From Quantum Materials to Solvation Dynamics: Experiments Proposed for Picosecond X-rays at the Advanced Photon Source

Linda Young
BESSY-VSR Workshop
Helmholtz-Zentrum Berlin
14-15 October 2013

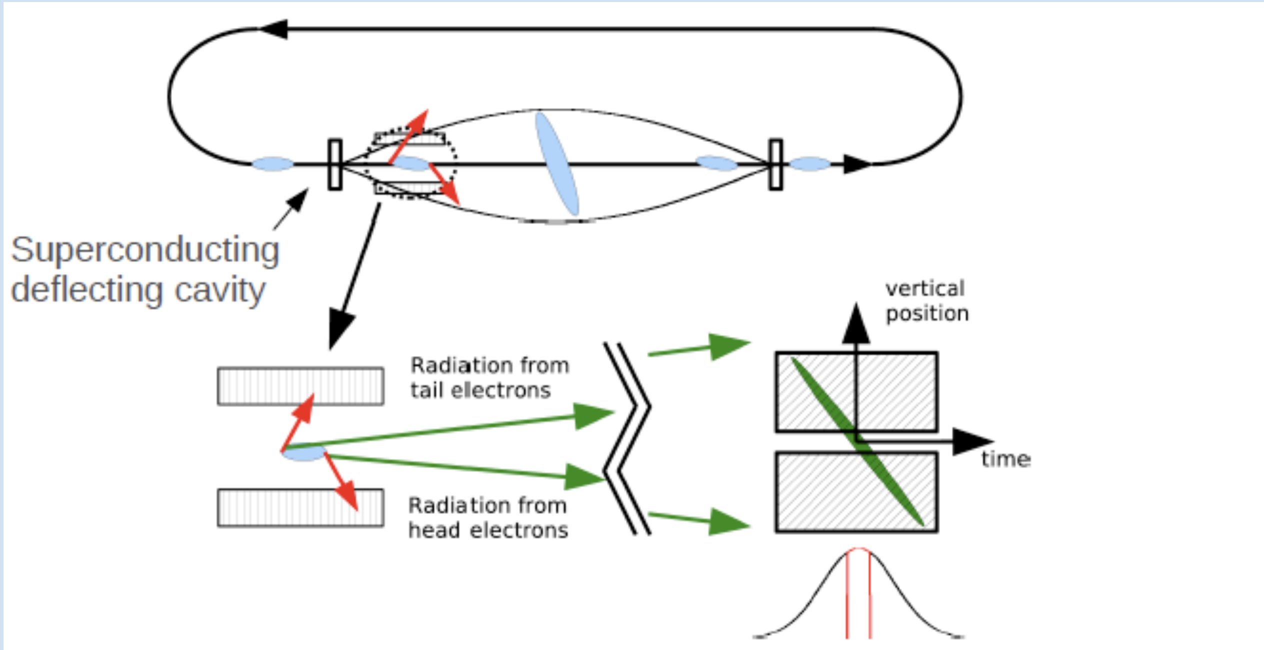
To direct and control matter at the quantum, atomic and molecular level.

2007 BESAC Grand Challenge Report

Outline

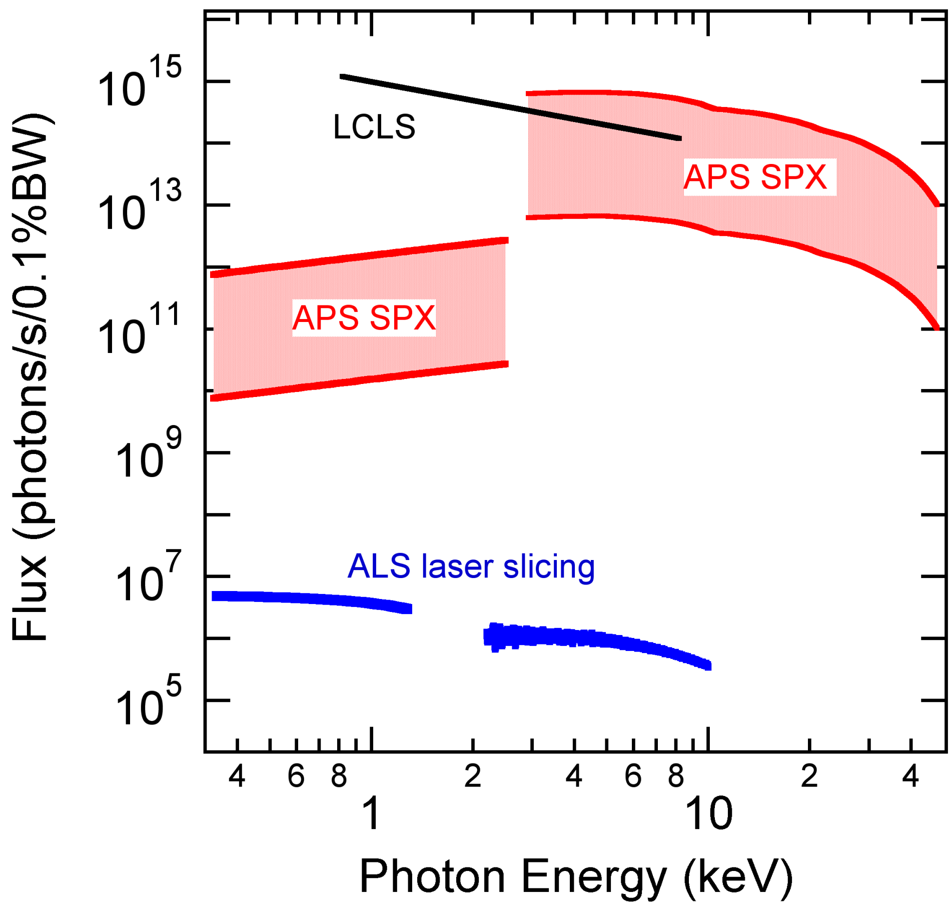
- A little history on picosecond science at the APS
- SPX First Experiments
  - Quantum materials
  - Nanoscale devices
  - Chemical dynamics in condensed phases
- SPX future – within the new MBA lattice design

Crab-cavity approach to short pulse x-rays in storage rings



Concept: A. A. Zholents *et al*., Nucl. Instrum. Methods A **425**, 385 (1999).
APS involvement: Lake Geneva Strategic Planning Meetings (August 2004).
APS simulation: M. Borland, Phys. Rev. ST Accel. Beams **8**, 074001 (2005).
APS implementation: Collaboration with JLab, Argonne Physics Division
Successful test of a traditional rf deflection cavity & tuner in February 2013

SPX: Short Pulse X-ray Facility
World's first high-repetition-rate, ultrastable, ultrafast x-ray source with pulse duration approaching 1 ps



FEATURES

- Gentle, linear x-ray probe
- Stable, high average flux, widely tunable, polarized (10^{13}/s)
- High energy capable 100 keV
- Strightforward synchronization to ~250 fs with external laser
- Multiple simultaneously operating beamlines providing soft and hard x-rays

APS fill patterns compared with other US storage rings

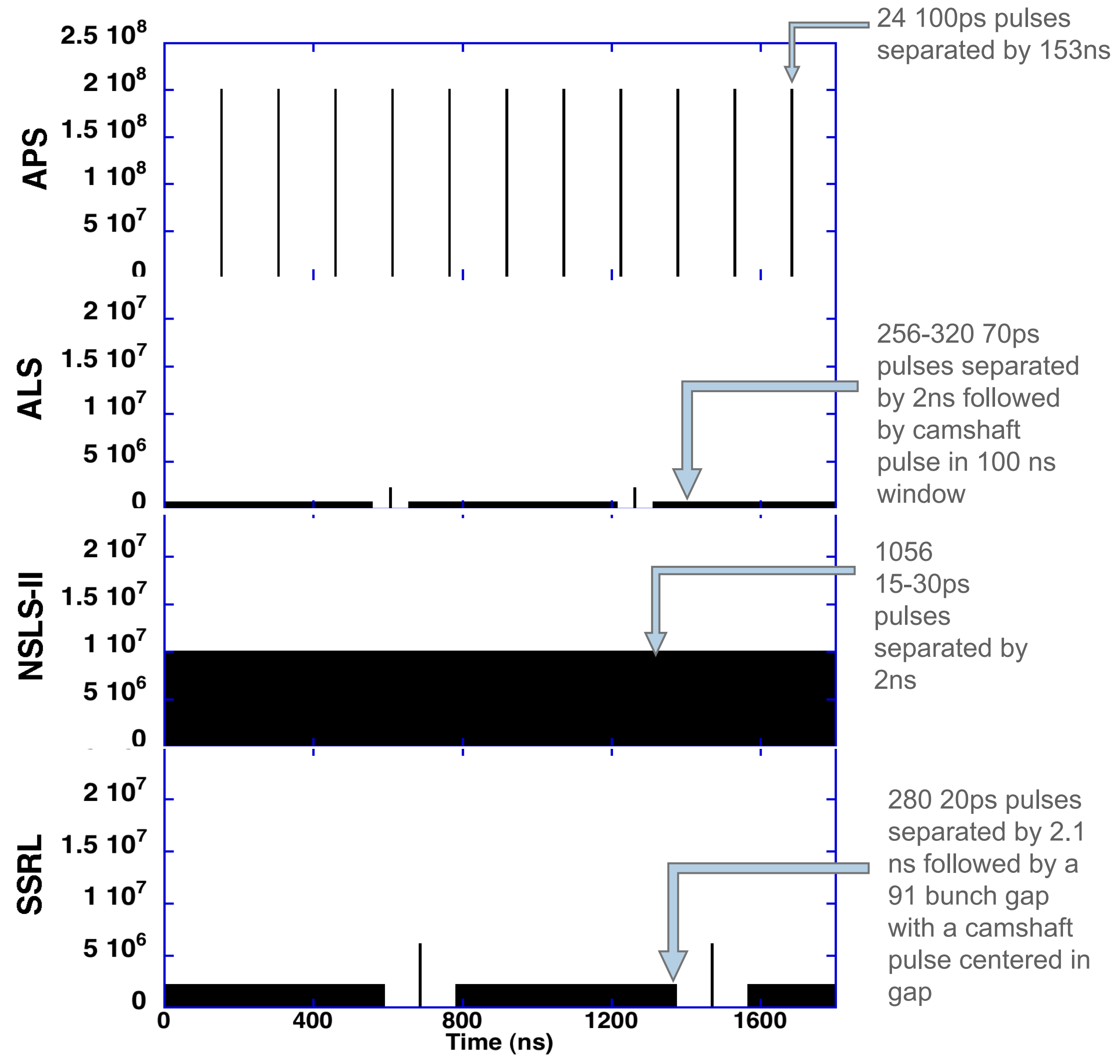
The APS x-ray pulse structure is advantageous for timing.

