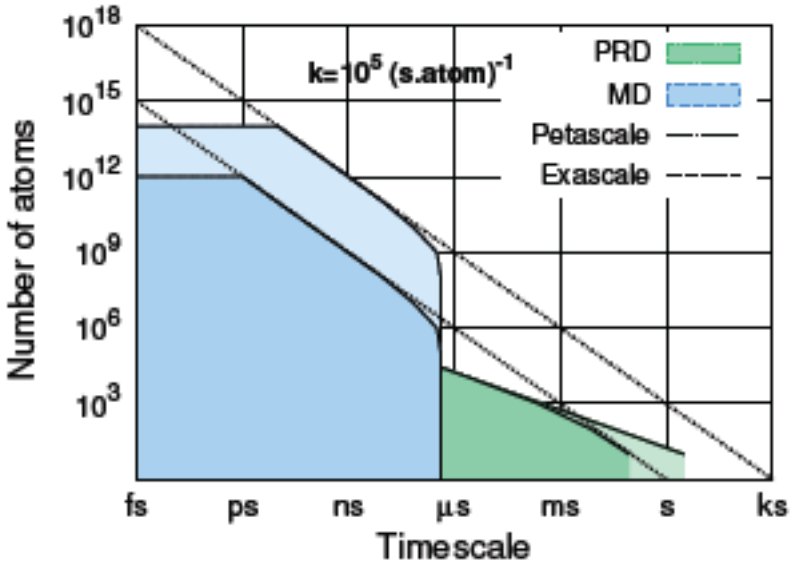
3

Note that VP-FFT is a development of an Argentinian colleague working at LANL (R. Lebensohn)

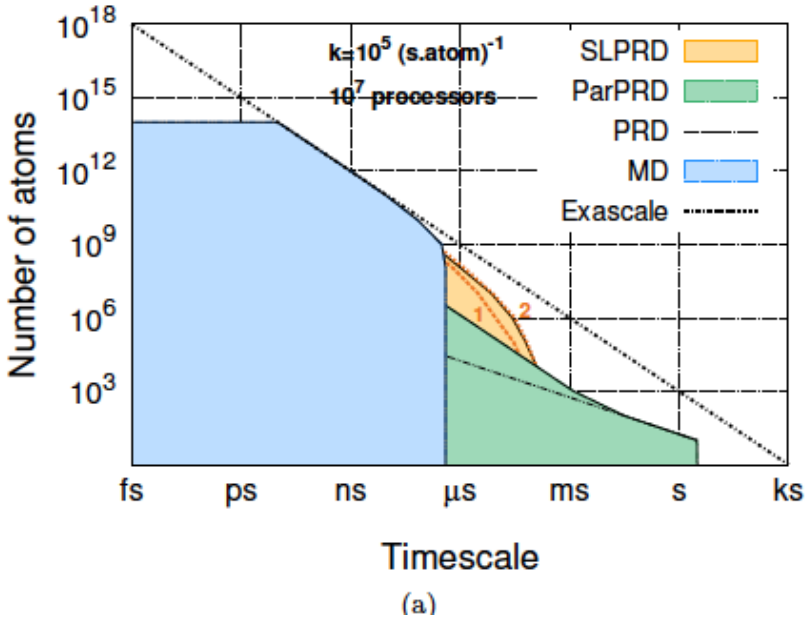
Example of exascale challenges: Sublattice parallel replica dynamics

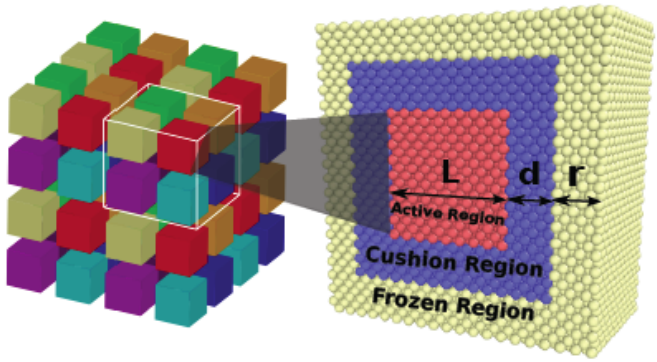
Enrique Martínez, Blas P. Uberuaga, and Arthur F. Voter (in press)

- Exascale computing presents a challenge for the scientific community as new algorithms must be developed to take full advantage of the new computing paradigm
- Molecular dynamics and parallel replica dynamics fail to use the whole machine speedup, leaving a region in time and sample size that is unattainable with current algorithms
- Molecular dynamics (MD) algorithms are extremely efficient in parallelizing space,4 and therefore large atomic systems can be simulated for short times with unprecedented accuracy
- However, these traditional algorithms are not suitable for studying long-time phenomena, such as vacancy diffusion, as they become communication bound and the characteristic time for the process becomes extremely hard to attain
- Parallel replica dynamics (PRD) exploits the fact that for many physical processes the system trajectory executes transitions from state to state on time scales orders of magnitude larger than the atomic vibrations; i.e., the dynamics of the system are dominated by infrequent events.



A new strategy for domain decomposition alows to fill part of the gap in the exascale triangle





Exascale triangle, assuming 10^7 processors, for the three different methods, MD (blue), ParPRD (green) and SLPRD (beige) for event rates of k = 105 $(s.atom)^{-1}$

Spatial domain decomposition. To avoid boundary conflicts the domains are further divided into subdomains. Simulation cycles among the colors, performing parallel replica dynamics in each of the subdomain independently

Back to the atomic scale: The modelers' playground

Many important applications of materials involve interfaces and mixtures between disparate bonding environments

Improving the ability to model such interfaces promises new capabilities in computationally prototyping new materials

