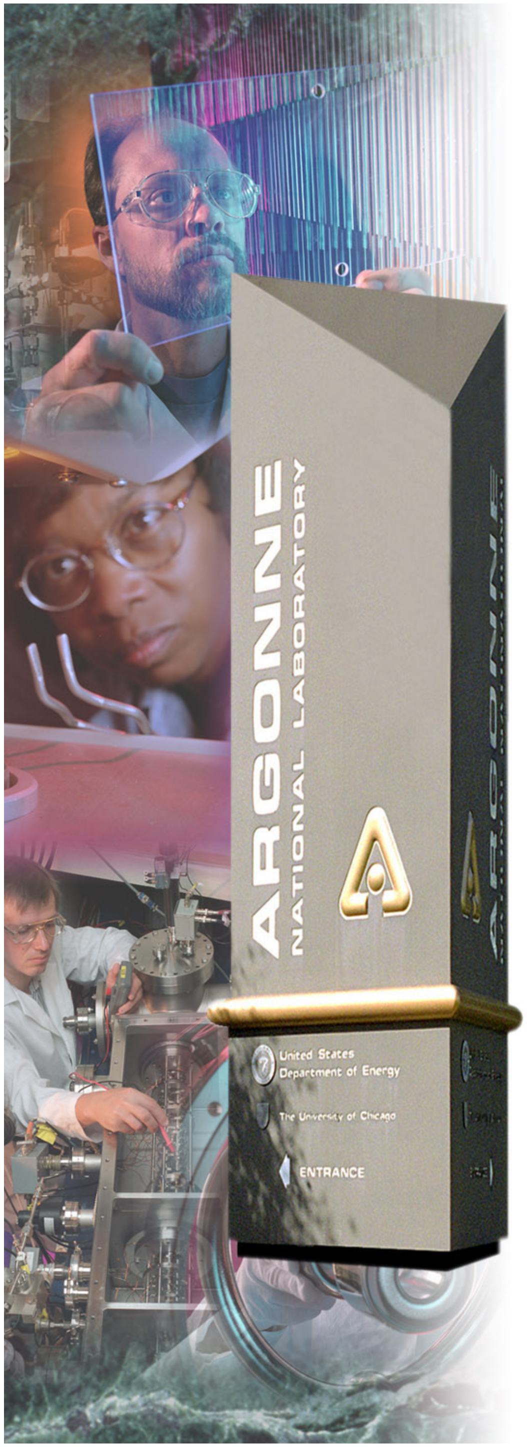


Overview of Current Capabilities and Experiments Using the Time Structure of the Beam

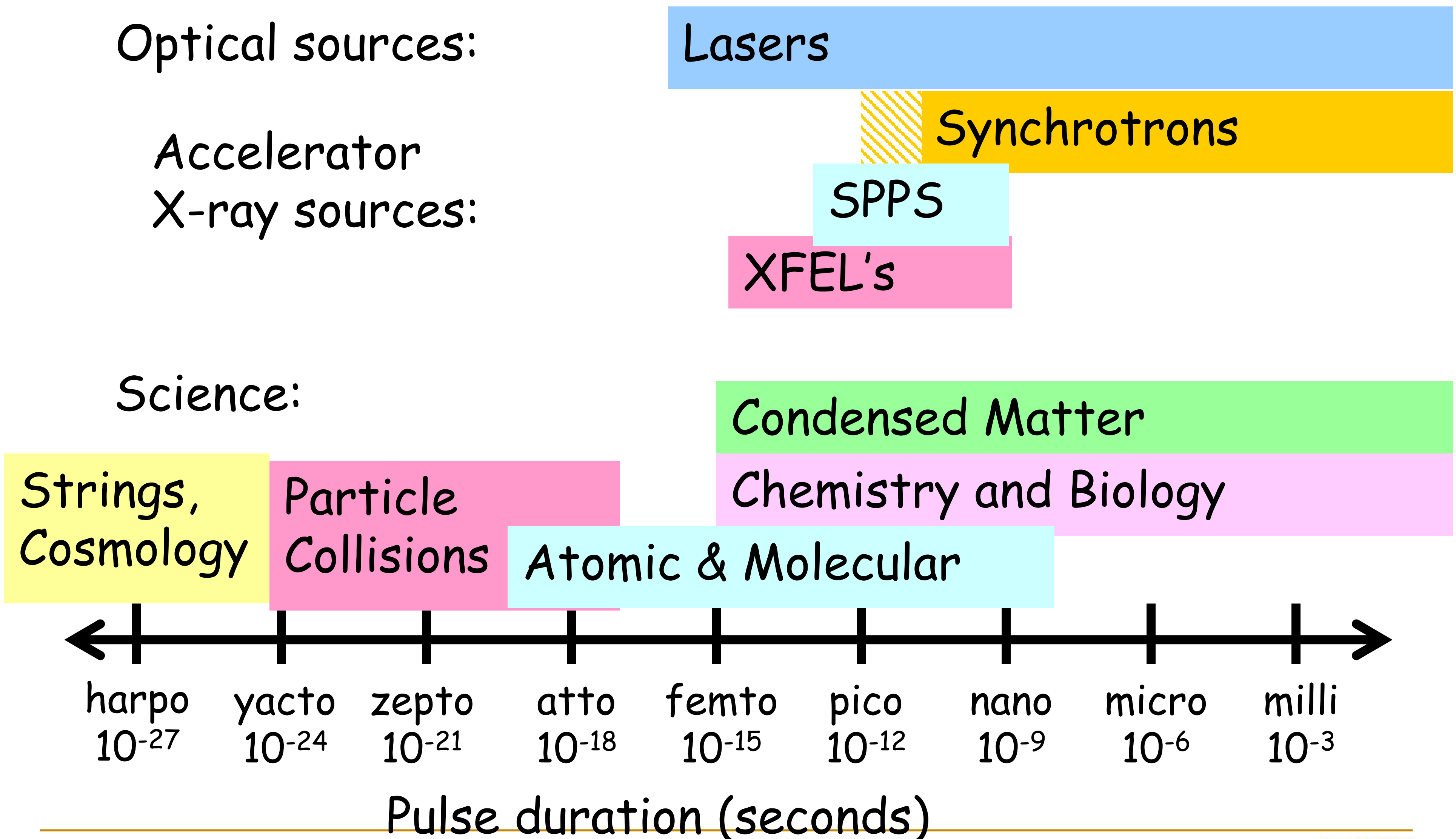
J. Murray Gibson

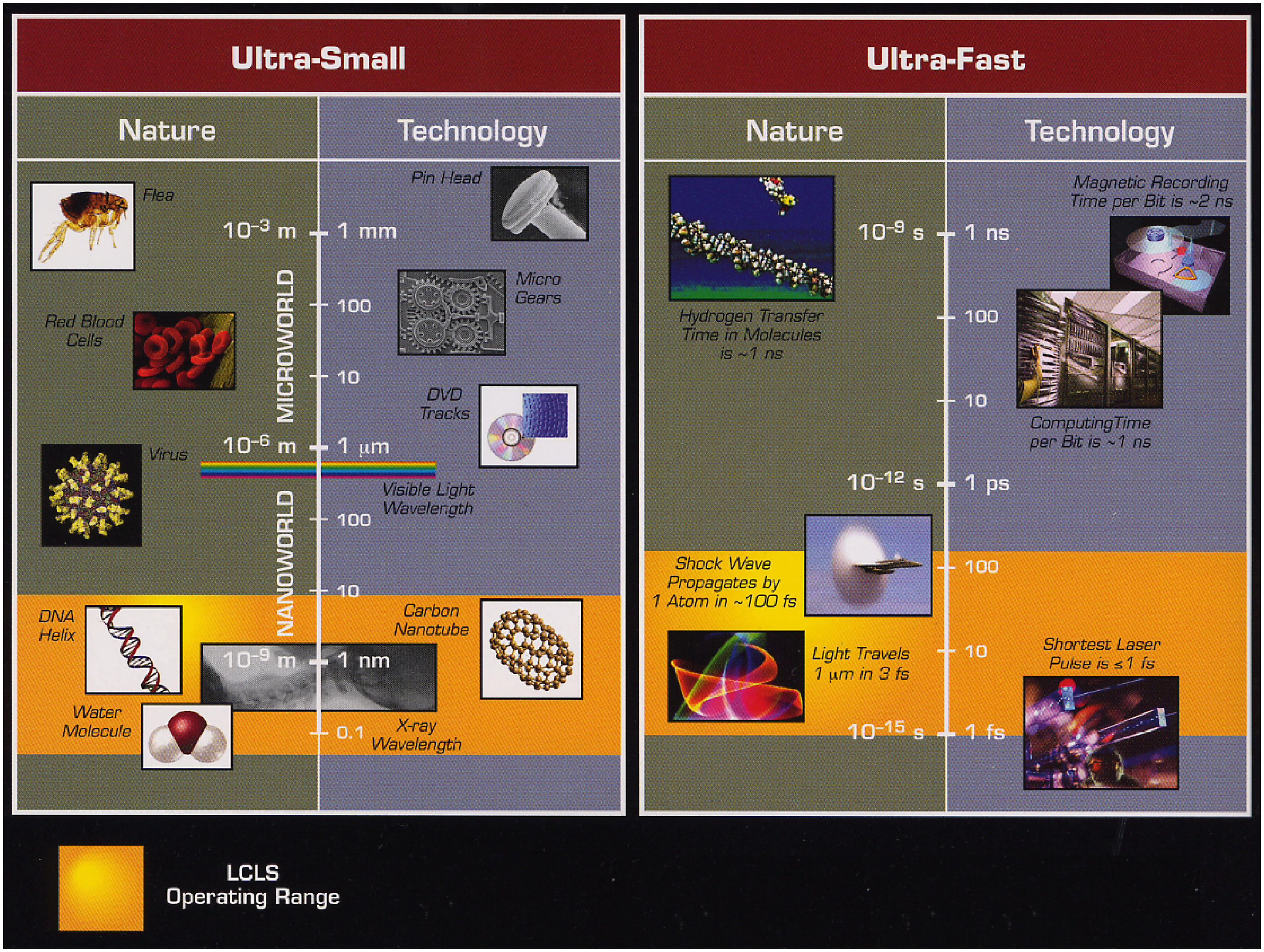
Argonne National Laboratory

Office of Science
U.S. Department of Energy

A U.S. Department of Energy
Office of Science Laboratory
Operated by The University of Chicago

