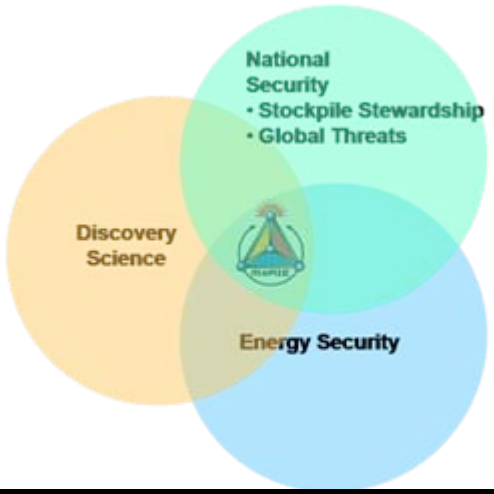
MaRIE Theory, Modeling and Simulation Integration

Mark B. Chadwick (T-DO) and Turab Lookman (T4)

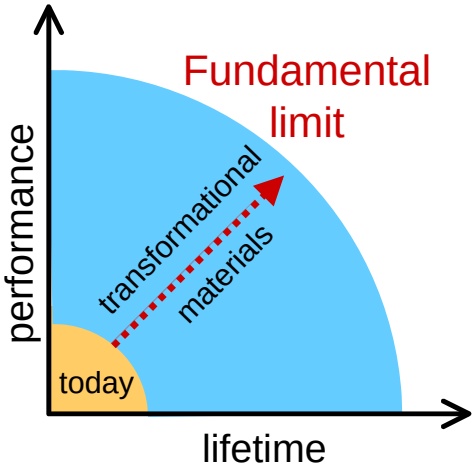
TMC BOD
F. Addessio, F. Alexander, M.J. Andrews, C. Bronkhorst, A. Balatsky, E.D. Dendy,, E. Kober, T. Kelley, M. Stan, S. Valone, J. Wills, J. Wendelberger

MaRIE: Materials R&D Capability for 21st Century National Security and Energy Security



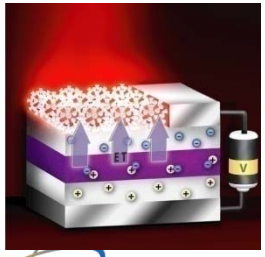
MaRIE addresses materials challenges across missions

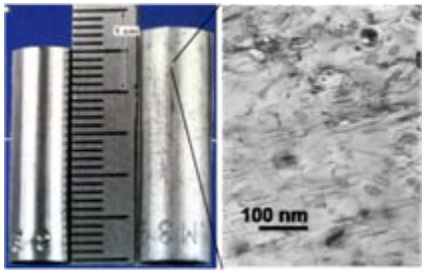
MaRIE enables the transition from "observation" to "control"



MaRIE will transform the science of microstructure, interfaces, and defects

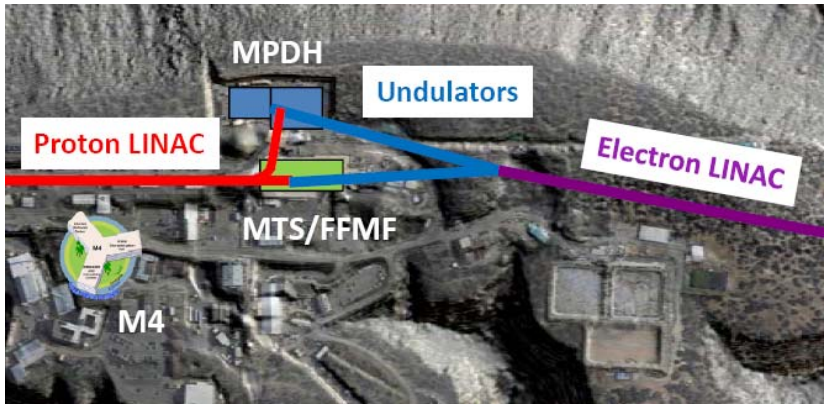
Next-generation solar cell architecture





Radiation-induced swelling

MaRIE provides tools for transformational materials performance in extremes



The transition from "observation & validation" to "prediction & control" is a central mission challenge AND the frontier of materials research

- **Conquering "the micron frontier" is essential for solving transformational materials grand challenges**

J. Sarrao

- **MaRIE will provide unique capabilities**
  - **Simultaneous *in situ* imaging and scattering measurements**
  - **Accessing materials irradiation/damage extremes**
  - **Incubating materials discovery and solutions through control of defects and interfaces**

MPDH C. Barnes

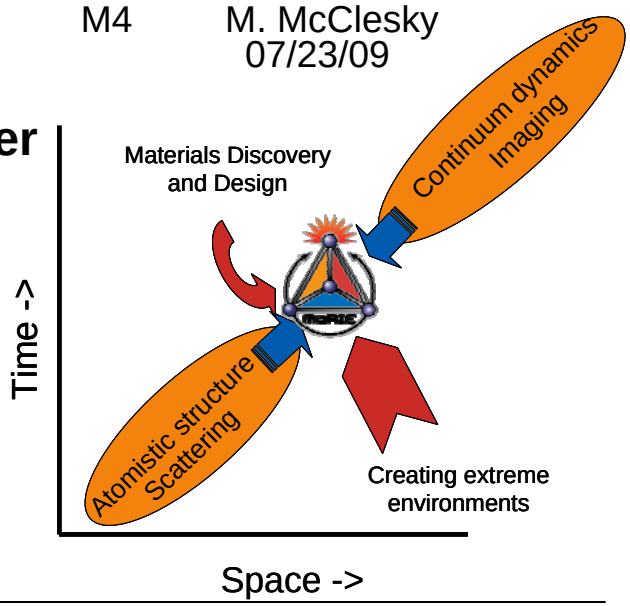
FFF M. Bourke

M4 M. McClesky 07/23/09

- **MaRIE provides unprecedented international user resources in a national security science setting**

LANSCE is essential for MaRIE's success

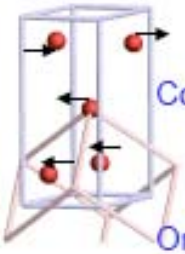
MaRIE will be the first capability with unique co-located tools necessary to realize transformational advances in materials performance in extremes



Bridging the micron gap: the challenge at the mesoscale

$\mathring{A}$ — nm — μm — mm

Atomic



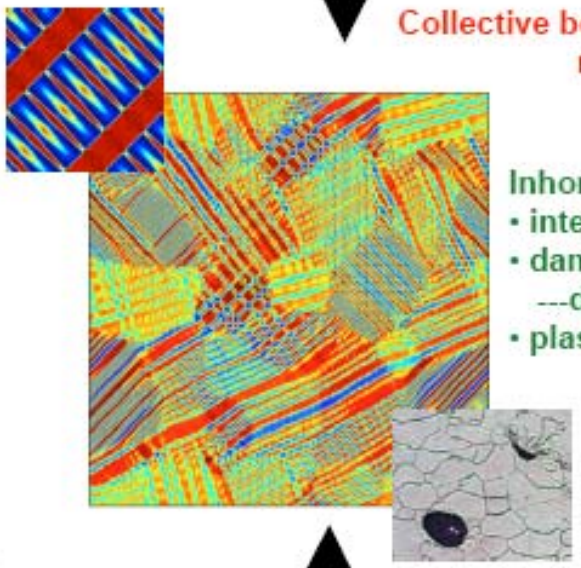
Collective modes

Order parameter fields

First principles
- Molecular dynamics
- Electronic structure

Experimental
- probes, characterization, 3D imaging

Collective behavior and microstructure



Inhomogeneities:
- interfaces, twins,
- damage (brittle, ductile)
  ---defects,voids, cracks
- plasticity (dislocations)

Theoretical
materials science, condensed matter & statistical physics
- Landau (phase field)
- self-consistent field methods
- nonequilibrium, statistical fluctuations
- dislocation dynamics

