Soft X-Ray Workshop Report:
Scientific needs enabled by coherent soft x-rays

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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

**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:**
  o What transformational research opportunities will be enabled by storage-ring-based, ultrahigh-brightness soft x-ray beams?

• **Attendance & Agenda:**
  o 80 scientists from around the world
  o 3 plenary talks on scientific opportunities & new source capabilities
  o 9 technique talks by international BL scientists (including early career)
  o 13 breakout sessions

• **Website:**
  o [https://sites.google.com/a/lbl.gov/coherent-soft-x-ray-workshop/home](https://sites.google.com/a/lbl.gov/coherent-soft-x-ray-workshop/home)

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

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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

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

Interfacial reaction/diffusion in aerosols

Precipitation/dissolution in geosystems

Imaging intercalation in battery electrodes

Intra- and intermolecular electron transport

Electron transfer in artificial photosynthesis

Operando analysis tools with chemical contrast to probe nanoscale transport
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 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

Proposed elements for skyrmion processing

Skyrmion source

Skyrmion processing

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

Memristor structure
Stan Williams, HP labs

Spintronic Memristor
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

High resolution to probe electronic inhomogeneities
Spectroscopy + transport to measure G(r,t)

Dynamical structure factor $S(q,\omega)$ 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 $\sim k_B T$

Challenges
- Probe soft losses at energy $< k_B T$ with quasi-elastic RIXS using FT detection
- Probe fluctuations at timescales down to $\hbar/k_B T$ 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*


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

- **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

Blue - FePO$_4$
Red - LiFePO$_4$
Increased signal and improved resolution for RIXS: double-dispersion technique that benefits from high brightness

- 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 $h/k_B T$ timescale

**Nucleation kinetics**
[10.1126/science.1230915]

**Selective Catalysis**
[10.1038/nchem.1956]

**Polymer reptation**

**Intercalation kinetics**
[10.1039/C0EE00473A]

**Self assembly**

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**Time Scale (sec)**

- $10^{-1}$
- $10^{-3}$
- $10^{-5}$
- $10^{-7}$
- $10^{-9}$
- $10^{-11}$
- $10^{-13}$
- $10^{-15}$
- $10^{-18}$

**Energy Scale (eV)**

- $10^{-12}$
- $10^{-10}$
- $10^{-8}$
- $10^{-6}$
- $10^{-4}$
- $10^{-2}$
- $10^{0}$
- $10^{2}$

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**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, 1000 pm x 8 pm, 500 mA (New)
- **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)

First new multi-bend achromat rings

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

- **Elettra**

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 coherent soft x-rays are key to new basic energy science
  o 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
  o New capabilities are being developed for transformational science

• Soft x-rays address a broad range of fundamental science and technology
  o 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