LA-UR-15-21334
Approved for public release; distribution is unlimited.

Title: MaRIE 1.0: The Matter-Radiation Interactions in Extremes Project, and the Challenge of Dynamic Mesoscale Imaging

Author(s):
Barnes, Cris William
Barber, John L.
Kober, Edward Martin
Lookman, Turab
Sandberg, Richard L.
Shlachter, Jack S.
Sheffield, Richard L.

Intended for: 11th LANSCE Neutron School, 2015-02-19/2015-02-27 (Los Alamos, New Mexico, United States)

Issued: 2015-02-23

MaRIE 1.0: The Matter-Radiation Interactions in Extremes Project, and the Challenge of Dynamic Mesoscale Imaging

Cris W. Barnes, John L. Barber, Ed Kober,
Turab Lookman, Richard Sandberg,
Jack Shlachter, Richard Sheffield

February 26, 2015

Abstract

The Matter-Radiation Interactions in Extremes project will build the experimental facility for the time-dependent control of dynamic material performance. An x-ray free electron laser at up to 42-keV fundamental energy and with photon pulses down to sub-nanosecond spacing, MaRIE 1.0 is designed to meet the challenges of time-dependent mesoscale materials science. Those challenges will be outlined, the techniques of coherent diffractive imaging and dynamic polycrystalline diffraction described, and the resulting requirements defined for a coherent x-ray source. The talk concludes with the role of the MaRIE project and science in the future.

MaRIE is the experimental facility for the time-dependent control of dynamic material performance

The Mission Need

Develop flexible and low-cost product-based solutions to materials problems in the stockpile through accelerated qualification, certification, and assessment.

The Challenge

Interfaces, defects, and microstructure between atomic scale and engineering continuum determine time-dependent material properties.

What It Is: The Capability Gap

MaRIE fills the gap with:

- 42-keV XFEL for coherent, brilliant, repetitive x-rays; and
- Multiple simultaneous probes (x-ray, proton, electron, optical); and
- Close linkage to synthesis, fabrication and characterization facility; and
- Full integration with advanced theory, modeling, and computing.

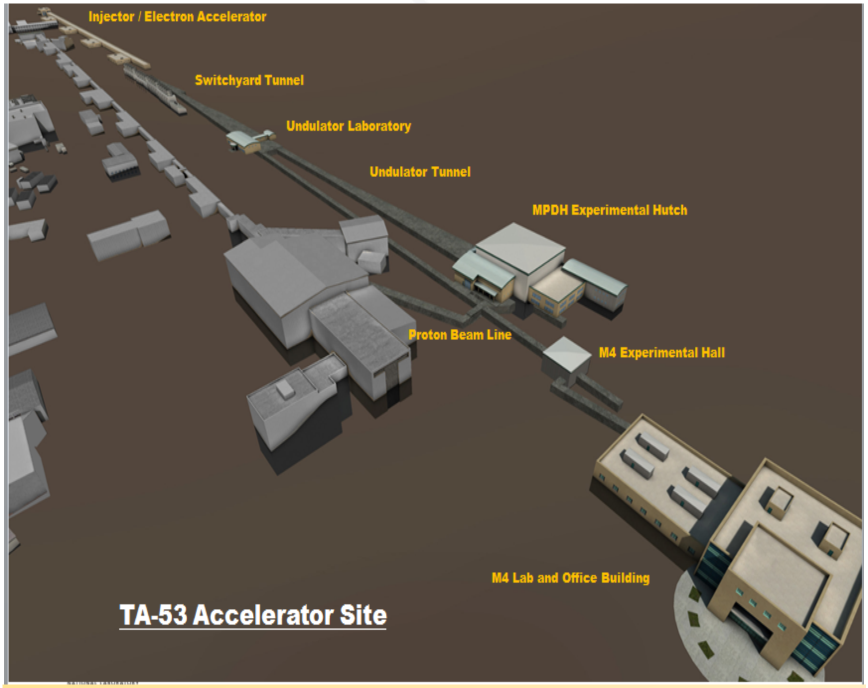
How It's Done

After time-dependent control of process, structure, and properties during synthesis of materials at the mesoscale, MaRIE will subject them to extreme environments and use both imaging and diffractive scattering to connect to the product performance.

MaRIE responds to the NNSA Tri-lab Facility Roadmap Gap of
"predict and control from materials and devices to manufacturing processes"

Production → Button → Boom → Disposition

- MaRIE will provide the world's first very-hard (42-keV) **XFEL** with GHz (few pulses) repetition;
- A **Making, Measuring, and Modeling Materials (M4) Facility** for materials synthesis and characterization with high-performance computational co-design focused on the mesoscale; and
- A **Multi-Probe Diagnostic Hall (MPDH)** coupling hard, coherent, brilliant x-ray photons with 12-GeV electron and 0.8-GeV proton radiographic tools in time-dependent extremes**.**



MaRIE facility definition derives from "First Experiments" functional requirements and identified performance gaps.

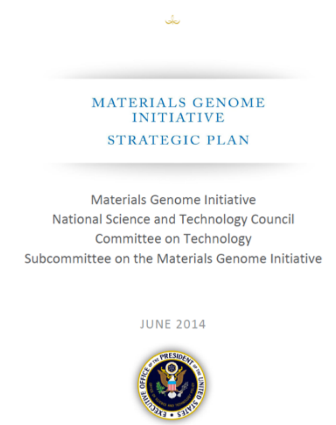
LANL-led efforts with the Scientific Community ~2006-2012 defined the Grand Challenges

Requirements:
Sub-μm space resolution
100's – 1000's-μm samples
Sub-ns time resolution,
~30 frames in
1−10-μs duration

meso2012.com

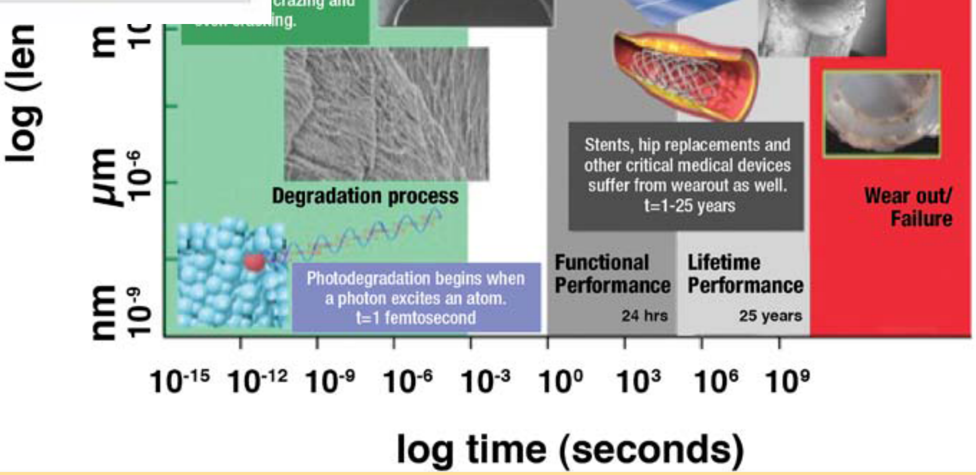
science.energy.gov

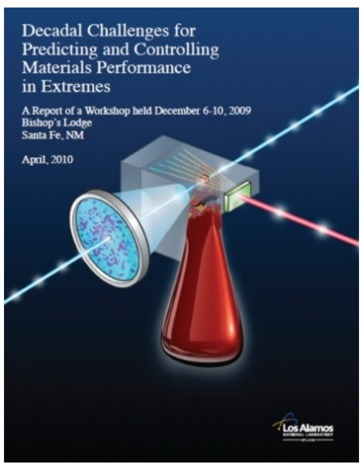
Materials Genome

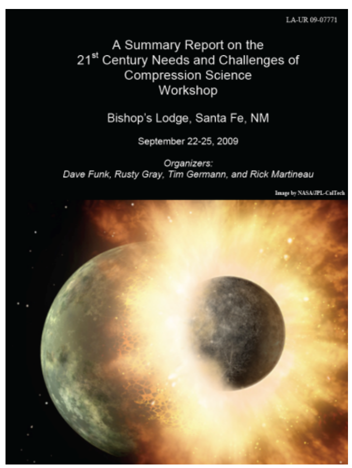


Advanced Manufacturing

Whitehouse.gov

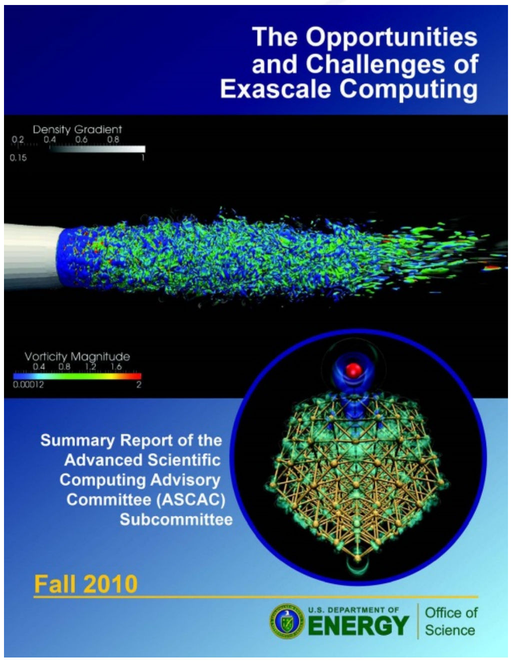


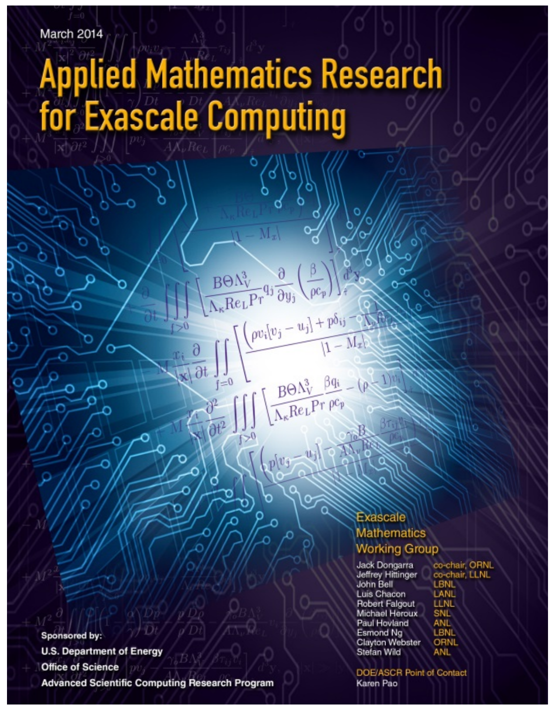


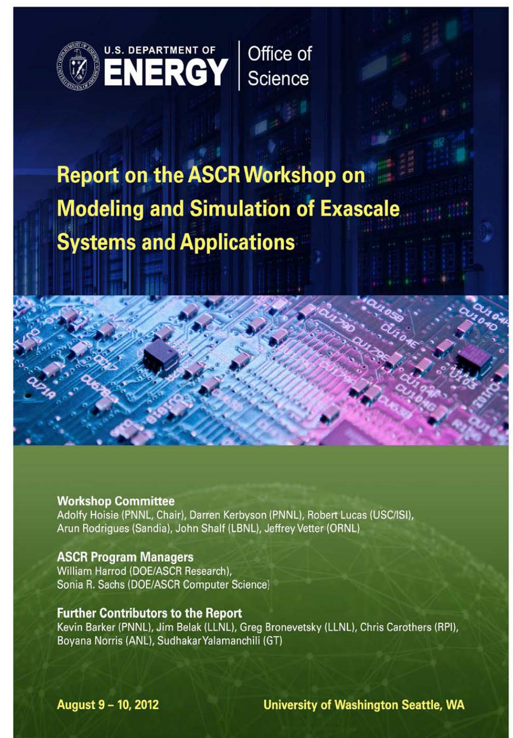


The challenge is to observe the dynamic evolution of polycrystalline materials at the granular and inter-granular level (the mesoscale)