Computational
- data mining
- inference
- learning
- visualization

Macroscale

Elastic free energies, kinetics

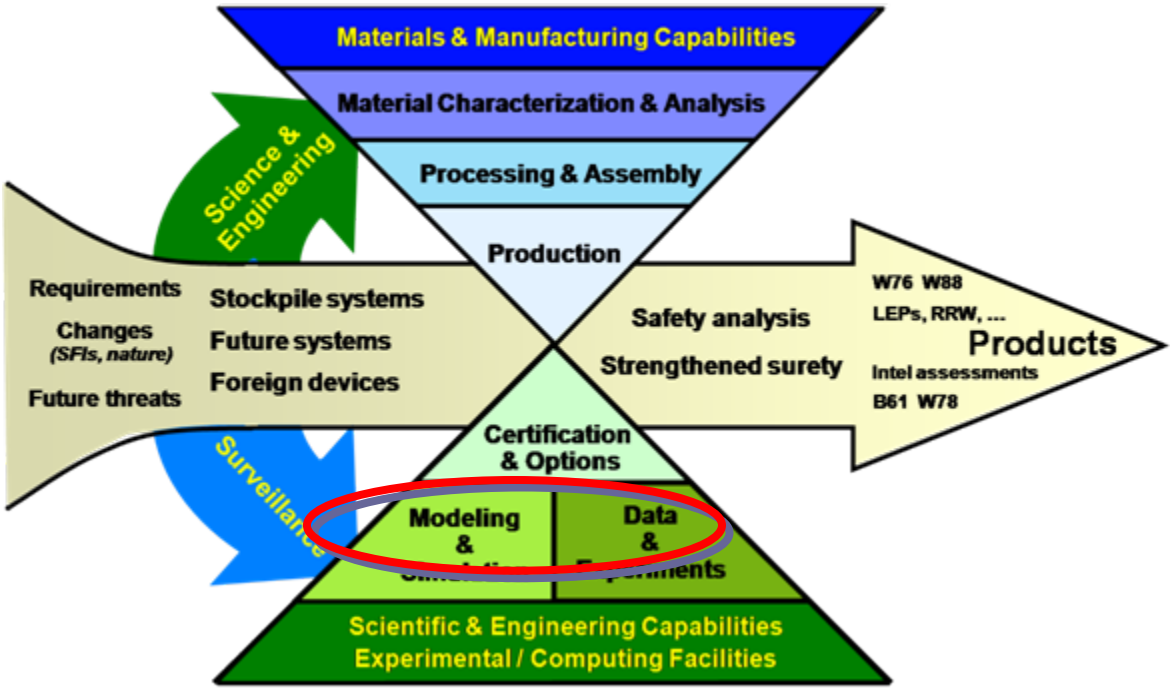
Inelastic potentials
- plasticity
- damage
- phase transformations
- twinning
- radiation

Extreme
$(\sigma, T, \dot{\varepsilon}, radiation)$

UNCLASSIFIED

Los Alamos NATIONAL LABORATORY EST. 1943
Operated by Los Alamos National Security, LLC for NNSA

Transition from "observation & validation" to "prediction & control" is a key mission challenge AND the frontier of materials research



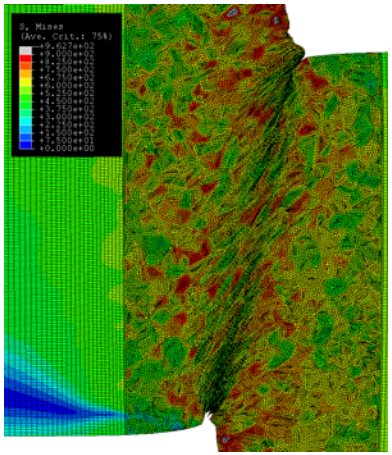
- **Majority of stockpile issues will likely continue to be materials based:**
  - Strength, damage: microstructure matters
  - Manufacturing processes matter: Cast/wrought, weld, alt. material
- **Future stockpile :**
  - Much national discussion regarding alternatives

ASC models are informed and validated by experimental Campaign data:
- **But only capture gross features** of material response (Maybe good enough now, but not adequate for assessing aging, and stockpile alternatives)
- **Not currently predictive** for most scenarios; calibration play a major role
- **Multi-scale material model development** Physics and Engineering Models (PEM) program in **ASC will need MaRIE data to inform and validate models**

Overview - Materials Models Used at LANL

Macro-Mech.

Numerous constitutive models

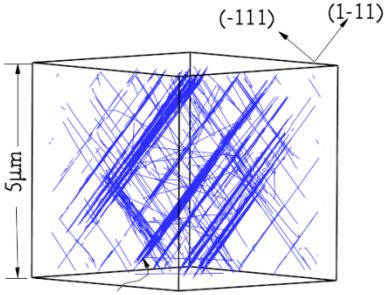


Polycrystal

Visco-plastic self consistent

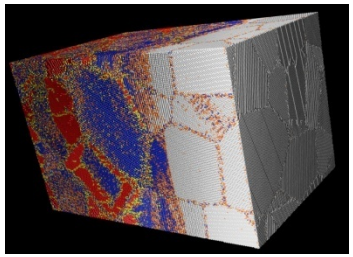
Single Crystal

Ginzburg-Landau Disl. Dyn. code



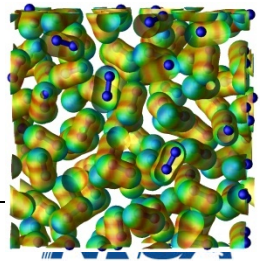
Molecular Dynamics

SPaSM
LAMMPS



Quantum Mechanics

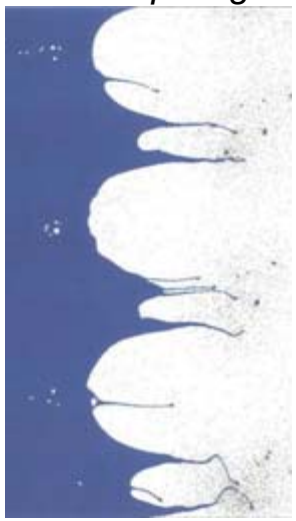
VASP
MondoSCF

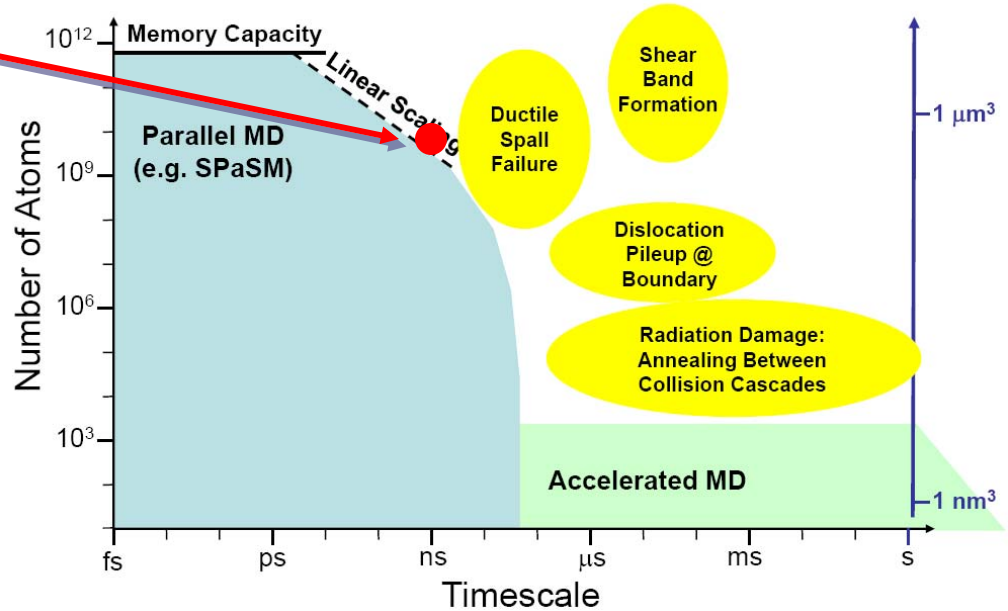


MaRIE data here

Present Los Alamos MD Simulations on Roadrunner - On the Verge of Accessing Phenomena at the Micron Scale

2D MD simulations of shock-induced ejecta and Richtmyer-Meshkov instability, showing bubbles and spike growth. (RR, 361 TF/s)





Future ejecta modeling challenges:

- 3D modeling of conical etching pits
- Polycrystal 3D ejecta (role of solid phase ejecta production)
- Use of more sophisticated potentials beyond EAM

Our MD simulations of ejecta mass and velocity distributions, together with experimental data, have helped develop an ejecta source model in our codes

Role of MaRIE Theory, Modeling and Computation (TMC) Board

- **Established this FY09**
  - Response to last year's EAB recommendation:

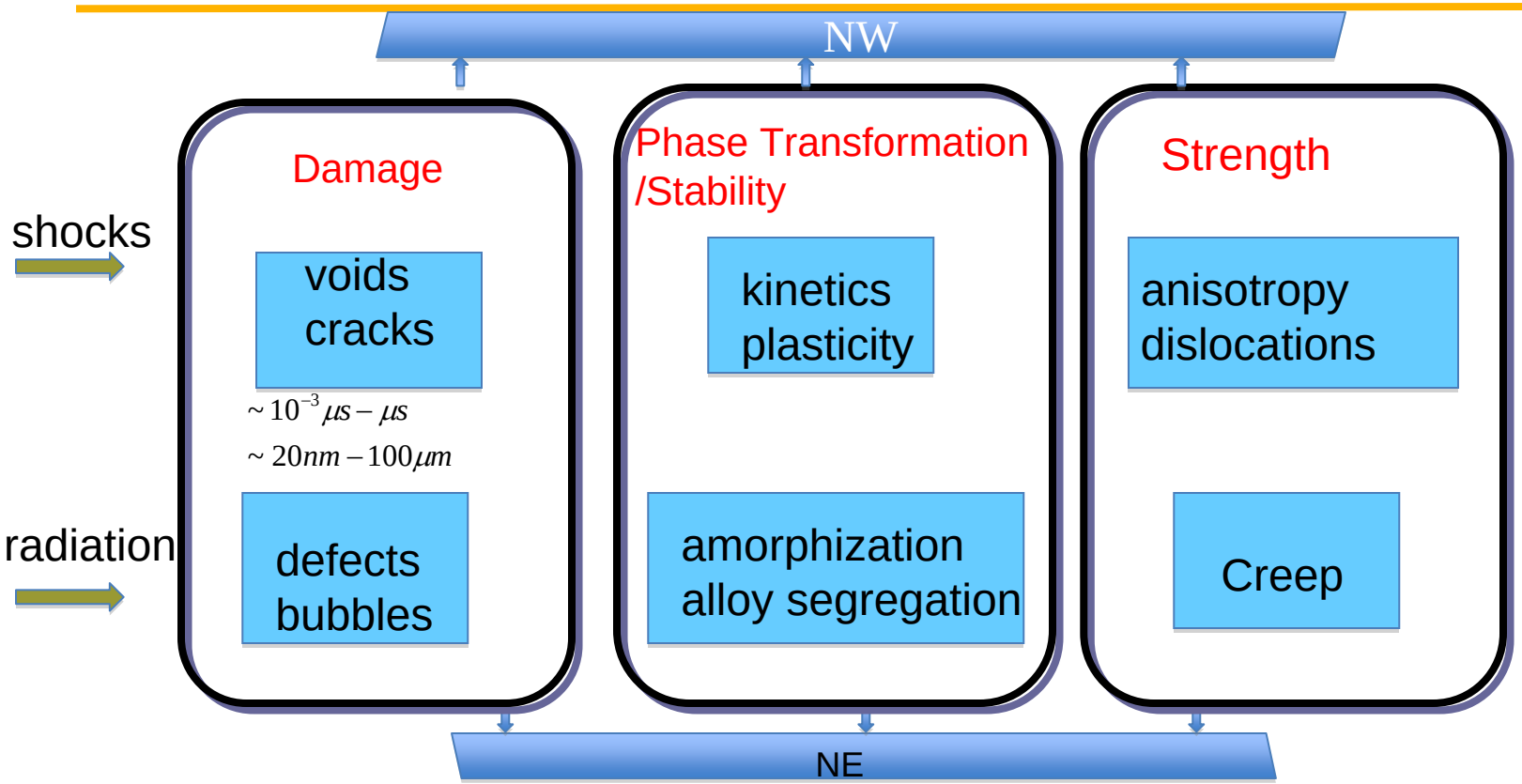
    "The Board strongly encourages a stronger emphasis on the connection between theoretical studies using LANL's high performance computing and the MaRIE experimental program. The overall challenges of understanding, creating, and exploiting materials over the full spectrum of length scales (i.e., going beyond the nanoscale to fill in the micron gap) under extreme conditions, is an important overarching concept"

- **Goals**
  - Articulate how potential MaRIE data can play a crucial role in **development** of, and **confidence** in, material models.
  - **Guide** experiments to validate theories and simulations.
  - Develop the theoretical and computational tools to **integrate** with experimental facilities.

- Comprised of **technical experts and leaders** in material theory & modeling/simulation, and IS&T
  - Engage leaders/researchers in ASC/PEM program and DoD munitions program, as well as advanced reactors (NEAMS), Office of Science/BES, actinide processing, catalysis research, Proctor and Gamble,…

F. Addessio, F. Alexander, M. Andrews, C. Bronkhorst, A. Balatsky, E. Dendy, E. Kober, T. Kelley, M. Stan, S. Valone, J. Wills, J. Wendelberger

TMC Tasks and integration with pillars



MPDH

F.Addessio
C. Bronkhorst*

FFF

J. Wills,S. Valone,
M. Stan*

- Current input to models ? How would they benefit from MaRIE data (define specs.)?
- Model developments needed to use this data ?
- What will they predict and how well (Uncertainty Quantification)

TMC Tasks and integration with pillars (contd)

Interfaces and nanoscale materials: $nm - \mu m, 10^{-12} s - 10^{-9} s$ to predict and control response.

Heterogeneities disorder

Photon to electronic energy conversion processes

→

Functionality: superconductivity,multiferroic charge and energy transport and transduction

Energy harvesting and radiation detection

M4

A. Balatsky
D.L. Smith*

Analysis

Inference, model selection
Characterization, imaging
Uncertainty Quantification

Resources
HPC Infra-structure

Storage
Communications
Processing

IS&T

F. Alexander, E. Kober*, ..

*also on other BOD's

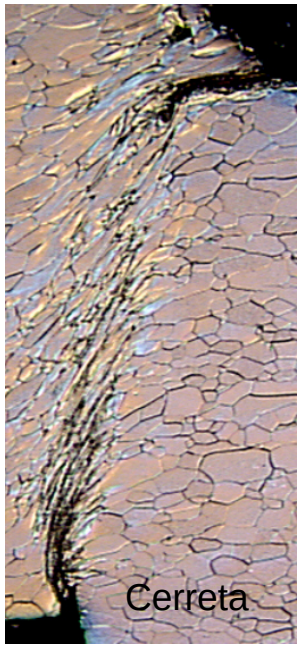
Example: Ductile damage leading to failure

Relevant materials challenges

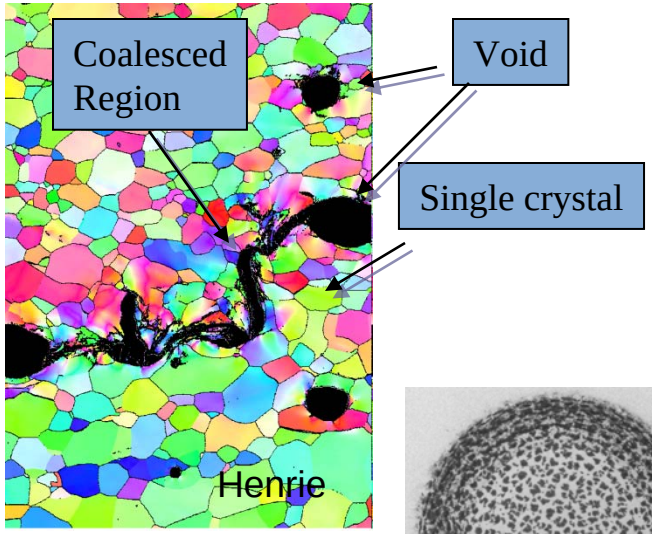
- Rate-dependent plasticity/anisotropy
- Damage: brittle / ductile / anisotropy
- Deformation twinning
- Phase transformations
- Re-crystallization
- Kinetics/ rate effects
- Radiation damage
- HE drive / high strain rate
- Shear banding

--nucleation, growth, coalescence, fracture

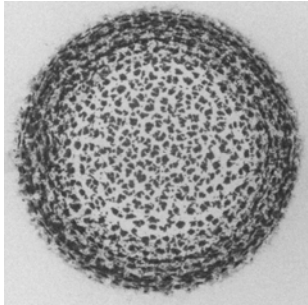
Shear localization



Porosity growth & failure



Ta



Physics-based models key to a predictive capability framework

Structural transformation and dislocations in shocked NiTi

Source: Thakur, A. M., Metall. Mater. Trans. A28:1445 (1997)

245 m/s

QuickTime™ and a
TIFF (Uncompressed) decompressor
are needed to see this picture.

needle twins

QuickTime™ and a
TIFF (Uncompressed) decompressor
are needed to see this picture.