Ultrafast Sources and Science:




Ultra-Small
Nature
Technology
Flea
10^{-3} m
1 mm
Pin Head
MICROWORLD
100
Micro Gears
Red Blood Cells
10
DVD Tracks
10^{-6} m
1 μm
Virus
Visible Light Wavelength
NANOWORLD
100
10
Carbon Nanotube
DNA Helix
10^{-9} m
1 nm
Water Molecule
0.1
X-ray Wavelength
Ultra-Fast
Nature
Technology
Magnetic Recording Time per Bit is ~2 ns
10^{-9} s
1 ns
Hydrogen Transfer Time in Molecules is ~1 ns
100
10
ComputingTime per Bit is ~1 ns
10^{-12} s
1 ps
Shock Wave Propagates by 1 Atom in ~100 fs
100
Light Travels 1 μm in 3 fs
10
Shortest Laser Pulse is ≤1 fs
10^{-15} s
1 fs
LCLS Operating Range

Time Domain Science at 1ps and Longer

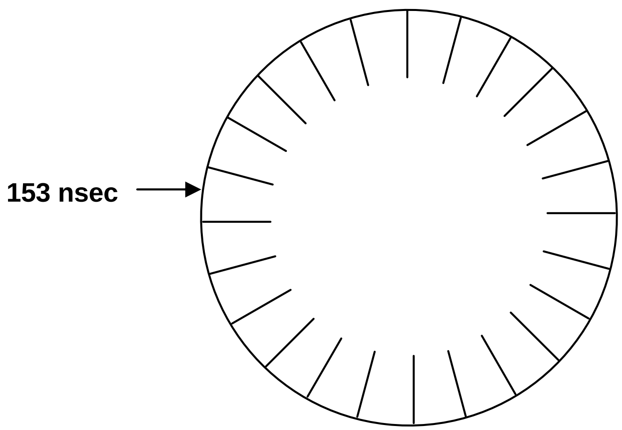
- **Spectroscopy and real-time measurement are complementary**
  - Just like diffraction and imaging
- **Spectroscopy works well below time scales of fs-ps**
  - one fs ~ eV; ps ~ meV
- **The time domain is the only way to access information on time scales much longer than 1 ps**

Scientific impact areas of APS work (that uses the time structure of the beam)

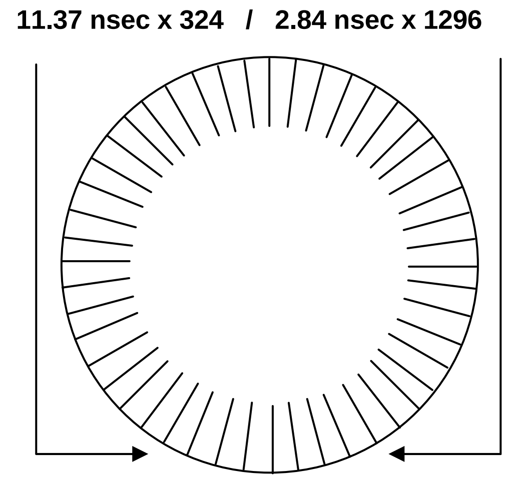
- **Nuclear Resonant Scattering**
  - Vibrational dynamics (geophysics, biophysics)
  - Local magnetism studies (materials)
- **Pump-probe experiments**
  - Atomic physics
  - Transient structures in chemical and biochemical reactions
  - Non-thermal materials behavior