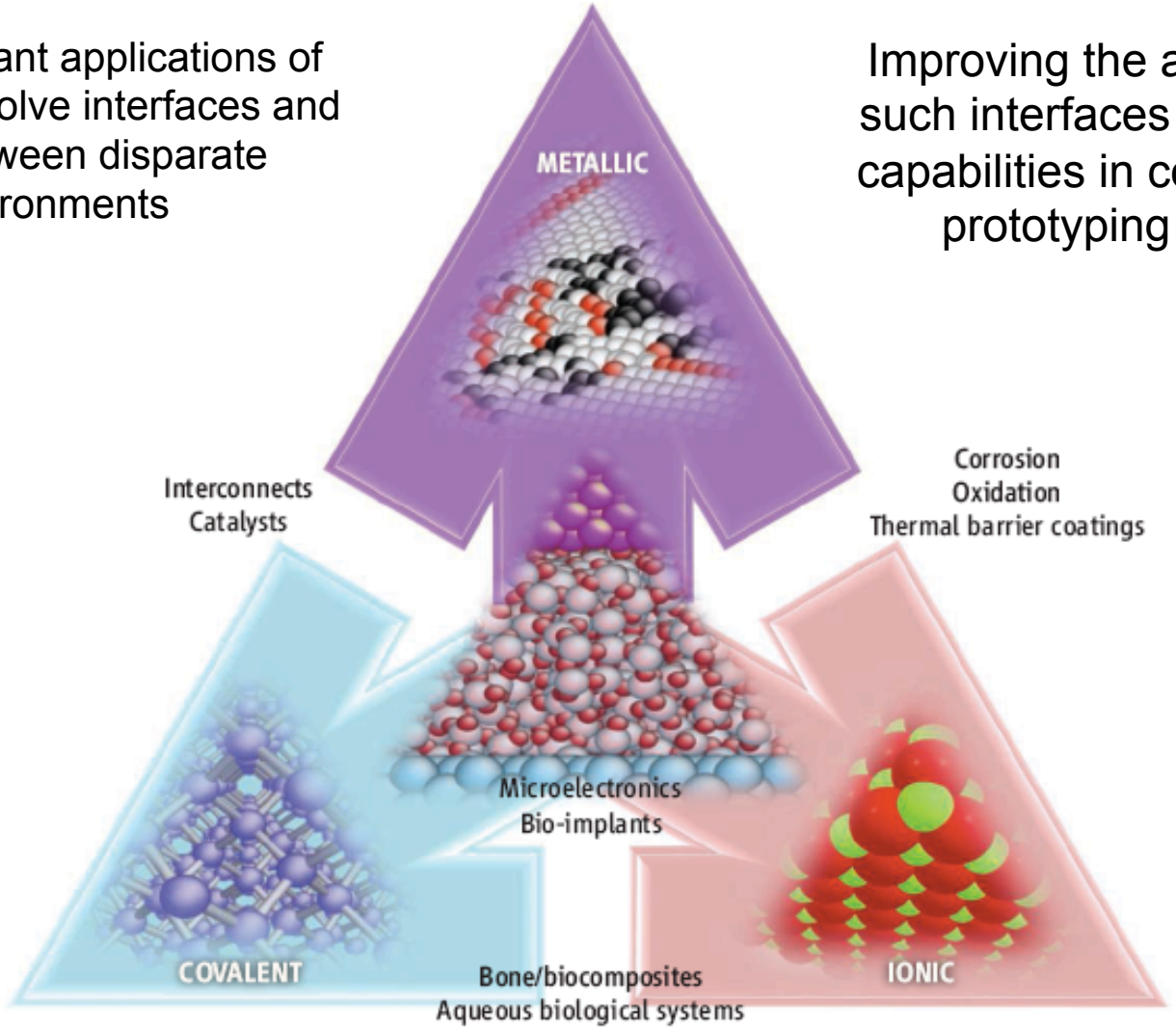
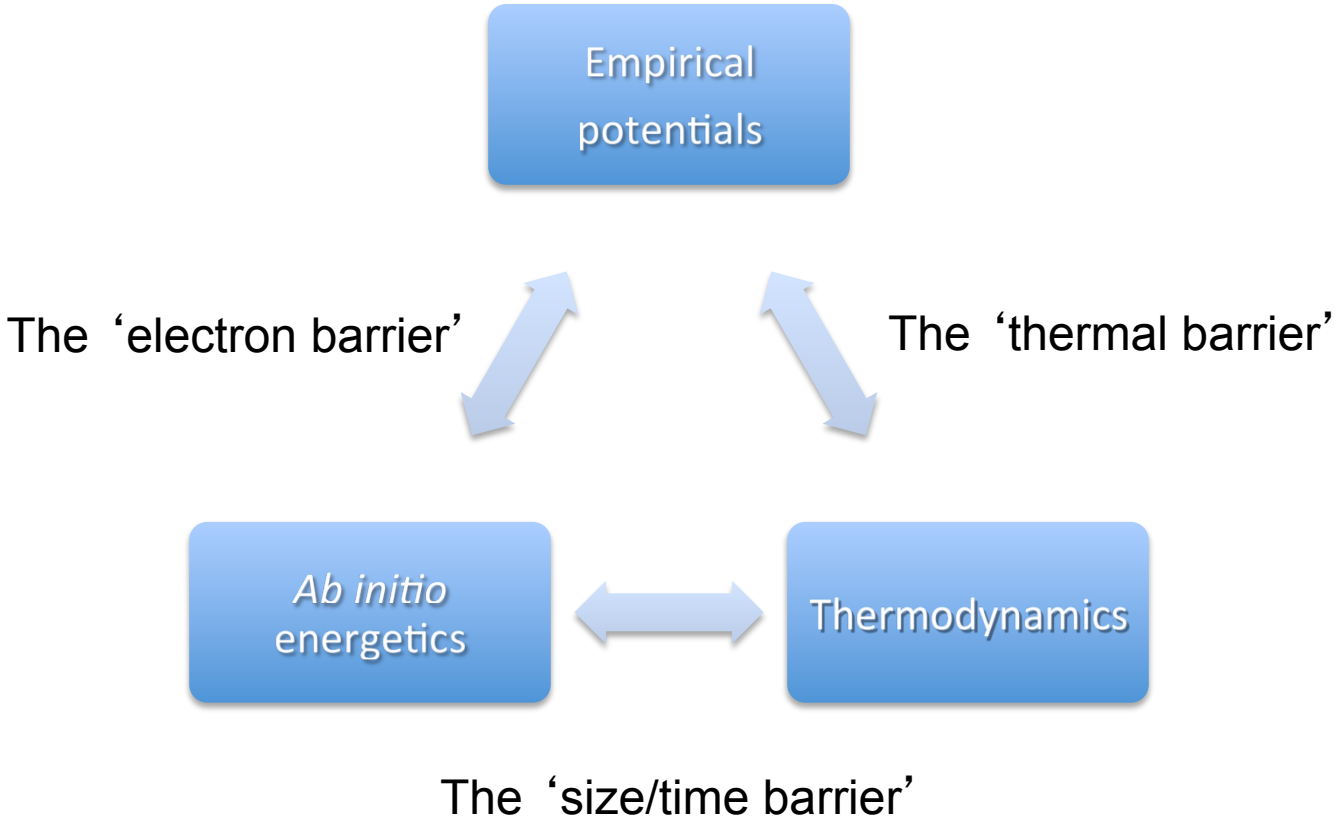


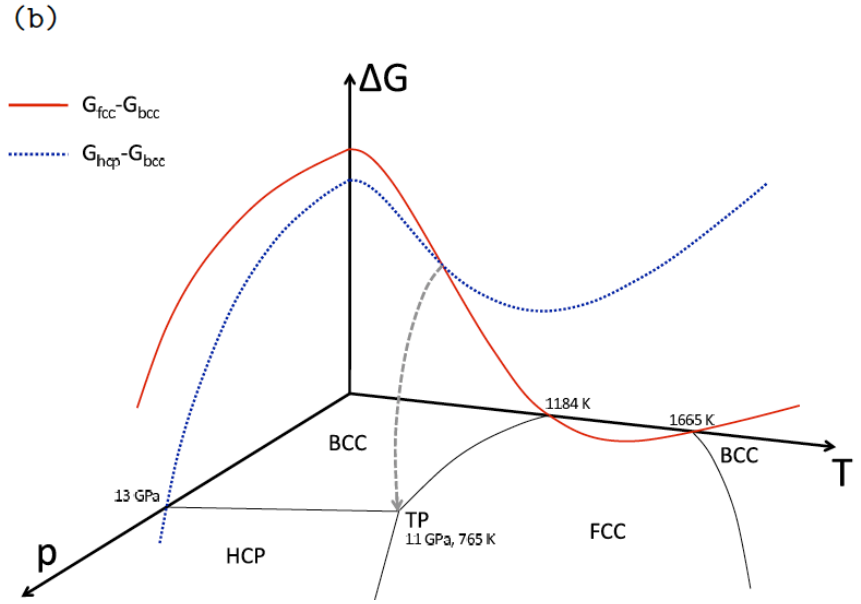
Figure from S. Phillpot and Su. Sinnott. Science 09/2009