100% of pulses are potentially useful.

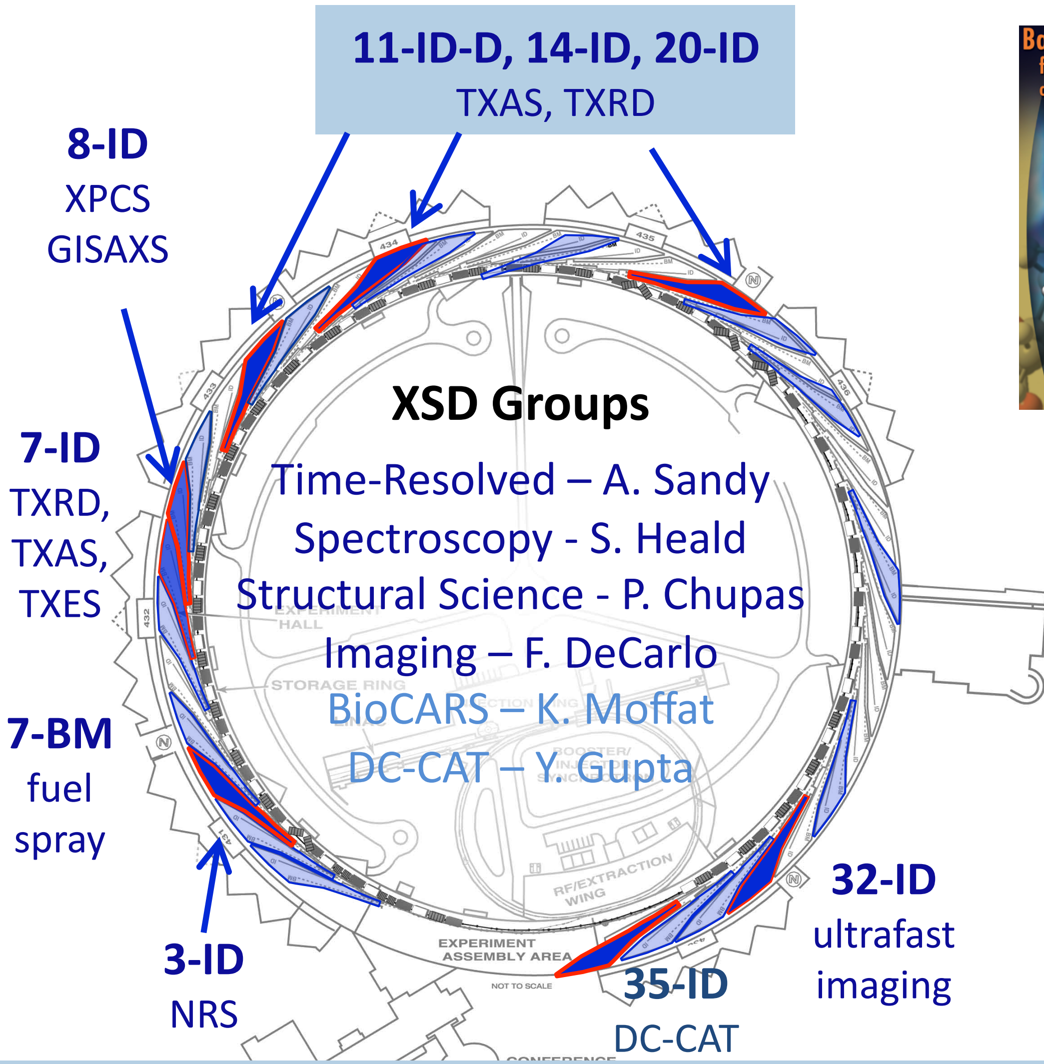
Timing mode 80% of runtime

Individual pulses can be readily chopped out
- sample recovery/replenish
- detector gating/readout

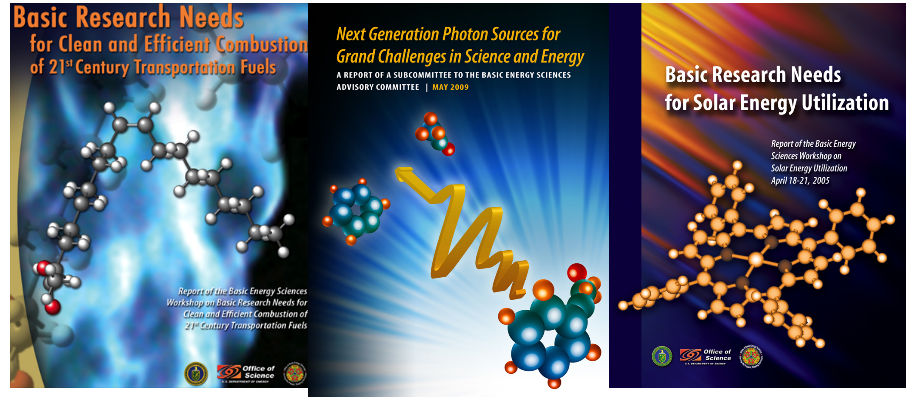
Previously timing underexploited.



XSD - Time-resolved research



DOE Basic Needs Reports



Challenges

- Understand and control the behavior of complex chemical, material and biological systems at a molecular level
- Understand nanoscale, polymer and fluid dynamics

Unique APS features: time-structure, single-shot phase contrast imaging, 10^{10} x-rays/pulse for time-resolved diffraction, 4 laser-pump/x-ray probe stations

Recent Science Workshops

X-rays in the Fourth Dimension

May 5-6, 2012, Chicago

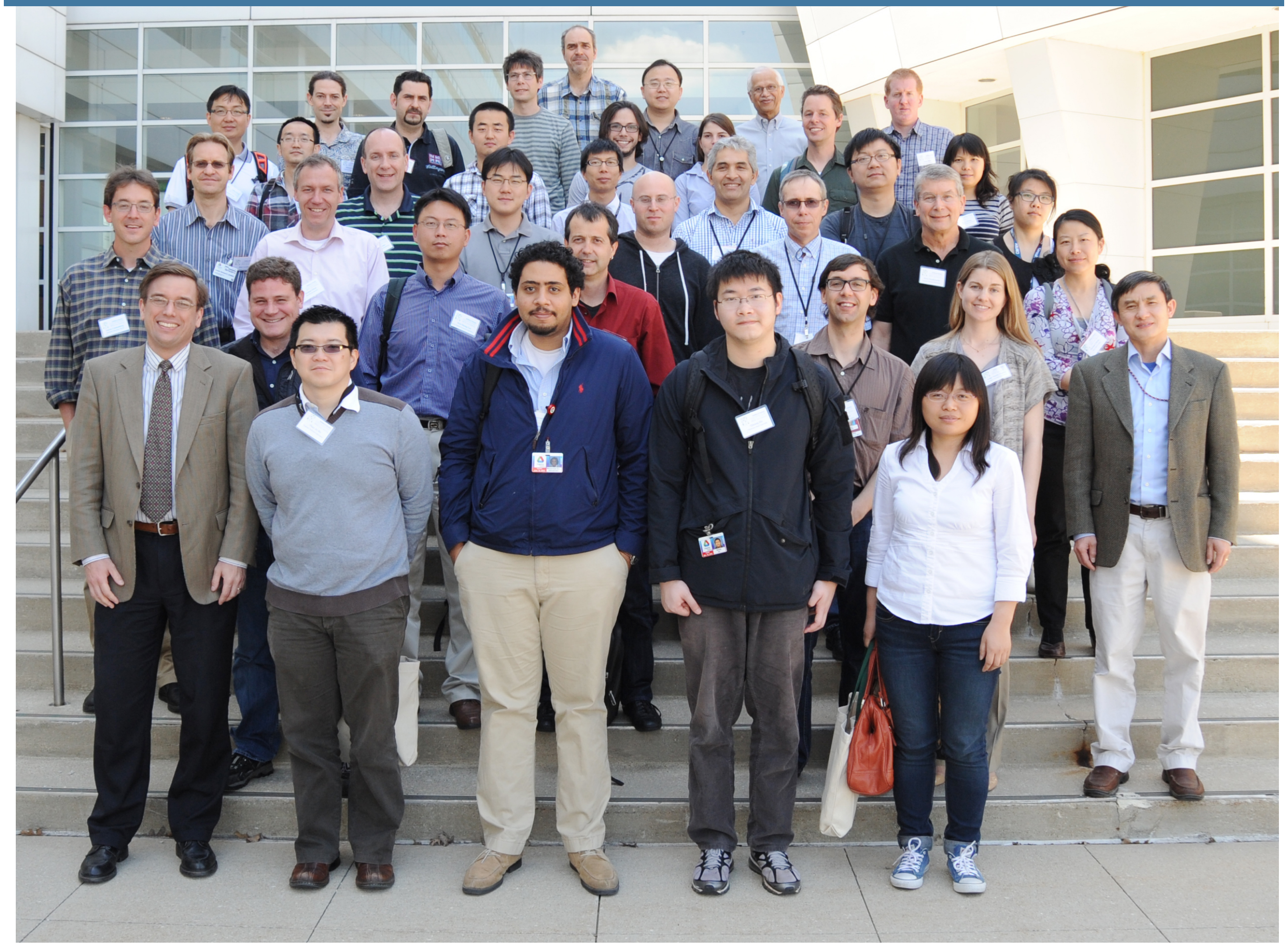
Organizers

Majed Chergui	**EPFL Lausanne, Switzerland**
Paul Evans	**University of Wisconsin, Madison, WI**
Aaron Lindenberg	**SLAC, Menlo Park, CA**
Bob Schoenlein	**LBNL, Berkeley, CA**
Linda Young	**Argonne National Laboratory, Argonne, IL**

