國家同步輻射研究中心
National Synchrotron Radiation Research Center

時間解析技術應用在真空紫外光科學研究
(Time-resolved technology used in the research of VUV)

Yin-Yu Lee (李英裕)

Experimental Facility Division, Office: S114
NSRRC: Tel (03)5780281 ext. ***7114*** *lab. 3165*
Fax(03)5783813
Email:yylee@nsrrc.org.tw
101 Hsin-Ann Road, Hsinchu Science Park,
Hsinchu 30076, Taiwan

Outline

- Time-resolved study overview
- Types for using SR in time-resolved experiments
- TPS capability & Potential experiments
- Developing of time-resolved facilities in NSRRC

  Required instruments for time-resolved experiments:

  - ---Timing control units- trigger and time delay
  - ---Fast chopper/shutter- bunch gated

Time-resolved experiments

Fundamental parameters in physical world

- **Energy, Momentum, Position, Time**
- Time can be applied to all the techniques, in principle
- Some time-resolved experiments take advantage of the pulsed nature of the synchrotron radiation

Laboratory experiments simply require two pulses: one to "pump" energy into the sample system and a second to “probe” the system's excited state, when viewed sequentially, show us how a given process evolves over time in ultrafast regime.

"ultrafast" regime opens the possibility to explore -

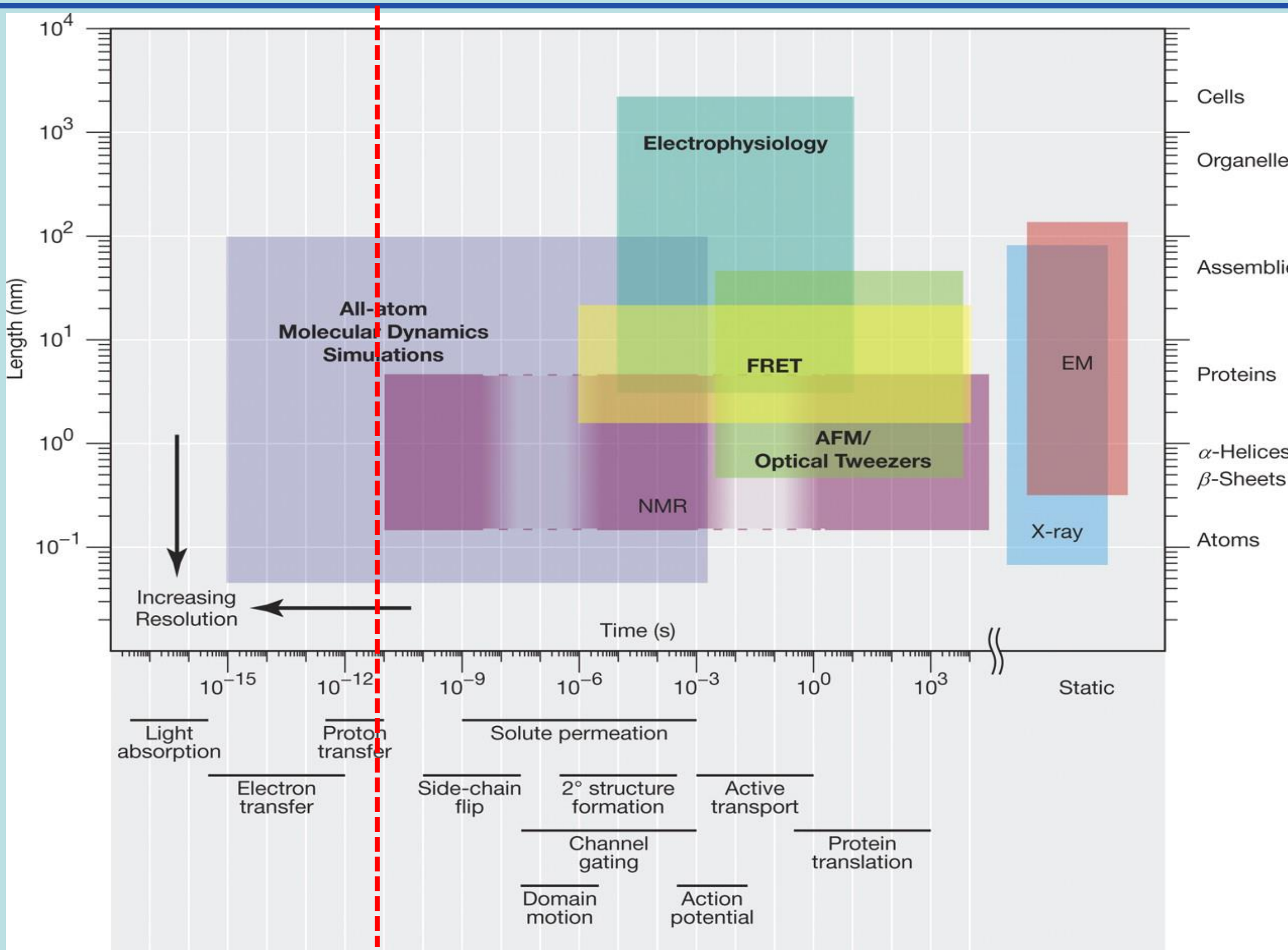
- **chemical bond’s making, breaking, and atomic rearrangement**
- **kinetic pathways of chemical reactions**
- **details of the phase transitions in solids**
- **function and efficiency of biological processes.**

Fundamental ultrafast processes in semiconductors

Microscopic process	Characteristic time(s)
Carrier-carrier scattering	$10^{-15} – 10^{-12}$
Intravalley & intervalley scattering	$\geq 10^{-14} – 10^{-13}$
Carrier-optical phonon thermalization	$\sim 10^{-12}$
Optical phonon-acoustic phonon interaction	$\geq 10^{-11}$
Carrier diffusion (0.1*μm*)	$\sim 10^{-11}$
Auger recombination (carrier density 10^{20} cm^{-1})	$\sim 10^{-10}$
Radiative recombination	$\geq 10^{-9}$
Lattice heat diffusion (1 μm)	$\sim 10^{-8}$

A. Othonos, JAP **83,** 1789-1830 (1998)

Spatial and Temporal Resolution for Phenomena

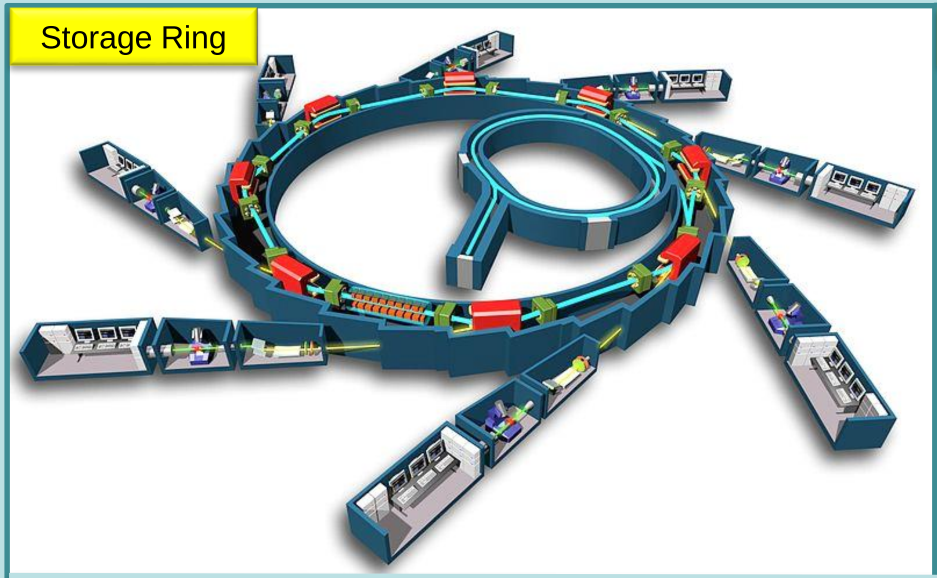


Spatiotemporal resolution of various biophysical techniques. Techniques capable of yielding data on single molecules (as opposed to only on ensembles) are shown in bold. The timescales of some fundamental molecular processes, as well as composite physiological processes, are indicated below the abscissa. The spatial resolution needed to resolve certain objects is shown at right. NRM, nuclear magnetic resonance; AFM, atomic force microscopy; EM, electron microscopy; FRET, Förster resonance energy transfer.