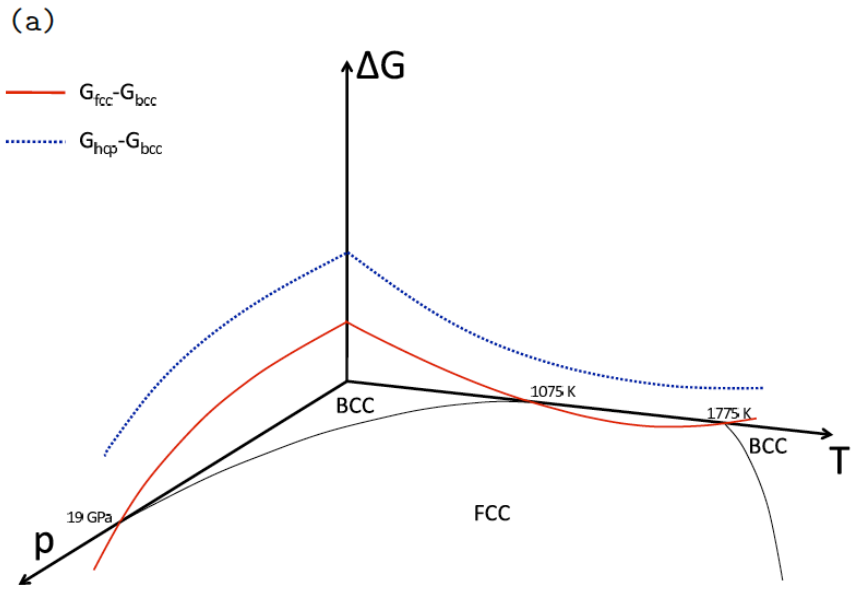
Our work on computational materials science at the atomic scale focuses on several challenges



The ‘thermal barrier:’ Fe polymorphism

T. Lee & S. Valone
MST-8 Materials Science and Technology





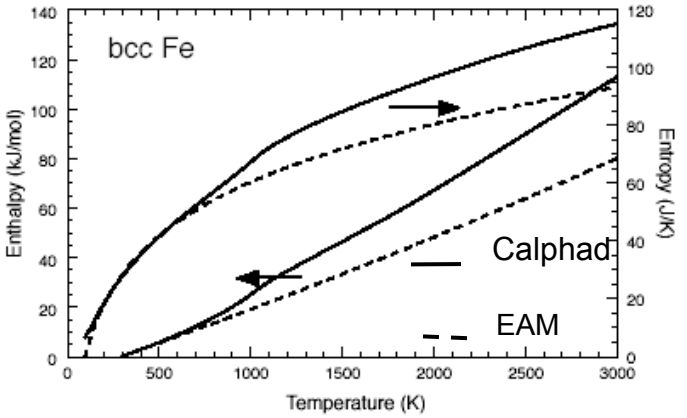
Fe has bcc-to-fcc transition as a function of T and a bcc-to-hcp transition as a function of P
→ a triple point

Classical potentials have so far been unable to capture both transitions simultaneously

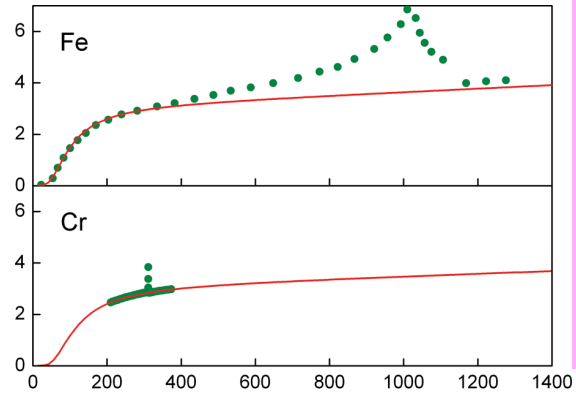
T. Lee et al., JPCM 24,225404 (2012)

Finite-T properties are difficult to capture when e-e correlations are important

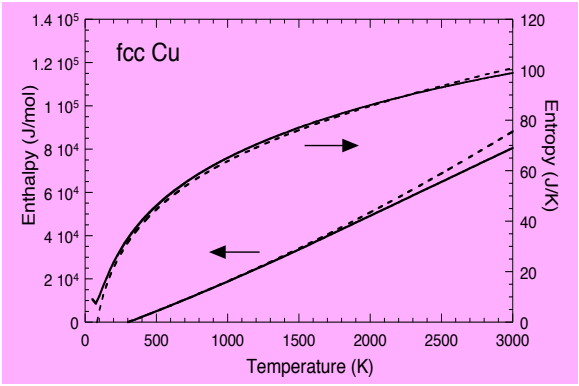
EAM and CALPHAD Fe enthalpies and entropies



Heat capacities of pure metals



Dots: experimental data
Lines: empirical potentials
From E. Martinez 2012

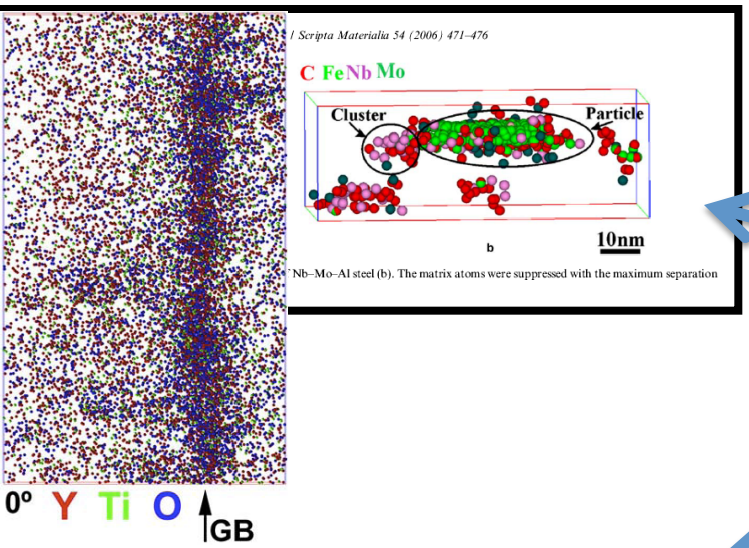


Cu, shown for comparison

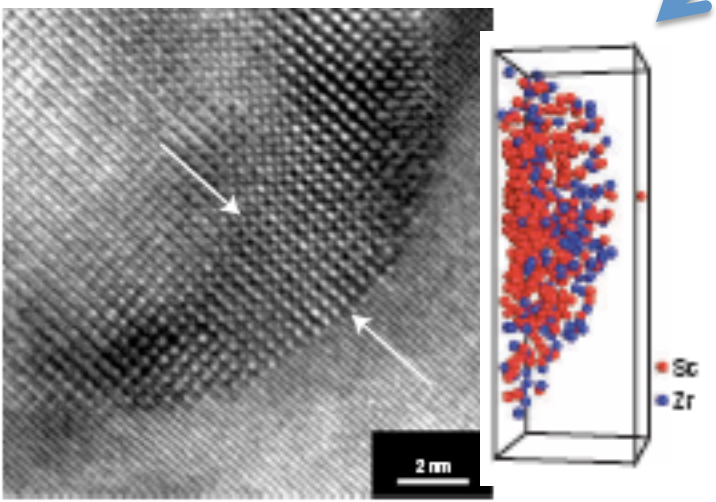
This behavior represents a major obstacle for empirical potentials for Fe, whose validity is therefore restricted to the FM phase, well below the Curie T

The ‘thermal barrier’

ODS ferritic steels, Miller 2009



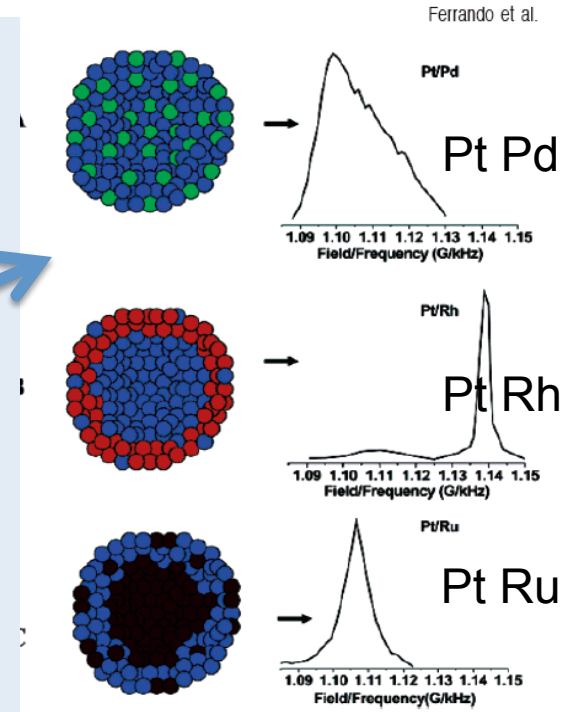
Al-Zr-Sc nanostructured alloy, Clouet 2006



Most of the nano-engineered materials today are

- non-equilibrium
- multiphase
- multicomponent

systems, where thermodynamics and kinetics play crucial roles in determining their microstructure



igure 10. Schematic representation of three different Pt−alloy anoparticle systems with their corresponding ^{195}Pt NMR spectra: A) Pt−Pd, (B) Pt−Rh, and (C) Pt−Ru. Pt, Pd, Rh, and Ru atoms e shown in blue, green, red, and black, respectively. (Reprinted with permission from ref 129. Copyright 2003 American Chemical Society.)

