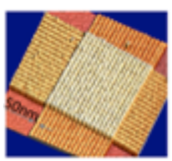
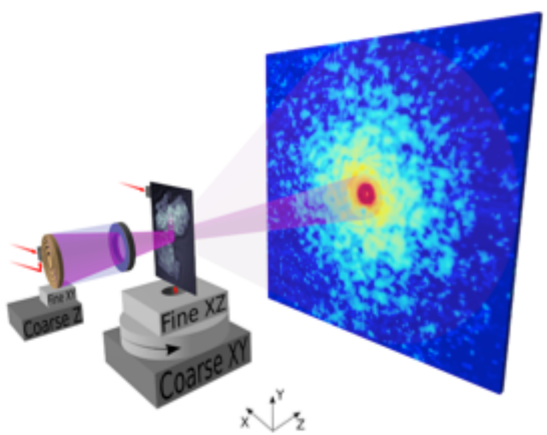
Soft X-Ray Workshop Report:

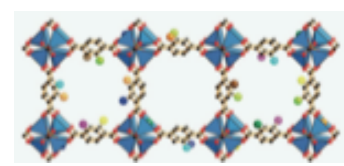
Scientific needs enabled by coherent soft x-rays

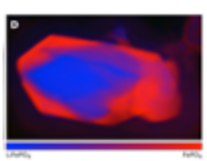
BESAC Feb. 26, 2015

Roger Falcone
Advanced Light Source
Lawrence Berkeley National Lab









Framework for x-ray sources and science

1) Science addressed by x-rays requires techniques that use both soft and hard x-rays, roughly differentiated as:

Soft x-rays

"Where are the electrons?"
"What is the chemical bonding, electronic structure, magnetism, correlation…?"

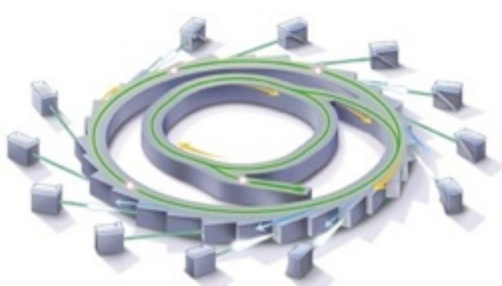
~ 1 keV and 1 nanometer

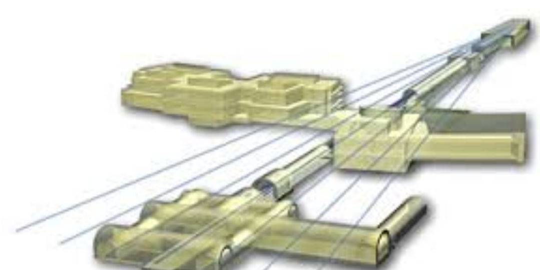
Hard x-rays

"Where are the atoms?"
"What is the structure, lattice strain, functionality…?

~ 10 keV and 1 Angstrom

2) Complementarity exists between quasi-CW beams from storage ring sources and pulses from linear free-electron lasers





BES supports 4 synchrotron ring x-ray sources

BERKELEY LAB Lawrence Berkeley National Laboratory

BROOKHAVEN NATIONAL LABORATORY

SLAC NATIONAL ACCELERATOR LABORATORY

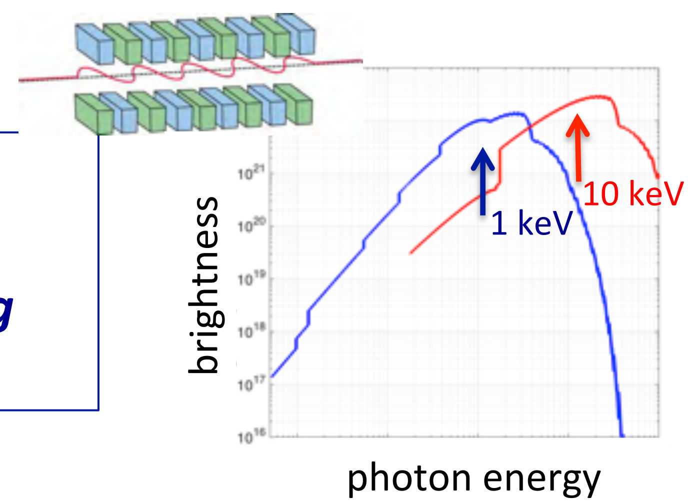
Argonne NATIONAL LABORATORY

Softest X-Rays
Best for mapping chemical and electronic structure

Intermediate X-Rays
More penetrating in materials for imaging and spectroscopy

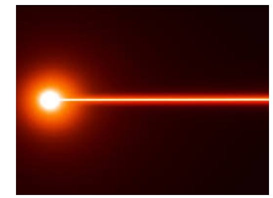
Hardest X-Rays
Best for structural determination

Each source has distinctive characteristics as well as overlapping capabilities allowing unique discovery science and serving a geographic location and user base

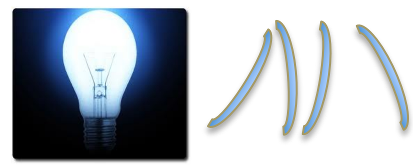


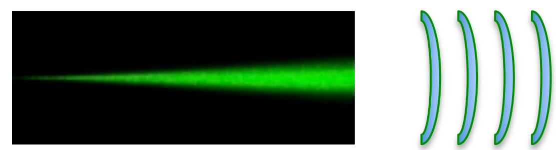
Brightness and coherence are key

Brightness:
- photons/sec/area/angle/bandwidth
- high light power in a small spot



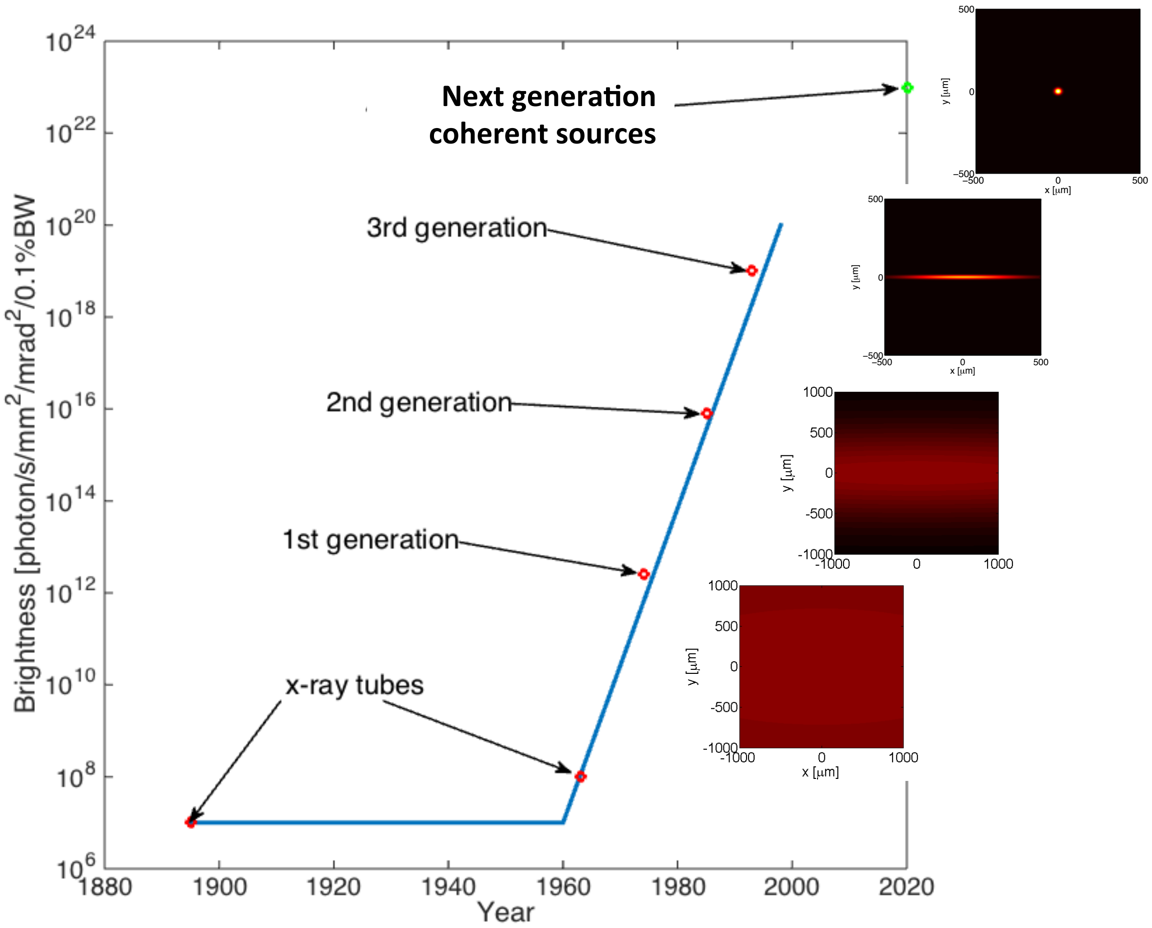
Coherence (transverse):
- uniform transverse wave front
- results in laser "speckle"