APS Storage Ring Fill Patterns

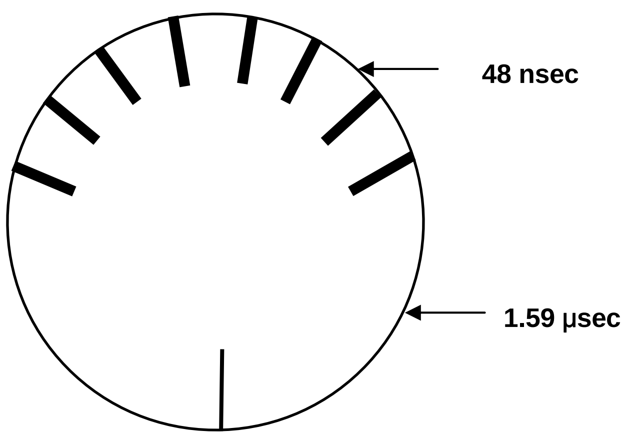
Singlet 24



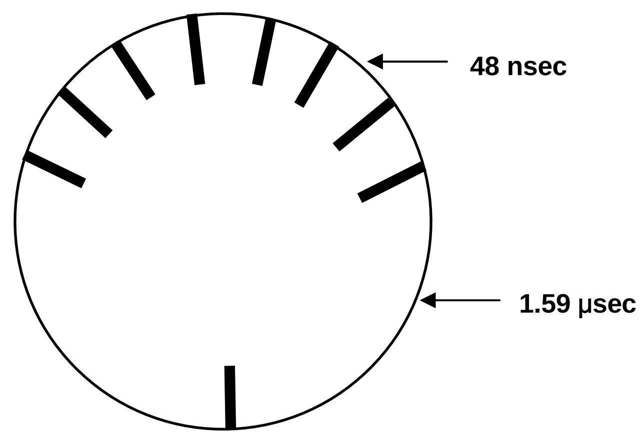
Multi bunch 324 / 1296



Hybrid with singlet 1 + 8*7



Hybrid with triplet 3 + 8*7

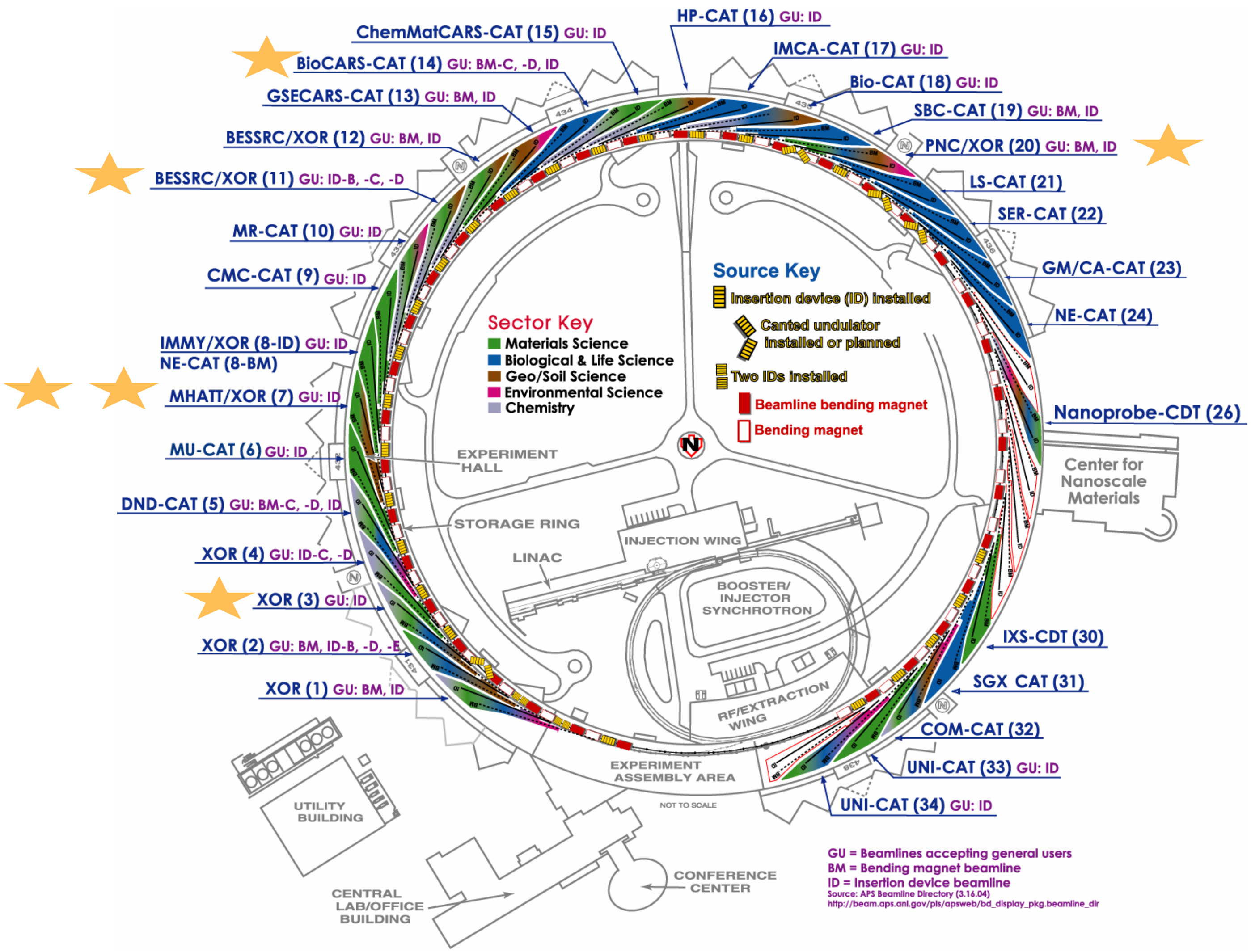


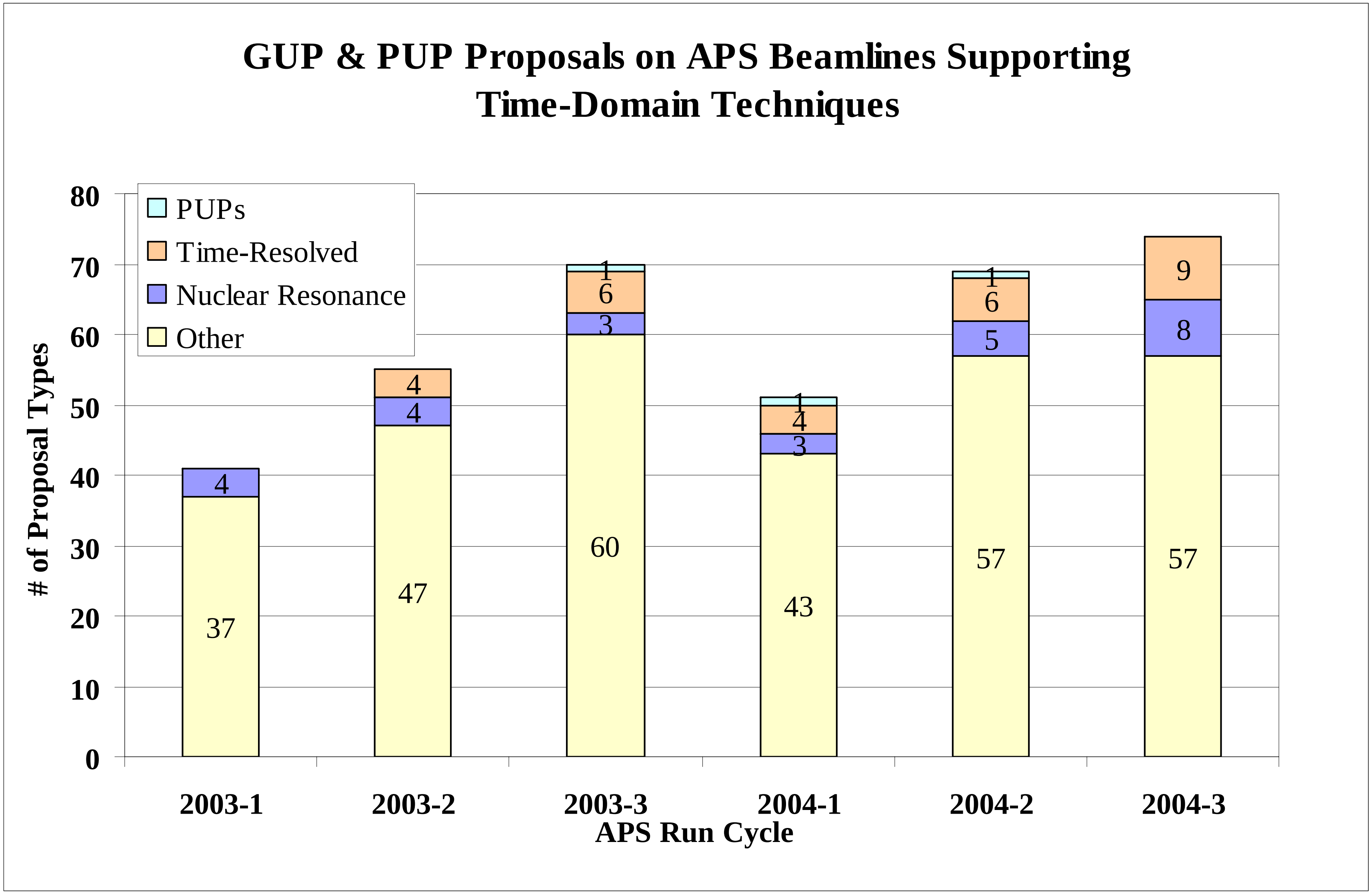
OPERATING MODE HISTORY
(# OPERATION SHIFTS)

	FY01	FY02	FY03	FY04	FY05
NON TOP-UP:					
Higher Emittance	429	90	69	0	0
Hybrid (triplet) 3 + (8x7)	39	75	0	0	0
Low Emittance (324 bunch)	0	0	72	168	111
Low Emittance (1296 bunch)	0	0	0	0	32
TOP-UP:					
Hybrid (singlet) 1 + (8x7)	45	36	63	84	86
23 or 24 Bunch	96	420	423	399	396
TOTAL	**609**	**624**	**627**	**651**	**625**

- As of FY04 all modes are low emittance.
- 24 bunch replaced 23 bunch top-up mode on 10/15/03.

Science that uses the time structure of the beam





GUP & PUP Proposals on APS Beamlines Supporting
Time-Domain Techniques
PUPs
Time-Resolved
Nuclear Resonance
Other
# of Proposal Types
80
70
60
50
40
30
20
10
0
37
4
47
4
4
60
3
6
1
43
3
4
1
57
5
6
1
57
8
9
2003-1
2003-2
2003-3
2004-1
2004-2
2004-3
APS Run Cycle

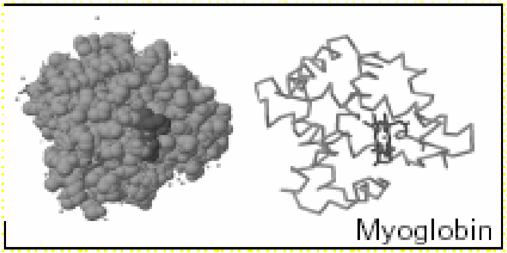
Nuclear Resonant Scattering Studies

- Sector 3 (XOR), sector 16 (HPCAT)
- Applications in
  - Geophysics (lattice dynamics under extreme conditions)
  - Biophysics (local vibrational dynamics in proteins)
  - Materials science (local magnetism in nanostructures)
- Timing requirements
  - Sufficiently separated bunches (>150 ns)
  - High bunch purity (no spurious bunches)

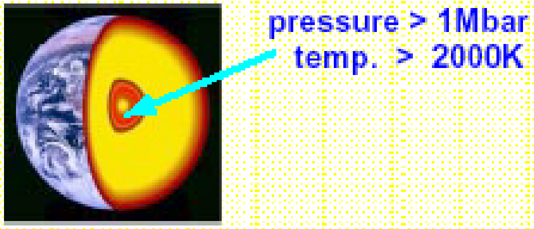
NRS Studies (Ercan Alp)

<u>target applications:</u>

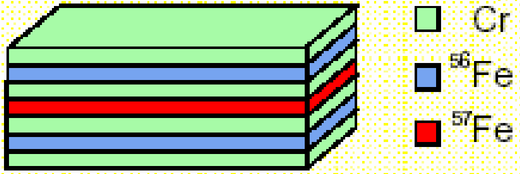
- perfect isotope selectivity & complete suppression of nonresonant signals
- excellent sensitivity (10^{12} nuclei in the focused beam)



☆ proteins and other large molecules



☆ materials under high-pressure



☆ nanostructures

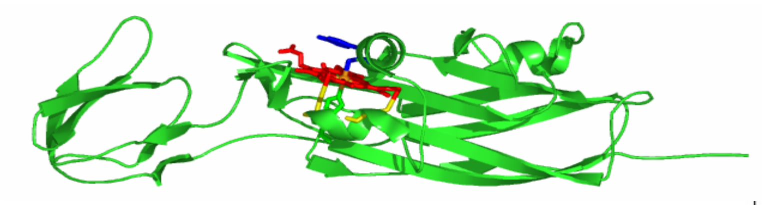
☆ disordered and amorphous materials

NRS Studies (Ercan Alp)

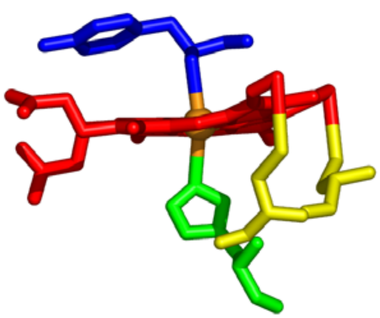
Vibrational dynamics in proteins

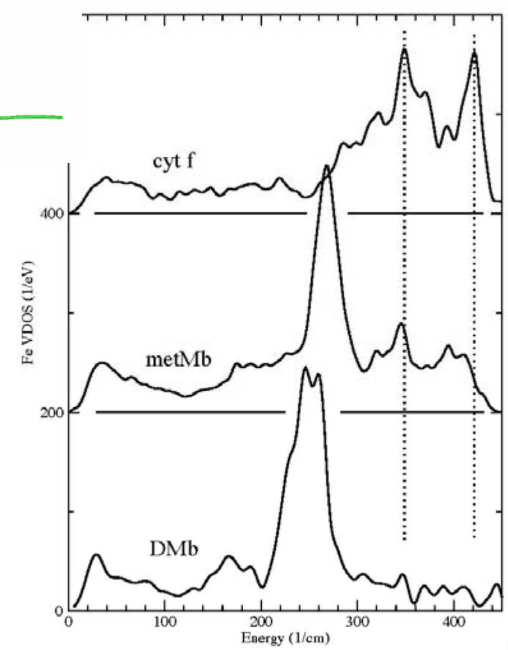
An electron-transfer membrane protein,part of the cytochrome b_6f complex of oxygenic photosynthesis

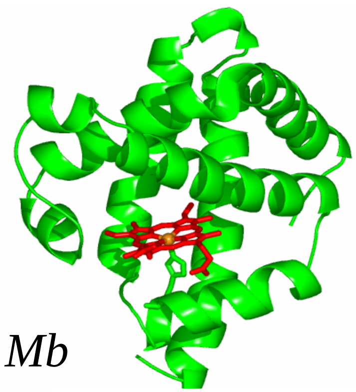
Myoglobin, an oxygen ligand-binding protein, found in muscle tissues, etc.