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TIFF (Uncompressed) decompressor
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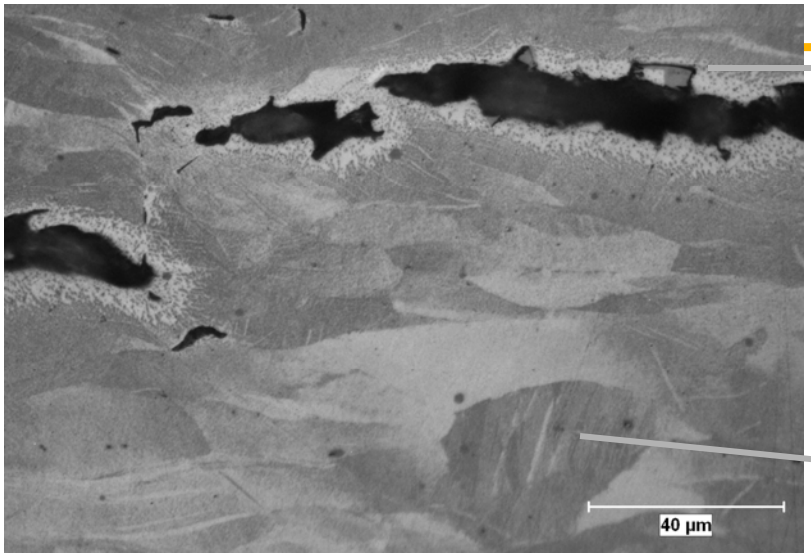
dislocations

- stress induced transformation:
  from cubic to monoclinic

Damage in DU: cleavage vs ductile dimples



- Cracking connected with inclusions
- Regions of plastic work interconnecting cracks
- Microstructure is heavily twinned
- ~50% of the cracks follow grain boundaries

D.Dennis-Koller et al.

Example: **Time evolution** of ductile damage
Q: How will MaRIE data help ?

•**Currently:** Void distributions obtained from recovered samples as input to models.
Not indicative of **nucleation, growth** or **coalescence** events

•**Need:** rate dependence of void number density $\frac{dn}{dt}$

growth rate of size of voids $\frac{dw}{dt}$

dependence on shear/volume stress

shape of voids and coalesced regions

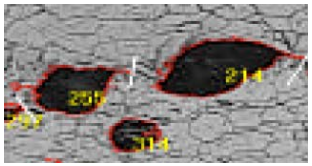
•**Model developments:** inertial effects $\frac{d^2n}{dt^2}$, $\frac{d^2w}{dt^2}$

spatial and time correlations between voids

statistical fluctuations of void density and size

Time/Spatial scales

$\leftarrow w \rightarrow$



Ta

$\sim 10^{-3}\,\mu s - \mu s$

$\sim 20nm - 100\mu m$

•**Impact:** **advanced predictive model for damage and failure at higher strain rates, & coupled to the microstructure of materials used in applications**

Presently data available before experiment & after recovery.
MaRIE will provide damage data at earlier & intermediate times

Example: Damage

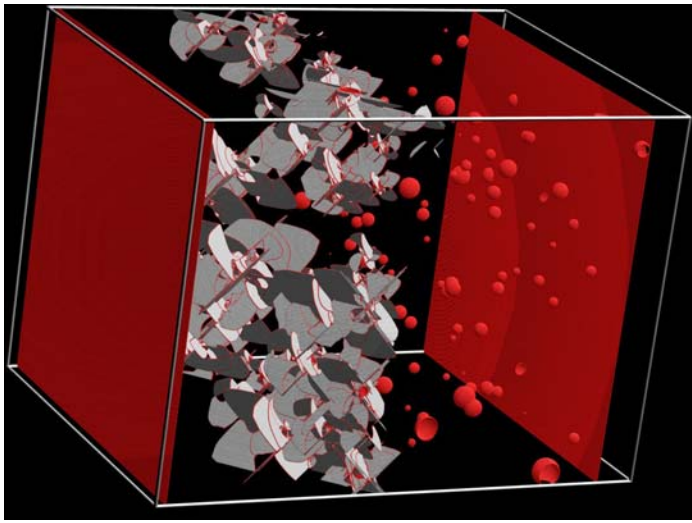
From Experiments:

- Elucidate (porosity and shear) mechanisms at early, late times (non-regular geometries lead to nearby voids to link in clusters/networks with enhanced growth)

- Data in nucleation regime will allow extension to large strain rates (by input of $n(\sigma)$ and inertial dynamics). Will improve transition regimes.

- Effects of heterogeneity: variations in void sizes due to differing nucleation rates

- Origin and measurement of plastic effects -- behavior of dislocations around voids and substructure evolution at high strains



T.Germann et al.

Theory/Modeling

- Nonequilibrium behavior: current models assume stress equil. Will lead to delayed fracture and applications to high strain rates

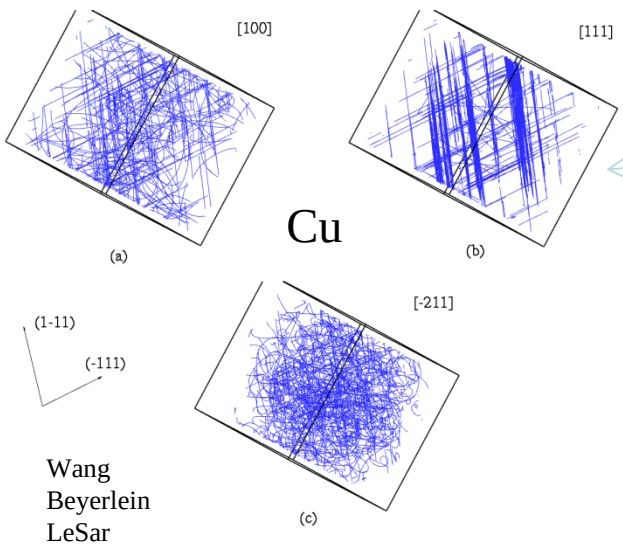
- Time dependent correlations --- currently ignored as nonlinear interactions of fields not amenable to solution.

- Statistical fluctuations; spread in fracture behavior
  -- current macro models deal with ave.behavior; spread in fracture behavior

- Microstructural models that predict nucleation, growth, .. & suggest approx. in analytical forms for input into macro. scale models

Plasticity at High Strain Rates

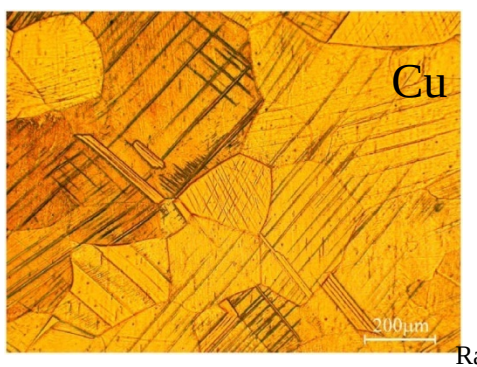
- Significant uncertainty and lack of data in plastic strength models at large deformation and high strain rates.
- Since high rate experiments difficult, Dislocation dynamics and MD motivate physics input to PTW model (no anisotropy, twinning, grain size distribution).



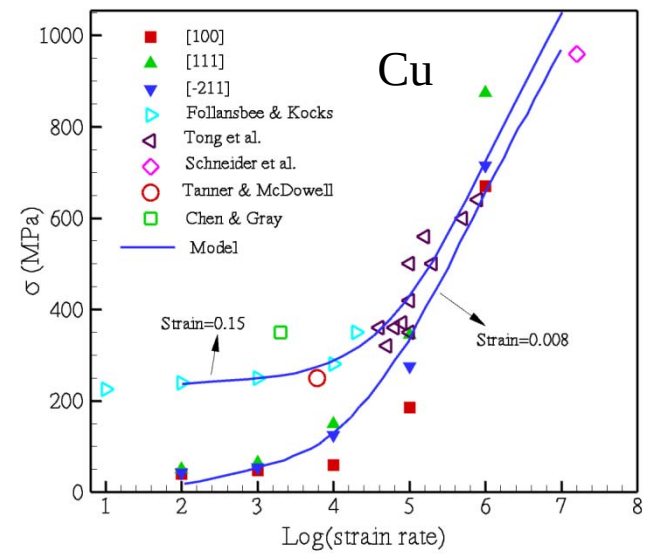
Wang
Beyerlein
LeSar

Dislocation behavior changes with crystal loading orientation

Slip banding



Slip band formation observed at high rate



Raevsky
Preston
Zocher

Small strain hardening similar to experiments

Where is the nonequilibrium science ?

- Thermal temperature does not determine statistical distributions, not sufficient to cause configurational rearrangements.

$$\frac{1}{T} = \frac{\partial S}{\partial E}$$

$$\frac{1}{\chi} = \frac{\partial S}{\partial V}$$

- Stirred or shaken powders (Mehta & Edwards)

  shearing, stirring causes the particles to ergodically explore configuration space

  entropic definition of temperature.