Invited Speakers: Chi-Chang Kao(SSRL), Uwe Bergmann(LCLS), Harald Reichert (ESRF), Tetsuya Ishikawa(Spring-8,SACLA), Edgar Weckert(DESY), Christian Bressler(XFEL), Alexander Föhlisch(BESSY), Fulvio Parmigiani(FERMI@Elettra), Gabe Aeppli (UCL), Tony Heinz (Columbia), Toni Taylor (LANL), Wilfried Wurth (Hamburg) …

U.S. DEPARTMENT OF ENERGY

Ultrafast Dynamics in Strongly Correlated Materials, Atoms, Molecules and Clusters
APS User Meeting, May 7, 2013



Organizers

Yuelin Li
Dave Keavney
Philip Ryan
Steve Southworth

Invited Speakers

Paul Evans, Aaron Lindenberg, Adrian Cavallieri, Chris Milne, Christoph Bostedt, Wei-Sheng Lee, Anne Marie March, Michael Forst, Richard Averitt, Urs Staub

SPX First Experiments

June, 2013

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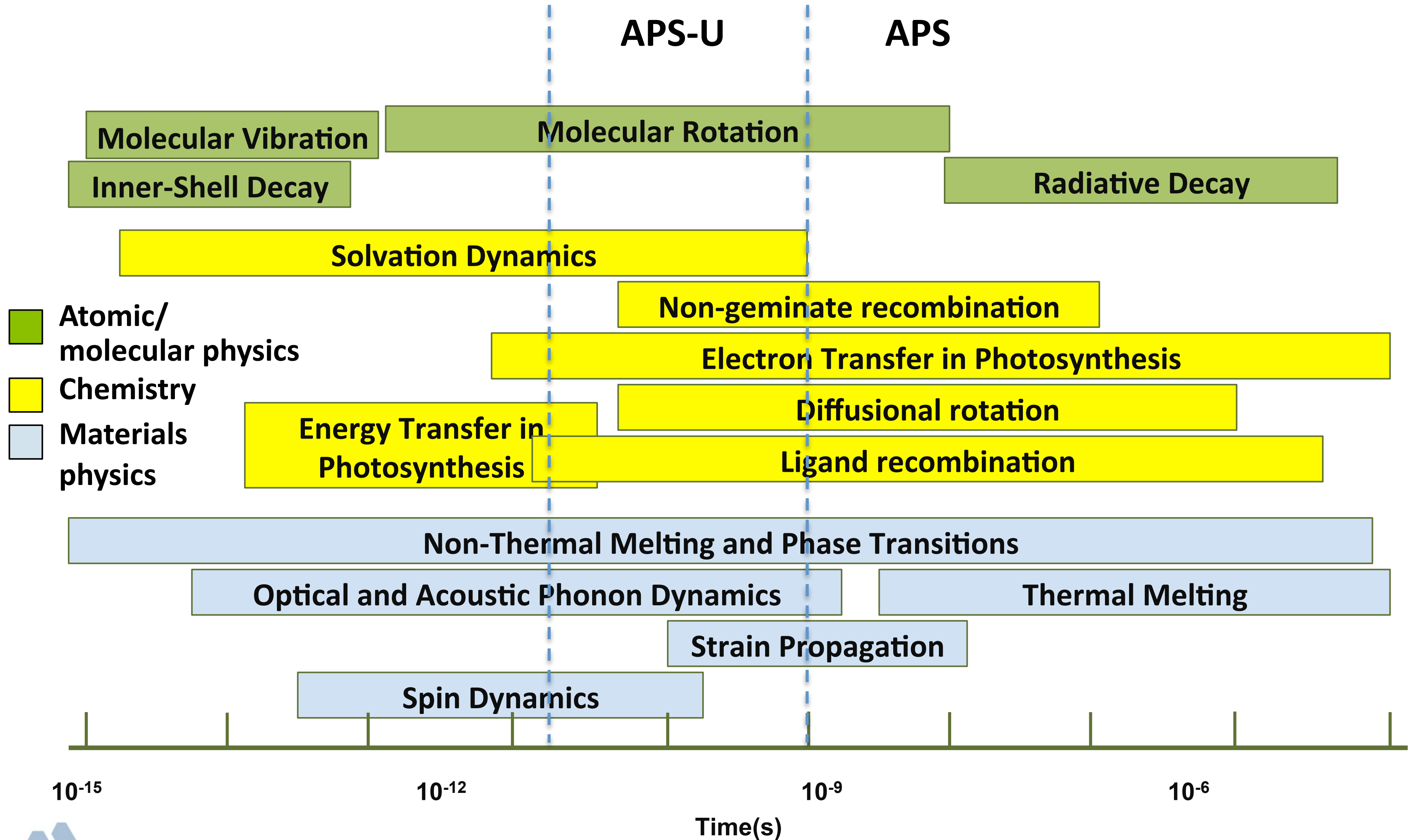
Viewing dynamics and transient states is fundamental to controlling chemical and material systems



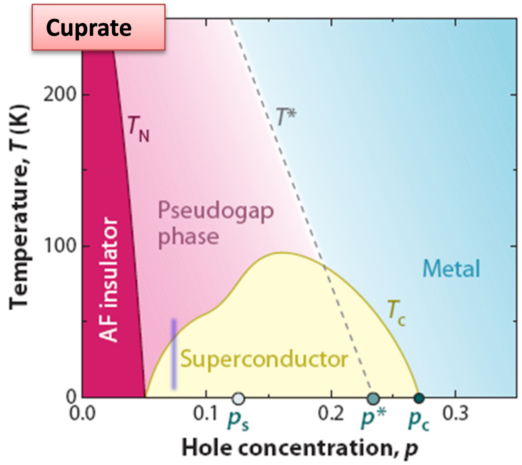
Quantum materials

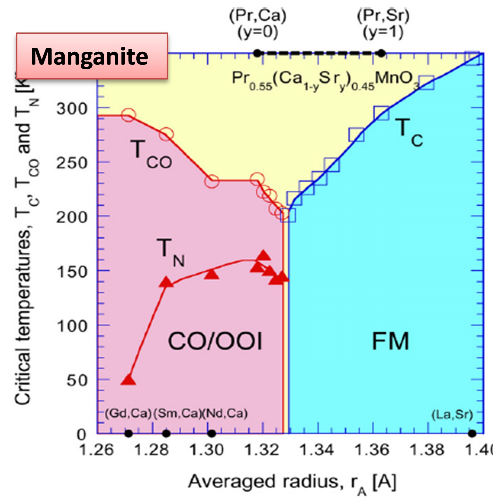
How do complex phenomena emerge from simple ingredients?

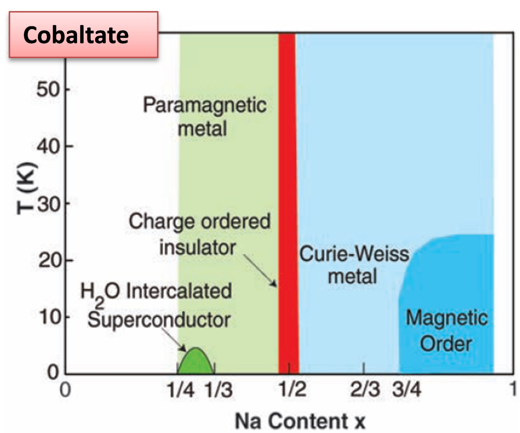
National Academies Press
CMMP 2010

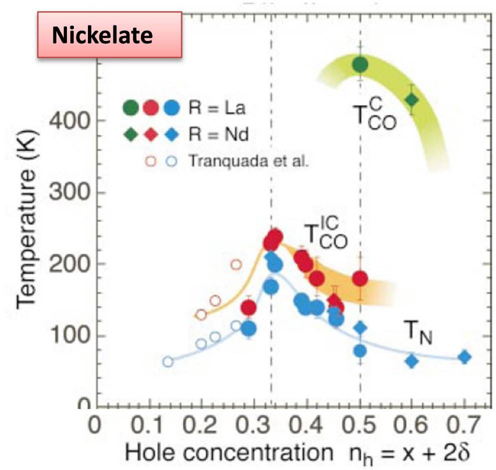
Quantum Materials

Can we understand and control the emergence of collective behavior in strongly correlated multiparticle systems?









