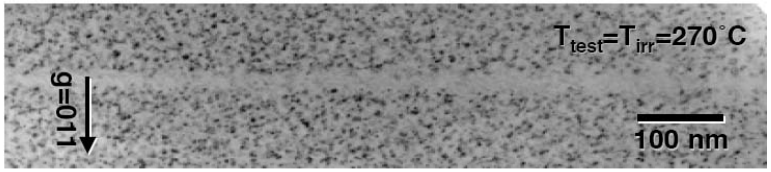
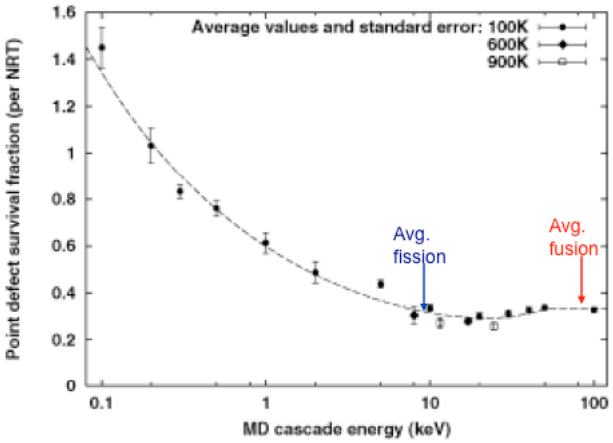
Panel Area:
Radiation Matter Interactions for Fission and Fusion

Radiation creates a unique environment that creates microstructures and subsequent property changes in a non-equilibrium manner

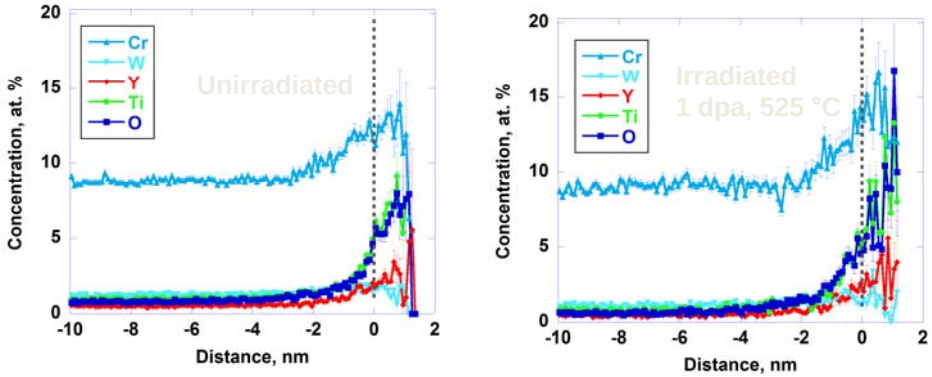
- Collision Cascades
- Radiation-induced segregation and associated phase changes
- Void Swelling
- Point defect-interface (sink) interactions
- Flow localization
- Corrosion & Crack propagation (IASCC)
- Embrittlement
- Degradation of electron and phonon transport



S. Zinkle



S. Zinkle



A. Certain and Mike Miller

Panel 1: Extreme Environment Tolerant Materials
Panel 2: Watching damage happen

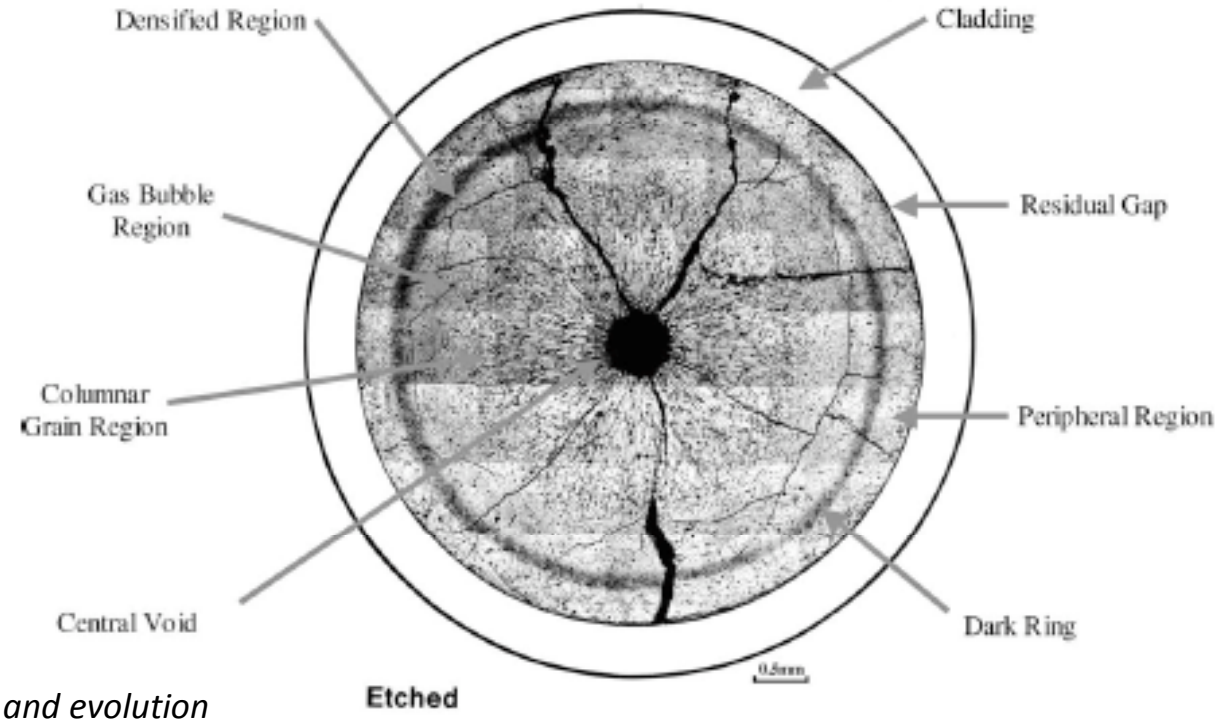
The need for higher performing materials in the extreme environments of advanced fission and fusion reactors is well documented. Approaches that accelerate the transition from the observation and validation of performance to the prediction and control of functionality through materials discovery and science-based certification remain grand challenges.

- Todd Allen, Chair
- Mike Burke
- Darryl Butt
- Rob Goldston
- Tom Holden
- Rick Holt
- Stu Maloy
- Amit Misra
- Christina Trautmann
- Gary Was
- Dieter Wolf

A particular challenge is the ability to make (and model/interpret) measurements of relevant phenomena in irradiation environments of interests and thereby advance fundamental understanding and predictability beyond today's 'cook and look' approaches.

- Steve Zinkle, Chair
- Don Brown
- Barney Doyle
- Scott Hsu
- Wayne King
- Mark Kirk
- Meimei Li
- Mike Nastasi
- Matteo Rini
- Yanwen Zhang

Example: Nuclear fuel-where everything changes



Microstructure formation and evolution

- *void swelling*
- *fission-gas bubble formation & release*
- *phase behavior , grain growth, precipitation*
- *alloy species redistribution*

- *Effects on thermo-mechanical behavior*
  - *thermal conductivity*
  - *crack nucleation, diffusion creep, etc.*
  - *fuel-cladding interaction*

Priority Research Directions

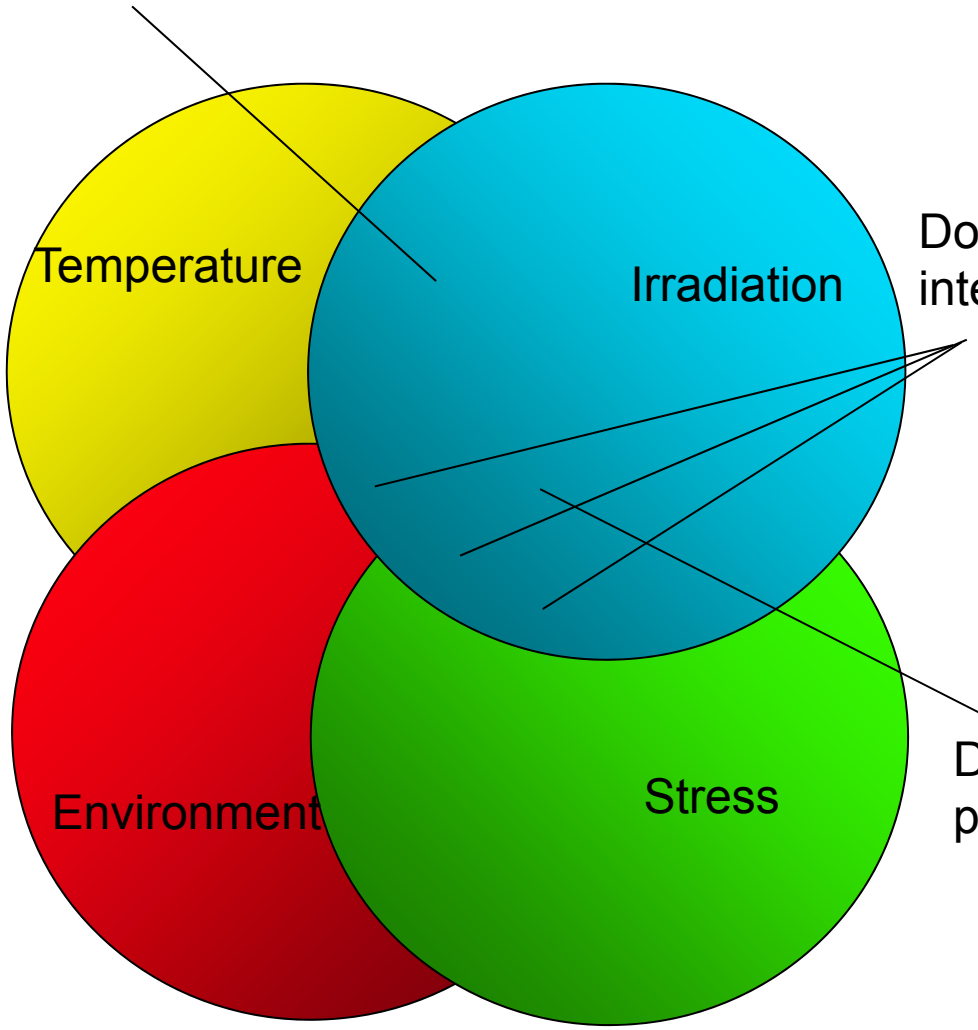
Thermo-Mechanical Properties and Dimensional Stability during Irradiation

Corrosion science under extreme environments

Microstructure science under irradiation

Understanding unit processes and their collective behavior

Traditional radiation effects research



Domain of corrosion-radiation interaction

Domain of mechanical properties

PRD 7: Thermo-Mechanical Properties and Dimensional Stability during Irradiation

Scientific Challenge

Irradiation and temperature change the active deformation systems (slip, twinning, climb) and cause dimensional changes including creep and swelling and loss of fracture resistance. Understanding these changes will allow prediction of the response of the material under static or dynamic loading. Both average properties and inhomogeniety are important, especially for crack initiation.

Research Direction

Long term irradiations are required to develop models for the influence of irradiation on deformation mechanisms, fracture and fatigue. Characterization is required using small angle scattering, stain mapping, in situ mechanical testing, metrology, electron microscopy, atom probe, tomography.

Capability Gap

- Facilities to expose materials up to 100-400 dpa for full-scale mechanical testing.
- Synchrotron and neutron diffraction on highly radioactive materials: easy specimen preparation and transfer.
- Quantified microscale testing and simultaneous ion beam irradiation in electron microscopes.
- Scaling from nano- to macro-scopic properties
- Models to predict the full range of mechanical behavior needed to design and operate reactors over all reactor operating conditions.

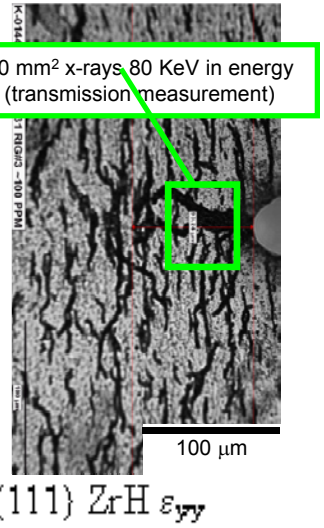
Potential Impact

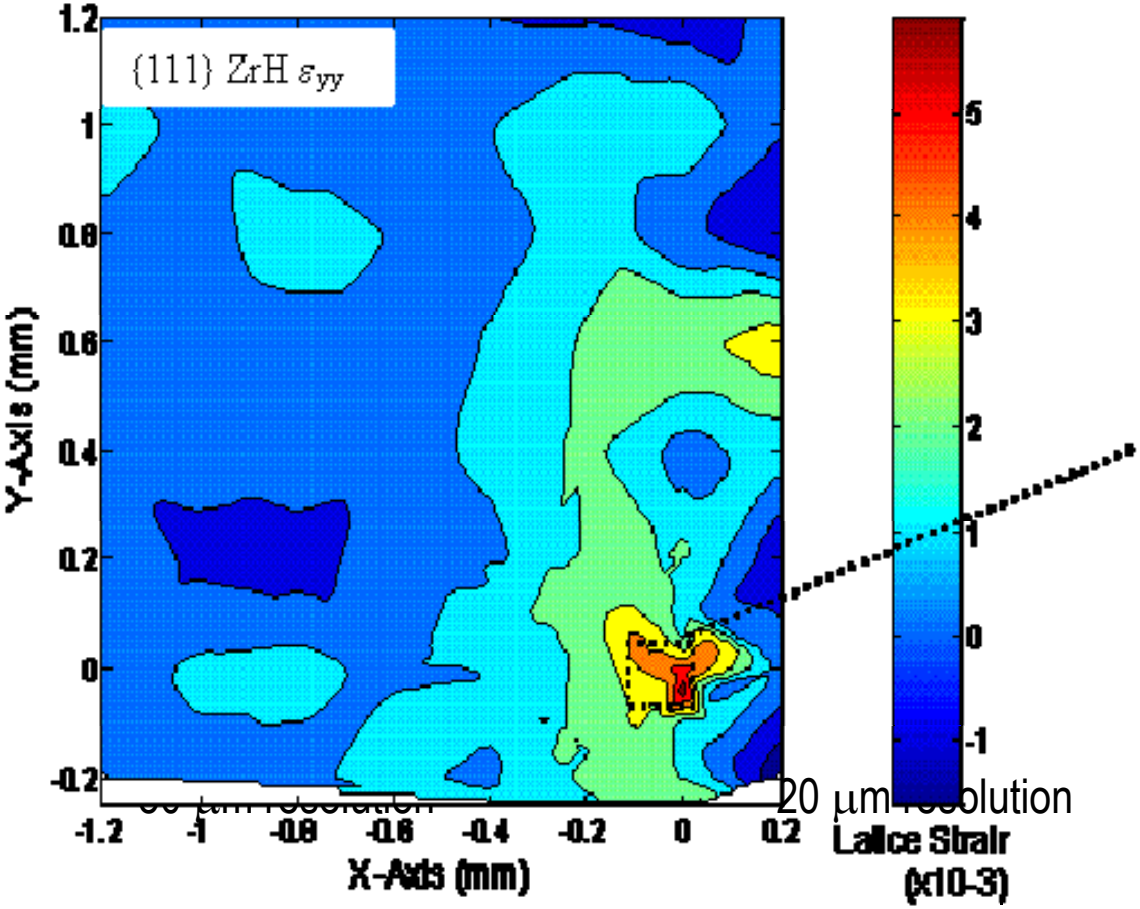
Substantial improvement in understanding and capability of modeling mechanical behavior of materials under irradiation will lead to:

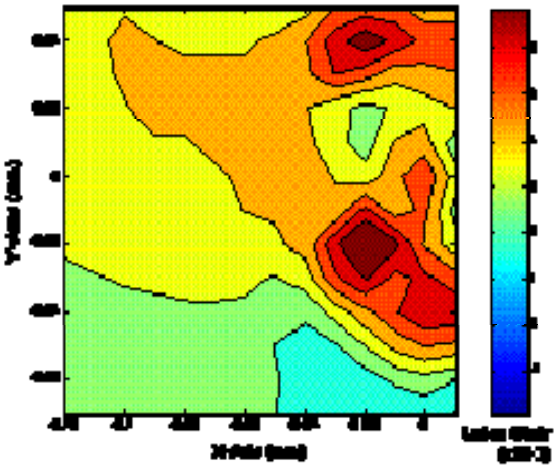
- Understand life limiting defects in nuclear plant, test new materials for engineering suitability in new plants or new kinds of plants (say, fusion reactor)
- life extension of existing nuclear plants and potential savings of billions of dollars in rebuild costs.

Hydrides and DHC in Zr-2.5Nb

Loaded to 8 MPa√m – lattice strain in hydride





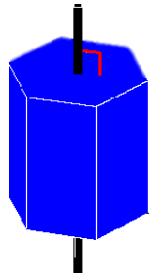


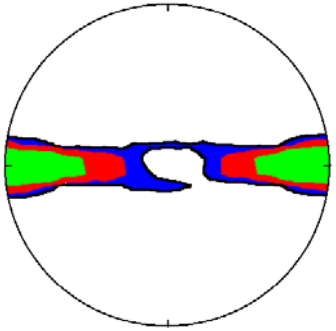
Introduce dislocation distribution into self-consistent polycrystalline models

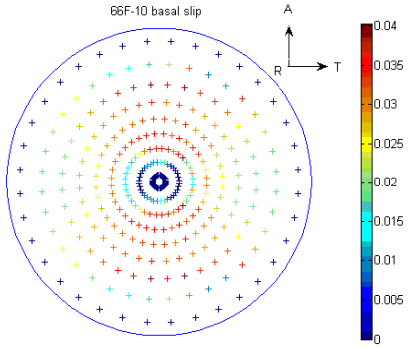
- **Single crystal properties**
- **+**
- **Crystallographic texture (ODF)**
- **+**
- **Dislocation distributions**
- **from manufacturing**
- **+**
- **Crystal interactions w/matrix**

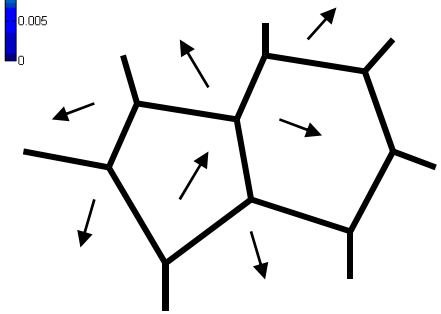
⬇

- **Macroscopic properties**

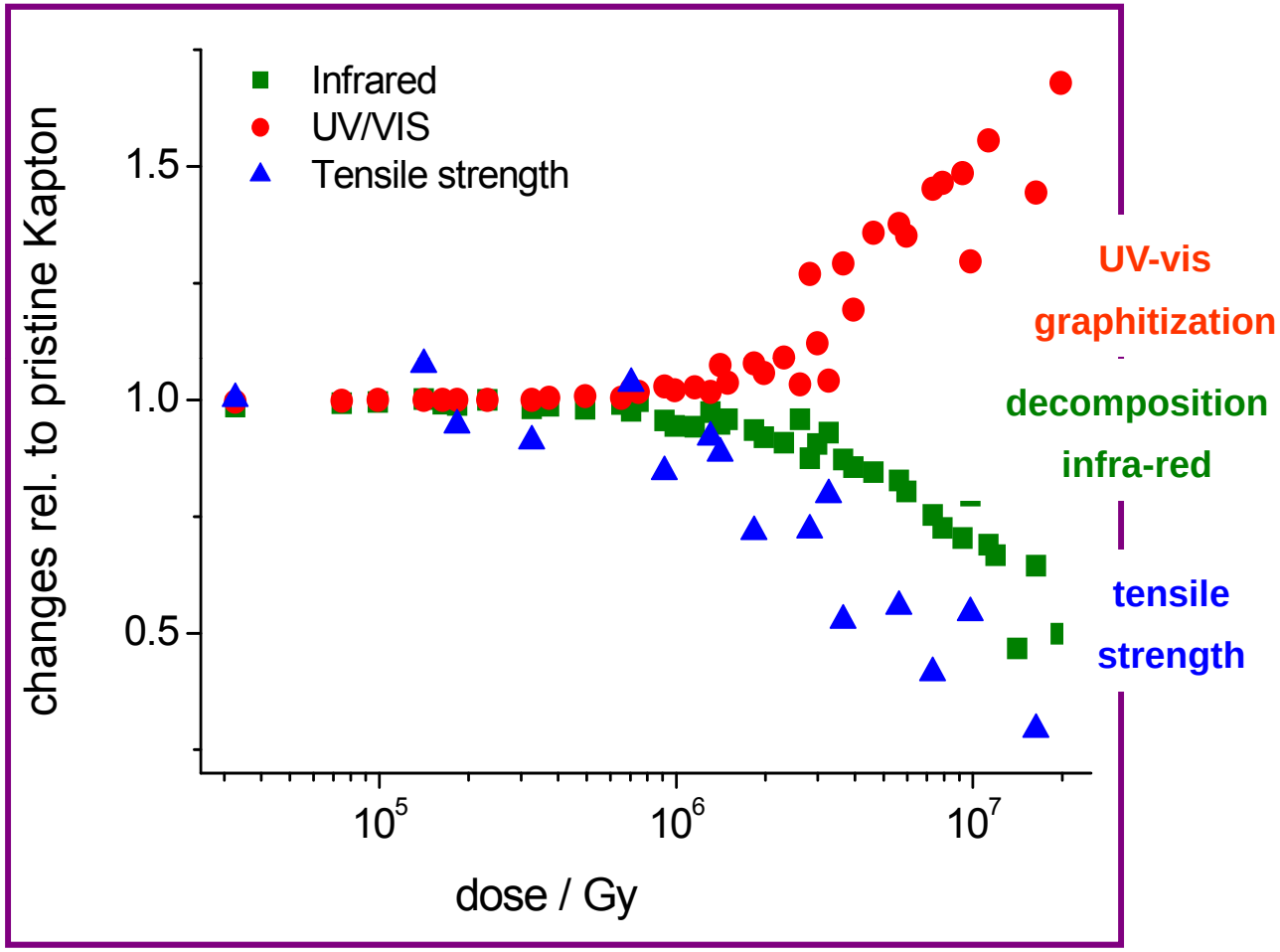








Irradiated Kapton



PRD 12.5: *Materials Design for Resistance to Corrosion and Surface Damage under Irradiation and Extreme Environments*

Scientific Challenge

Understanding and predicting how irradiation affects the material-environment interfaces – including chemical reactions, charged species transport, and plasma-surface interactions.