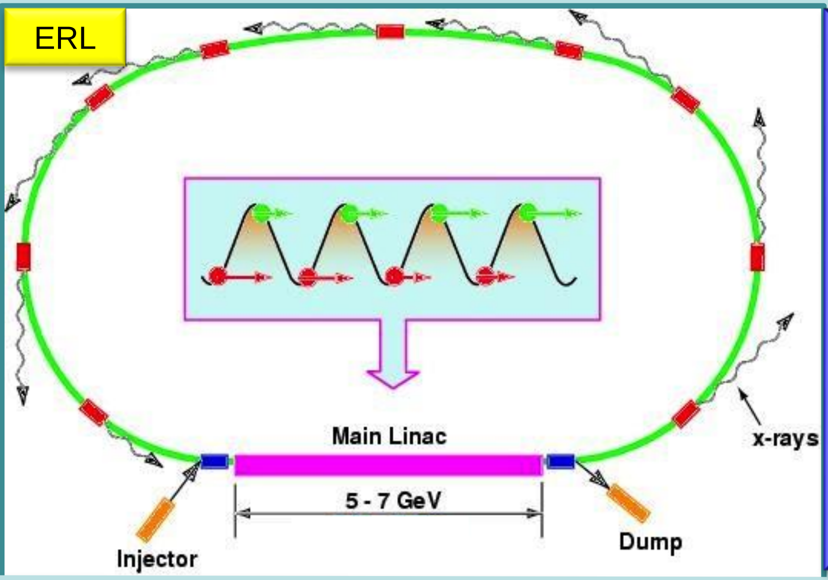
R. O. Dror et al, JG Physiology **135**, 555 (2010)

Light Sources: Storage Ring, ERL, & FEL

Storage Ring

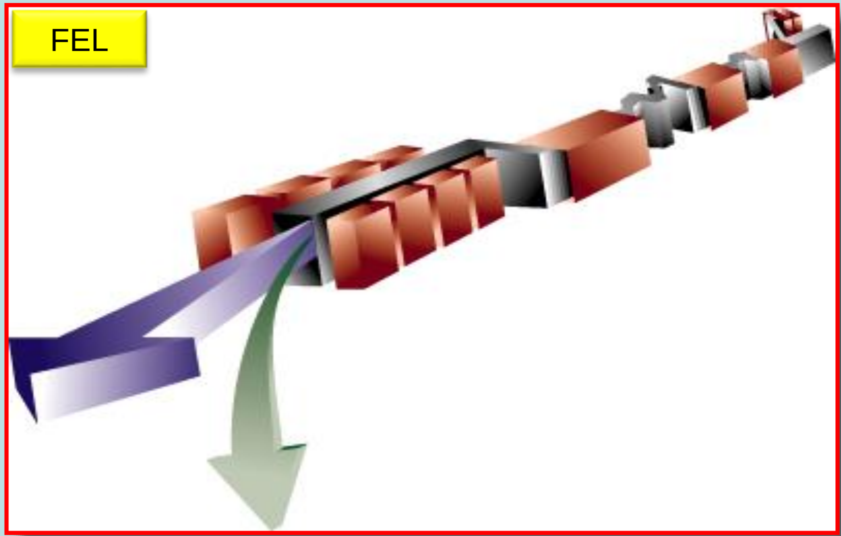


ERL



Spontaneous Emission

FEL

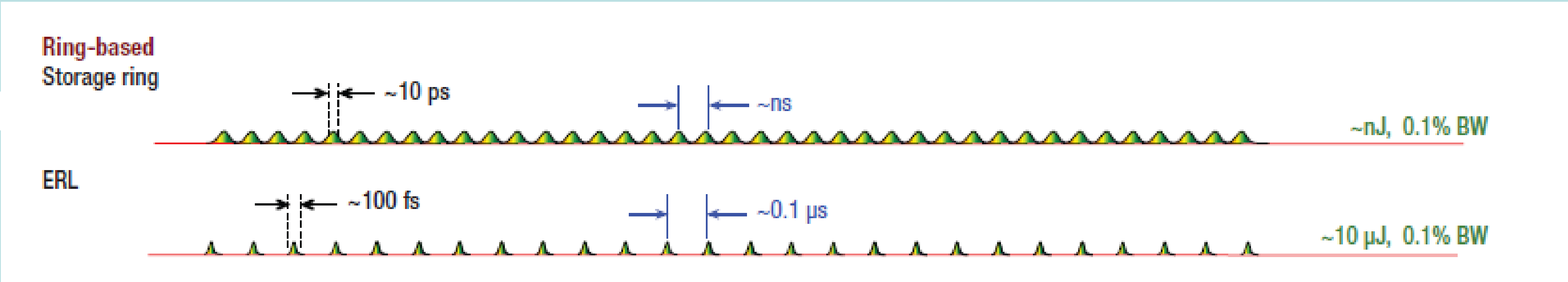


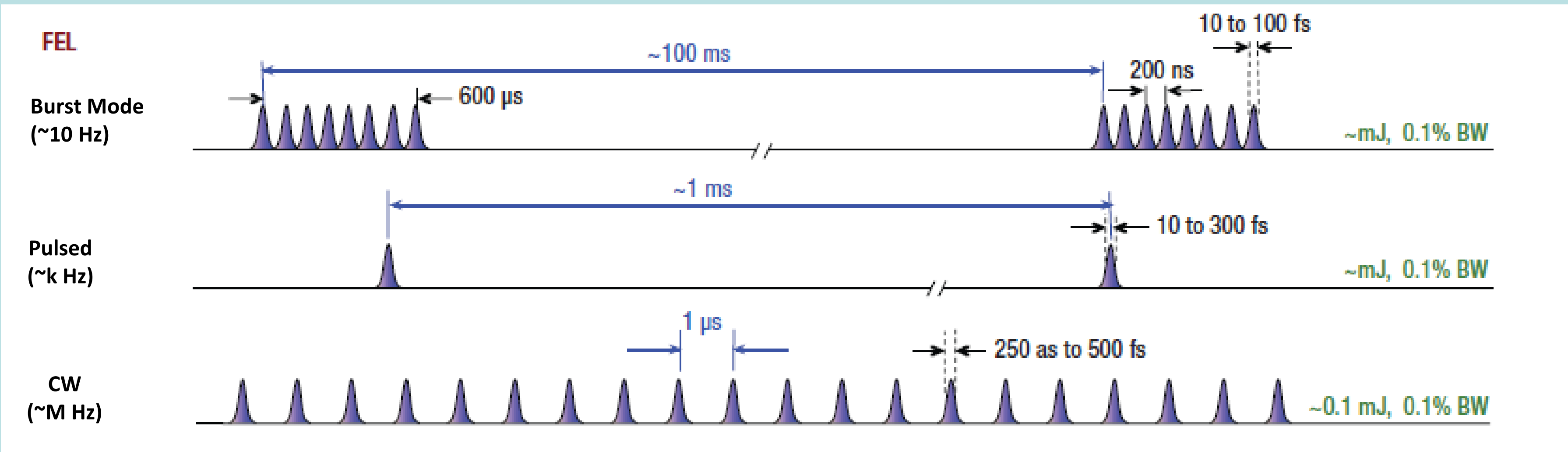
Stimulated Emission

How do Storage Rings and FELs Compare Today?

Parameter		Storage Rings	X-ray FEL
Wavelength Range		2-5+ decades typically	1+ decades (multiple undulators)
Peak Brightness ($ph/s/mr^2/mm^2/0.1\%BW$)		10^{22} – 10^{24}	10^{31} – 10^{33} (10^9 times higher than SR)
Average Brightness ($ph/s/mr^2/mm^2/0.1\%BW$)		10^{19}– 10^{21}	10^{20}– 10^{22}
Minimum Pulse Width (fs)		~10,000	~5
Coherence		Limited transverse spatial coherence	Transverse spatial coherence, limited temporal coherence without seeding
Stability	Energy	<.01% (with ~0.1% energy spread)	0.01-0.03% wo / self seeding
	Position	< 0.1 σ (~10 μm H, ~0.3 μm V)	~0.1 σ
	Time	< 0.1 σ (~1 ps, ~0.2 ps low α)	~100 fs
Number of Beamlines		Large (~30-60)	Limited (6 endstations per undulator)

Light Source Pulse Structures





Types: for storage ring based time-resolved exp.

1. Synchrotron radiation micro-pulse only

- 10s pico-sec time resolution
- Few pico-sec time resolution by low-α mode operation
- Coupling with fast detector and data acquisition always; streak camera, fast scope, or pico-timing analyzer, time gating ….. TAC or TDC based techniques

2. SR + oscillator laser pulse — pump-probe scheme I

- High repetition rate, up to 100 MHz, 10s nJ/laser pulse
- Mode-locked oscillator peak power in kW-MW, 10s pico-10s fs
- Fully use the SR flux, few ps to 10s ns time span

3. SR + amplified laser pulse — pump-probe scheme II

- Lower repetition rate, ~kHz, ~mJ/pulse
- Amplified laser peak power up to GW-TW
- Reduced usage of SR flux in $10^{-3\sim-6}$
- Chopper required for over exposure and avoiding sample damage