Quantum Material - Nickelate

Nickelate: $La_{1.75}Sr_{0.25}NiO_4$

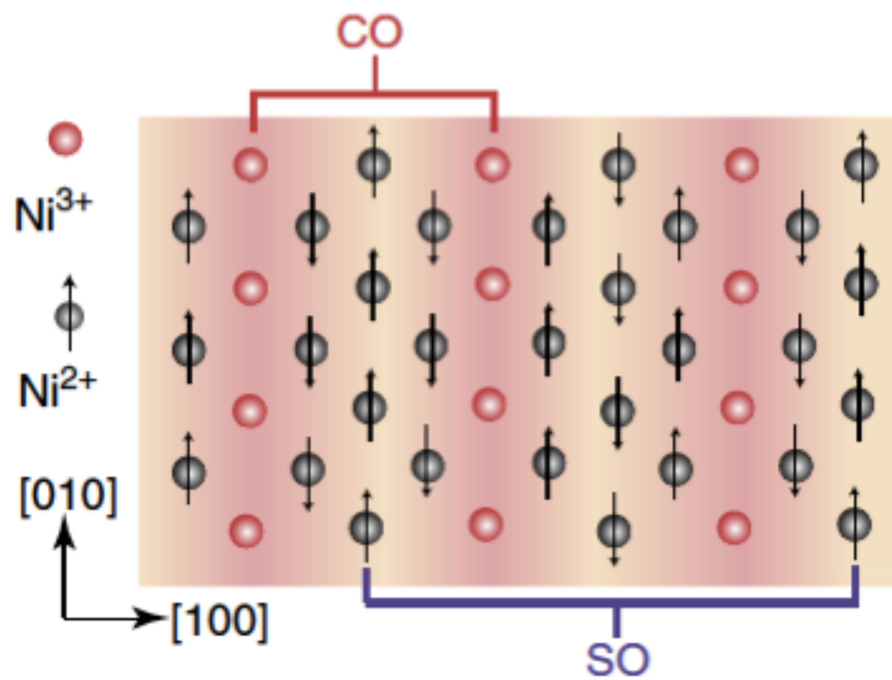
Electronic order on nm length scales

Charges & spins form regular patterns to minimize total energy

Manipulate order with light

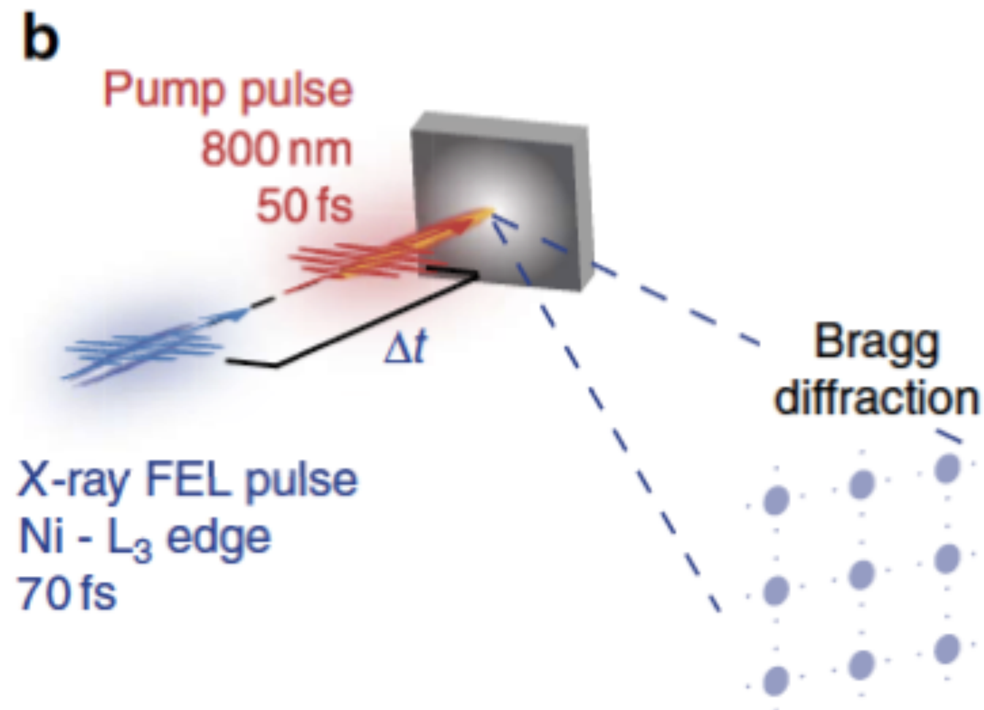
Probe charge order dynamics with x-ray diffraction (& optical reflectivity)

Compare light-induced vs thermally-induced phase change

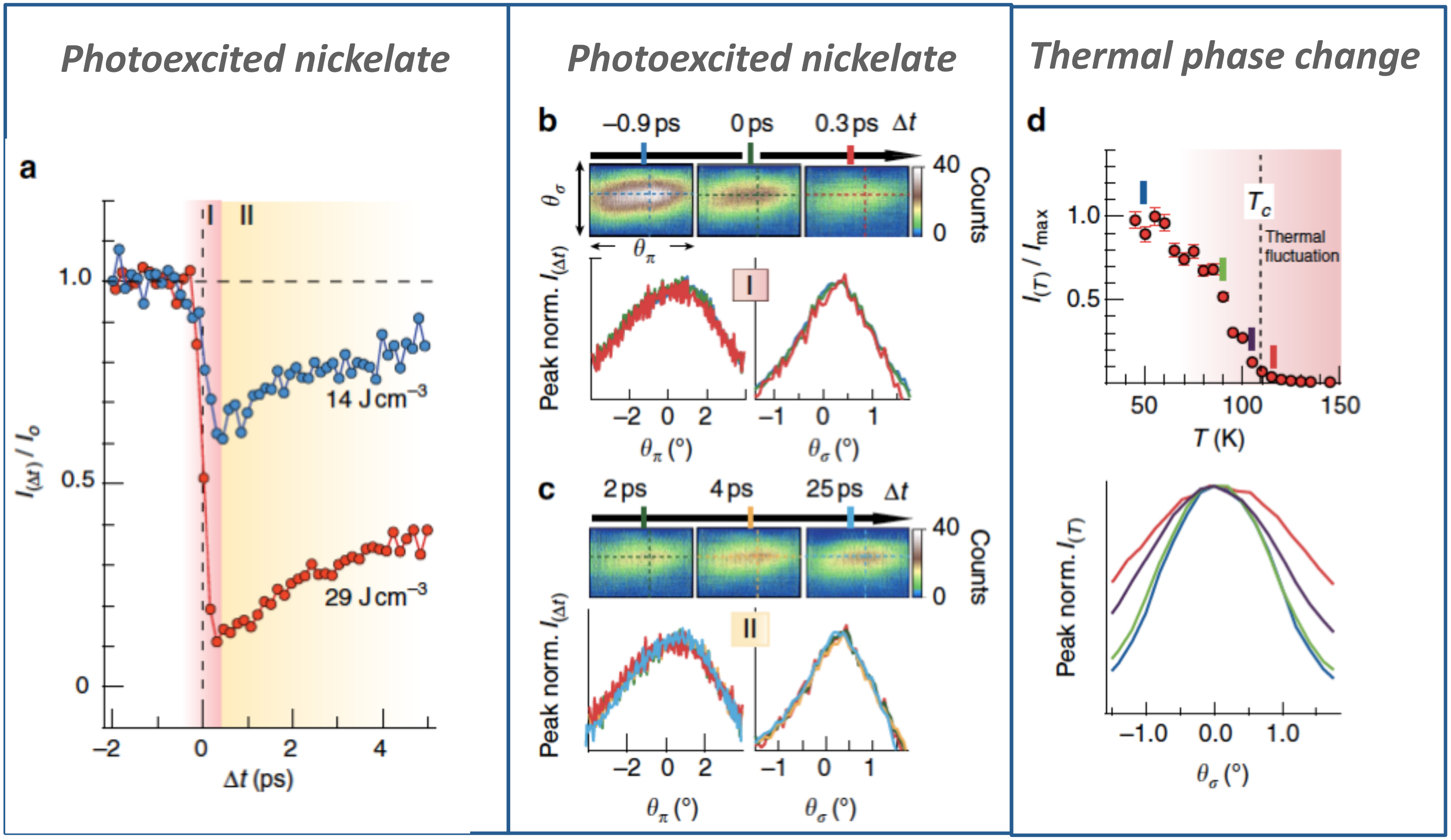


W. S. Lee et al., Nat Comm **3**, 838 (2012)

Observing charge order with resonant diffraction @ LCLS



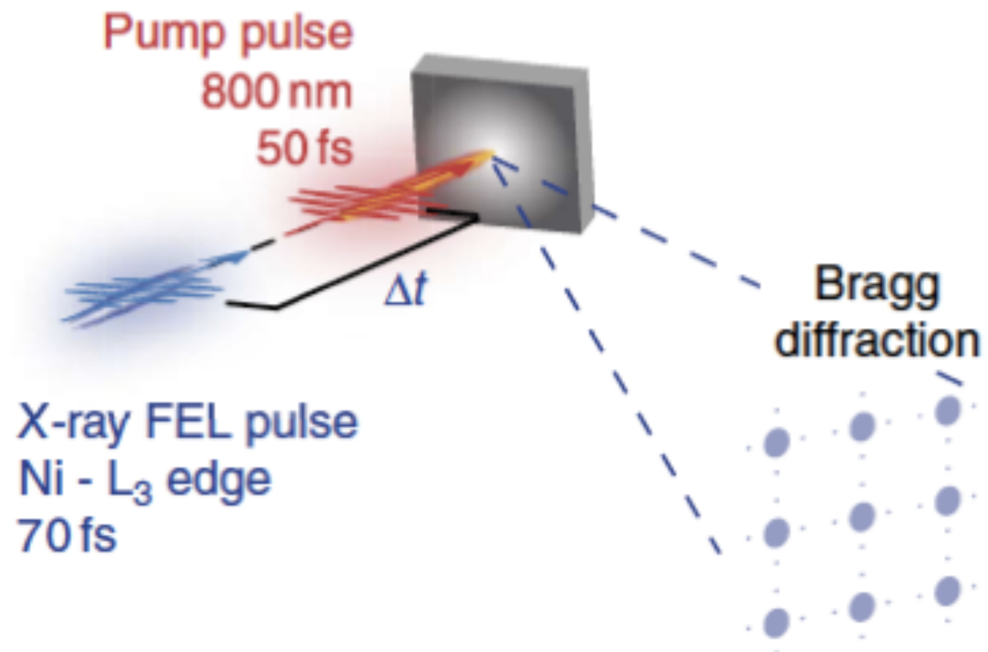
Light-induced Charge Order dynamics: Timescales



