EXPLORING MATERIALS WITH HIGH-ENERGY X-RAYS

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Aarhus University, March 17 2023

ARGONNE NATIONAL LABORATORY

History: First US national lab (1946); extend Fermi's UC work on nuclear reactors (outside of population center)
Today: Multidisciplinary, one of 10 Office of Science national labs; 3400 staff and ~1.2B budget

Artificial Intelligence for Science

Autonomous Discovery

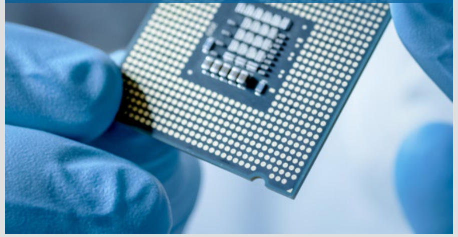
Clean Energy and Sustainability

Climate Action

Detection and Imaging of Signatures

Hard X-Ray Sciences

Microelectronics



Quantum Information Science

Radioisotope Discovery

ARGONNE FACILITIES

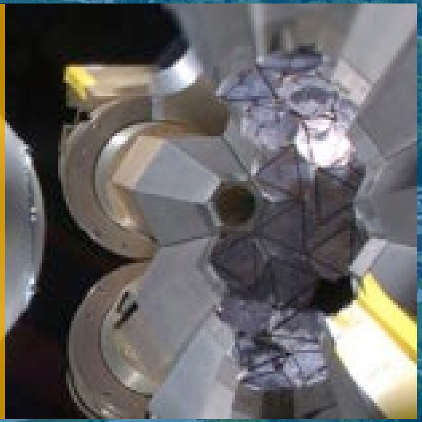
Enabling science from nanoscale to exascale

Advanced Photon Source

APS

Argonne Tandem Linear Accelerator System

ATLAS



Argonne Leadership Computing Facility

ALCF

Center for Nanoscale Materials

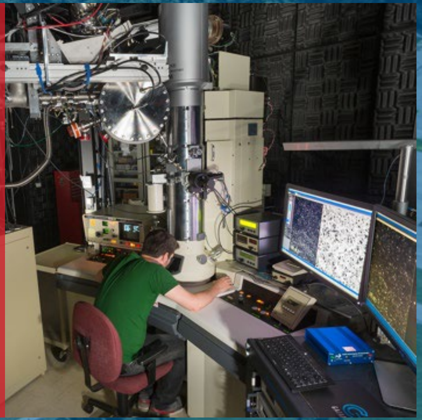
CNM

Atmospheric Radiation Measurement – The Southern Great Plains

ARM

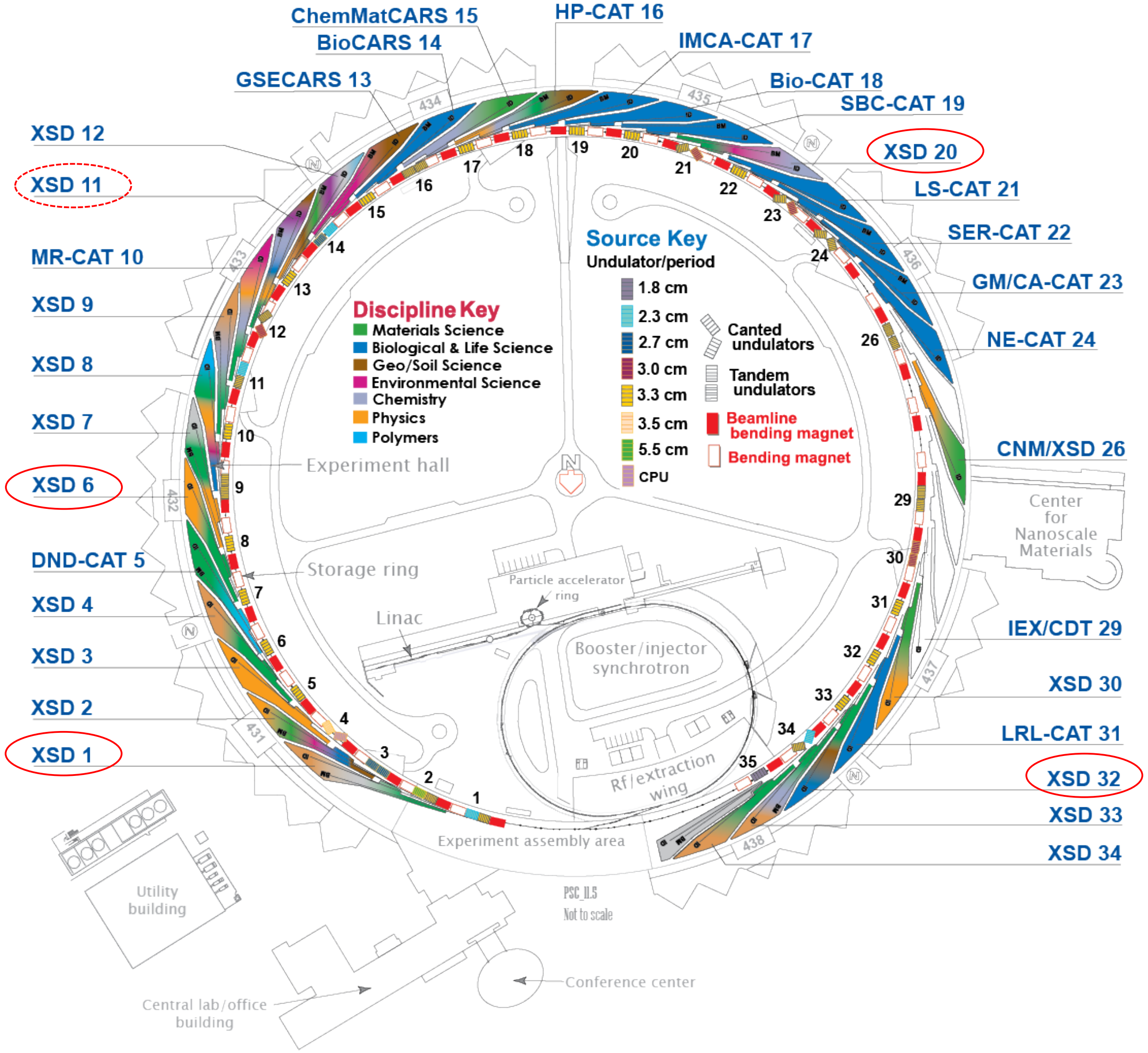
Intermediate Voltage Electron Microscopy-Tandem Facility

IVEM



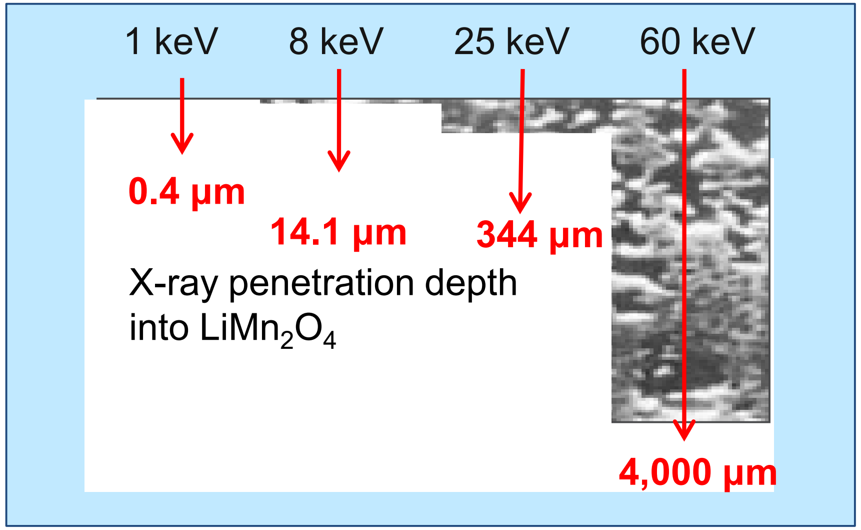
THE ADVANCED PHOTON SOURCE

- Comprises ~1/4 of ANL in terms of staff/budget
- Primarily DOE-BES funded
- Multi-disciplinary science
- The nations leading high-energy x-ray source
- 66 simultaneously operating beamlines, 5000 hours per year
- ~5500 users annually
- 1400 protein structures solved per year, many new drugs discovered
- Began operating 1995 -> MBA upgrade 2023-24



Primarily general user access - through peer-reviewed proposals – no charge

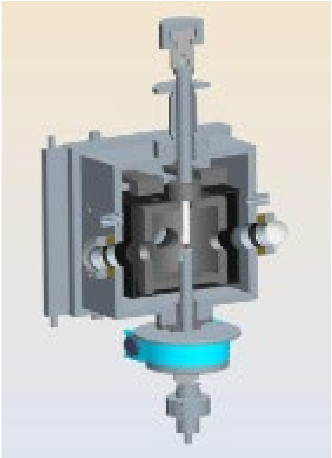
HIGH-ENERGY X-RAYS



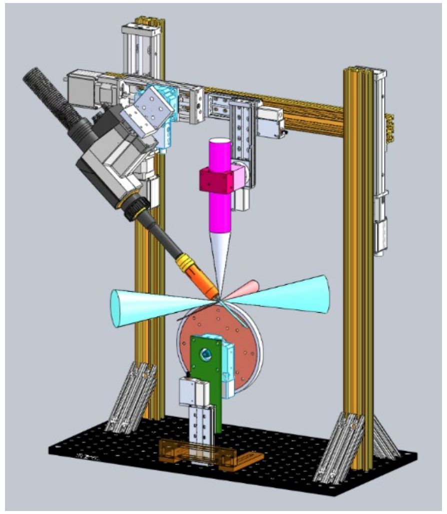
Interaction properties:
- low attenuation
- small scattering angles - high *Q access*
- improved validity of Born approximation (< mult. scattering)

Enables:
- large and extreme environments, e.g. in thick-walled containment
- scattering from bulk / away from surface in thick or high-Z samples
- operation in air

Microwave reactor



Stress+temp+activated



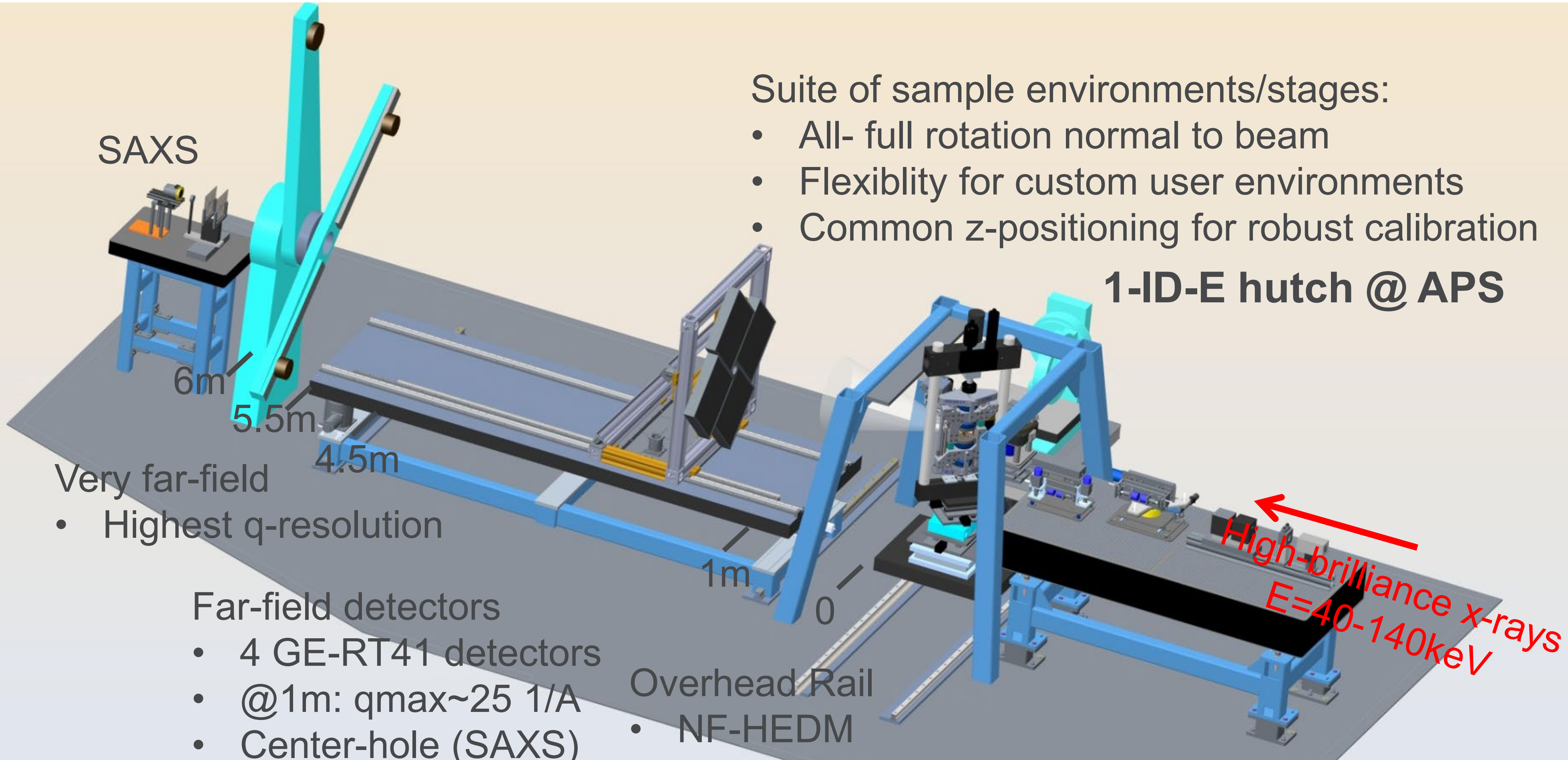
Additive manufacturing

High ring energy synchrotrons like APS (7 GeV) are brilliant HEX sources -> space and time resolution

1-ID: COMBINED IN-SITU HIGH-ENERGY TECHNIQUES

Suite of sample environments/stages:
- All- full rotation normal to beam
- Flexiblity for custom user environments
- Common z-positioning for robust calibration

1-ID-E hutch @ APS



Very far-field
- Highest q-resolution

Far-field detectors
- 4 GE-RT41 detectors
- @1m: qmax~25 1/A
- Center-hole (SAXS)

Overhead Rail
- NF-HEDM
- Conical slit
- Tomography
- Lasers

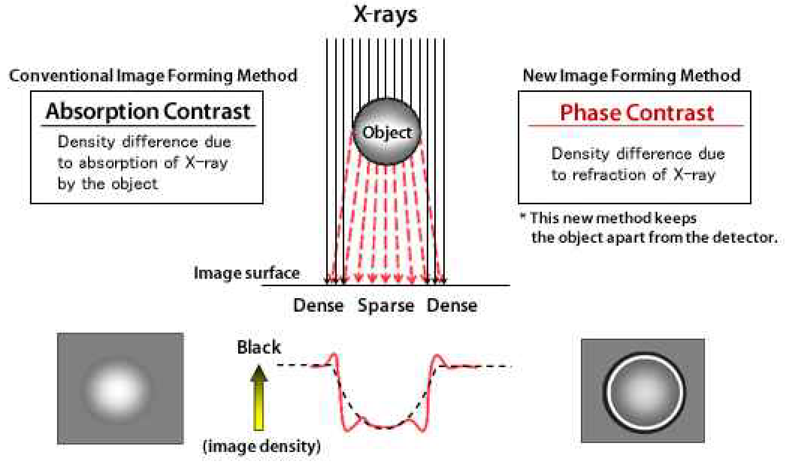
Beam-conditioning system
- Focusing optics for <um beams
- White and mono capable
- BPMs, slits, DIC, etc