Measuring and characterizing material-environment reactions under extreme conditions including T, P, radiation.

Design of materials and microstructure with enhanced resistance to corrosion and surface damage in extreme environments

Capability Gap

Capabilities are needed to manipulate environments and *simultaneously* observe, accelerate, control and mitigate kinetics, phase transformations and mechanisms.

Development of experimental stations for in situ measurements in extreme environments.

A range of irradiation including high flux sources (neutrons, ions, gammas, plasmas) and diagnostic tools will be required.

Requires significant computational modeling tools to *simultaneously* treat multiple processes in multiple phases.

Research Direction

Observe and measure modifications in materials structure and chemical reactions at interfaces under irradiation and extreme environments, including both effects on the material and its environment.

Developing more fundamental models for corrosion, erosion/redepositior and surface gas accumulation under irradiation and in extreme environments.

- Point defect and chemical transport
- Chemical reactions and phase equilibria
- Stress fields
- Electrical transport
- Localized and uniform processes

Validate models using measurements of reaction modes and kinetics.

Potential Impact

Ability to treat corrosion as an integral part of materials design (physics based models replace empirical and data based predictive approaches)

Provides predictive models to support current plant life extension (employing existing materials), and guidance for the development of new materials for advanced fission and fusion reactor concepts.

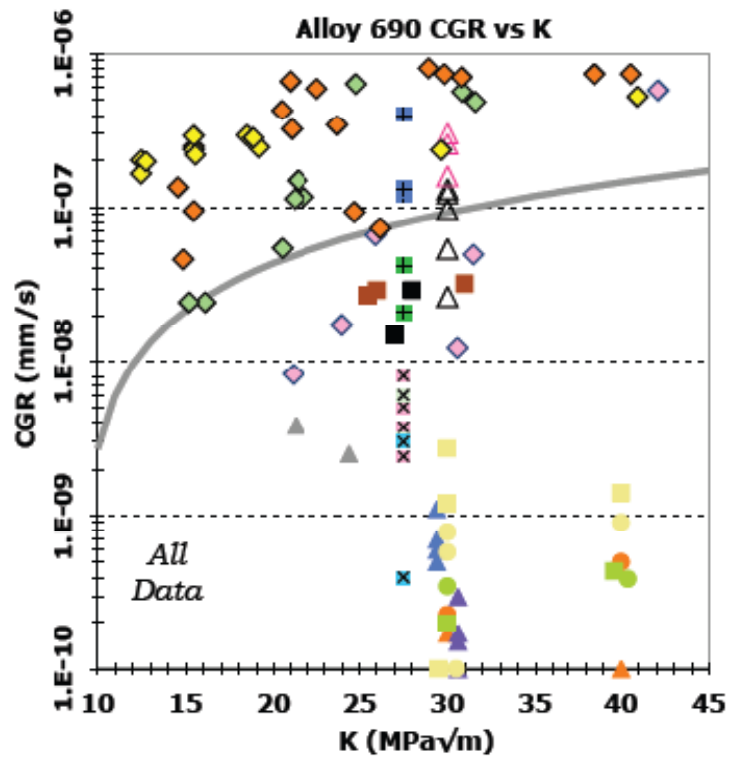
Provides predictive models for stockpile stewardship.

Improved energy storage materials, gas separation membranes.

Reduce infrastructure costs and broaden the operating window for materials applications in extreme environments

Variability of Materials Properties Employed in Plant Life Analyses

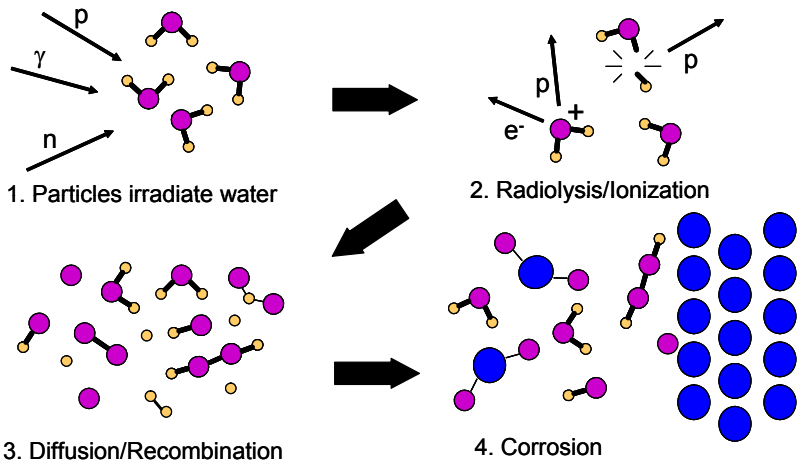
- Example : Alloy 690 Crack Propagation data



Summary of Recent Processing Affected Alloy 690 Crack Propagation Data

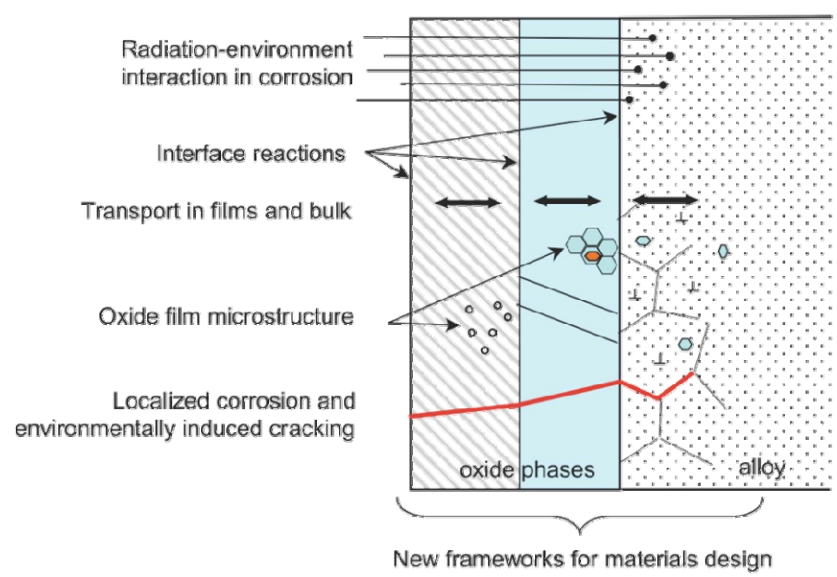
PRD2: *Materials Design for Resistance to Corrosion and Surface Damage under Irradiation and Extreme Environments*

Radiolysis at a Corrosion Interface

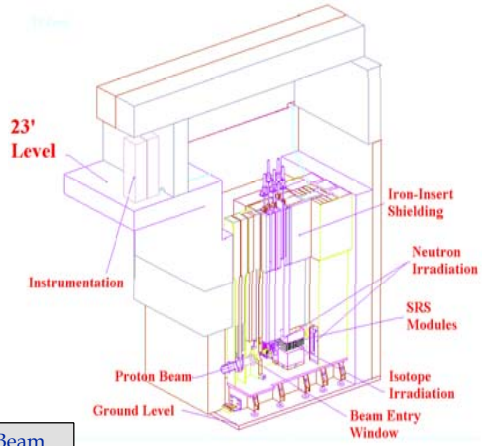


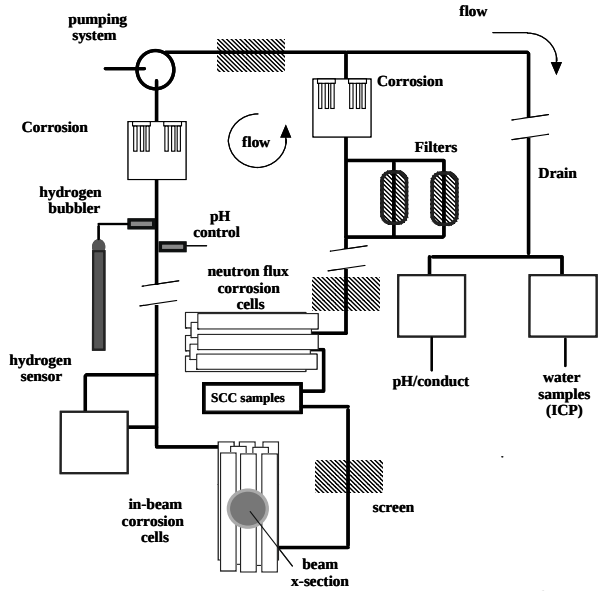
$$e^- + e^- \rightarrow \mathbf{H_2} + 2\,OH^-$$
$$e^-_{aq} + H^+_{aq} \rightarrow H$$
$$e^-_{aq} + H \rightarrow \mathbf{H_2} + OH^-$$
$$e^-_{aq} + OH \rightarrow OH^-$$
$$e^-_{aq} + \mathbf{H_2O_2} \rightarrow OH^- + OH$$
$$H + H \rightarrow \mathbf{H_2}$$
$$OH + H \rightarrow \mathbf{H_2O}$$
$$H + \mathbf{H_2O_2} \rightarrow \mathbf{H_2O} + OH$$
$$OH + OH \rightarrow \mathbf{H_2O_2}$$
$$H_3O^+ + OH^- \rightarrow \mathbf{H_2O}$$

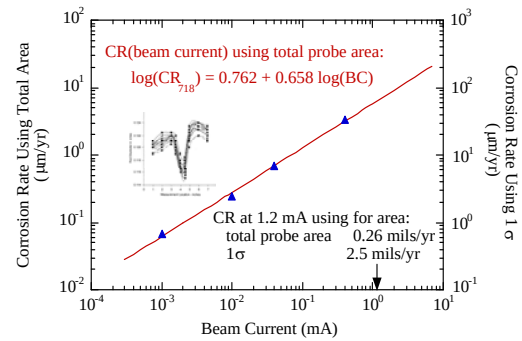
Irradiation-accelerated chemical reactions in materials

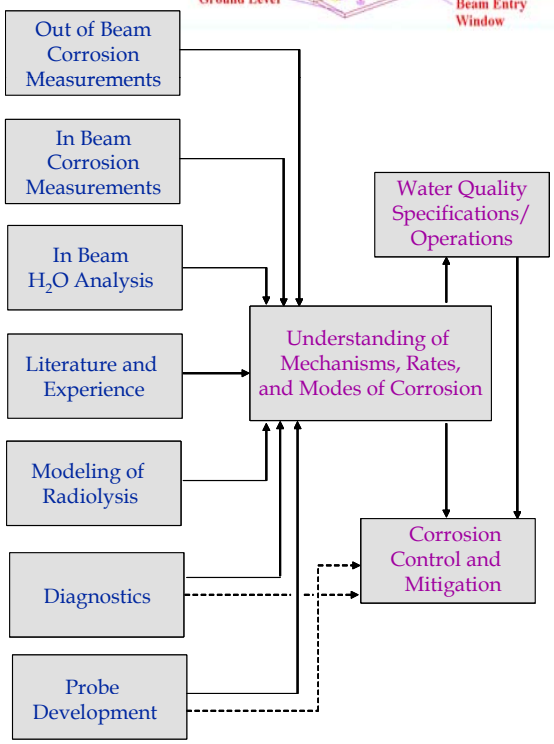


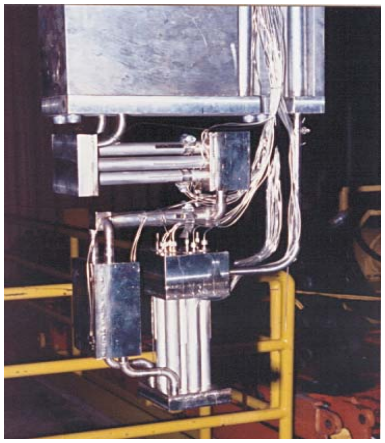
LAMPF Demonstration

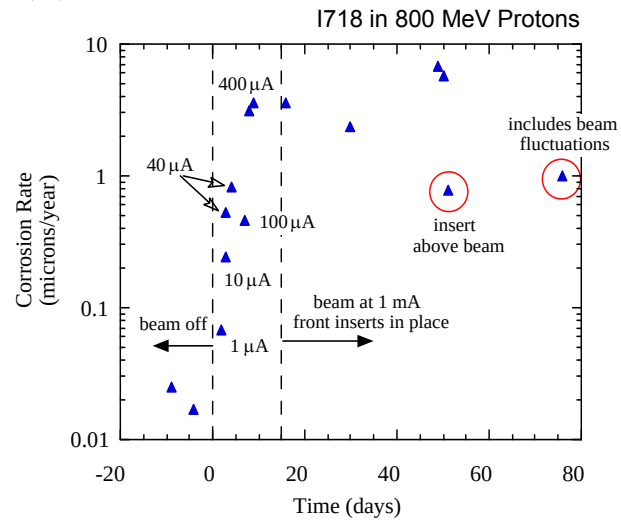


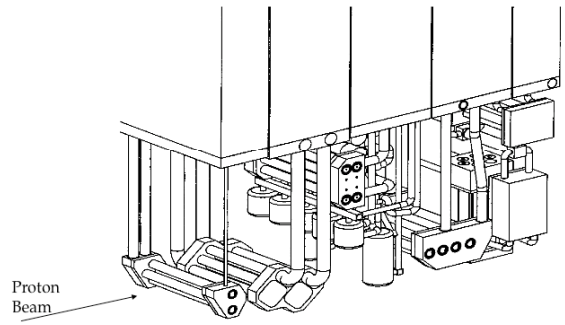












R. S. Lillard and D. P. Butt, "A Method for Measuring Corrosion Rates of Materials in Spallation Neutron Source Target/Blanket Cooling Loops," *Materials Characterization*, **43** [2/3] 135-145 (1999).

R. S. Lillard, G. J. Willcutt, D. L. Pile, and D. P. Butt, "The Corrosion of Alloy 718 During 800 MeV Proton Irradiation," *J. Nucl. Mater.*, **277** [2/3] 240-262 (2000).

R. S. Lillard, D. L. Pile, and D. P. Butt, "The Corrosion of Materials in Water Irradiated by 800 MeV Protons," *J. Nucl. Mater.*, **278** [3/4] 277-289 (2000).

PRD π: *Microstructure Science under Irradiation*

Scientific Challenge

Microstructure is the critical link between material science and materials performance

Developing an experimentally validated, multi-scale modeling approach for microstructure evolution under irradiation (void-, fission-gas and phase behavior, stress development, ...) to predict how these affect, e.g., the thermo-mechanical and thermal-chemical behavior. Incorporation of microstructural processes based on atomic-level mechanisms is critical towards developing a predictive fuels- and materials-performance capability.

Research Direction

Unified meso-scale approach that combines phase field with heat and chemical species transport theory to predict *concurrent* formation and evolution of microstructure under irradiation, high-temperature, environment and stress

- Microstructurally-controlled properties output to engineering-scale performance code
- Bulk and interfacial mechanisms and materials parameters input from lower-length scale (experiment or model, both unit mechanisms and collective effects)

Experimental validation of microstructural predictions using in-situ tools such as ion beams, electrons, x-ray, and neutron scattering

Capability Gap

- Experimental techniques, both in-situ (ion and neutron) and ex-situ, to characterize evolving microstructures in irradiated materials with sufficient statistical detail including unit mechanisms
- Develop foundations for understanding ion beam (light or heavy) correspondence to neutron irradiation effects
- Understanding of transport and defect properties, and phase behavior in complex, heterogeneous materials
- Robust scale-bridging algorithms
- Figures of merit that describe critical properties driving microstructural changes under irradiation

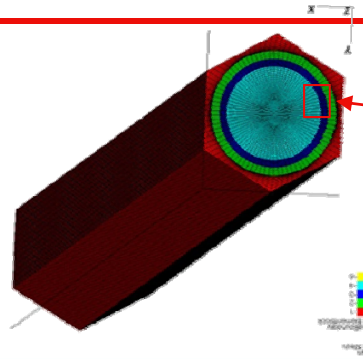
Potential Impact

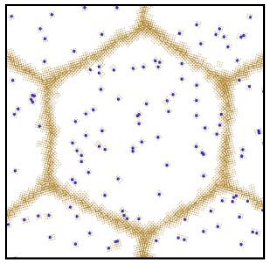
Microstructural processes are critical unknowns in the development of a predictive modeling capability for material performance

Explicit incorporation of microstructural processes based on lower-length mechanisms enables a *predictive* fuels- and materials-performance capability

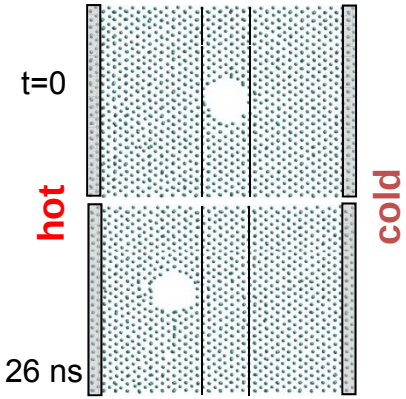
Ability to design material (composition, microstructure, phases, and interfaces) that optimize engineering performance

Hierarchical multi-scale simulation of nuclear fuel

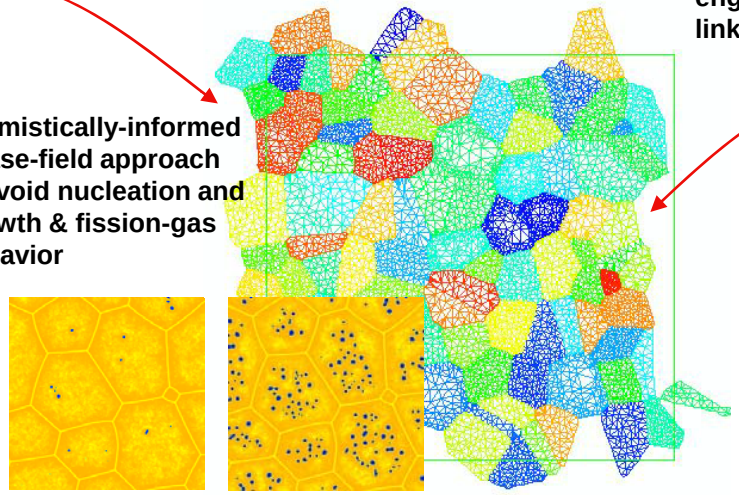




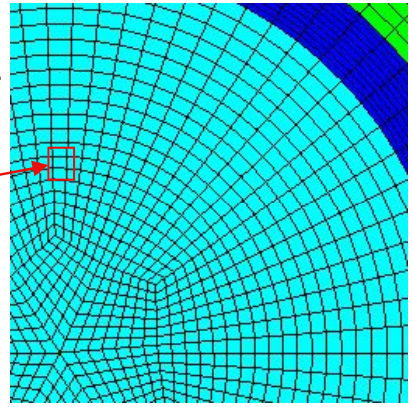
Molecular-dynamics simulation of radiation damage



Atomistically-informed phase-field approach for void nucleation and growth & fission-gas behavior



Materials science - engineering scale linkage



Atomic/electronic level
(Newton's laws)

Radiation damage, microstructural mechanisms and materials parameters

'Mesoscale'
(viscous force laws)

Effect of microstructural processes (fission-gas, voids, cracks, diffusion, …) on thermo-mechanical properties

Continuum level

Continuum mechanics, PDEs, constitutive laws

Explicit incorporation of microstructural processes based on atomic-level mechanisms is critical towards establishing a predictive fuels-performance capability

Materials Heterogeneity

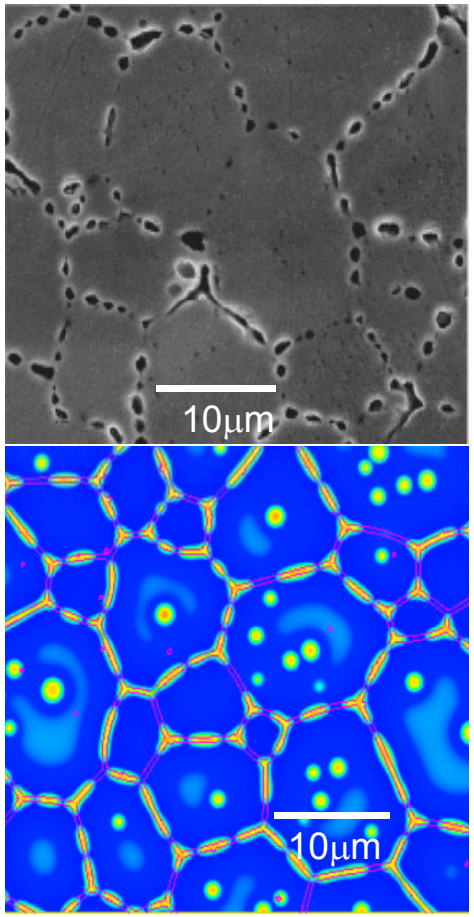


Fig. 1. Top: UO_2 fuel irradiated in PWR[2] Bottom: Phase Field simulation of UO_2 microstructure evolution[3]

[1]M. Stan, J. Nucl. Eng. Technology, **41** (2009) 39-52.
[2]I. Zacharie *et. al.,* J. Nucl. Mater. **255** (1998), 92-104.
[3]S.Y. Hu et al., J. Nucl. Mater. **392** (2009) 292–300.