MaRIE will be operating in an environment of exascale computing and future codes

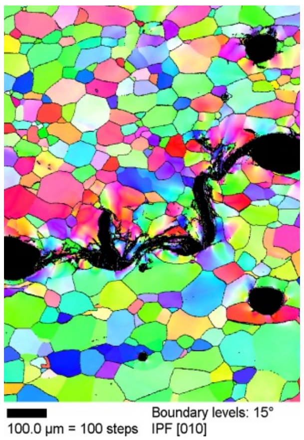


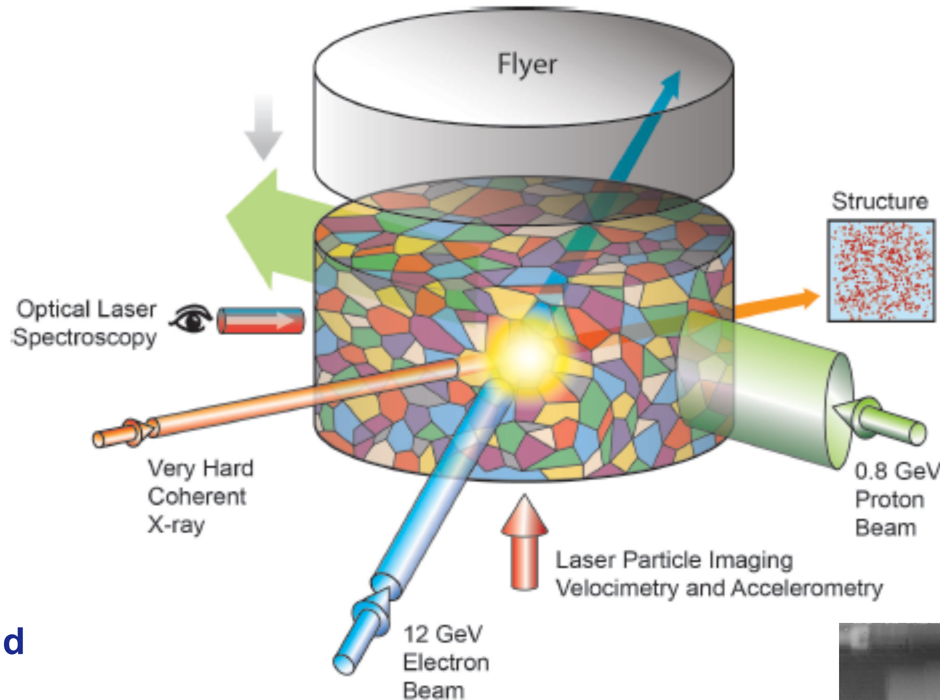




Experimental Facilities are needed to Discover mechanisms, Validate and Calibrate sub-grid models

The challenge is to observe the dynamic microstructure and phase evolution in materials down to the sub-granular level while connecting to the macroscale





Requirements:
Sub-µm space resolution
100's – 1000's-µm samples
Sub-ns time resolution,
~30 frames in
1−10-µs duration

The model:

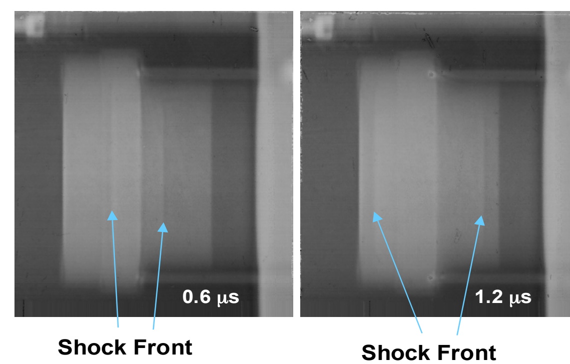
Accurate sub-grain models of microstructure evolution coupled to molecular dynamics

The goal

Predict dynamic microstructure and damage evolution

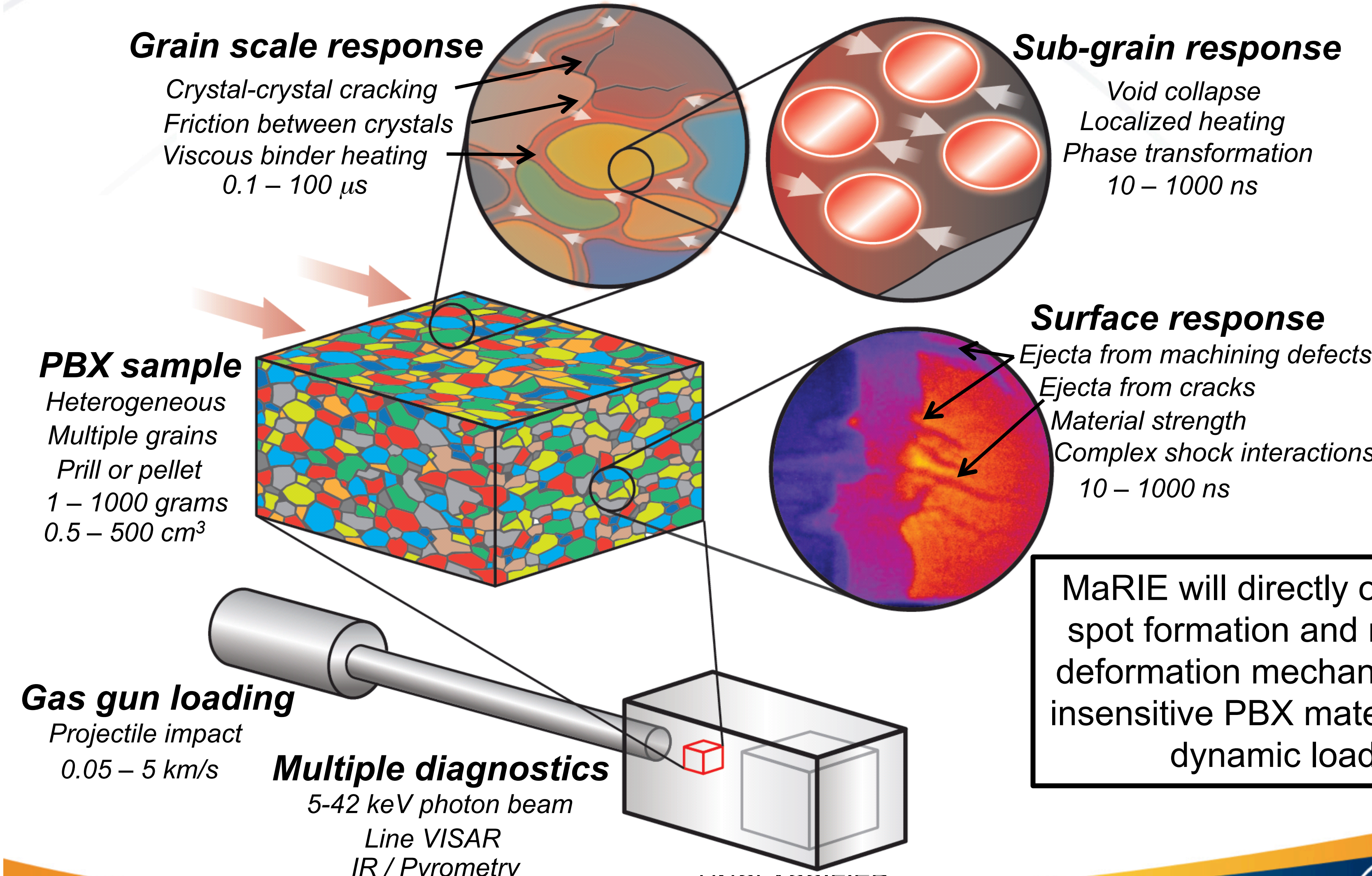
The first experiment

Multiple, simultaneous dynamic *in situ* diagnostics with resolution at the scale of nucleation sites (< 1 µm; ps – ns)



MaRIE allows us to break apart the problem

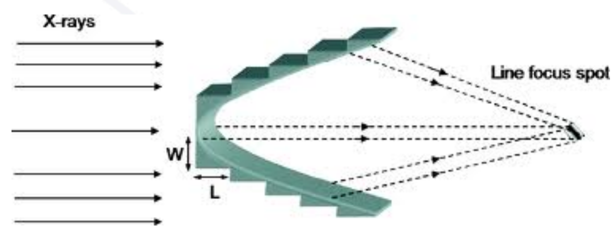
MaRIE allows *in situ* study of hot spot formation and other microstructural phenomena



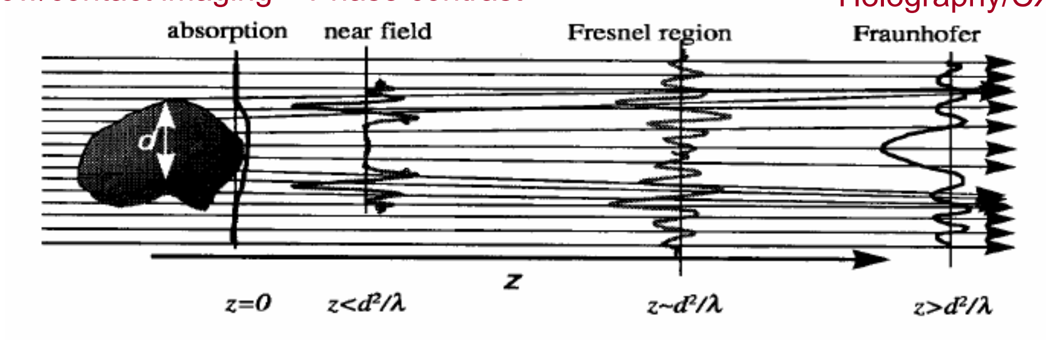
MaRIE will directly observe hot spot formation and mesoscale deformation mechanisms within insensitive PBX materials during dynamic loading.

X-ray Imaging Techniques Can Give Sub-micron Resolution

- **Traditional Imaging (think lens)**
  - Fresnel, Kirz, Laue, Kinoform lenses for hard x-rays (up to 0.5 MeV)
  - Resolution: down to ~100's nms



- **Radiography Approaches (Line-projection, tomography, PCI and divergent beam PCI)**
  - Resolution typically scales with pixel size (max r ~ pixel/100 > 100 nm, typically 1 μm)
- **Coherent Approaches (Holography, PCI, CXDI)**
  - Resolution: down to <10 nm

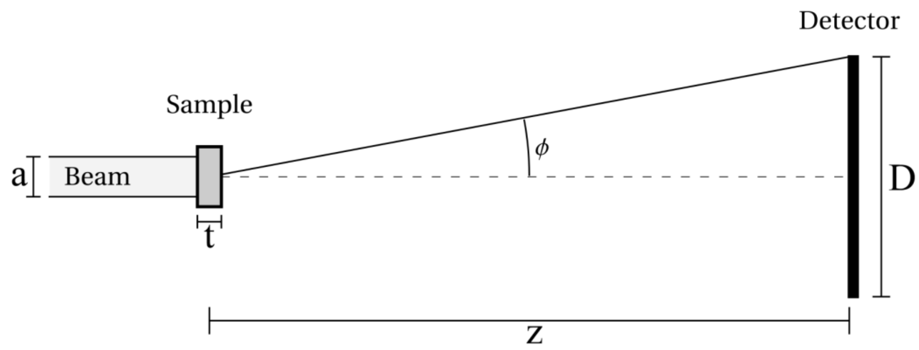


CXDI can provide 3D information, but has only been demonstrated for softer x-rays (<1 keV) and small samples (few micron)

Scattering & Diffraction Theory

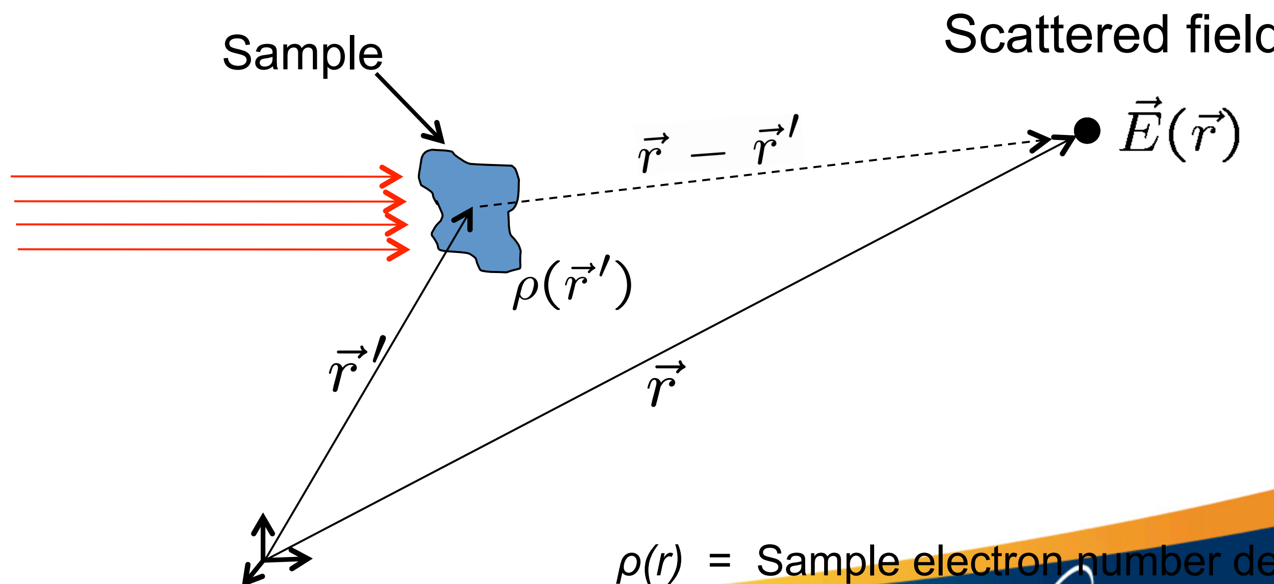
Schematic of a basic experimental imaging setup:

a = Spot/sample size
t = Sample thickness
z = Sample-to-detector distance
D = N_p p = Detector size
N_p = # of detector pixels
p = Detector pixel size



Incident coherent beam

$$\vec{E}_o(\vec{r}\,') = \hat{\varepsilon}\, E_o\, e^{i\vec{k}_i \cdot \vec{r}\,'}$$



