Soft X-ray Science at High Brightness Storage Rings

Coherent Soft X-ray Workshop

Stretch Goals

Soft X-ray Science Opportunities

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ALS Division Deputy for Science

DLSR Workshop
APS/ANL
November 19-21, 2014

https://sites.google.com/a/lbl.gov/coherent-soft-xrayworkshop/home

Charge

1) What transformational research opportunities will be enabled by storage-ring-based ultrahigh brightness soft and intermediate x-ray beams?

2) What are the primary challenges to accomplish this science

Workshop was not directly about the proposed ALS upgrade

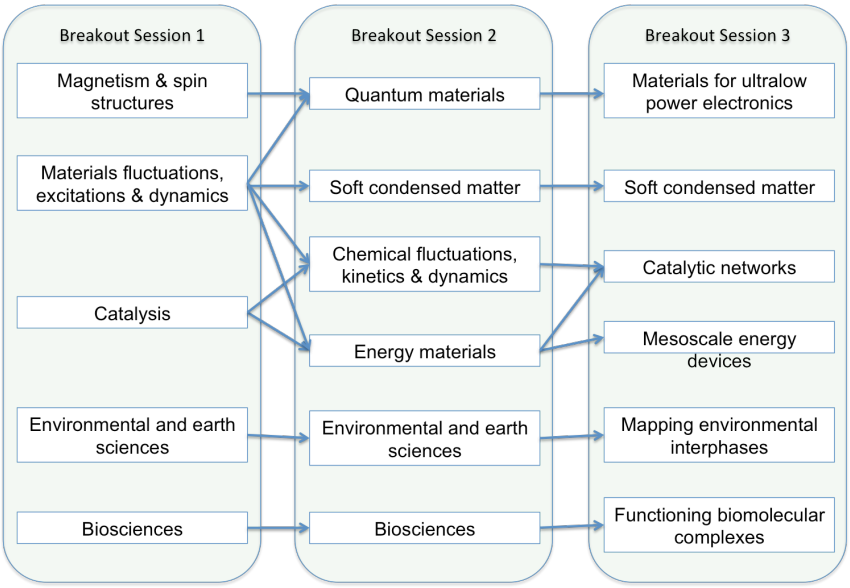
Workshop on Soft X-ray Science using Diffraction Limited Storage Rings
Advanced Light Source, Lawrence Berkeley National Laboratory
October 1- 3, 2014

AGENDA

Wednesday, October 1
Plenary Session (8:00 AM – 1:00 PM) in Building 15 Room 253
Plenary overflow in Building 6 Room 2202 (video-streaming via FUZE)

7:45	Continental breakfast	
8:15	Welcome and Introduction	
8:25	Materials with ultrabright soft x-ray beams	Wolfgang Eberhardt
9:00	Bioscience with ultrabright soft x-ray beams	Wayne Hendrikson
9:35	Storage Rings: near and far future	Mikael Eriksson
10:10	Break	
10:30	Diffractive imaging	David Shapiro
10:45	Soft x-ray nano-spectroscopy	Andrea Goldoni
11:00	Magnetic microscopy	Bastian Pfau
11:15	Photon correlation spectroscopy	Stuart Wilkins
11:30	Chemical dynamics/fluctuations	Oliver Gessner
11:45	Fluctuation SAXS	Peter Zwart
12:00	nanoARPES	Eli Rotenberg
12:15	(nano-)RIXS	Thorsten Schmitt
12:30	nano-APXPS	Hendrik Bluhm
1:00	Lunch	
2:00	Breakout session 1:	

Paths Through the Workshop



$N_{breakout} >> N_{session}$

Encourage cross-talk between breakouts:
Each breakout invited only 5-6, but was filled by attendees from other breakouts

Move attendees outside their comfort zones:
Final breakouts on functioning (mesoscale) devices with societal impact

What did we really want from this exercise?

Transformational research ideas enabled by high soft x-ray brightness/coherence

What didn't we want?