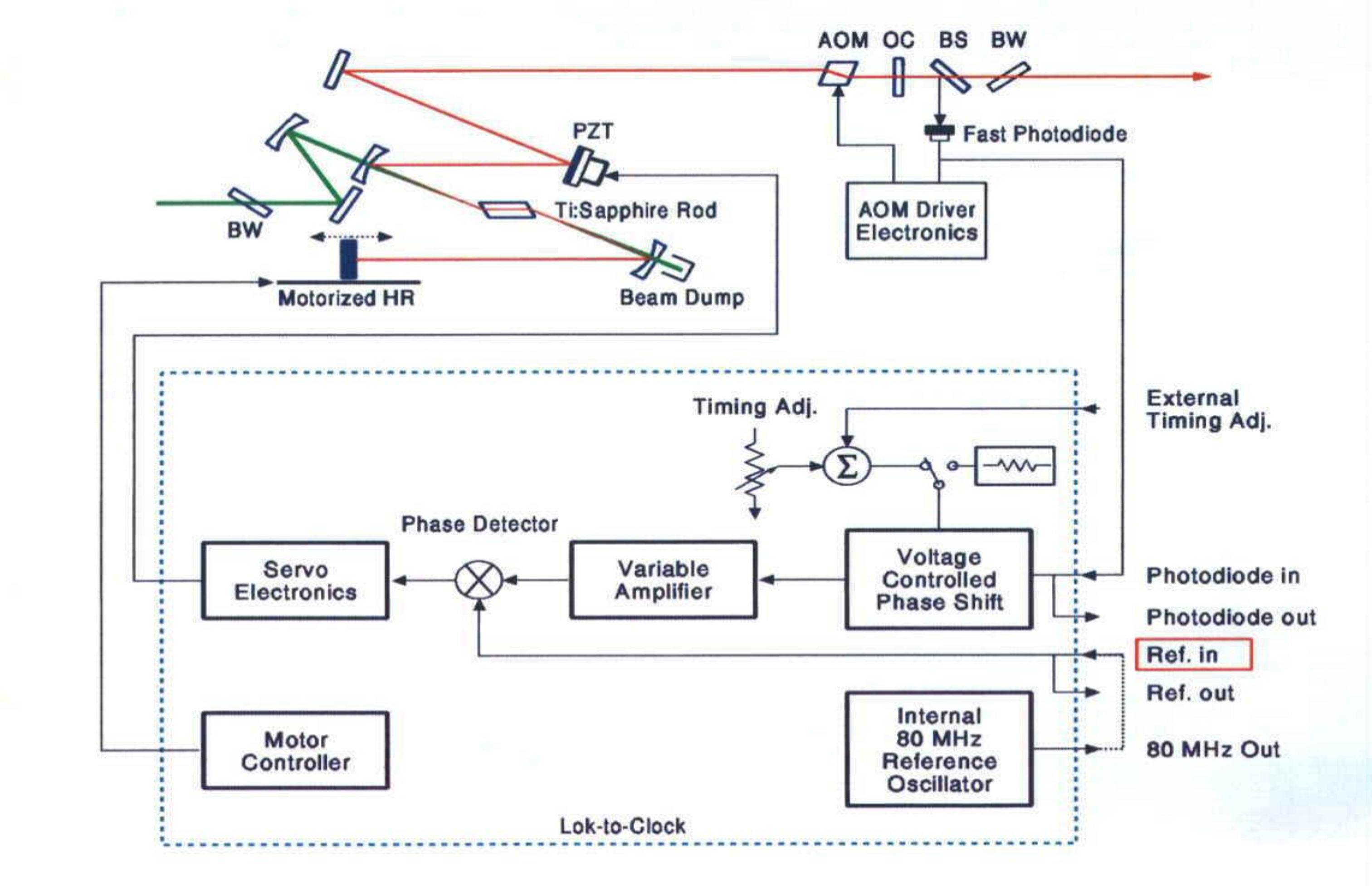
☑ **Time-resolution is limited by the X-ray pulses duration at NSRRC currently.**

1. Temporal width of a synchrotron X-ray pulse: 20-40 ps FWHM
2. Low-a mode: 1-5 ps FWHM
3. Slicing mode: 100 fs FWHM
4. →FEL: sub fs-10s fs FWHM