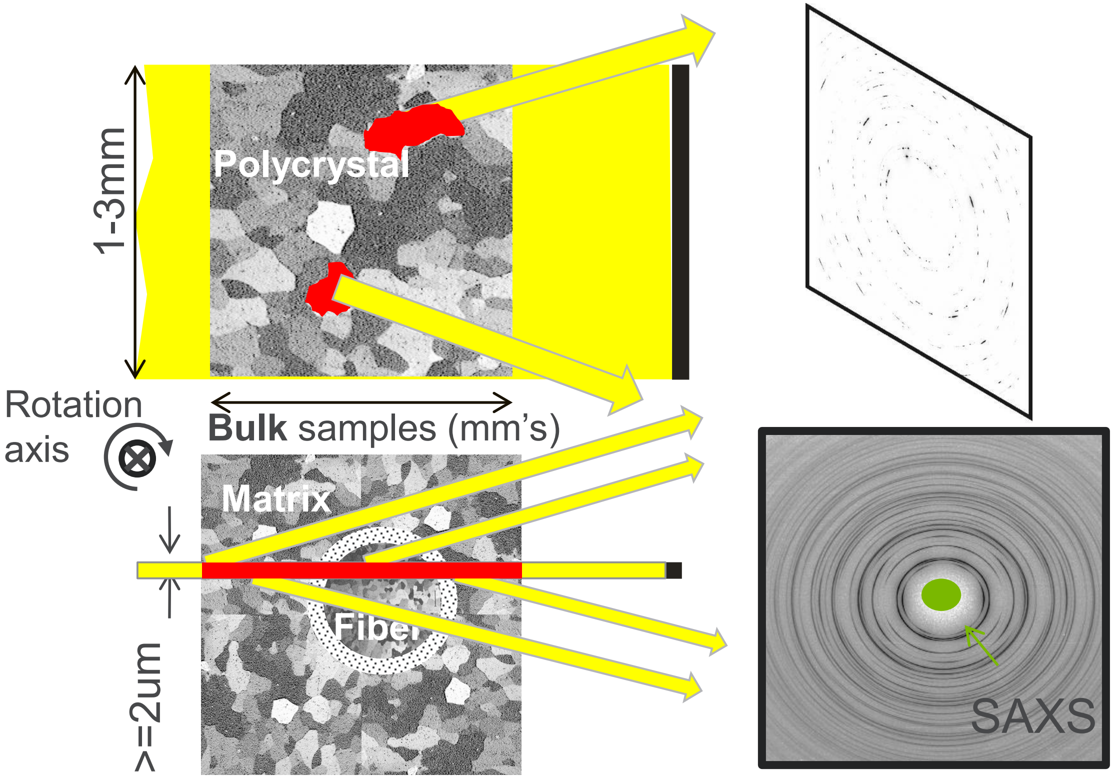
IMAGING MICROSTRUCTURE WITH HIGH-ENERGY X-RAYS

- Direct-beam based
  - Volume based on beam size, typically 0.1-10 mm^3
  - Linear resolution ~(beam size) /1000
  - Absorption or phase contrast w/synchrotrons
  - Rotation series-> reconstruct
  - 3D volume of morphological features (cracks, 2^{nd} phases etc)

- Diffraction-based, grain resolved (3DXRD or HEDM)
  - Make xray volume (just) small enough to resolve distinct reflections (synch <~10k grains)
  - Rotation series -> reconstruct
  - 3D grain-resolved: size, position, orientation, strain
  - As detector is moved further away, more sensitive to strain than position (nf-, ff-, vff- modes)

- Scattering-based, grain averaged (SAXS and WAXS)
  - Crystalline ('powder') & non-crystalline materials
  - Strain and volume for each phase present
  - Translate (& rotate) to image in 1D->2D->3D (scattering tomography)
  - Angle and energy dispersive modes

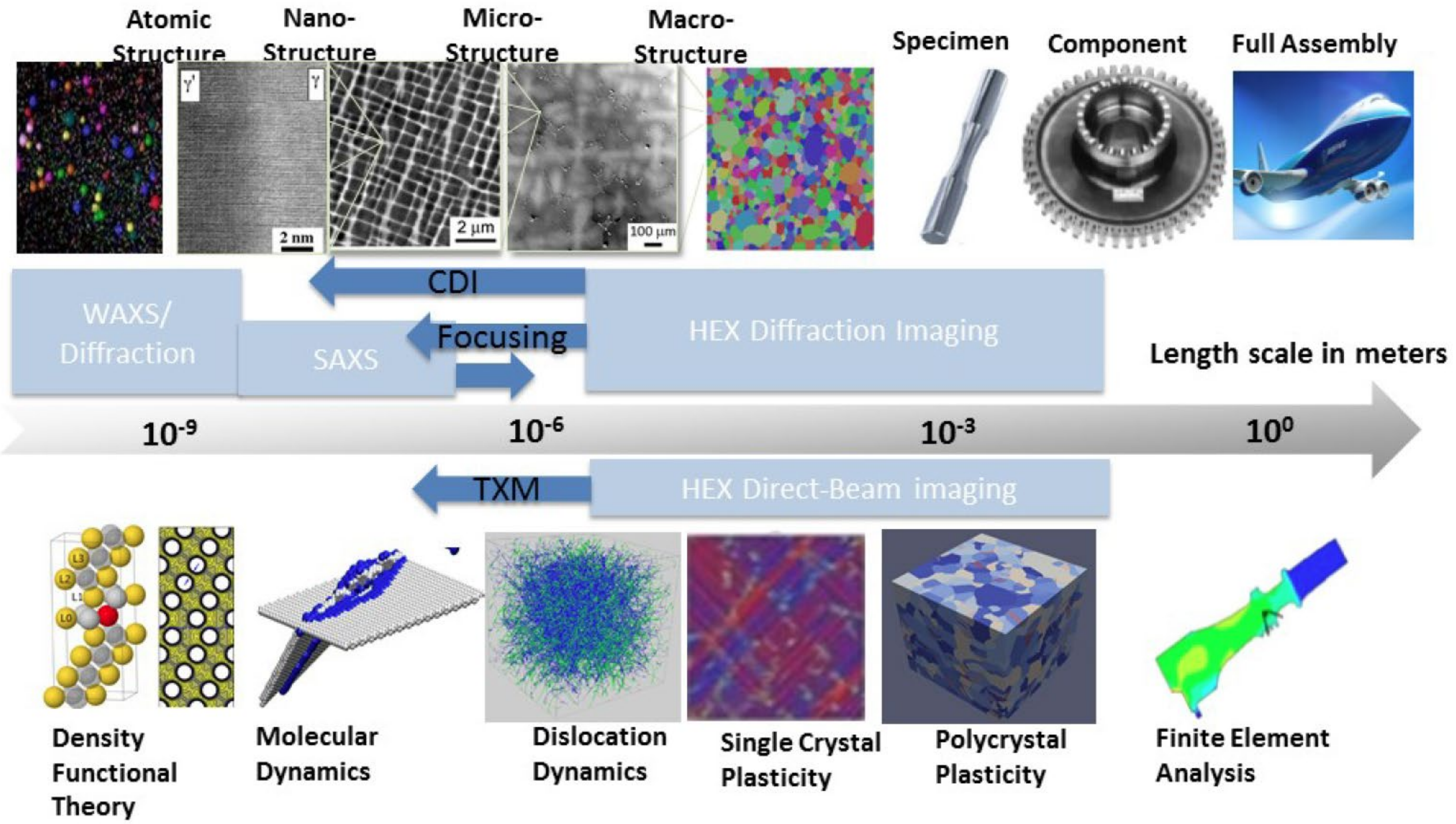




Incident beam E= 40-120 keV; Scattering angles <10 deg

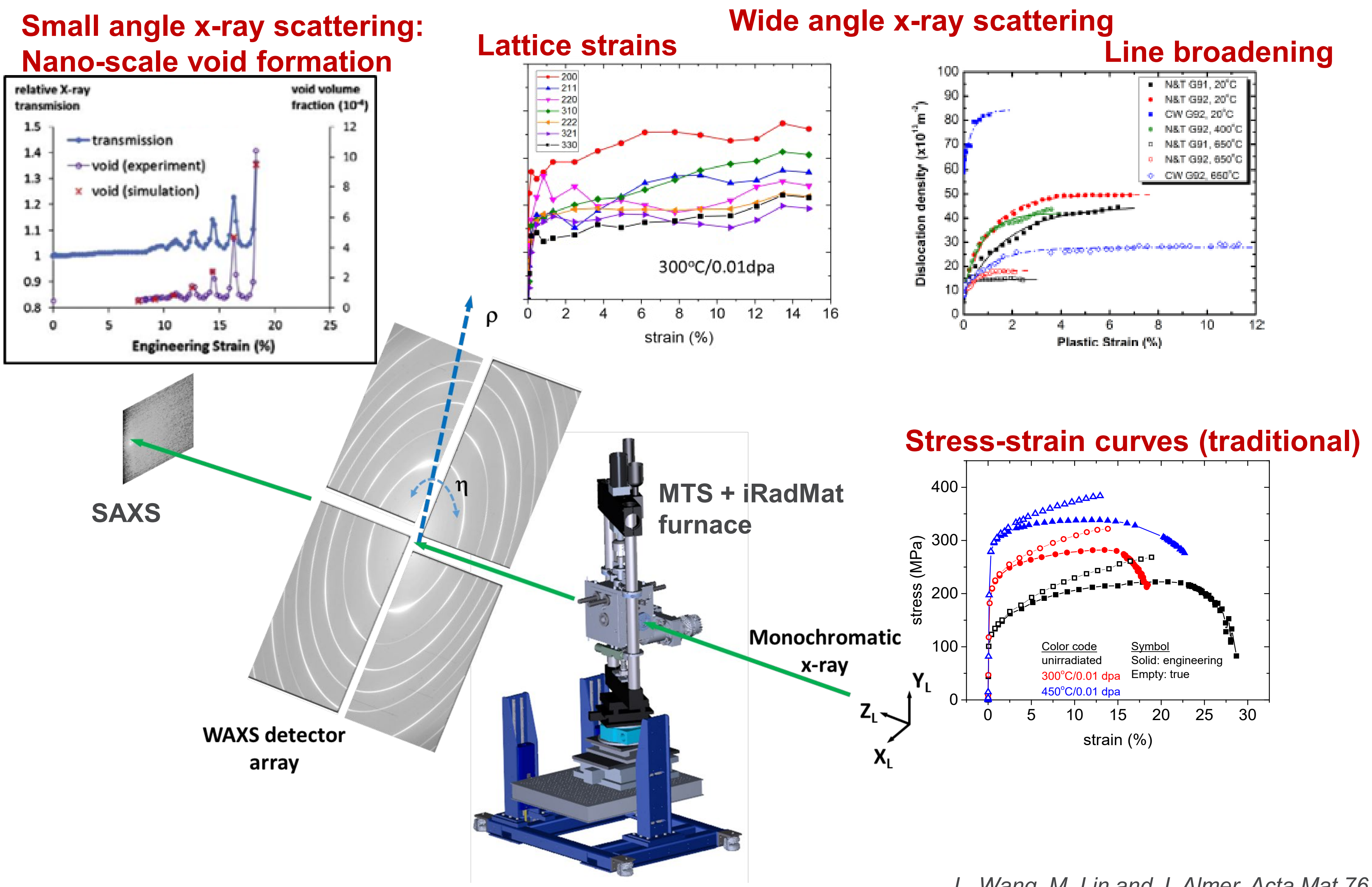
HEX: MULTI-SCALE, MULTI-MODAL IMAGING

- Non-destructive and (often) in-situ
- Wide size and time-scales through direct- and reciprocal-space methods
- Multi-dimensional imaging to test and improve materials models



Scientific cases for hierarchical needs include: aerospace metals, batteries, SOFCs, nuclear materials and bio-materials

IN SITU SAXS/WAXS CAPABILITIES

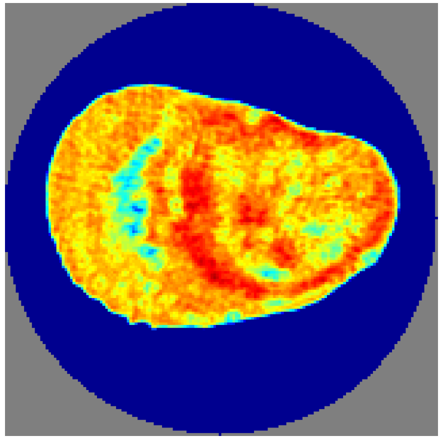


L. Wang, M. Lin and J. Almer, Acta Mat 76 (2014).

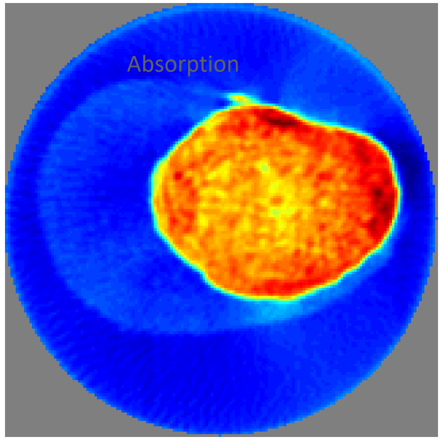
SCATTERING-BASED TOMOGRAPHY

- Useful for nano-grains, high deformation, weakly ordered systems (WAXS/PDF,SAXS)
- These are typically energy materials, geomaterials or biological materials
- Reconstruct a 2D scattering pattern per voxel: rich microstructural information

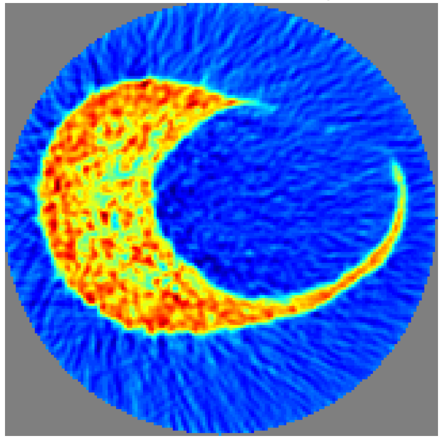
Absorption



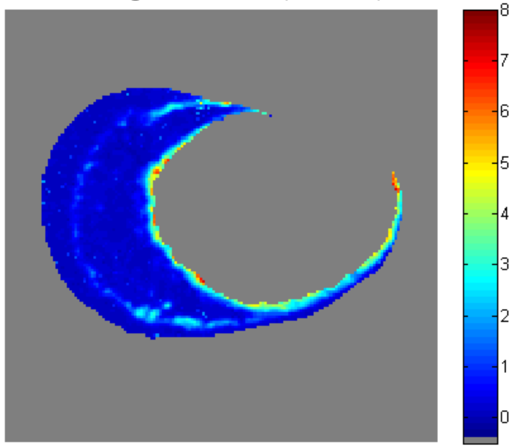
Aragonite intensity



Calcite intensity



Mg content (mol%)

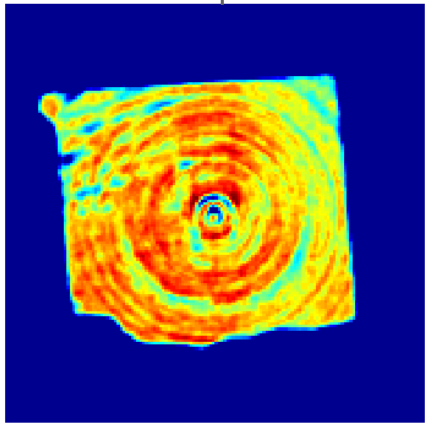


Byssus

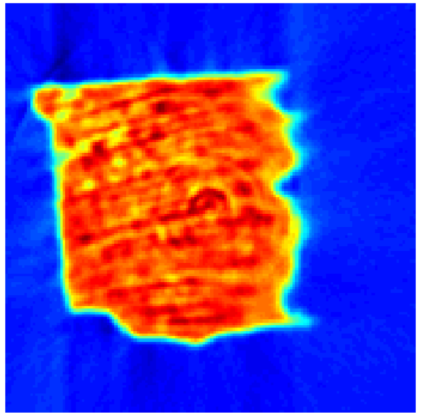
25μm resolution: phases with similar density can be easily distinguished. Peak shifts provide intraphase concentration gradients.