Diffraction limited:
- limit to uniformity of light waves
- focus to smallest spot

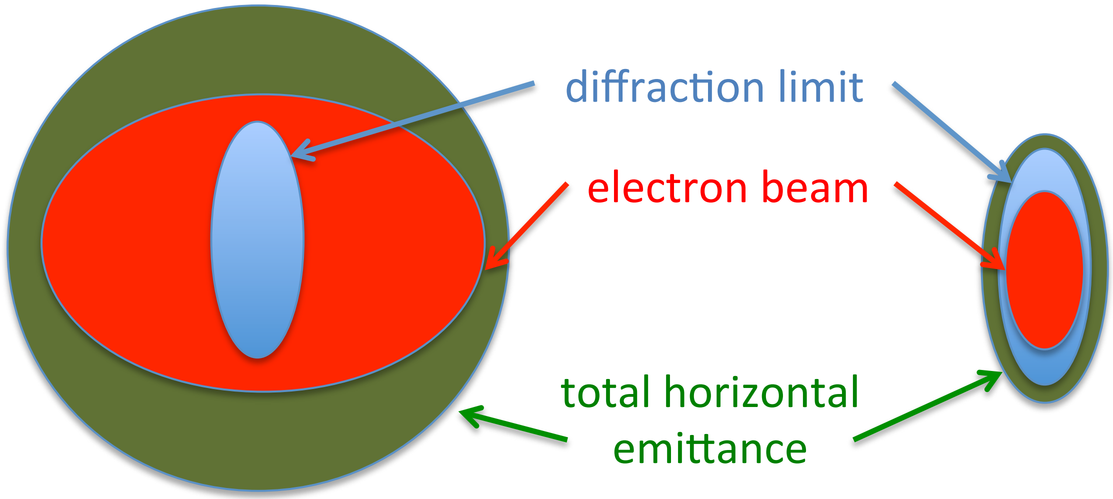
Brightness and coherence of x-ray synchrotron sources have increased by many orders of magnitude over decades



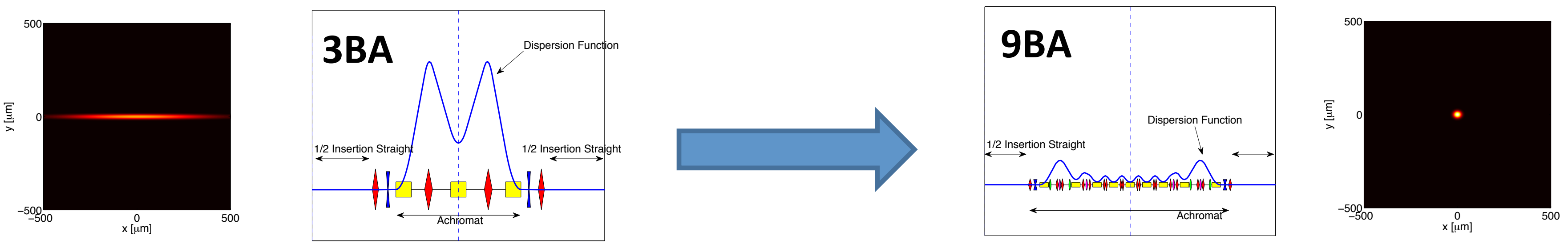
Brightness and coherence will determine the length of time necessary to do an experiment for a given spatial, temporal, and energy resolution

Sources with “diffraction-limited storage beams” are coherent

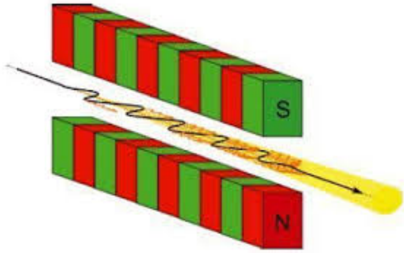
The fundamental limit of source size and divergence depends on wavelength



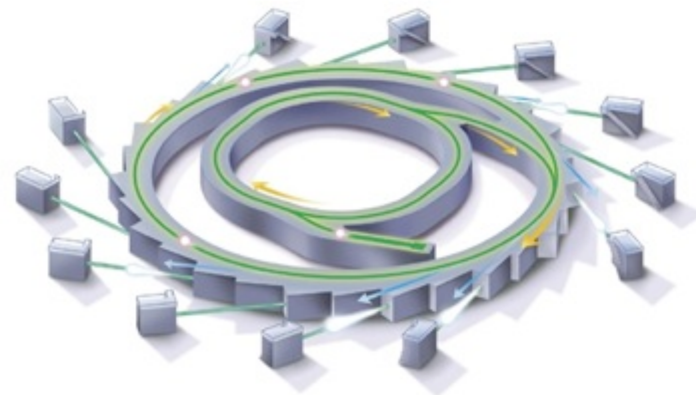
Multi-bend achromat magnets will allow us to get to the diffraction limit

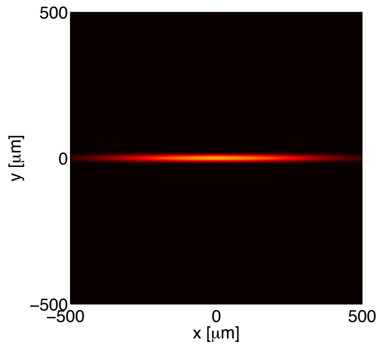


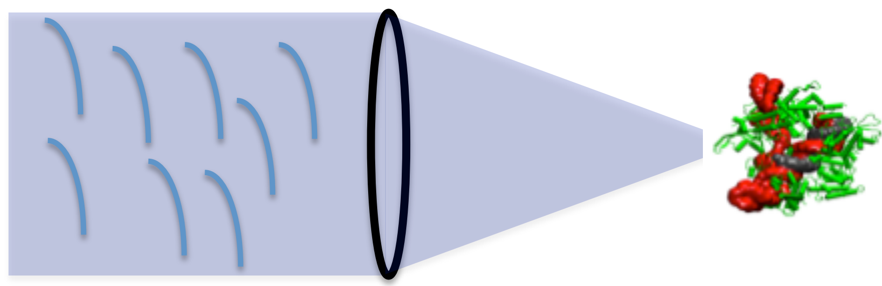
“DLSBs” have circulating electron beams small enough that the spot size and divergence of emitted x-rays is dominated by the fundamental diffraction of light, and not by the electron beam