PRD ⌘: *understanding atomistic and collective fundamental mechanisms of radiation effects in materials*

Scientific Challenge

Key fundamental unit processes involving solute atoms and point radiation defects, and phenomena that control the collective microstructural evolution, are poorly understood. These represent source terms for long-term development of persistent microstructures and accompanying physical and mechanical property changes due to radiation.

Research Direction

Quantify defect production characteristics
Measure and model solute and radiation point defect interactions with line and planar sinks (dislocations, grain boundaries, precipitates, etc.)
Observe defect and solute agglomeration processes
Understand the life cycle of microstructural features and their impact on material properties

Capability Gap

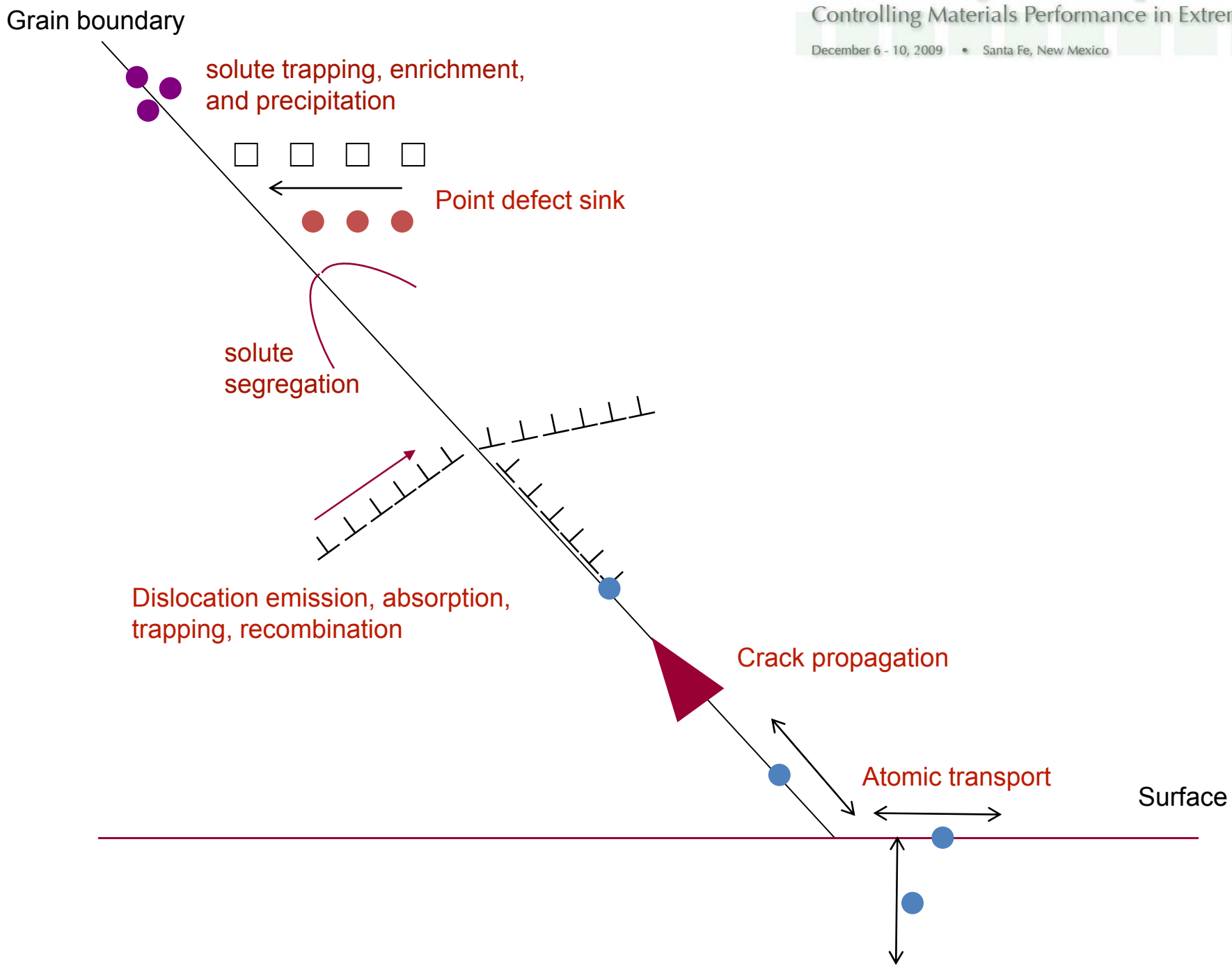
Currently very limited in-situ test capabilities

New measurement tools capable of simultaneous spatial and temporal resolution approaching 0.1nm and <1ps are needed

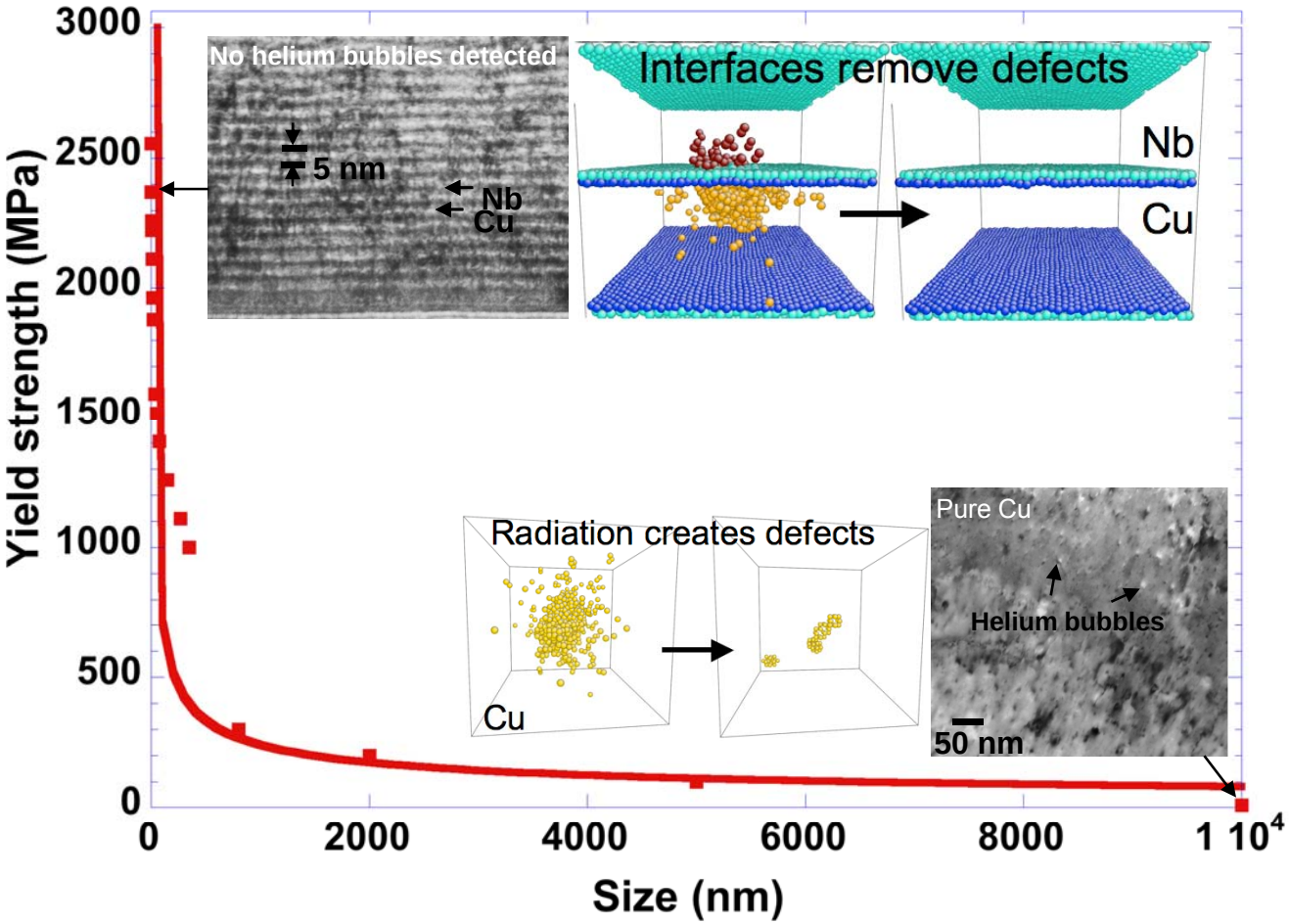
Capability is needed for simultaneous measurement by multiple probes (in-situ or ex-situ) in order to firmly establish links between microstructure and properties

Potential Impact

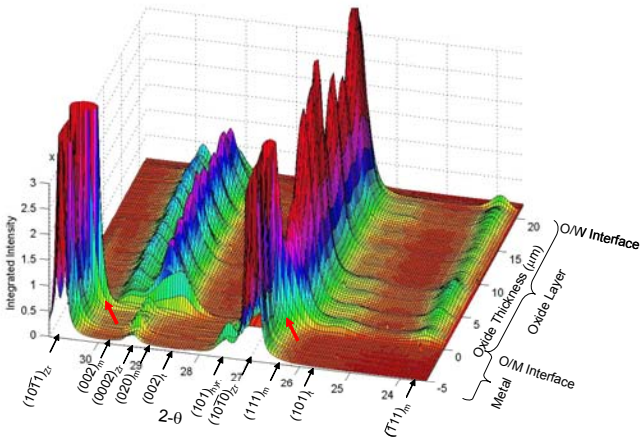
Development of validated physics-based multiscale models
- Extrapolation beyond current knowledge base
- Understand flux effects (including pulsing)
- Understanding how to use accelerated testing
- Bottom-up design of materials with unprecedented tolerance to extreme environments


Grain boundary
solute trapping, enrichment, and precipitation
Point defect sink
solute segregation
Dislocation emission, absorption, trapping, recombination
Crack propagation
Atomic transport
Surface

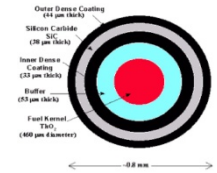
Interfaces act as obstacles to slip and sinks for radiation induced defects. Hence, nanolayered composites provide orders of magnitude increase in strength <u>and</u> enhanced radiation damage tolerance compared to bulk materials.

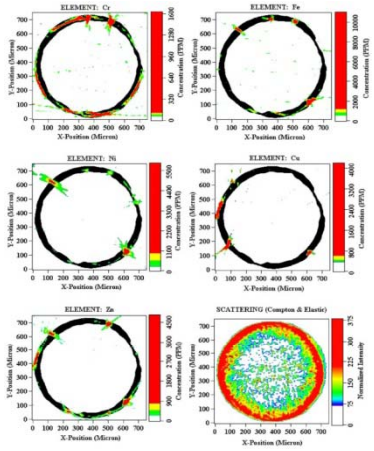


Applications of Synchrotron Radiation in Nuclear Materials

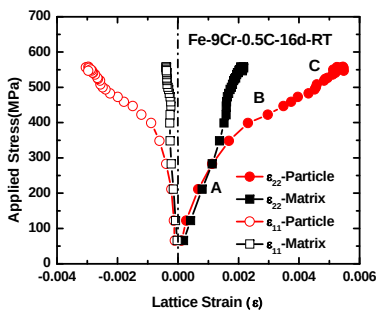


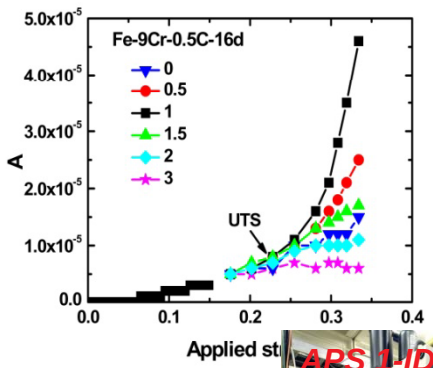
Micro-beam XRD to obtain information of oxide structure in a zirconium alloy oxide layer formed in 360C pure water. (A. Motta et al, 2004).

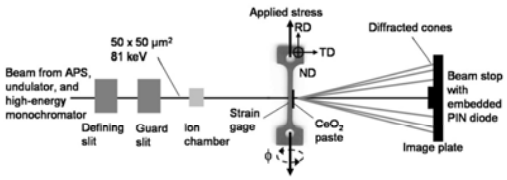




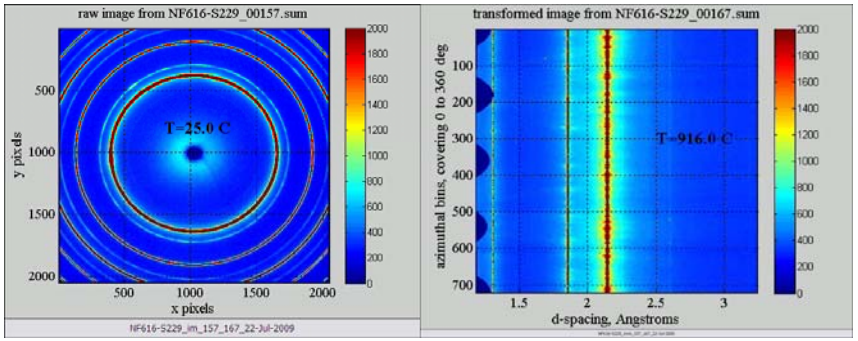
X-ray fluorescence micro-tomography shows spatial distribution of trace elements in TRISO fuel particles (M. Naghedolfeizi et al 2003)







Lattice strain and void evolution in ferritic alloys studied by XRD and SAXS (J. Stubbins et al 2009)



Time-resolved XRD measurements during annealing and straining in NF616 ferritic steel (Meimei Li et al, 2009)

Cross-cutting themes/issues & meta topics

- Prediction and control of the response of materials in a nuclear environment (temperature, stress, irradiation, corrosion) requires development of basic understanding of property changes driven by microstructural development informed by atomistic processes

- Tools (models, measurements, and organizational structures) that improve our understanding at each level must be developed
  - Probes or multiple probes at multiple scales
  - Ion beams, photon sources, plasmas, and neutrons

Final thoughts (work remaining to be done)

YES

Backups

PRD final (text) product

When translated to a text document (~ 3 pages), each PRD will follow the below format:

PRD title

Problem Statement (a few sentences describing the problem)

Executive Summary (abstract/executive summary of scientific challenge, research direction, and capability gaps)

Research Directions (upper right of quad chart)
A few enumerated research directions described in ~ one paragraph/each

Scientific Challenges (upper left of quad chart)
Including a bulleted list of specific questions to which the answers are presently unknown and a brief discussion of why they're significant

Capability Gaps (lower left of quad chart)
Explicit discussion of needed new tools (both experimental and computational); gaps should be non-facility specific – allowing multiple routes to close the gap

Potential Impact (lower right of quad chart)
Discuss how solving this problem would impact the field

Things to do

Finish PRDs
Commence writing text to support PRDs
Provide Todd text and key images
Draft 1 paragraph that supports your initial presentation

Radiation Matter Interactions for Energy Conversion (George Crabtree, Dave Teter, Paul Follansbee)

Panel 3: Materials discovery and processing for advanced functionality
The need for predictive design, discovery, and growth of new materials, especially single crystals, to achieve advanced functionality is a grand challenge. Further, exploiting the full suite of synthesis, fabrication, and processing capabilities is essential if we are to discover by design next generation materials and enable the transition from observation to control.

Panel 4: “Making every photon count”
Translating quantum phenomena that arise uniquely on the nanoscale to bulk performance requires a predictive understanding of interface and microstructure effects that limit integrated efficiency and system lifetime that are central to achieving the full potential of advanced functional materials.