CO recovery timescales measured by XRD: τ_{fast} = 2 ps, τ_{slow} = 57 ps

CO melting by temperature rise leads to different long range ordered state

Observing charge order with resonant diffraction @ LCLS



Fluence ~0.5 mJ/cm^2/pulse
300 X 300 μm^2 target
Fraction of LCLS pulse energy used ~ 10^{-3}-10^{-4}
Number of shots per time step: ~ 40
Repetition rate: 60 Hz

Repetive measurement, linear x-ray probe, ps timescale

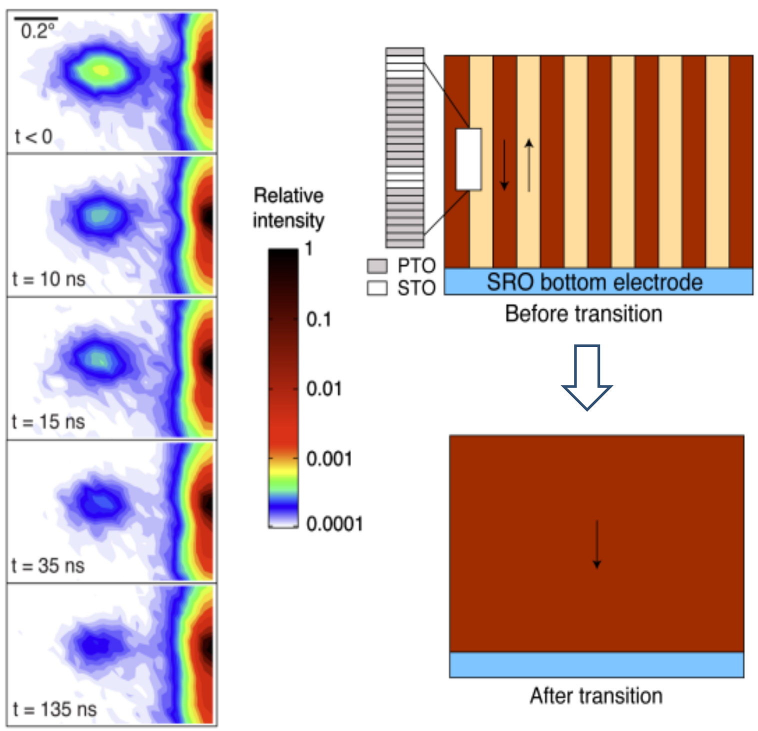
Beyond macroscopic averages to microscopic dynamics

Repetitive measurement, linear x-ray probe, ps timescale, nanometer focusing

Understanding nanoscale switching dynamics

Current capability:

Macroscopically averaged measurements of nanoscale order.

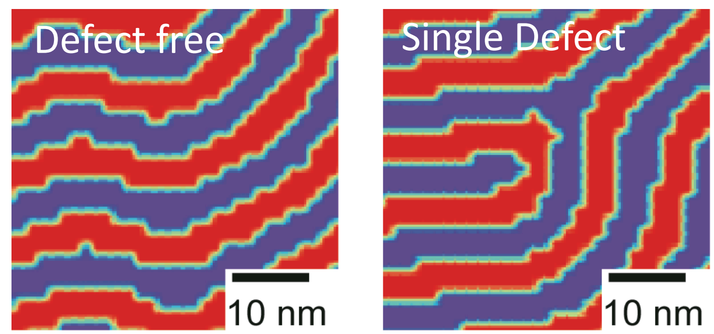


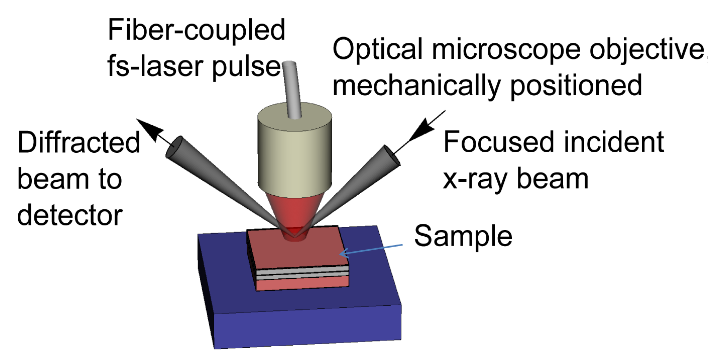
Ferroelectric stripe nanodomains (APS)

Jo *et al*., Phys. Rev. Lett. **107**, 055501 (2011).

Capability with APS-U

Nanofocused, picosecond pulses at high rep rate enable one to locally address functional materials at timescales of the intrinsic excitations.





Chemical dynamics in condensed phases

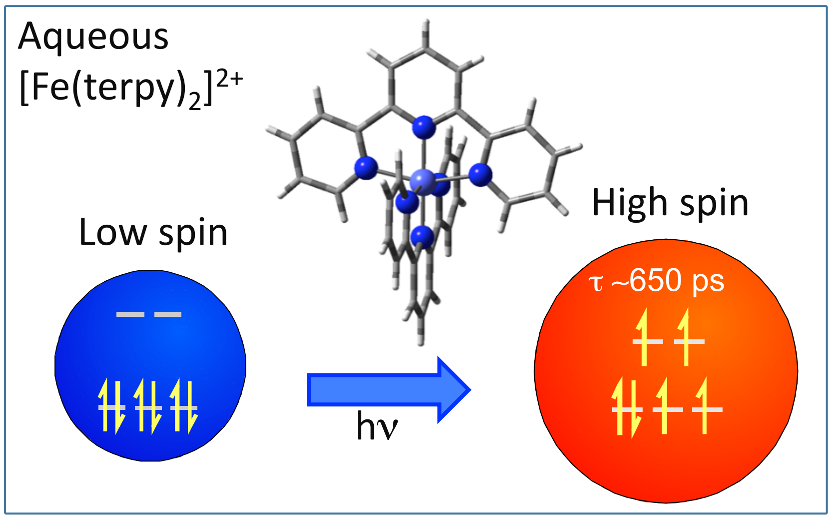
Repetitive measurement, linear x-ray probe, ps timescale, ultrastable

Understanding chemical dynamics in solution

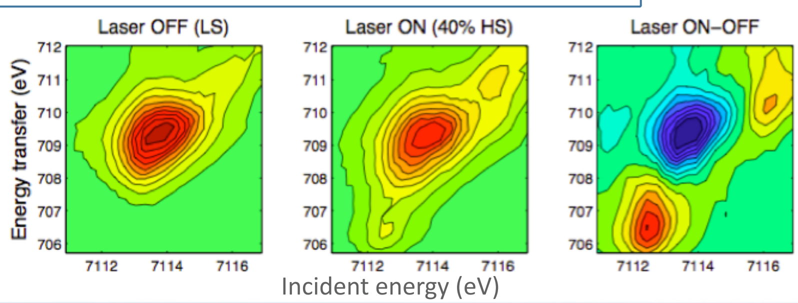
Can we observe and control pathways for efficient chemical reactions?

Current capability

- High-rep-rate studies provide local atomic and electronic coordinates for transient species of >100 ps lifetime via simultaneous XAS, XES, RIXS, XDS).

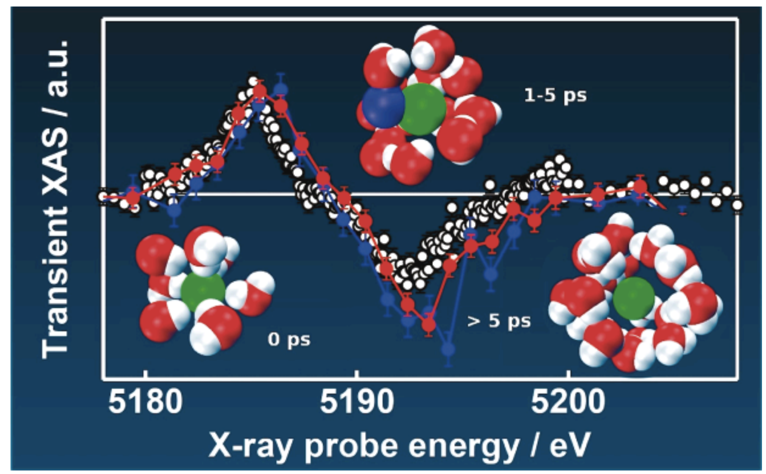


K. Haldrup et al., *J.Phys Chem.* ***A116**, 9878 (2012)*
G. Vanko et al, *JESRP (2012)*



Capability with APS-U

- SPX enables probe of transient phenomena down to ~2 ps timescale (charge transfer, electron transfer, ligand substitution, solvent caging)



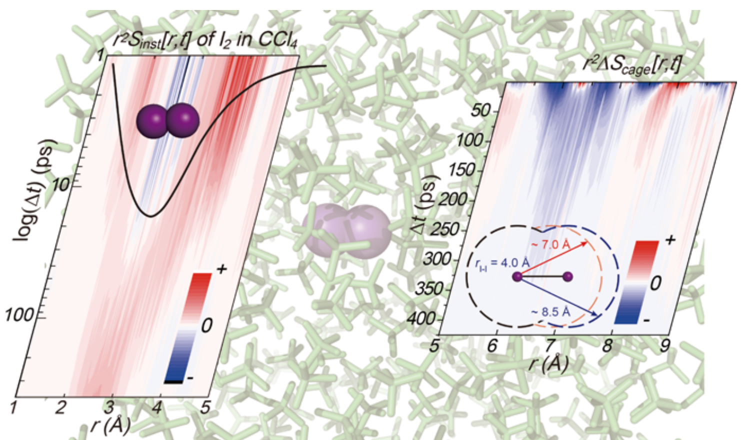
Solvation of iodine atom in water – MD simulations predict transient formation $I^o(OH_2)$ complex that lives 3-4 ps, followed by expansion of solvent shell; *V.-T. Pham et al, JACS (2011)*