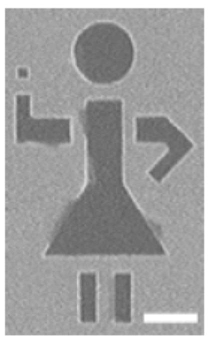
$\rho(r)$ = Sample electron number dens

The Far Field

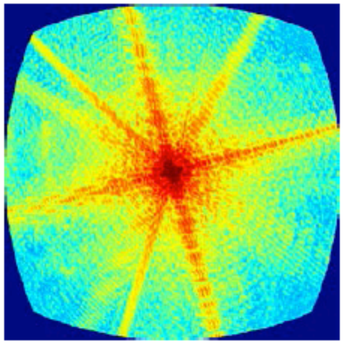
Define the Fresnel number, $Fr \equiv \dfrac{a^2}{\lambda z}$. When $Fr \ll 1$:

$$\vec{E}(\vec{r}) \propto \int d\vec{r}\,'\rho(\vec{r}\,')\,\mathrm{e}^{-i\vec{q}\cdot\vec{r}\,'} = \mathrm{FT}\,\{\rho\}$$

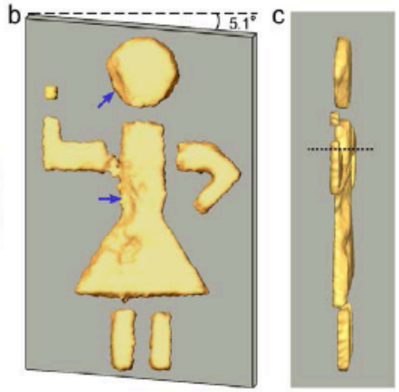
where $\vec{q} = \vec{k}_f - \vec{k}_i = k\hat{r} - \vec{k}_i$ is the *scattering vector*. Since $\vec{q}$ is a 3D vector, 3D imaging is possible in the far field*:



Sample (SEM image)



Scattering Pattern



Reconstruction

For "MaRIE-like" imaging problems, achieving $Fr \ll 1$ requires prohibitively-large values of z (100s to 1000s of meters) due to the high photon energies (and low wave-lengths λ) required to penetrate a sample of dense material.

* "Three-dimensional structure determination from single view," Raines, et al. *Nature* **463**, 214-217 (2010).

The Fresnel Regime

Define the "Small angle number" $An \equiv \dfrac{D^4}{8\,\lambda\, z^3}$. When $An << 1$:

$$\vec{E}(\vec{r}) \propto \int d\vec{r}\,'\rho(\vec{r}\,')\,\mathrm{e}^{i\frac{k|\vec{r}\,'|^2}{2z}}\,\mathrm{e}^{-i\vec{K}\cdot\vec{r}\,'} = \mathrm{FT}\left\{\rho\,\mathrm{e}^{i\frac{k|\vec{r}\,'|^2}{2z}}\right\}$$

where $\vec{K} = k\frac{\vec{r}}{z} - \vec{k}_i$.

It can be shown that $\vec{K}$ is a purely two-dimensional vector, i.e. it lies on a 2D manifold. This means that in the Fresnel regime, the scattered and diffracted field encodes only 2D information about the sample, and **3D imaging is not possible in the usual experimental setup***.

Nevertheless, the condition $An << 1$ is often much less stringent than the far-field condition $Fr << 1$. For fixed detector size $D \sim 2.8$ cm (a typical value), values for z of 10s of meters to 100 meters are sufficient to ensure that $An << 1$.

* Single shot, *in situ*, etc.

Coherent X-ray Diffractive Imaging

Detector preserves intensity – but not phase – of scattered field:

$$I(\vec{r}) \;\propto\; \left|\vec{E}(\vec{r})\right|^2 \;\propto\; \left|\int d\vec{r}\,'\,\rho(\vec{r}\,')\,\mathrm{e}^{-i\vec{q}\cdot\vec{r}\,'}\right|^2 \;=\; \left|\mathrm{FT}\{\rho\}\right|^2 \;=\; \left|\hat{\rho}(\vec{q})\right|^2$$

Provided additional information about the support of the sample/beam intersection is available in the form of an oversampling condition, these missing phases can be recovered via a process of *iterative phase retrieval* (Fienup 1978).

<u>Transverse imaging resolution:</u>

$$\frac{\Delta x}{\lambda} = \frac{1}{D}\sqrt{\left(\frac{D}{2}\right)^2 + z^2} \approx \frac{z}{D}$$

Far-field

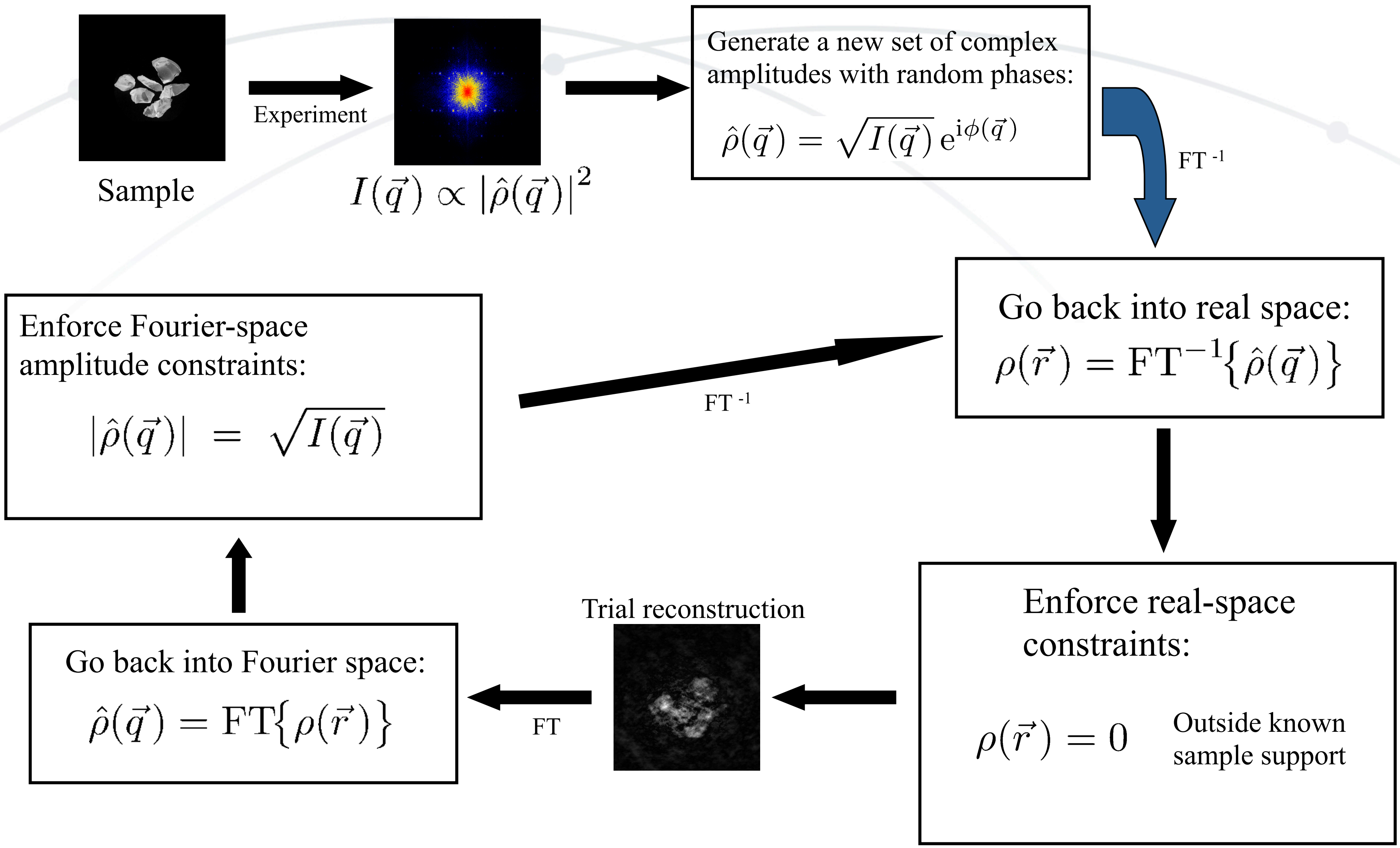
$$\frac{\Delta x}{\lambda} = \frac{z}{D}$$ Fresnel

<u>Longitudinal imaging resolution:</u>

Far-field

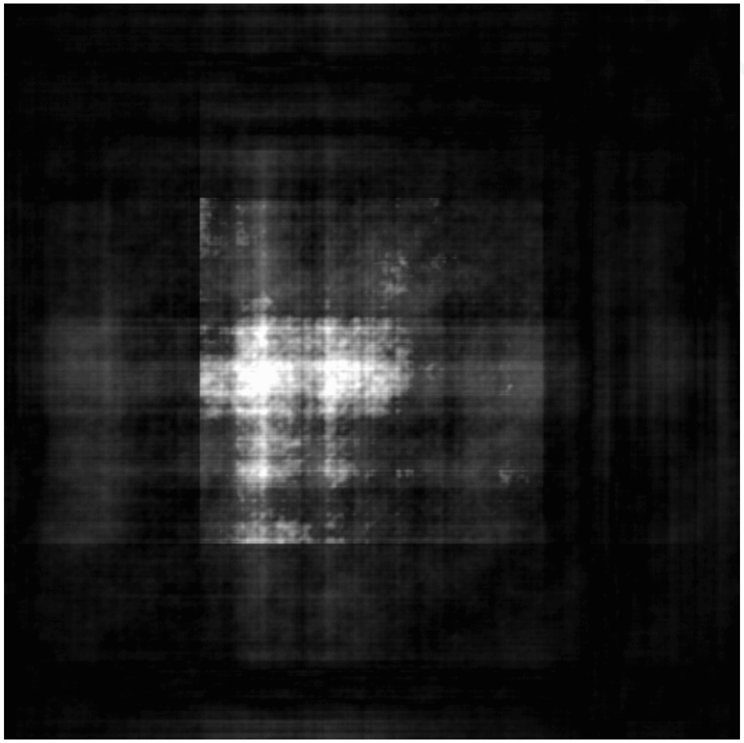
$$\frac{\Delta z}{\lambda} = \frac{1}{2}\left(1 - \frac{z}{\sqrt{\left(\frac{D}{2}\right)^2 + z^2}}\right)^{-1} \approx \frac{4z^2}{D^2}$$

$$\frac{\Delta z}{\lambda} = \infty$$ Fresnel



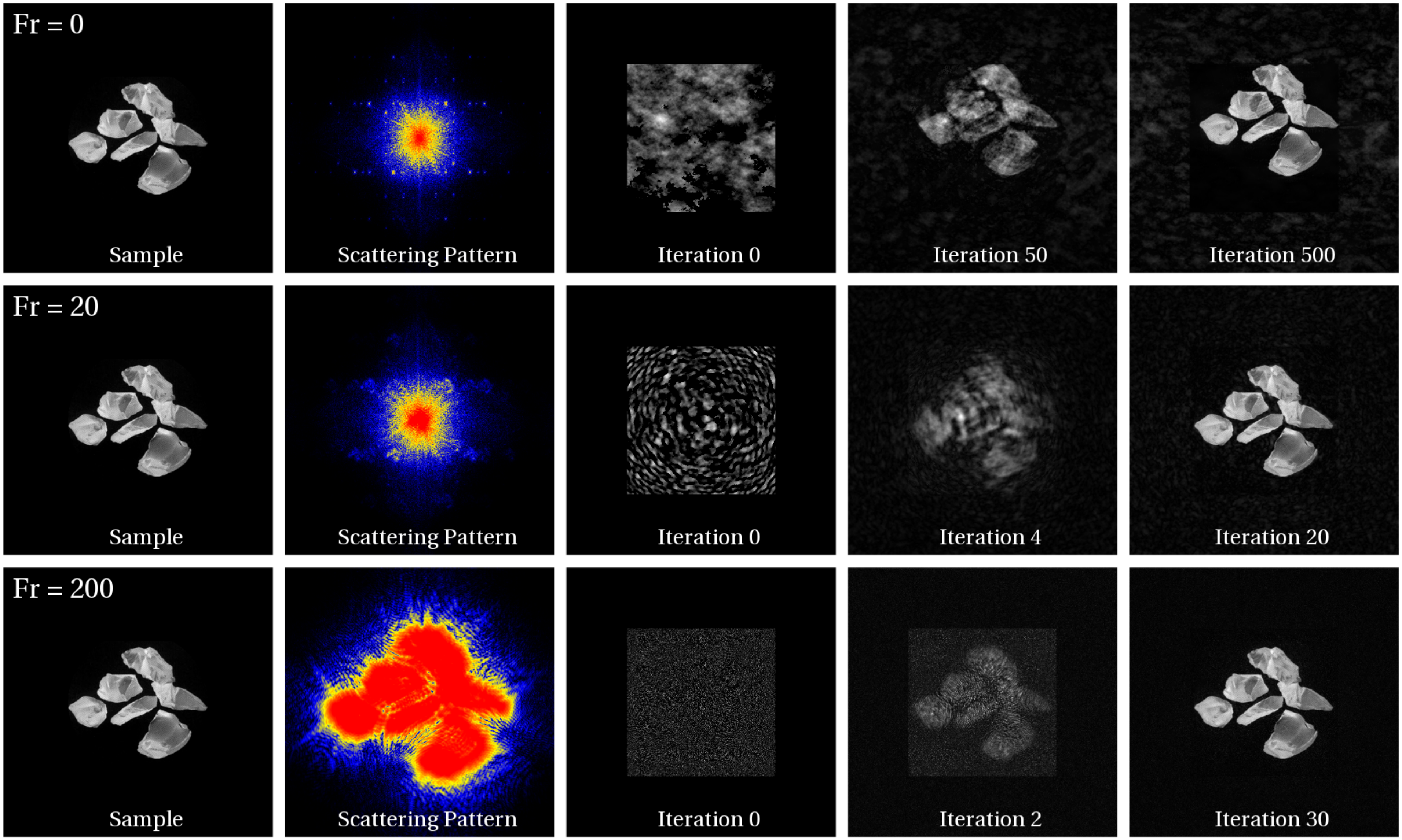
As shown here, this assumes the far field. However the same procedure works with slight modification in the Fresnel regime (Xiao & Shen 2005).