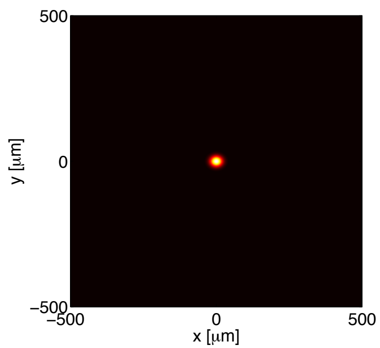


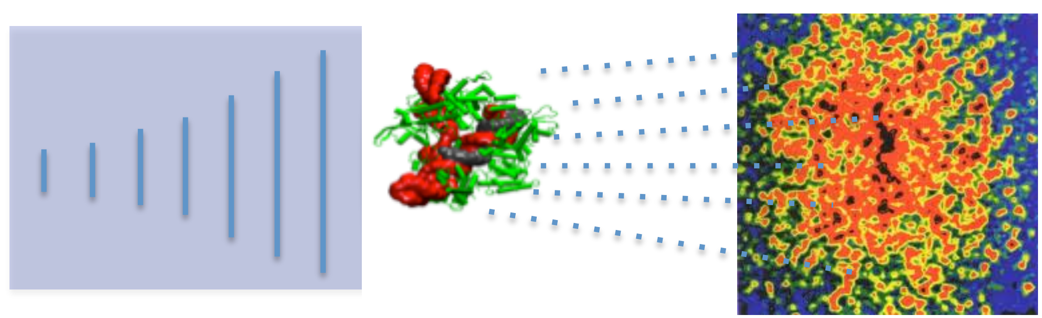
High brightness and coherence enable imaging with high spatial & temporal resolution





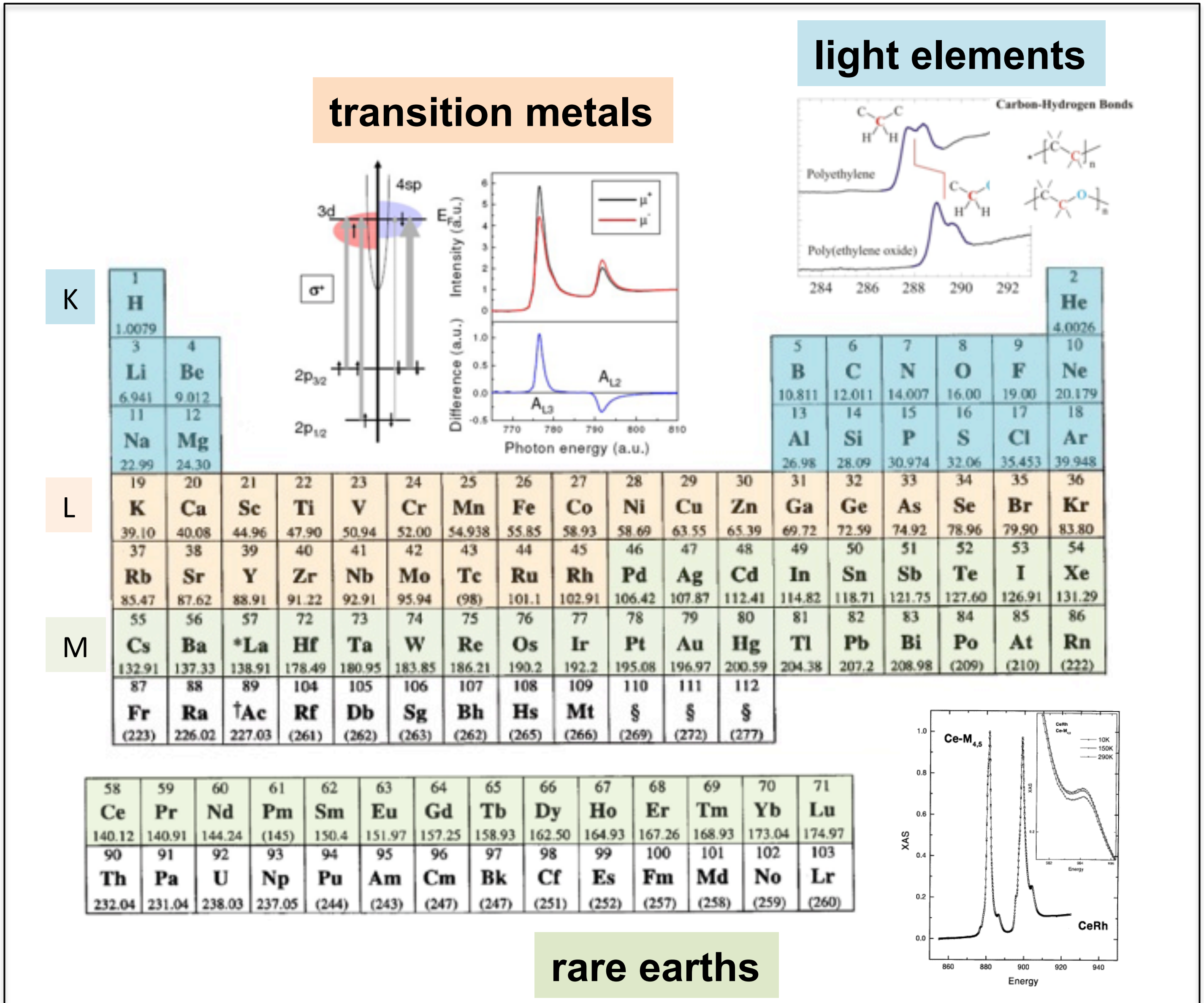






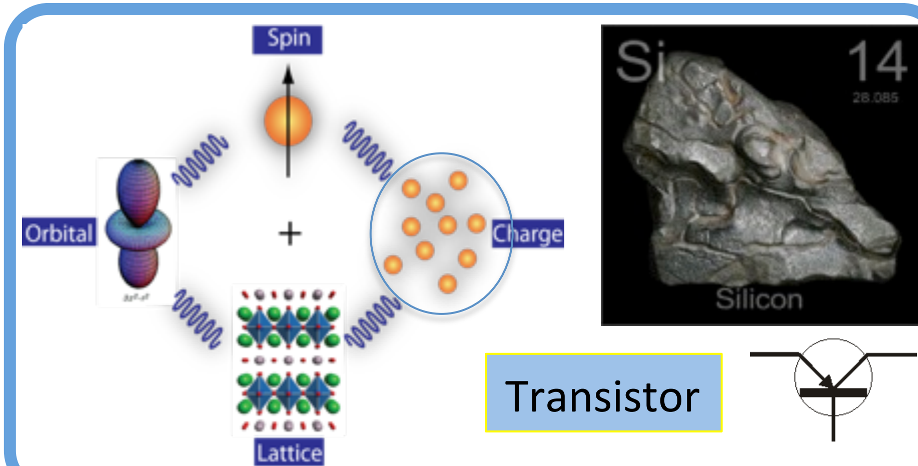
Soft x-rays access resonances in the elements

for imaging, scattering and spectroscopy with structural, chemical and electronic contrast