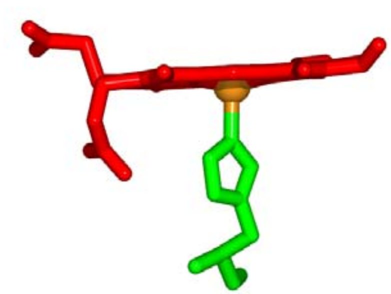
Cytochrome f





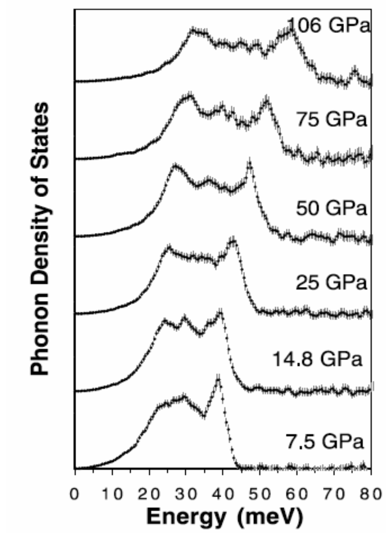


Mb

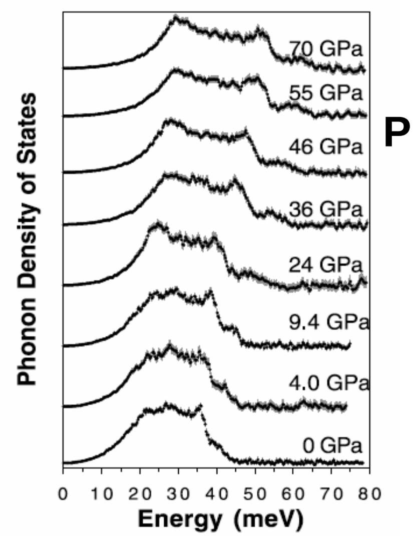


NRS Studies (Ercan Alp)

$Fe_{0.92}Ni_{0.08}$

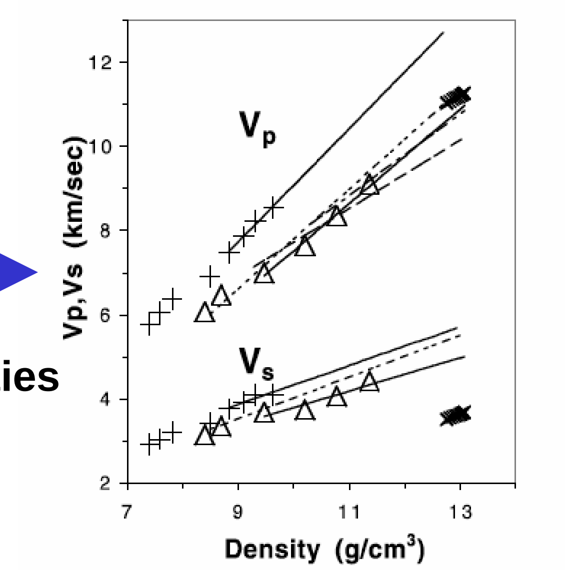


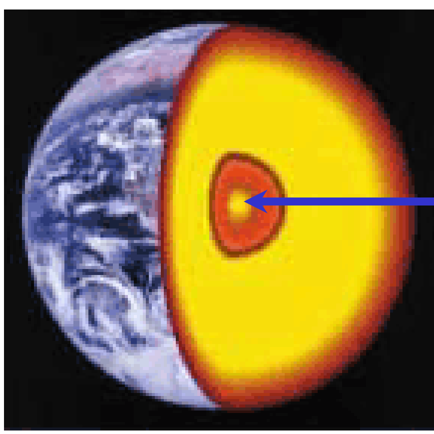
$Fe_{0.85}Si_{0.15}$



Phonon DOS

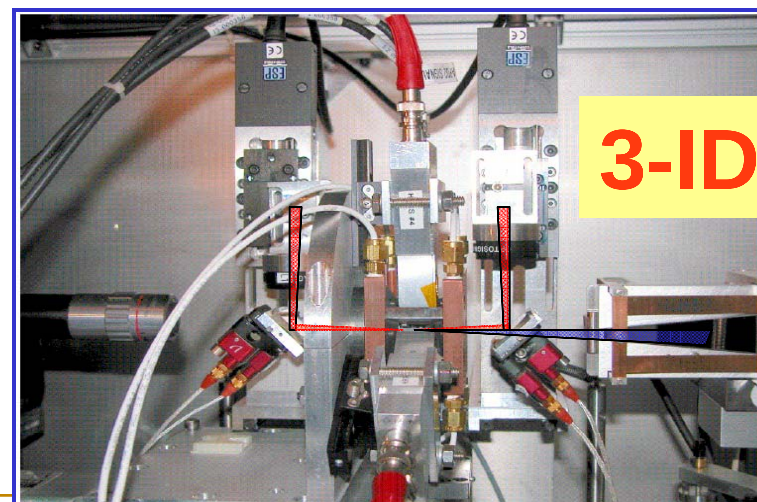
Sound velocities





Extreme Conditions

364 GPa
~ 6000 K



3-ID-B

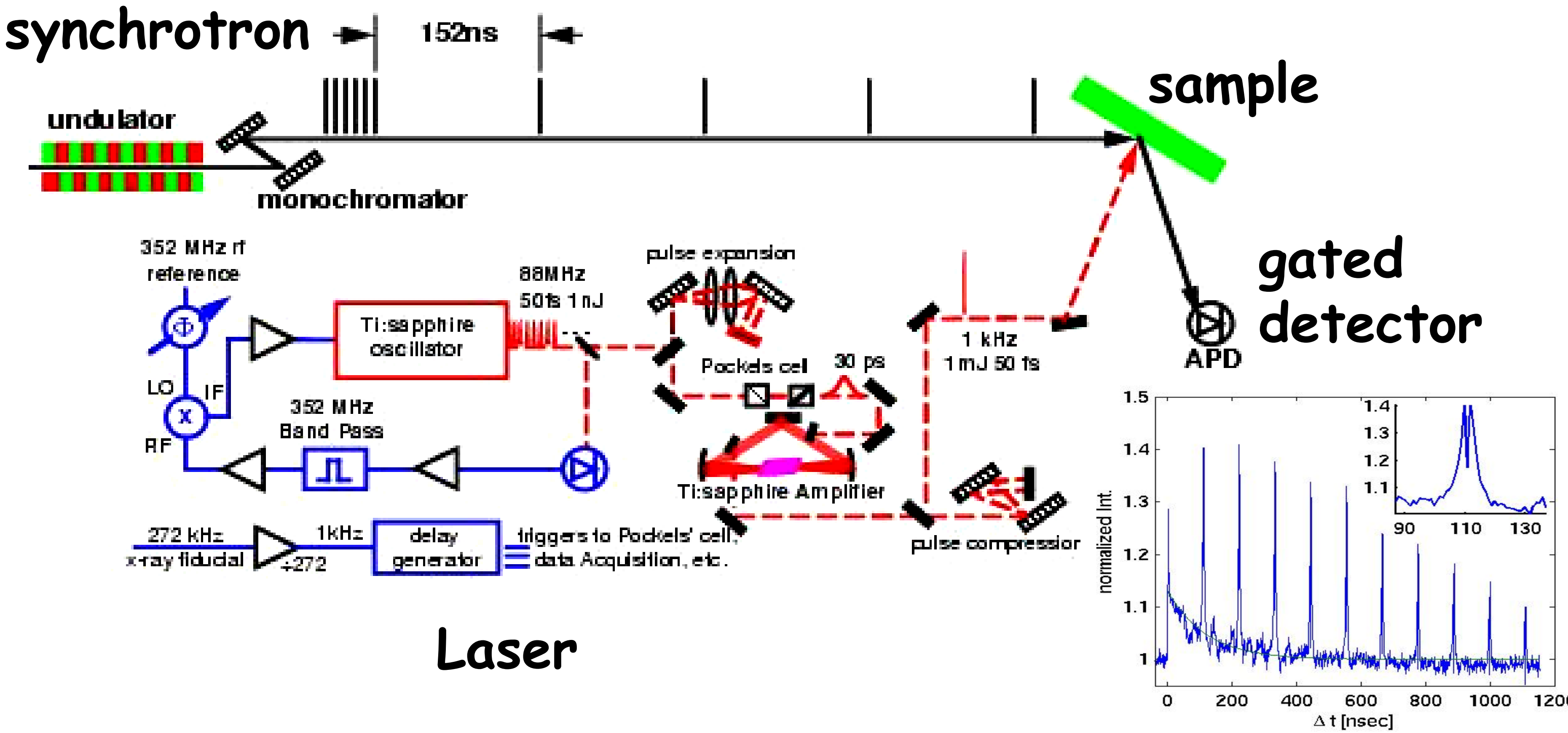
Laser-pump–X-ray-probe Studies

- **Basic phenomena in high EM fields**
- **Rapid non-thermal heat transfer processes**
- **Transient structures in chemical and biochemical reactions**
- **Chemical excited states**
- **Understanding technological applications**
  - Non-thermal drilling
  - Sub-ps materials processing
  - Isotope separation

Laser facilities at the APS

- Sector 7
  - Fs pulse width
  - Spectroscopy with sub-ps time-resolution
  - Using special timing modes
- Sector 11
  - ps pulse width
  - Spectroscopy with ps - µs time-resolution
  - Using special timing modes
- Sector 14
  - ns, ps pulse width
  - Time-resolved crystallography with ns - ms time-resolution
  - Using a chopper to gate the x-ray pulses
- Sector 20
  - Sub-ps pulse width, high-rep, low peak power
  - Spectroscopy with > 100 ps
- ……….