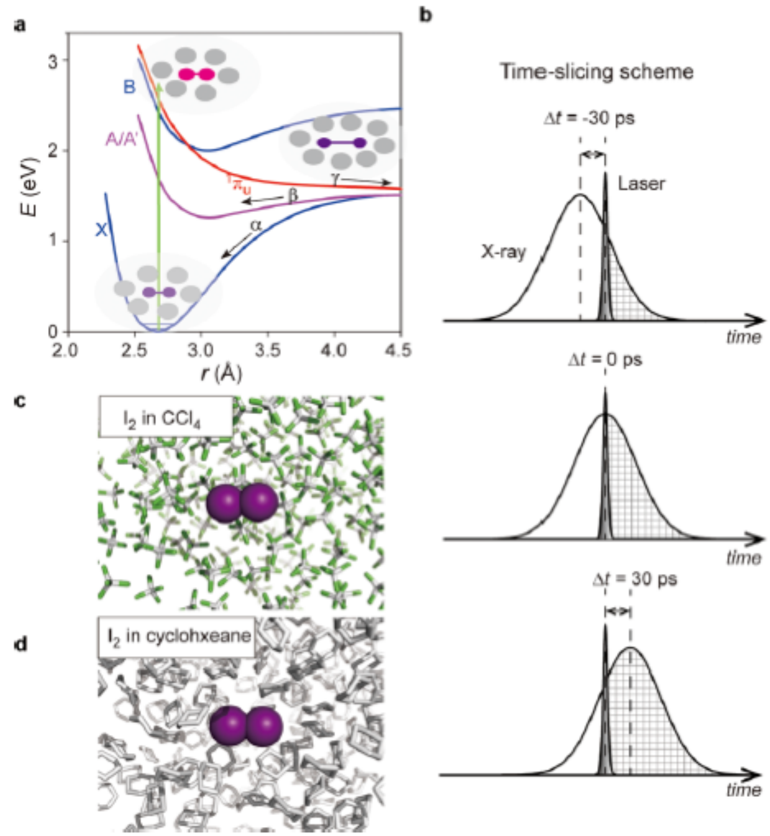
Ultrastable pulses allow sub-picosecond visualization of solute-solvent rearrangements

TIme-slicing and deconvolution using ultrastable synchrotron pulses has improved time resolution to ~10% of the x-ray pulse duration. Can be readily ported to SPX.

- Visualizing solvent-solute interplay critical for understanding chemical reactions.
- Test case: I_2 geminate recombination & vibrational relaxation
- Observe correlated I_2 and solvent cage expansion ($I_{2:}$ 2.67 – 4Å) and solvent ~1.5Å and contraction

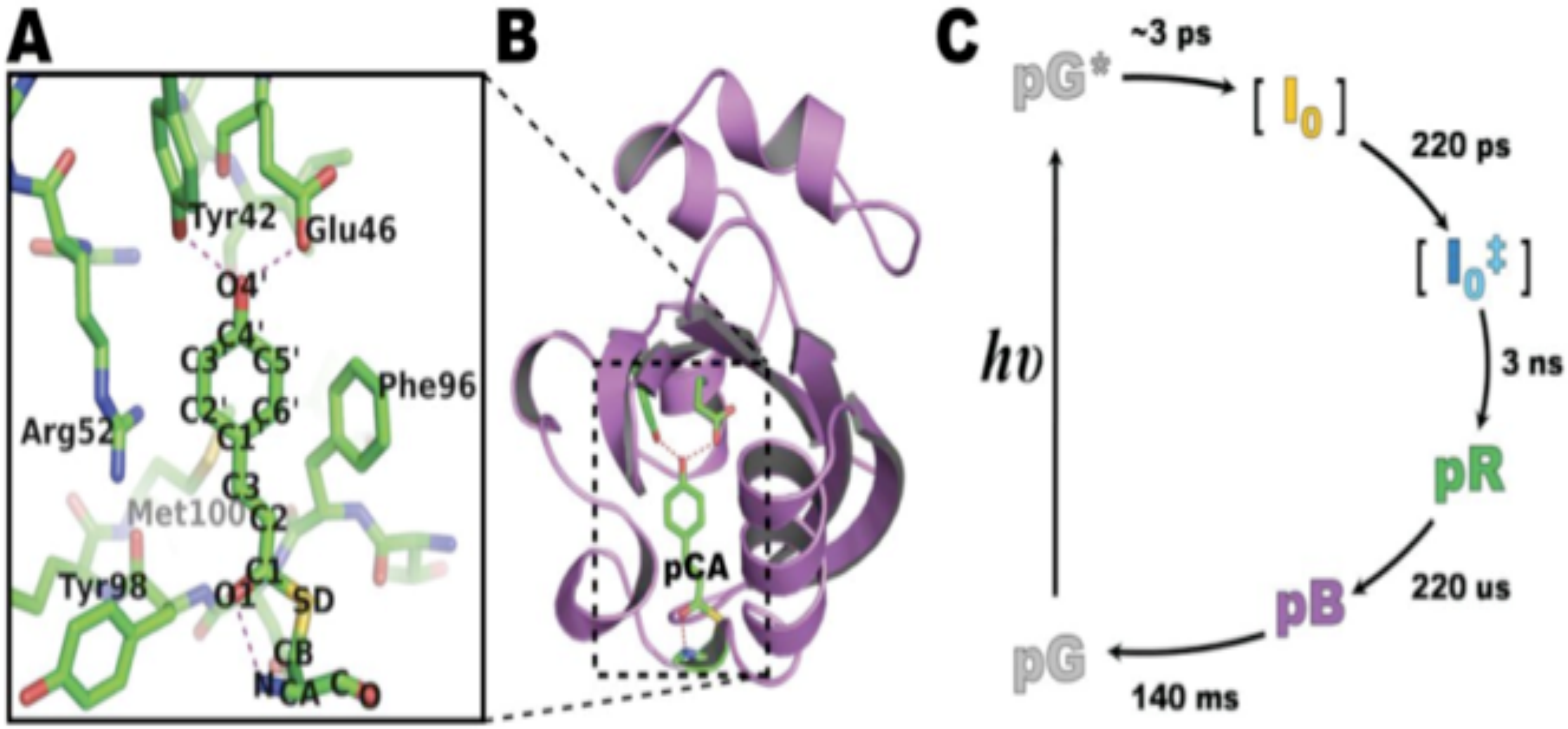
$\tau(CCl_4)$ ~ 16 ps, 76 ps,
τ(cyclohexane) ~ 55 ps





From H. Ihee Group, JACS **135**, 3255 (2013)

Dynamics of Protein Structural Changes

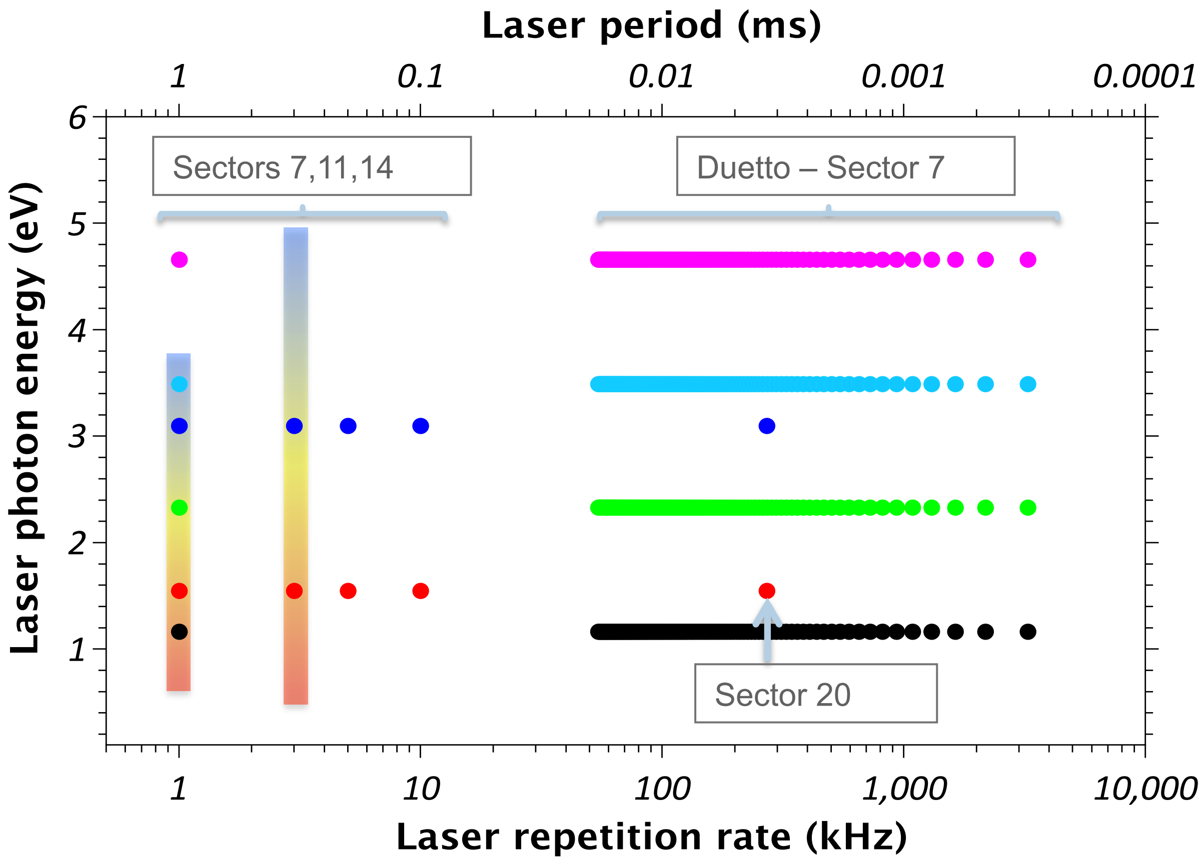


Photoactive Yellow Protein

Moffat, Srajer, Henning

Time-resolved Laue diffraction from highly stable undulator sources yields structure amplitudes accurate enough to reveal small structural differences.