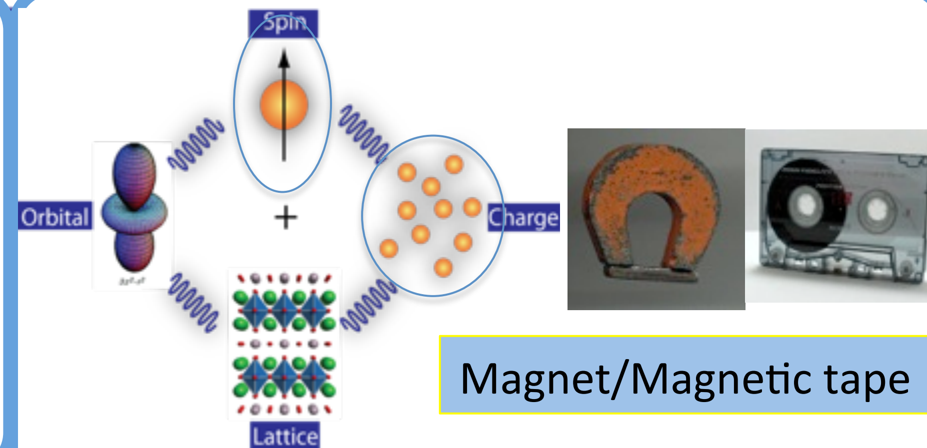


Unraveling Phenomena in Condensed Matter

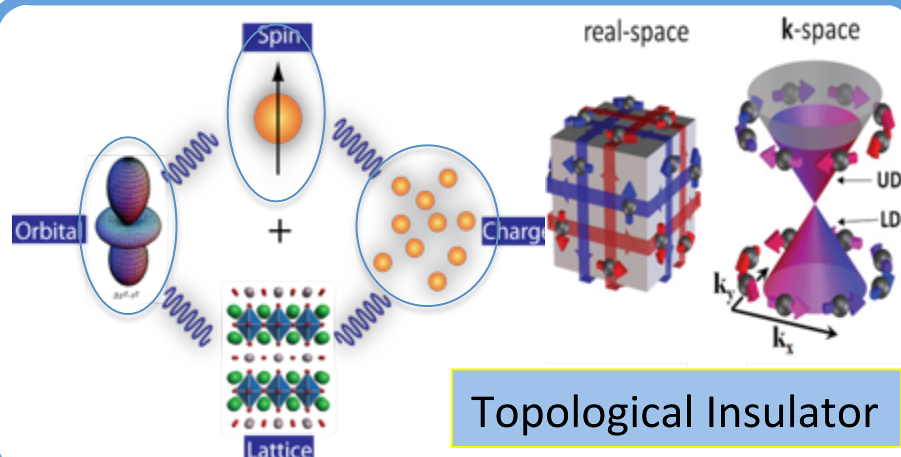
Many interacting degrees of freedom - novel phases emerge out of complexity



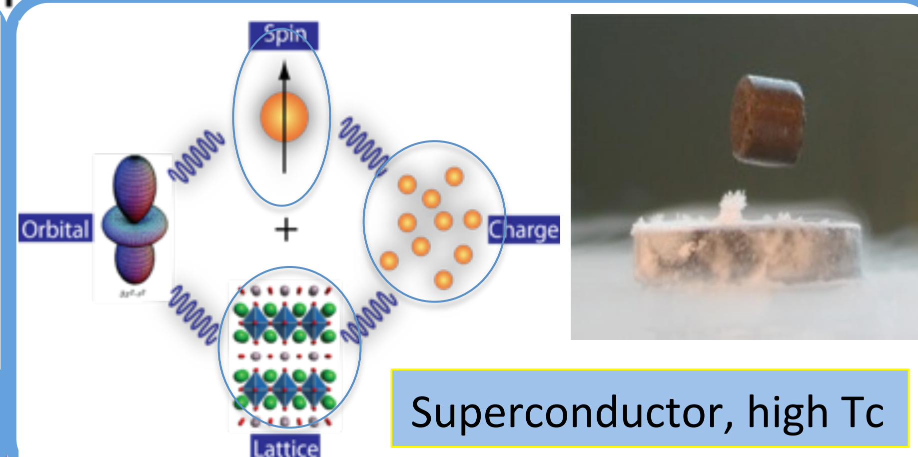
Transistor



Magnet/Magnetic tape



Topological Insulator



Superconductor, high Tc

Need to measure the electronic correlations at different ***length*** (momentum), ***energy*** and ***time*** scales → **Soft x-rays provide the techniques of choice**

- **Charge**:
  - What transformational research opportunities will be enabled by storage-ring-based, ultrahigh-brightness soft x-ray beams?
- **Attendance & Agenda**:
  - 80 scientists from around the world
  - 3 plenary talks on scientific opportunities & new source capabilities
  - 9 technique talks by international BL scientists (including early career)
  - 13 breakout sessions
- **Website**:
  - https://sites.google.com/a/lbl.gov/coherent-soft-x-ray-workshop/home
- **Conclusion:**
  - Coherent soft x-rays will enable frontier science in materials and energy research by providing spectroscopic contrast, nanoscale spatial resolution, and broad temporal sensitivity for spontaneous dynamics

Going beyond current measurement limits

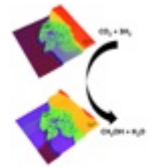
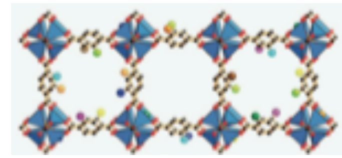
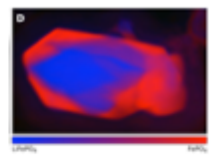
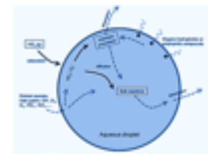
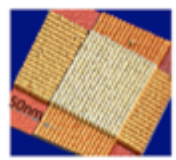
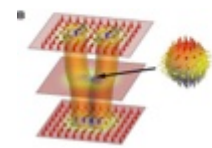
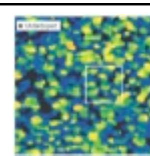
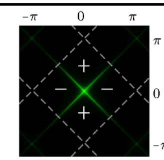
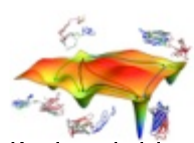
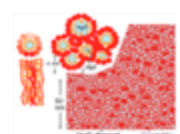
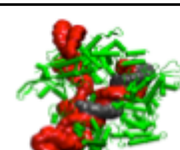
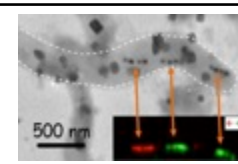
- **Probe deep into the nanoscale**
  - where surface properties of catalytic or other functional nanoparticles, rather than bulk properties, determine functionality
  - where diffusion and coherence length scales, and domain walls, defects, and grain boundaries determine functionality

- **Image inherently heterogeneous materials**
  - utilize coherent scattering for wavelength-limited resolution

- **Reveal functionality through natural or spontaneous dynamics**
  - thermally or diffusion driven kinetics, and spontaneous processes that determine chemical reactions and functionality

- **Benefit from the higher coherent flux and stronger scattering of <u>soft</u> x-rays**

Six scientific opportunities with societal impacts: the need for bright and coherent soft x-rays

Image spatiotemporal catalytic correlations	 Multicenter catalysts  MOF catalysis	***Enabling directed chemistry***
Measure transport across complex interphase regions	 Mapping charge  Aerosol chemistry	
Image low power charge and spin processing	 Memristor array  Skyrmion annihilation	***Materials to enable low power processing***
Image complex electronic textures and symmetries	 Electronic inhomogeneity  Pairing symmetry	
Probe soft modes to optimize functional systems	 Protein folding  Nanoporous membranes	***Addressing global biological and environmental challenges***
Image functioning biopolymers and biopolymer complexes	 Biopolymer complexes  Magnetosomes	

Imaging spatio-temporal catalytic correlations

Opportunities

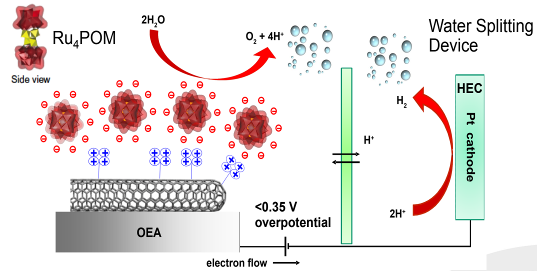
- Multi-center catalysts which achieve efficient and selective synthesis rivaling that of biological systems

Challenges

- *In situ* tools with chemical specificity and spatiotemporal resolution to understand kinetic correlations in heterogeneous catalysis