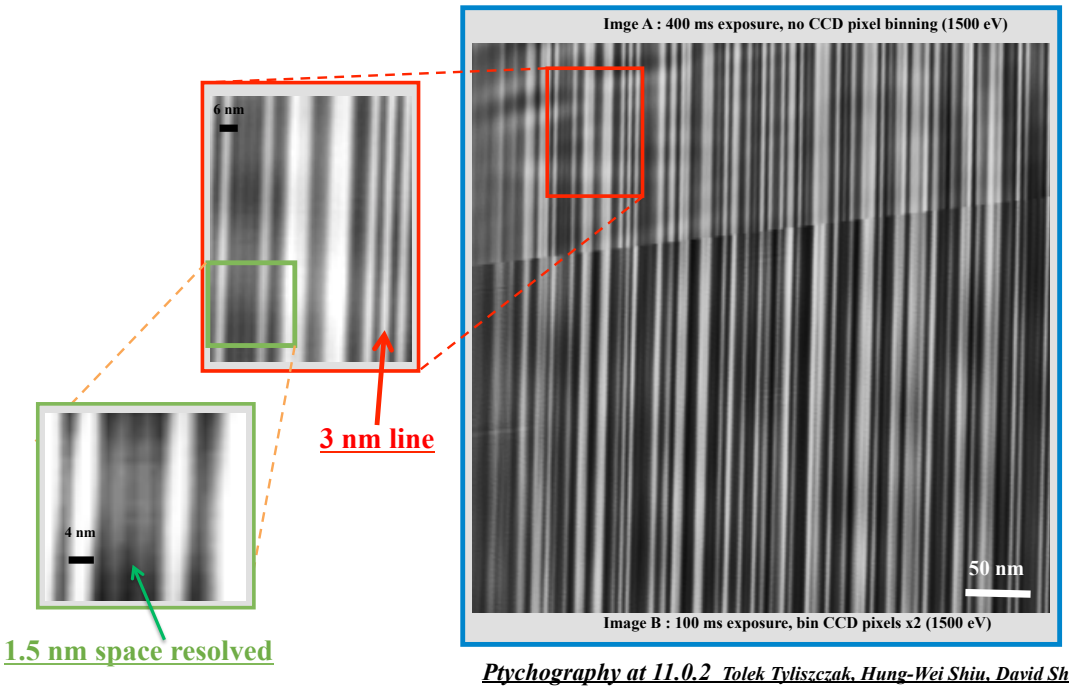
"Incremental" improvements in existing techniques (e.g., high throughput) are important but not very relevant for the science case

"Incremental" improvements in metrics, e.g., 1 nm, 0.1 meV, etc., are also important but not very relevant

What did we get (so far)?

Some "stretch goals" and good research examples

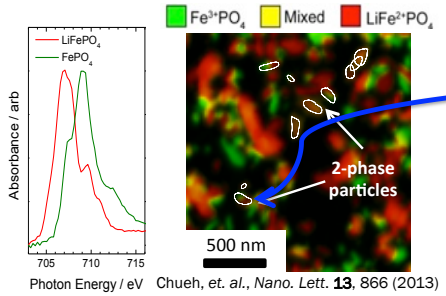
The Best Spatial Resolution of Soft X-ray Microscopy
1.5 nm resolved !! (4 weeks ago)



Ptychography at 11.0.2 *Tolek Tyliszczak, Hung-Wei Shiu, David Shapiro*

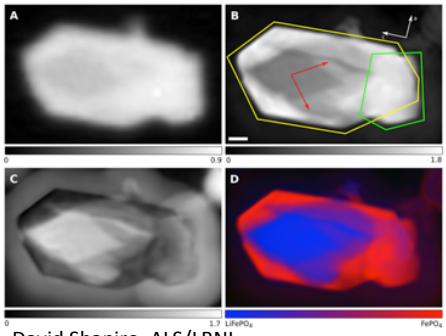
Sample from Valeriy Yashchuk

Coherent X-Rays and Ptychography Enable Probing Deep into the Nanoscale



Chueh, *et. al.*, *Nano. Lett.* **13**, 866 (2013)

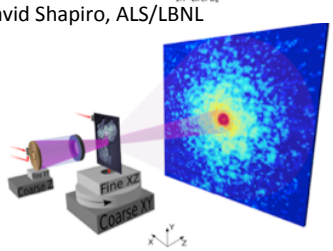
Chemical maps with STXM



David Shapiro, ALS/LBNL

	30 nm	10 nm	3 nm
ALS	180 s	4 hours	23 days
ALS-U	1.8 s	144 s	5 hours

X-ray exposure time required to image this particle in 3D with full spectral contrast



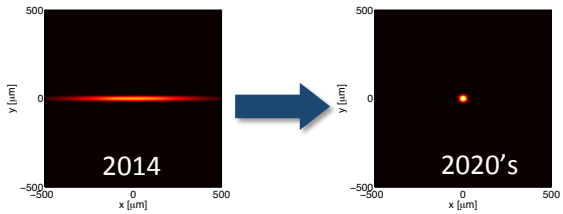
Ptychography

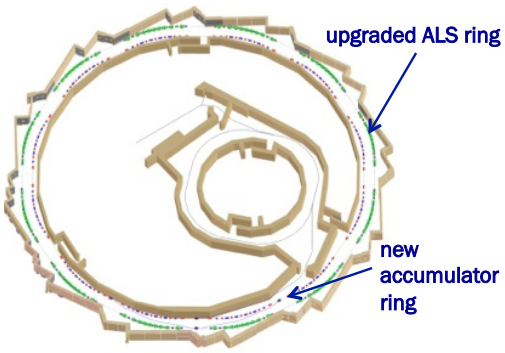
A revolution in lensless x-ray imaging Is being enabled by coherence and high-performance computing