Radiation Matter Interactions for Energy Conversion Panel Members

Paul Follansbee, *Saint Vincent College*
John Hack, *Bettis Atomic Power Laboratory, Bechtel*
Quanxi Jia, *LANL*
Tom Lograsso, *Ames Lab*
Greg Olson, *Northwestern University & Questek Innovations, LLC*
Ian Robertson, *University of Illinois*
David Teter, *LANL*
George Crabtree, *ANL*
Alan Bishop, *LANL*
Zachary Fisk, *UC Irvine*
David Singh, *ORNL*
Darryl Smith, *LANL*
Toni Taylor, *LANL*
Alp Findikogllu, *LANL*
Sasha Balatsky, *LANL*
James Belak, *LLNL*

With help from many others

Original Priority Research Directions

Decadal Challenges for Predicting and Controlling Materials Performance in Extremes

December 6 - 10, 2009 • Santa Fe, New Mexico

Microstructure science under irradiation
Materials design for resistance to corrosion under irradiation
Mechanical properties and dimensional stability during irradiation
Understanding unit processes in radiation effects in materials
Understanding collective radiation effects phenomena in materials
Development of robust radiation effects model of radiation damage evolution

Materials design and lifetime prediction
Rapid materials discovery and synthesis
Corrosion science under extreme environments
Controlling nucleation phenomena

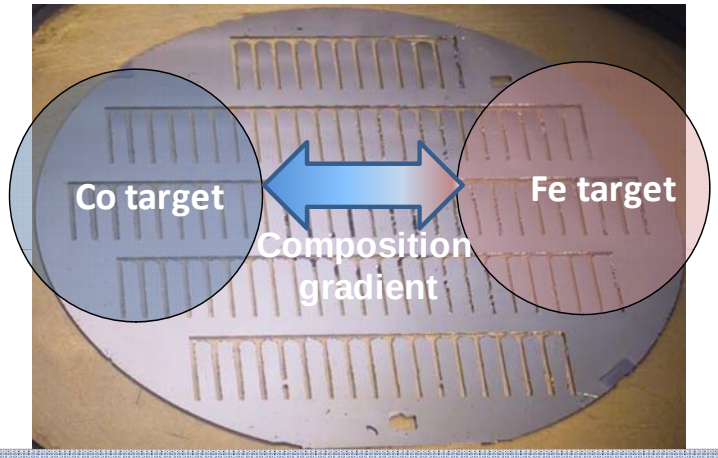
Controlling functionality from complexity
Accelerating materials discovery
Controlling photon-matter interactions: making every photon count

Quantification of structure at extreme P-T and strain rate
Control of end state performance through predictive manipulation of process-aware structure property relations
Manipulating novel states, structures, and order-disorder beyond thermodynamic constraints
Predict characterize & control the performance of matter under conditions between condensed matter & ideal plasmas
Determine the transport of energy, momentum and mass under extreme density and temperature
Understand and control the chemistry of materials under extreme conditions

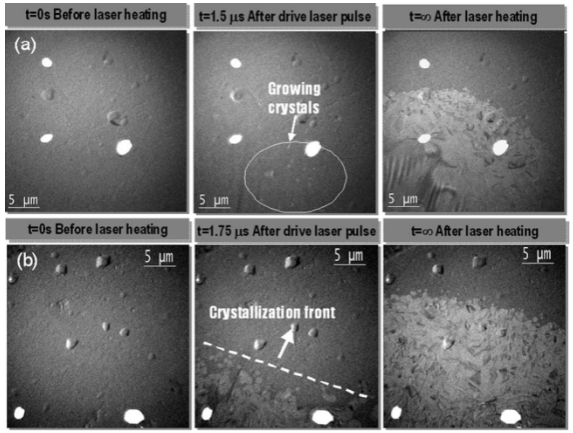
Develop statistical approaches for the analysis and modeling of extreme valued properties
Materials variability
Predict and control properties of heterogeneous materials
Atomistic simulations of slow deformation and flow of matter
Bridging time scales from nucleation to microstructure
Accurate simulations of complex materials in radiation environment

Radiation Matter Interactions for Energy Conversion Panel
Four PRDs

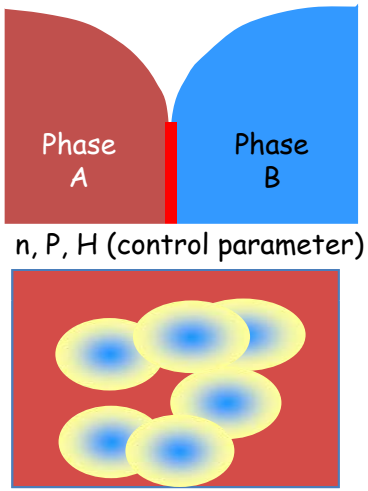
Accelerating Materials Discovery



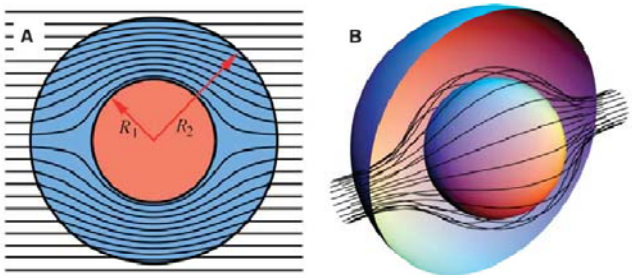
Controlling Nucleation Phenomena



Controlling Functionality from Complexity



Controlling Photon-Matter Interactions



PRD: *Controlling nucleation phenomena*

Scientific Challenge

Nucleation is fundamental in controlling many material phenomena, yet predictive capability remains elusive. Areas of : Phase transformations in both solid-liquid, solid-solid, solid-vapor, Defects and defect structures
Fatigue and fracture, corrosion pitting,
Grain structure and orientation relationships

Capability Gap

•Development of experimental probes and models that can probe nuclei at small length scales (10 nm) and time scales (~ps-ns) but sampling large volumes (microns – mm) over time scales (secs to days).
• 3D X-ray diffraction measuring lattice strain (~10 nm res.) to detect onset of nuclei (current state 2D, 20 nm).
• Compositional and structural fluctuations can be measured via TEM down to nm scale, but not at time scales needed. How do we improve temporal resolution?
• Connection of theoretical models with real heterogeneous fluctuations.
• Develop theoretical models, models and experimental probes aimed at understanding interfacial mechanisms: nucleant interface and nucleus interface.

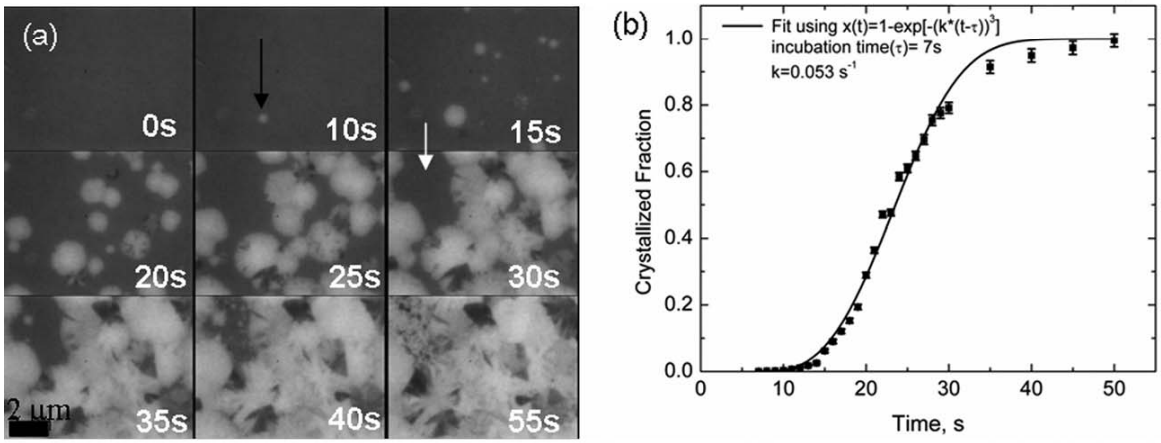
Research Direction

- Determine the form of the pre-embryo, nucleant and nuclei and interrelationships (structure, composition, strain)
- Determine methods to characterize these various forms.
- Develop theories for heterogeneous nucleation that account for multiply distributed nucleant potency.
- Experimentally characterize and quantify distributed heterogeneous nucleation behavior.
- Explore methods for seeding the structure with defect controlling nucleation
- Use undercooling techniques to watch embryo development at time scales that we can measure.

Potential Impact

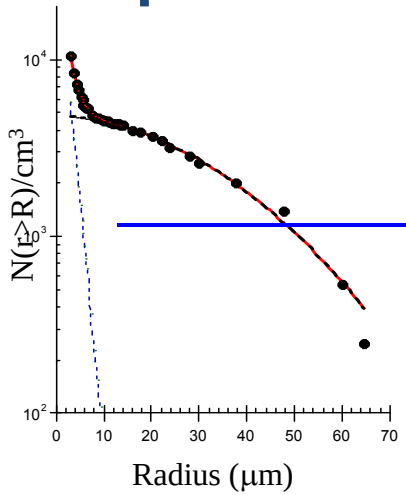
Remove key barrier to predictive capability for microstructural control across numerous application areas.

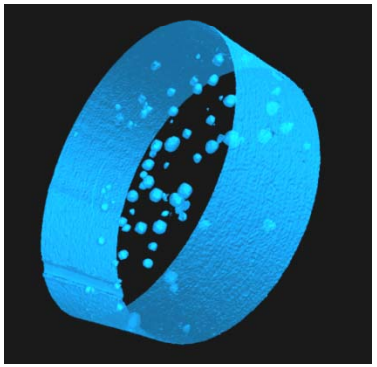
Enable control of novel distribution of phases for improved functionality.

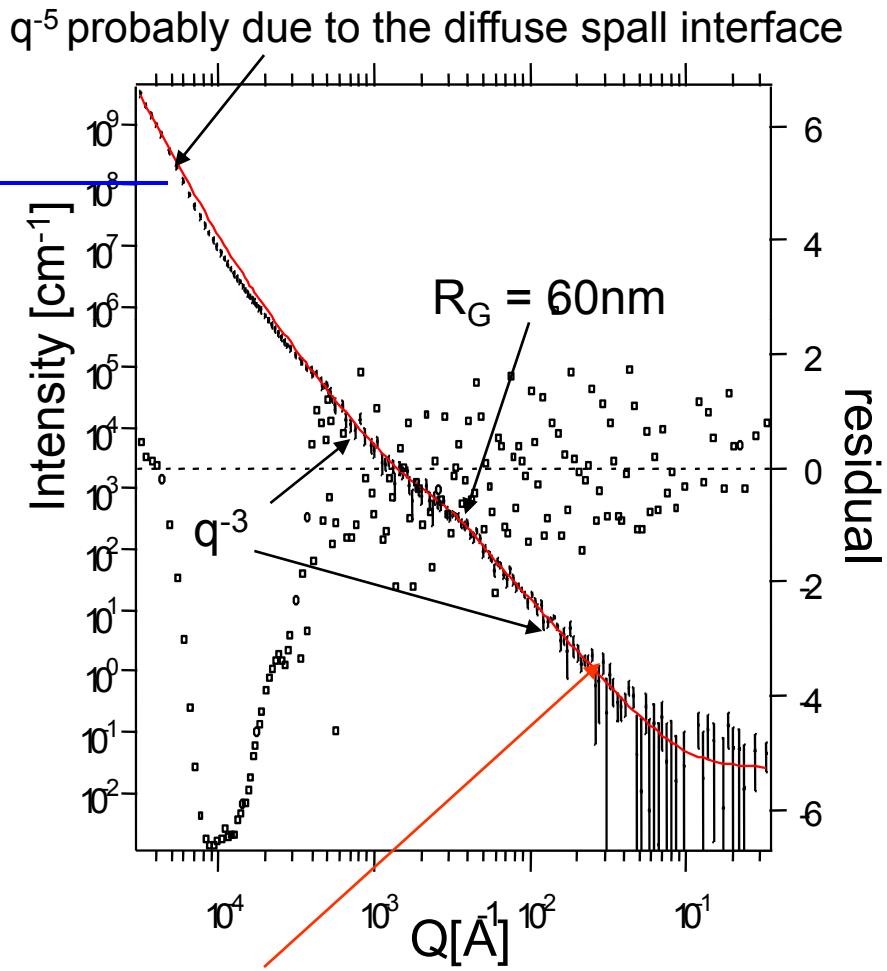


Crystallization of amorphous NiTi, when heated by pulsed laser irradiation, in:
Strongly Driven Crystallization Processes in Metallic Glass, Thomas LaGrange, David S. Grummon, Bryan W. Reed, Nigel D. Browning, Wayne E. King,and Geoffrey H. Campbell, Applied Physics Letters 94, 184101 2009

Analysis of SAXS/SANS data yields information on particle size, spacing and morphology







Unified Fit (Beaucage, Ilavsky)

Extracting the void distribution from the scattering data requires "fitting" the experiment. We have overlapping tomography and scattering data!

Scattering and tomography together results in a void size distribution spanning 4-5 orders of magnitude in scale!!

PRD: *Accelerating Materials Discovery*

Scientific Challenge

Discover next generation high performance materials
Establish a new paradigm for materials discovery, tight integration of theory, synthesis, and characterization

Capability Gap

Timely integration of theory with synthesis and characterization

Guided exploration of materials space

In situ, real time environmental characterization tools

Insufficient scale of materials synthesis capability nationwide

Rapid methods for assessing synthesis routes including prediction of multi-component thermodynamics and kinetic pathways

Process modeling to assist in identifying synthesis routes for targeted compounds.

Rapid screening tools, including very small samples, for assessing material functionality; e.g. multi-component system on chip exploring properties.

Research Direction

Discover guiding principles for materials functionality, e.g., Zintl concept

Synthesis and process modeling of novel phases from extreme conditions: pressure, photon, electric, radiation fields. electrolytic

Exploration of unstable or metastable phases-anomalous valence states

Morphological control of crystal growth to include defect and impurity control.

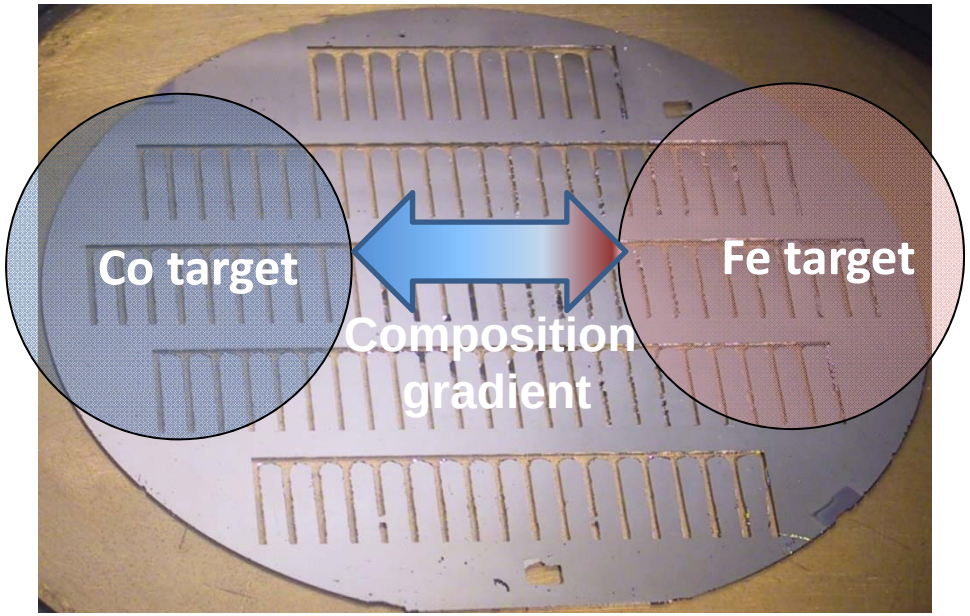
Potential Impact

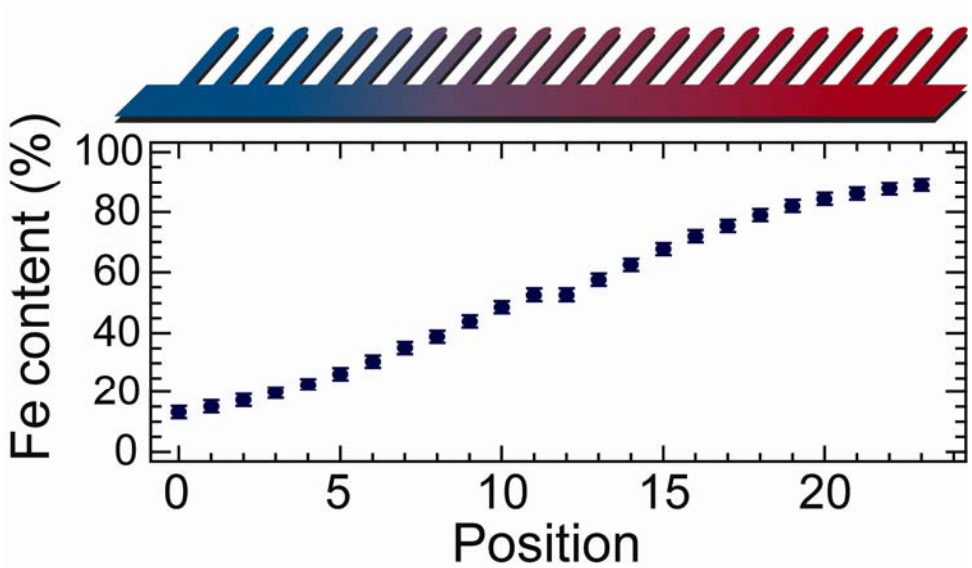
Accelerated identification of new materials with novel properties or order of magnitude improvement of properties.

Training of new generation of materials synthesizers

Rapid introduction of new materials into the basic research and technology streams

Magnetostrictive thin-film composition spreads





Deposition Condition:

- Films deposited on cantilever array
- Magnetron Co-sputtering deposition
- Deposition temp: RT (film quenched)
- Film thickness ~.5 to .8 μm

Cantilevers:

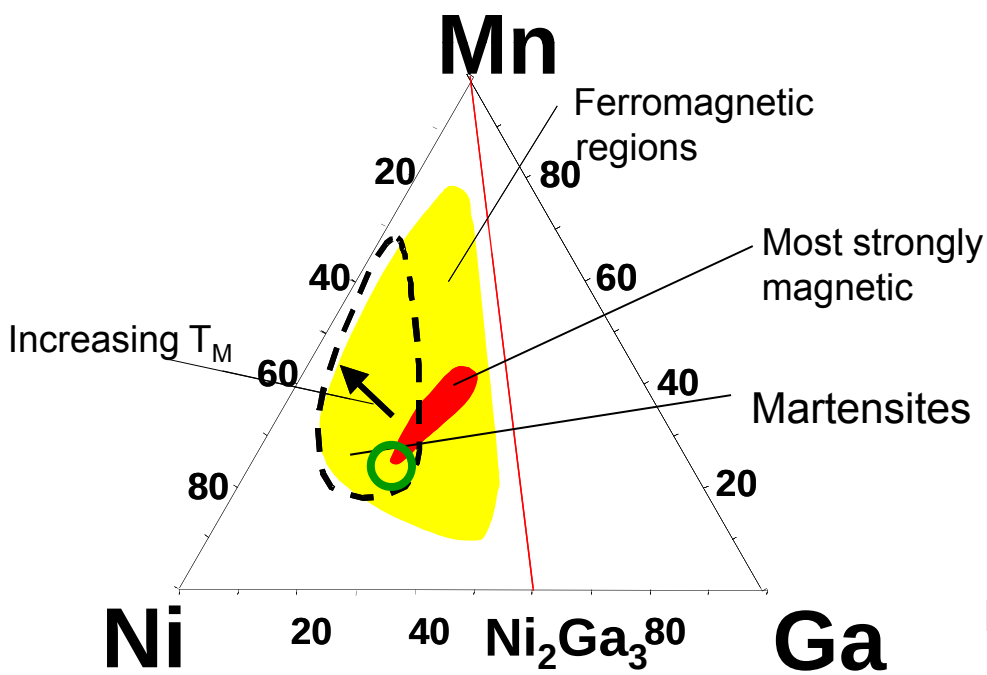
- Si/SiO_2 substrates
- Substrate thickness ~75 μm

I. Takeuchi, U. Maryland

High-throughput mapping of physical properties

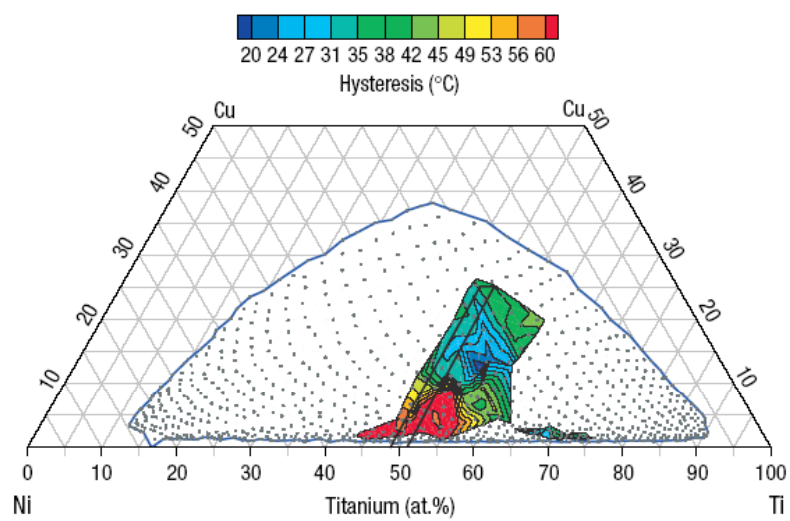
A variety of rapid screening techniques are employed: scanning SQUID, cantilever libraries, etc.