10mm resolution (smaller sample): individual lamellae can be distinguished; strains shown

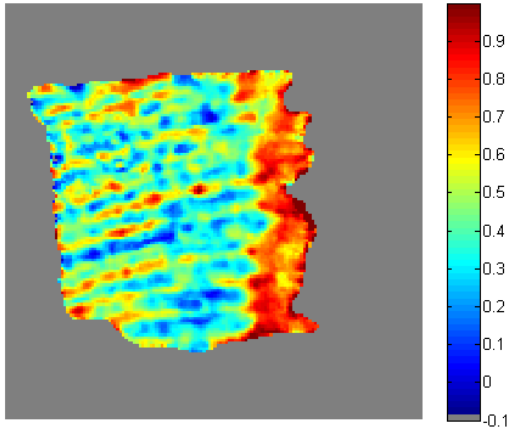
Absorption



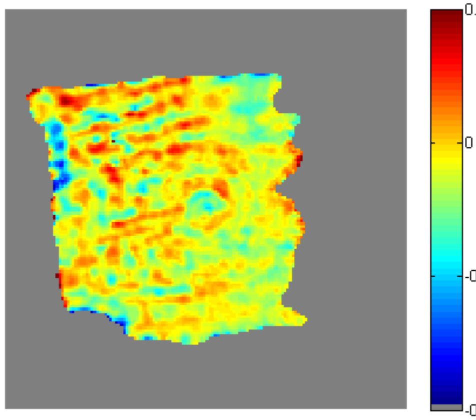
Aragonite intensity



Micro strain, aragonite



Macro strain, aragonite

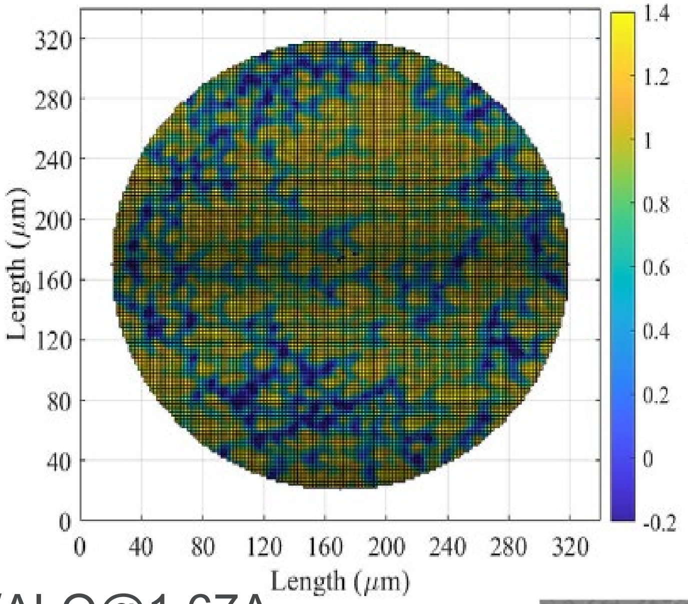


Leemreize, Almer, Stock & Birkedal. J. R. Soc. Interface 10 (2013)

And @ higher resolution? See Henrik!

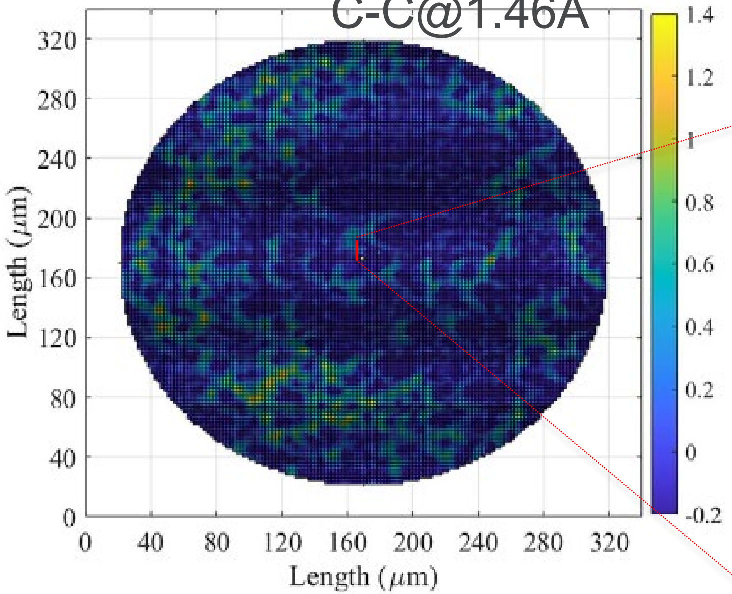
SCATTERING-BASED TOMOGRAPHY

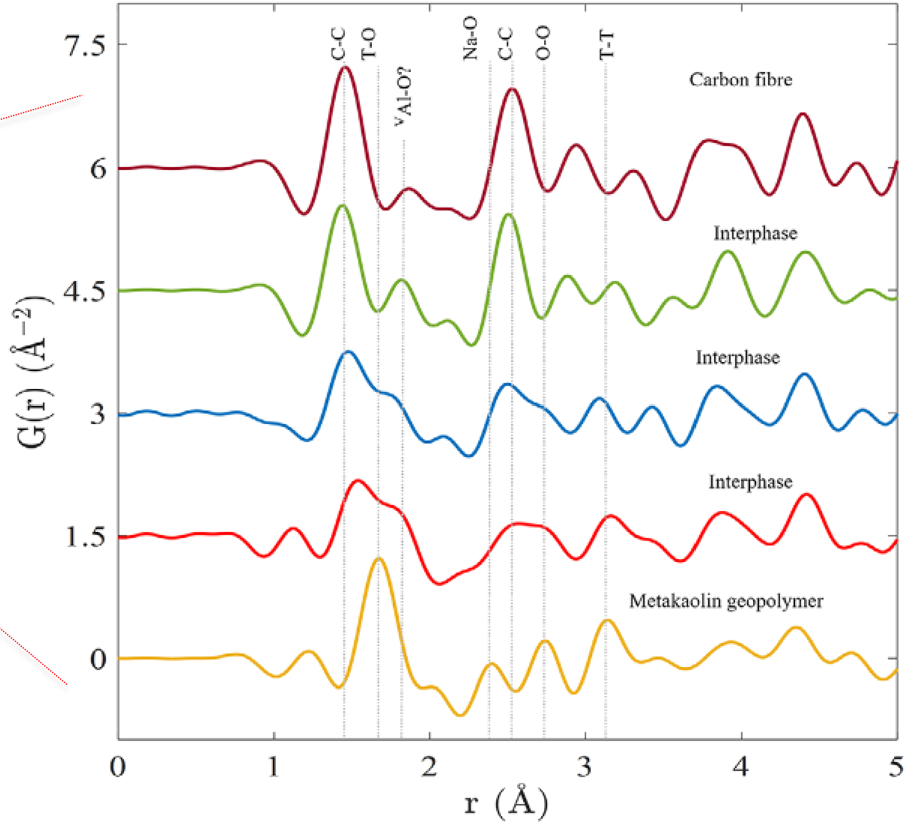
- C-fiber metakaolin geopolymer composites (‘green concrete’)
- Local PDF measurements identify local atomic structure at fiber/matrix interface
- Si/Al-O bonding associated with C-O-Al linkages (from DFT) @ interface
- Responsible for good mechanical properties?



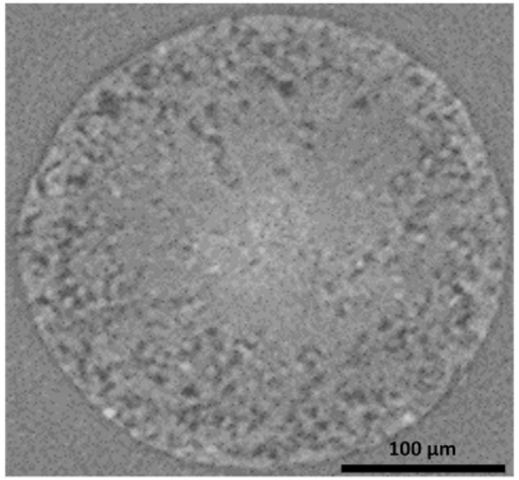
Si/Al-O@1.67A

C-C@1.46A



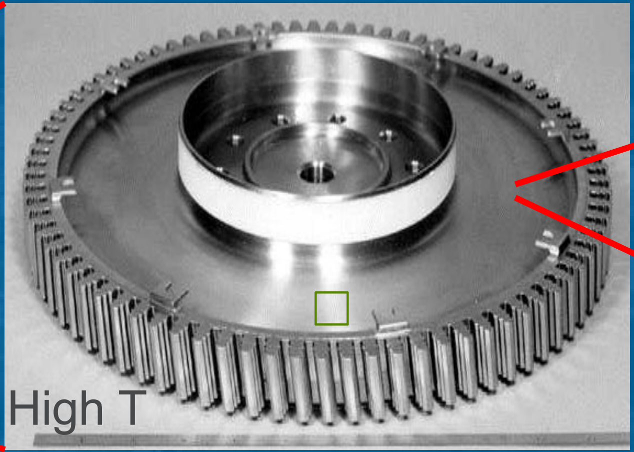


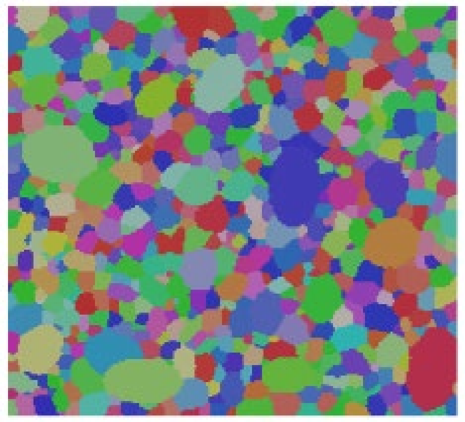
ST: 2 x 10 μm^2 beam
AT: ~1um resolution



Absorption

Hajimohammadi, Masoumi, Almer, White et al, JACeS, 2022; 105:1519-1532





DETAILED SCIENCE CASE: FATIGUE IN AN EMBEDDED INCLUSION

ELSEVIER

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat

Full length article

Investigation of fatigue crack initiation from a non-metallic inclusion via high energy x-ray diffraction microscopy

CrossMark

Diwakar Naragani [a], Michael D. Sangid [a, *], Paul A. Shade [b], Jay C. Schuren [b, 1], Hemant Sharma [c], Jun-Sang Park [c], Peter Kenesei [c], Joel V. Bernier [d], Todd J. Turner [b], Iain Parr [e]

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[d] *Engineering Directorate, Lawrence Livermore National Laboratory, Livermore, CA 94550, USA*
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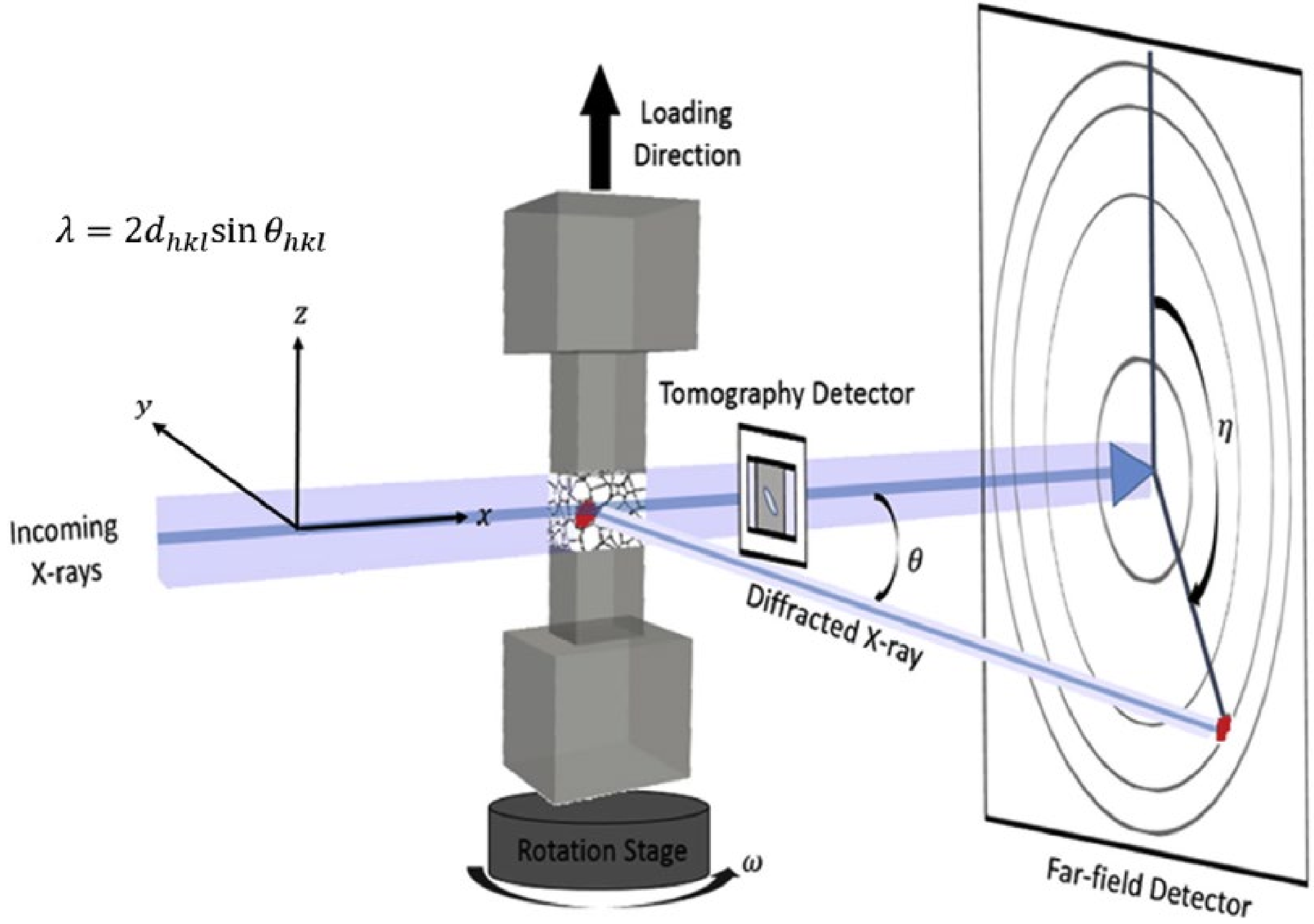
Argonne NATIONAL LABORATORY

FATIGUE IN EMBEDDED INCLUSION

Experimental Details



E=65.3 keV

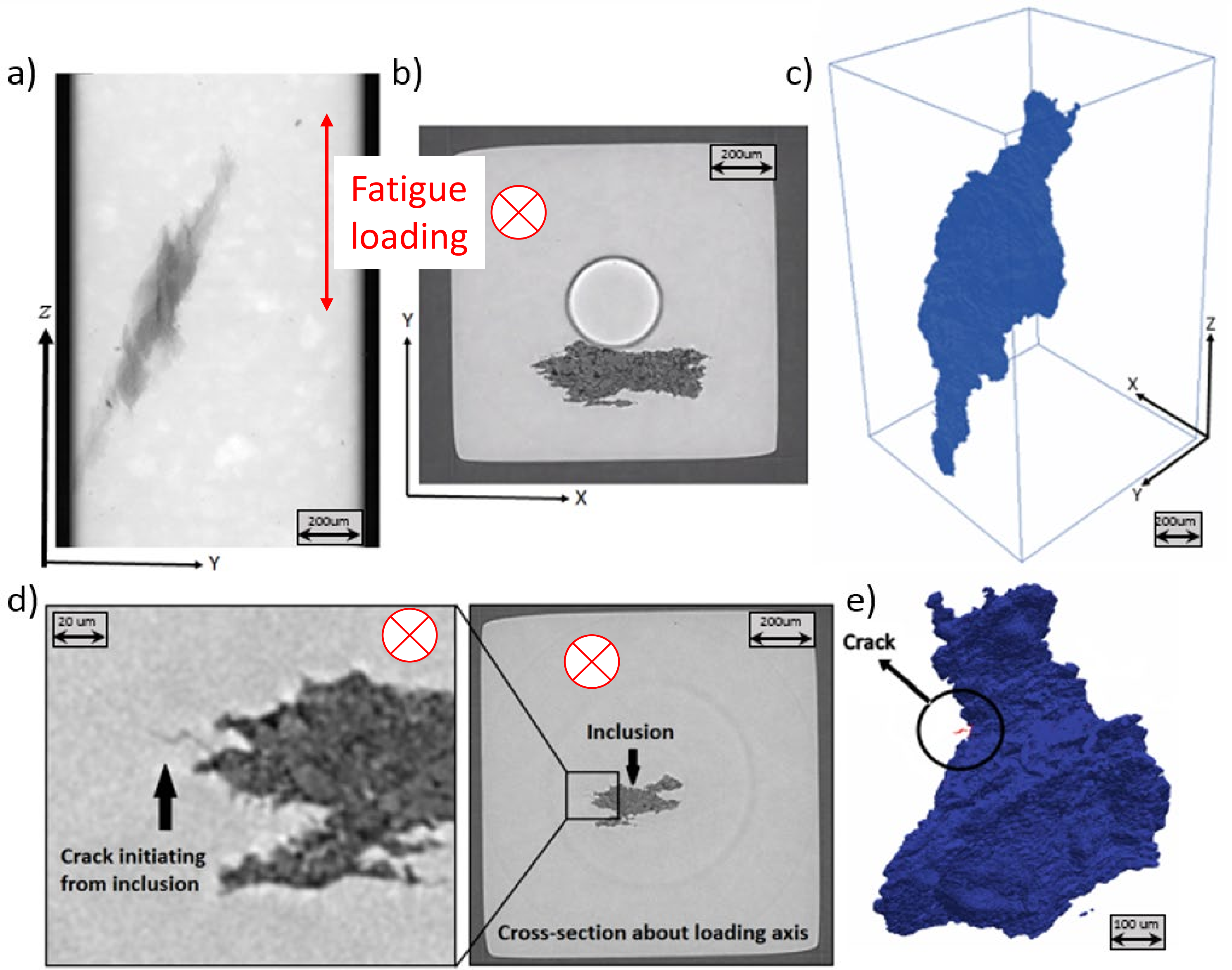
Direct beam tomography
- 1.5 (H) x 1(V) mm2 beamsize
- 1800 images over 360 deg
- ~5 min per 1 mm3 volume