Synchronization Mechanism: Lock-to-clock

(1) 2.5MHz x 32
(2) 500MHz /25 x 4
⟶ **80 MHz(12.5ns)**

500 MHz /6= 83.33 MHz, 12 ns



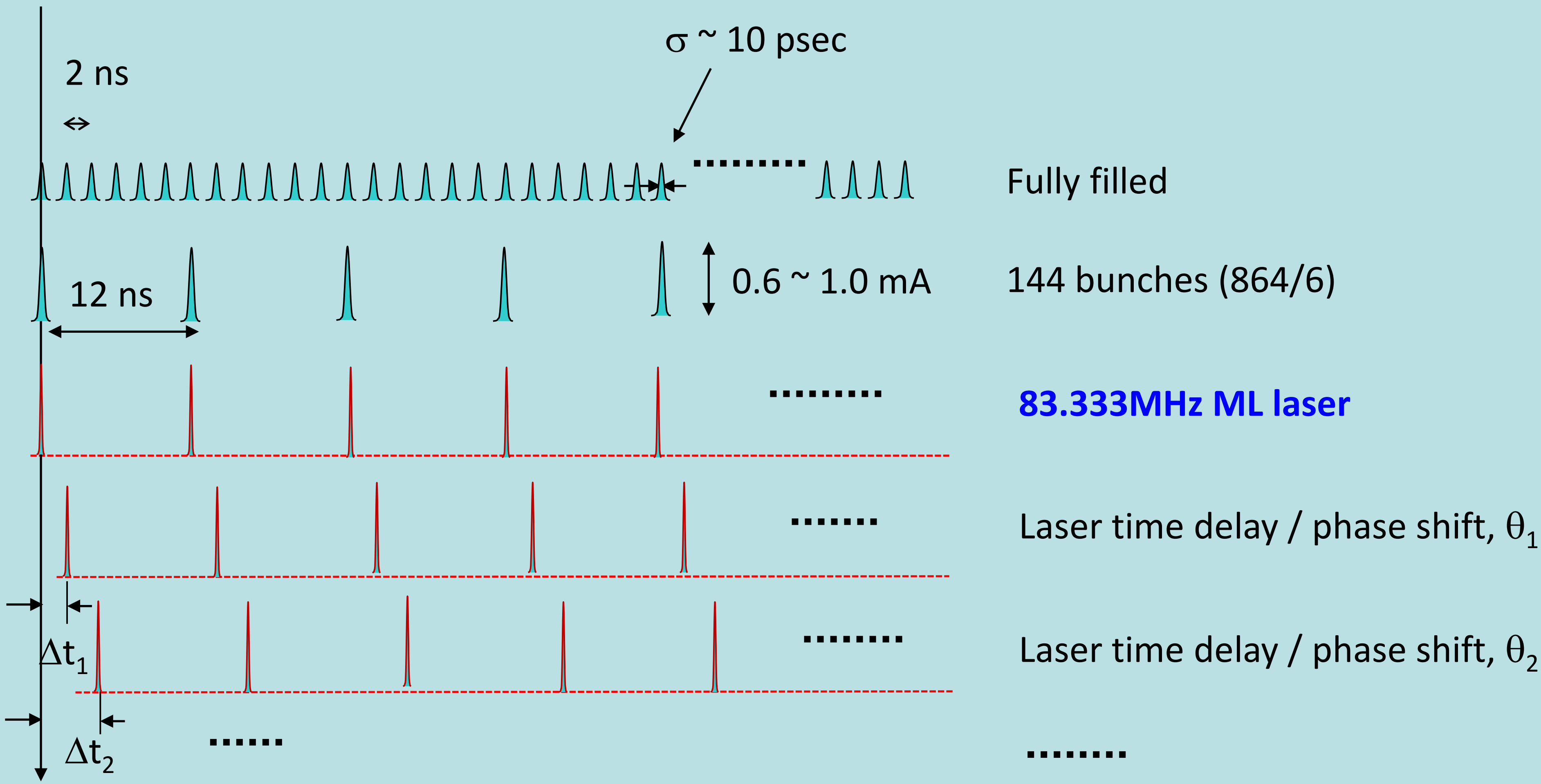
Cavity modes: $\lambda_n = 2L/n$

$\Delta f = c/2L$ ----*repetition rate*

Pump-probe experimental scheme I

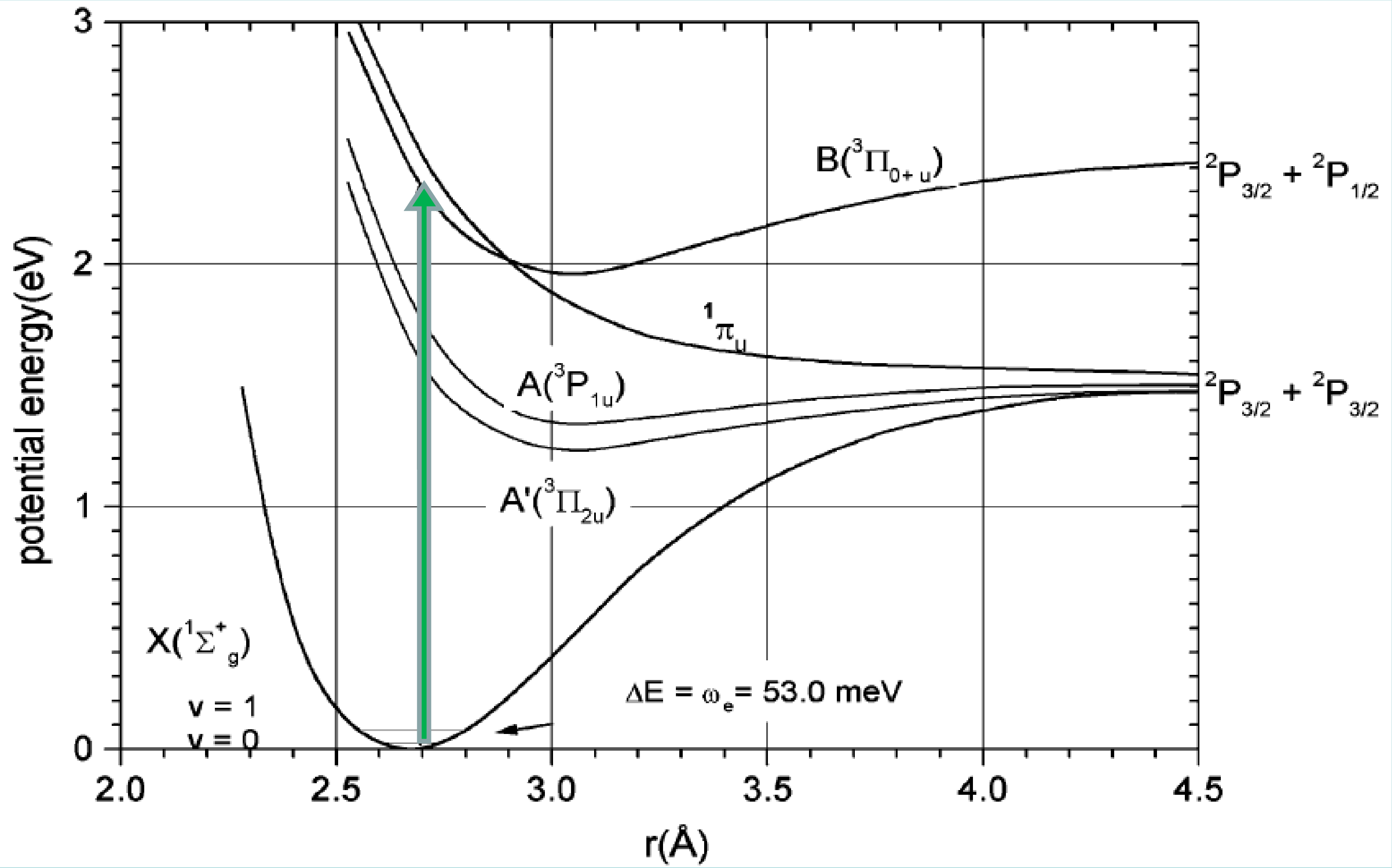
Low peak power, High repetition rate

144 bunch mode: equal spacing, 12 ns; TPS



Reversed for laser pump - X-ray probe up to time span 12ns.

Electronic structure of Iodine, I_2



The potential energy of the iodine molecule for different wavefunctions X, A/A0 etc. The molecule is dissociated by a 150 fs pulse at 530 nm that excites the molecule to the B state (vertical arrow).

Structure Kinetics *-Recombination of photodissociated I_2 in CCl_4*

$I_2 \rightarrow 2I$ or $I_2^* \rightarrow I_2$

Diffracted signal depends on
scattering wave vector, q and time delay, τ .

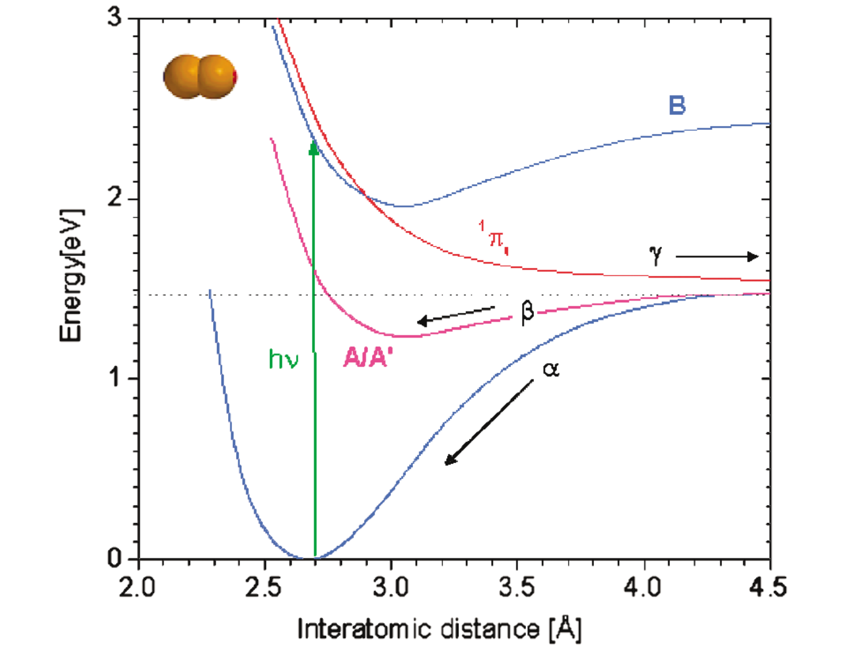


FIG. 1 (color). Low lying electronic energy surfaces of I_2: the states X, A/A', and B are attractive, whereas the state $^1\pi_u$ is repulsive. The processes α, β, and γ denote vibrational cooling along the X potential, geminate recombination through the states A/A', and nongeminate recombination, respectively.

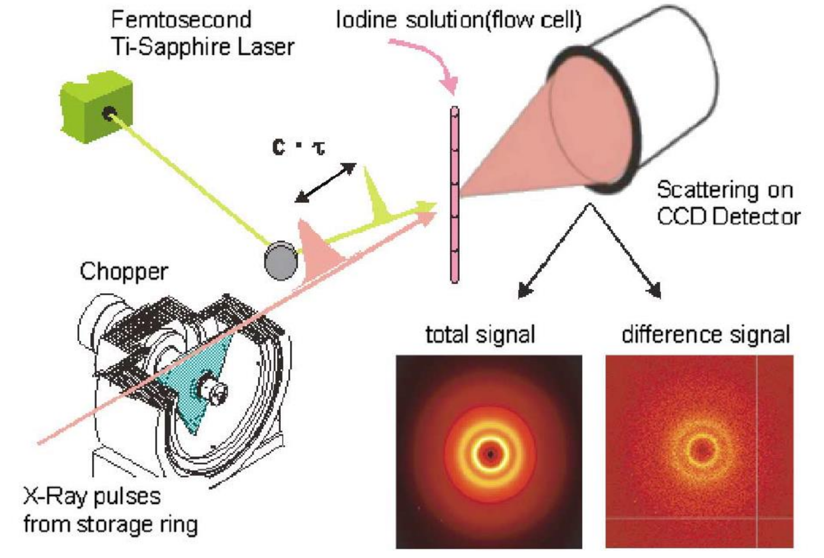


FIG. 2 (color). Experimental setup: the x-ray pulses are generated by an undulator. The spectrum is centered at 0.67 Å (18.5 keV) and its bandwidth width is $d\lambda/\lambda = 0.03$. The flux on the sample is 5×10^8 per pulse and the pulse length is 150 ps. The solution is excited by 150 fs laser pulses at 520 nm, populating the electronic states $^1\pi_u$ and B. The common laser/x-ray repetition frequency is 896.6 Hz and the exposure time 10 s per CCD frame.

Solution molecules

Transient trap the pair of nonbonded atoms and force them to recombination--gemination

Time-resolved optical spectroscopy & time-resolved X-ray diffraction

----monitor atomic motions in liquids

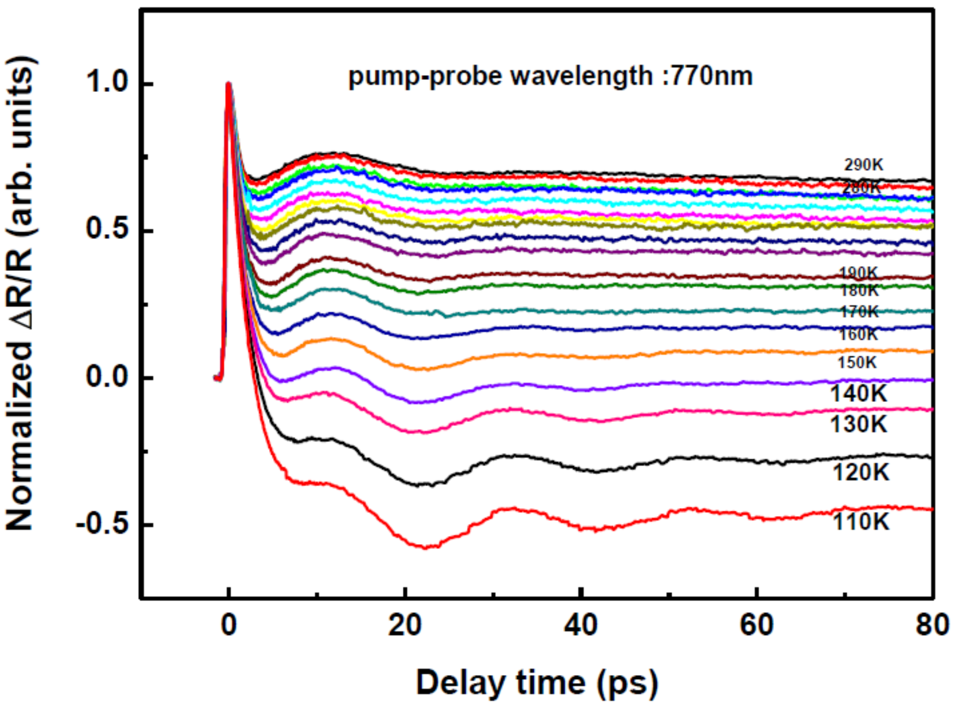
A. Plech etal., PRL 92,125505 (2004), ESRF