Non-thermal heat transfer (David Reis)

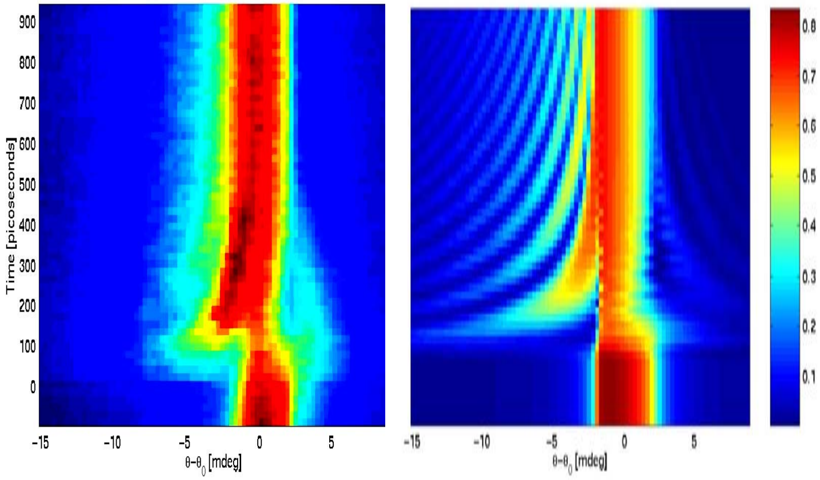


FOCUS

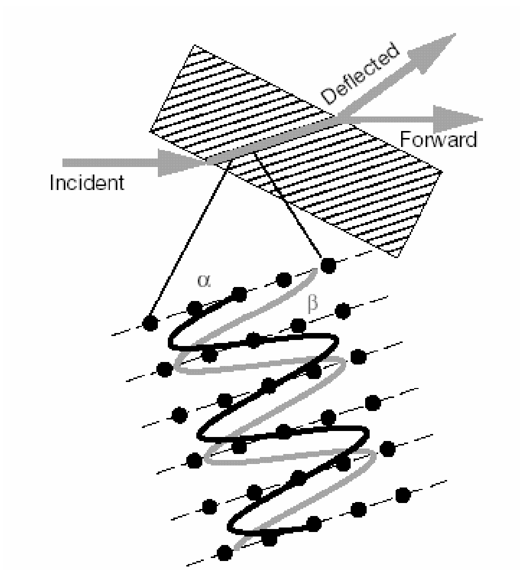
MHATT-CAT

Non-thermal heat transfer (David Reis)

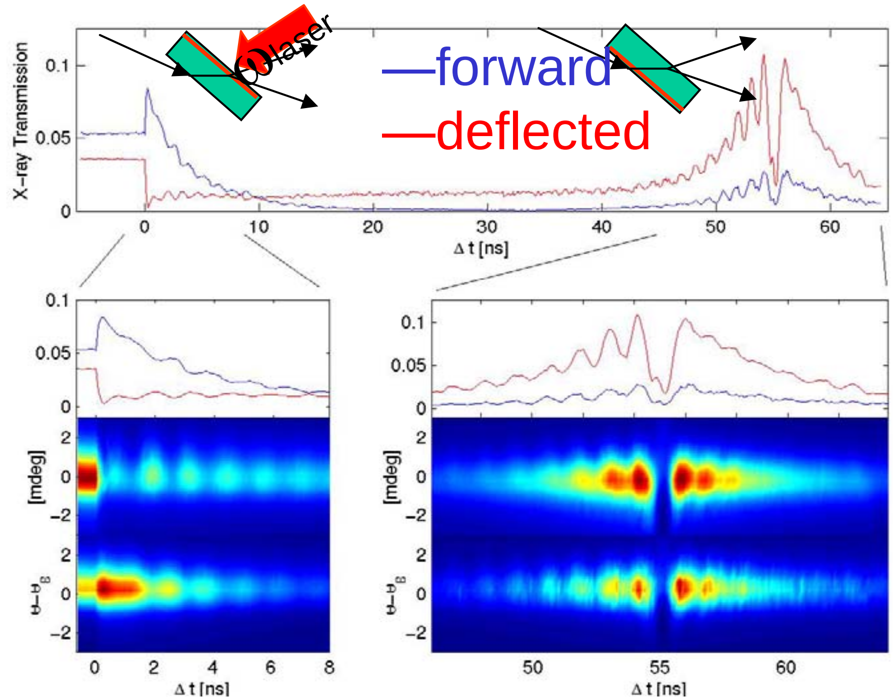
Coherent Acoustic Phonons



D.A. Reis, *et al*, Phys. Rev. Lett. 86, 3072–3075, 2001.



Coherent Control of Pulsed X-ray Beams



M.F. DeCamp *et al.*, Phys. Rev. Lett. 91, 165502, 2003.

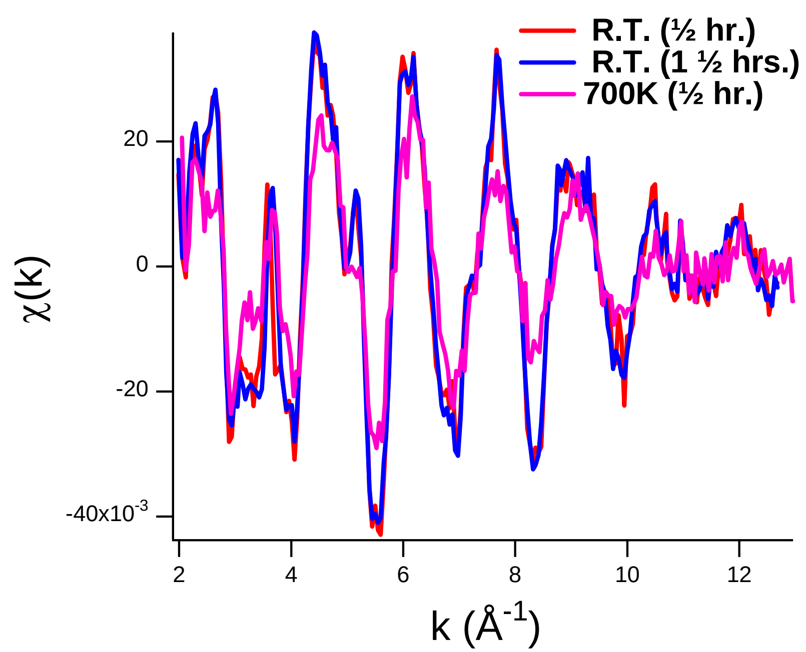
FOCUS

MHATT-CAT

Time-resolved XAFS (Steve Heald)

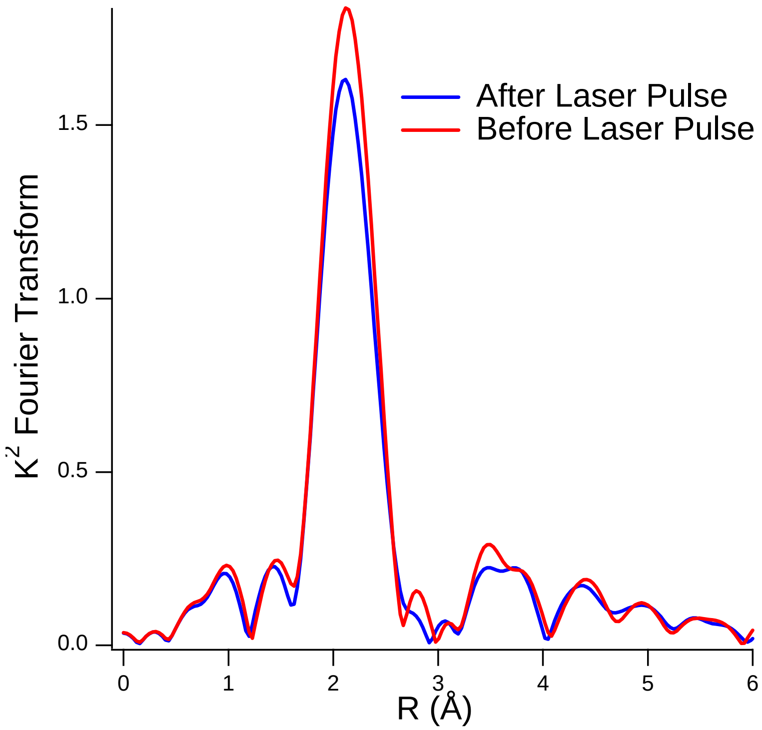
- High-rep rate laser (272 kHz) allows use of all photons from a single bunch
- Micro-focused laser and x-rays provide high flux density
- XAFS useful for disordered materials (amorphous, liquid, gas)
- Time resolution limited by bunch length

Time-resolved EXAFS from laser-heated 200nm Ge film. Data collection times shown in parentheses.