Nanoalloys. Ferrando 2008

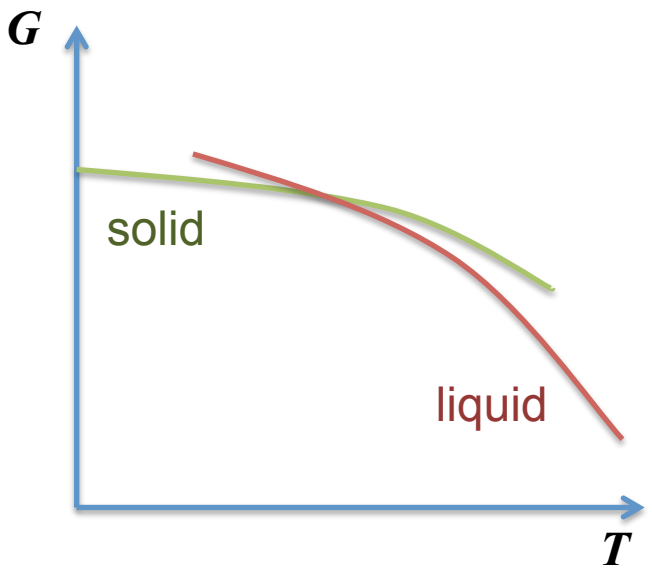
The 'thermal barrier'
Computational thermodynamics

The free energy G is a thermodynamic potential that determines the thermodynamic properties of a material

$$G = U + pV - TS$$

The chemical potential can be derived from G as

$$\mu_i = \left(\frac{\partial G}{\partial N_i} \right)_{T,P,N_{j \neq i}}$$



G is minimium at equilibrium

It can thus be used to determine phase stabilities, phase diagram of a system, and the driving forces when out of equilibrium

Can we obtain G from atomistic simulations?

Computational thermodynamics

Free energy G cannot be obtained as an ensemble average
We use Mixed Hamiltonian with switching parameter λ

$$\lambda U + (1 - \lambda) W$$

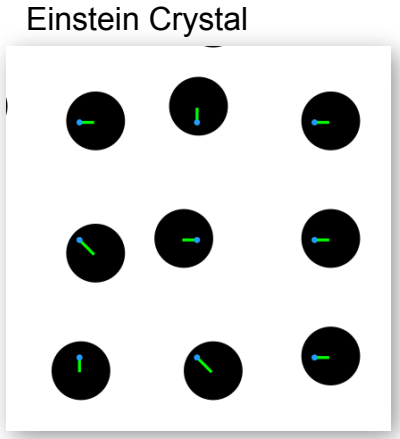
Free energy F_W of the harmonic oscillator system can be computed analytically

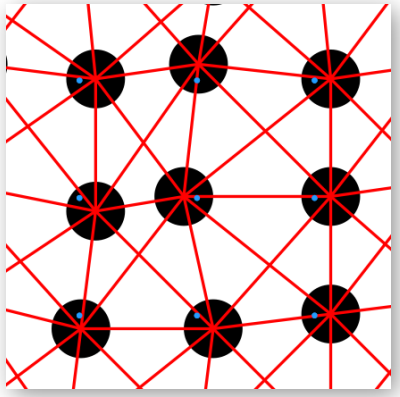
$$G_W = -kT \log \Omega_W \quad , \quad \Omega_W = \left(\frac{T}{T_{\text{Einstein}}} \right)^3$$

Evaluate the ensemble with respect to the pure Hamiltonians U (full interaction) and W (harmonic oscillators) and compute

$$G_U = G_W + \underbrace{\int_0^1 \langle U - W \rangle d\lambda}_{\text{switching work}}$$

We implemented this technique in Lammps (free distribution MD code)



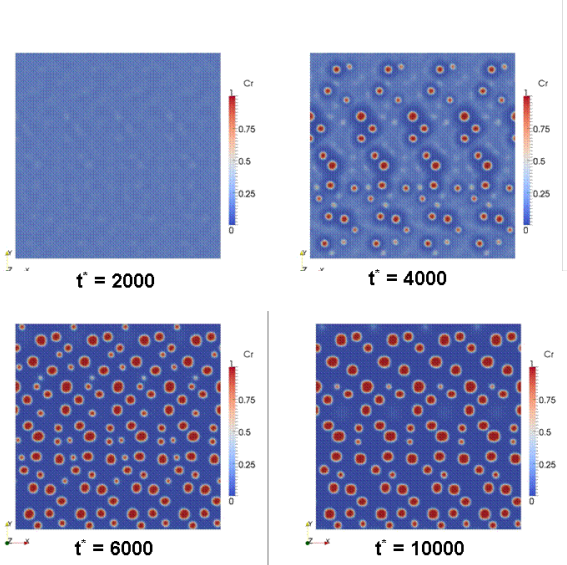

Fully interacting system

Microstructural evolution of $Fe_{80}Cr_{20}$ at 535 K

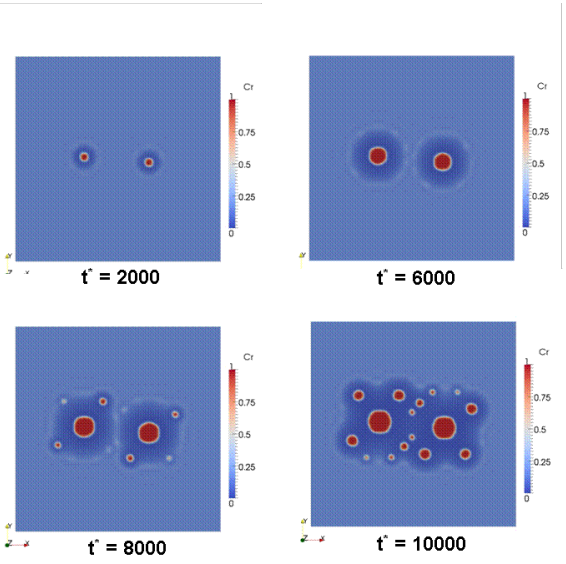
Work on Phase Field modeling by M. Tonks and P. Millet (INL)
as part of the NEAMS program

Work on LKMC modeling done at LANL
by E. Martinez in collaboration with CEA Saclay (Fr)
as part of the EFRC program

In a single crystal



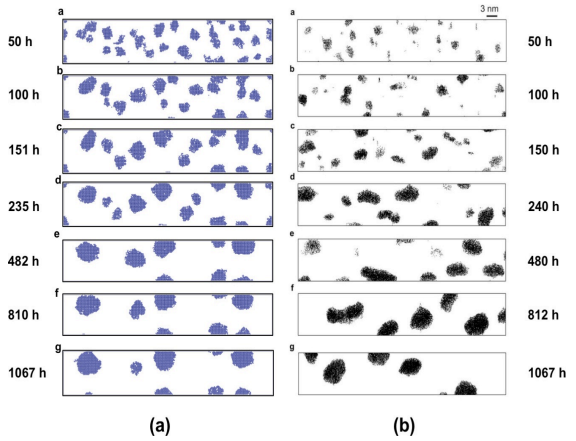
In presence of an edge dislocation dipole



LKMC

3D atom probe

Novy et al (2009)



The size / time barrier
Atomistic Kinetic Monte Carlo

Louis Vernon, Enrique Martinez, Blas Uberuaga, Art Voter, and Alfredo Caro
T and MST Divisions

Objective:

Many systems of interest share a common feature: their long-time dynamics consists of infrequent jumps between different states (i.e., activated processes).