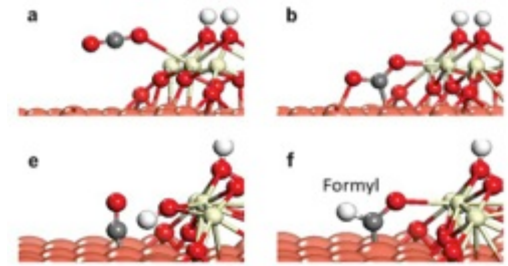
Strengths of coherent SXRs

- Chemical maps with nanometer imaging resolution and modest time resolution
- Nanometer/nanosecond catalytic kinetics with chemical contrast using XPCS



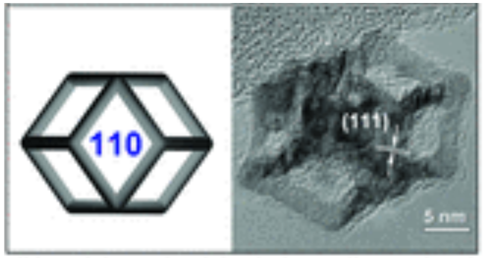
Electrocatalytic water splitting

(Andrea Goldoni, ELETTRA)



CeO_x/Cu CO_2 reduction catalyst

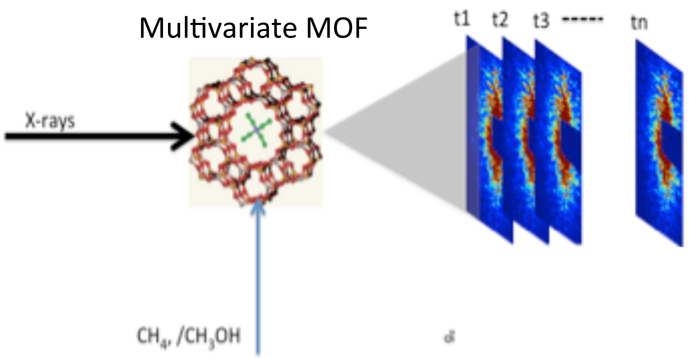
(Graciani, et. al., *Science* **345**, 546 (2014))



$PtNi_3$ nanoframe with very high ORR electrocatalytic activity

[Chen, et. al., Science **343**, 1339 (2014)]

Chemical XPCS



XPCS with chemical contrast will reveal kinetic correlations between catalytic centers

Measuring transport across complex interphase regions

Opportunities

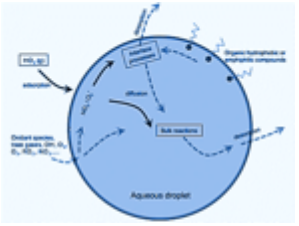
- Probe interfacial reaction/diffusion in aerosols to model atmospheric chemistry
- Optimize charge separation and transport in energy storage/conversion
- Design optimized light harvesting materials
- Understand transport/precipitation/ dissolution of CO_2 in brines

Challenges

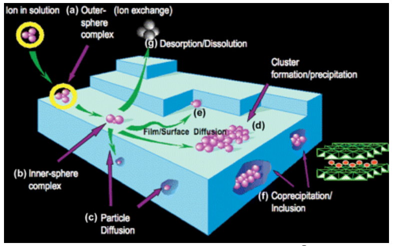
- *Operando* analysis tools with chemical contrast to probe nanoscale transport
- Probe nanoscale dynamics and kinetics near individual nanostructures

Strengths of coherent SXRs

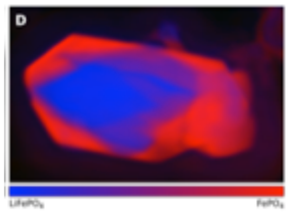
- Movies of nanoscale chemical structure and kinetics
- Potential picosecond pump-probe imaging with high spatial resolution and chemical contrast



Interfacial reaction/diffusion in aerosols

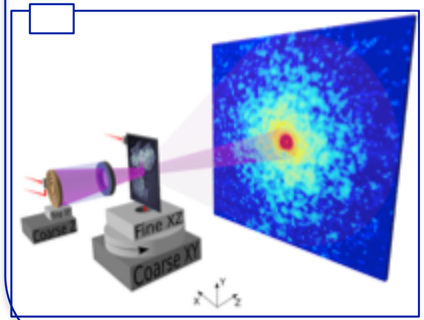


Precipitation/ dissolution in geosystems



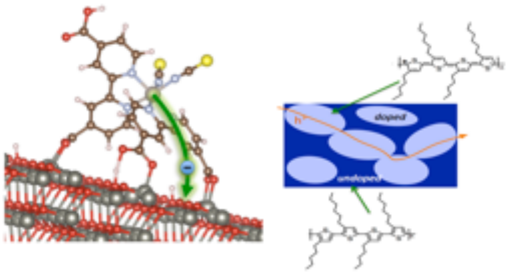
Imaging intercalation in battery electrodes

Potential for MHz Frame Rate Movies with CDI

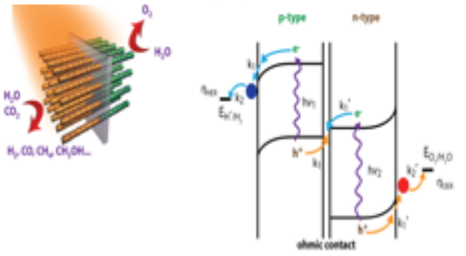


3D images with low nm resolution, full spectrum: ~1 hr

2D image, ~ 10 nm resolution, one energy: ~1 μs



Intra- and intermolecular electron transport



Electron transfer in artificial photosynthesis

Imaging low power charge and spin processing

Opportunities

- Skyrmions are nanoscale spiral spin textures proposed for low power memory, classical and quantum processing
- Memristors provide a platform proposed for for 3D high density digital memory and low power neural processing

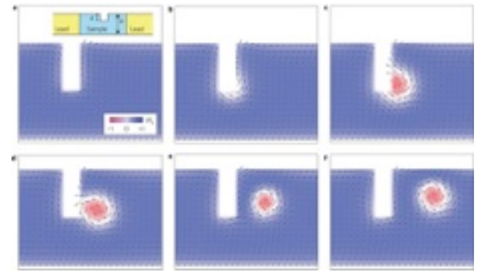
Challenges

- Image skyrmion drift, diffusion, interactions, and processing
- Probe evolving 3D structure of memristor junction with nanometer resolution and oxidation state contrast

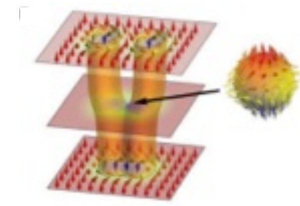
Strengths of coherent SXRs

- High magnetic/spin contrast of soft x-ray spectroscopy, scattering, imaging
- High sensitivity to oxidation state with low-nm resolution

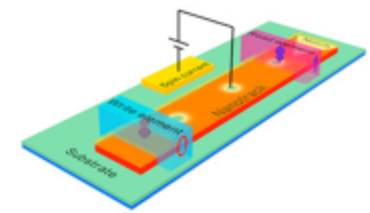
Skyrmion source
Iwasaki, et. al. Nat. Nano 8, 742(2013)



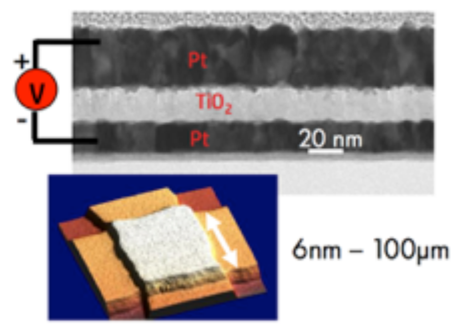
Skyrmion processing
Milde *et al. Science* **340**, 1076 (2013)



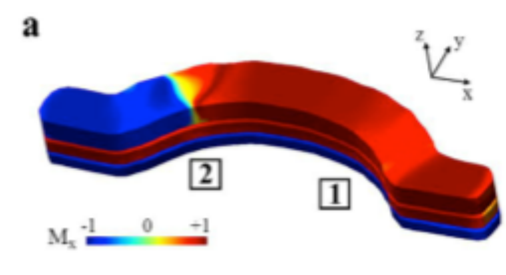
***Proposed* elements for skyrmion processing**