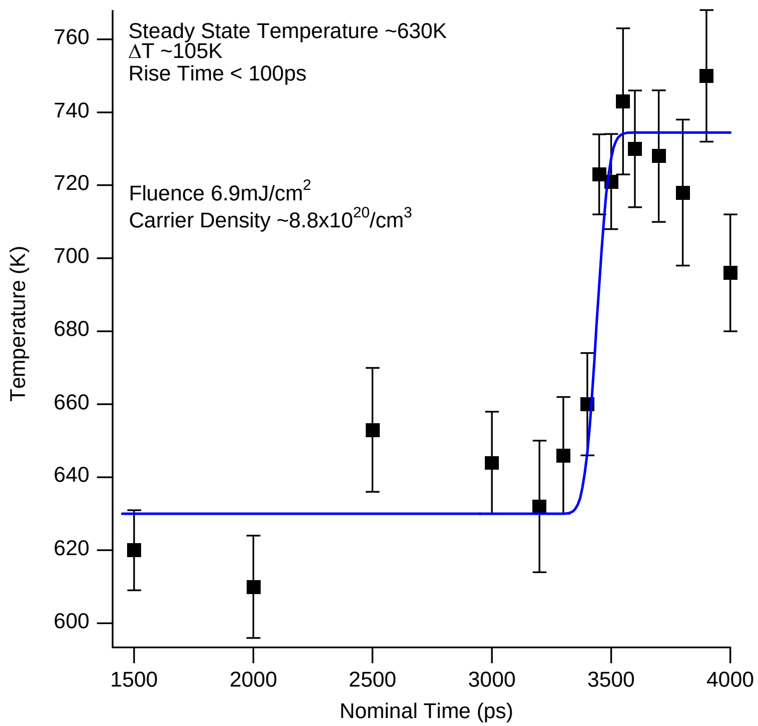


Time-resolved XAFS (Steve Heald)

Fourier transform of EXAFS on Ge at different delay times:



Time dependent temperature:



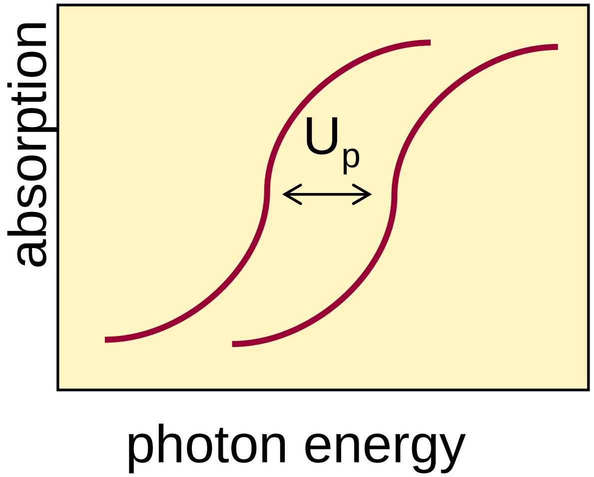
Atoms in strong EM fields (Linda Young)

Pulsed x rays permit us to test theoretical predictions on atoms in the presence of strong laser fields :

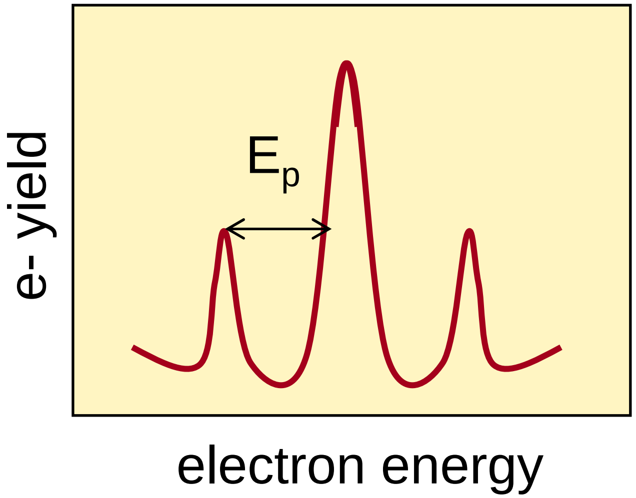
ponderomotive shift in threshold -> absorption spectrum

free-free transitions in continuum -> electron spectra

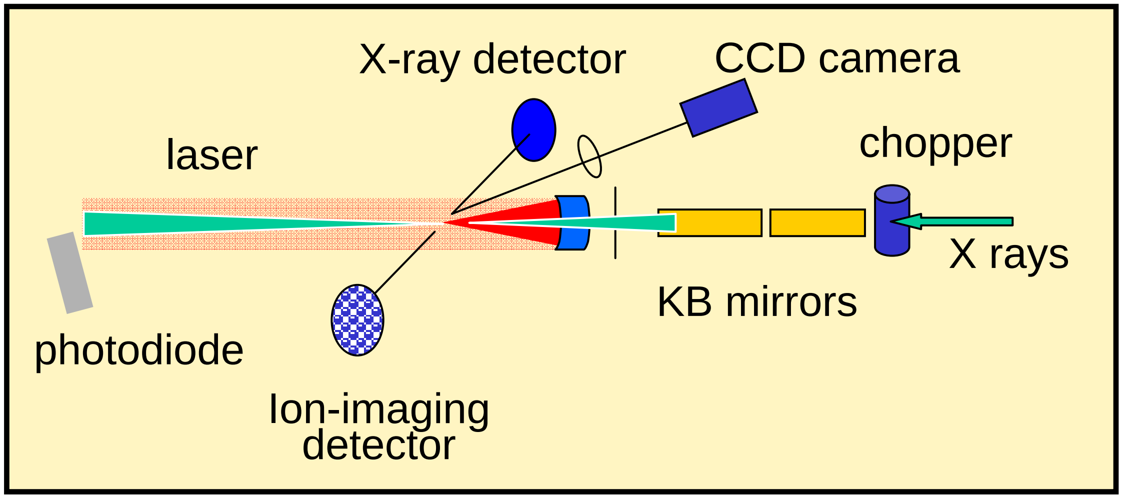
Ponderomotive shift



Electron satellites

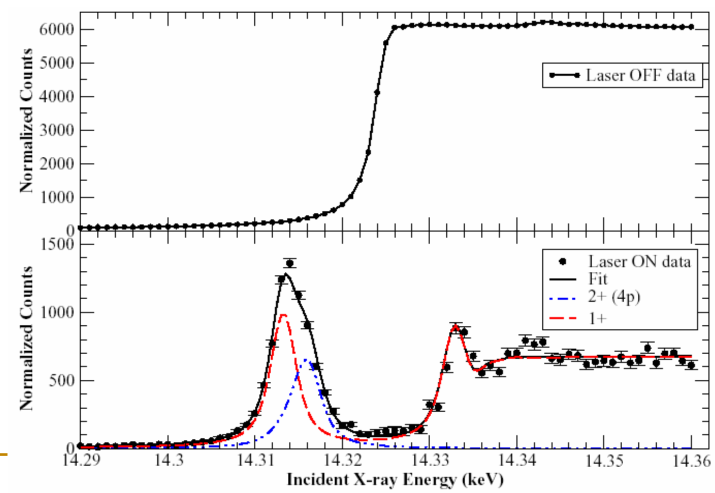


Atoms in strong EM fields (Linda Young)



X-ray parameters:
10^6 x-rays/pulse
10 μm spot size

Laser-modified Kr near-edge absorption

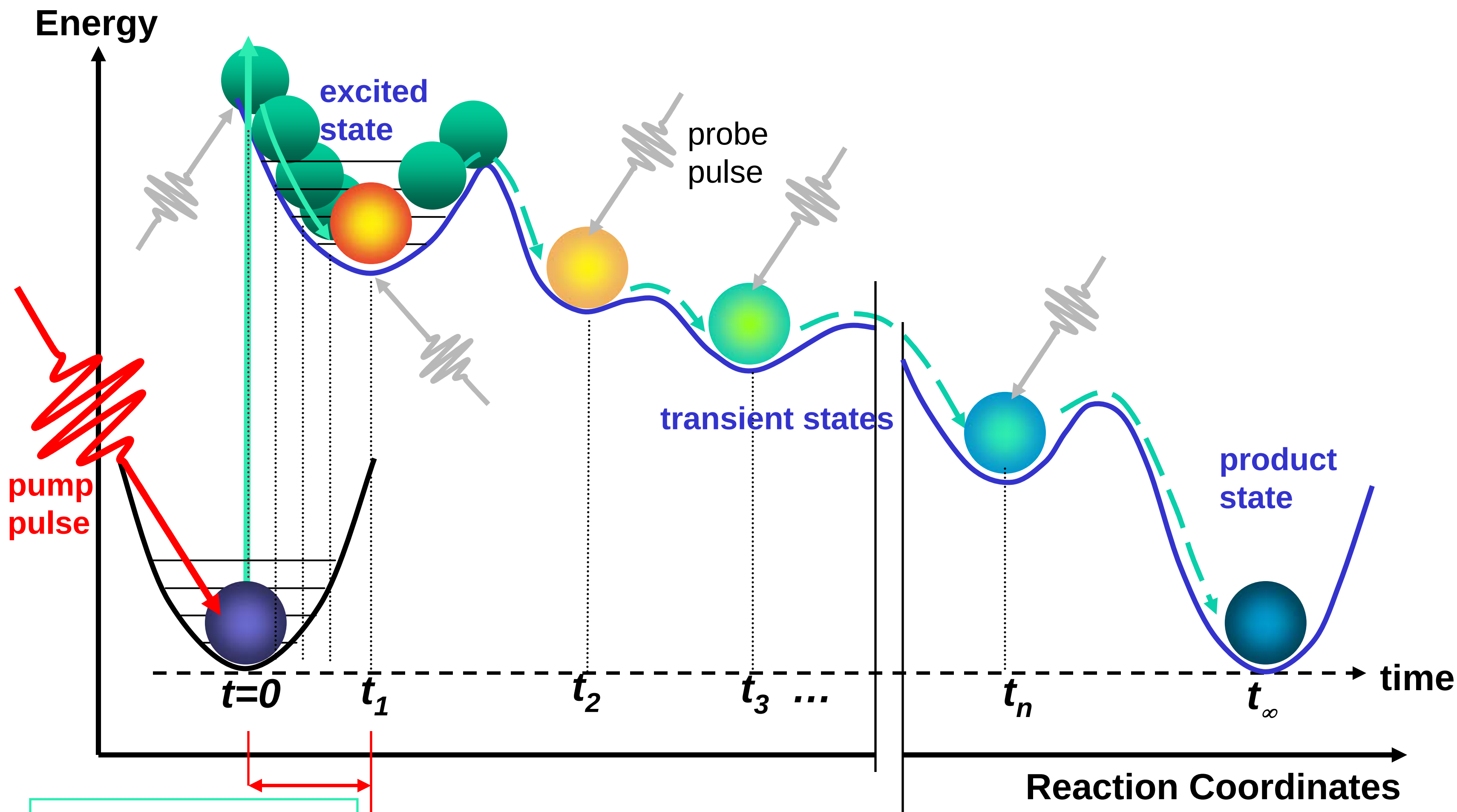


Laser off

Laser on

***Laser parameters**:*
~2 x 10^{14} W/cm^2
800 nm, 1 mJ, 50 fs,
100 μm spot size

Transient Molecular Structures (Lin Chen)