Goals for Proposed ALS Upgrade

(these look a lot like the goals for other upgrades)

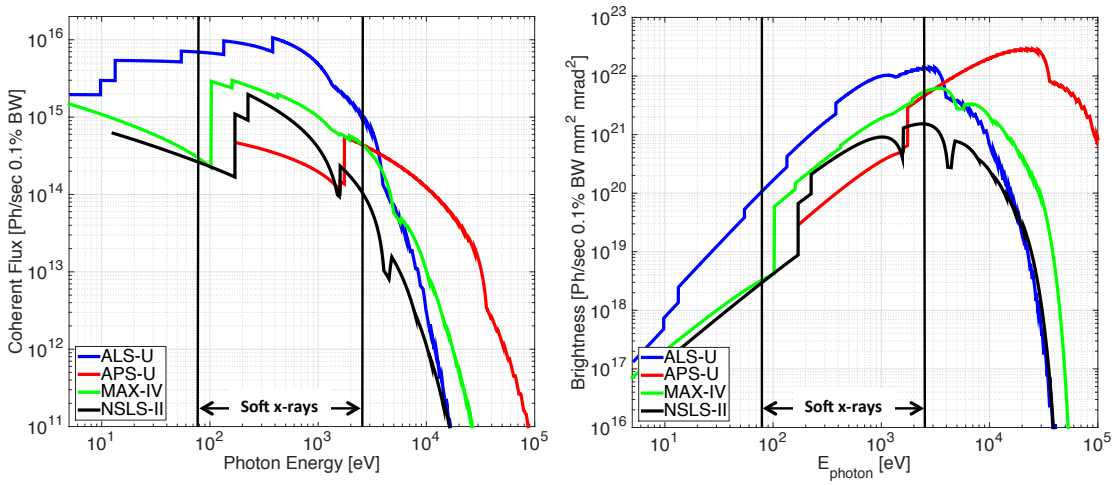
- **Enable DOE leadership in basic science and its mission in meeting societal needs in energy and the environment**
- **Develop the highest brightness and most capable soft x-ray synchrotron facility**
  - SXR brightness beyond any synchrotron, current, planned, under construction
  - up to 1000x increase in brightness over current ALS





- **Use advanced imaging techniques to address essential science and technology**
  - chemical, electronic, and magnetic maps of functional systems
  - nanometer resolution in 3-dimensions
  - dynamics and kinetics on natural timescales from picoseconds to minutes

An Upgrade of ALS Could Have Highest Coherent SXR Flux from a Synchrotron



Best for imaging and understanding dynamic systems

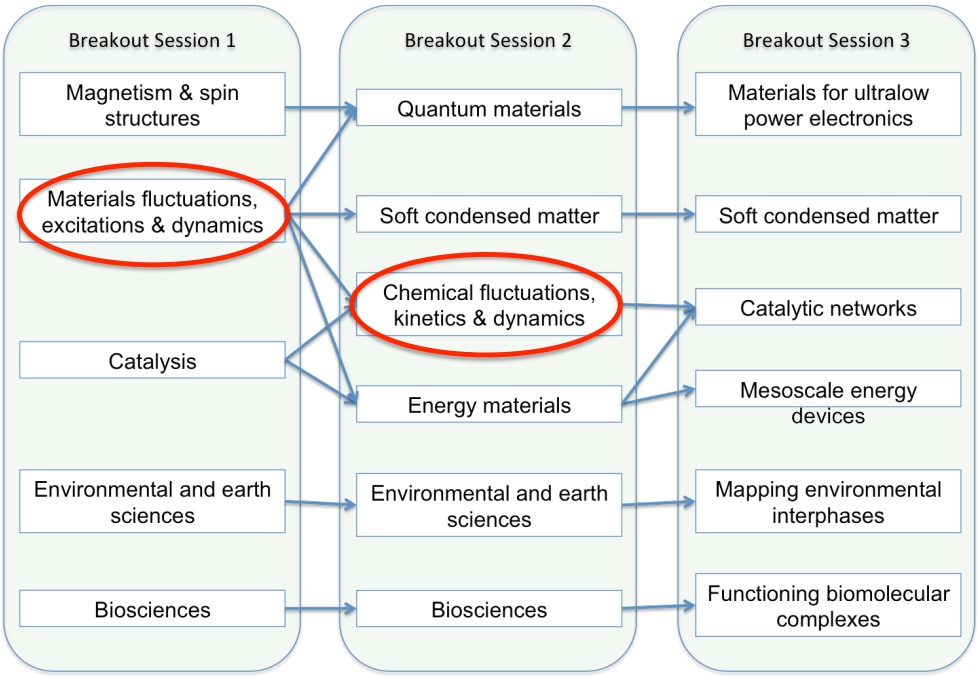
ALS-U Stretch Goals

Brightness ~ (photons/sec) x (1/[area x solid angle]) x (1/BW)