- Amorphous plasticity $T \longrightarrow T_{eff}$ J. Langer

- High strain dislocation plasticity ???? $\rho : \exp(-B / T_{eff})$

Modeling and Simulation for FFF: Structural phase stability and constituent diffusion

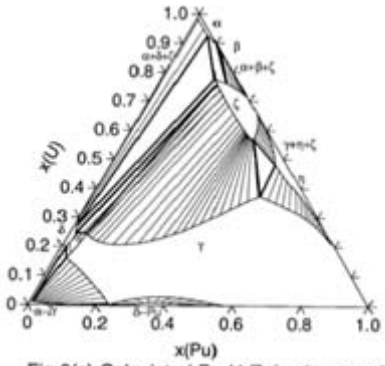
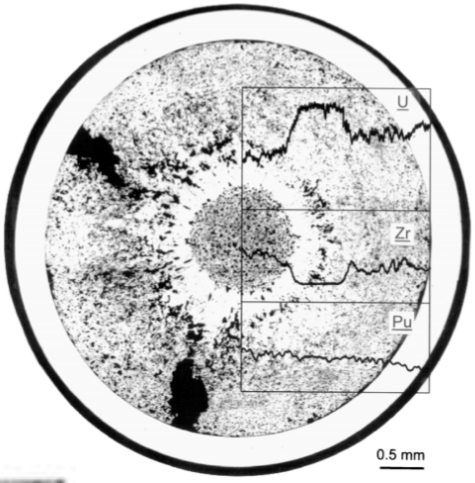


Fig.9(c) Calculated Pu-U-Zr isotherm at 868K.

•**Phase / composition** changes cause potentially critical changes in properties

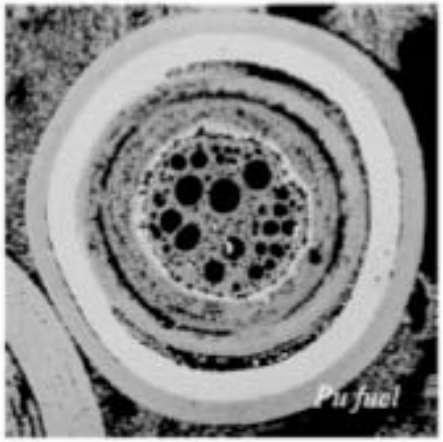
-- in-situ monitoring of phase composition change evolution



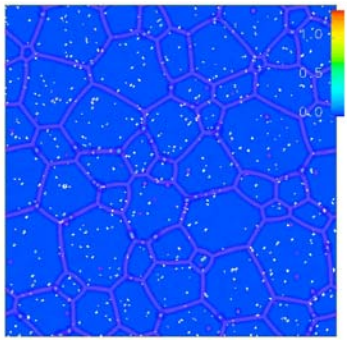
Constituent redistribution in a U-Pu-Zr fuel

• **Porosity and fission gas** diffusion, coalescence, and void swelling play a critical role in fuel lifetimes.

-- in-situ monitoring of coupled damage evolution / material burn up



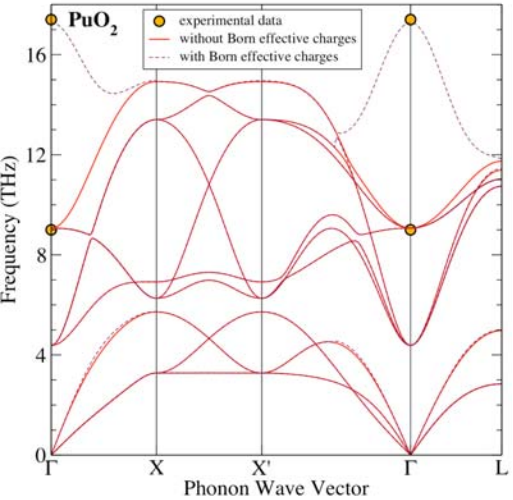
Swelling and damage in a Pu Triso fuel


Phase field (Hu et al., 2009)

- **Thermal transport**

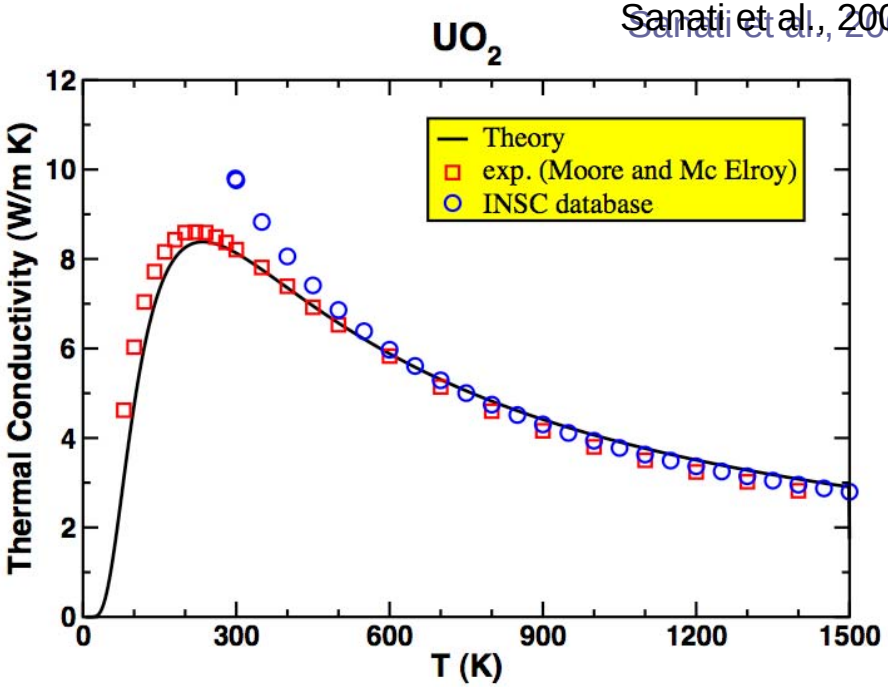
  Critical material properties for reactor design

  ---in-situ monitoring of thermal gradients and FP distribution under irradiation.



Calculated and experimental phonons in PuO(2)

Sanati et al., 2009

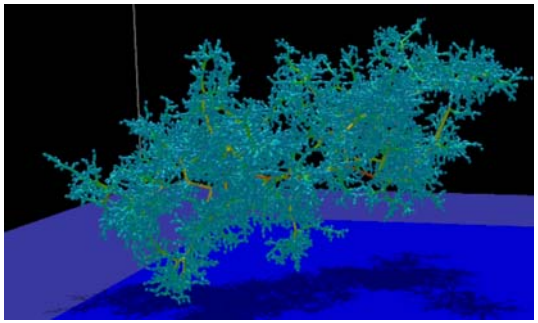


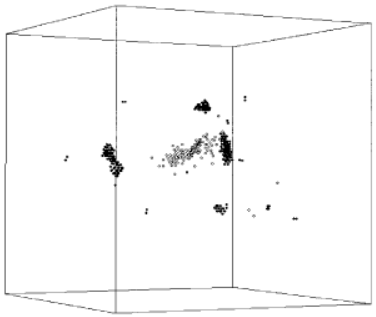
Need: prediction as a function of damage

Radiation Damage with Molecular Dynamics

S. Valone

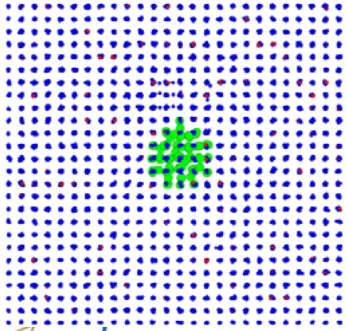
Metals and Alloys





High energy cascade in Pu metal w/Kubota and Wolfer/LLNL, Baskes

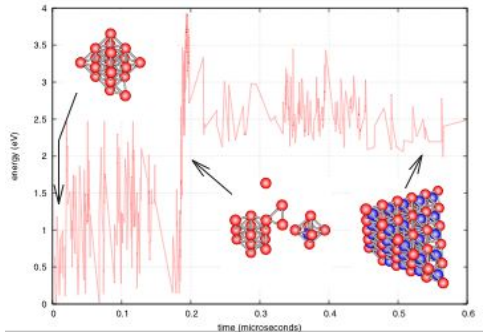
Predicts inversion vacancy and interstitials in spatial distribution



He bubble behavior in Pu metals and alloys

(Baskes, Martin)