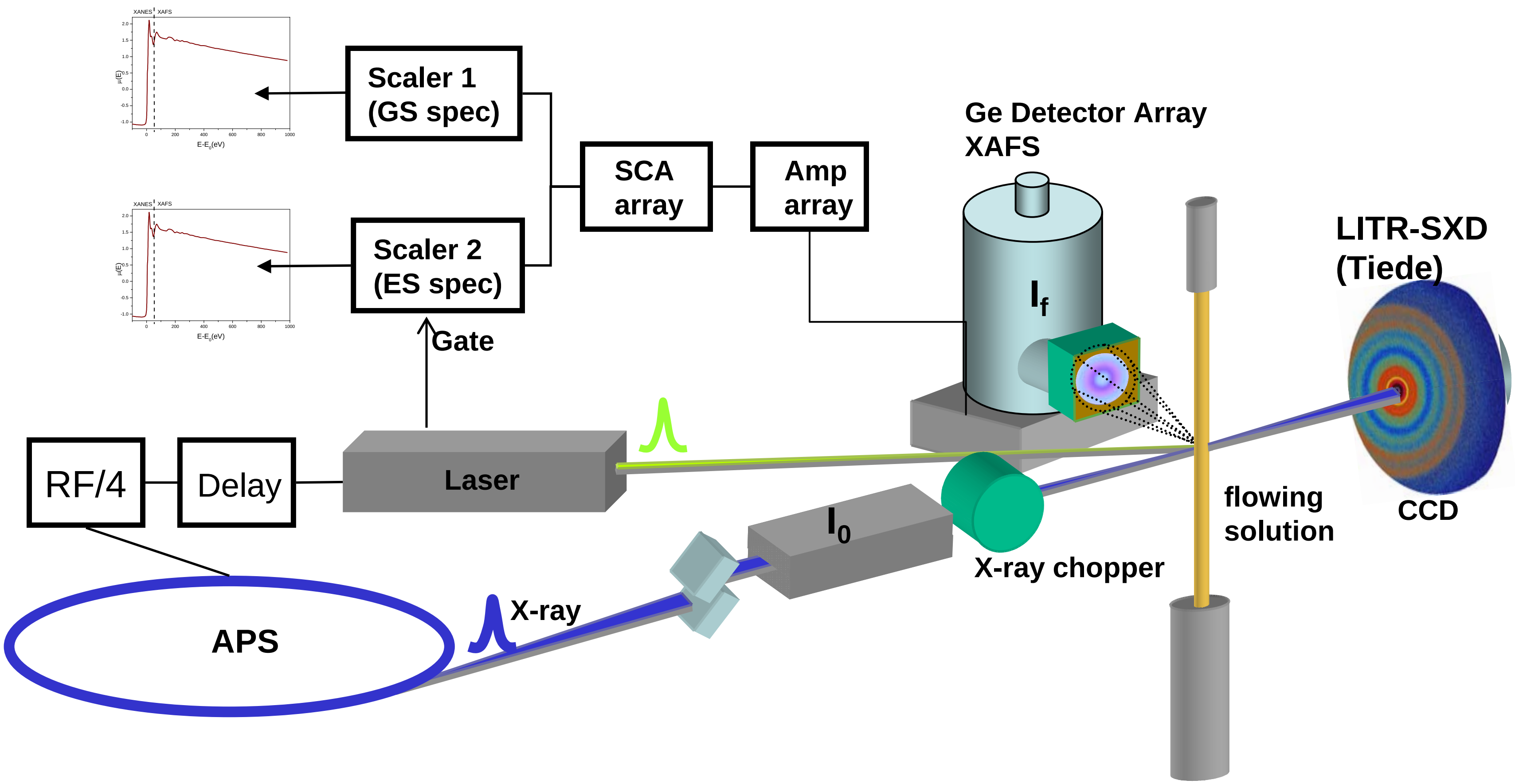


coherent atomic motions on fs to ps time scales → ”molecular movie”

incoherent atomic motions leading to discrete thermally equilibrated transient states on ps to s time scales →”molecular snapshots”

Transient Molecular Structures (Lin Chen)

Experimental Setup for LITR-XAS/WAXS (Beamline 11ID, APS)



Time-Resolved Macromolecular Crystallography (Keith Moffat – BioCARS)

- **All chemical, biochemical and biological reactions involve changes in atomic positions as the reactions proceed; intermediate structures thus differ from those of the static reactants and products. To understand mechanism requires that these intermediate structures, the pathways by which they interconvert and the rates at which they do so be determined experimentally**

- **A brief laser pulse illuminates a stationary crystal of a (light-sensitive) molecule and thus initiates or pumps the reaction. After a suitable time delay t, a single polychromatic synchrotron X-ray pulse (~100 ps) or a pulse train is delivered to the crystal and probes its space-average structure by generating a Laue diffraction pattern. A complete data set then contains |F(hkl, t)| for all unique values of hkl and the entire time range, with suitable redundancy and accuracy. From that data set, the changes in electron density – and hence in structure – with time can be calculated i.e. a "molecular movie"**

Technical Challenges Affecting Experiments that Use the Time Structure of the APS Beam

- Single-bunch current
- Bunch purity
- Bunch length

Mike Borland

- Detectors

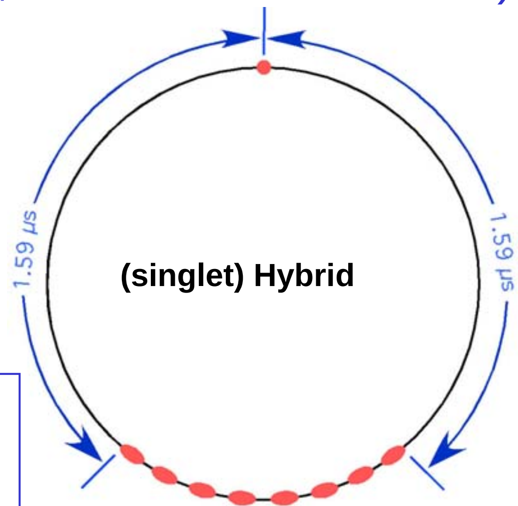
- Choppers

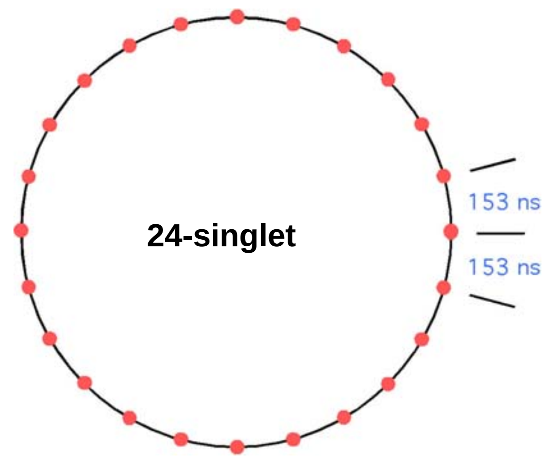
Detectors for timing experiments

- Streak camera
  - Sub-ps resolution possible
  - Low efficiency, no spatial resolution
  - Limited dynamic range
- Avalanche Photodiode (APD)
  - 100 ps resolution demonstrated, 1 ns standard
  - Good efficiency below 15 keV
  - Excellent dynamic range (more than 9 orders of magnitude)
  - Limited spatial resolution
- Integrating detectors
  - High spatial resolution possible
  - Good efficiency at high energies
  - X-ray beam chopper required

Single-Bunch Experiments need X-ray Choppers

- Laser-pump-x-ray-probe experiments with ps-temporal resolution require
  - Ultrafast detectors (x-ray streak camera, gated CCD, diode-based detectors)
  - X-ray choppers
- Select single x-ray pulse from synchrotron pulse train
  - (singlet) hybrid fill (1.59 µs each side)
  - Routine 24-singlet mode (153 ns each side)
  - Phase-lock with the synchrotron RF

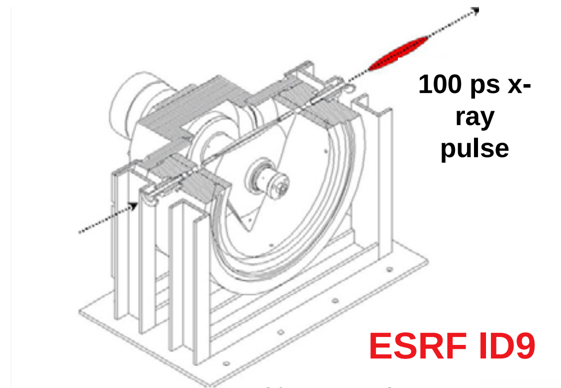




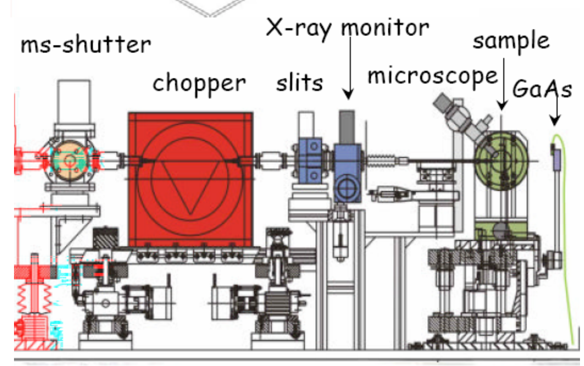
Chopper Types

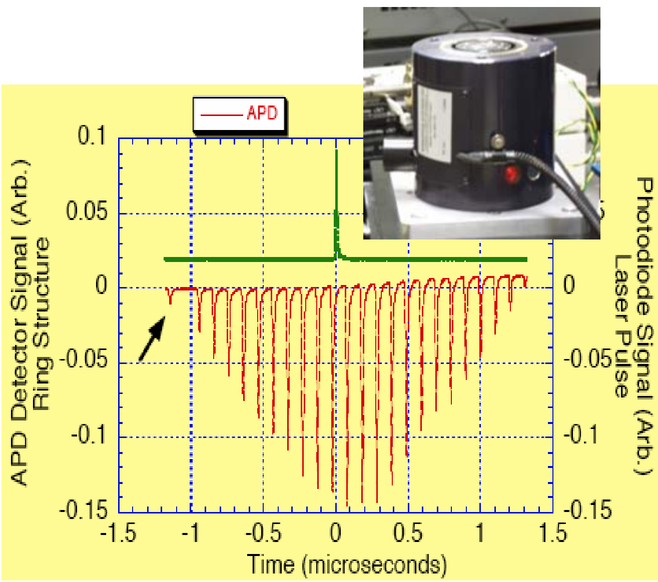
- Mechanically Rotating
  - ESRF/Julich: adjustable time window, white beam
  - APS high speed: fixed window, mono or pink beam
- Mechanical/optical hybrid
  - APS rotating crystal: mono beam, narrow window
- Optical
  - Surface-acoustic-wave-perturbed diffraction
  - Optical phonon induced diffraction (fs-slicing)