Accelerated Molecular Dynamics methods (A. Voter's presentation) and kinetic Monte Carlo (KMC) algorithms can be used to extend the simulated time.

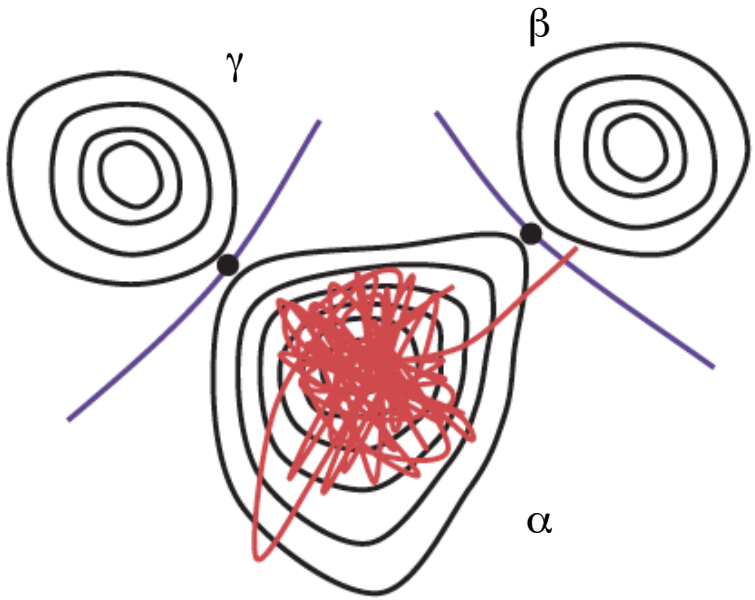
In this presentation we focus in the KMC algorithm

The MD timescale problem

Classical MD can only reach nanoseconds to microseconds due to the stiffness of the equations of motion (time step is limited to fs)

Processes of interest often take much longer:

- Vapor-deposited film growth (s)
- STM/AFM surface manipulation, nanoindentation (ms – s)
- Bulk and surface diffusion processes
- Radiation damage annealing (ns to years)
- Protein folding (μs - s)
- Precipitation

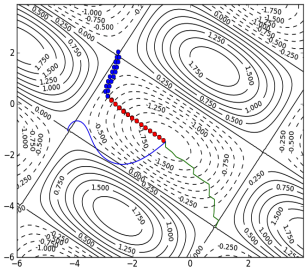


Our Kinetic Monte Carlo Approaches

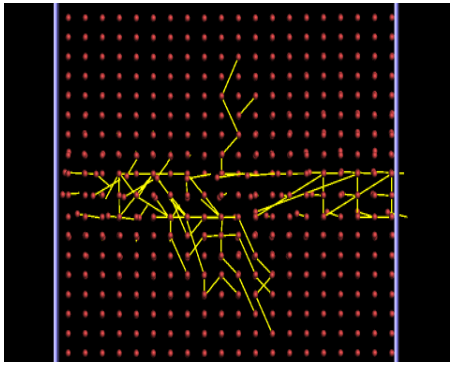
Self-learning KMC

- Dynamically explores the potential energy surface to discover all processes.
- Calculate the rates accurately.
- Is accurate and computationally demanding.

Transition searches on a distorted eggbox potential.



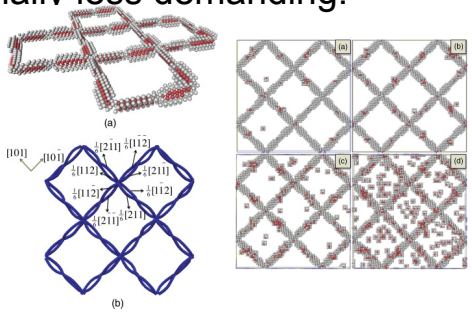
Defects diffuse to and within a complex grain boundary - evolved using SL-KMC.



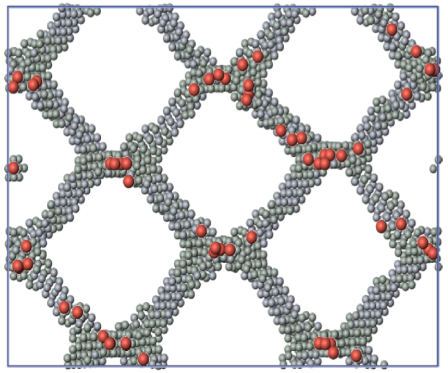
Event-driven KMC

- Uses the local microstructure to guess possible processes
- The accuracy in the rate calculation can be chosen
- Computationallv less demanding.

Cluster of vacancies in a (100) twist boundary in Cu, showing the preference for vacancies to go to constrictions



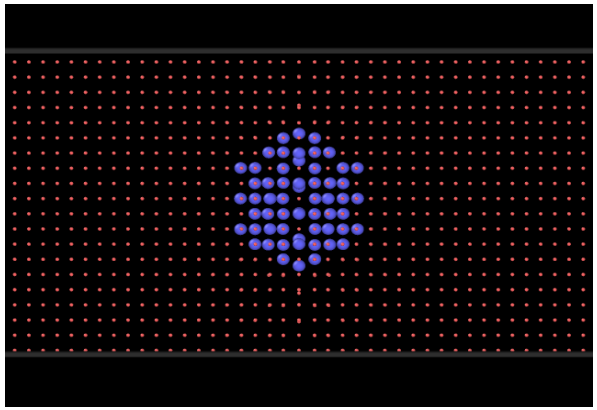
Clustering of vacancies at a (110) twist boundary in Fe, composed of a network of screw dislocations



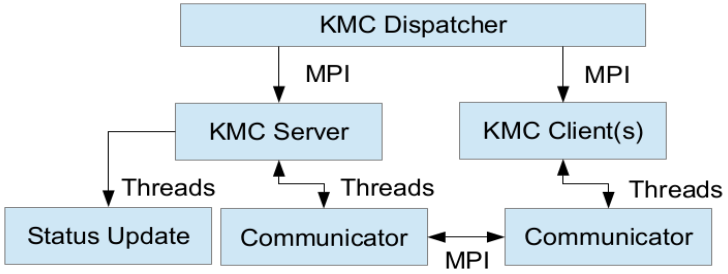
Self-Learning kinetic Monte Carlo

Transition searches are pricey

- Computationally more demanding than predefined processes.
  - Optimisation is essential.
    - Efficient transition searches.
      - Defect localisation.
    - Information recycling.
    - Parallelised workload.