Multiple optical excitation sources operating at the APS



Gilles Doumy

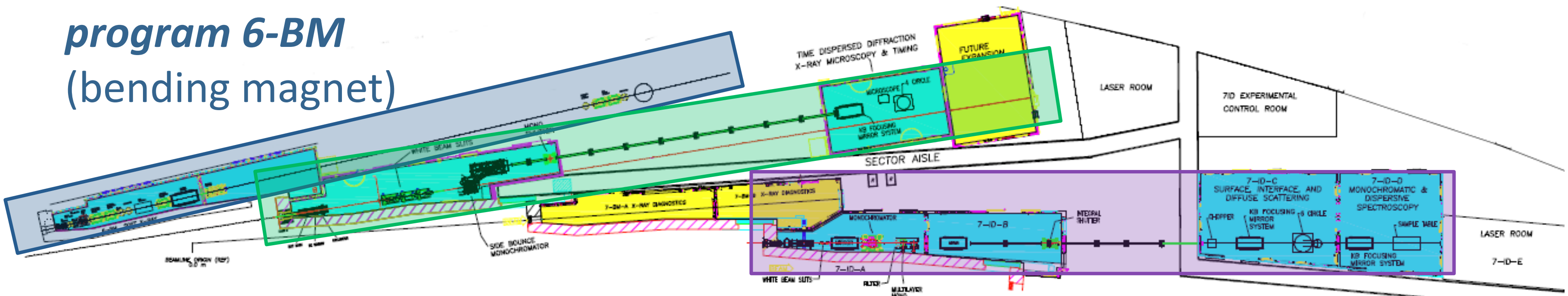
- Usable repetition rate governed by sample recovery/replenishment.
- Lasers can produce optically excited states, pressure jumps, temperature jumps, vibrational excitation; align, orient, polarize molecules; inject electrons; desorb molecules; ablate materials

SPX --- Evolving Plans

SPX: Multiple simultaneously operating beamlines

Soft x-ray science program 6-BM
(bending magnet)

Hard x-ray imaging & microscopy 6-ID (time-dispersed diffraction)
(insertion device)



Hard x-ray spectroscopy and scattering 7-ID
(insertion device)

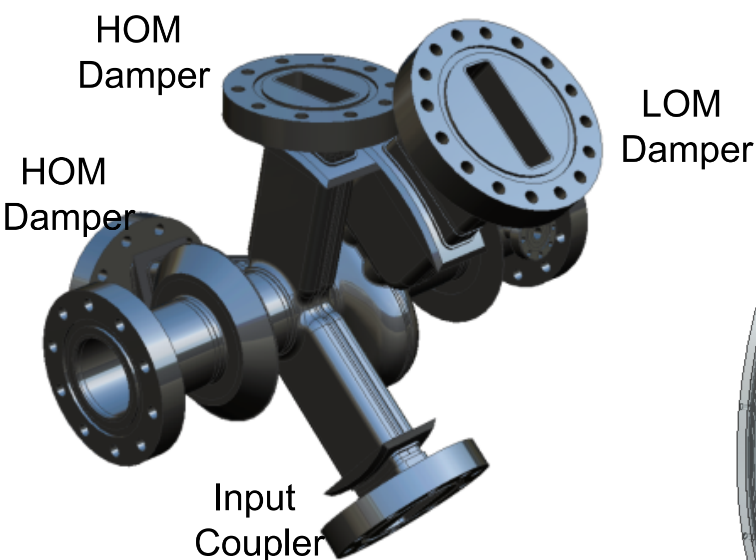
- **Three independently operating beamlines being planned in APS-U**
  - Hard x-ray spectroscopy and scattering (two endstations)
  - Hard x-ray imaging and microscopy (novel uses of chirped pulses)
  - *Soft x-ray program (one bending magnet, two endstations)*
- **Co-located laser facilities**

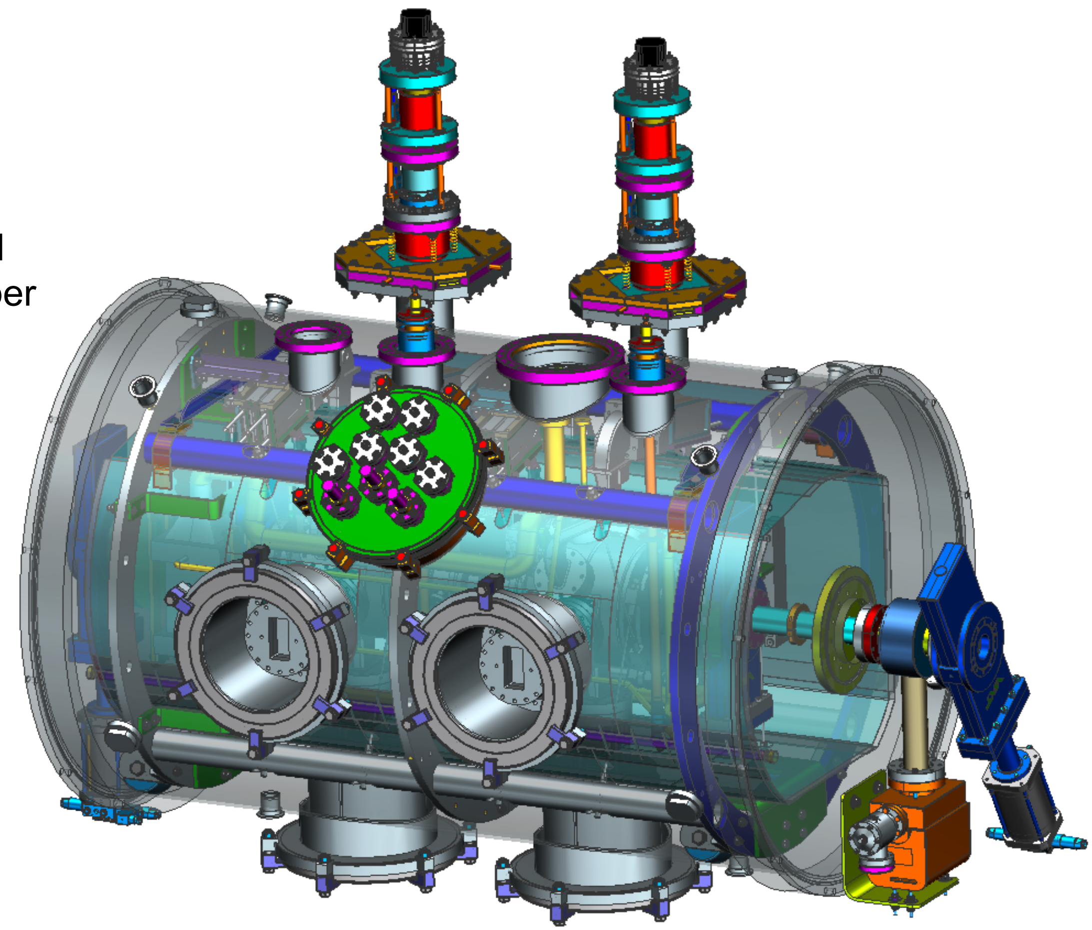
SPX beamline specifications - flexibility is key

Hard and Soft X-ray Beamlines	Energy Range, Bandwidth (dE/E)	Pulse duration, Repetition Rate, Spot size, Flux, Comments
SPX Spectroscopy and Scattering	4-35 keV, 10^{-4}, 10^{-2}	2, 10, 100 ps; 6.5 MHz – 271 kHz C-station: 6.4 x 6.4 µm D-station: 2.7 x 3.4 µm 10^{11}, 10^{12}, 10^{13} x-rays/s for 10^{-4} BW
SPX Imaging and Microscopy R. Reininger et al. RSI 84, 053103 (2013)	7-14 keV 10^{-4}	2-80 ps, 6.5 MHz Spotsize: 33 x 14 µm Time dispersion – 0.052 ps/µrad 50 nm spot w/zone plate microscope
SPX Soft X-ray Spectroscopy R. Reininger et al. JSR 20, 654 (2013)	200-2000 eV $5x10^{-4}$, $2x10^{-4}$	1.5 - 100 ps, 6.5 MHz Source disp: 25 ps/mm 10x10µm $2x10^{9}$ x-rays/s for $5x10^{-4}$ BW Linear and circular polarization

Original SPX0 prototype cavity

2 cavity cryomodule, producing 1 MV chirp





Full SPX implementation: 4 m length

MBA lattice design - October 1 revision

MBA: 6 GeV, 200 mA, 80 pm, flexible timing modes
Present: 7 GeV, 100 mA, 3 nm, flexible timing modes

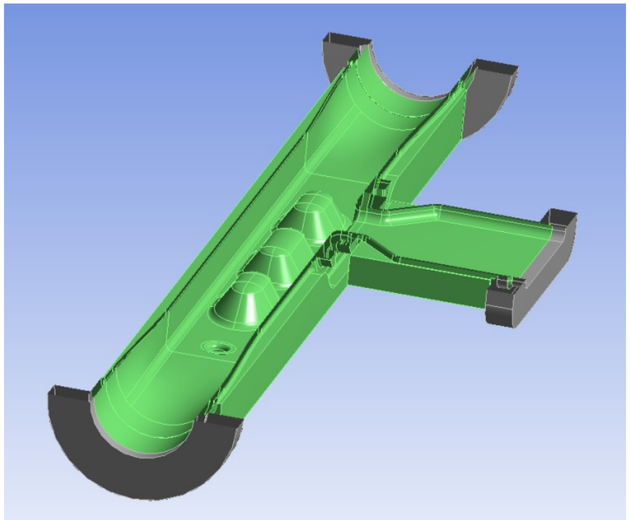
Quantity	Symbol	Range	Units
Total current	I	200	mA
Number of bunches	N_b	48-324	
Bunch rate	f_b	13-88	MHz
Rms bunch duration	σ_t	70-18	ps