Ultrafast Choppers - Mechanical



- **ESRF/Julich rotating chopper**
  - In-vacuum triangular Ti rotor with beam tunnel in one side
    - maximum radius 96.8 mm, tunnel length 165 mm
  - Magnetic bearing running from 10 to 900 Hz
  - Mono and white beam compatible to 40 keV
  - Opening window from 100 ns to 170 µs with trapezoidal slits
  - Focusing mirror required for sub-µs operation
    - Capable of both singlet and singlet-hybrid patterns
  - White beam crystallography and scattering

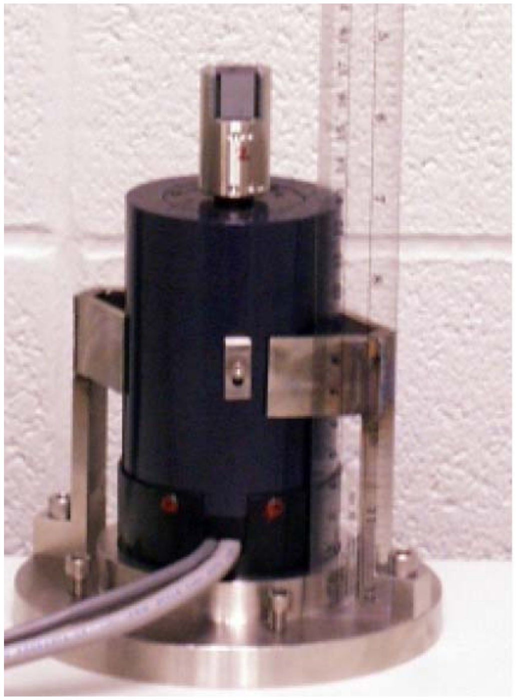


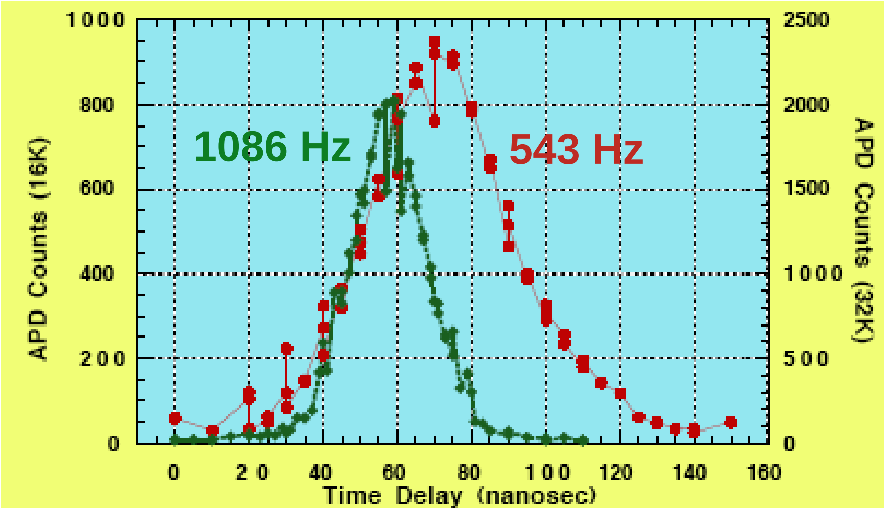


- **APS Rotating Chopper**
  - In-He disk rotor (R=25 mm) with 0.5x2.29 mm^2 slot
  - Air bearing running at fixed 1331 Hz (< 3 ns jitter)
  - 2.45 µs opening for (singlet) hybrid fill pattern only
  - Mono and white beam compatible to 30 keV
  - Very compact, portable, and in-expensive

Ultrafast Choppers – Hybrid

- **APS rotating crystal beam chopper - Fastest chopper so far!**
  - In-air Si(111) crystal cube mounted on a fast rotor
  - Fixed frequencies at 271.5, 543 and 1086 Hz (< 3 ns jitter)
  - Opening window 24 ns at the highest rotation speed
  - Opening window determined by crystal rocking curve width
  - Optimal condition being nondispersive double-bounce geometry
  - Only monobeam compatible
  - Not effective with beams focused in the diffraction plane
  - Not practical for experiments involving energy scan
  - Very compact, portable and inexpensive

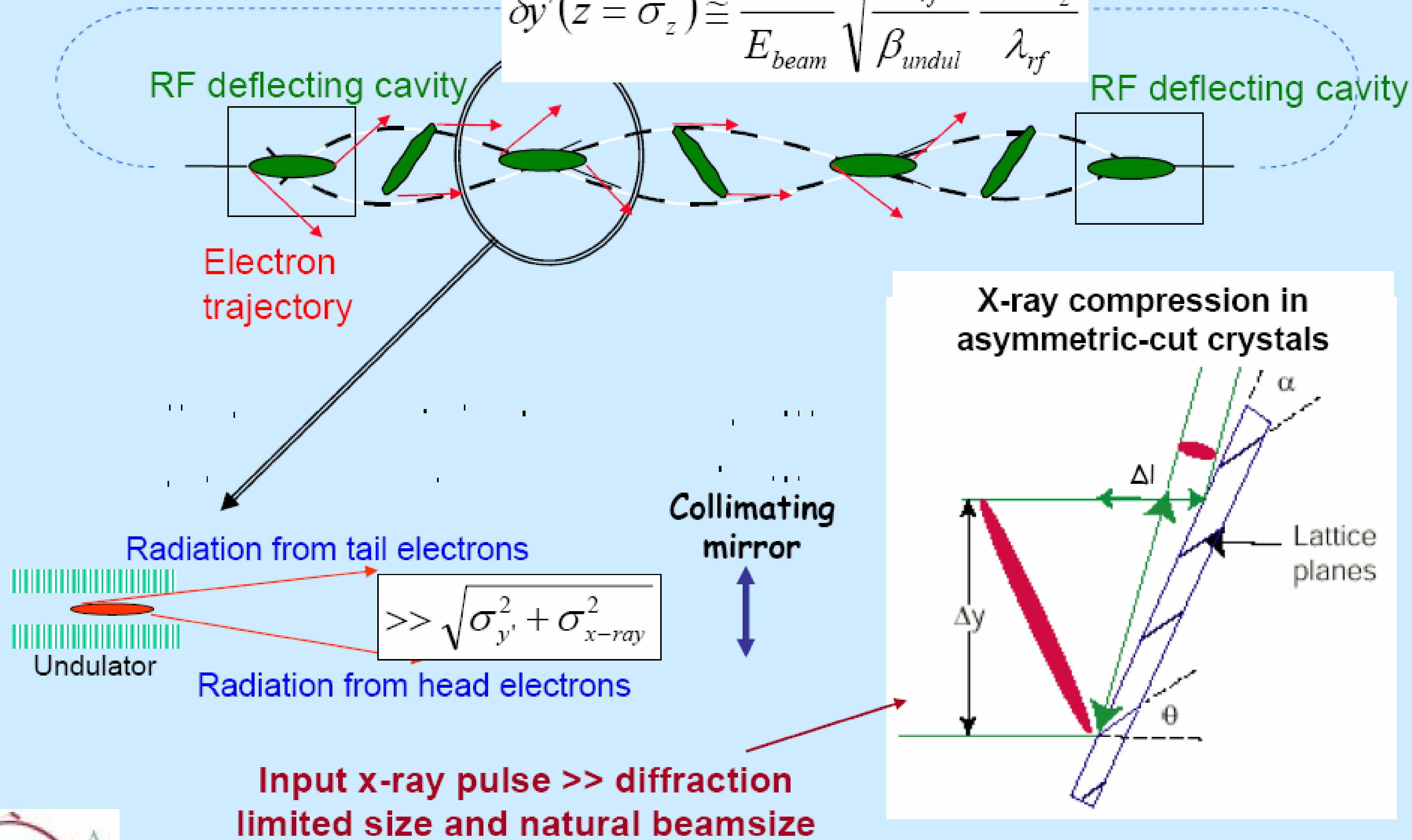




- **Optical chopper based on transient Bragg scattering**
  - Potentially very fast
  - Practicality yet to be demonstrated because of switch contrast

Obtaining short x-ray pulse from a “long” electron bunch

$$\delta y'(z=\sigma_z) \cong \frac{eU}{E_{beam}} \sqrt{\frac{\beta_{rf}}{\beta_{undul}}} \frac{2\pi\sigma_z}{\lambda_{rf}}$$



Zholents, et al., NIM A 425, pp 385-389 (1999)

Conclusion

- **Exciting areas of science are touched by these experiments which use the time structure of the beam**
- **We seek the review committee's guidance on the impact so far, the opportunities for growing future impact, and the possibility of growth for existing or new user communities**