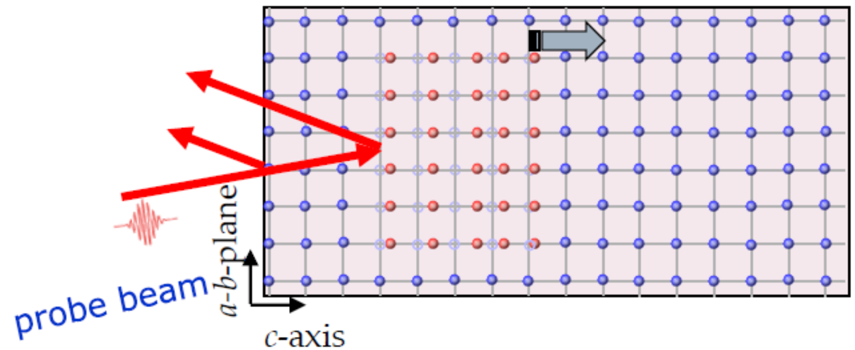
Temporally Coherent XRD

---Ultrafast dynamics of multiferroic material

Generation and detection of coherent acoustic phonons by ultrafast laser,
Probing the propagating strain by using time-resolved X-ray diffraction

$$\frac{\Delta R}{R}(T,t) = A_f(T)e^{-\frac{t}{\tau_f}} + A_{s_1}(T)\left[1-e^{-\frac{t}{\tau_{s1'}}}\right]e^{-\frac{t}{\tau_{s1}}} + A_{s_2}(T)\left[1-e^{-\frac{t}{\tau_{s2'}}}\right]e^{-\frac{t}{\tau_{s2}}} + A_o(T)e^{-\frac{t}{\tau_d}}\cos(\omega_o t+\varphi_o)$$





- **Propagating Strain Pulse Model**

longitudinal coherent acoustic phonons

Interference between surface and strain layer

HC Shih, TH Lin, CW Luo, et al., New J. Phys. 13, 053003 (2011)

Laser-Synchrotron electron-ion momentum imaging with few bunch operation

Magnetic bottle electron time-of-flight spectrometer

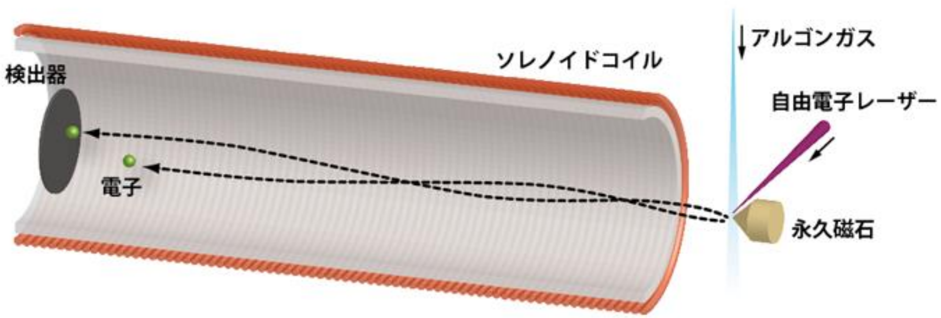
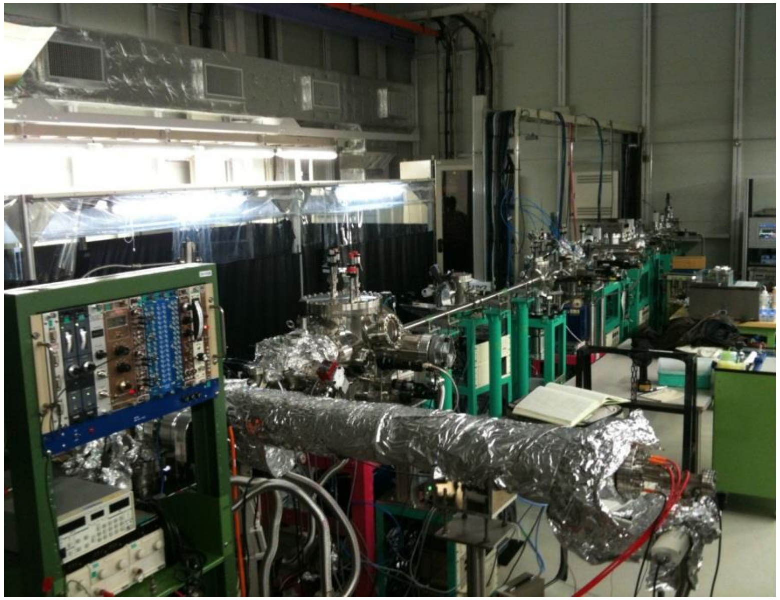


図１　磁気ボトル型光電子分光器

Magnetic bottle electron time-of-flight spectrometer @ test SCSS Spring-8.



Perspective

Magnetic bottle electron time-of-flight spectrometer

- ➡ Large detection energy range (0 ~ 1000 eV) of electron
- ➡ Full solid angle (4 π) of whole energy range of electron
- ➡ Adaptive ion extraction for e-ion coincidence detection

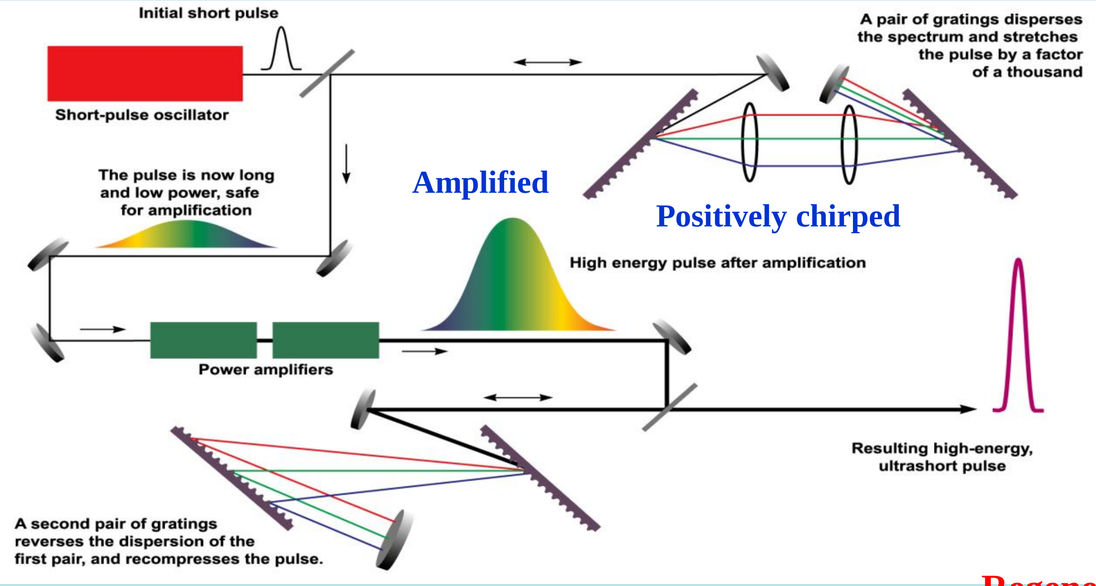
Extremely high sensitive to perform single-shot experiment by free electron laser and multi-electron coincidence (> 2 electrons) experiment by normal synchrotron radiation.

Pump-probe experimental scheme II

High peak power, low repetition rate

Mode-locked Laser System

Ti:Sapphire regenerative amplifier



Negatively chirped

	Legend (laser amplifier)
Pulse width	35 fs
Central wavelength	790 nm
Bandwidth	30 nm
Energy per pulse	4 mJ
Repetition rate (tunable)	1 kHz
Average power	4 W
Peak power	$1.15*10^{11}$ W