Predicts T, metal/He, and alloy composition dependencies

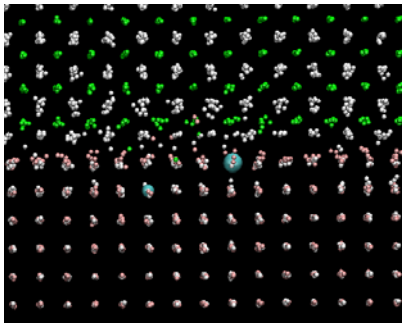


Void behavior in Pu metals

(w/Uberuaga, Hoagland, Voter)

Predicts novel mechanism of transformation to SFT

Ceramics



MD exploration of ceramic-ceramic interface mixing: Predicts kinetic trapping, substantial recover of interface at low dose

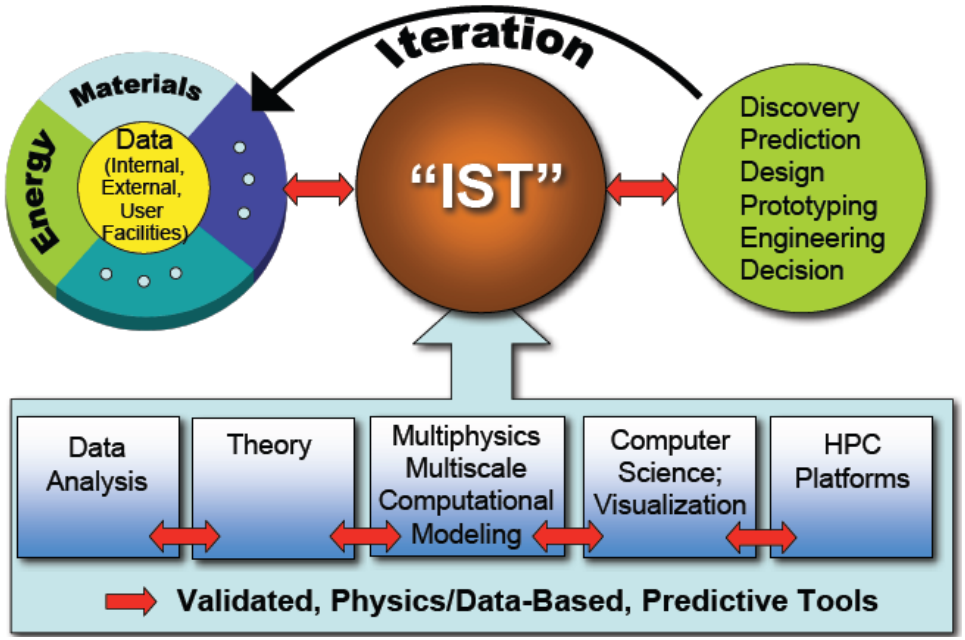
Theory and Modeling Challenges

- Electronic structure methods for extended disordered systems (defects, interfaces, clusters, heterogeneity).
- First principles methods for strongly correlated systems (actinides, frustrated materials).
- Far from equilibrium methods for high photon fluxes and phonon interactions and excited states.
- Vibrational methods for thermodynamics at high temperatures and pressures.
- First principles approaches to radiation damage: interaction of neutrons, ions with atoms.

- Accurate semi-empirical potentials: coupling electronic structure and MD
- Potentials with charge transfer
- Acceleration algorithms

- Kinetics of nucleation, growth, coalescence (of voids, bubbles) with spatial and temporal correlations and fluctuations -- theories of microstructure evolution
- Methods across scales: electronic structure and molecules to transport; atoms to continuum.

Information Science & Technology Capabilities for MaRIE



Multi-scale materials modeling: an *integrated* approach to modeling, simulation, and experimentation for multi-scale materials understanding

Materials Discovery: Methods for predicting, optimizing, and accelerating design and discovery of new materials

IS&T Analytical Tools: theory and application of methods for planning, visualizing, analyzing, combining, and understanding data; aging and lifetime prediction; reliability modeling and assessment

MaRIE Computing: Hardware/Software Architecture, Databases, Networks to meet the information storage and processing needs of MaRIE

IS&T: Feasibility studies (Frank Alexander)

- **Storage – Gary Grider**
  - What's needed to store data from various MaRIE experiments ?
  - What type of speed for analysis ?
- **Networking – Scott Pakin**
  - What is the optimal networking plan --- to the Science Complex ?
  - Movng data to the user community
- **Data Intensive Architectures – Adolfy Hoisie**
  - What architectures are needed for optimal analysis of massive and streaming data sets?
- **Collaborations – Jim Ahrens**
  - How can the MaRIE user community best collaborate--- efficient plan for visulaization, processing?
- **Visualization and Analysis – Jim Ahrens**
  - What resources are needed to visualize (dynamic) materials behavior and materials structure?

Topics (continued)

- **Inference, Learning Reduced Order Models – Frank Alexander**
  - How can we reduce the complexity of models required to predict materials behavior?
  - How can we predict materials behavior with confidence
- **Algorithms / Codes / Code Development – Andy White**
  - How will the new generation of codes (Molecular dynamics, Density functional) need to be structured to take advantage of likely heterogeneous architectures?
- **Computing Cycles – Josip Loncaric**
  - What processing power will be needed to carry out the above program?
- **Design of Experiments – Joanne Wendelberger**
  - How can we design experiments to yield maximum information?
  - How can we “make every photon count”?

Opportunities

- Seeing new signatures or features in complex or multi-dimensional data that stimulate further studies
- Comparative and quantitative scientific visualization
- Going from data tables or 1D plots to 2D or 3D plots can add new information that may get lost in stacks of tomographic 1D or 2D slices

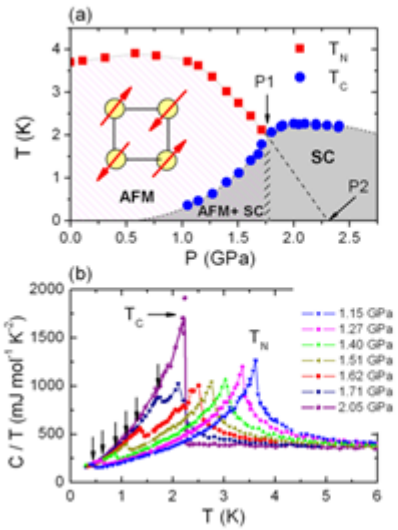
Challenges

- Data formatting, data conversion, data presentation
- Visualization feedback loop to experiment or theory
- Real-time visualization of experiments or simulations
- User-friendly visualization tools and technical support
- Distributed visualization available on desktop computer

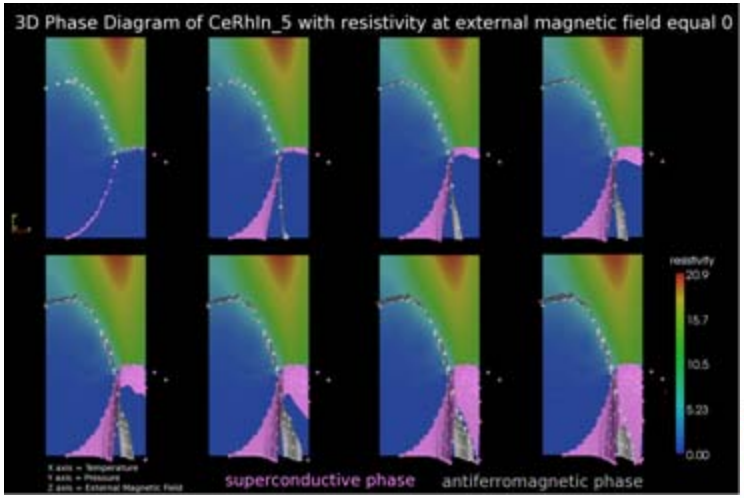
For more details on viz at CINT contact Sasha Balatsky (T-4/MPA-CINT) or Matthias Graf (T-

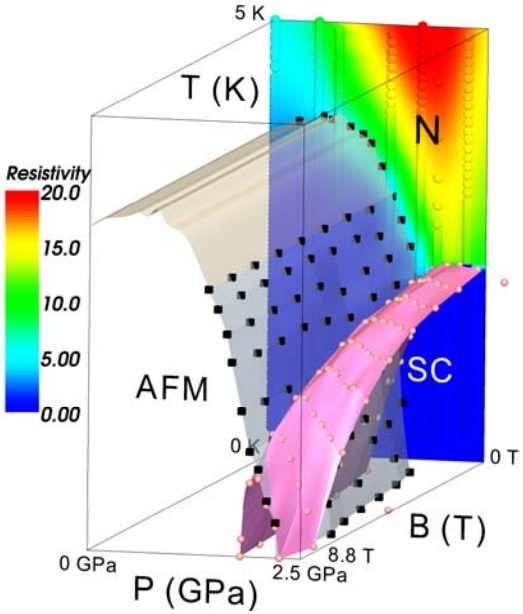
Bringing out the details: Phase diagram of $CeCoIn_5$