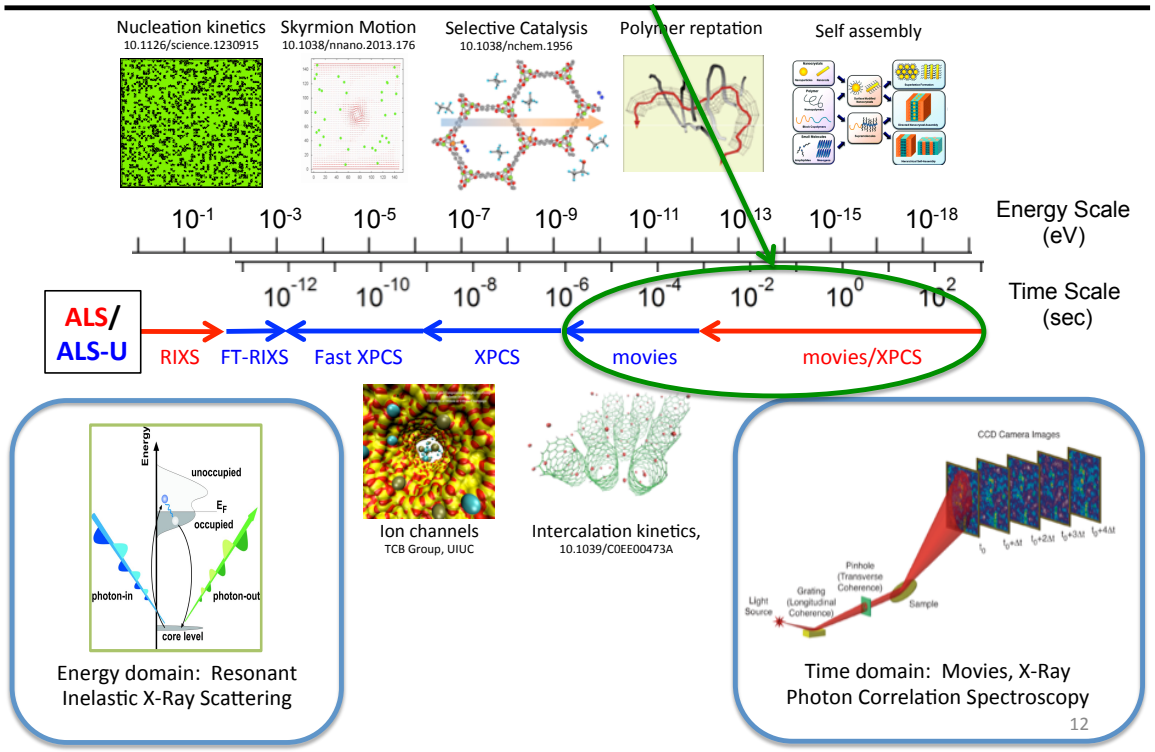
Time resolution Spatial resolution Spectral resolution

- Nanoscale chemical/material fluctuations and correlations at h/k_BT time scale
- Nanoscale chemical/material fluctuations and correlations at k_BT energy scale
- Interferometry for high sensitivity, spectral resolution

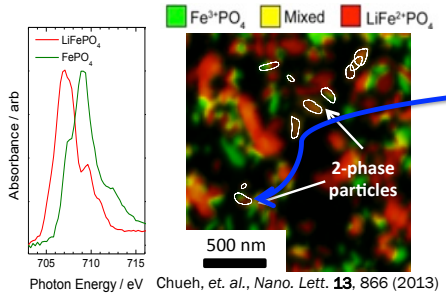
Workshop and ALS-U Stretch Goals



Capturing Spontaneous Nanoscale Kinetics: Time Domain



Coherent X-Rays and Ptychography:
Nanoscale Cinema



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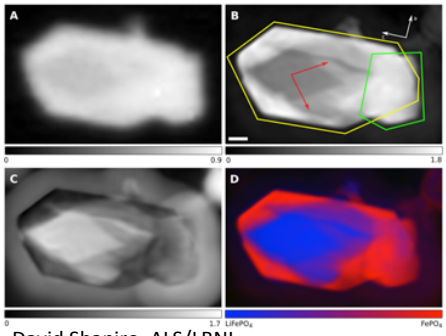