Skyrmion racetrack memory
Zhang, et. al., Sci. Rpts DOI: 10.1038/srep07643)

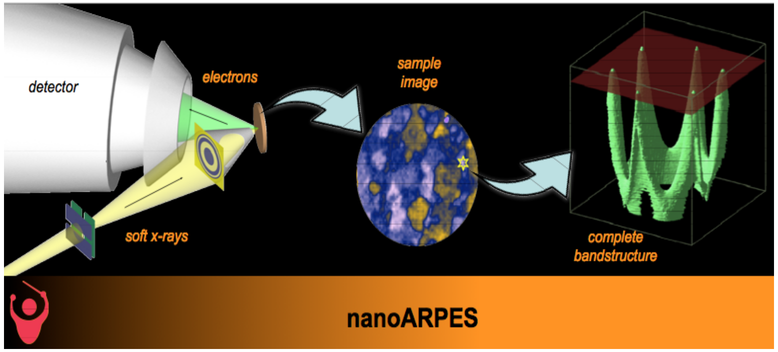


Memristor structure
Stan Williams, HP labs



Spintronic Memristor
Metaxas et al, Sci. Reports 3, 1829 (2013)

Imaging complex electronic textures and symmetries



Opportunities

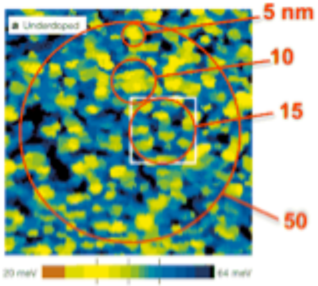
- Higher temperature superconductors by controlling 3D electronic textures
- Control heterogeneity for new functionality, advanced processing in diverse materials

Challenges

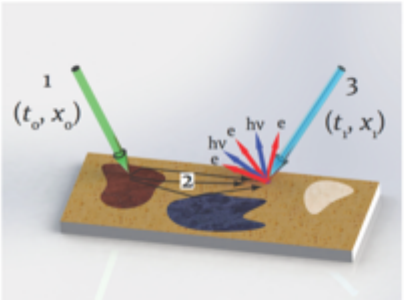
- Soft x-ray probes with 5 - 50 nm resolution
- Sub-10 meV resolution RIXS

Strengths of coherent SXRs

- High coherent flux, emerging diffractive optics enables SXR spectromicroscopy with low-nm spatial resolution
- Photon polarization and vorticity offer sensitivity to probe exotic phases



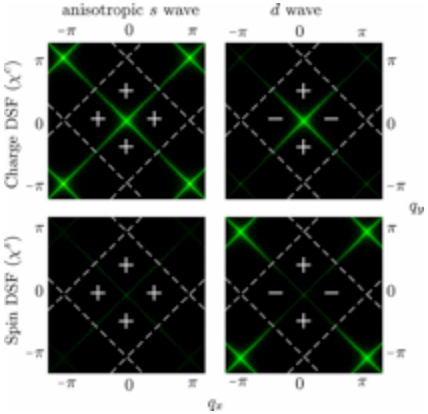
Lang, et. Al., Nature 415, 412 (2001).



Eli Rotenberg, ALS.

High resolution to probe electronic inhomogeneities

Spectroscopy + transport to measure G(r,t)



Marra, et. al., PRL 110, 117005 (2013)

Dynamical structure factor S(q,ω) for spin and charge excitations in unconventional superconductors: revealing the mechanism of HT superconductivity

Probing soft modes to optimize functional systems

Opportunities

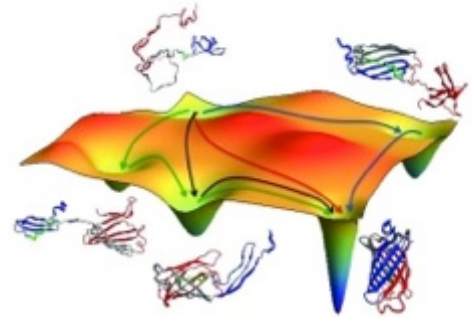
- Probe crossover between dynamic and kinetic regimes
- Understand how function emerges from degenerate, interacting modes at energy ~ $k_B T$

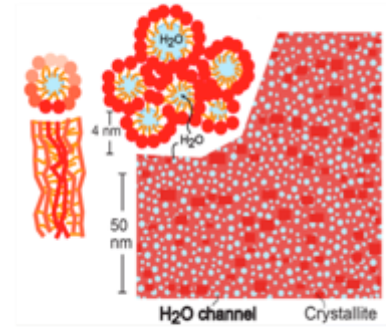
Challenges

- Probe soft losses at energy < $k_B T$ with quasi-elastic RIXS using FT detection
- Probe fluctuations at timescales down to $h/k_B T$ with quasi-elastic XPCS using streak camera and pulses from source

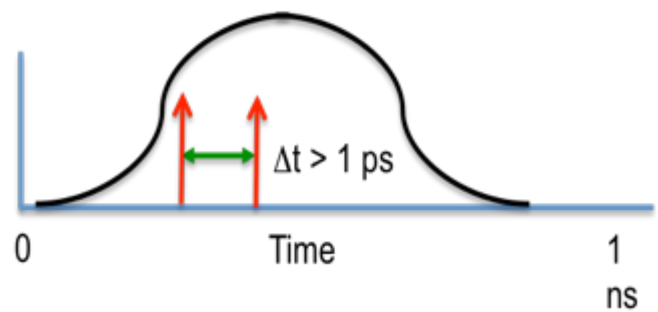
Strengths of coherent SXRs

- Highest possible coherent flux provides best XPCS/FT-RIXS signal
- Diverse contrast mechanisms to extend neutron scattering
- Intense, focusable beams to study nanostructured materials

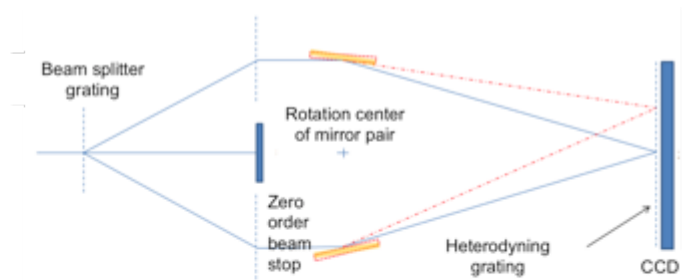




Bioinspiration: diversity of modes at 1 ps and longer conspire to drive protein folding, membrane function, and beyond



Fast XPCS: delayed coincidences inside single SR pulses



FT RIXS: atom-scale control systems enable new precision measurements

Imaging functioning biopolymers & biopolymer complexes

Opportunity

- Measure biopolymer structure in solution
- Connect biopolymer structure to function

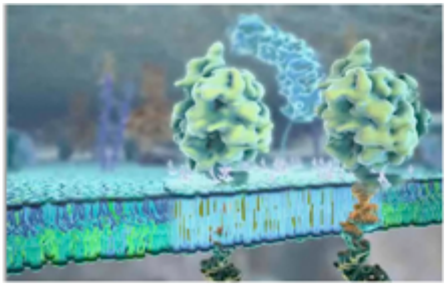
Challenge

- Many interacting low energy degrees of freedom
- Motion occurs over a wide range of time scale
- Functional biosystems are not periodic