Example of iterative phase retrieval

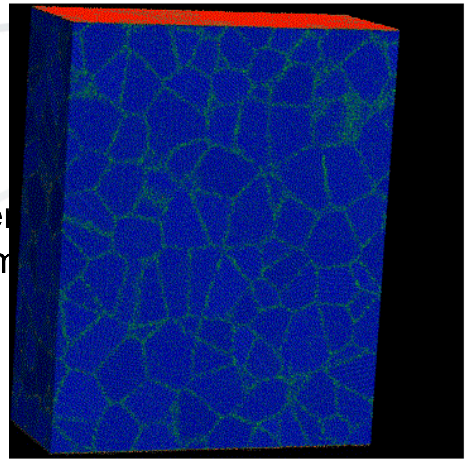


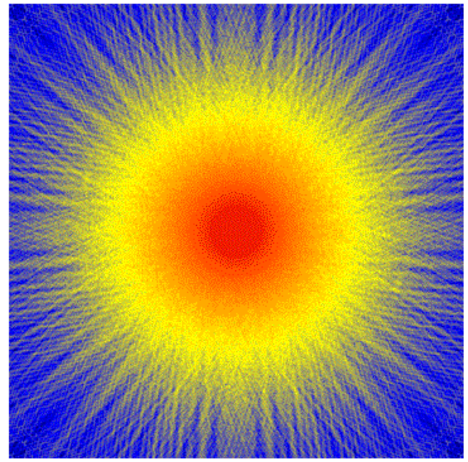
What do you think the image is?

Simulated CXDI Examples:



Simulated CXDI on a 2D slice of an MD copper polycrystal. Top hat beam profile. *Fr* ~ 4:


MD Polycrystal


Diffraction Pattern

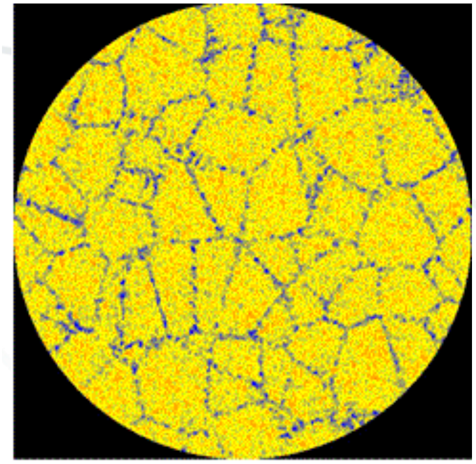
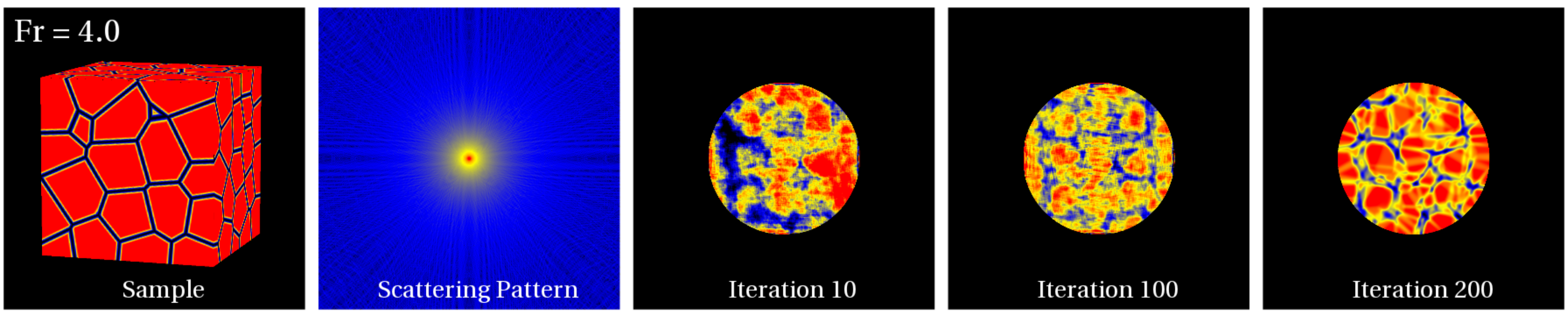


Image Reconstruction Iteration 100

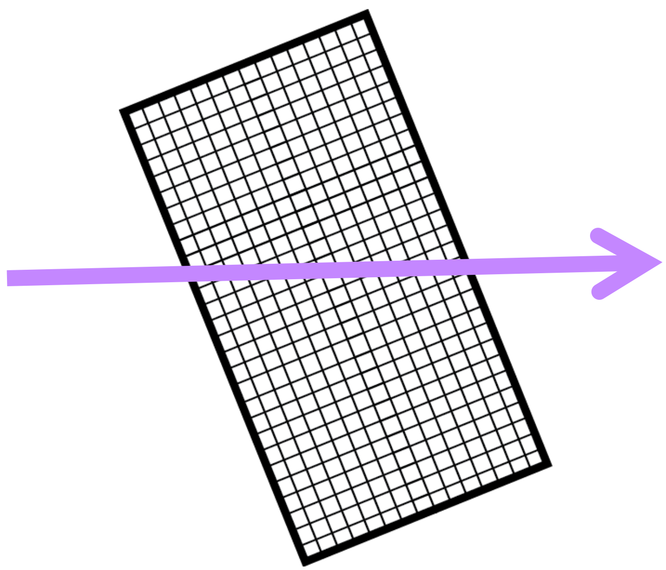
A more realistic CXDI example shows how a Fresnel regime reconstruction of a 3D polycrystal becomes a <u>projection</u> of the grain boundary structure:



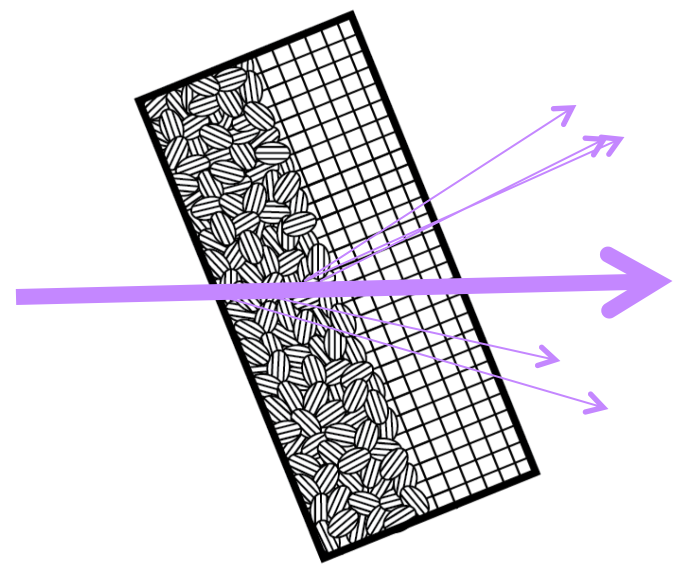
a = 100 μm *z* = 100 m Energy = 50 keV Δx ~ 200 nm
D ~ 2.0 cm *Fr* ~ 4.0 *An* ~ 10^{-3}

Techniques for probing shocked single crystals on MaRIE are being developed at LCLS

- Matching the Bragg condition for ambient single crystal diffraction creates signal that is too strong for detector
- Set up experiment for no diffraction from the ambient single crystal
- Diffraction signal occurs when Bragg condition is satisfied for transformed phase

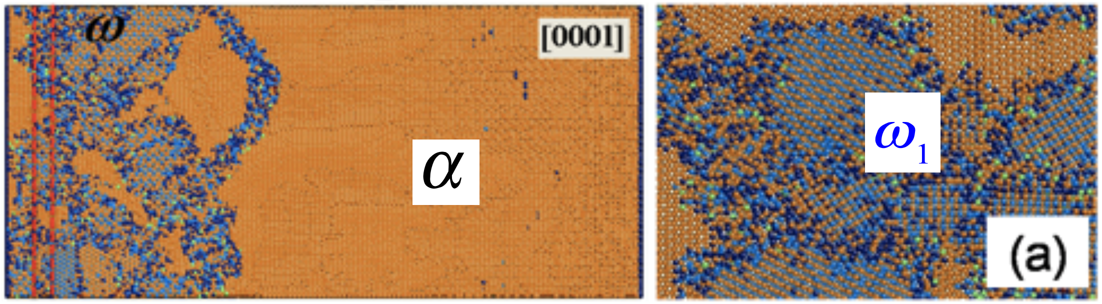


Bragg condition is not satisfied for ambient single crystal



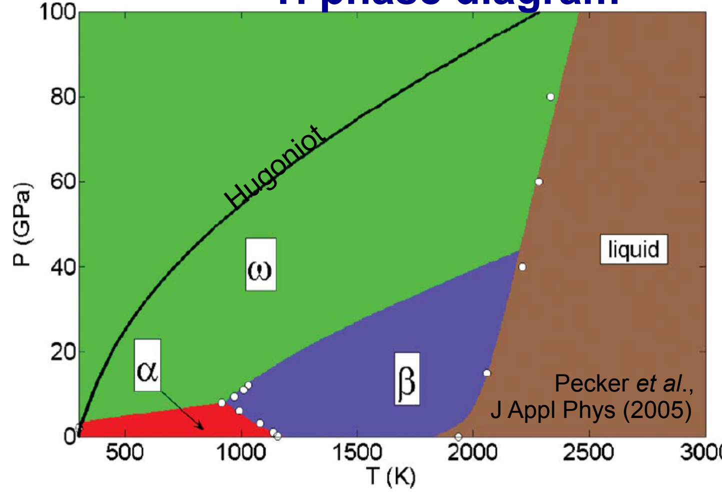
Some crystals of the shock transformed phase satisfy the Bragg condition, causing diffraction

Experiments at LCLS on phase transformation dynamics demonstrate utility of MaRIE

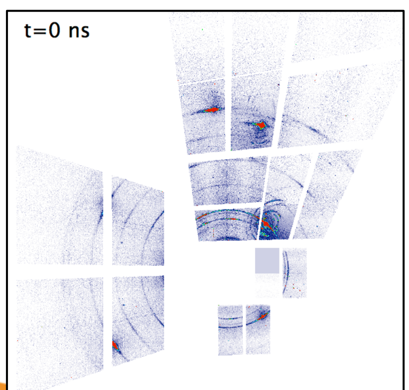


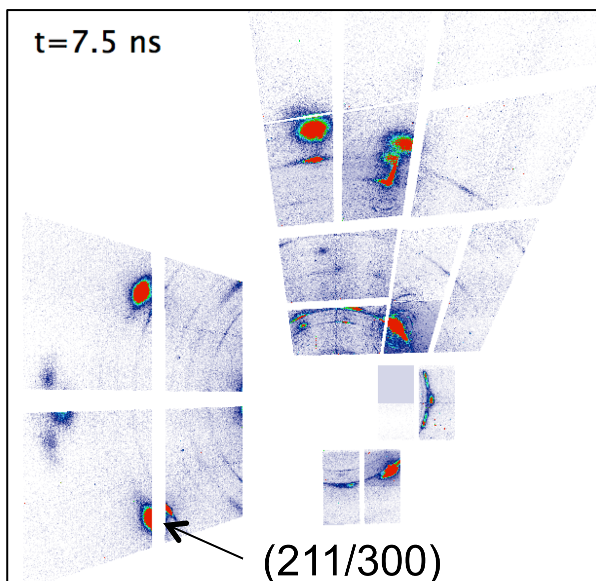
Large dependence of the evolving structure on the crystallographic shock direction in MD calculations