Usual presentation



Multi-dimensional comparative and overview presentation



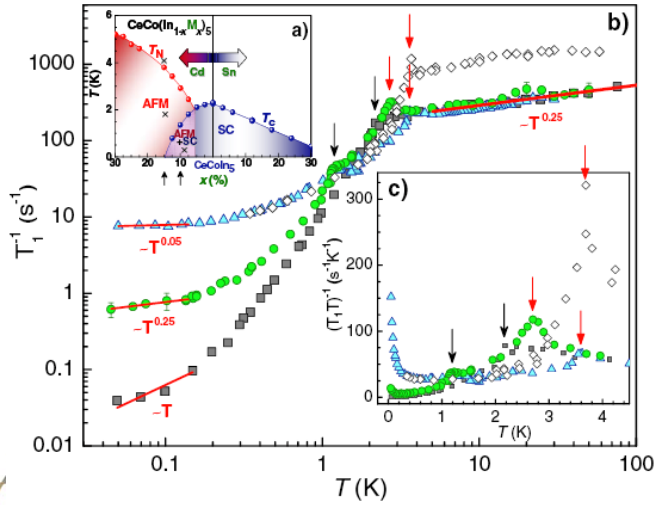


Advantages:
1. Comparative studies
2. Zooming in to study details
3. Zooming out for "big picture"

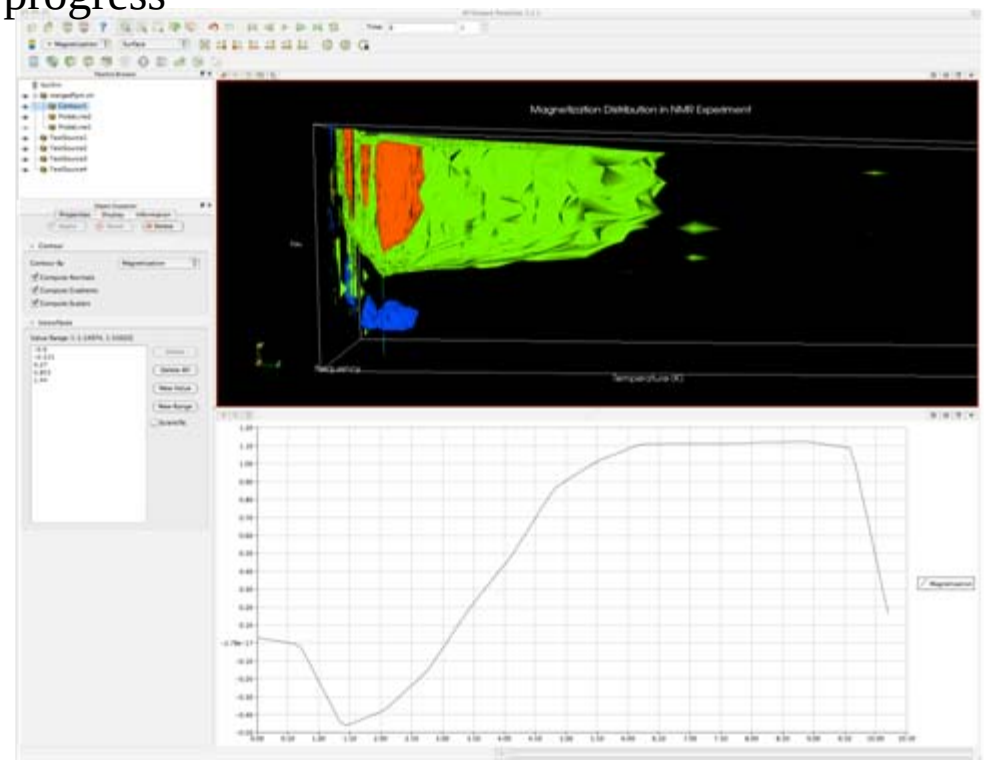
Challenges:
1. Working with sparse data sets
2. Slow turn-around with data collection

Analysis of NMR spectra of $CeCoIn_5$

Usual presentation of NMR spectra by integrating out two degrees of freedom to get spin-lattice relaxation rate $1/T_1$. Green symbols show 2 peaks at AFM and SC transition.

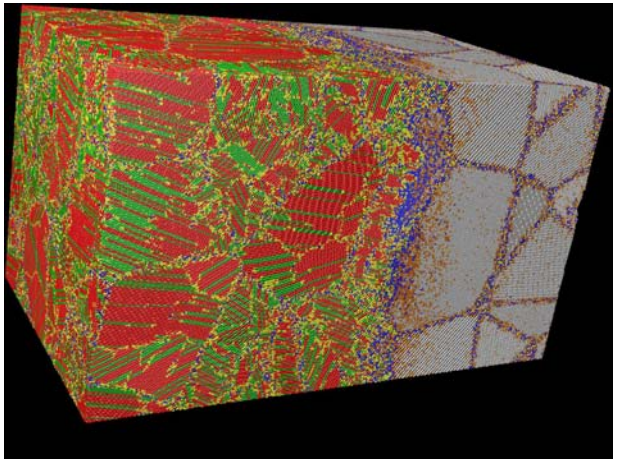


Spectral presentation of magnetization shows 3 signatures (red areas). Only 2 are expected for AFM and SC phase transitions … work in progress



Screenshot of viz tool ParaView

Quantifying Molecular Dynamics Studies



K. Kadau et al., *Phys. Rev. Lett.* (2006).

Shock-induced transformation in Fe predicted by large-scale molecular-dynamics simulations

Bcc(8nn, gray)->comp Bcc (10nn, blue)->hcp,fcc (12nn, red,green): simple algorithm

NEMD results confirmed experimentally by ultrafast (nanosecond) X-ray diffraction (shocks and high intensity X-rays produced by high energy laser systems OMEGA/Janus/Vulcan)

D.H. Kalantar et al., *PRL*. **95**, 075502 (2005)

Massive MD studies (SPaSM on Roadrunner): Trillions of atoms = several cubic microns

Tool to characterize sensitivity to polycrystalline morphology, realistic defect concentrations

However, massive data sets with limited storage capacity (1:1000 snapshots)

Identify “interesting” events in real time and save additional information in those areas: machine learning for anomaly detection and classification

More quantitative description of local geometry in terms of spherical harmonic expansions, enhanced classification of geometries

Group into classes of response behavior: phase transitions, slip planes, dislocation loops

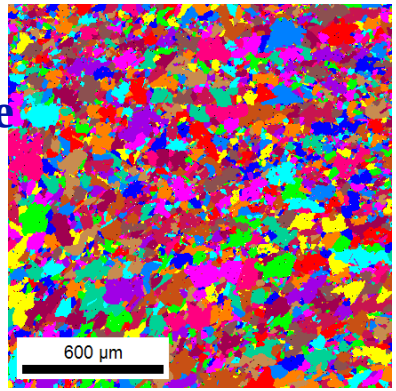
Dominant modes for reduced order modeling: basis vectors for higher level models (e.g. plastic flow). What are the associated thermodynamic properties?

Sensitivity of results to the form and parameters of chosen potential functions.

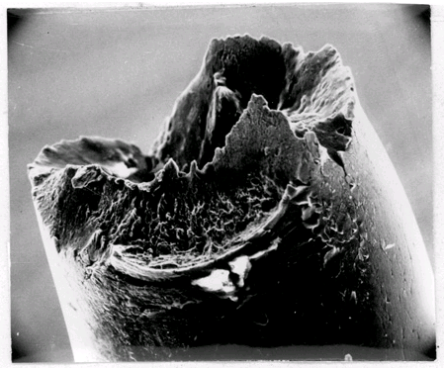
Comparison and iteration with experimental results: diffraction, OIM, Kober, Barber, Steinwart

Multiscale Imaging and Multiscale Design of materials: identify structure-property correlations across length scales

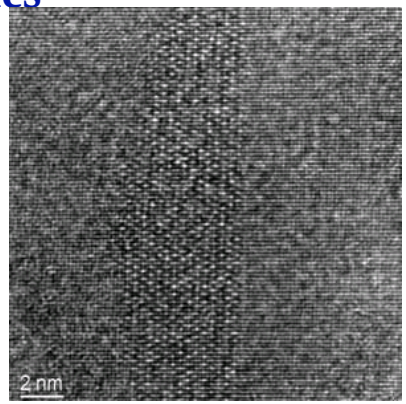
Prediction and classification of materials behavior under extreme radiation conditions through integration of massive imaging and spectroscopy data across length and time scales