Far-field HEDM
- 1.5(H) x 0.1 (V) mm2 beamsize
- 720 images over 180 deg
- ~10 min per 0.1mm3 volume (~2k grains)
- Build up larger volumes by vertical translation

Fatigue loading
- RAMS device (in-grip rotation)
- Interrupt at several fatigue cycles for x-ray measurements at min/max load
- N=1,2,5,10,20,50,100,200,500,1k,2k,5k,10k

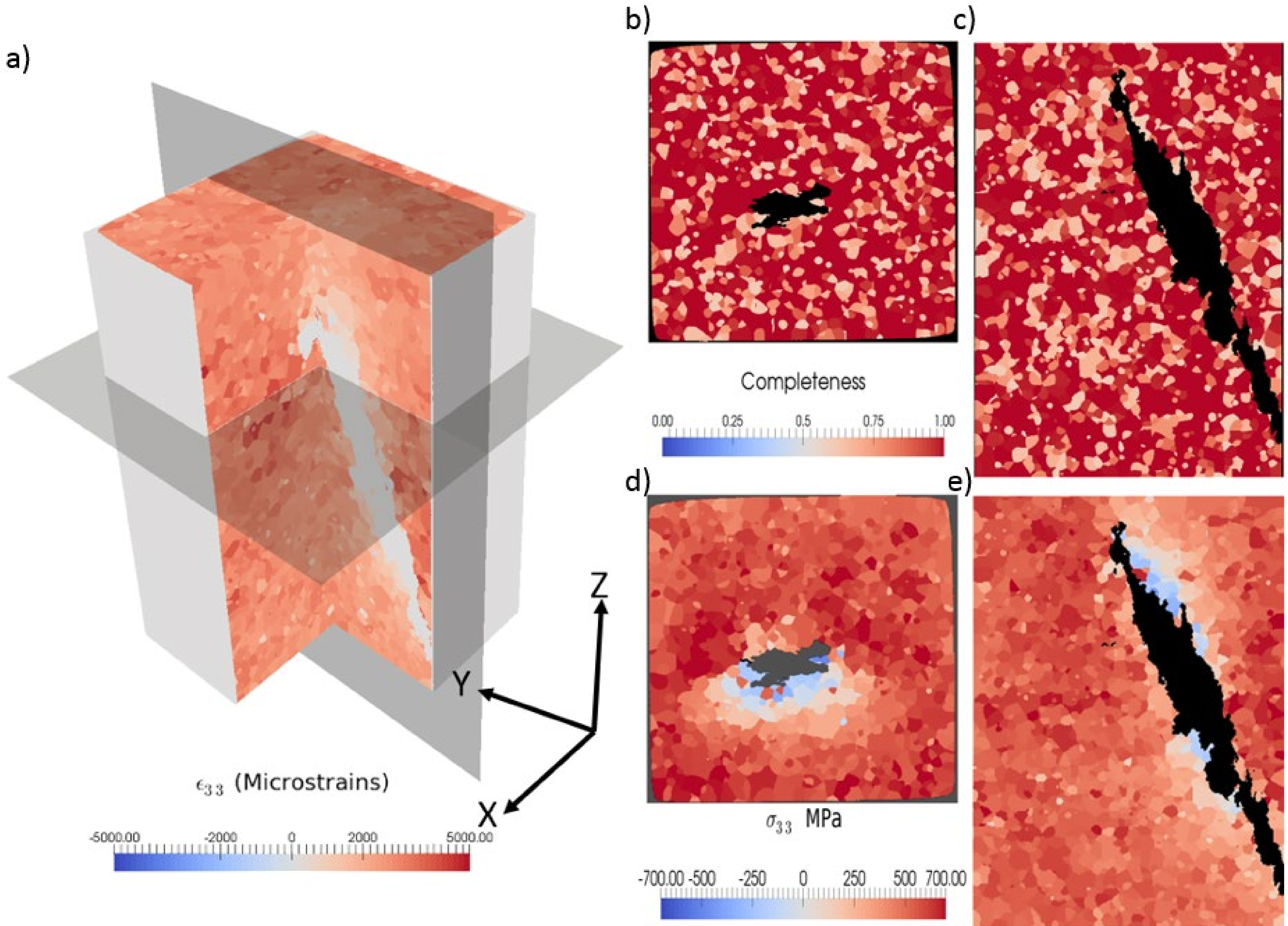
Al2O3 inclusion (~1.3mm tall) seeded in Ni superalloy (HT to equiaxed ~30um grain size)
Found with ultrasound then EDMd to ~center in 1x1 mm2 cross section sample

FATIGUE IN EMBEDDED INCLUSION: TOMOGRAPHY



- Sample radiograph and renderings of reconstructed volume (resolution ~1.5um).
- Complex shape of inclusion mapped
- After cycling to 10k cycles, a crack was found to initiate at the matrix inclusion interface

FATIGUE IN EMBEDDED INCLUSION: 3D STRAINS



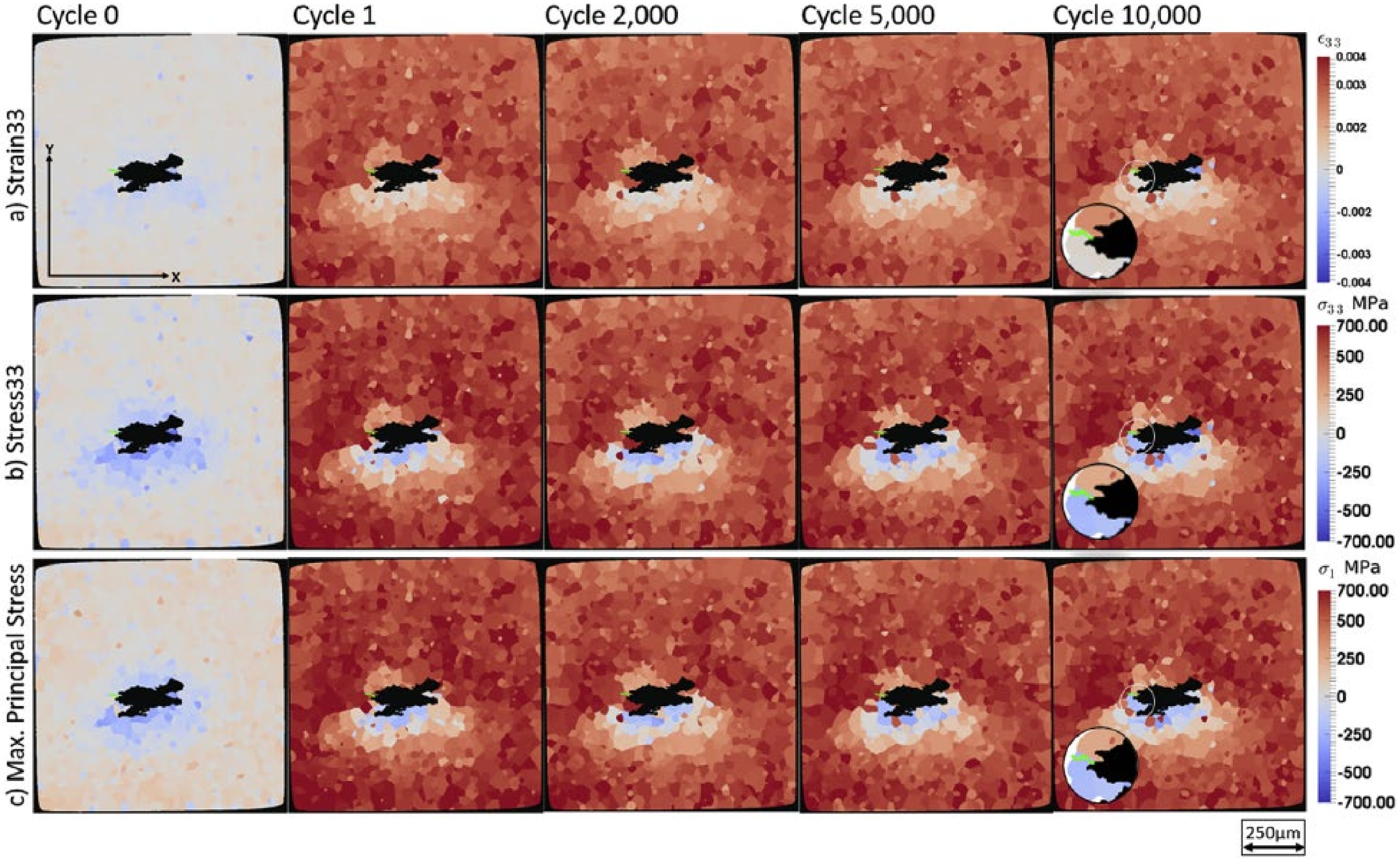
Leuguerre tessellation for grain maps.

Strain tensors for each of ~2k matrix grains per 0.1mm layer.

Significant gradients near Al2O3.

Al2O3 too fine to determine grain-resolved strains.

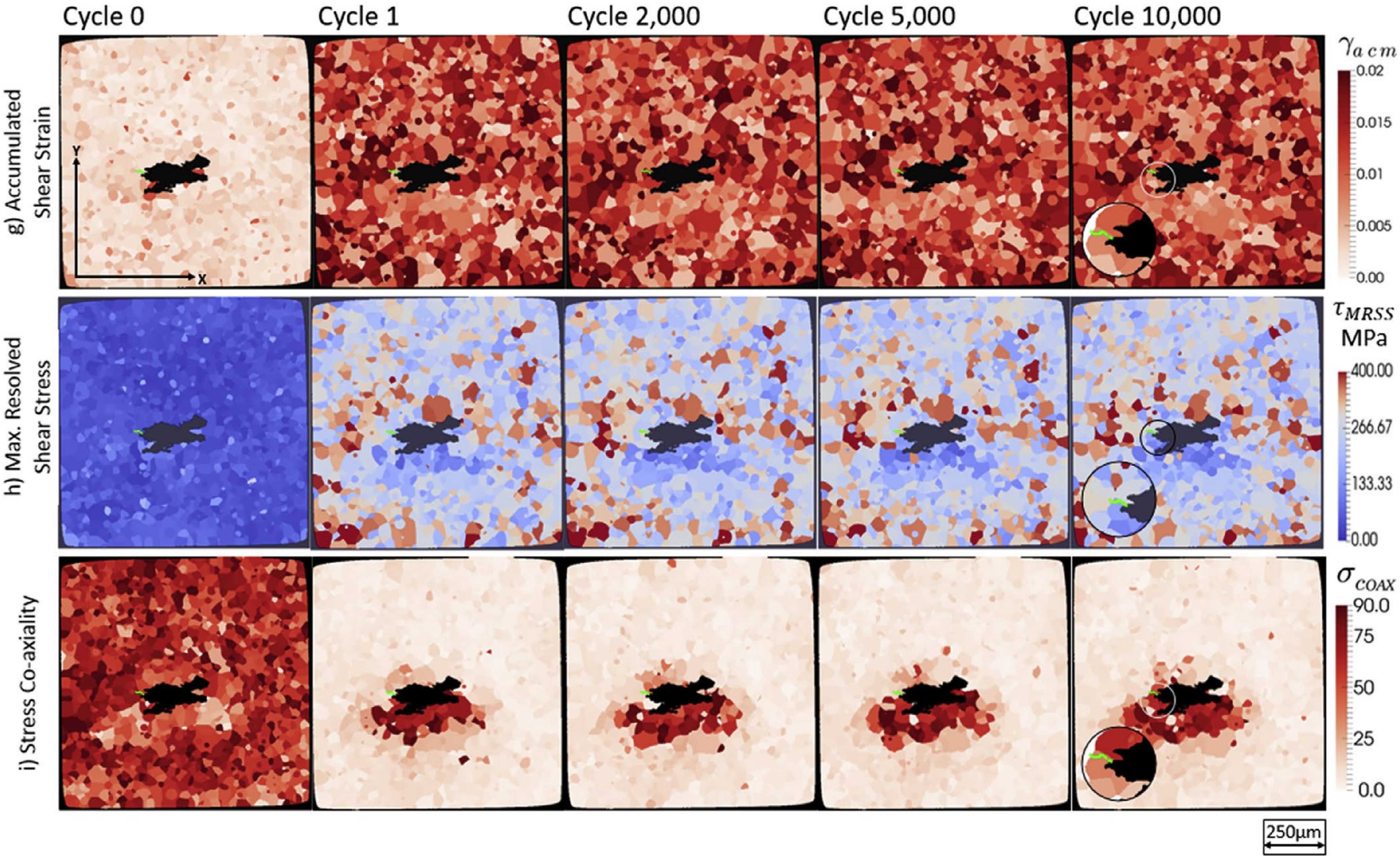
FATIGUE IN EMBEDDED INCLUSION: ε–σ EVOLUTION



ε–σ gradients due to inclusion (starting as residuals)

ε–σ in given grain are ~stable until crack initiation at ~10k cycles

FATIGUE IN EMBEDDED INCLUSION: ε–σ EVOLUTION

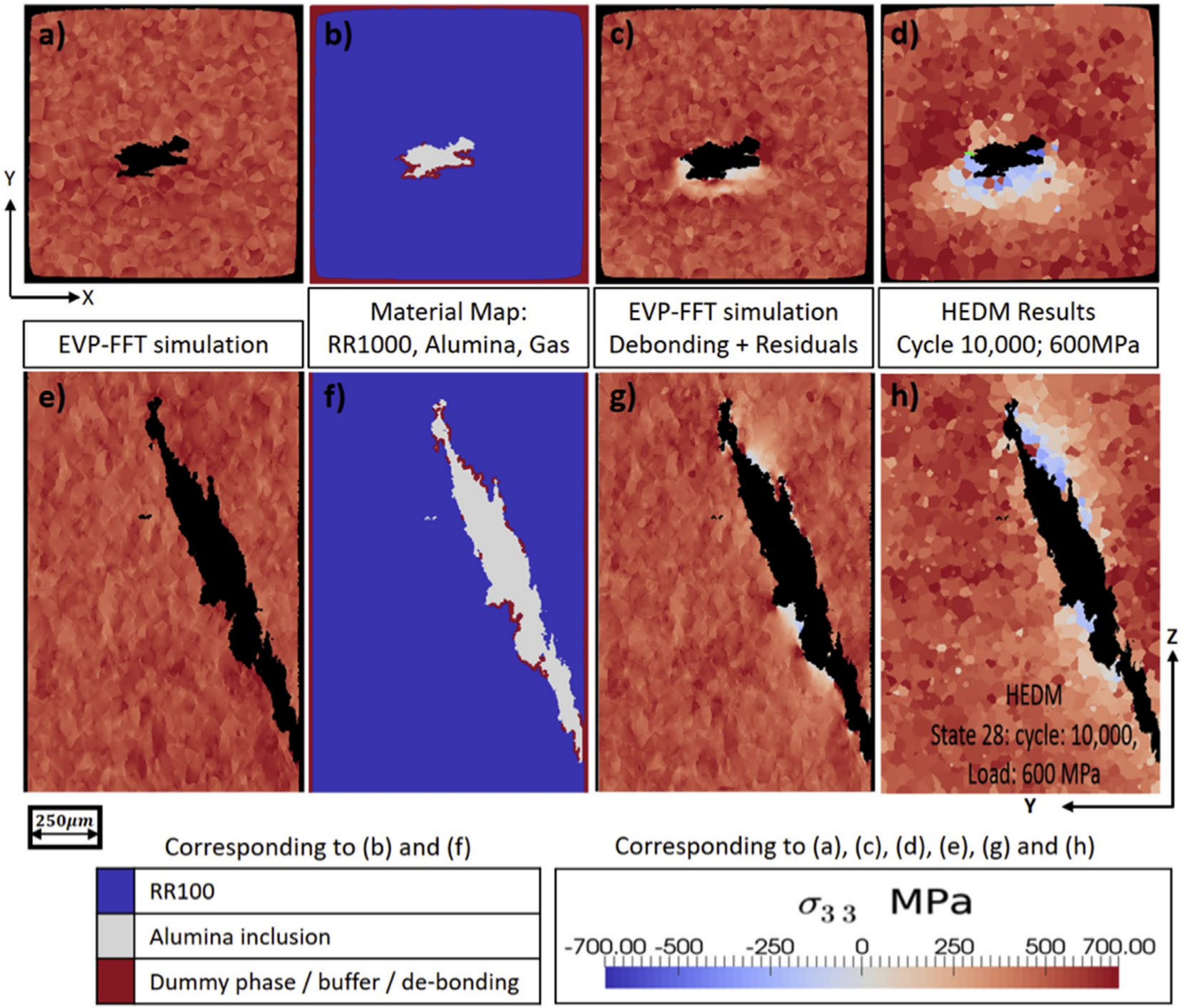


High shear stress and co-axiality gradients at intergranular crack site

Good indicators of crack initiation

FATIGUE IN EMBEDDED INCLUSION: STRESSES || LOADING

Elasto-viscoplastic FFT grain-level model



When model incorporates:

- Microstructure & geometry
  - from tomography & ff-HEDM
- Residual stresses
  - from ff-HEDM @ no load
- Debonding
  - From ff-HEDM @ load

Grain-level stresses compare well to those from ff-HEDM

Enhances ability to locate hot-spots (e.g. crack formation) eg: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ELSEVIER

Full length article

X-ray characterization of the micromechanical response ahead of a propagating small fatigue crack in a Ni-based superalloy

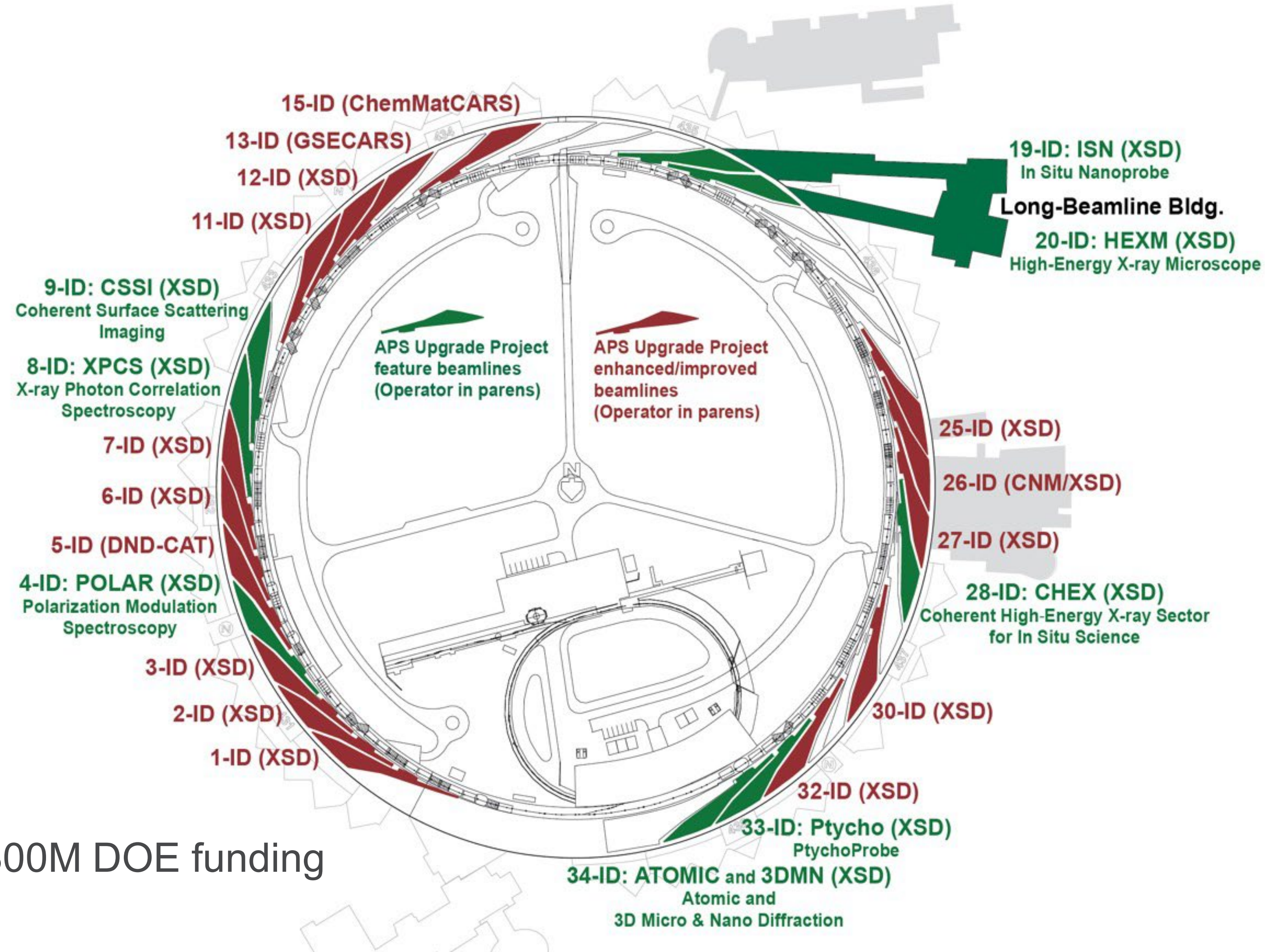
Diwakar P. Naragani [a], Paul A. Shade [b], Peter Kenesei [c], Hemant Sharma [c], Michael D. Sangid [a,*]

[a] School of Aeronautics and Astronautics, Purdue University, West Lafayette, IN, 47907, USA
[b] Materials and Manufacturing Directorate, Air Force Research Laboratory, Wright-Patterson AFB, OH, 45433, USA
[c] Advanced Photon Source, Argonne National Laboratory, Argonne, IL, 60439, USA

OUTLOOK: APS UPGRADE AND NEW HEX CAPABILITIES

APS UPGRADE PROJECT (APS-U)

Dark-period April 2023-June 2024



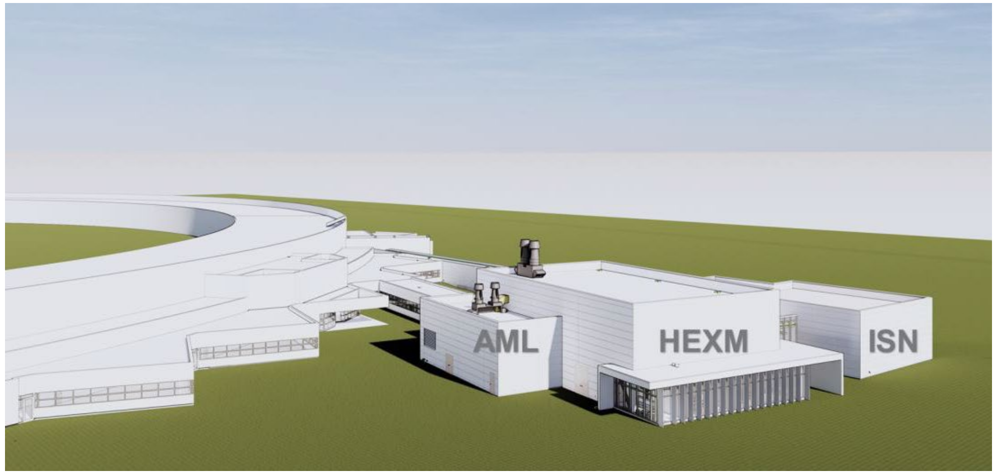
~800M DOE funding

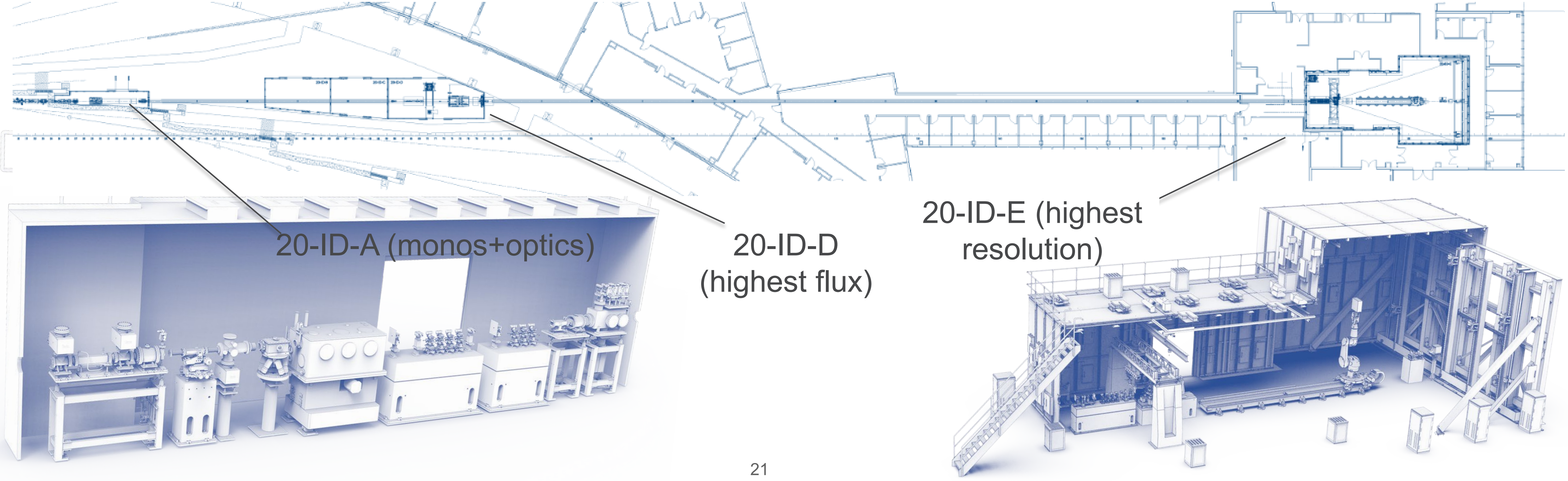
- New storage ring, **42 pm** emittance @ 6 GeV, 200 mA
- New and updated insertion devices, including SCUs
- Combined result in brightness increases of up to 500x
- 9 new feature beamlines + Long Beamline Building
- 15 enhanced and improved beamlines
- Exploit high performance computing, AI

HEXM 'first experiments' discussion @ APS-Users Meeting May 4 2023 (virtual, free registration)

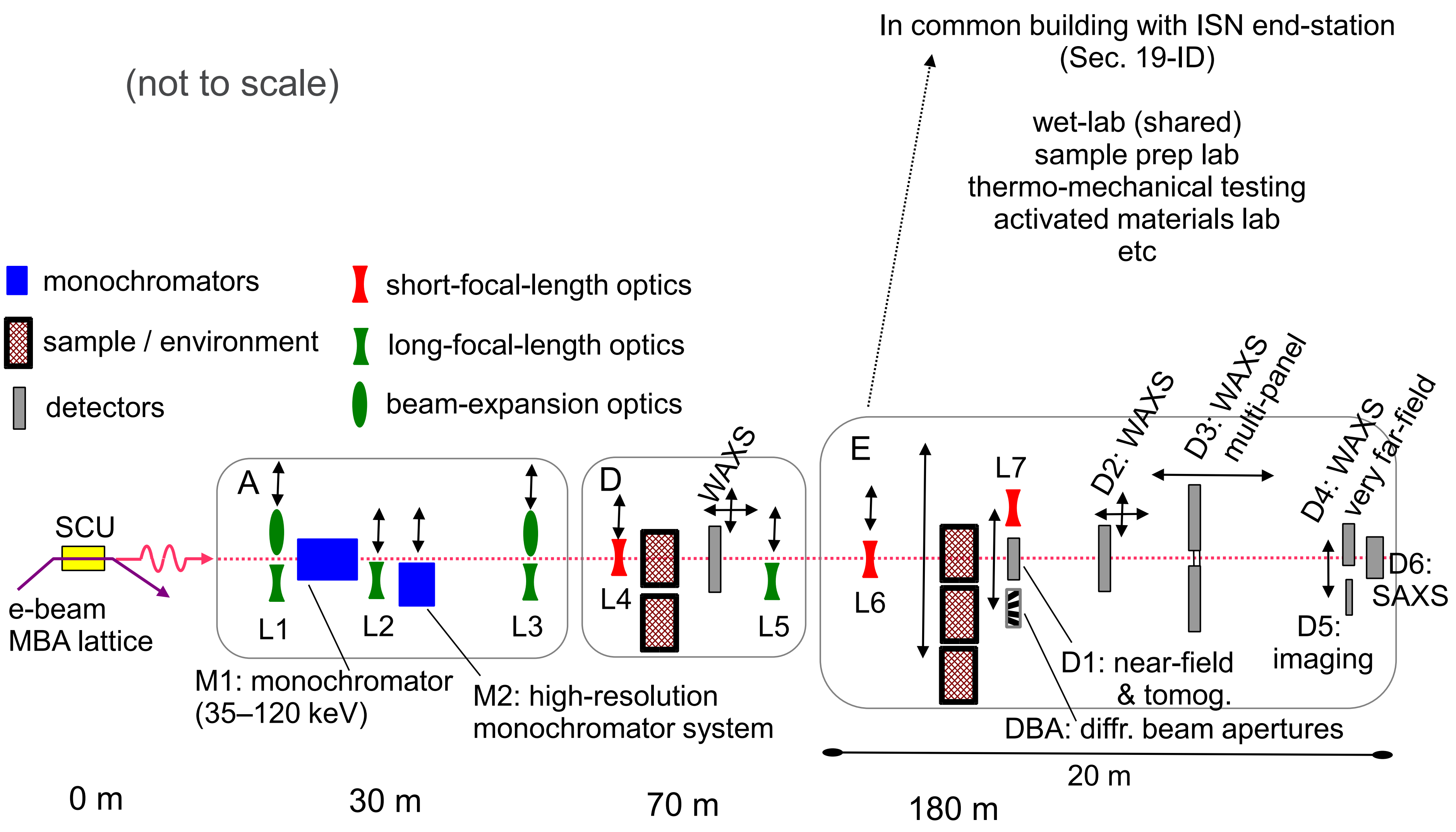
HEXM BEAMLINE @ 20-ID

- One of two ‘long beamlines’ under APS-U
- Two new white-beam hutches, at nominal distances D@70 meters and E@180m
- 20-ID-E in Long Beamline Building shared with 19-ID In Situ Nanoprobe
- Activated Materials Lab next to 20-ID-E (NSUF/DOE-NE)