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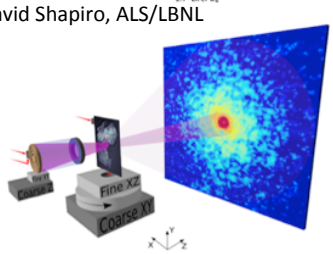
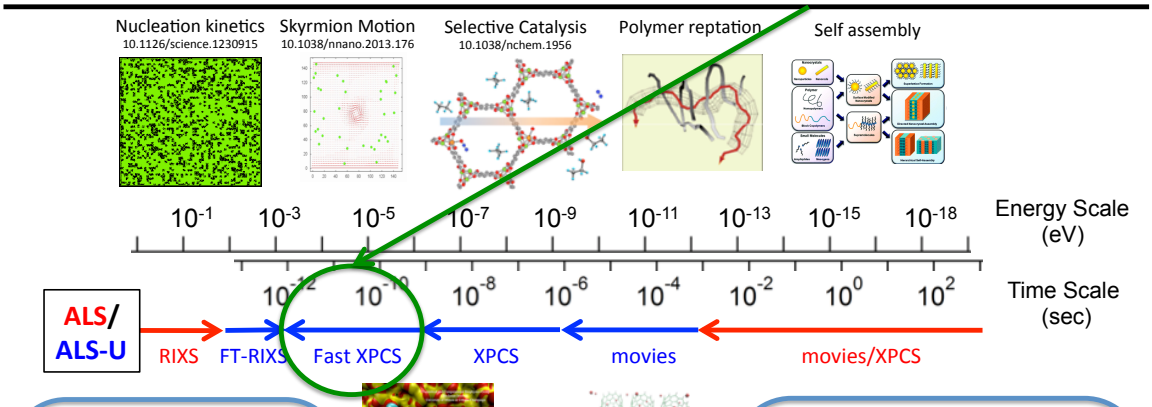
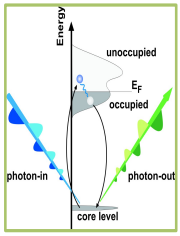


Ptychography

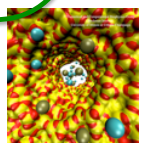
A revolution in lensless x-ray imaging Is being enabled by coherence and high-performance computing

Brightness Allows Capture of Spontaneous Nanoscale Kinetics: Approaching the k_BT Energy Scale

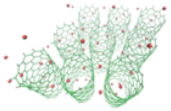




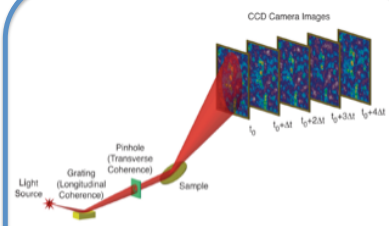
Energy domain: Resonant Inelastic X-Ray Scattering



Ion channels
TCB Group, UIUC

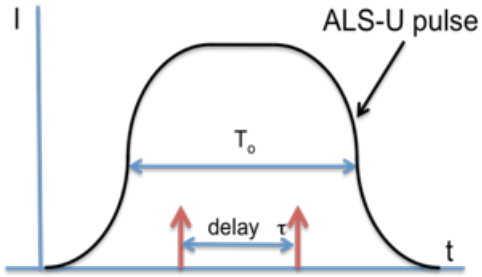


Intercalation kinetics,
10.1039/C0EE00473A



Time domain: X-Ray Photon Correlation Spectroscopy

Leveraging High Brightness To Probe the h/k_BT Time Scale



Time resolution near h/k_BT: fast XPCS

- Measure delayed coincidences inside a single ALS pulse
- High frame rate streak camera to achieve ~ 1 ps resolution
- Extreme event rate will push the limit of data handling

Continuous source (τ >> pulse period):

$$S/N\Big|_{cw} \sim \langle I_{sp} \rangle \sqrt{T \delta t N_{sp}}$$

$S/N \sim \langle I_{sp} \rangle^2$

Pulsed source w/streak camera:

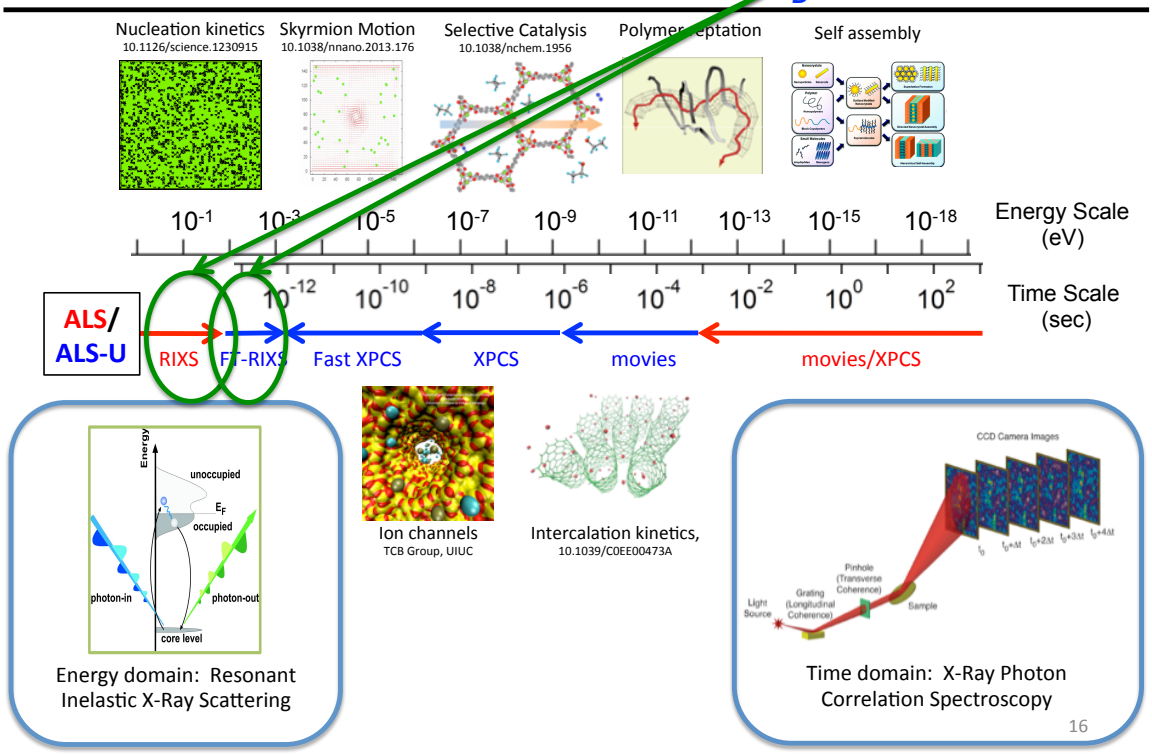
$$S/N\Big|_{pulse} \sim \langle I_{sp} \rangle \sqrt{T_o \delta t N_{sp} N_p}$$

$S/N \sim \langle I_{sp} \rangle^2$/duty cycle

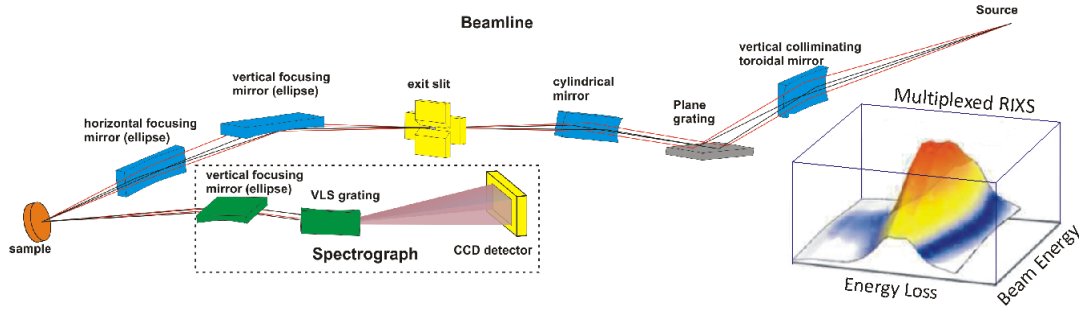
What ring time structure is optimal?

Require highest possible coherent flux

Brightness Allows Capture of Spontaneous Nanoscale Kinetics: Approaching the k_BT Energy Scale



Double-Dispersion RIXS



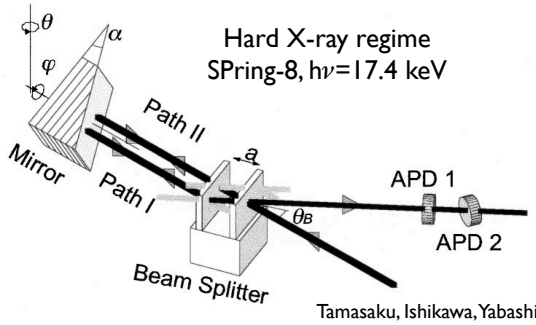
Multiplex both incident and scattered energy