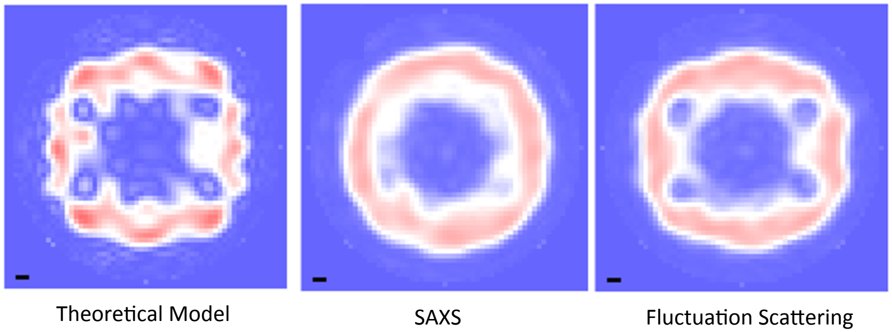
Strengths of coherent SXRs

- Multimodal probes of nanoscale order, assembly, and dynamics via microscopy, spectroscopy, and scattering
- Large SXR scattering cross sections and resonant scattering improves dynamic range, chemical selectivity
- Fluctuation x-ray scattering can interpolate in space and time between crystallography and protein SAXS

CAS9+RNA+DNA Structure: *gene slicer*
(Sternberg, et. al., Nature, 507, 62-67, 2014)



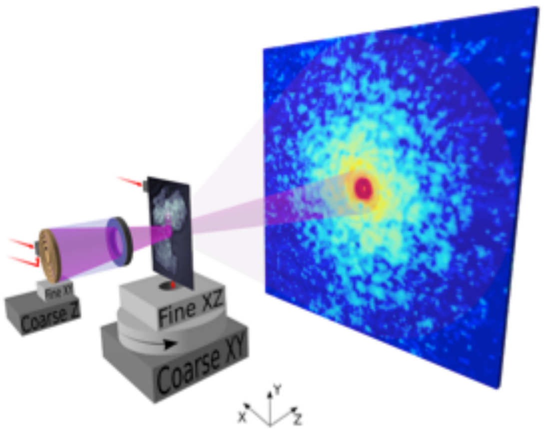
Coupled membrane/ protein motion



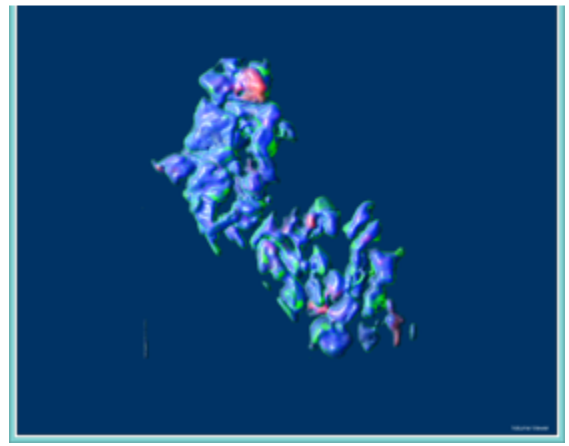
Fluctuation X-ray Scattering

- single snapshots from FEL or DLSR freeze rotational motion
- reconstruction provides non-angle-averaged structures close to macromolecule morphology

Imaging with a current, partially-coherent source: $LiFePO_4$ particles in 3D at 18 nm resolution

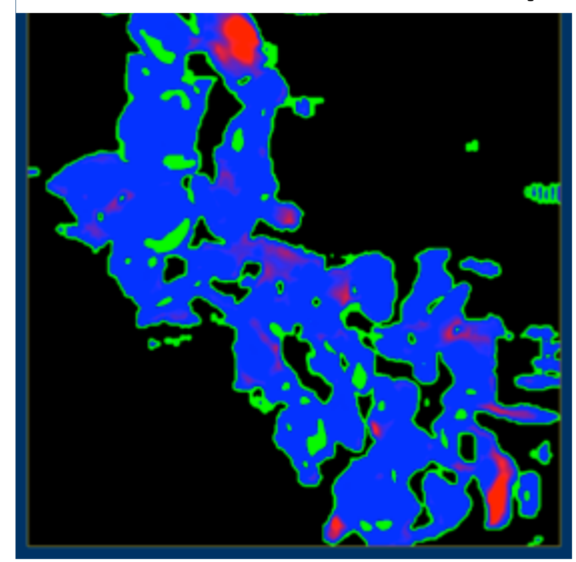


3d difference map



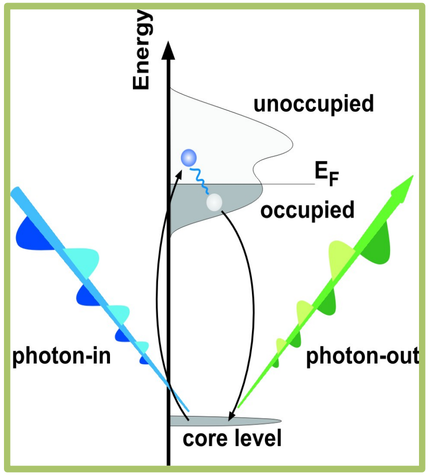
Blue - $FePO_4$
Red - $LiFePO_4$

2d slice of difference map

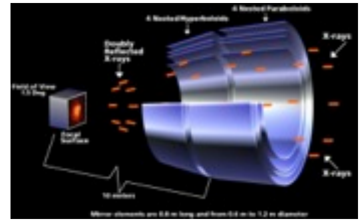


- *Nanosurveyor* instrument at ALS
- ~ 100 x 100 x 20 nm partially de-lithiated particles
- 24 hours collection time
- bend magnet source
- Shapiro, Yu, et al, Nano Letters

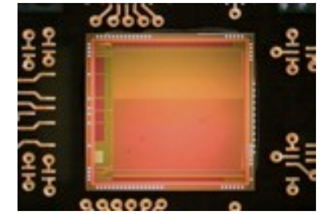
Increased signal and improved resolution for RIXS: double-dispersion technique that benefits from high brightness



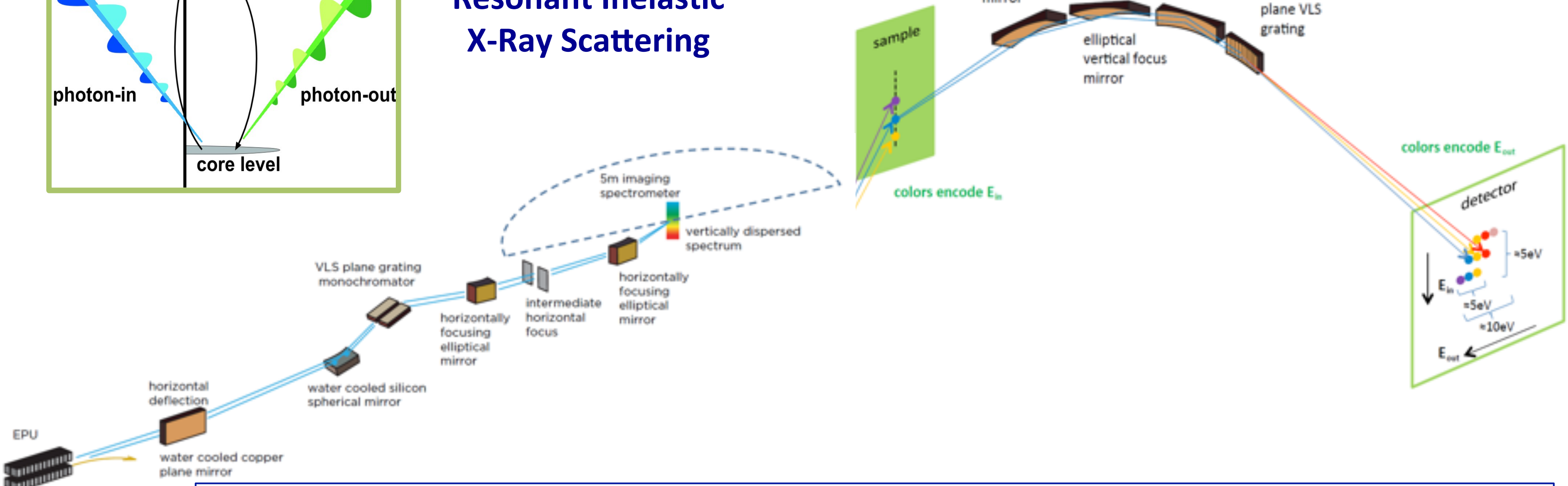
RIXS Process: S(q,ω)
Resonant Inelastic X-Ray Scattering