Mapping of FSMA



Nature Materials **2**, 180 (2003)

Mapping of FSMA



Lattice parameters and hysteresis mapping

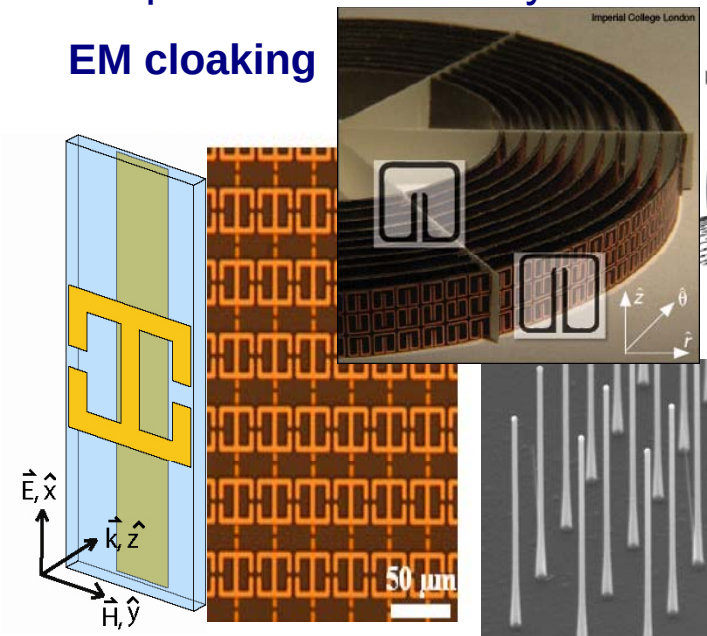
Nature Materials **5**, 286 (2006)

I. Takeuchi, U. Maryland

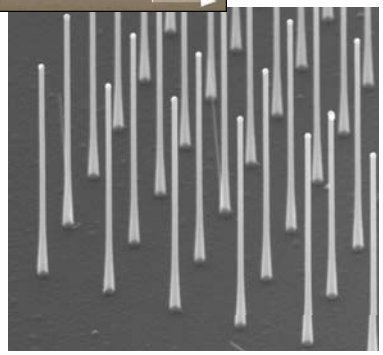
Controlling photon-matter interactions: making every photon count

The fields of plasmonics and metamaterials have allowed rudimentary control over classical interactions of light with matter while a combination of materials design and coherent control enables optimization of quantum mechanisms and/or wavefunctions for specific functionality such as reactivity, phase transformation or charge separation.

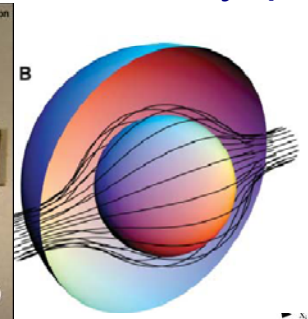
EM cloaking

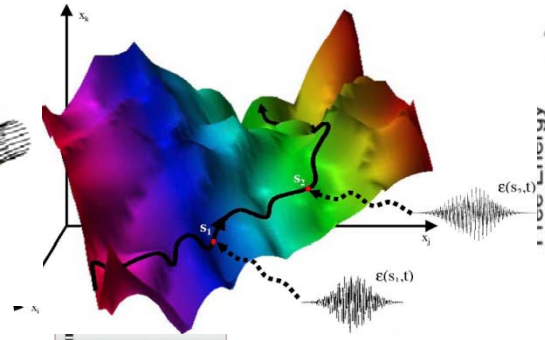


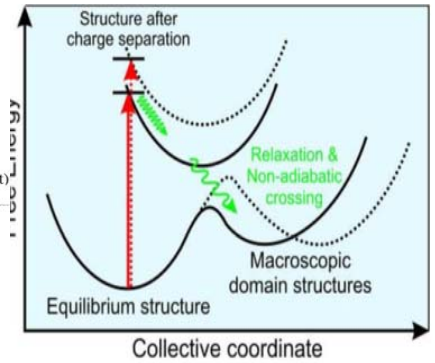
'Perfect' absorber

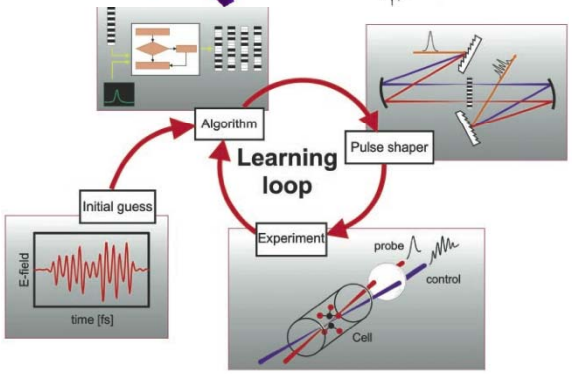


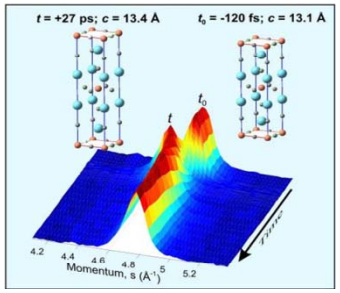
Nanowire-based photovoltaics











Ultrafast coherent control of wavefunctions

Goal: Active control of photon-matter interactions:
i) classically through tunable photonic materials with multifunctional, broadband electromagnetic response; ii) in the quantum regime through the development of a predictive capability to design materials and/or pulse sequences to coherently control the outcome of the interaction, either electronically or structurally

PRD:
Controlling photon-matter interactions: making every photon count

Scientific Challenge

Goal: Exquisite control of photon-matter interactions: i) classically through tunable photonic materials with multifunctional, broadband electromagnetic response; ii) in the quantum regime via quantum control of chemical reactions & material functionality. Recent efforts span nascent R&D in metamaterials and plasmonics to optimization of optical excitations in materials thru empirical coherent control of wavefunctions.

Capability Gap

- Predictive theory of nanoscale EM interactions beyond effective medium theory and of coherent photon-matter interactions.
- Optimization of materials for conversion of photons to other excitations (energy conversion)
- 3D nanofabrication with ability to achieve 2D placement of nanostructures
- Ultrafast coherent spectroscopic characterization with far-field and near-field nanoscale imaging
- Design/implementation of ultrafast pulses to control structural and/or electronic excitations.

Research Direction

Classical regime: developing new approaches to address issues of bandwidth, loss, isotropy, dispersion, inhomogeneity, bianisotropy, active control, and nonlinearity

Quantum regime: develop predictive capability to design materials and/or pulse sequences to coherently control the interaction of photons with electronic or structural degrees of freedom.

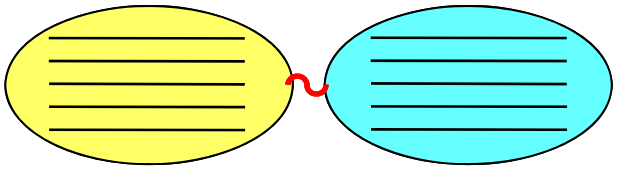
Potential Impact

•Controlling photon-materials will be essential for next generation renewable energy technologies, the design of catalytic materials and multifunctional materials for sensing, advanced computing and communications, the understanding of reaction mechanisms, and new directions such as the nanomechanics of Casimir force control.

•20 years timescale; accurate and then predictive simulation, synthesis, fabrication and advanced characterization of complex materials, measurement & control of ultrafast phenomena

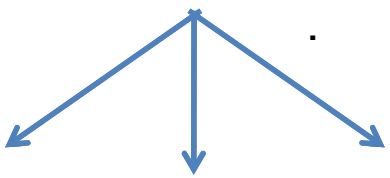
Controlling Functionality from Electronic Complexity

spacing ~ kT, h√, eE, . . .



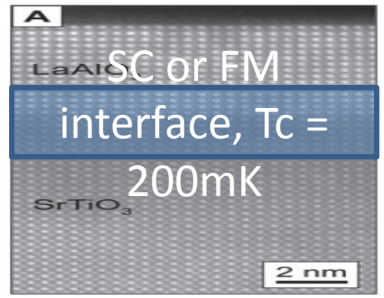
metal, . . .

magnet, . . .



phase transition
nanophase separation
superconductor, insulator
. . .

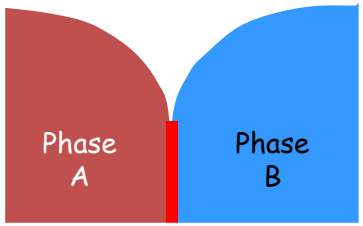
flexible response
spatially resolved functionality
response to defects – self-healing
positive or negative feedback
create new state from external stimulus, emergent



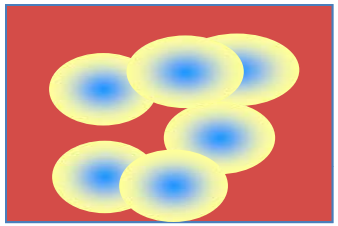
J.Rehren ,Science 317, 1197, 2007

biology – ultimate example
sequential conformational functionality
systems design
→ inorganic analogs

hierarchical structure
long range -> robustness
short range-> functionality



n, P, H (control parameter)



macroscale functionality
new properties / giant susceptibilities

sequential/temporal functionality
temporal dynamics
e.g., photon-chemistry-fuel
fast switching behavior

Understand and control the functionality of complex materials
Harness nanoscale electronic complexity for robust macroscale functionality

PRD: *Controlling electronic functionality from complexity*

Scientific Challenge

- Understand and control the functionality of complex materials, such as nanoscale phase separation, quantum criticality, coupled degrees of freedom, coupled hierarchical length and energy scales
- Harnessing nanoscale complexity for robust macroscale functionality
- Exploiting the functionality from degeneracy of energy scale (softness)
- Materials beyond Bloch theory, “thinking outside the Bloch”
- Multi-functionality, mutuality of contra-indicated properties

Capability Gap

- Nonlinear, nonadiabatic, nonequilibrium, dynamic functionality
- Understanding and achieving robustness via feedback from nonlinearity
- Tools to identify the relevant degrees of freedom (nanoscale controlling macroscopic)
- Theoretical framework for understanding length, time and energy scales of emergent functionality

Research Direction

- Correlating structural and spectroscopic information
- Range of time and length scales, e.g., pump probe, fluctuation spectroscopy, multidimensional NMR-like
- Identify and understand model systems for functionality, e.g., relaxor ferroelectrics, heavy fermions, . .
- Guiding design principles for complex functional systems

Potential Impact

- Engineering high functionality in nanostructured materials
- Structure - function cycles from intrinsic feedback (training, ageing, self-healing, . . .)
- New materials with novel ground states
- New materials with unprecedented performance, such as 50% solar conversion efficiency, thermoelectric figure of merit greater than 5, superconductors for long distance power transmission
- Materials for sustainable energy application

Matter Interactions in Extremes

Process-aware material certification in extremes

When solids stop being solids

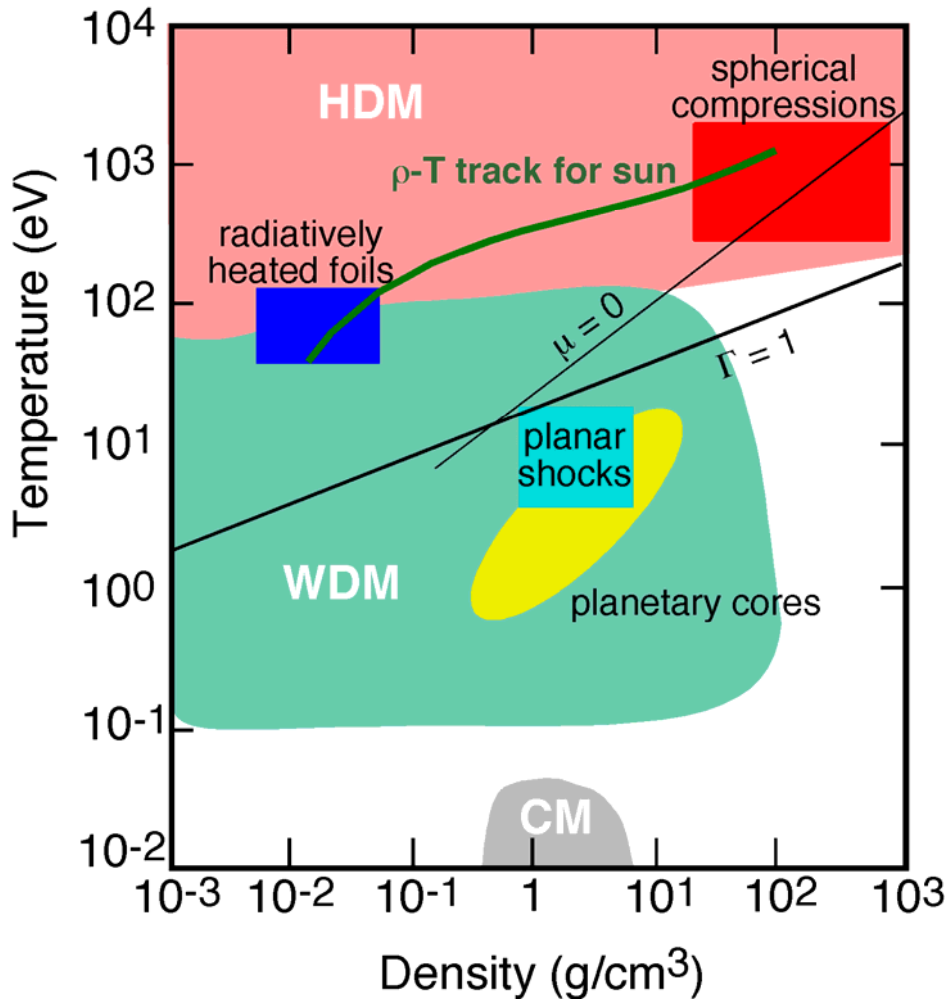
Rus Hemley (CIW) and Roger Falcone (UC Berkeley), Chairs
Malcolm McMahon (U Edinburgh) and Bob Cauble (LLNL), Co-chairs

Malcolm Andrews (LANL)
Cris Barnes (LANL)
Jim Belak (LLNL)
John Benage (LANL)
Jon Boettger (LANL)
Neil Bourne (AWE)
Curt Bronkhorst (LANL)
Rip Collins (LLNL)
Michael Di Rosa (LANL)
Juan Fernandez (LANL)
David Funk (LANL)
Jim Glownia (DOE/SC)
Alex Goncharov (CIW)
Rusty Gray (LANL)
Donald Hickmott (LANL)
Chi-Chiang Kao (BNL)
Haeja Lee (SLAC)
Guoyin Shen (CIW/APS)
Choong-Shik Yoo (WSU)

Matter Interactions in Extremes

Process-aware material certification in extremes

When solids stop being solids



Matter Interactions in Extremes

Process-aware material certification in extremes

When solids stop being solids

New physics and chemistry as one moves from near-STP to extremes of temperature, density, and pressure, away from the comfort of well-explored STP condensed matter and confidence of an ideal plasma. New territory, where the matter states we can create are transient, requiring theory well beyond present capabilities, encompassing new opportunities.

Matter Interactions in Extremes

Principal Research Directions

Predict, characterize, and control the performance of matter between solids and plasmas

Control the transport of energy, momentum and mass under extreme density and temperature

Exploit the chemistry of a new periodic table at extreme P, T

Predict, characterize, and control the performance of matter between solids and plasmas

Scientific Challenge

Understand and utilize state of matter - structure, phase transitions, kinetics - as we traverse pathways through the phase space described by extreme density and temperature

Research Direction

Measure EOS, atomic and electronic structure, opacity of WDM

Develop self-consistent theory that permits prediction of WDM properties

Capability Gap

Ability to treat degeneracy, strong coupling, and quantum effects simultaneously

Creation of homogeneous WDM using arbitrary pathways

Multi-beam sources for WDM sample and probes for simultaneous measurements of multiple state variables

Develop time-dependent Thomson scattering (theory with experiment) and diffractive imaging

In situ diagnostics for atomic-to-macroscopic structure higher fluence, higher energy light sources

Potential Impact

Understand and quantify load-path-dependency of properties of matter

Predict, manipulate, and control high-pressure matter

Ability to optimize inertial fusion targets

Understand creation mechanisms, internal structure, dynamics and evolution of astrophysical objects

PRD: Predict, characterize, and control the performance of matter between solids and plasmas

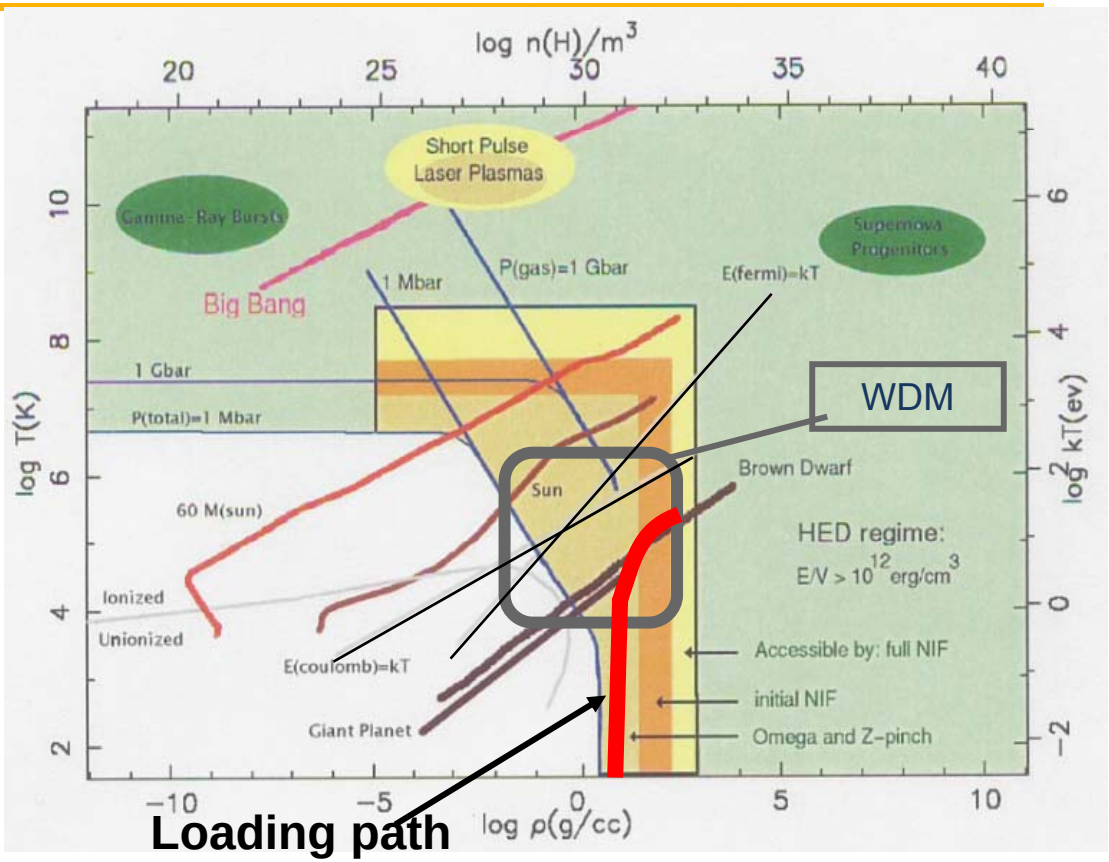
Research direction: measure EOS, atomic and electronic structure, opacity of WDM (illustrates capability gaps)

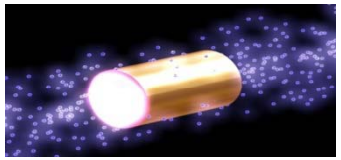
Illustrative **experiment** on **H**:

arbitrary loading **pathway**,

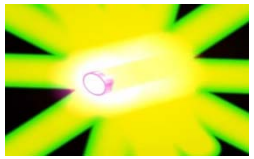
cryo H cylinder inside Pb liner

- Isochoric heating to few eV with proton beam
- Isentropic compression to few g/cc
- Prad diagnostic of ρr
- Probing with keV xrays & heavy ion-beam
- X-ray Thomson scattering, dE_b / dx, opacity

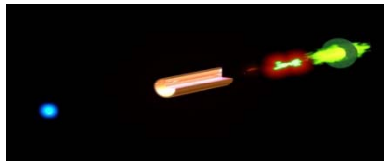




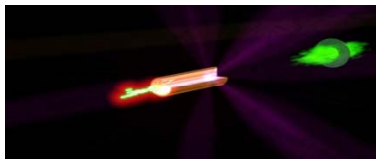
Isochoric pre-Heating (ions)