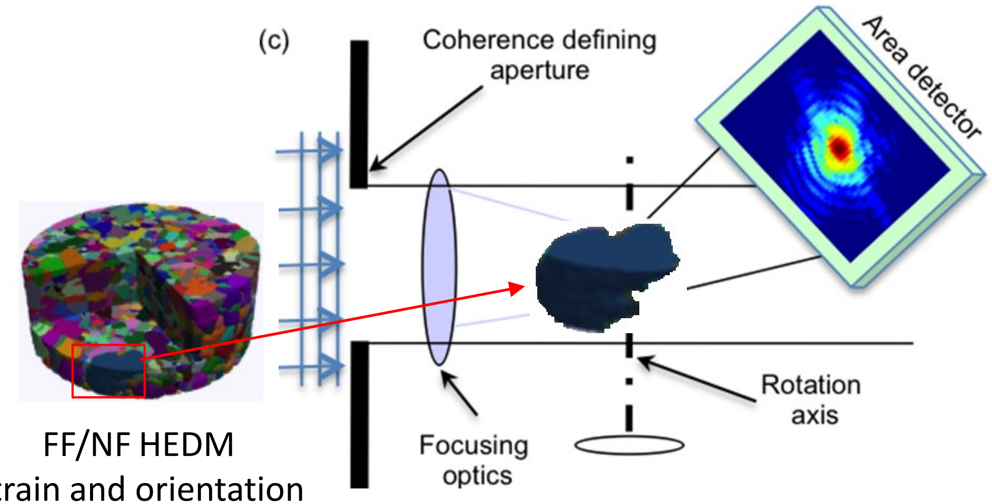
HEXM BEAMLINE @ 20-ID



DARK-FIELD IMAGING

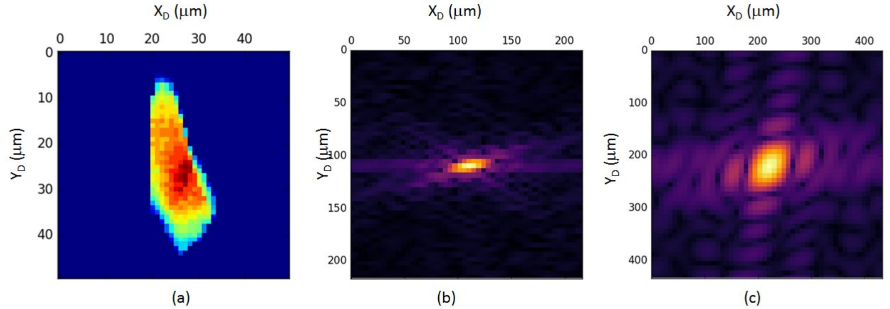
Enabled by new sources & beamlines: HEXM @ APS-U

- Lensless: BCDI -> MBA transverse coherence at 50 keV will match that of todays APS at 10keV
- Lens-based: Dark-field microscopy (DTU, ESRF, APS-LDRD)
- Enabled by:
  - MBA Coherence, pre (post) sample lens optics, large hutch, high E resolution, high resolution detectors
- Explore various measurement strategies (e.g. ptychographic) and reconstruction algorithms
- 'Zoom-in' complement to today's HEDM : access to sub-grain & nano-grain information not achievable today
- Helping users choose technique(s)/modes to use is a challenge we are addressing



FF/NF HEDM
Strain and orientation information

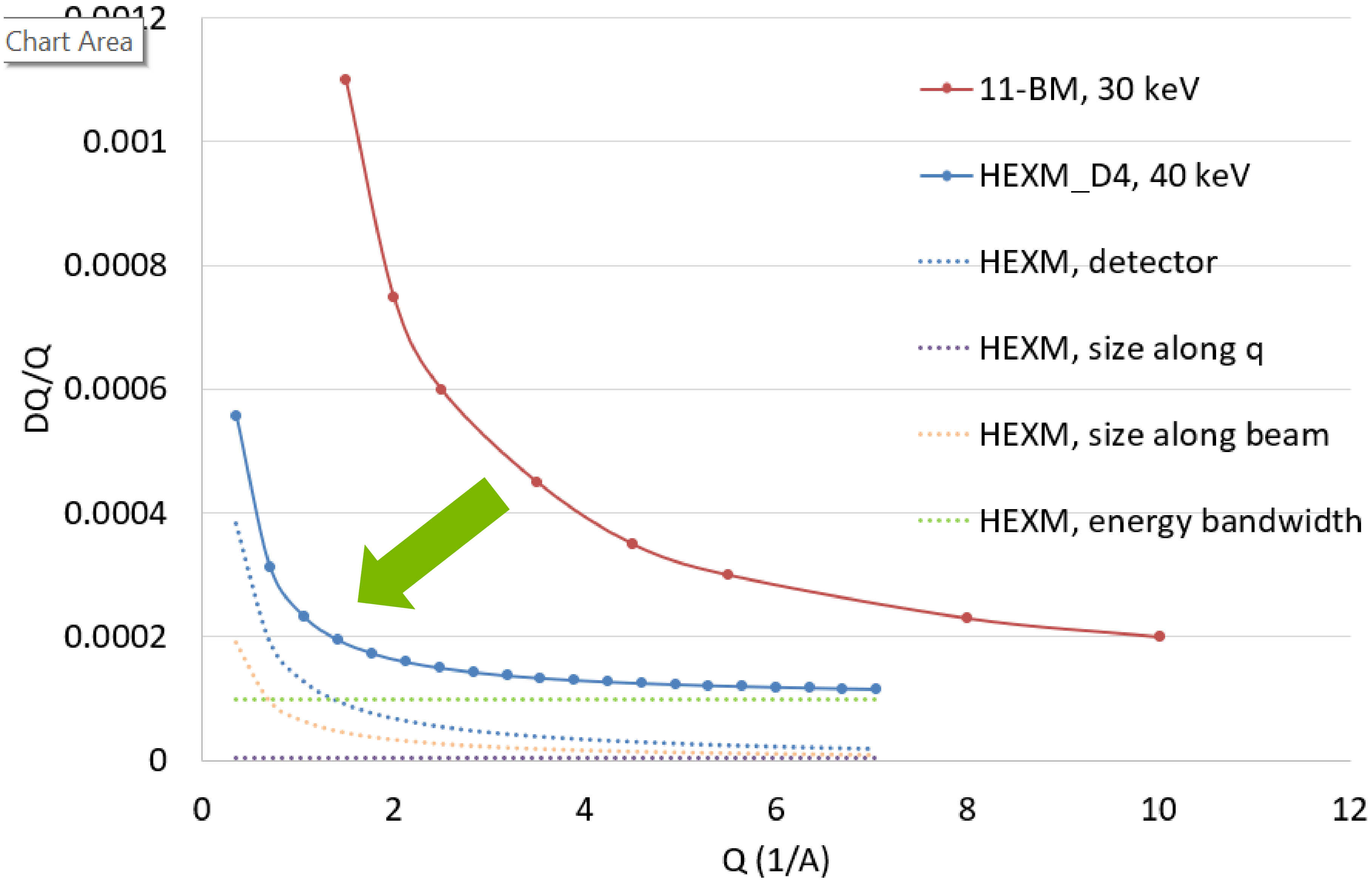
Simulations of BCDI patterns at E = 40 keV, sample-detector = 14 m, detector resolution = 10 μm, $2\theta = 10^o$, grain size = 10 μm. (a) 2D shape of the grain along the projection direction. (b) The grain is fully illuminated using perfectly coherent 50 μm. (c) The grain is partially illuminated with pytchographic measurement strategy in mind.



Maddali et al, "Sparse recovery of undersampled intensity patterns for coherent diffraction imaging at high X-ray energies", Sci Rep. 8 (1) (2018)

RECIPROCAL SPACE RESOLUTION

HEXM @15m samp-det, collimated beam



(10um pixel size)

(50um grain)

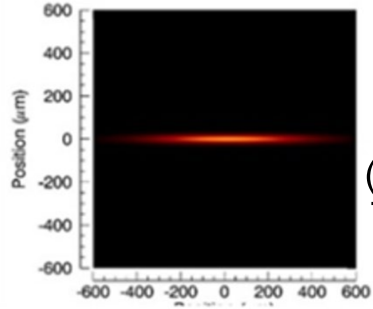
($\Delta E/E=1e-4$)

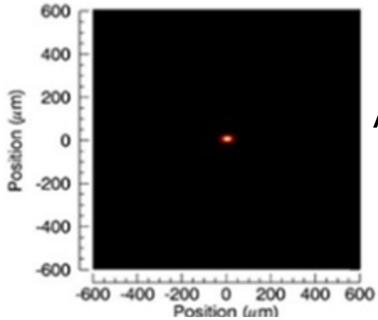
note $\Delta E/E$ to 3e-5 should be possible

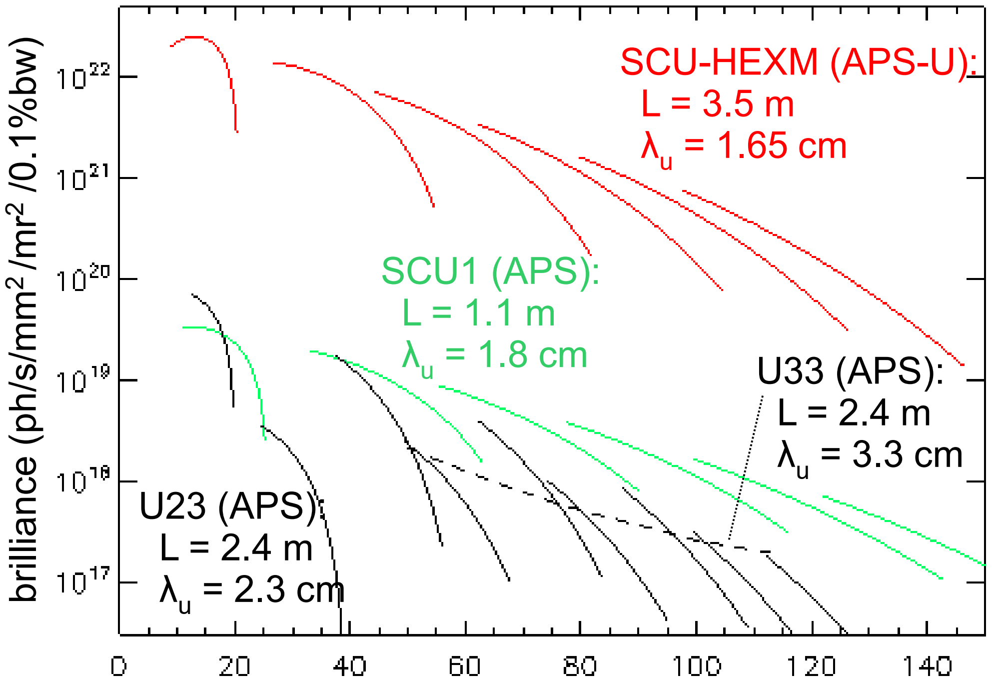
EXPERIMENTAL TECHNIQUE GAIN FACTORS

- Gains at 70 keV relative to today's 70 m distance at 1-ID-E with SCU1
- SCU1: 1.08 m long, 1.8 cm period, n = 5
- Future SCU-HEXM: 3.5 m long, 1.65 cm period, n = 7

Figure of merit	Experiment	"flat" (70 m)	"flat" (180 m)	"round" (180 m)	timing "round" (180 m)
central cone flux	- large unfocused beam **(tomography)**	4.6	4.6	4.4	4.3
flux density	- apertured unfocused beam - 2D-focused flux (spot size unimportant) **(ff-HEDM, SAXS, PDF, fluorescence)**	10	1.5	1.3	1.0
brilliance, coherent flux	- 2D-focused flux density (credit for spot size) **(ST, ff-HEDM, SAXS, PDF, Fluorescence)** - coherence **(BCDI)**	530	530	250	190
vertical flux density	- line-focused flux (focal width unimportant) **(nf-HEDM)**	4.6	1.8	1.4	1.3
vertical brilliance	- 1-D focused flux density (credit for focal width) **(nf-HEDM)**	13.6	13.6	5.2	4.5



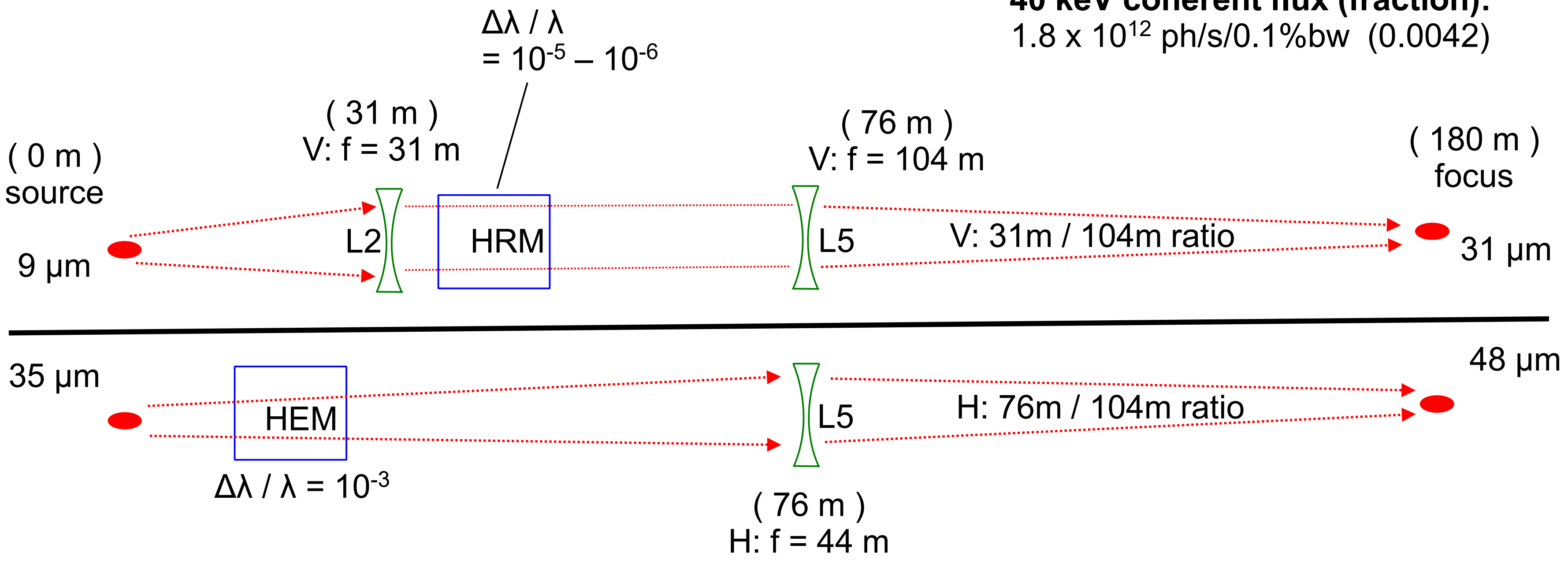




LARGE, COHERENT FOCAL SPOTS (ZOOM-IN)

- Bragg coherent diffraction imaging (BCDI) on relatively large grains (tens of μm) requires similarly sized coherent focal spots.
- **MBA lattice (“flat”)** source offers ~ x 500 (150) - fold improved coherent flux (fraction), but requires low-demag, or even magnification geometries with respect to the small source.
- A long beamline facilitates such a configuration.

40 keV coherent flux (fraction):
1.8 x 10^{12} ph/s/0.1%bw (0.0042)

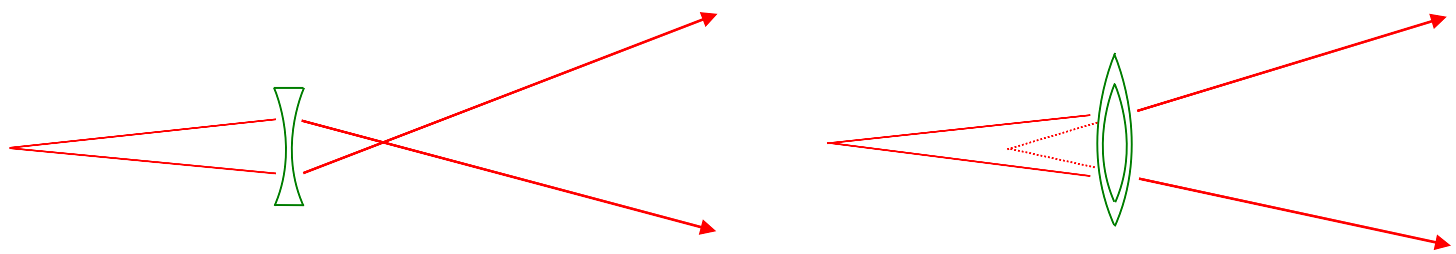


For a 70 m beamline, such large focal spots (in vertical) are not achievable.