SEM/OIM

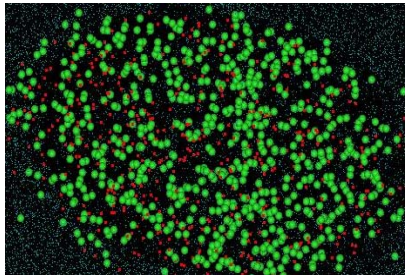


Failure analysis



TEM/ Diffraction

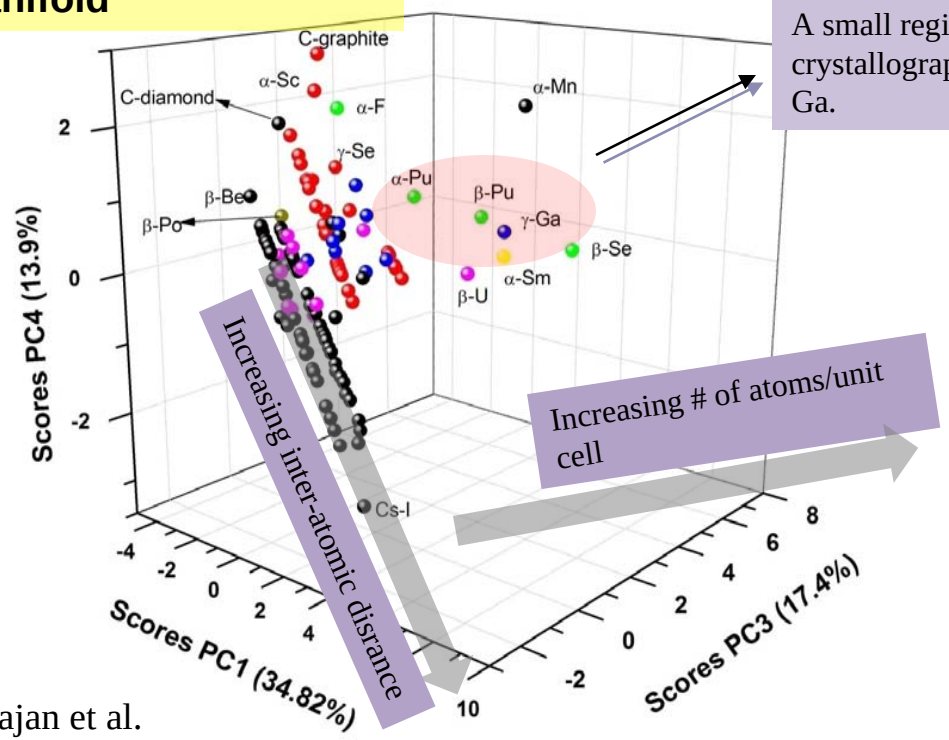
Atom probe tomography



Model based materials imaging informatics: Linking theory with experiment for structure-property via statistical learning based analysis of massive amounts of in-situ / ex-situ spectroscopic and imaging data

Informatics aided design of alloys

Model analysis done using crystallographic properties in a linear manifold



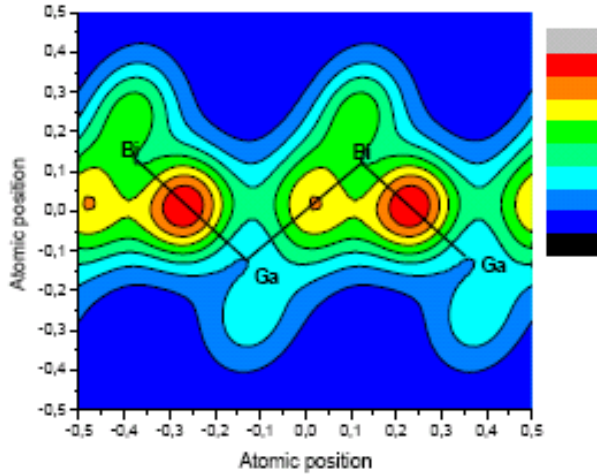
A small region in PC space indicating the crystallographic similarity between Pu and Ga.

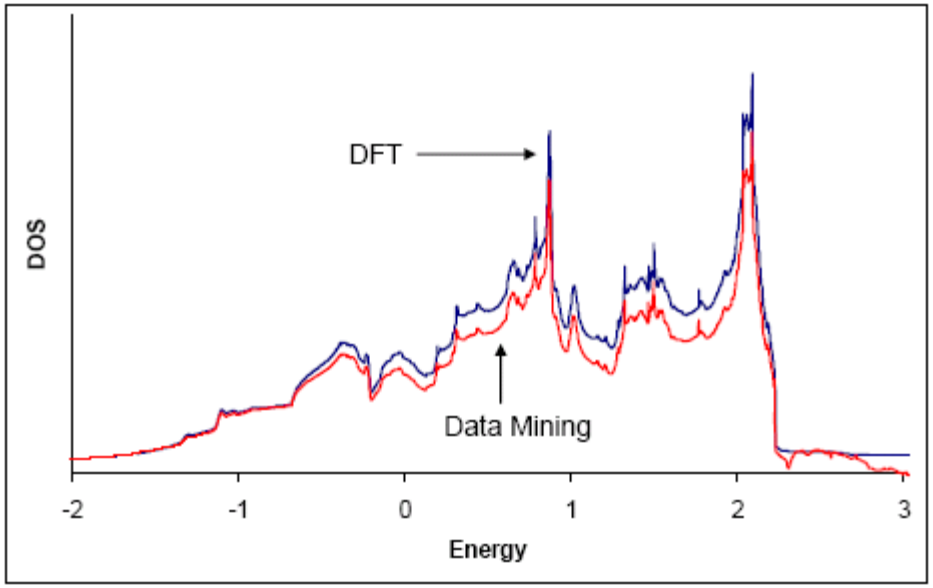
- Cubic
- Hexagonal
- Monoclinic
- Orthorhombic
- Tetragonal
- Trigonal
- Rhombohedral

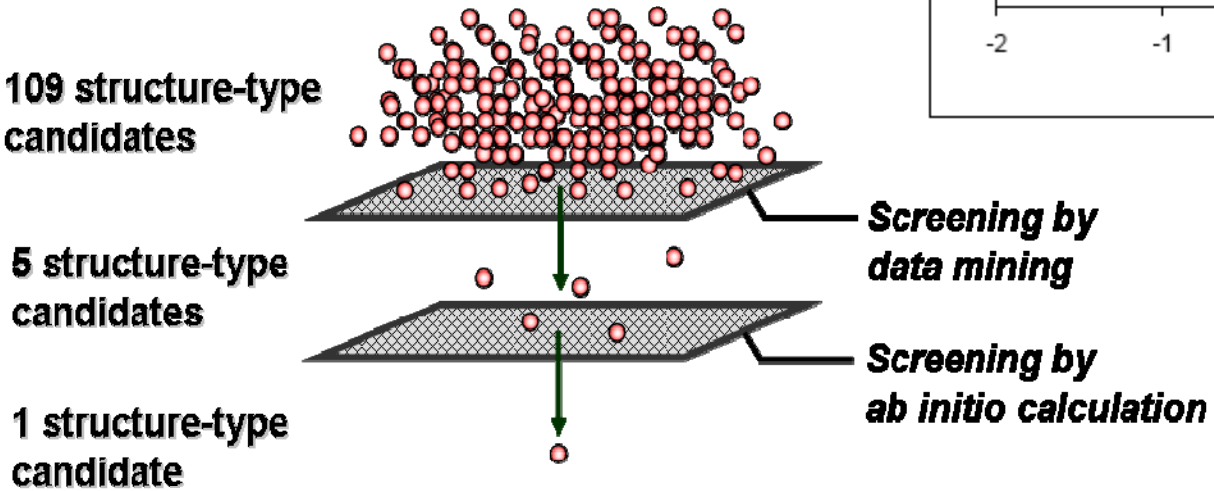
Further step includes, appending this data with physical, chemical and thermodynamic properties and modeling the behavior of complex Pu.

K. Rajan et al.

Model based materials informatics: Linking statistical mechanics with statistical learning







Conclusions:

Small-scale feasibility studies currently underway to galvanize and integrate theory, modeling, and simulation work with MaRIE

- Initial examples illustrated

- Later this year we will have further worked examples

At many length scales, LANL has developed theory & models for simulations on our HPC (petascale) platforms for material responses.

Advances are needed to address:

- What is it about the material structure, initial defect distributions, grain boundaries, *etc*, that impacts material performance?

- How can we advance material design based on this information?

- How can we optimize MaRIE experiment design and characterization tools that extract maximal amounts of information?