Isentropic compression



Probing with x-ray pulse & ion beam

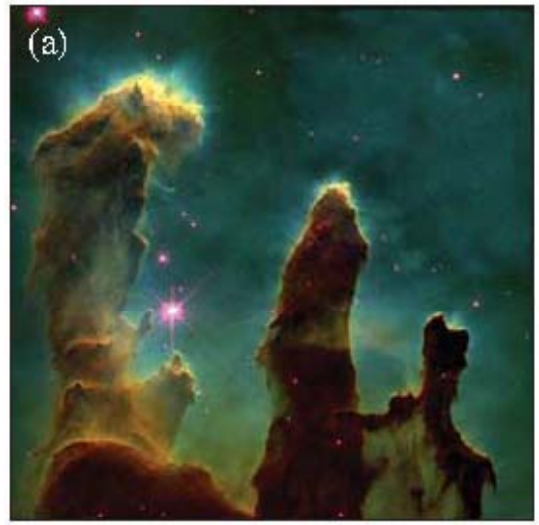


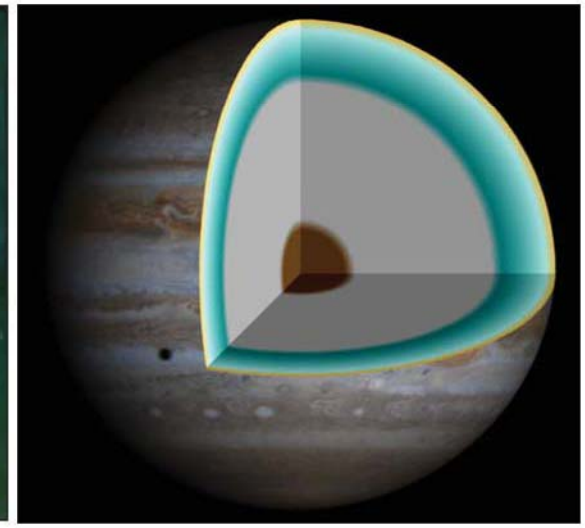
X-ray TS, & dE_b / dx measurement

PRD: Predict, characterize, and control the performance of matter between solids and plasmas

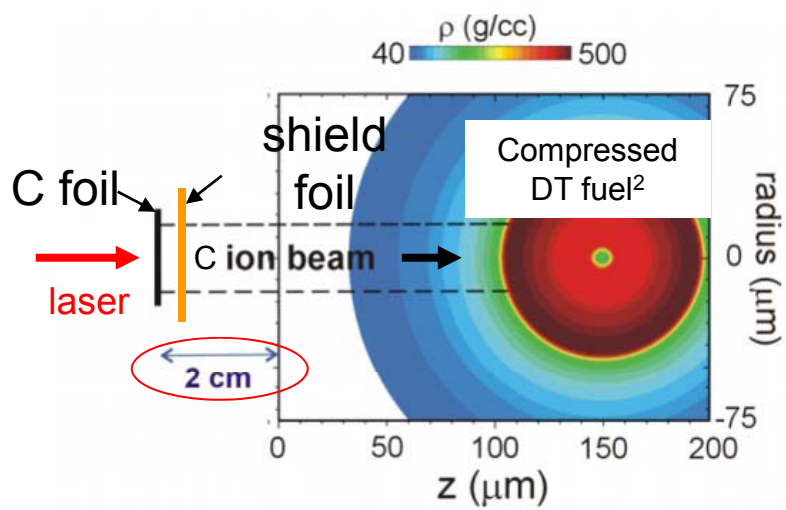
Potential Impact: Understanding WDM advances our understanding of the universe and enables important applications

- Understanding the universe
  - Star creation and
  - Planetary physics





- Advances use-inspired research
  - Inertial fusion
  - Weapons physics
  - Materials damage, manufacturing under extreme conditions (*e.g.*, lasers & pulsed power)



Control the transport of energy, momentum and mass under extreme density and temperature

Scientific Challenge

Develop a methodology for calculating transport properties in a quantum system where usual order parameters fail

In situ measurement of transport properties in WDM before hydrodynamic disassembly, including spatial gradients.

What are the hydrodynamic properties of extreme matter?

Capability Gap

Creation of homogeneous WDM using arbitrary pathways

Multiple beams for making WDM sample and probing

In situ diagnostics for atomic-to-macroscopic structure higher fluence, higher energy light sources

Develop time-dependent Thomson scattering (theory with experiment) and diffractive imaging

Ability to treat degeneracy, strong coupling, and quantum effects simultaneously

Research Direction

Develop a self-consistent methodology for calculating viscosity, electrical and thermal conduction, stopping power, and electron-ion equilibration in extremes of density and temperature.

Measure transient processes in WDM

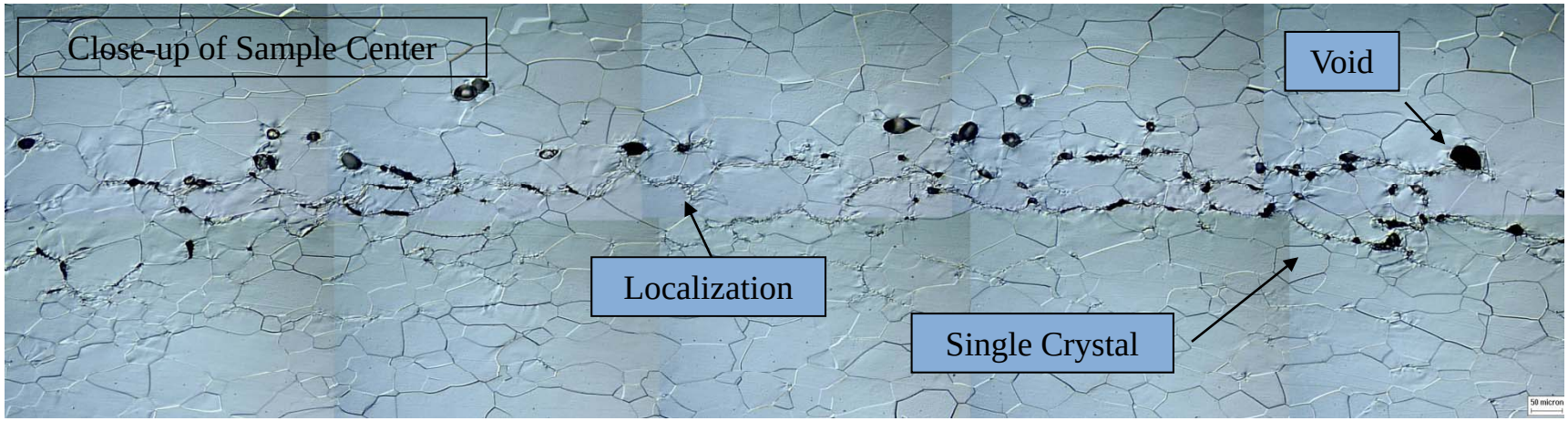
Potential Impact

How matter flows under extreme conditions is significant uncertainty

Applications of variable-density fluid flows in rapidly transforming objects: ICF explosions, stellar explosions, nuclear reactors, cooling devices, and casting.

Control the transport of energy, momentum and mass under extreme density and temperature

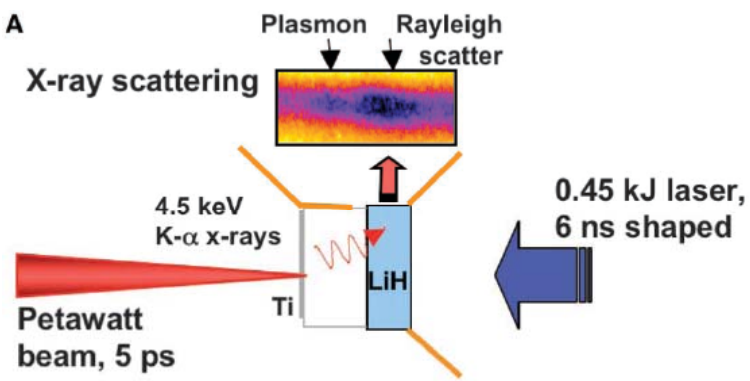
Once you make and characterize warm dense matter (matter at extremes of density and temperature where the usual order parameters fail) the next problem is determining its dynamic behavior: what are the fluxes of mass, momentum and energy in conditions where materials may loose strength, may melt and re-freeze, are in transition regimes in fluid parameters, and may introduce quantum or coupling effects that are new?



The transport of mass, momentum and energy are scientific challenges with cross-connects to constitutive properties and chemistry

Control the transport of energy, momentum and mass under extreme density and temperature

Research Direction: Transient measurements
Capability Gap: *In situ* diagnostics with high time and spatial resolution



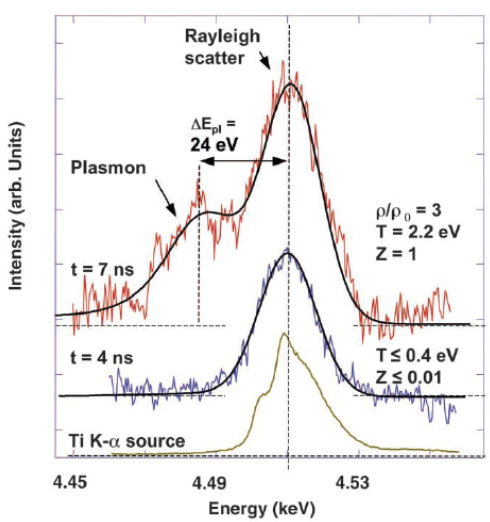


Fig. 2. X-ray scattering spectrum from shocked LiH, showing elastic Rayleigh scattering and inelastic plasmon scattering features. At $t = 7$ ns (top), the plasmon energy shift of 24 eV indicates 3× compression, whereas the intensity of the elastic scattering feature shows heating to temperatures of 2.2 eV. Earlier in time only elastic scattering is observed (middle) as demonstrated when compared with the K-α source spectrum (bottom). The observation of plasmons at $t = 7$ ns indicates the transition to the metallic free electron plasma in the solid phase.

From Kritcher et al., Science 322 (2008) pg. 69

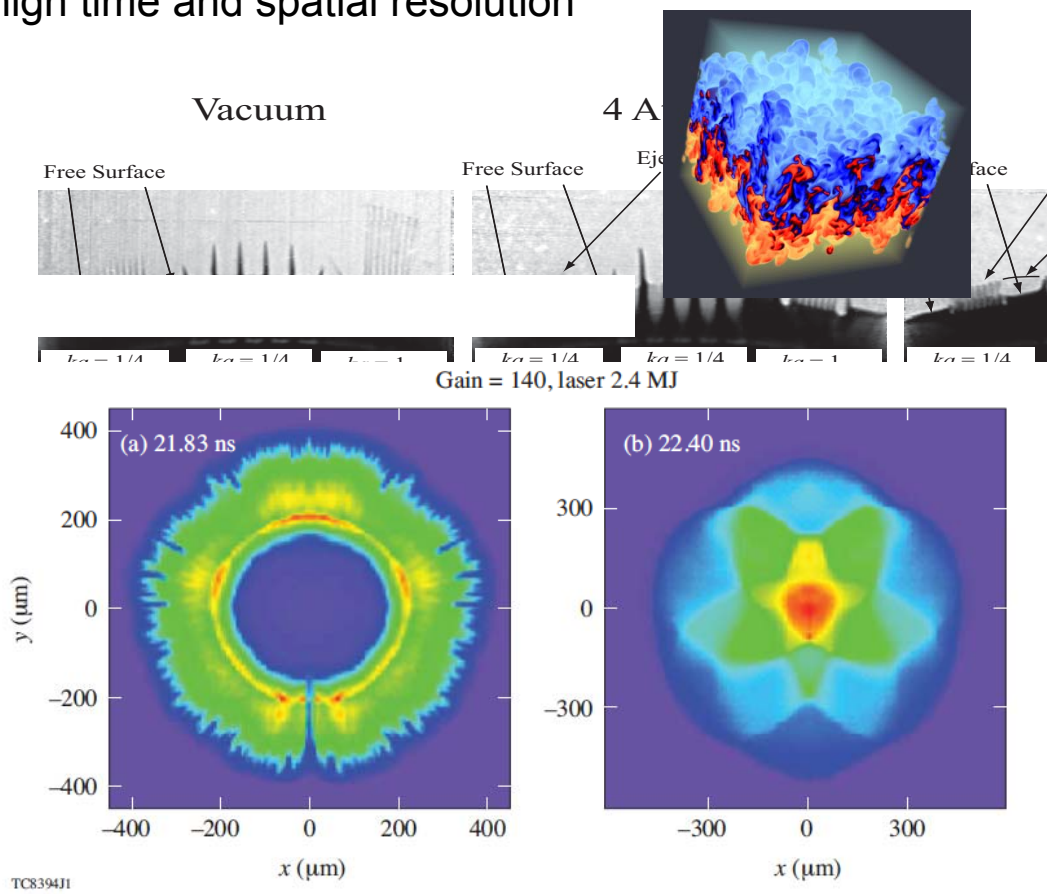


Figure 4. Snapshots of an IFE target implosion (isodensity contours) from a two-dimensional simulation (courtesy, A. J. Schmitt, NRL).[2] Hydrodynamic instabilities cause density nonuniformities in the dense shell during the implosion (at 21.83 ns) and the burn phase (at 22.40 ns).

Matter Interactions in Extremes

Cross-cutting issues

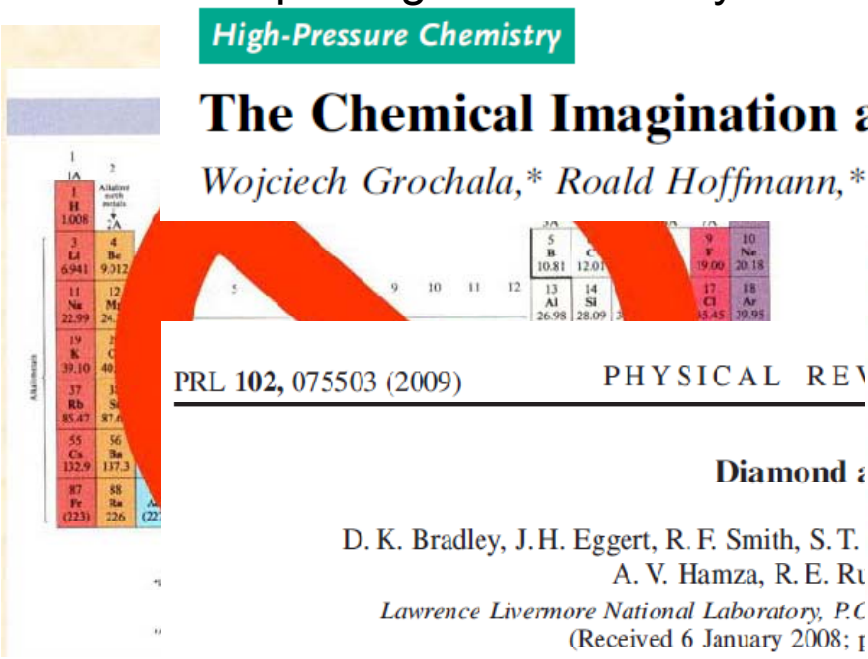
Structure – microstructure, form, evolution
process path
properties, control

In situ diagnostics – high-fluence, high-energy light sources
multi-beam sources for simultaneous measurements of multiple variables

High-performance computing – hardware, software infrastructure, beyond DFT codes

Multiscale ‘modeling’ – atomic-to-macroscopic

Exploiting the Chemistry of a New Periodic Table at Extreme P-T



High-Pressure Chemistry

DOI: 10.1002/anie.200602485

The Chemical Imagination at Work in *Very* Tight Places

Wojciech Grochala, Roald Hoffmann,* Ji Feng,* and Neil W. Ashcroft**

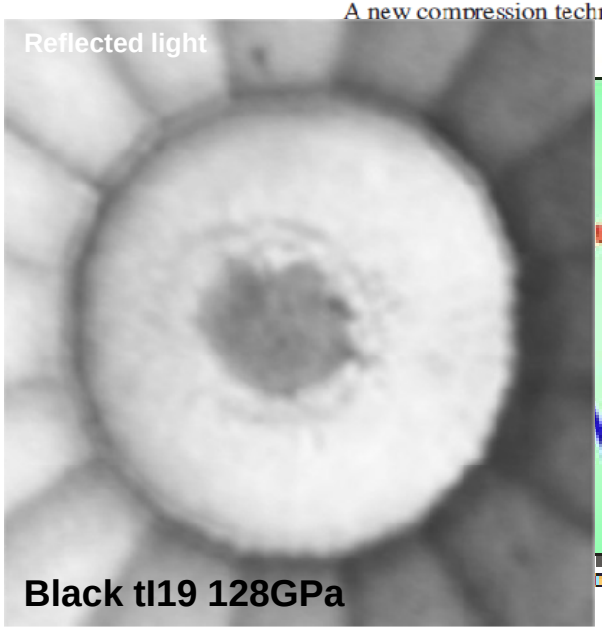
PRL **102,** 075503 (2009) PHYSICAL REV

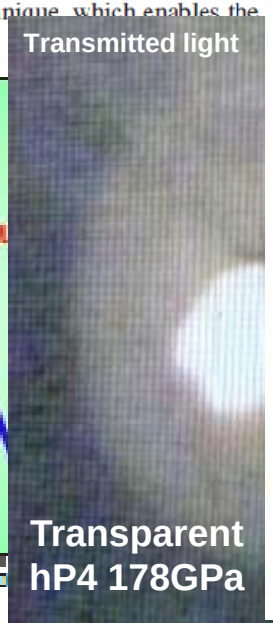
Diamond a

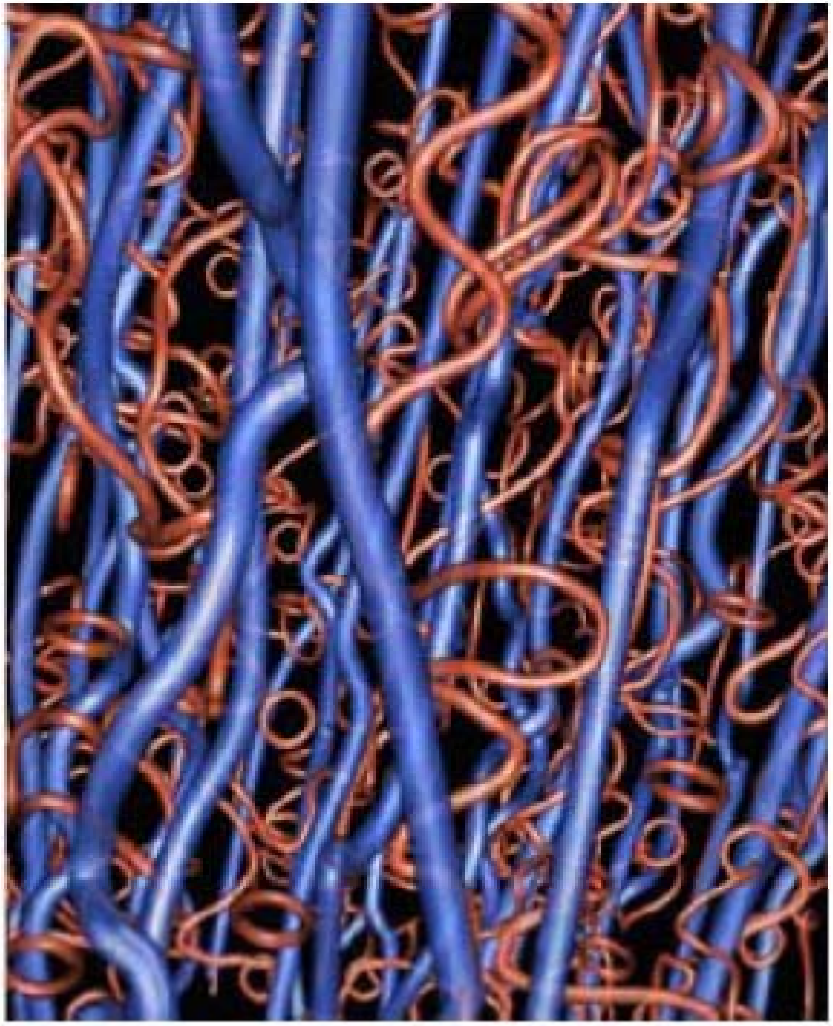
D. K. Bradley, J. H. Eggert, R. F. Smith, S. T.
A. V. Hamza, R. E. Ru
Lawrence Livermore National Laboratory, P.C