Ti phase diagram



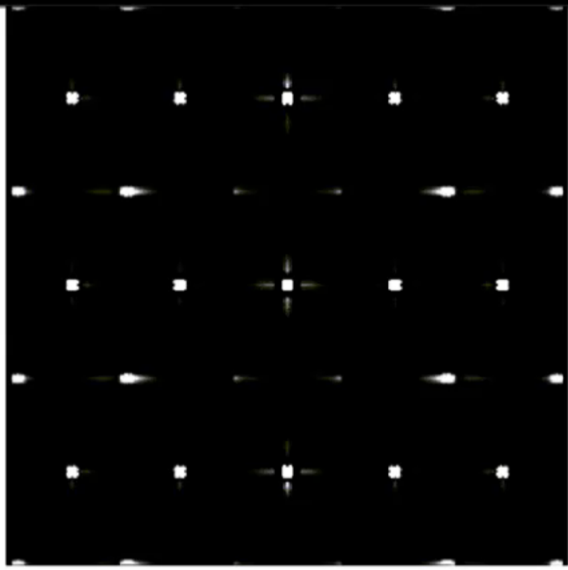
Ti [10-10]

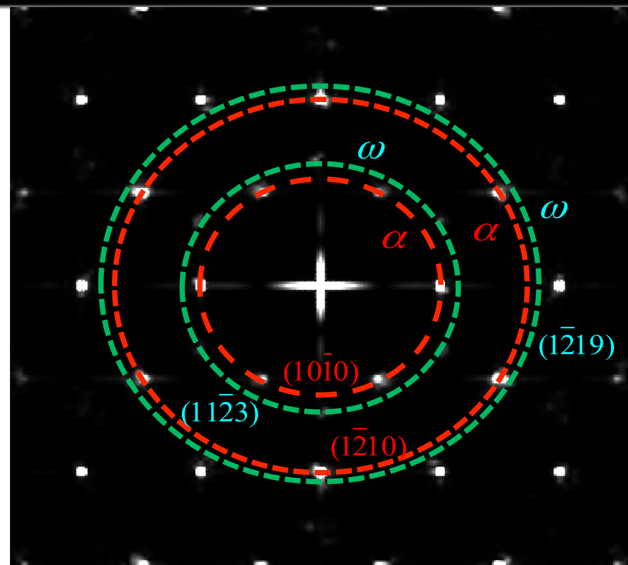




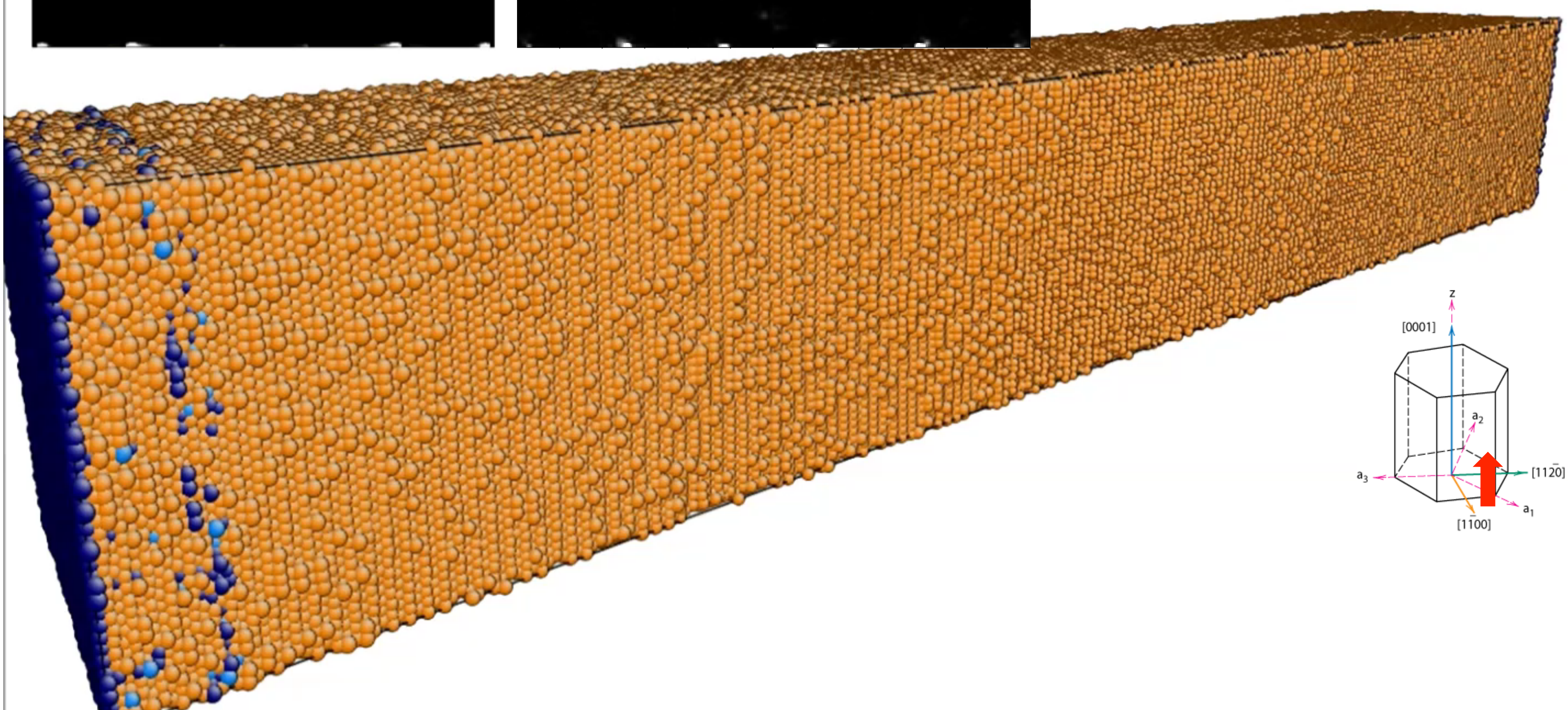
Goal: Determine the mechanisms of plasticity and phase transformation

Kinetics of α−ω shock-driven phase transformation were observed

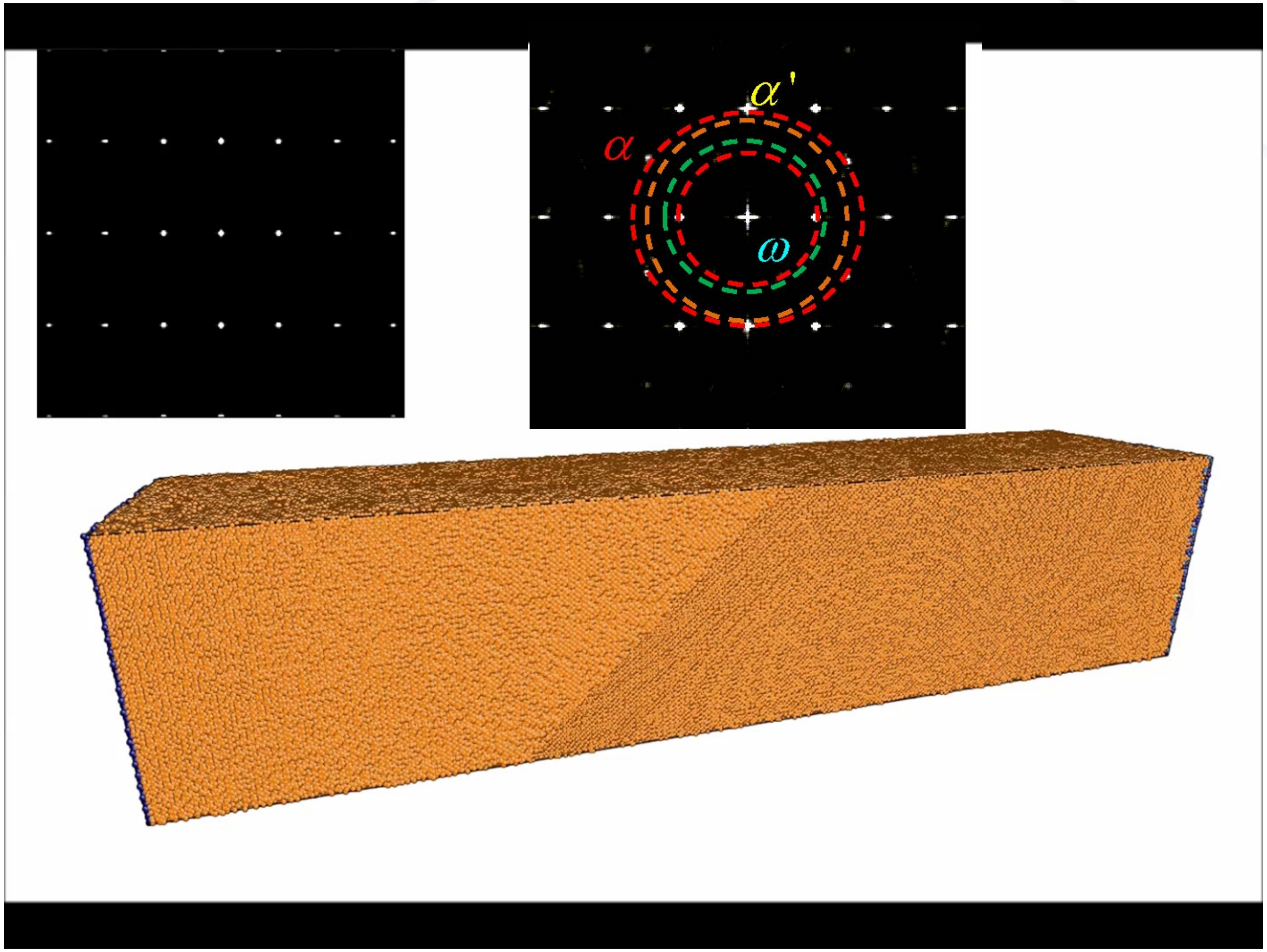




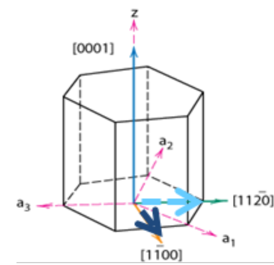
Simulated diffraction patterns show clear phase transformation



Movies allow probing new mechanisms in Ti single crystal under shock: 90^0 rotation PRECEDES phase transformation!



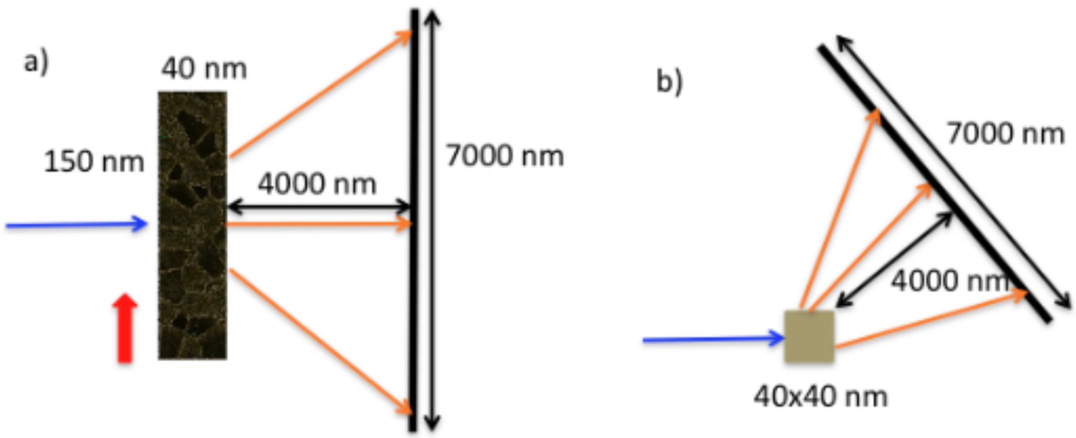
initial hcp : α1 phase
reoriented hcp: α2 phase
transformed hex: ω phase



PHYSICAL REVIEW B 89, 220101(R) (2014)

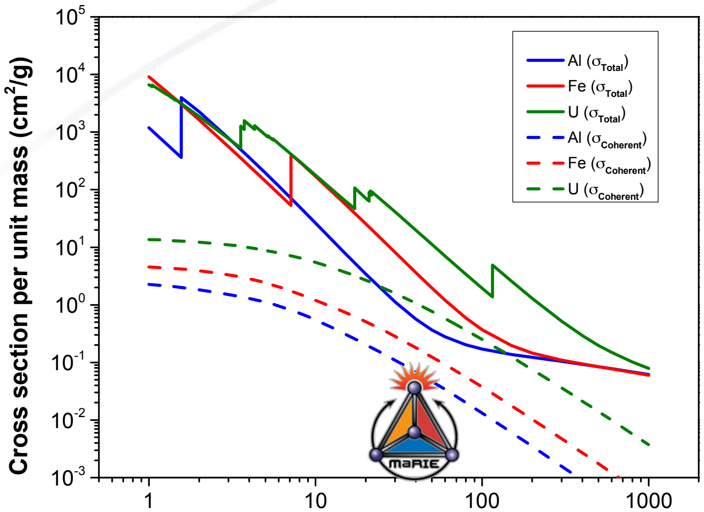
With prior sample characterization, polycrystalline dynamics can be followed