BEAM-EXPANDING REFRACTIVE OPTICS (ZOOM-OUT)

- Some techniques requires a large beam in one or both directions:
  – standard μ-CT
  – nf-HEDM uses line-focus, vertical ≤ 1 μm, many mm horizontal

- Beam expanders are efficient on a long beamline with a beam that starts small:
  – low refractive power needed
  – small aperture needed

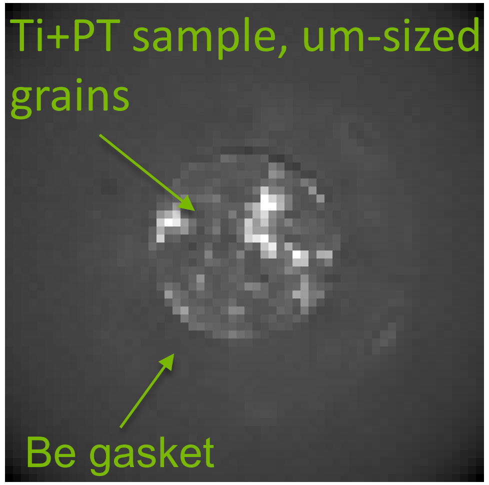


MBA - 41 pm brightness mode, 3.5 m SCU, 70 keV

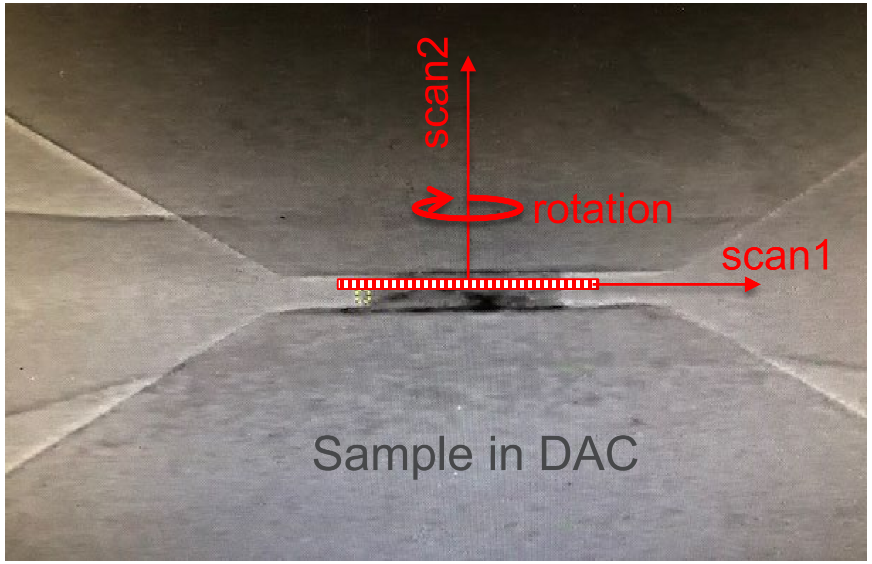
horizontal beam size at 35 m:	0.47 mm FWHM
horizontal beam size at 180 m:	2.46 mm FWHM
expand with:	diamond lens f = - 55.2 m at 34 m 0.6 mm aperture
expanded size at 180 m:	4.4 mm FWHM
efficiency:	69 %

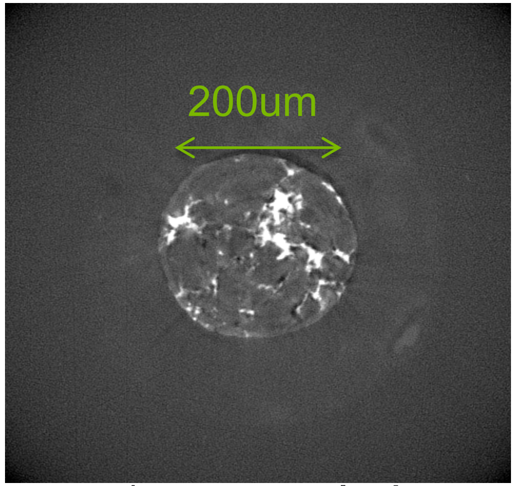
For a 70 m beamline, beam expanders are less efficient, not straightforward.

MULTI-MODE IMAGING IN EXTREME ENVIRONMENTS

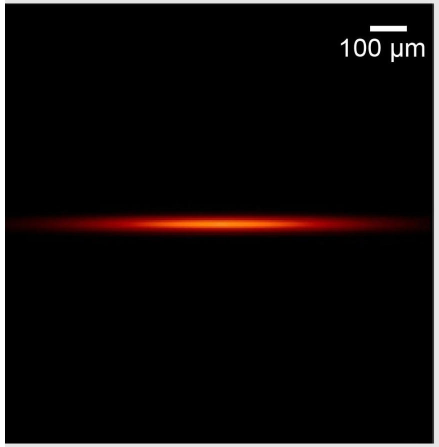


10um resolution



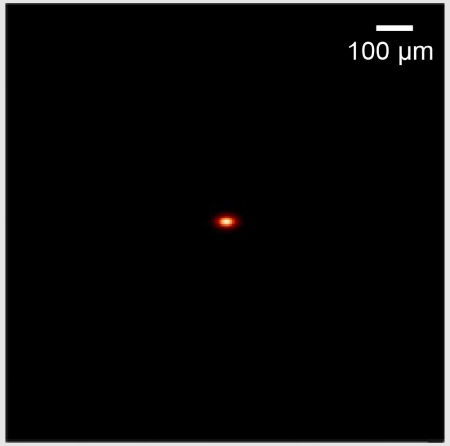


1 um resolution



APS Beam

APS U



APS-U Beam

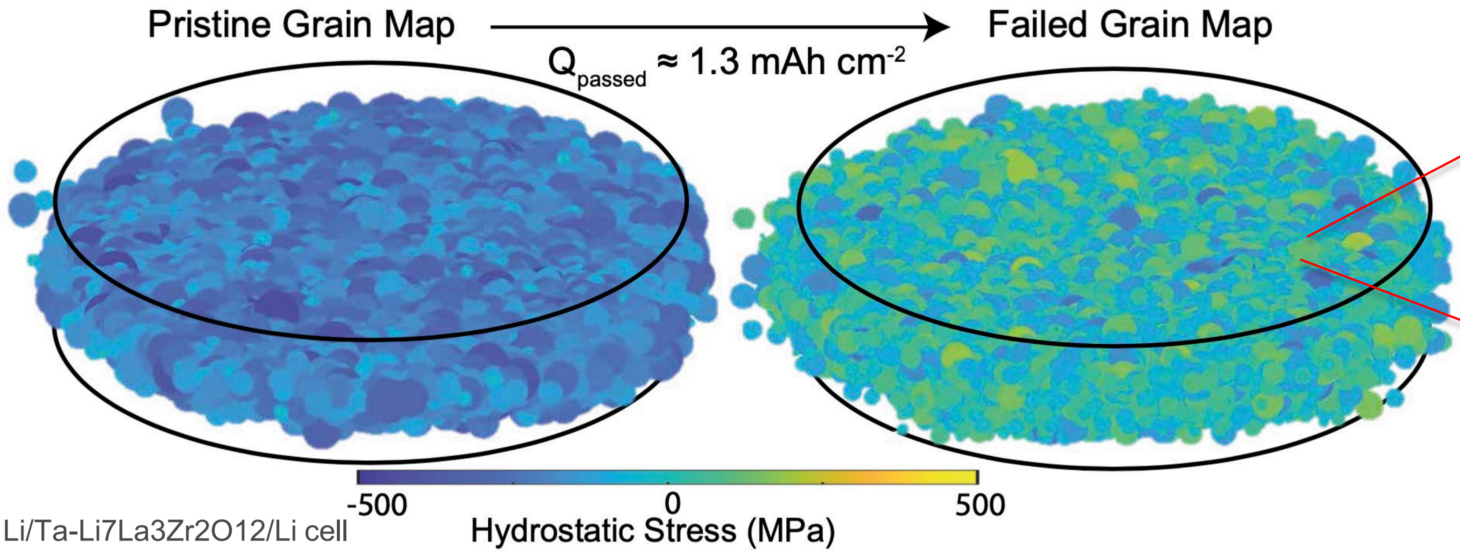
Rotate and Scan (OD, 1D or 2D)

- Smaller APS-U horizontal beamsize -> enhanced resolution/flux
- Voxellized scattering information (phases, strains, etc):
  - Higher resolution for scattering tomo (shown)
  - Higher disorder for PF-HEDM (grain-resolved)

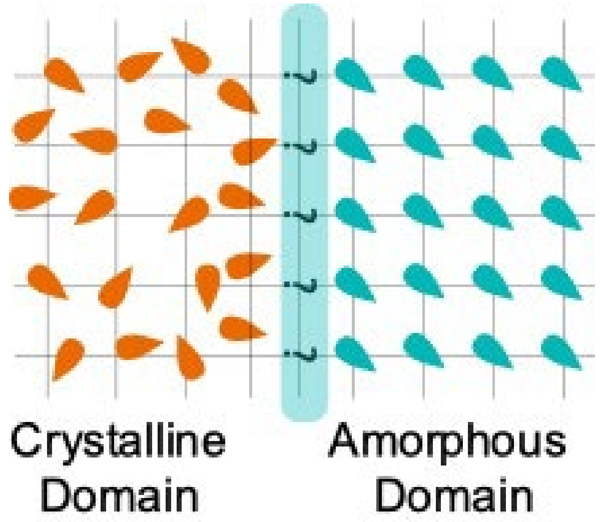
LDRD 2021-3: “High pressure material characterization in 3-dimensions using X-ray diffraction-contrast computed tomography”, Y. Meng, C. Chuang, H. Sharma

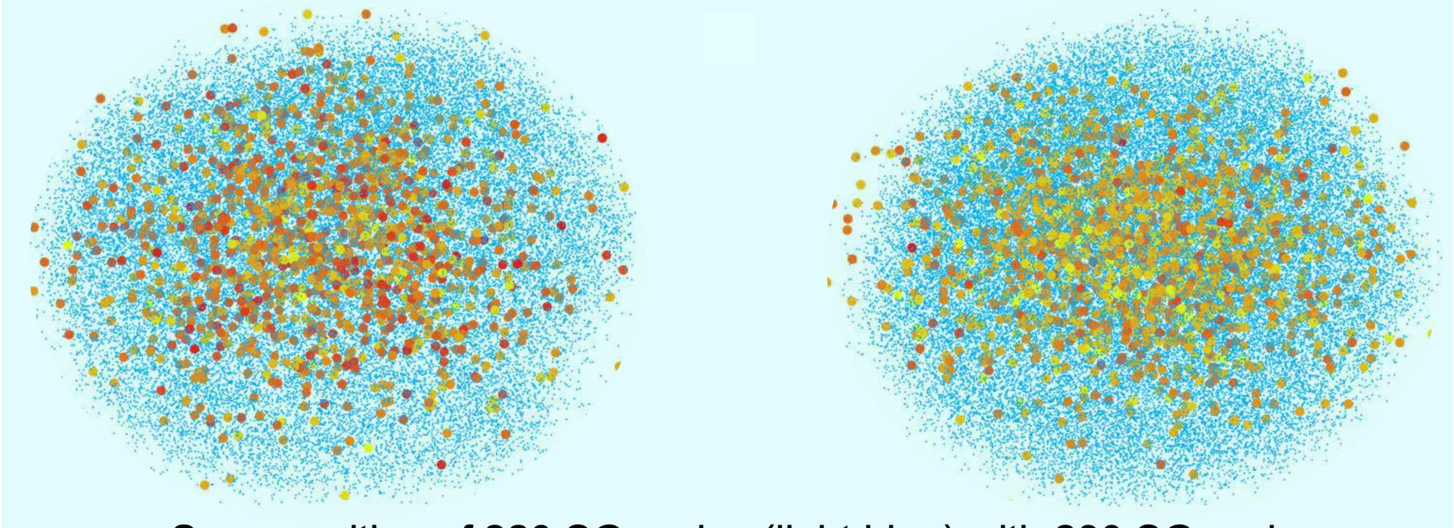
“Concurrent determination of nanocrystal shape and amorphous phases in complex materials by diffraction scattering computed tomography”, M.E. Birkback et al, JAC 50(1), 2017

MULTI-MODE IMAGING OF SOLID STATE BATTERIES



APS-U




Superposition of 220 SG grains (light blue) with 230 SG grains

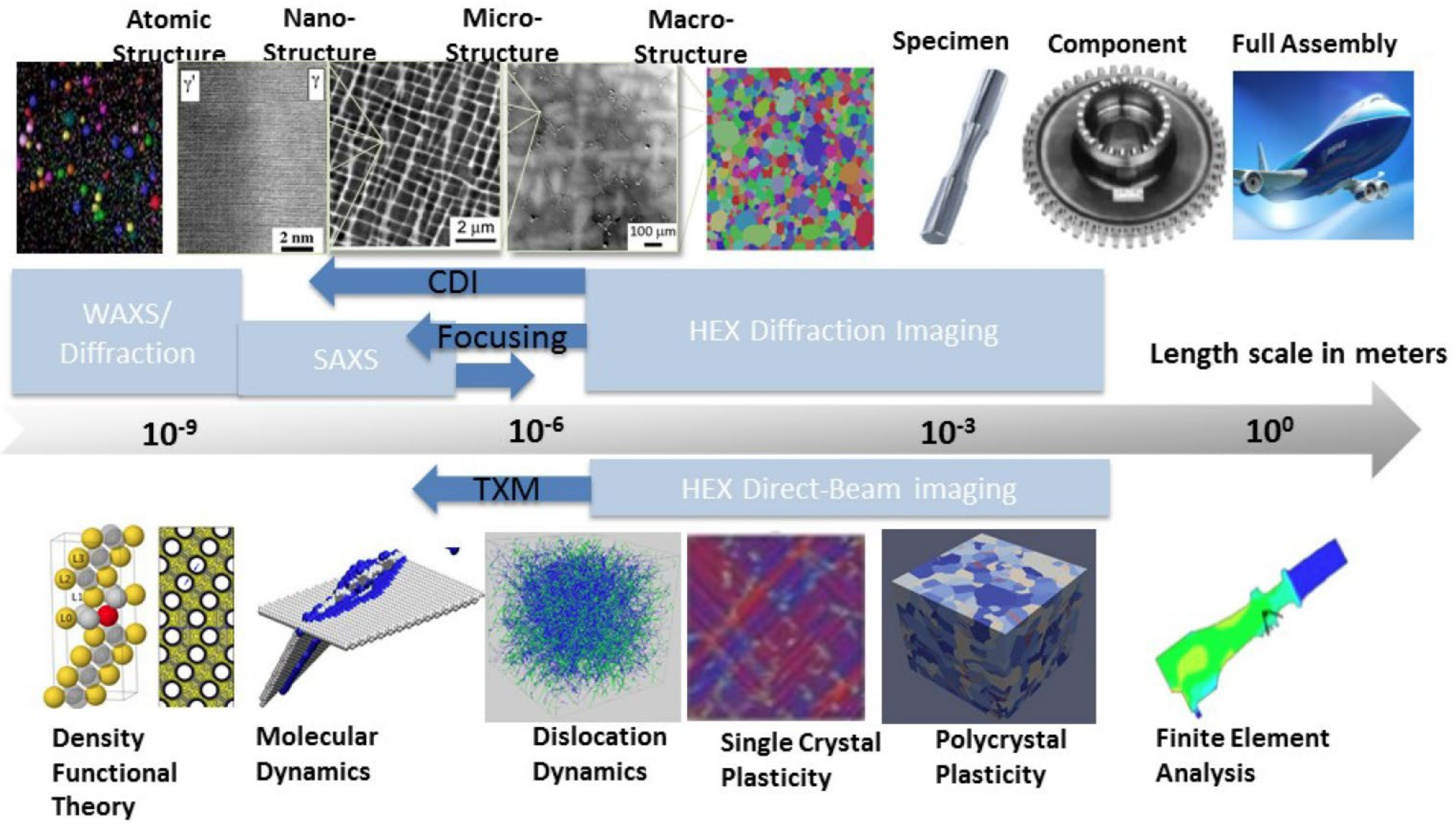
M.B. Dixit et al, 'Status and Prospect of in-situ and operando characterization of solid-state batteries', Engineering & Environmental Science 14 (9), 2021.