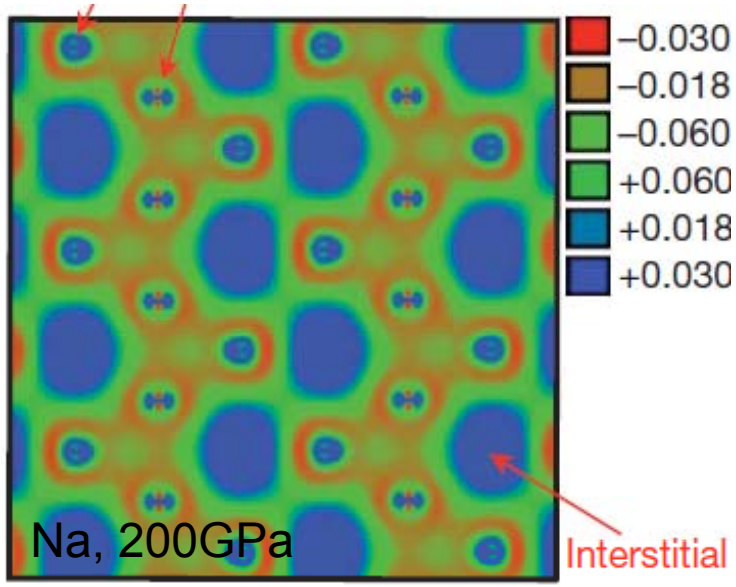
(Received 6 January 2008; r

A new compression technique, which enables the



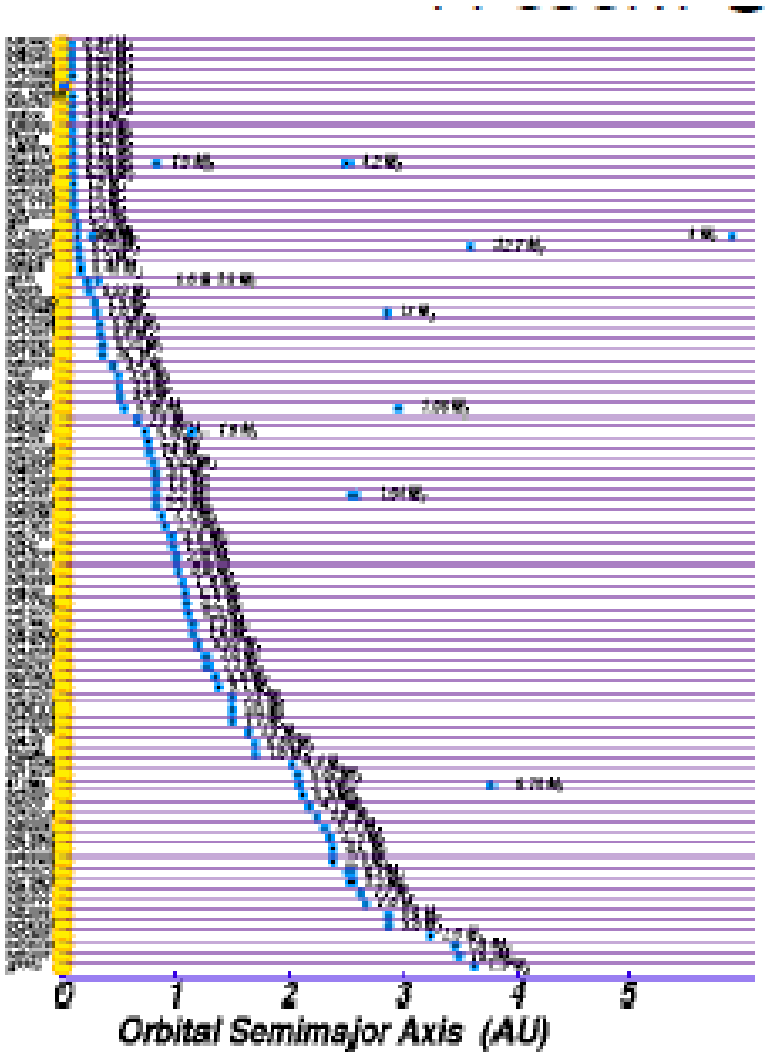





−0.030
−0.018
−0.060
+0.060
+0.018
+0.030
Na, 200GPa
Interstitial

Ma et al 2009

LCLS


0
1
2
3
4
5
Orbital Semimajor Axis (AU)

Exploiting the Chemistry of a New Periodic Table at Extreme P-T

Scientific Challenge

Predict and create novel chemical states at extreme PT beyond the periodic table

What are the effects of high pressure on atomic structure, bonding, and reactivity?

Is chemistry and structure different at a Gbar where core electrons may be involved?

Control of chemistry by matter/radiation coupling in concert with P and T

Research Direction

Develop computational tools for describing electronic structure that enable predictive capability of chemical reactivity, bonding, over a range T, P, ρ states

Develop ultrafast diagnostics to observe bond breaking/formation, local energy creation, stoichiometry, kinetics

Observe production and effects of transient states

Capability Gap

Ability to control state conditions with necessary precisely over a large range of T, P, ρ.

Time-dependent spectroscopic diagnostics capable of *in situ* (sub-surface) probing of extreme conditions at high spatial resolution

Development of time-domain diagnostics for extreme chemistry

Move beyond existing theories to overcome theory/experiment timescale gap

Potential Impact

Control creation of new, stable materials

Understand planetary interior chemistry, including mixtures

Understand matter where reactive energy – burn – is significant relative to other sources

Cross-Cutting Themes

Richard LeSar & Tony Rollett

Panels 7 & 8:

Panel 8 Members

- Michael Fluss (LLNL), co-Chair
- Richard LeSar (ISU and Ames Laboratory), co-Chair
- Vasily Bulatov (LLNL)
- Tim Germann (LANL)
- Turab Lookman (LANL)
- Sidney Yip (MIT)
- Jim Belak (LLNL)
- Ed Kober (LANL)
- Kimberly Budil (DOE)
- Arthur Kerman (MIT/UT/ORNL)

Panel 7 Members

- Tony Rollett (CMU), Chair
- Marius Stan (LANL), co-Chair
- Alfredo Caro (LLNL)
- Brent Adams (BYU)
- Surya Kalidindi (Drexel)
- Irene Beyerlein (LANL)
- Alan Patterson (LANL)
- Tony Redondo (LANL)
- Wendy Cieslak (LANL)

Landau Ratio

as amended by Bulatov & Kerman

$$\text{Landau_ratio} = \frac{\text{No. Independent Points Matched by Model}}{\text{No. Adjustable Parameters} + \text{Adjustable Assumptions}}$$

Scope of Each Panel

- (P7) Bringing about the transition from (empirical) observation to (quantitative) control will require substantial developments in characterization, analysis, modeling, computation and design of (critical) experiments. Successful examples are available but materials under extreme environments pose challenges.
- (P8) The frontier of multi-scale modeling will likely be achieved by creating an experimentally-validated framework that spans spatial and temporal scales through an integrated approach that moves beyond the passing of parameters between distinct models at distinct scales.

Priority Research Directions

1. Materials Complexity
2. Bridging between Length & Time Scales
3. Interface between Computation & Experiment
4. Materials Design and Lifetime Prediction

PRD: *Materials Complexity*

Scientific Challenge

- Heterogeneity-dependent variability in material response, more so under extremes of loading.
- Size dependence (microstructure, sample size) based on weakest link statistics.
- Time dependence (scatter increases at both ends, creep and in shock)

Capability Gap

See following charts

Research Direction

See following charts

Potential Impact

- Quantification of reliability based on validated theoretical models that fully capture the heterogeneous character of materials and handle realistic boundary conditions and gradients.
- Statistically-based definition of properties and performance based on materials history.
- Size & Time Scaling Laws, e.g. how fast 'strength' decreases as sample size increases, or as microstructural length scales (grain size) increases, e.g. how failure times in the laboratory correlate to failure times in service.

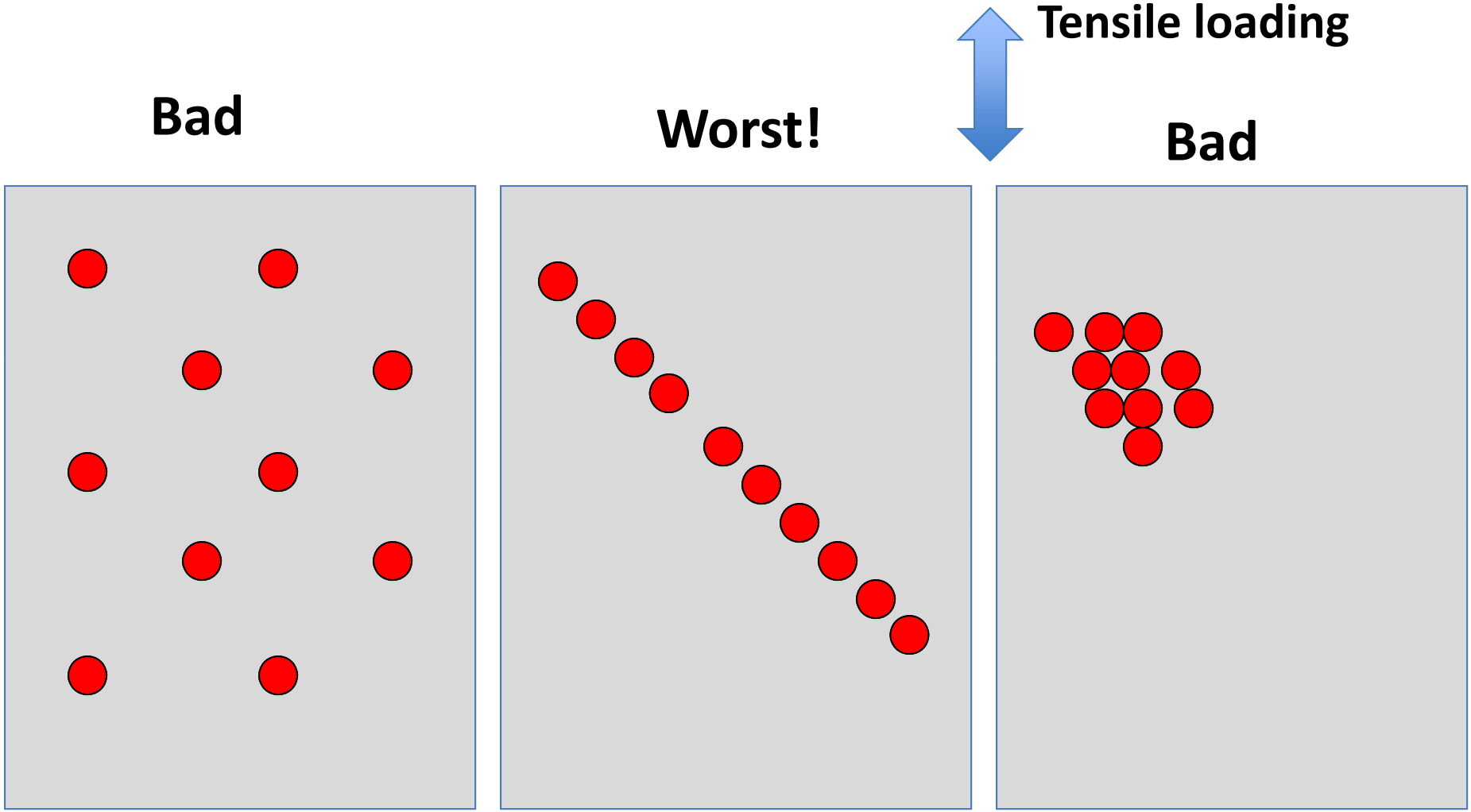
Materials Complexity: Capability Gaps

- **Lack methods to accurately model dynamic situations in which the electrons and the ions are not in equilibrium, e.g. ion bombardment in radiation damage**
- **Lack probabilistic theories of material failure that relate the 3D statistical distribution of defects to the statistical distribution in material performance, e.g. RVEs vs. SVEs.**
- **Lack ability to accurately model multicomponent systems, e.g. phase relationships and diffusion**
- **Lack robust approaches to quantify the uncertainties of materials properties, e.g. thermal properties**

Materials Complexity: Research Directions

- **Combine experimental, numerical, and theoretical efforts towards developing theory for improbable events, e.g. defining tails of distributions.**
- **Develop new approaches to fast retrieval of higher-order moments of structure and response fields, e.g. analysis of synchrotron experiments.**
- **Develop accurate Hamiltonians for multi-component materials, e.g. diffusion in a multi-component alloy**
- **Develop instantiation of actual microstructures in simulations to increase accuracy, e.g. deformation of polycrystalline materials**
- **Incorporate defect-driven variability explicitly within simulation framework, e.g. effects of defect distribution on shock response**
- **Develop simulations that include quantified errors and uncertainties across scales, e.g. how dislocation substructure affects response**

Materials Complexity: *Spatial Arrangement*



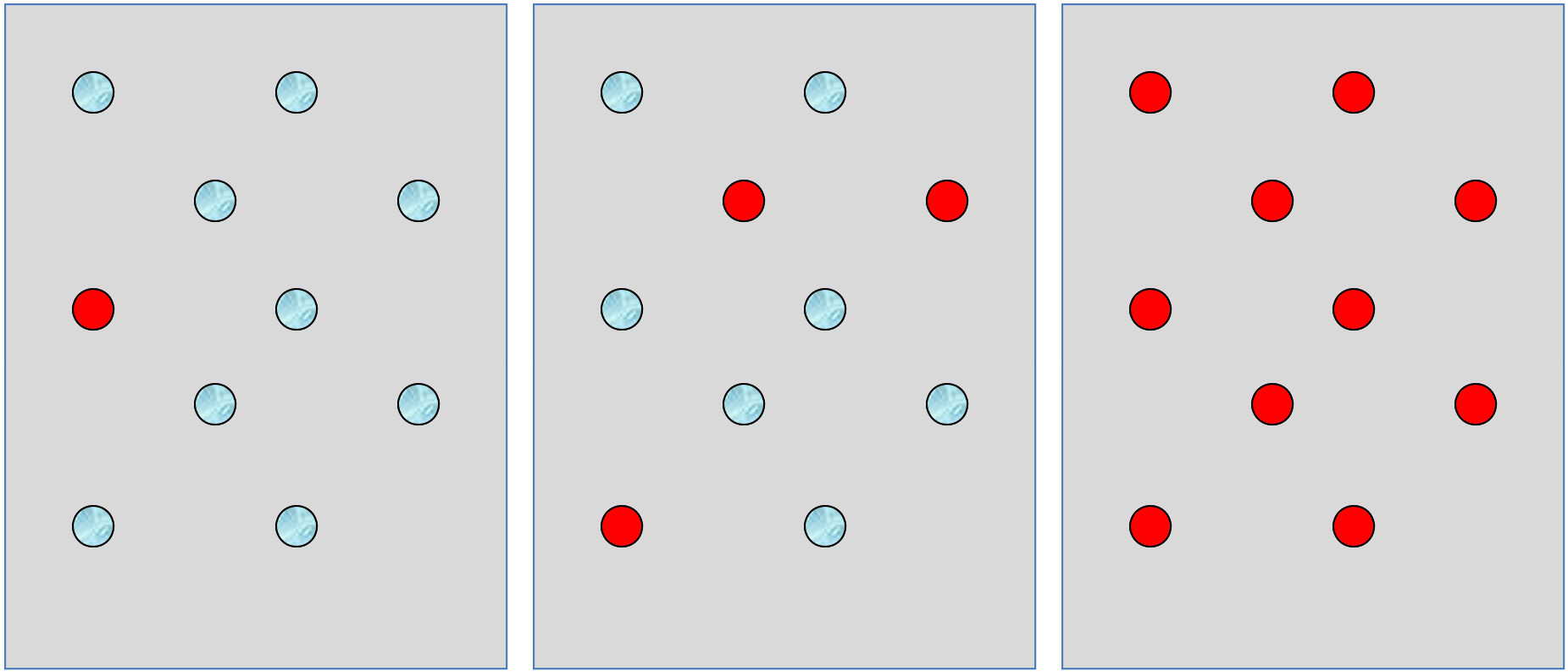
Same volume fraction of voids, different distribution, very different outcomes!

Materials Complexity:
Same Arrangement, Different Loadings

Low stress
e.g. high cycle fatigue

Medium stress

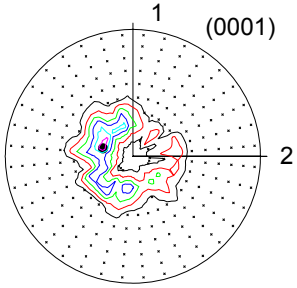
High stress
e.g. low cycle fatigue



Same distribution of latent defects, very different outcomes as a function of loading!

High strain-rate deformation of clock-rolled Zr (*)

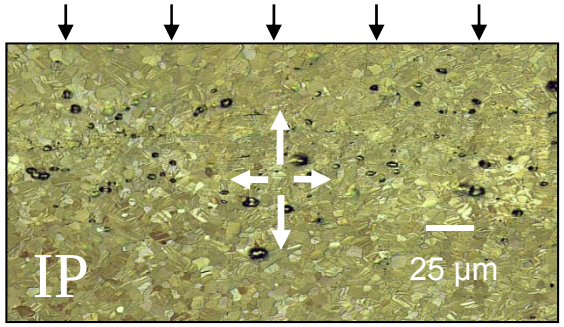
Clock-rolled Zr

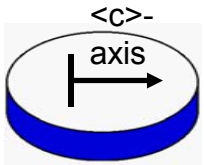


Spallation of an IP sample:

tension along a softer PX direction (<a>-axes predominant), prismatic slip (higher rate-sensitivity)

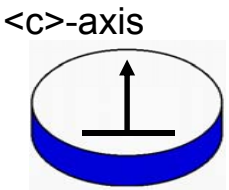
slower porosity evolution

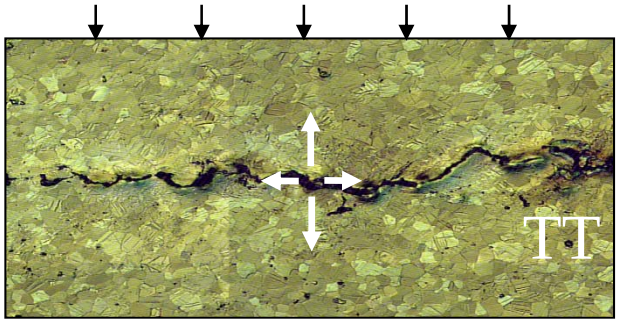




Slide courtesy of Ricardo Lebensohn

Gray et al., Shock Compression Cond. Matter, 1999





faster porosity evolution

Spallation of a TT sample:

tension along a harder PX direction (<c>-axes predominant), twinning (lower rate-sensitivity)

(*) Dynamic PropertiesTeam, MST-8, LANL

PRD: *Bridging Length & Time Scales*

Scientific Challenge

- Need new tools for analyses of large datasets at multiple spatial and temporal scales.
- Development of a new generation of data-mining tools based on established knowledge in DSP and SE.