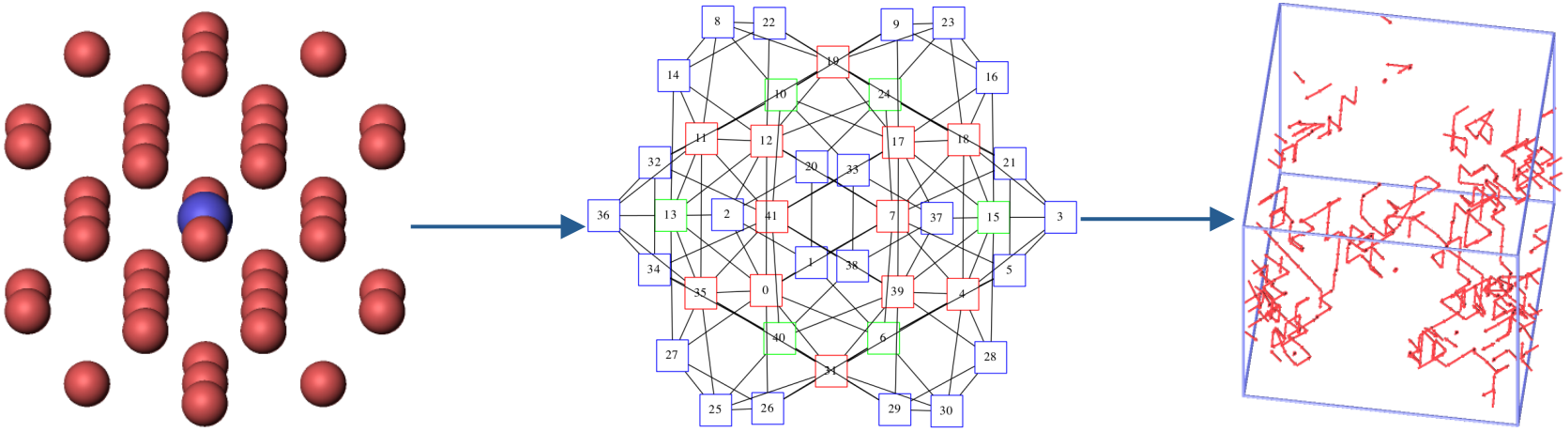
Transition searches are localised to regions containing defects.



Transition searches and minimisations are distributed across many processors using MPI.

Localisation allows categorisation

- Localising defects:
  - Reduced search dimensionality.
  - **Defect classification.**
    - Defect recognition.



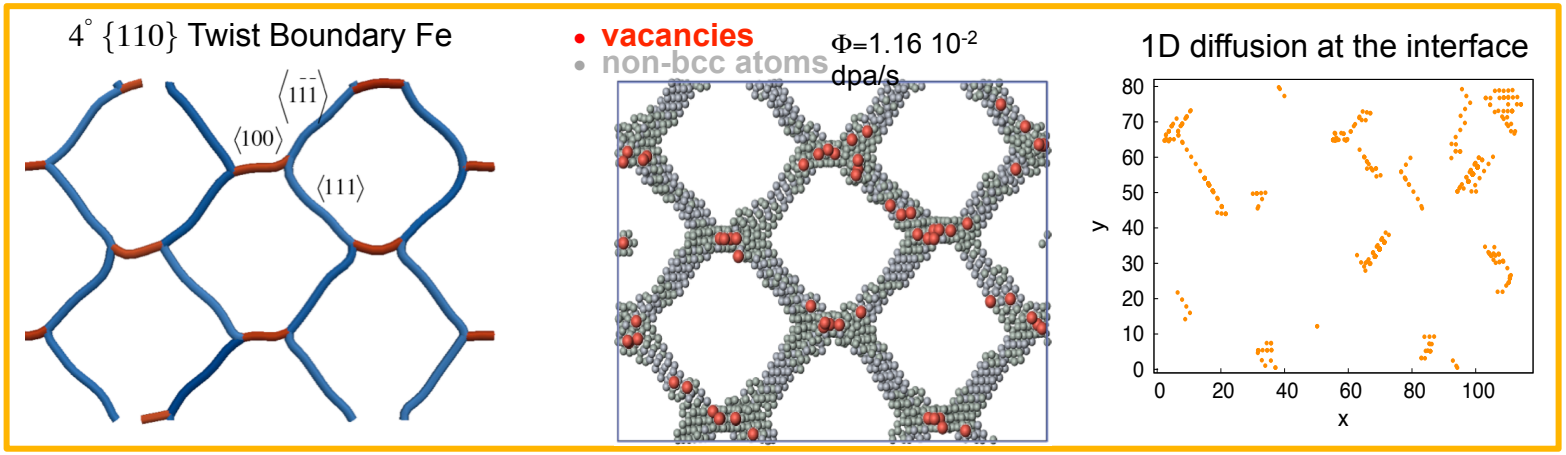
Atoms local to a vacancy (highlighted) are extracted.

A connectivity graph/hash representation is determined.

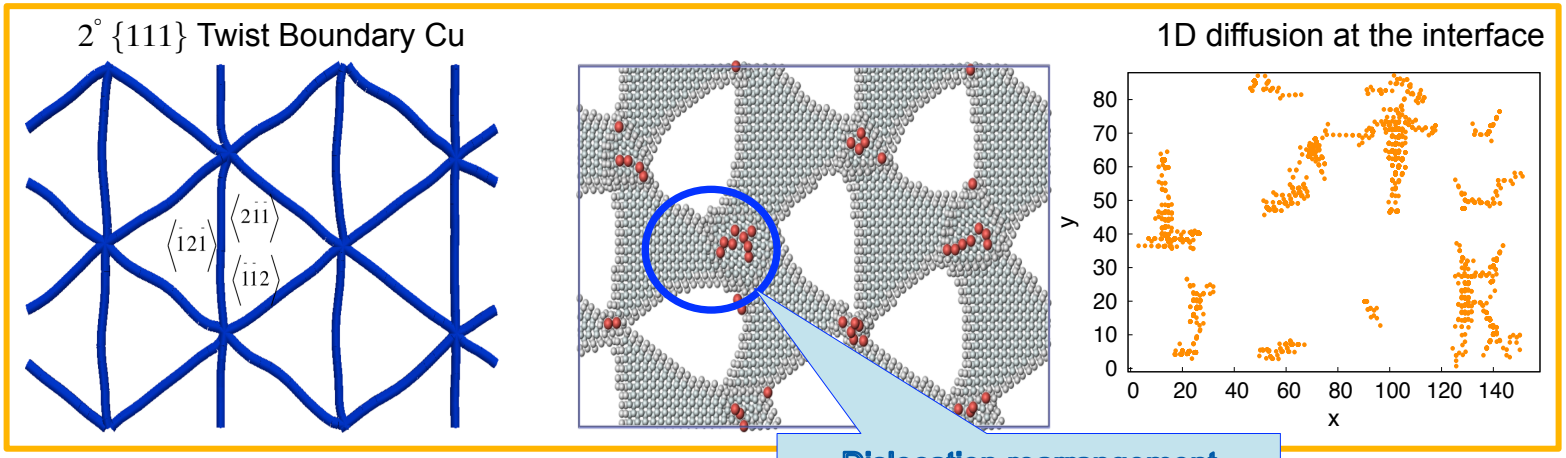
The vacancy rapidly diffuses with no new transition searches.