Increase signal/throughput by ~x100 at ALS

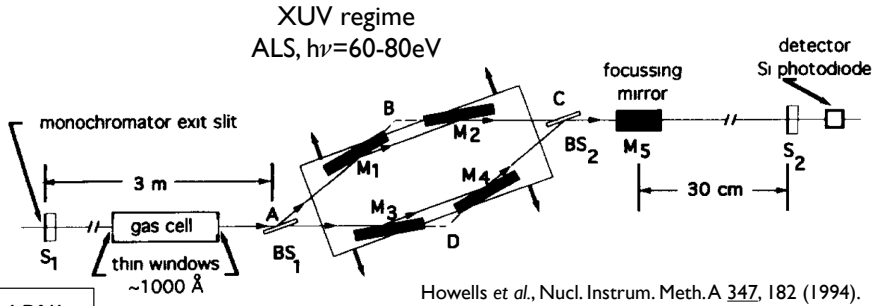
Gain another factor of ~10 at ALS-U

Best chance to achieve ~1 meV resolution with grating spectrographs

X-ray FT Spectrometers



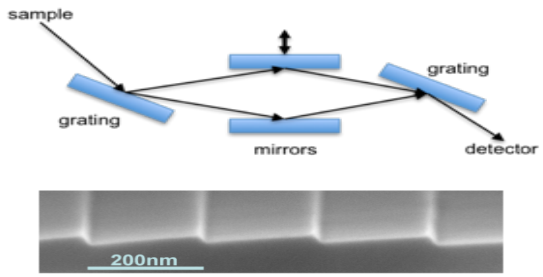
Tamasaku, Ishikawa, Yabashi, Appl. Phys. Lett. (2003).



Howells *et al.*, Nucl. Instrum. Meth. A 347, 182 (1994).

Gessner, LBNL

Stretching Energy Resolution to < 1 meV: FT - RIXS

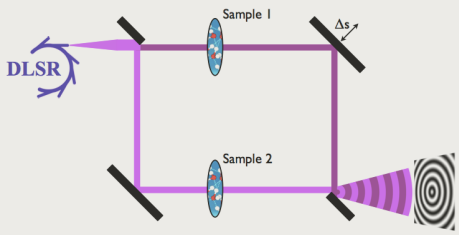


- Interferometer provides high resolution and potentially high angular acceptance
- Not clear whether double-dispersion approach will work
- Optics based on emerging grating beam splitters developed at ALS
- Requires high stability; potential for < 1 meV resolution

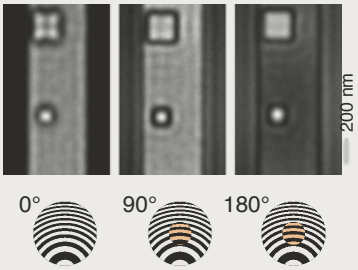
Phase Sensitive Detection at ALS-U

Coherent phase measurement
to improve sensitivity and reduce perturbation of sample

Interferometry for ultrahigh SXR sensitivity



Phase imaging with Zernike zoneplates



- Expand newly established optical phase techniques to enhance x-ray imaging techniques
- Go beyond speckle and XPCS in use of coherence

Controlling Chemistry with Nanostructure: Compartmentalized Catalytic Networks

Opportunity

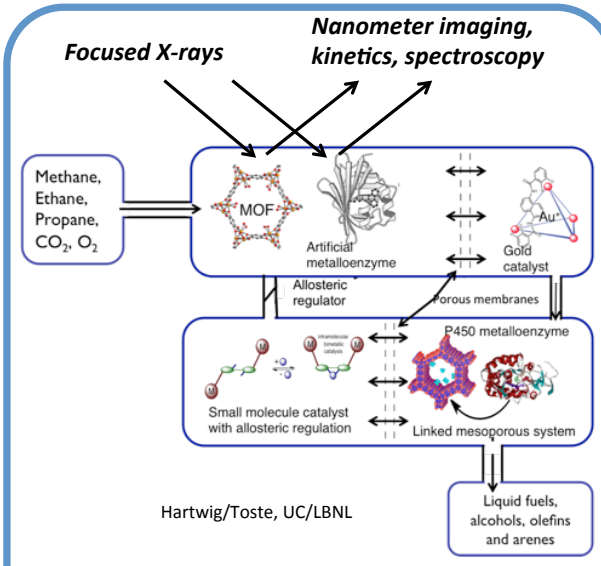
- Catalysis using compartmentalized nano-reactors based on feedback, regulation, and self-repair
- Large increase in catalytic efficiency and with selectivity rivaling that of living cells

Challenges

- *In situ/in operando* analysis tools with chemical specificity to understand and optimize network function
- Probe nanoscale chemical kinetics near individual structures

ALS-U Strengths

- Chemical maps with nanometer resolution
- Nanometer/nanosecond reaction-diffusion kinetics



Hartwig/Toste, UC/LBNL

Conceptual Catalytic Network
Low-temperature, selective coupling of methane and ethane to higher alkanes and alkenes.