- SSB materials -> complex interfaces: amorphous/crystalline, polymorphic
- Decomposition reactions at interfaces (thermally/electrochemically driven) lead to disparate regions affecting transport
- Zoom-out -> identify "problem" zones
- Zoom-in -> probe inter/intra-grain dynamics as well as grain boundaries

2mm diam. samples in-operando: 'real' electrochemical conditions

HEXM (20-ID) & 1-ID: MULTI-SCALE, MULTI-MODAL IMAGING

- Exploit MBA-emittance for world-leading high-energy x-ray characterization
- Bridge current length-scale gaps through direct- and reciprocal-space methods
- Multi-modal techniques: combinations of spatial zoom-in/out methods -> including NEW HE-coherence based



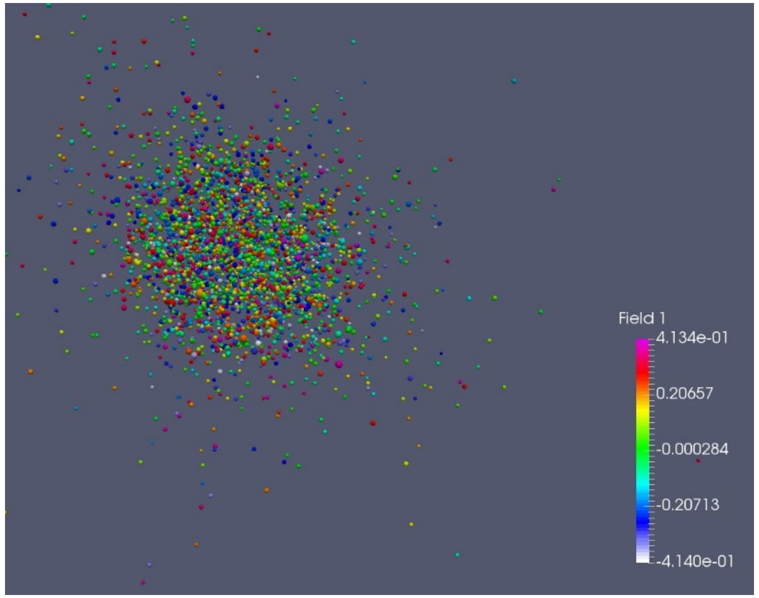
Scientific cases for hierarchical needs include: aerospace metals, batteries, SOFCs, nuclear materials and bio-materials

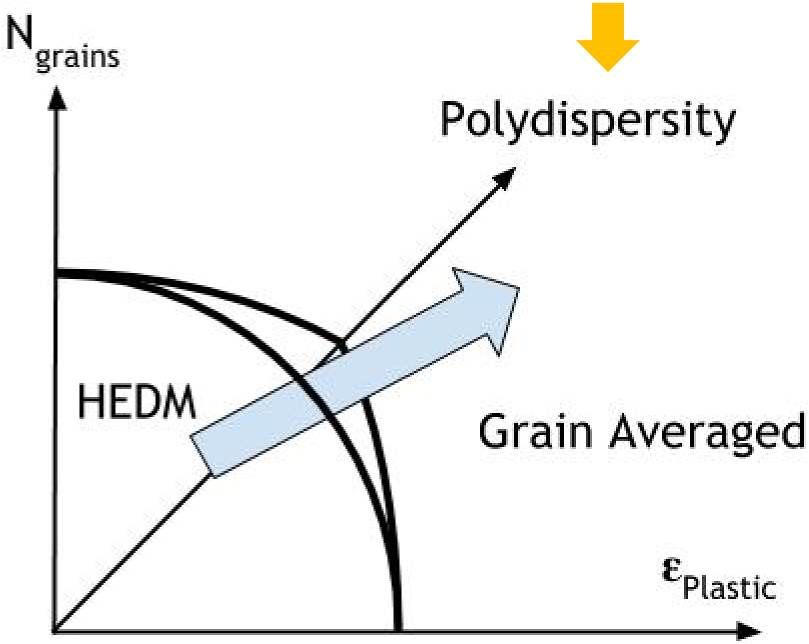
HEDM (GRAIN-RESOLVED) LIMITS

Wish to push HEDM ‘envelope’ out
- Software to treat spot overlap (MIDAS)
- Improved detectors, energy resolution, focusing

Diffraction intensity proportional to grain volume:
- Now use GE-RT41 with dynamic range 1e12
  - 10:1 max ratio in grain size (1000:1 in intensity)
- Pilatus (CdTe) have DR 1e20 (at least 100:1 possible)

Monodispersed LaB6 ~1um grain size reconstructed with 10k grains
Battery materials at limit:1um vertical slice of Tin electrode (~0.5-5um grains)

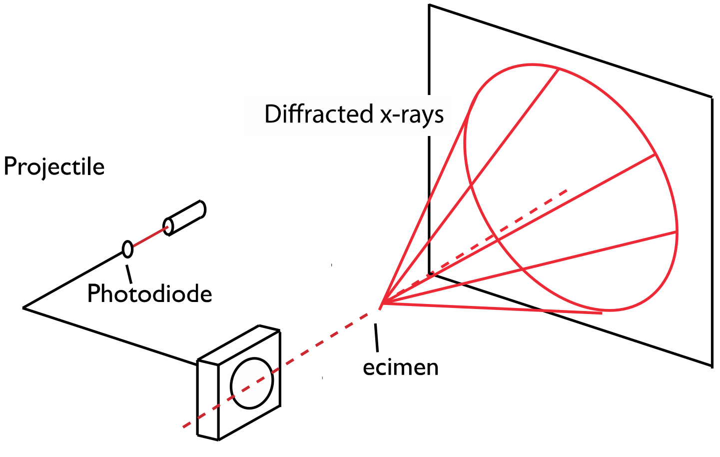


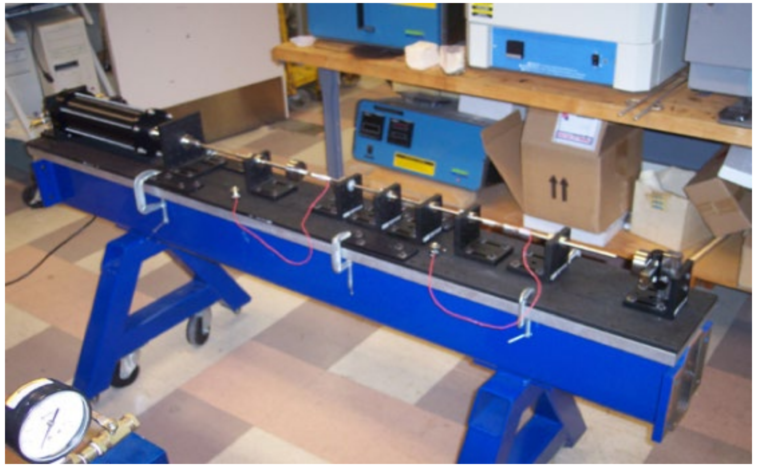


Higher plastic strains using 2D focused beams and raster scanning and/or DFM techniques

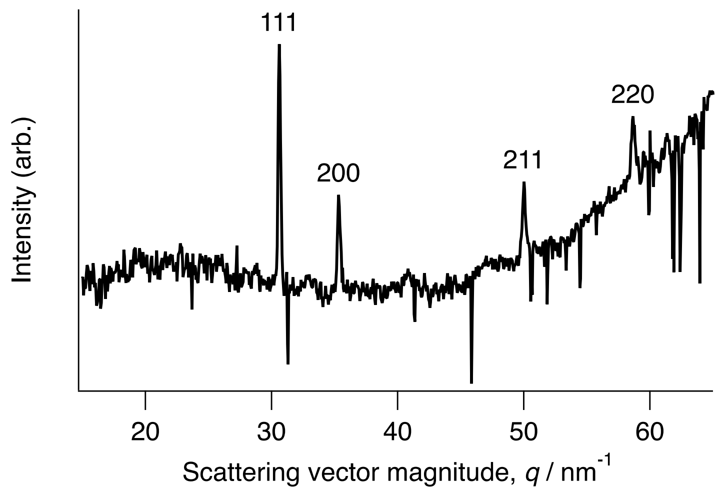
TEMPORAL LIMITS

In situ diffraction during dynamic deformation





Integrated lineout from Monel:(40 μs exposure)



Today (since these measurements):

- **~3x more flux (SCU); confirmed during in-situ AM**

APS-U

- **5-10x additional flux (pink beam)**

~ 1μs should be possible based on flux limit

Detector readouts (continuous):

- **GE a-Si==200 ms**
- **Dexela CMOS==50 ms**
- **Pilatus CdTe==2ms**
- **Lambda==1ms**

New HEX detectors needed to take fullest advantage of flux

P. Lambert, T. Hufnagel, K. Vecchio et al, Rev. Sci. Inst 85, 093901 (2014)

ACTIVATED MATERIALS LABORATORY (AML)

Objectives

- Facilitate users to safely conduct experiments on activated materials at APS
- Improve sample accessibility and operational flexibility -> ***enhance scientific productivity and enable expansion of in situ testing capabilities***

Scope and Function

- A Radiological Facility
  - Receiving/shipping samples
  - (Dis)assembling sample holder/containment
  - Testing/maintaining *in-situ* equipment
- Handle nuclear reactor materials and fuels in solid form
- A central lab providing encapsulated Rad samples for characterization at APS beamlines

•Realization

- DOE/NSUF provided funding for construction
- Designed and approved using ALARA principles

Proximity to HEXM 20-ID-E endstation helpful for conducting experiments there, but AML can service any APS beamline

Data Handling and Real Time Processing

- HEXM beamline is projected to produce 250 TB/day (peak) and 4 PB per year
- Planning and R&D is in progress to scale existing HEXM HPC tools and utilize compression
- Heterogeneous computing model - CPU+GPU to handle APS-U

Polaris, > 44 Petaflops
2022

Aurora, > 1 Exaflop
2023

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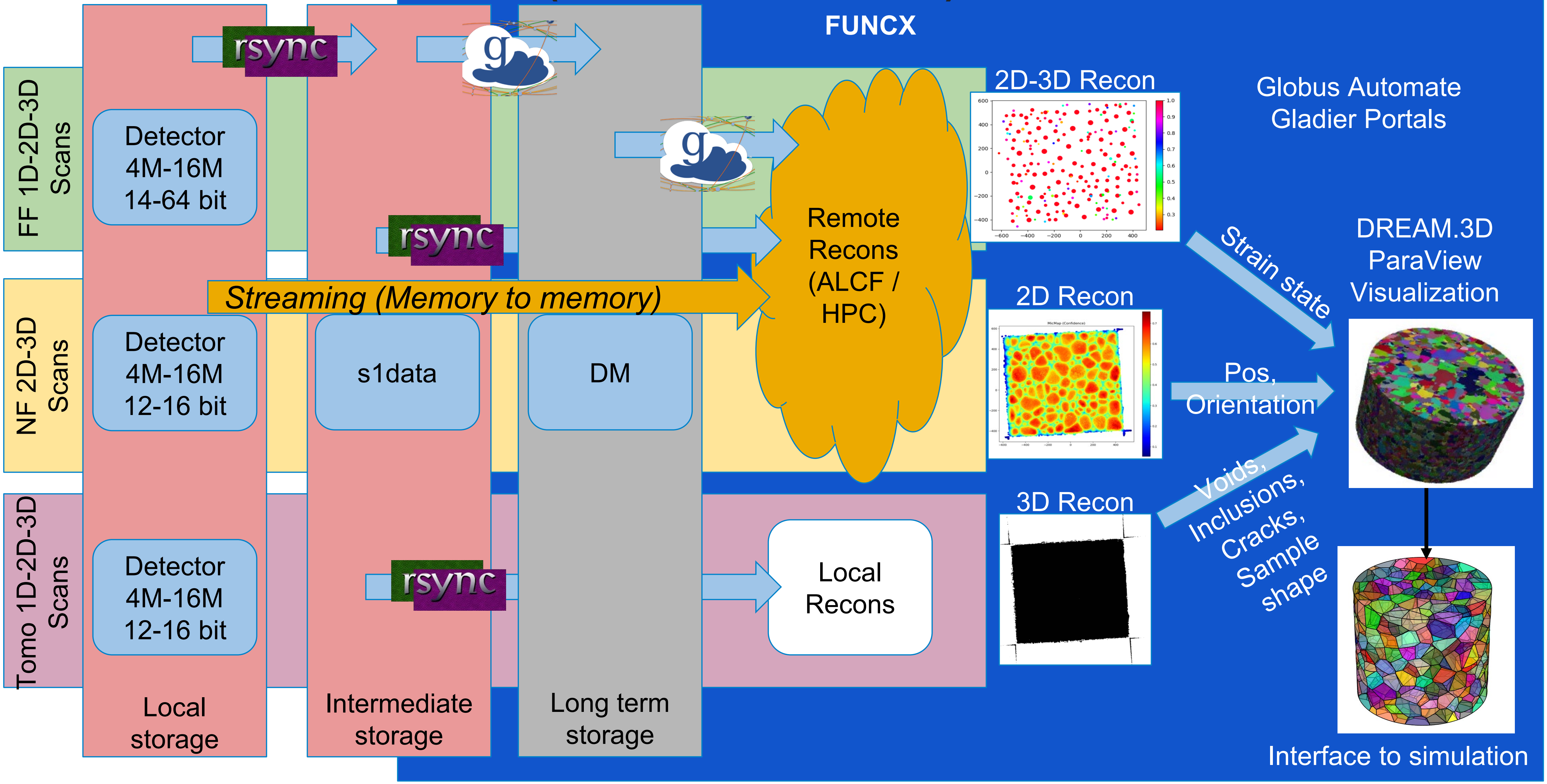
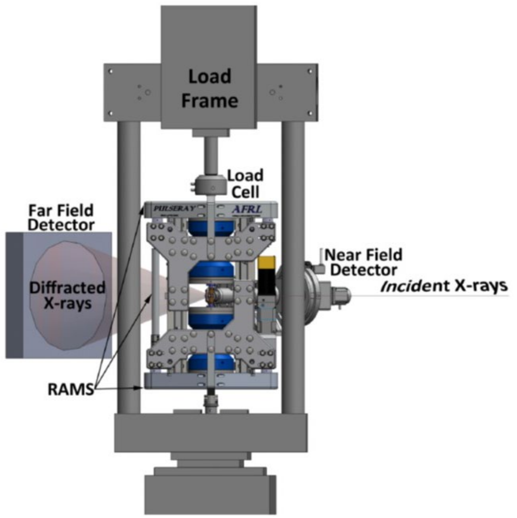
COMPUTE WORKFLOW (IN DEVELOPMENT)

Courtesy of H. Sharma

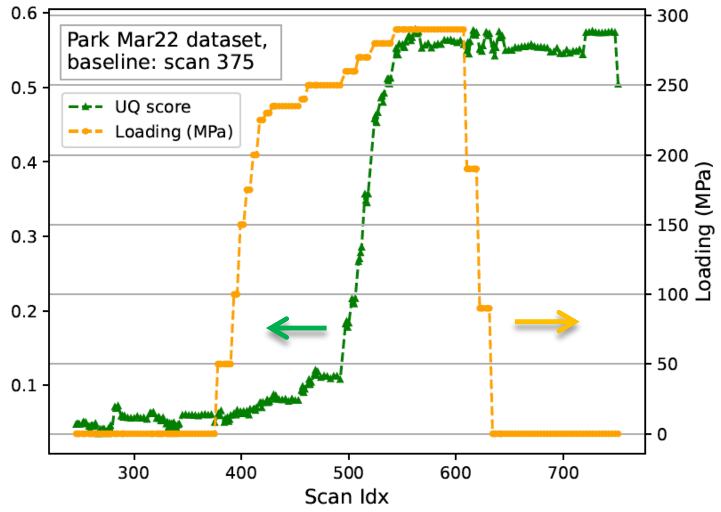


DETECTING DEFORMATION w/ HEDM DATA AND ML

- Can we use AI/ML to rapidly detect structural deformation (e.g., onset of plastic deformation) without complete data reduction?
  - Provide actionable information to the users during the experiment.
- One HEDM dataset per loading ~ 12 gigabyes!
- New ML method for rapid event detection
  - Transform bulky, redundant image dataset into compact, semantic-rich representations of visually salient characteristics.
  - This transformation permits subsequent rapid event detection based on proximity within compact feature spaces.



Ti-7 alloy



Hemant Sharma, Weijian Zheng, Dennis Trujillo, Zhengchun Liu, Peter Kenesei, Jun Park, Nino Miceli, Raj Kettimuthu

SUMMARY

- High energy x-rays provide unique properties:
  - High space and time resolution
  - 3D and (typically) non-destructive
  - A variety of in-situ environments
- Multi-modal studies can provide key scientific insight:
  - Hierarchical materials: bio-materials, aerospace & nuclear alloys, batteries, composites
  - Example of fatigue in embedded inclusion: understanding & enhancing models for crack initiation/growth
- APS Upgrade (as well as others worldwide) will enhance HEX capabilties
  - New beamlines for “smaller needles, bigger haystacks”
  - Starting late 2024 (1 year dark year)
  - Collaborations are essential for highest impact
    - Expert users – technical and scientific commissioning +
    - Non-traditional users – bring science (and we + experts bring techniques+software)