48 bunch mode: x, x' = 7.4 μm, 5.7 μrad
y, y' = 10.9 μm, 3.8 μrad

Michael Borland – white paper on APS website

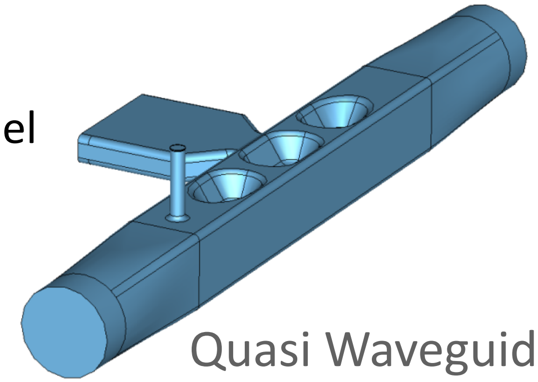
QMiR Deflecting Cavity Geometry, Collaboration with FNAL and ANL/APS

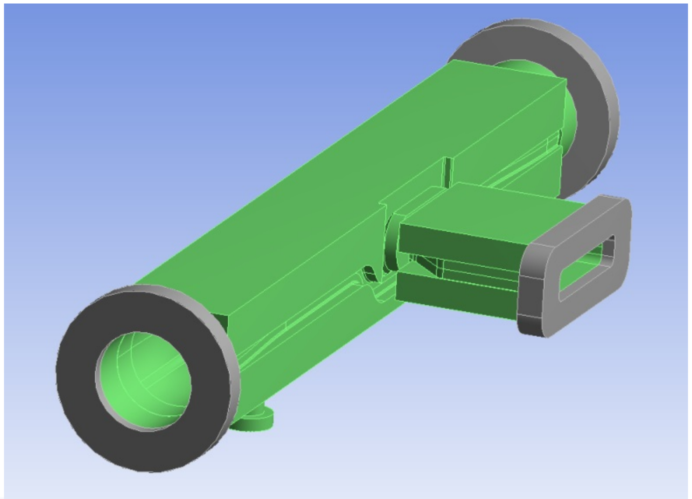
- QMiR- Quasi Waveguide Multi Cell Resonator
- Extremely high shunt impedance that can provide 2 MV of deflecting voltage with just one QMiR.
- Beam-induced HOMs propagate along the beam pipe and can be easily dumped outside the cryostat.
- Compact cryostat and significantly reduced cryogenic load compared to elliptical deflecting cavities.



This is what we are building

Shell model

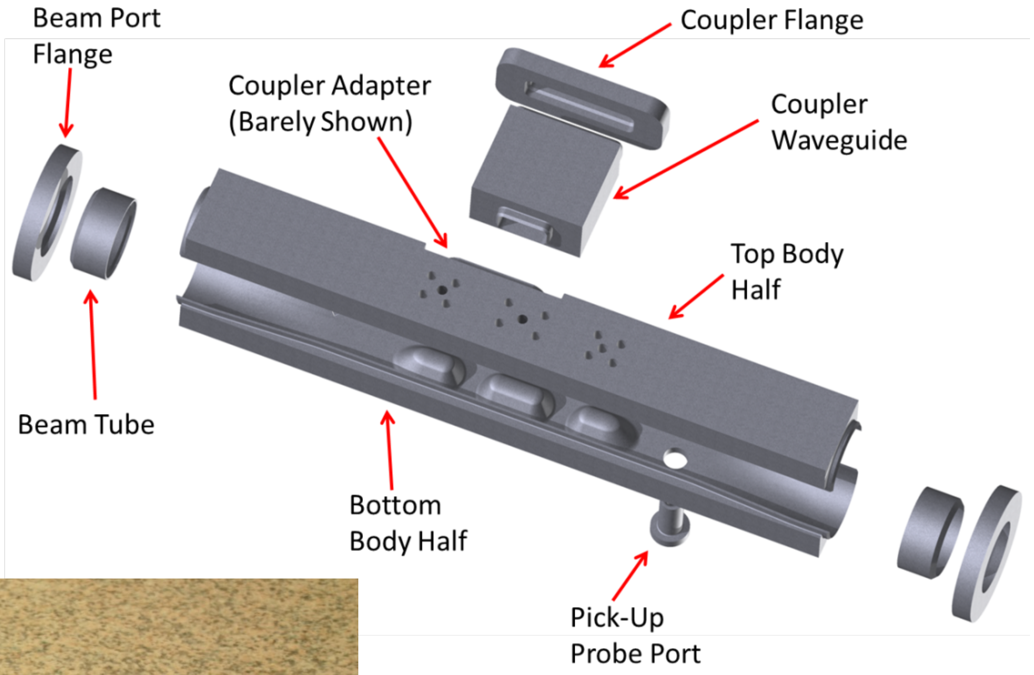


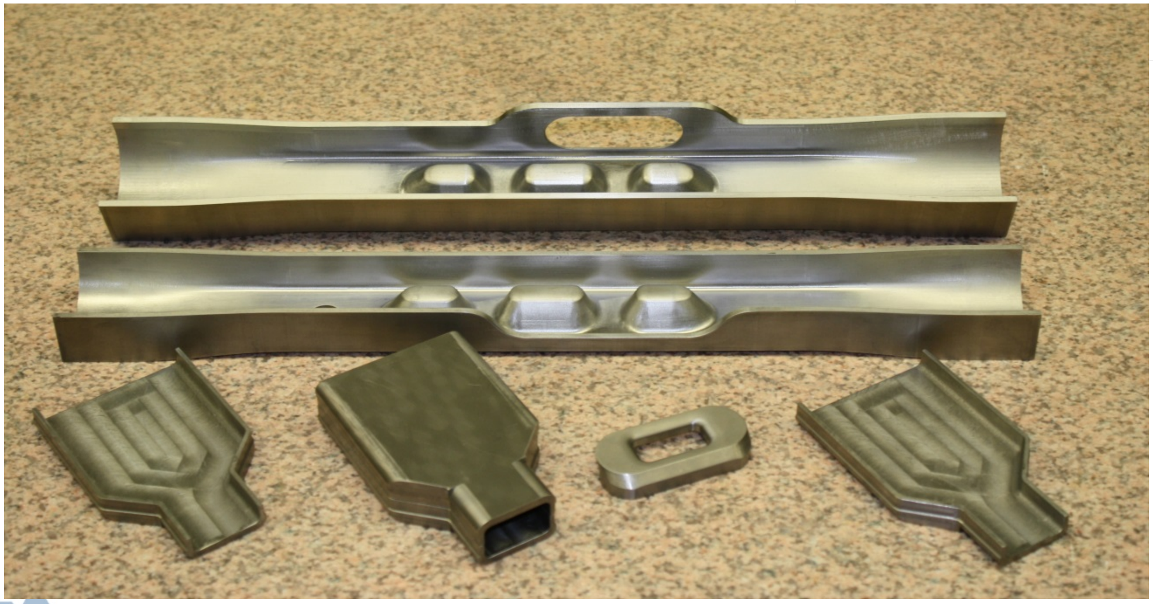


Quasi Waveguide Multicell Resonator
Patent application is pending

Fabrication

- 9-month schedule:
  - Start date: April
  - Nb cavity should be cold/RF tested by the end of December





Summary

- X-rays underutilized in time-domain. Emphasis has been on structure. Synchrotrons will continue to play an important role for time-domain studies.

- Picosecond timescales are ideal for nanoscale dynamics: quantum materials, nanoscale devices, chemical dynamics in solution phase …

- Synchrotron-based picosecond x-ray sources are complementary to XFELs. Comparable in flux and superior in stability, tuning and use of x-rays as a simply-understood, linear response probe of matter. APS-SPX appears feasible/enhanced within the new MBA lattice design.

- Flexibiliy important: Timing (pulse duration and interval). Bandwidth (monochromatic, pink). Polarization (linear, circular). Stability (time resolution below pulse duration)

- Highly experienced user community eager and ready for an ultrastable, widely tunable, polarized, high-average-flux x-ray source.

APS-U MBA lattice workshop: October 21-22

Acknowledgements: Recent Community Involvement

- SPX STAC (Scientific and Technical Advisory Committee)
  - Paul Evans (U Wisconsin) - Chair
  - Christian Bressler (XFEL – Hamburg)
  - Lin Chen (Argonne/Northwestern)
  - Hermann Durr (SLAC)
  - David Fritz (LCLS-SLAC)
  - Steven Johnson (ETH – Zurich)
  - Aaron Lindenberg (Stanford-SLAC)
  - David Reis (Stanford-SLAC)
  - Stephen Southworth (Argonne)

Additional Contributors to 2011 Science Case: Roy Clarke (U Michigan), Victor S. Batista (Yale University), Felix N. Castellano (Bowling Green State University), Edward W. Castner, Jr. (Rutgers University), Robert A. Crowell (Brookhaven National Laboratory), Christopher M Laperle (Providence College), Christoph Rose-Petruck (Brown University), Roseanne J. Sension (University of Michigan), Dario Arena (Brookhaven), Bill Bailey (Columbia), Paul Crowell (Minnesota) Jin Wang, Eric Dufresne , Yuelin Li, Haidan Wen, Dave Keavney, John Freeland (Argonne)