Transitions can be mapped onto equivalent defects

Event-Driven Kinetic Monte Carlo Off-lattice Relaxations



t = ~10^{-2} s, beyond MD capabilities



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Physical Review B 86 (2012) 214109

Radiation effects on nanoscale metallic foams

A Caro, M Caro, Y. Wang, E. Fu, E. Martinez, D. Schwen, K. Baldwin, MST
w/collaborations @ LLNL, VT

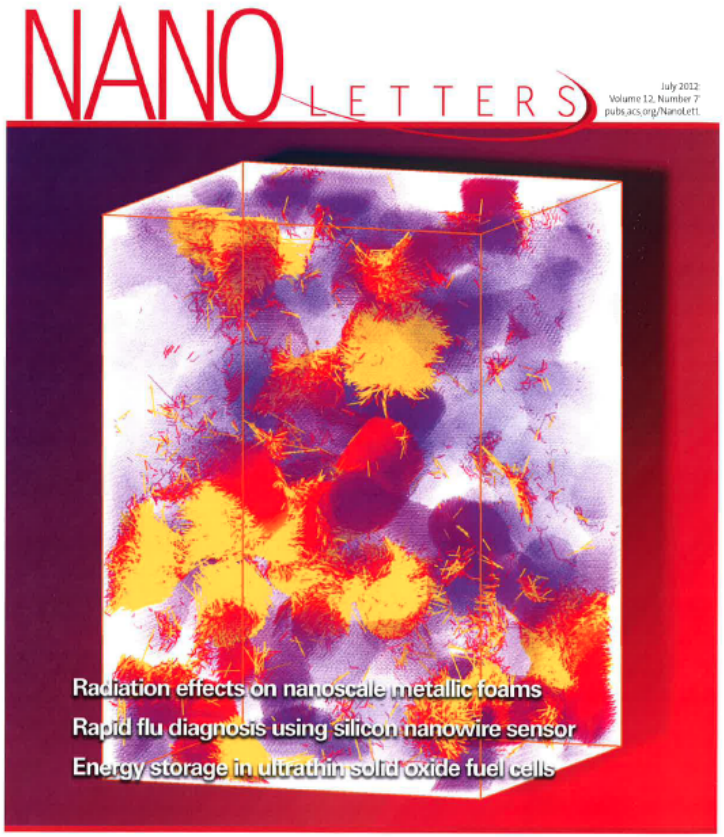
Objective:

- Explore the physics of surface-driven bulk physical behavior, such radiation tolerance

Opportunity:

- Nanoporous materials could become a new class of extremely radiation tolerant materials
  - Nanoporous materials offer a large amount of free surfaces
  - Free surfaces act as sinks providing opportunities for irradiation induced defects to annihilate through diffusion

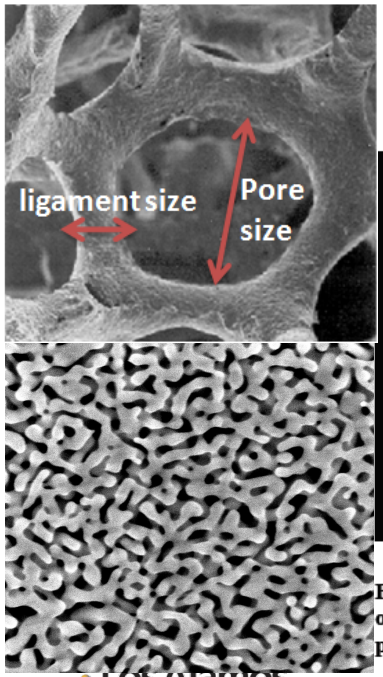
Cover of Nano Letters July 2012



Making foams in the computer

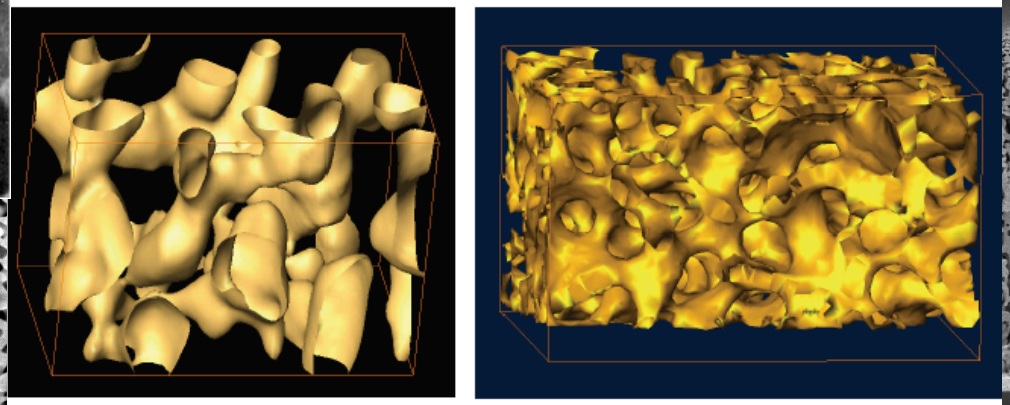
Two methods have been reported:

- Spinodal decomposition (an AB solution with a miscibility gap
- Gas condensation (solidification of a low density gas



Experimental sample

Computer sample



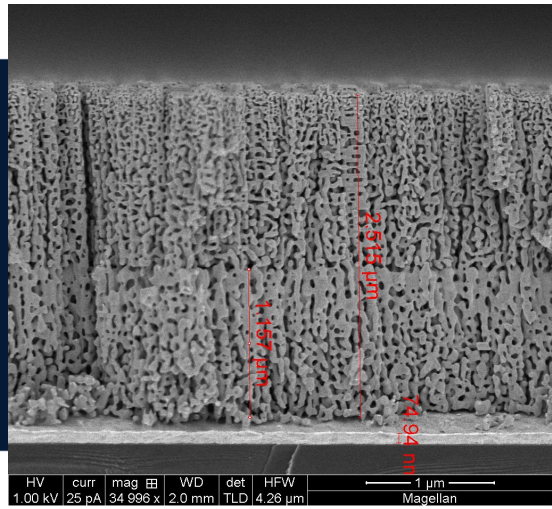
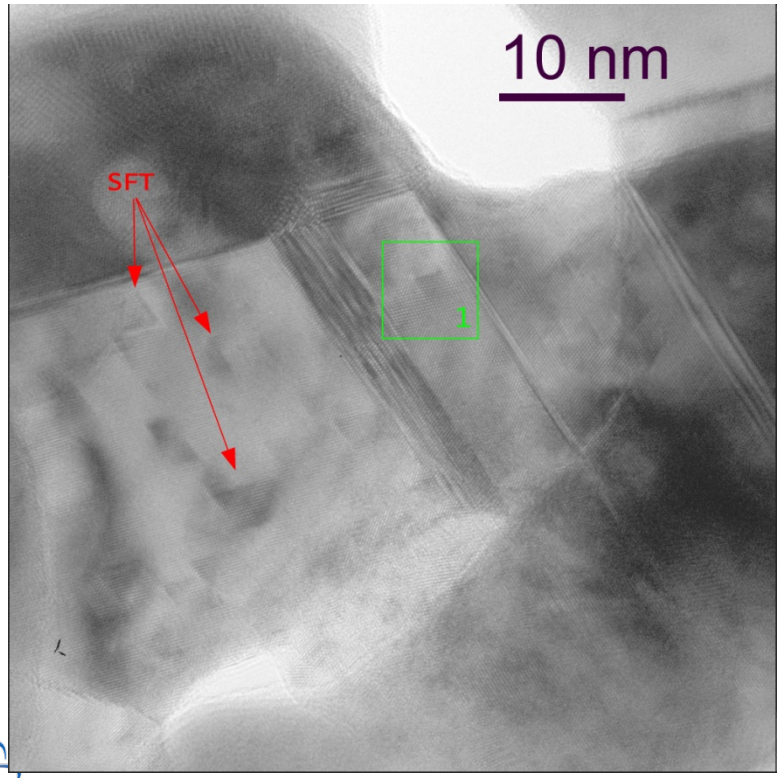
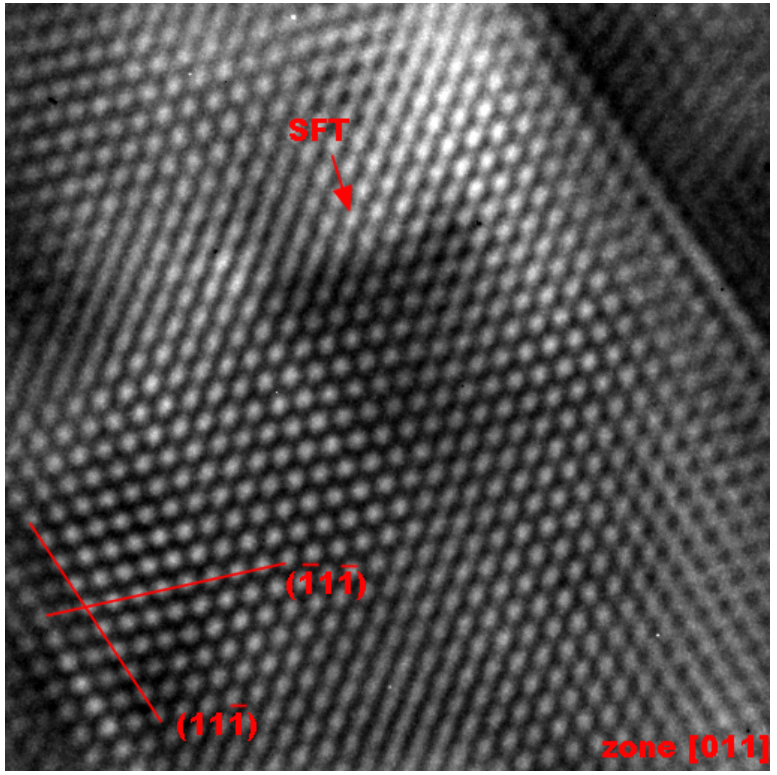