Ti Polycrystal, 1500 m/s



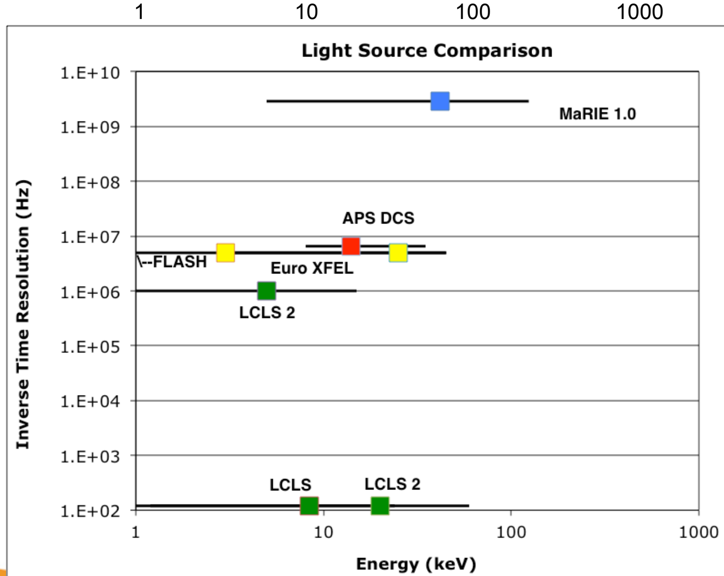
18 million atom simulation with explicit diffraction signal calculated
40 x 40 x 150 nm sample; 7 x 7 µm dectector panel at 4 µm
Fresnel number ~ 5: same as 4 x 4 x 15 µm sample and 7 x 7 cm panel at 4 cm
~Near-field conditions: diffraction peaks reflect crystal shapes
Intensity: log scale spanning 4 orders of magnitude
Crystals undergo initial elastic compression, followed by plastic deformation and phase transformation
Brown = non-bcc atoms; green = twin planes

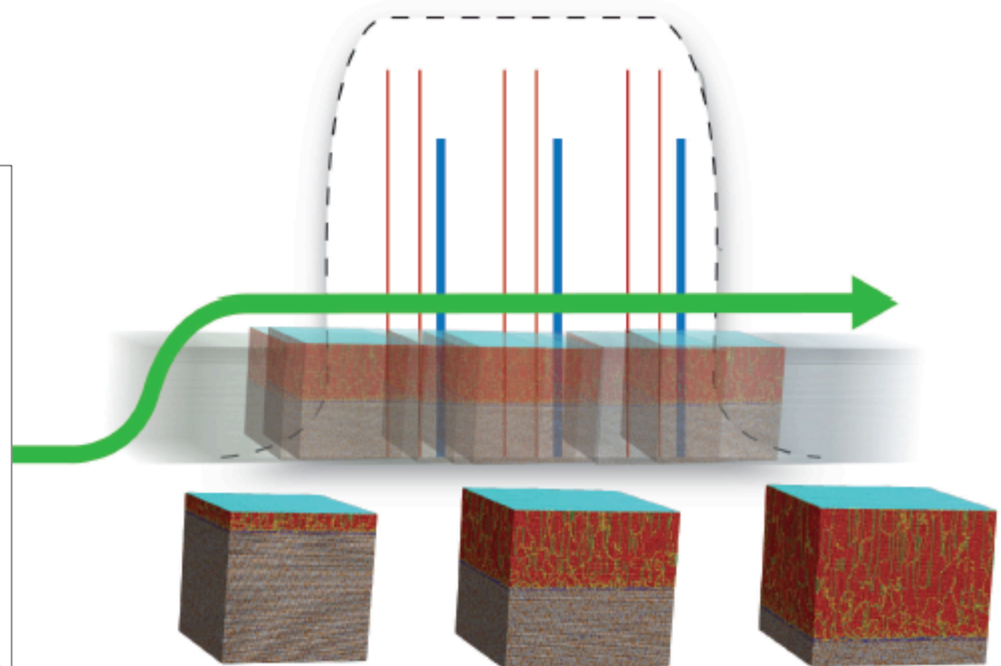
To see with time-dependence into and through the mesoscale requires: x-rays; coherent; brilliant and high repetition-rate; of sufficient high energy; and multiple probes at multiple scales



The MaRIE XFEL is harder and higher repetition rate than peer photon sources

MaRIE multiplexes 42-keV x-ray photons (red), 12-GeV electrons (blue), and 0.8-GeV protons (green) during a single dynamic event

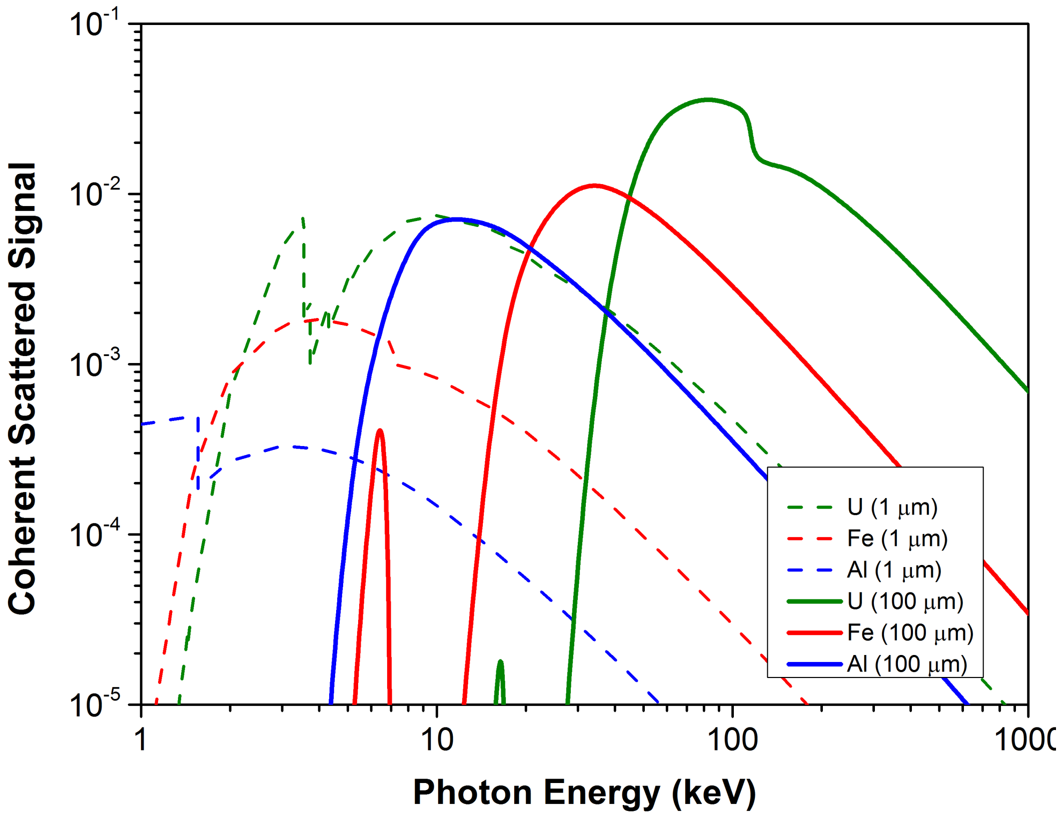




Optimizing the Coherently-Scattered Signal

N_0 = # of photons incident upon the sample
$N_U(t)$ = # of unscattered / unabsorbed photons in the beam at a depth t into the sample
$N_{1C}(t)$ = # of singly-coherently-scattered photons in the beam at a depth t into the sample.

$$\frac{N_{1C}(t)}{N_0} = \mu_C\, t\, e^{-\mu_T\, t}$$

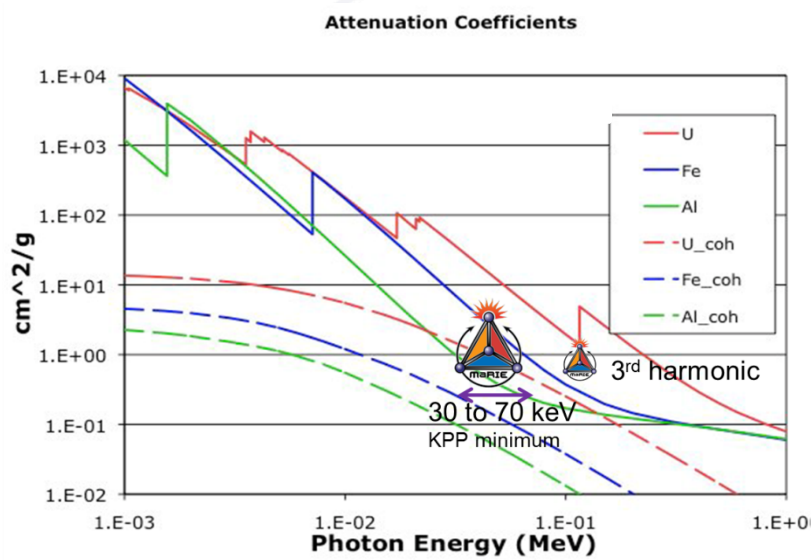


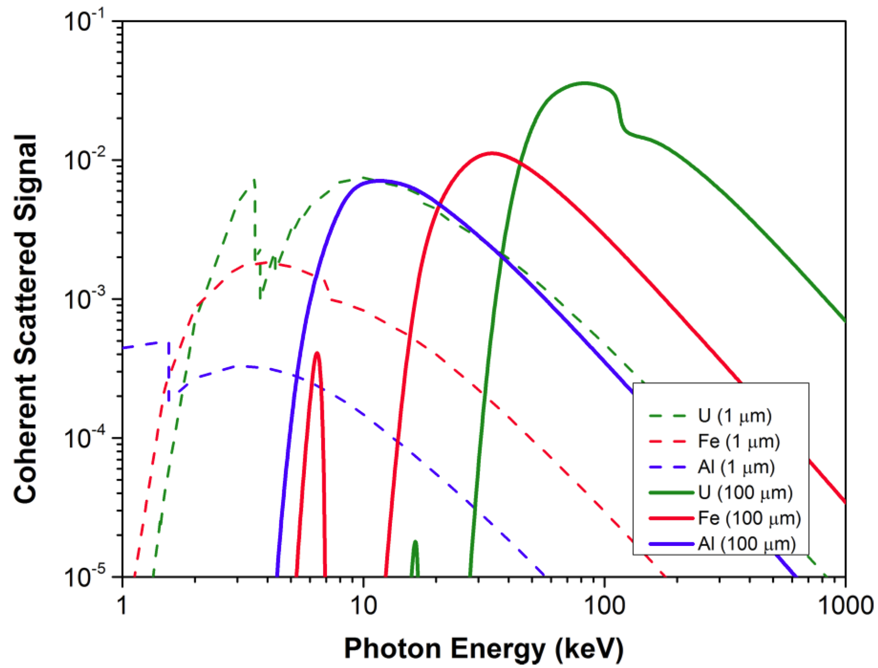
Note: It can be shown that for CXDI the rule of thumb for required coherence is that

$$\frac{\Delta\lambda}{\lambda} < \frac{2}{N_p} \sim 10^{-3}$$

where N_p is number of usable pixels on the detector.

The 42-keV design energy is a trade-off between maximizing elastic scattering for diffraction, minimizing absorptive heating, and sample thickness

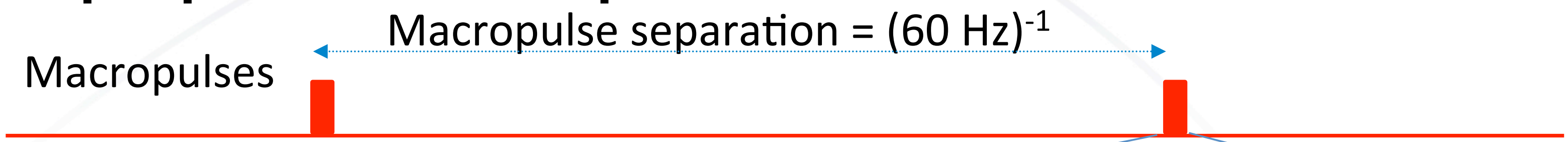




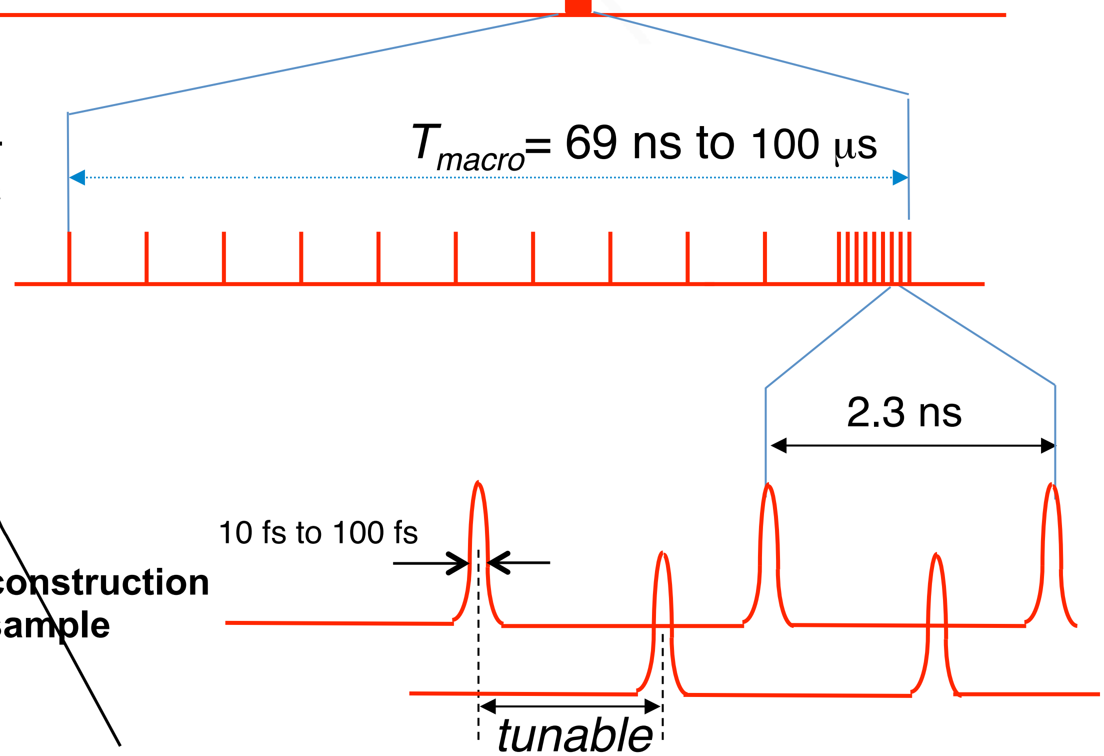
A high resolution image requires a minimum number of coherently scattered photons per sub-ps pulse. This sets the incident number of photons on a sample of ~2×10^{10}.