Capability Gap

See following charts

Research Direction

See following charts

Potential Impact

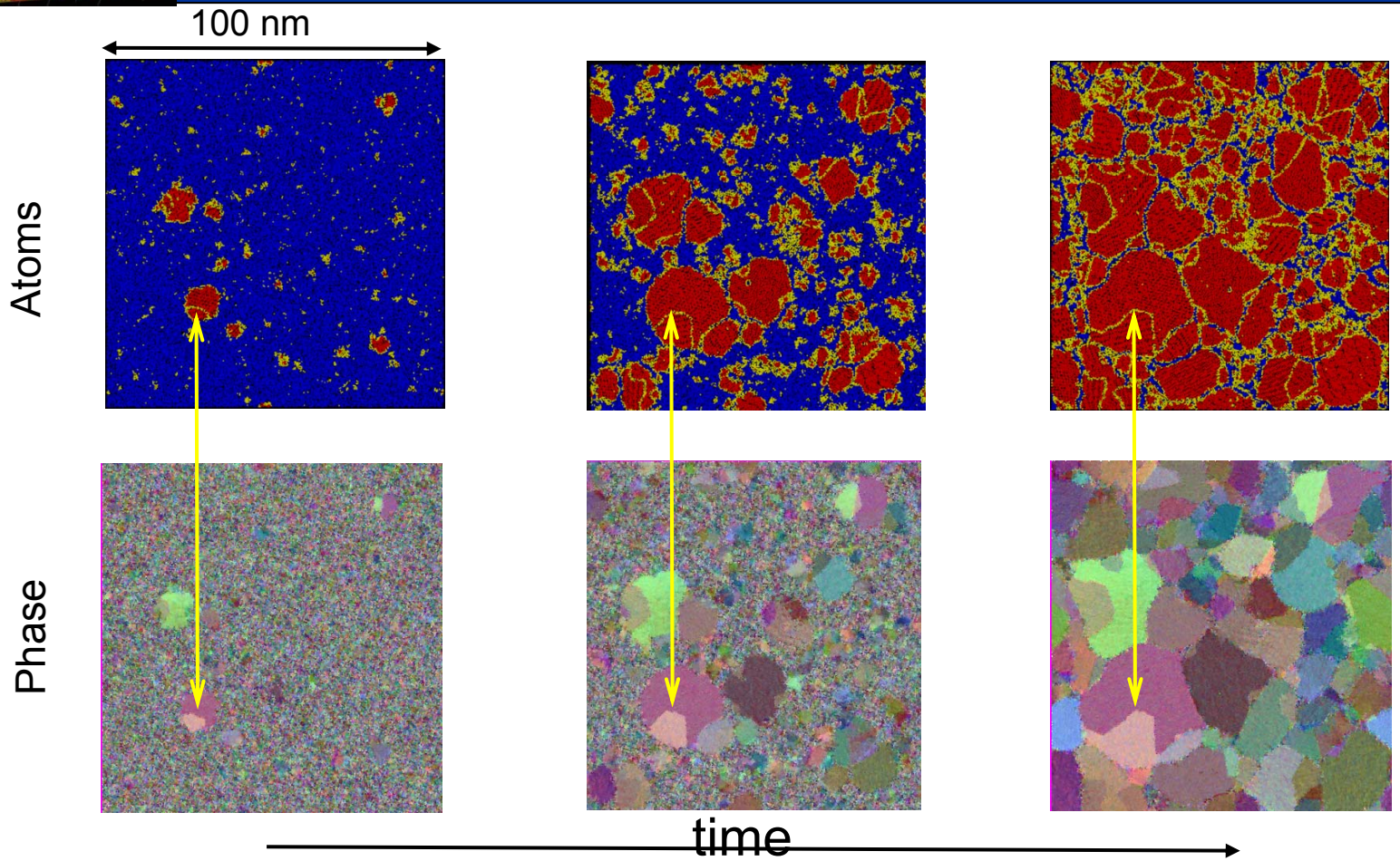
- Will allow the transformation from observation to prediction and control of extreme-valued properties.
- Will allow robust extraction of underlying physical principles at the mesoscale.
- Will bring a mathematically grounded approach to materials science & eng.

Bridging Length & Time Scales: Capability Gaps

- **Lack theoretical and computational approaches to bridge the length and time scale disconnect between atomistic unit mechanisms and mesoscale dynamics, e.g. radiation damage**
- **Lack of atomistic methods at long times scales, e.g. glasses, aging, creep, …**
- **Lack of methods for inverse mapping from larger to smaller scales, e.g. connecting phase-field to atomistics**
- **Lack fundamental understanding of threshold phenomena, e.g. lack of treatment of competing mechanisms, e.g. breaks in passive oxide films.**
- **Lack ability to couple properties at the mesoscale with the underlying (dynamic) electronic structure, e.g. the 'electronic barrier': classical approaches do not fully capture quantum effects.**
- **Lack of algorithms for exascale computing, e.g. programming for strongly heterogeneous clusters**

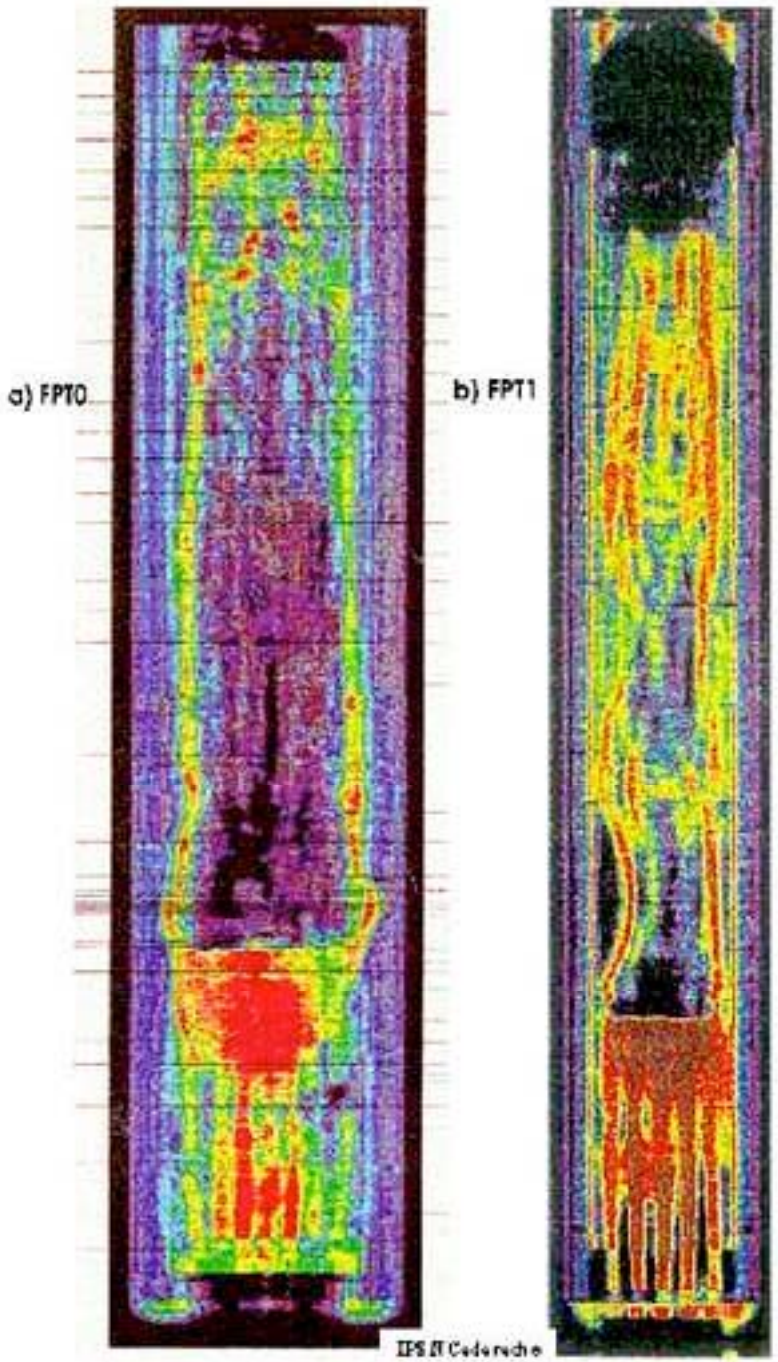
Bridging Length & Time Scales: Research Directions

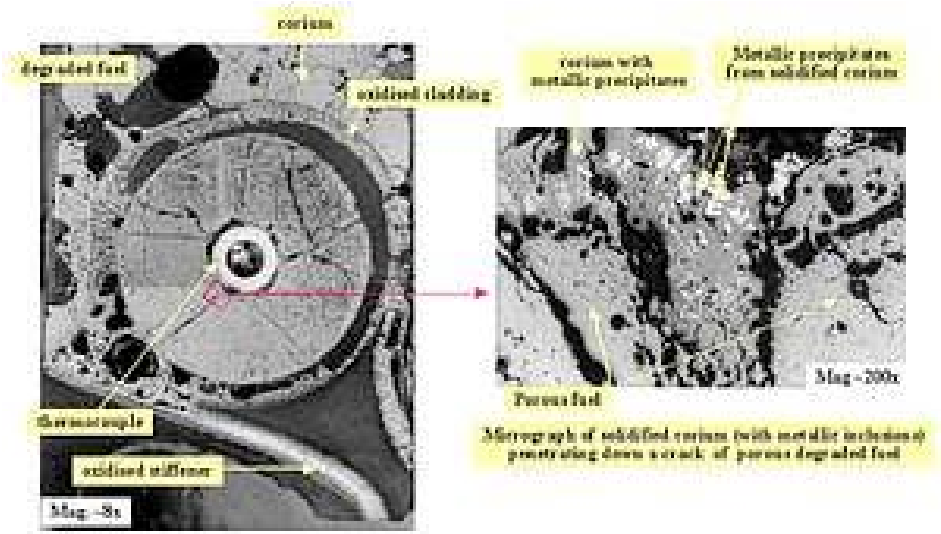
- **Develop strategies for bridging length and time scales, e.g. close the gap between moment analysis and percolation analysis.**
- **Develop and characterize infrequent-event sampling algorithms for extending length and time scales, e.g. hyperMD, metadynamics.**
- **Develop statistically reliable accelerated algorithms for cross-scale simulations from picoseconds to years, e.g. materials aging, corrosion.**
- **Develop methodologies for tracking uncertainties in simulations across scales, e.g. nucleation phenomena**
- **Exploit Information Science to do optimal multiscaling: adaptive model refinement and self-learning algorithms, e.g. incorporating microstructure-based simulations into engineering modeling systems.**



Validate micro-physics within Phase Field Model with MD simulations that overlap in time and space

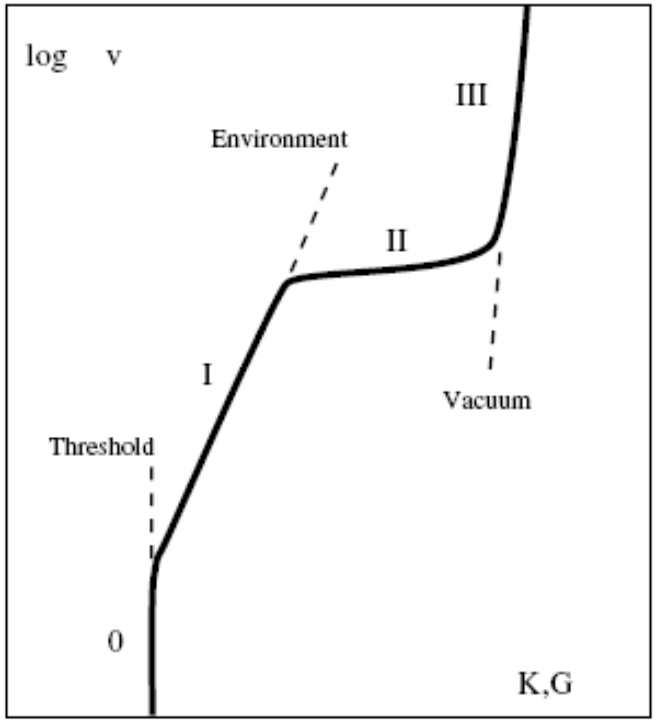
Burnt-up Nuclear Fuel



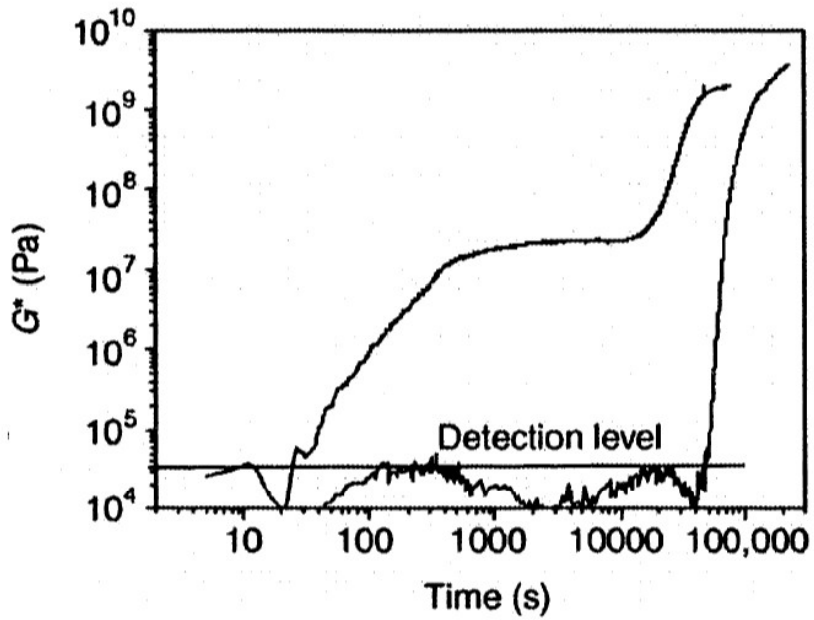


Courtesy of M. Fluss

Molecular mechanisms on long time scales



Stress corrosion cracking (b) [Ciccotti, 2009]



Rheology of cement

Courtesy of S. Yip

PRD 3: *Interface between Experiments & Simulation*

Scientific Challenge

- Optimize the assumptions in models, especially in design activities
- Develop a framework to discover and integrate knowledge from experimental and modeling datasets.

Research Direction

See following charts

Capability Gap

See following charts

Potential Impact

- Create predictive capabilities to control properties (free energy, chemical species mobility, thermal/electrical conductivity) of heterogeneous materials that operate in extreme conditions (fusion/fission reactors, chemical reactors, weapons, etc).
- Enable the design of materials for national security and energy applications.

Interface between Experiments & Simulation: Capability Gaps

- **General lack of theory and models to bridge between experiments and simulations.**
- **Lack of quantitative methodology for validation, for comparison of simulations with experiments.**
- **The 'thermal barrier': current techniques do not describe realistic free energy landscapes, e.g. for off-stoichiometric materials.**
- **The 'empirical barrier': Current simulations often involve empirical models that cannot be extended to extreme radiation, temperature, or pressure.**
- **"Brute force" experiments and simulations are prohibitively expensive for defining tails of distributions.**
- **Calculated or measured performance of "Real Systems" must account for interfaces, free surfaces (edge effects), high gradients in drive stimulus, etc.**
- **Need for in-situ, real-time, 3D experimental microstructural quantification, including representation and instantiation of simulations.**
- **A lack of interface between integral tests (experiments) and physics-specific models**
- **Lack adequate theories at the scale of experiments and engineering**

Interface between Experiments & Simulation: Research Directions

- **Combine techniques such as computed tomography and diffraction, with simulation of processes e.g. nucleation of damage.**
- **Develop joint efforts between experimentalists and modelers and common frameworks for exchange of information towards a common goal of establishing high fidelity predictive capabilities, e.g. dynamic failure.**
- **Develop simulation approaches with accurate representation of materials, e.g. 3D microstructures.**
- **Design science-driven experiments to test predictive simulations of specific mechanisms and cross-over state variables, e.g. stress, temperature, pressure, ...**
- **Establish active feedback between simulations and experiment in materials design, e.g. mitigation of radiation damage in ODS steels or laminated composites.**
- **Develop simulations and acceleration experiments that account for detailed mechanisms, e.g. radiation damage**

Thermal Properties in Fuels

Heterogeneous materials consist of multiple homogenous phases with specific properties. The global (effective) properties of heterogeneous materials are currently related to the local properties of their phases via empirical models. Over the next decade it is expected that the complexity (and heterogeneity) of materials operating in extreme conditions can only increase. We need a theory and predictive computational tools to address the challenge.

Example: Radiation-induced changes in UO_2 nuclear fuel microstructure continunously decrease the effective thermal conductivity, K_{eff}, slowing down the removal of heat and leading to overheating of the fuel element[1].

-Heterogeneous characteristics (Fig. 1) include: multiple phases (bulk, void, gas bubbles), different interface types, and non-uniform dislocations and cracks.

-K_{eff} depends on the space and time distributions of the grain (and pore) size, shape, and orientation.

-Predicting K_{eff} as function of burnup (time) would have tremendous impact on improving fuel performance and design.

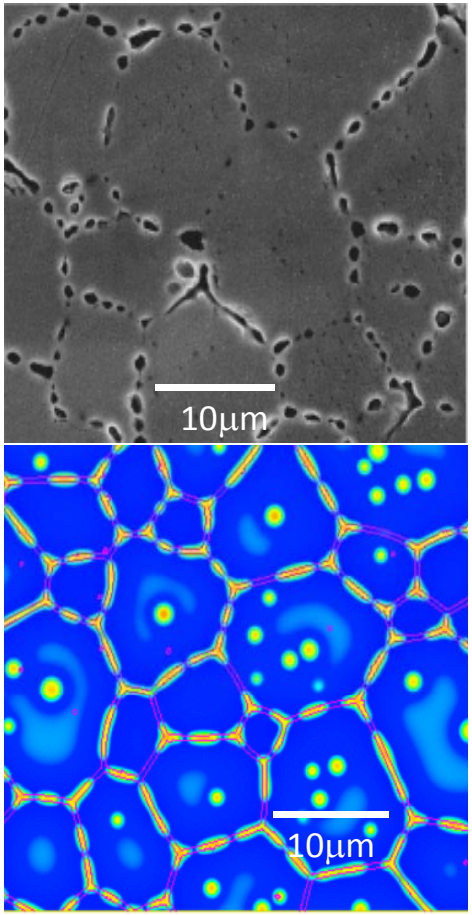


Fig. 1. *Top*: UO_2 fuel irradiated in PWR[2] *Bottom*: Phase Field simulation of UO_2 microstructure evolution[3]

[1]M. Stan, J. Nucl. Eng. Technology, **41** (2009) 39-52.
[2]I. Zacharie *et. al.,* J. Nucl. Mater. **255** (1998), 92-104.
[3]S.Y. Hu et al., J. Nucl. Mater. **392** (2009) 292–300.

PRD 4: *Materials Design and Performance / Lifetime Prediction*

Scientific Challenge

- Science-based engineering of materials; founded on scientific mechanisms as opposed to empirical relationships.
- Understanding of the fully integrated relationships processing / structure / properties / performance
- Model-assisted accelerated qualification and testing for performance through life, e.g. AIM.

Capability Gap

- Requires a change in education philosophy to integrate science and engineering of materials
- Accelerated discovery of controlling mechanisms.
- Ability to quantify the full spectrum of the *dynamic* statistical distribution of the structure and properties with the necessary spatial and temporal resolution.
- Predictive capability for inverting information flow from performance to structure to processing
- Novel synthesis tools to access predicted optimal microstructure.

Research Direction

Materials Design

- Couple experimental identification of critical controlling mechanisms with models optimizing processing, structure, properties, performance
- Integral validation / calibration throughout experimental and modeling iterations
- Quantified uncertainties

Materials Lifetime Prediction

- Deterministic mechanisms
- Statistical microstructures
- Probabilistic properties/performance
- Path dependency

Potential Impact

- Materials creation with tailored performance and accelerated transition into application.
- Confident application of materials outside of existing design allowables, e.g. life extension, new operating conditions.

Materials Design:

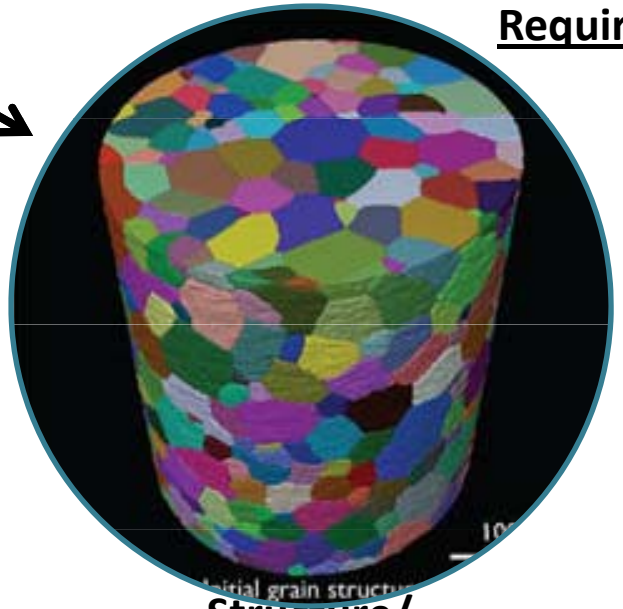
In Situ techniques to probe structure and properties at sufficient temporal and spatial resolution to quantify controlling mechanism

Performance Meets Requirements or Modify

Materials/Process Specification

Design Performance Requirements

Materials Synthesis Models



FEM models

Processing

Primary

Secondary

Fabrication

Material Properties

Mechanical

Physical

Chemical

Structure/ Composition

Architecture

Microstructure

Nanostructure

Process/Structure Models

Structure/Property Models

Image courtesy of E. Lauridsen

Cross Cutting Themes

- Large data sets (drives informatics).
- Rare events and extreme value statistics
- Uncertainty quantification
- Real-time, in-situ observation
- Active feedback in both directions between experiment and simulation, with quantitative figures of merit

Meta-Topics

- Education of the broader community about materials

Final Thoughts