Interphase Transport Critical to Multi-Scale Energy Storage and Conversion Structures

Opportunity

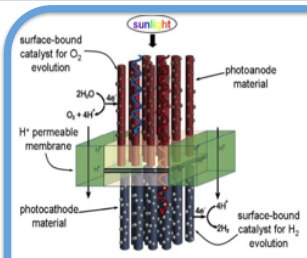
- Reveal electronic, chemical and atomic interfaces in energy storage materials that control function and evolve during operation
- Understand and optimize durability, capacity, and charge/discharge rate

Challenge

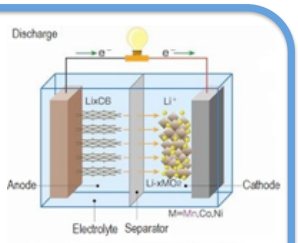
- *In operando* 3D nm-resolution chemical imaging and structure determination
- Image reaction and diffusion at electrochemical interphases

ALS-U Strengths

- *In operando* 3D tomographic chemical mapping of interfaces/interphases
- Nanofocusing for *in situ/in operando* experiments

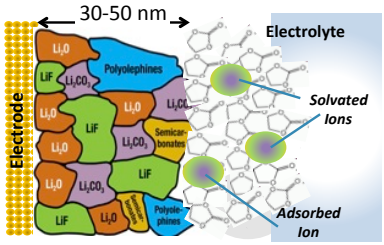


Artificial photosynthesis (JCAP)



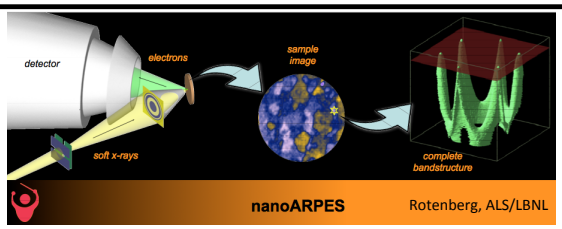
Batteries and fuel cells (JCESR, BATT)

Structure and Function of the Solid-Electrolyte Interface



What controls the formation, composition, and structure of the SEI? (from JCESR)

Control Electronic Heterogeneity to Achieve Novel Functionality of Correlated Systems



Opportunity

- Finally understand cuprate superconductors by unraveling the 3D electronic texture
- Discover new superconductors by controlling 3D electronic textures
- Control heterogeneity for new functionality, advanced processing in many material classes

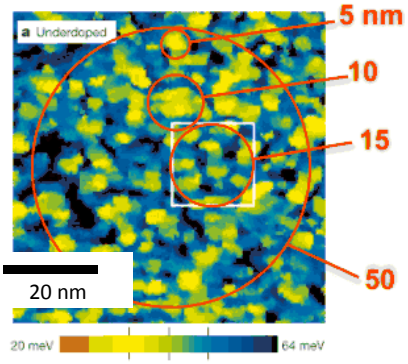
Challenge

- Soft x-ray probes with 5 - 50 nm resolution.

ALS-U Strength

- High coherent flux enables SXR spectromicroscopy with sub-5 nm spatial resolution
- Photon polarization and vorticity offer sensitivity to probe exotic phases

Local Variation of Superconducting Gap in Copper-Oxide Superconductor Imaged by STM



Lang, K., Madhavan, V., Hoffman, J., Hudson, E., Eisaki, H., Uchida, S., & Davis, J. (2002). Imaging the granular structure of high-Tc superconductivity in underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$. Nature, 415(6870), 412–416.

SXR spectromicroscopy with resolution deep in the nm scale is essential to unravel electronic heterogeneity common in complex materials.

Functionalized Nanoporous Materials: Selective Filtering and Controlled Chemical Transport

Opportunity

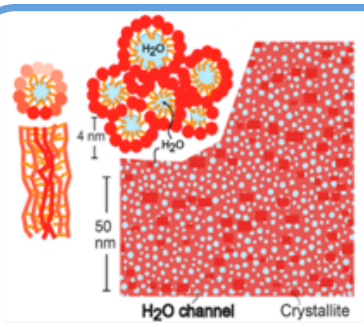
- Synthetic nanoporous membranes that rival the selectivity of biological membranes
- Recover dissolved critical materials from sea water, brine, waste water, . . .
- Optimize energy storage and conversion device

Challenges

- Visualize evolving chemical states in pores with high spatial and temporal resolution
- Measure coupled reaction/adsorption/ desorption/diffusion kinetics at the pore scale

ALS-U Strengths

- Chemical contrast and coherent power to probe *in operando* chemical structure and kinetics with nanometer, nanosecond sensitivity



Schmitt-Rohr, Nature Mat. **7**, 75 (2008)

Nanoporous Membranes

- Functionalize pores with spatially controlled chemical structures
- Control selectivity for highly selective chemical separations of useful materials

Addressing the "Water-Energy Nexus"

- Water purification
- Carbon capture & sequestration
- Biofiltration
- . . .

Summary: Soft X-ray Science Case

Focus on the distinguishing feature of high brightness – transverse coherence: ptychography, new kinds of phase imaging, XPCS, interferometry

Highlight niche science areas that are not well-addressed by other (DOE) sources

Emphasize new capabilities, but frame the science case with transformational science opportunities

Seek science at the boundaries between disciplines that addresses issues with societal impact (i.e., think outside the box)