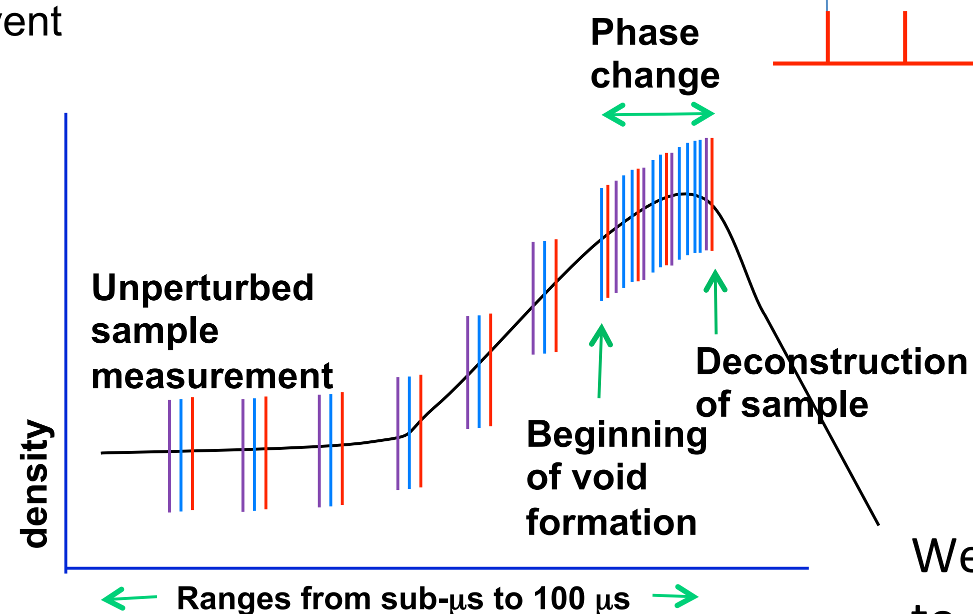
The fraction of incident photons coherently scattered just once, the coherent scattering signal, as a function of incident photon energy for various materials at thicknesses of 1 *μm* (dashed lines) and 100 *μm* (solid lines)

To see with time-dependence into and through the mesoscale requires: x-rays; coherent; brilliant and high repetition-rate; of sufficient high energy; and multiple probes at multiple scales



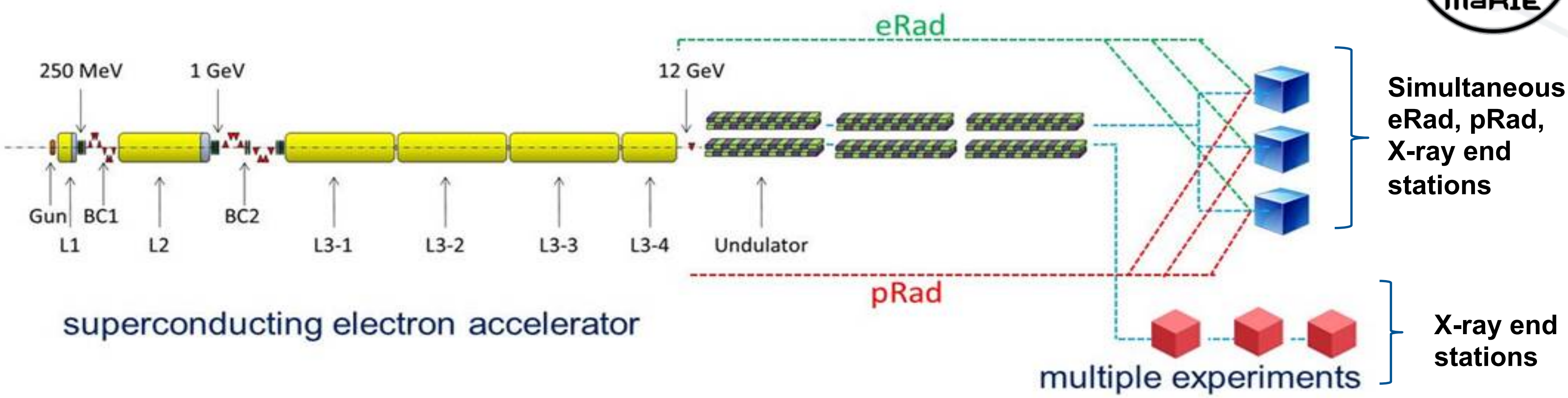
MaRIE multiplexes 42-keV x-ray photons (blue), 12-GeV electrons (purple), and 0.8-GeV protons (red) during a single dynamic event





We plan on using an optical split and delay to generate sub-ns pulse separation

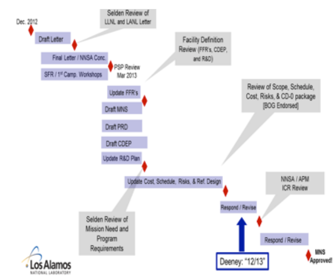
The SC linac runs parallel to the existing LANSCE proton accelerator



LANL will submit a CD-0 package to DOE/NNSA by summer of 2015

- A new direction for the nuclear enterprise
- A “new start” in the 2020’s

Path to CD-0

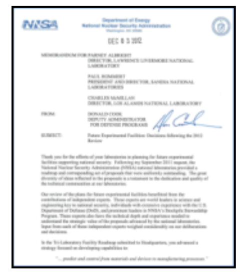


(2015)

Setting the stage for Mission Need

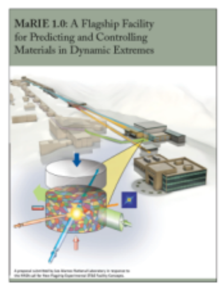
(2014)

“Move to CD-0”



(12/2012)

Pre-conceptual Proposal

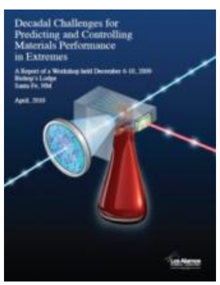


(2012)

Facility Definition

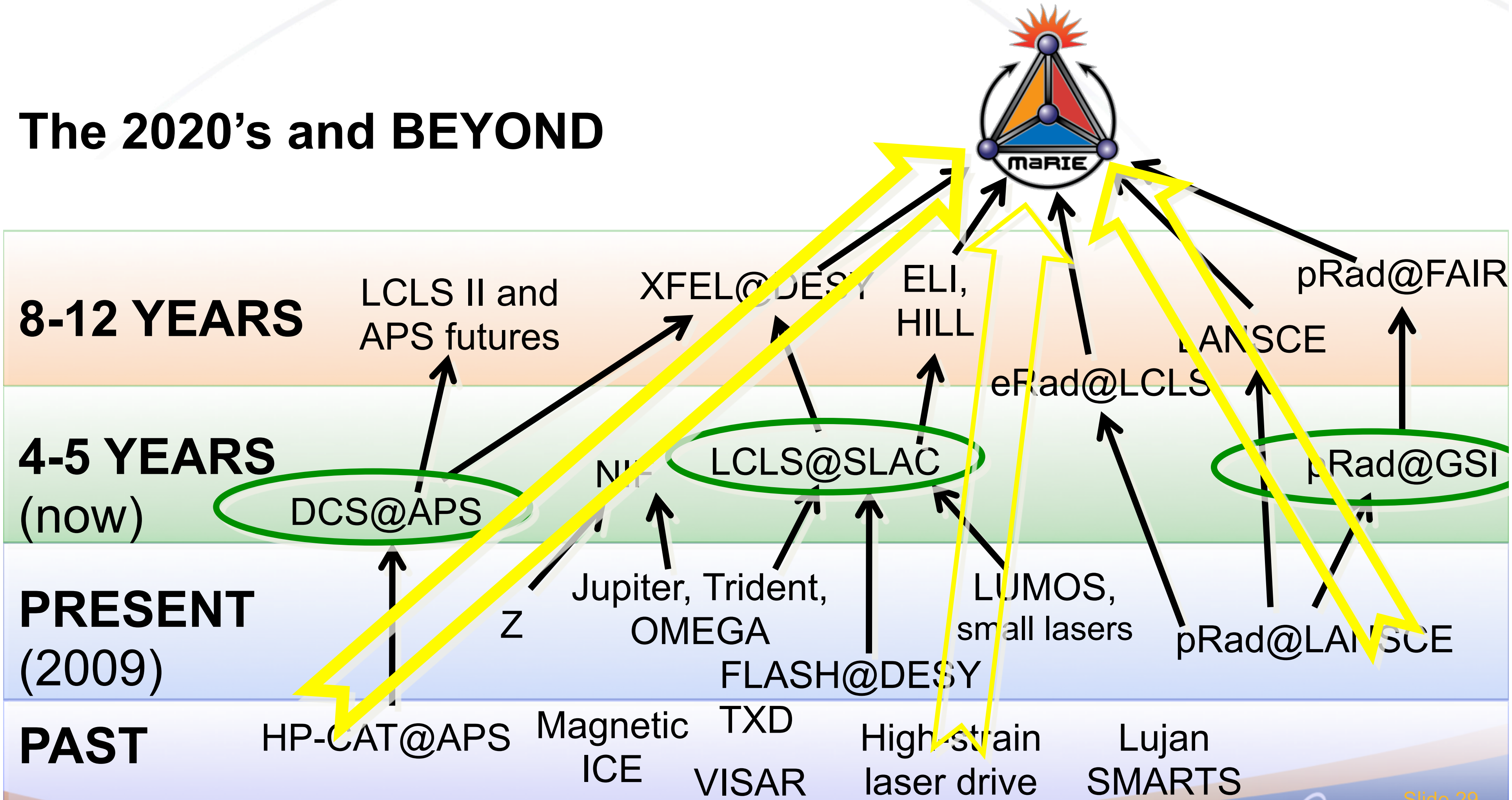
(2010)

Developing the Science Case



(2009)

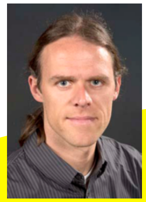
We are doing science and technology on the Roadmap to MaRIE today



MaRIE 1.0 science and mission is already attracting the best and the brightest across broad disciplines

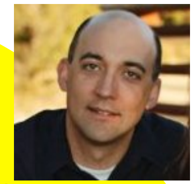

Ricardo Mejia Alvarez
P


Amy Clarke
MST

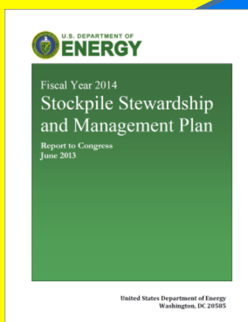

Sky Sjue
P


Saryu Fensin
MST


Cindy Bolme
WX

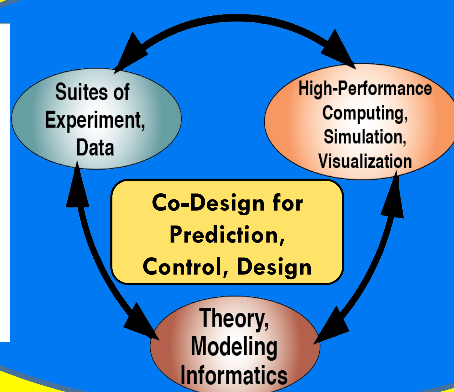

Garry Maskaly
XTD

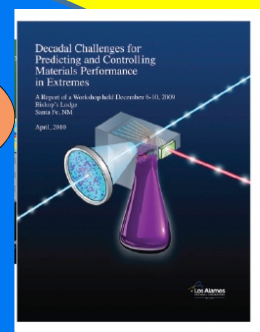

Kyle Ramos
WX


U.S. DEPARTMENT OF ENERGY

Fiscal Year 2014
Stockpile Stewardship and Management Plan
Report to Congress
June 2013