Chandra x-ray optics



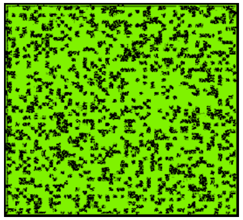
5 μm CMOS detector- Denes



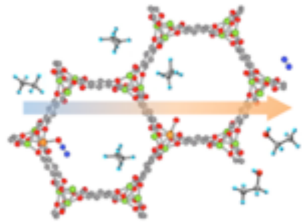
- **Dispersed at both sample and detector planes**
- **Produces full RIXS map and increases signal by ~300 over single wavelength**
- **Practical approach designed to reach 1 meV resolution with grating optics**
- **Round beams simplify design for improved signal and resolution**

Brightness allows capture of spontaneous nanoscale kinetics: approaching the h/k_BT timescale

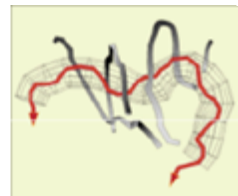
Nucleation kinetics
10.1126/science.1230915



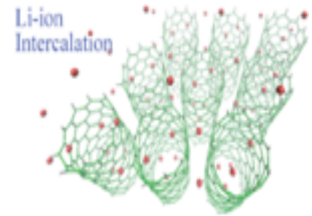
Selective Catalysis
10.1038/nchem.1956



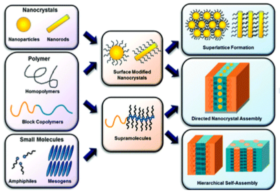
Polymer reptation

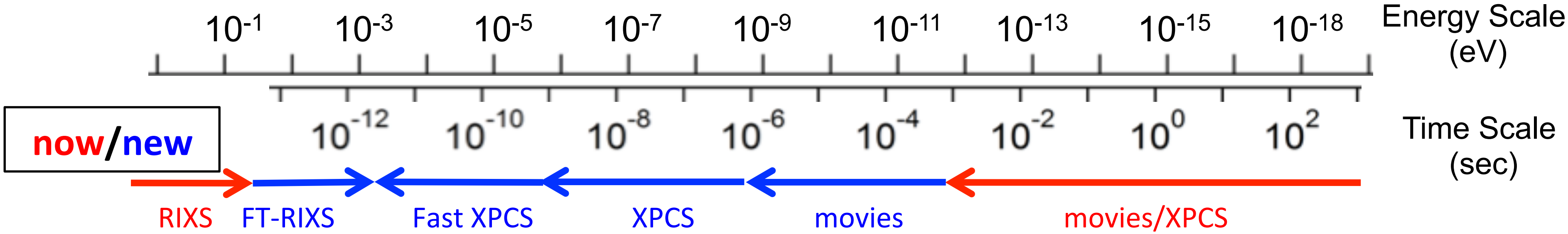


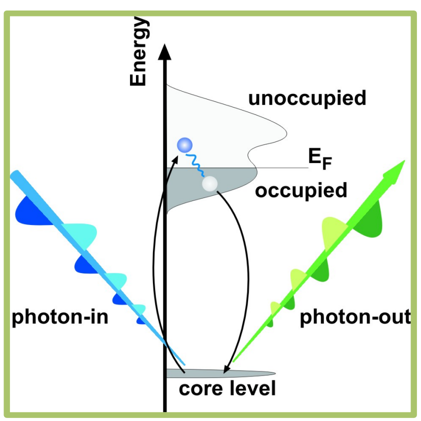
Intercalation kinetics,
10.1039/C0EE00473A



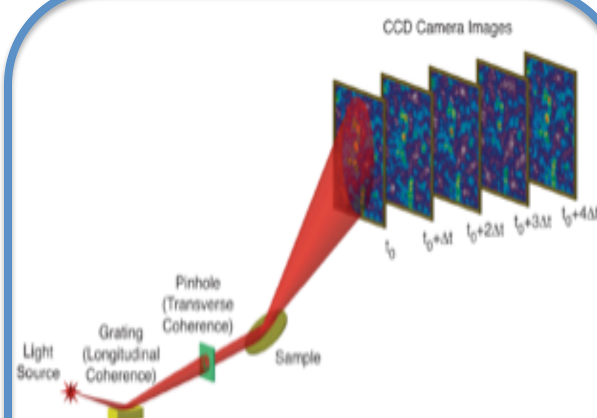
Self assembly



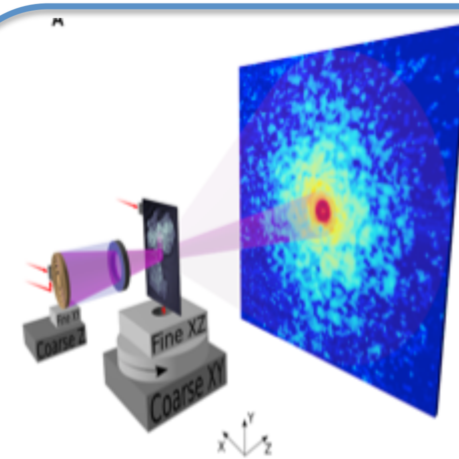




Resonant Inelastic X-Ray Scattering: $S(q,\omega)$



X-ray Photon Correlation Spectroscopy: $S(q,t)$



Time-resolved X-ray Microscopy: $G(r,t)$

International context: facilities are becoming brighter

Large circumference + damping wigglers

BNL: **NSLS-II** (2014): 3 GeV, 1000pm x 8 pm, 500 mA (**New**)

First new multi-bend achromat rings

Sweden: **MAX-IV** (2016): 3 GeV, 230 pm x 8 pm, 500 mA (**New**)

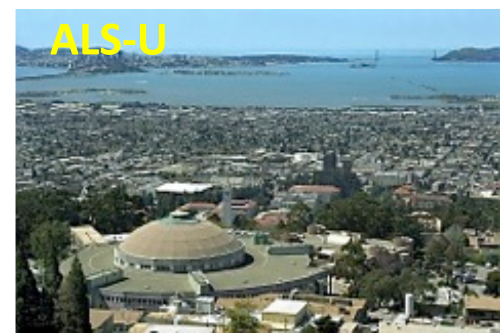
Brazil**: SIRIUS** (2016/17): 3 GeV, 280 pm x 8 pm, 500 mA (**New**)

1st multi-bend achromat ring upgrade

France: **ESRF-II** (2020): 6 GeV, 160 pm x 3 pm, 200 mA (**Upgrade project**)

U.S. upgrade landscape for the future

APS-U: 6 GeV, 60 pm x 8 pm, 200 mA (**Upgrade project**)



ALS-U: 2 GeV, 50 pm x 50 pm, 500 mA (**Conceptual design**)

Other international plans: *Japan (Spring 8, 6 GeV), China (BAPS, 5 GeV), Germany (PETRA-III), France (SOLEIL), Switzerland (SLS, 2.4 GeV), Italy (ELETTRA) are developing brightness upgrade plans*

Summary

- **Bright and and coherent soft x-rays are key to new basic energy science**
  - Observe nm-structures in 3-dimensions
    - Chemical, electronic, and magnetic maps of functional systems
    - Beyond lenses by using coherence
    - Dynamics and kinetics on natural timescales of picoseconds to minutes
  - New capabilities are being developed for transformational science

- **Soft x-rays address a broad range of fundamental science and technology**
  - with huge potential societal impacts in energy, health, materials, computing, and the environment

- *"…impressive international activity in the development of diffraction limited storage rings… will allow powerful new classes of experiments that take advantage of the full coherence and brightness."* **BESAC Subcommittee July 2013**