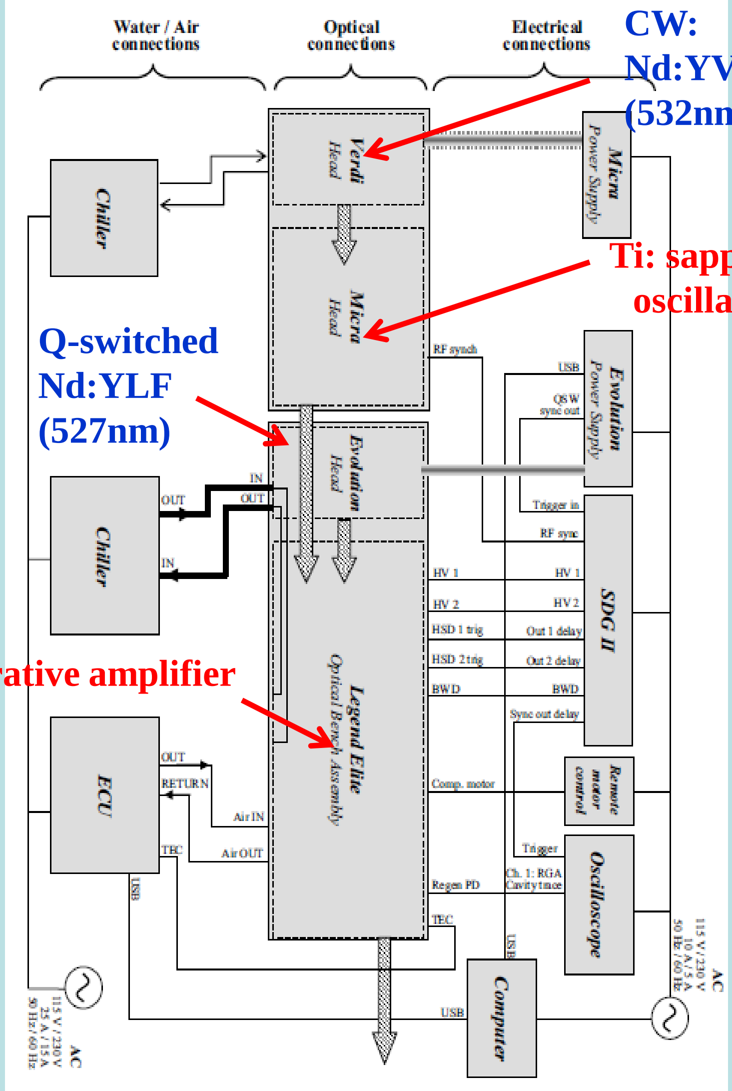
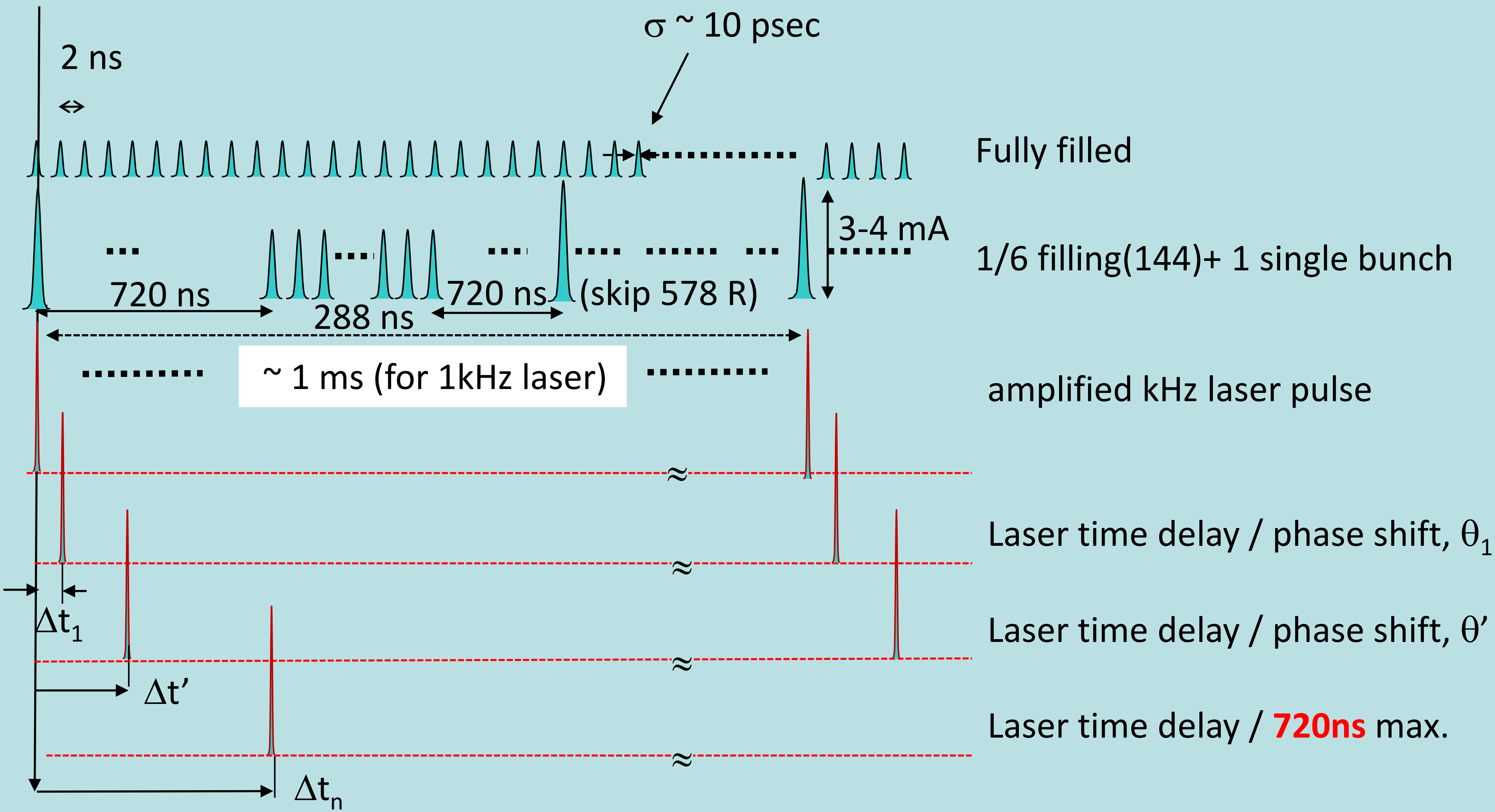


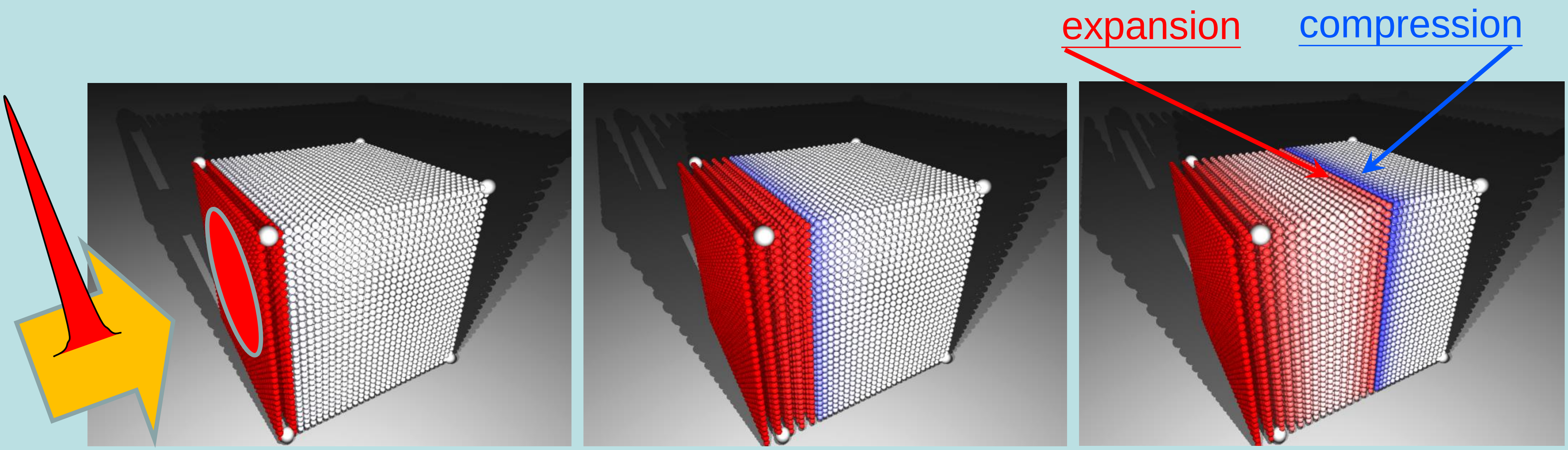
Figure 3-3. Legend Elite HE Connectivity Diagram

Single bunch mode:1/6-filling(144) + 1 bunch; TPS



Laser excitation of a solid (below damage)

A coherent set of acoustic phonons is generated in a semiconductor crystal via laser excitation



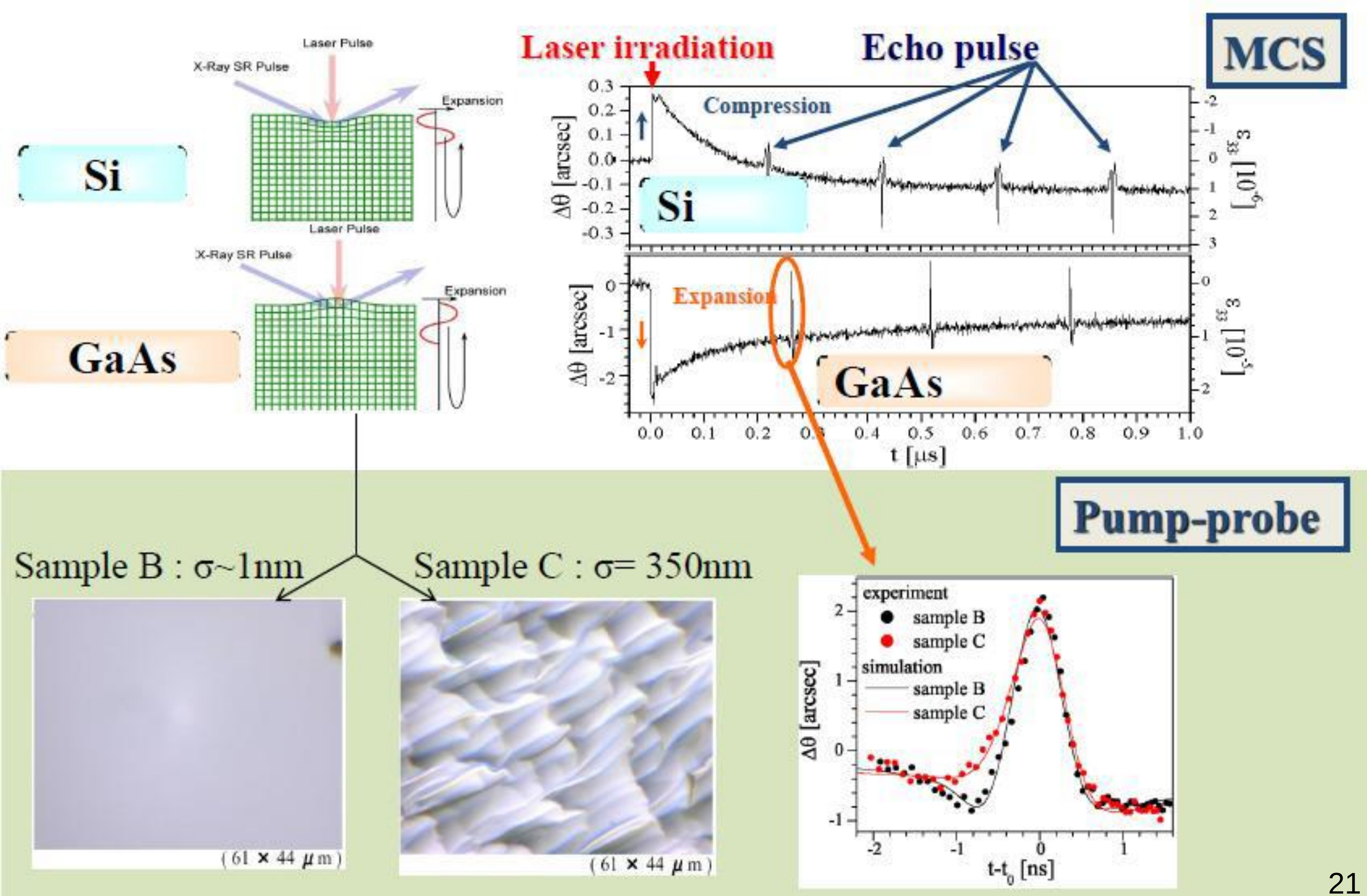
a) **Electrons are excited→ electronic stress due to deformation**
b) **Electrons heat lattice**
c) **Thermal stress**