Figure 4: Left: Surface rendering of a typical volume element in a nanoporous gold sample, as obtained by electron tomography. Ref.[7]. Right: np-Au digital sample prepared using the technique proposed here.

HRTEM shows SFTs are formed in irradiated np-Au

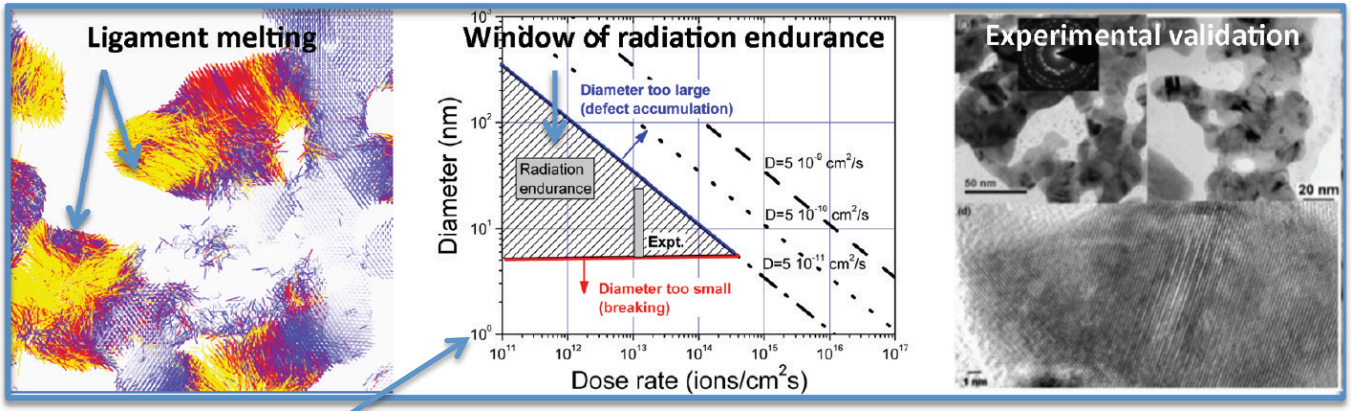
- **SFTs are observed in np-Au foams irradiated at RT at intermediate and high flux values; i.e. flux > 6×10^{10} ions/cm^2/s**
- **No SFTs observed at low dose-rates and/or LNT irradiations**





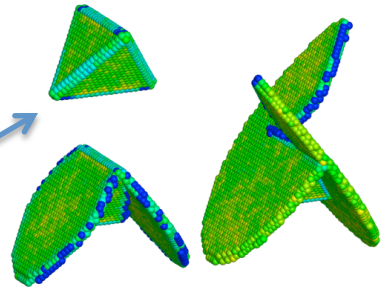
Box 1: ~ 5 nm large SFT along (110) projection at the highest flux of 3×10^{12} /cm^2/s

Are nanofoams radiation resistant?

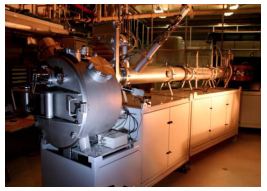


Major results:

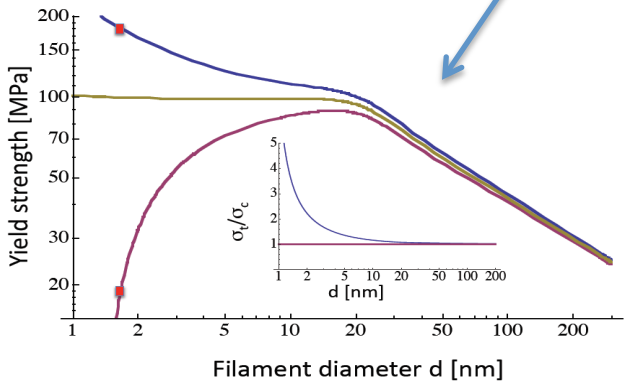
- A window of radiation resistance
- Radiation produces little damage in nanoscale foams, due to the large surface to volume ratio
-
- Main damage mechanisms is the creation of stacking fault tetrahedra, which induce a softening of the foam

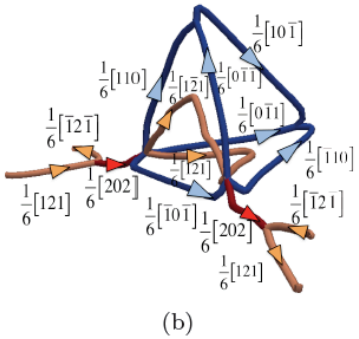


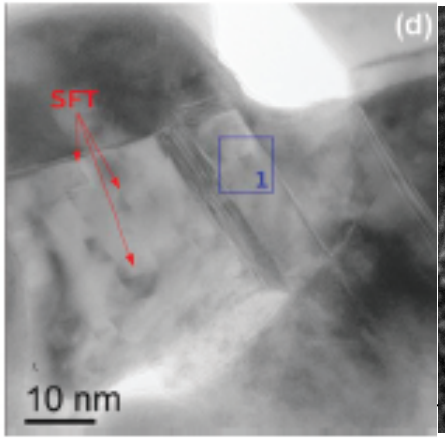
Material synthesis at CINT, irradiations at IBL

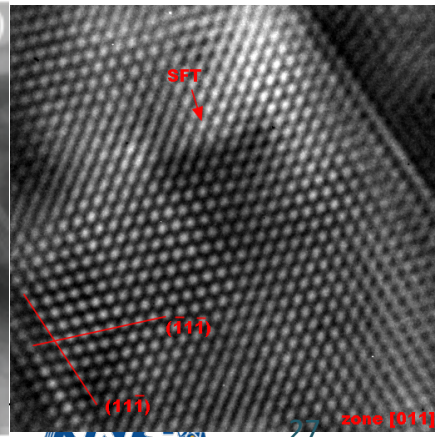


- Strong a-symmetry in the mechanical response in tension and compression









Summary and Future Directions

Our computational materials science work is based on both,

- The use of well established techniques such as:
  - *Ab initio* electronic structure calculations to predict energetics of materials
  - Classical molecular dynamics
  - Computational thermodynamics

- Our research portfolio on multiscale computational modeling is characterized by a significant effort on the development of new models, algorithms and tools at almost every scale in the multiscale paradigm, aiming at trespassing the current limitations on fidelity and accuracy

- The development of new models, algorithms, and tools to address problems such as:
  - The ‘electron barrier’
  - The ‘thermal barrier’
  - The ‘size / time barrier’