United States Department of Energy
Washington, DC 20585




Decadal Challenges for Predicting and Controlling Materials Performance in Extremes


Marianne Francois
CCS


Abigail Hunter
XCP


Irene Beyerlein
T

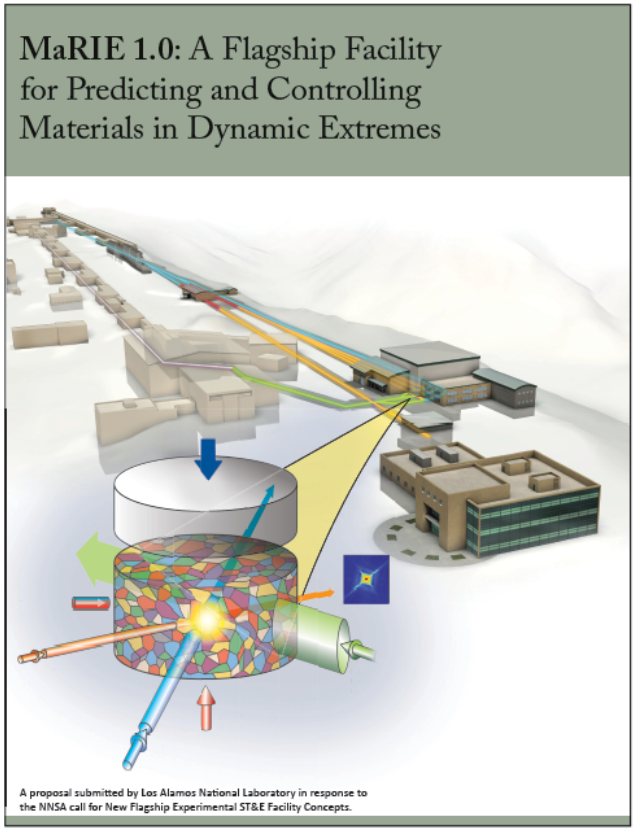

John Barber
T


Quinn Marksteiner
AOT


Richard Sandberg
MPA


Nikolai Yampolsky
AOT

MaRIE 1.0 will enable us to observe and ultimately control the time-dependent properties of materials that affect materials performance



A mission need exists for a facility focused on predicting and controlling materials in extreme environments, exploiting *in situ* transient measurements on real materials in relevant dynamic extremes to **address national security challenges through accelerated qualification, certification, and assessment**.

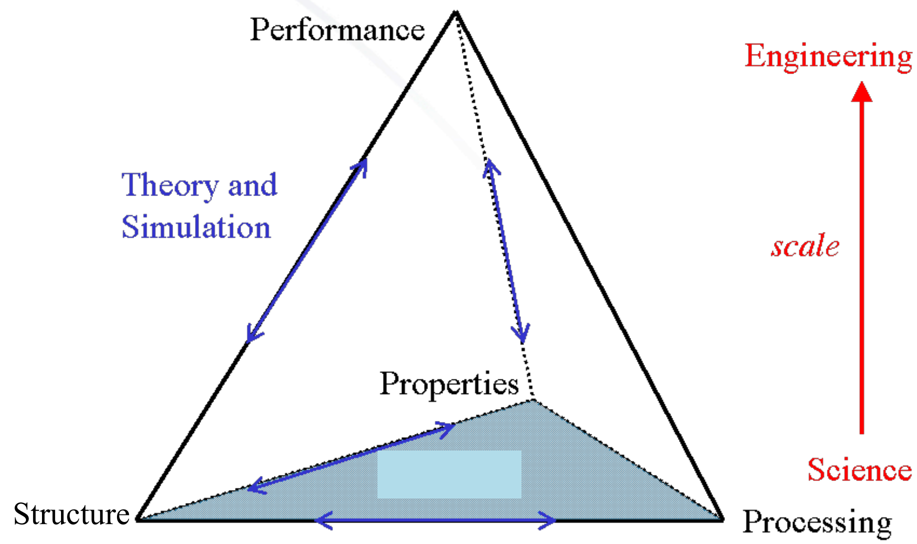
Achieving controlled functionality at the mesoscale through co-design is the **frontier of materials research**.

MaRIE 1.0 meets this need with a robust preconceptual reference design that is grounded in **community-defined mission and scientific requirements**.

Backups

MaRIE 1.0 is about *both* time-dependent control of making materials, and measurements of their time-dependent performance

- Materials Design is the integration of **structure** and **processing** to achieve a desired **property** of a material, all of which determines the **performance** of an engineering system with the intent to
  - (A) develop new materials and properties,
  - (B) select functions of materials for applications, or
  - (C) optimize performance of a given material.



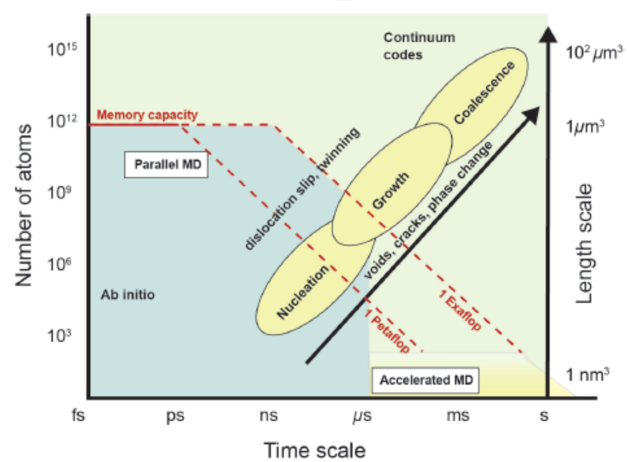
Vision: *MaRIE provides process-aware manufacturing R&D capability that integrates in-situ diagnostics with real-time adaptive modeling for concurrent feed-back control at the mesoscale for "born-qualified" materials.*

Some parameters from a few example experiments*:

Quantity	Description	Example 1	Example 2	Example 3
Z	Sample atomic number	26 (Fe)	92 (U)	92 (U)
λ	Source wavelength (Å)	0.25	0.25	0.3
E_γ	Photon energy (keV)	50	50	41.3
d	Spot size (μm)	100	100	120
t	Sample thickness (μm)	500	100	200
iFOM	Imaging figure-of-merit	400	500	120
$\mathrm{O}_\perp$	Transverse oversampling factor	2	2	2
p	Pixel size (μm)	12.5	12.5	12.5
$D = \mathrm{O}_\perp\ \mathrm{iFOM}\ p$	Detector size (cm)	1.0	1.25	0.3
L	Sample-detector distance (m)	100	100	100
$\phi_{\max} = \tan^{-1}(D/2L)$	Maximum scattering angle (rad)	5×10^{-5}	6.3×10^{-5}	1.5×10^{-5}
$\Delta x = L\lambda/D$	Transverse image resolution (nm)	250	200	1000
$\Delta z = 4\lambda L^2/D^2$	Longitudinal resolution (cm)	1.0	0.64	13
$\mathrm{Fr} = d^2/\lambda L$	Fresnel number	4.0	4.0	5
$\mathrm{An} = D^4/\lambda L^3$	Small angle number	4×10^{-4}	1×10^{-3}	3×10^{-6}
η_{QE}	Detector quantum efficiency	90%	90%	90%
P	Average photons per pixel	400	100	100
$1/\sqrt{P}$	Average-per-pixel noise-to-signal	5%	10%	10%
N_0	Required incident photons for imaging [Eq. (33)]	1.3×10^{10}	6.4×10^{9}	1.5×10^{10}
N_{1C}	Coherently-scattered photons for imaging [Eq. (31)]	2.8×10^{8}	1.1×10^{8}	6.4×10^{6}
N_{1C}/N_0	Fraction coherently scattered	2.3%	1.7%	0.04%
ΔT	Average temperature rise (K) [Eq. (35)]	3.4	27	23

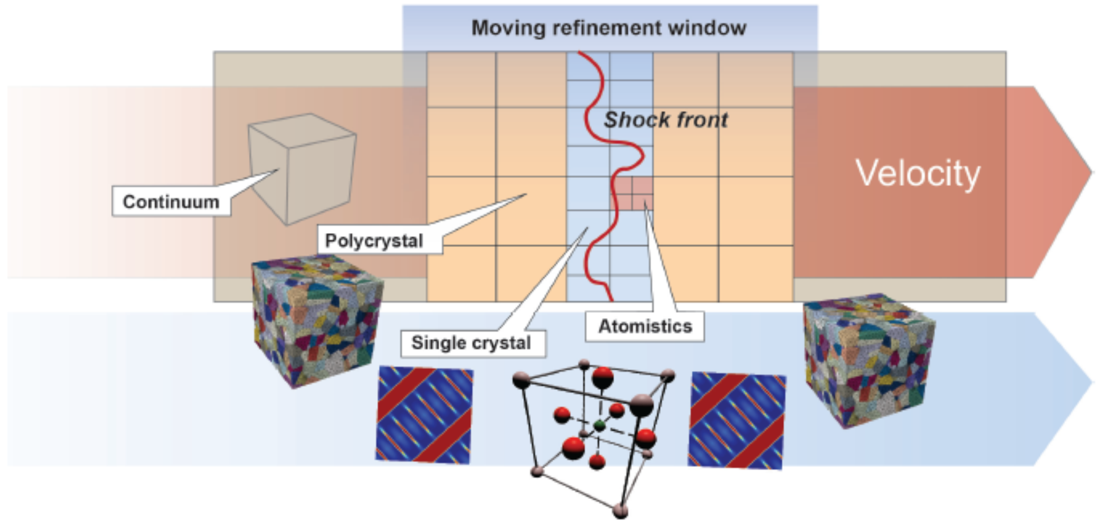
* Taken from PRB **89** 184105 (2014).

The coupling of multi-scale theory and multi-probe experiments on next-generation computing architectures for future integrated codes is key to our success.



Mesoscale materials phenomena need extreme-scale computing

Variable-resolution models are synergistic with multi-probe, *in situ*, transient measurements



The development of MaRIE-validated models will **reduce uncertainty** in integrated codes and provide **predictive descriptions of materials & components** during **manufacturing** and in **dynamic extremes**

Experimental ties to Theory for both manufacture and performance is critical

A number of codes are used

Macro-Mech.

TEPLA
SCM
DCA
GMC
MTS
PTW
Sesame

Polycrystal

VPSC
Taylor
S-Taylor
D-Poly

Single Crystal

G-L
DeMecha
Laminate
Classical

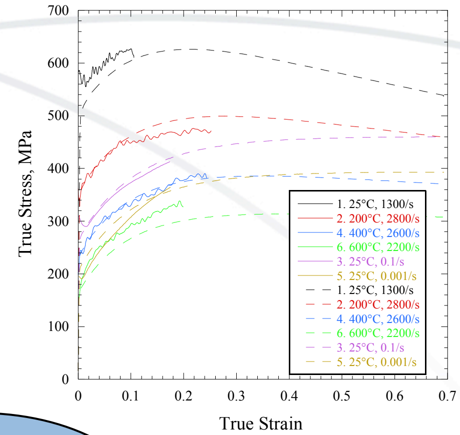
Molecular Dynamics

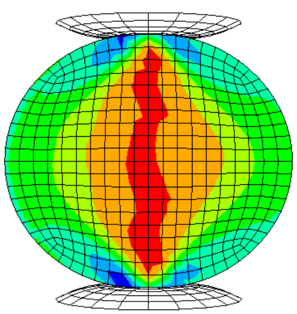
SPaSM
LAMMPS
Lucretius

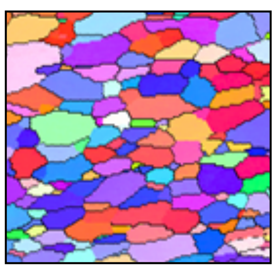
Quantum Mechanics

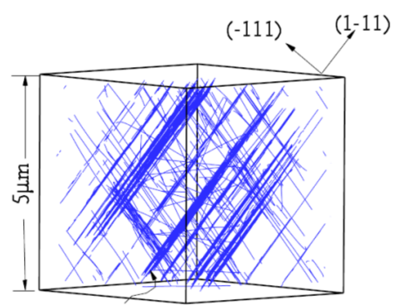
VASP
MondoSCF

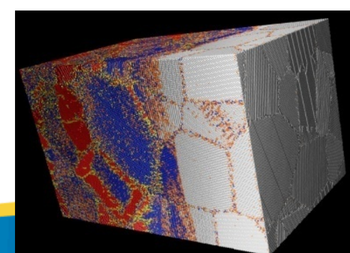
Experiment

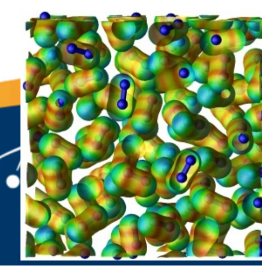












UNCLASSIFIED