Stress relaxes by lattice expansion

Expansion triggers acoustic wave (coherent phonons) --due to Newtons 3rd law

C. W. Siders et al.

Time-resolved XRD

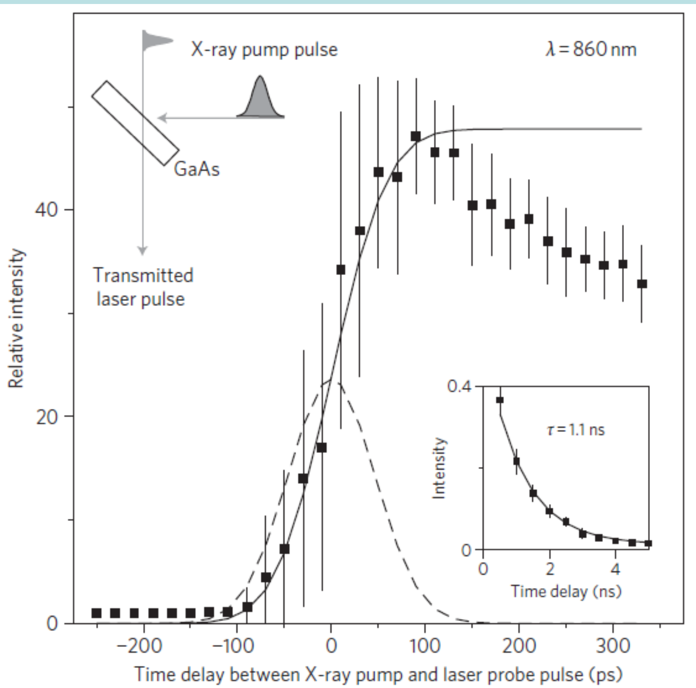


Y. Hayashi, Y. Tanaka, T. Kirimura, N. Tsukuda, E. Kuramoto, T. Ishikawa: Phys. Rev. Lett. 96 (2006) 115505.

X-ray induced optical transparency/opacity of GaAs

A study on ultrafast many-body response of semiconductors to x-ray absorption by X-ray pump-optical probe cross correlation.

Thin GaAs crystal, X-ray pump/Laser probe



Transmission at 860 nm versus time after X-ray pump pulse.

just above the bandgap→ **Transparent**
just below the bandgap→ **Opacity**

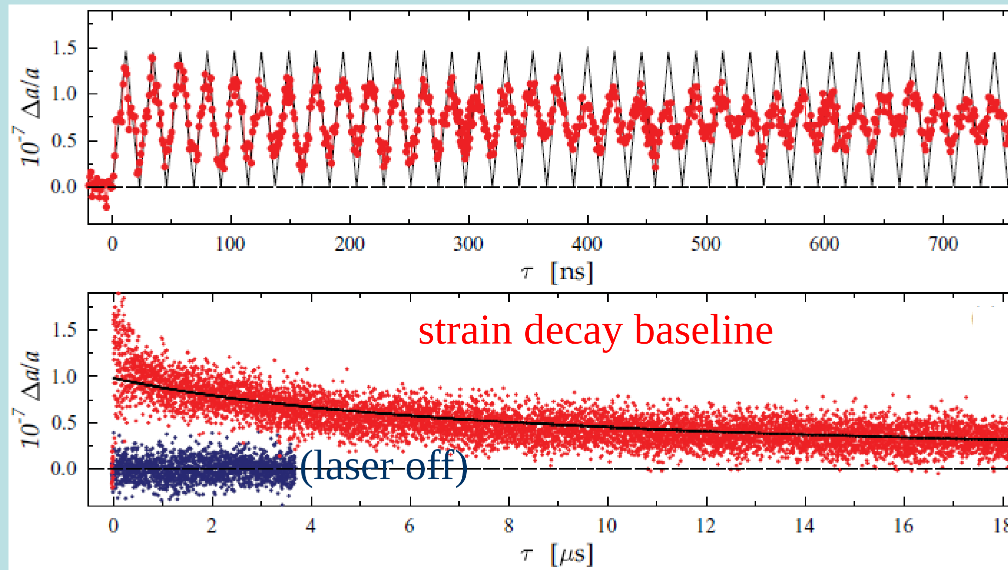
S. M. Durbin et al, Nature Photonics **6**, 111 (2012). (APS)

Dynamics of thermal expansion of Diamond lattice

lattice response probed by diamond (008) reflection

time scale 100 ps ~ 18 μs
X-ray $\Delta E/E \sim 10^{-8}$
low flux laser $\lambda = 400$ nm

Strain oscillation due to the strain standing wave



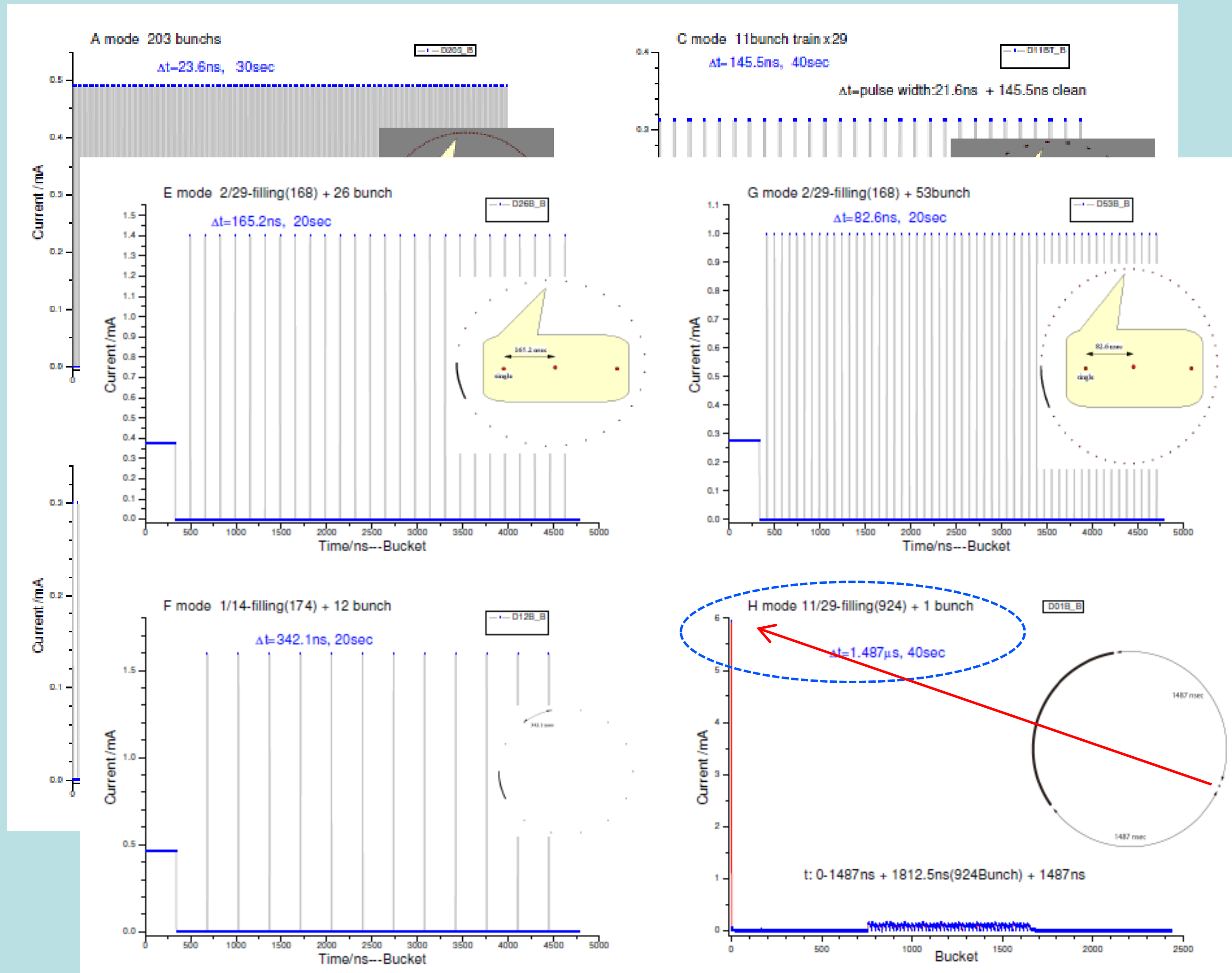
The results of experiment averaged for the low-energy and high-energy slopes are shown by red circles and lines. The sawtooth-like curve (black line) represents the strain profiles averaged across the thickness of the diamond crystal (Reflectivity→ Lattice parameter).

S. Stoupin et al, PRB **86**, 054301 (2012). (APS)

Current experimental facility

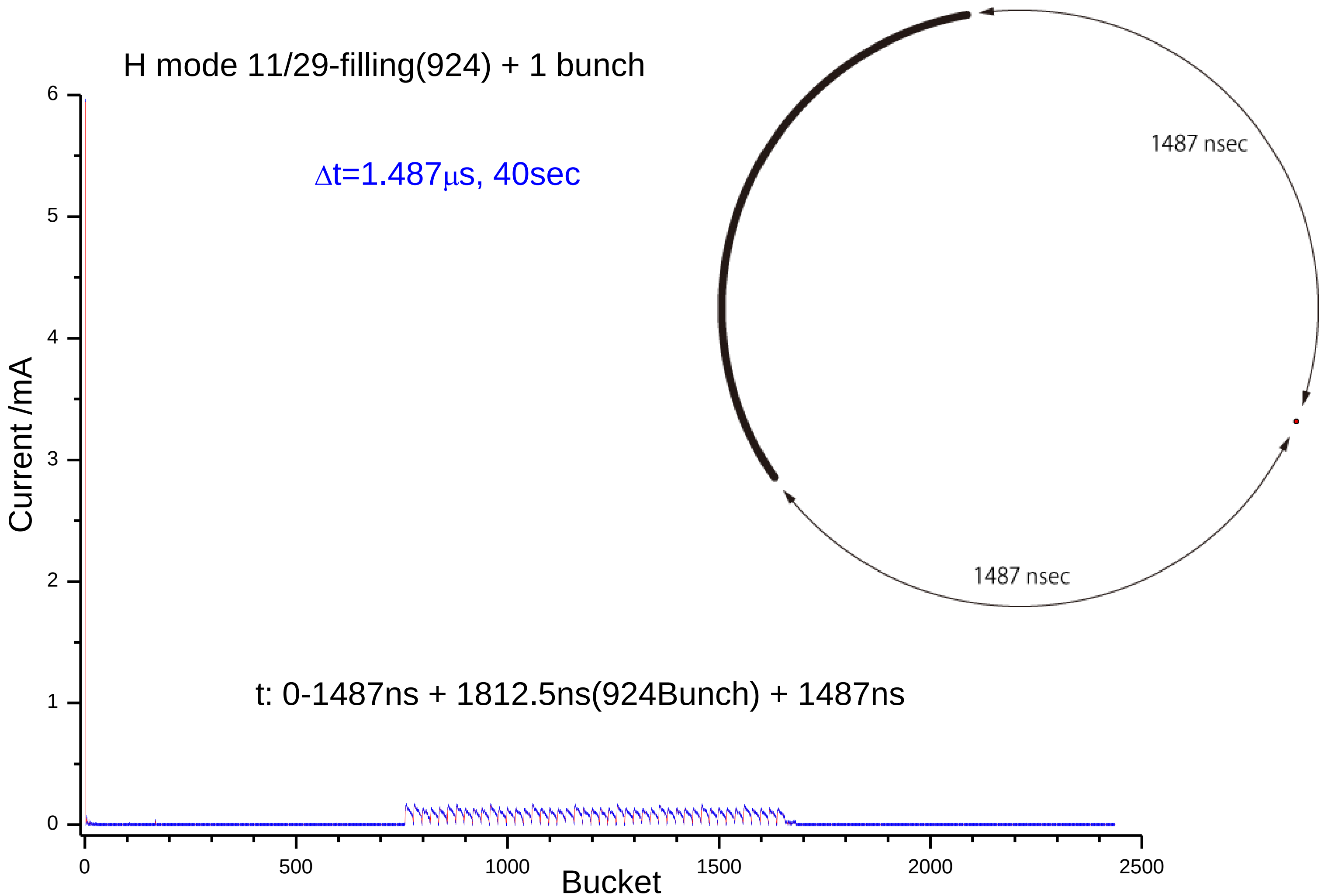
Spring-8 Several Bunch Modes

A. 203 bunches
B. 4 bunch train x 84
C. 11 bunch train x 29
D. 1/7-filling + 5 bunches
E. 2/29-filling + 26 bunches
F. 1/14-filling + 12 bunches
G. 4/58-filling + 53 bunches
H. 11/29-filling + 1 bunch



2012B

H-mode : 11/29-filling(924) + 1 bunch /SPring-8



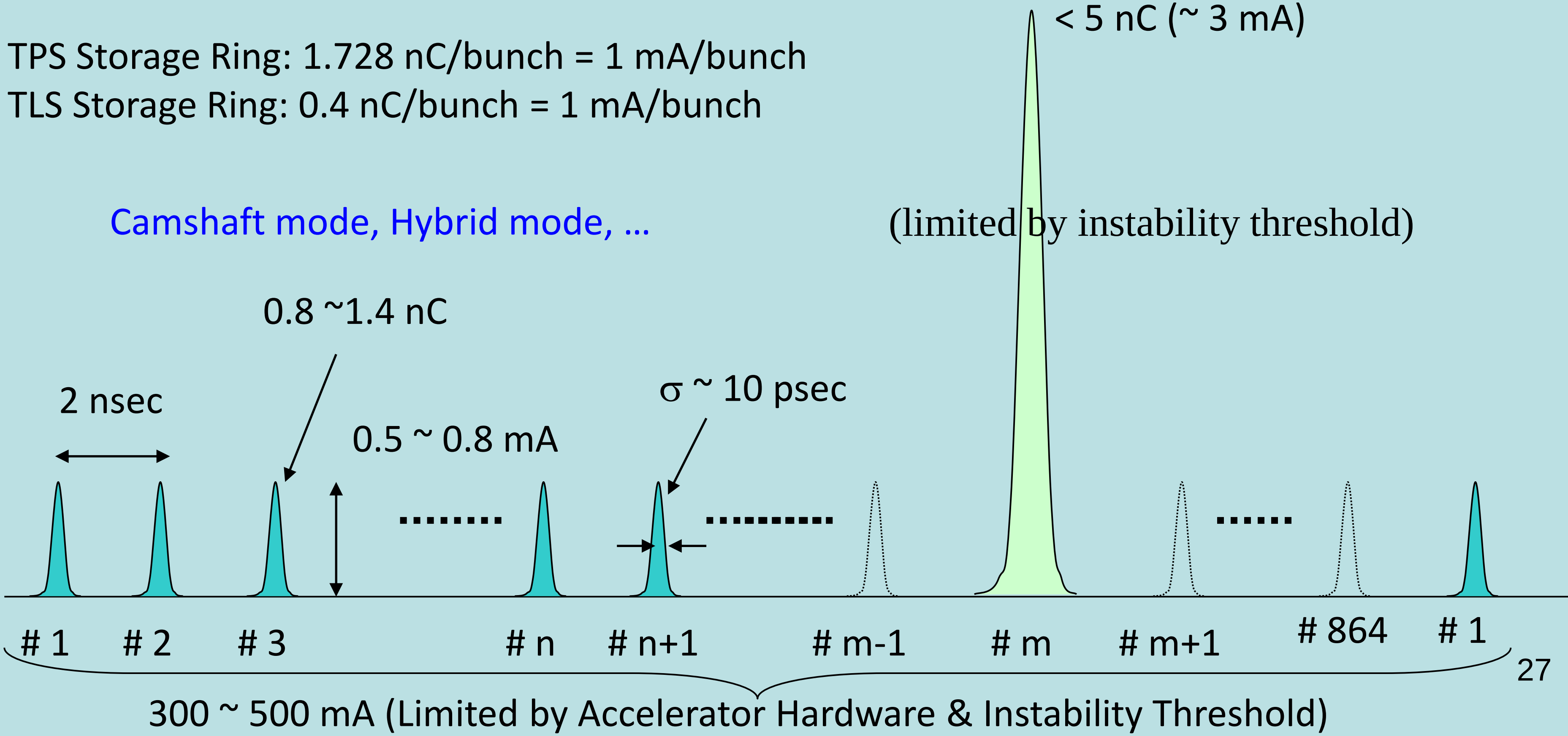
TPS Experimental Capability

TPS Capability

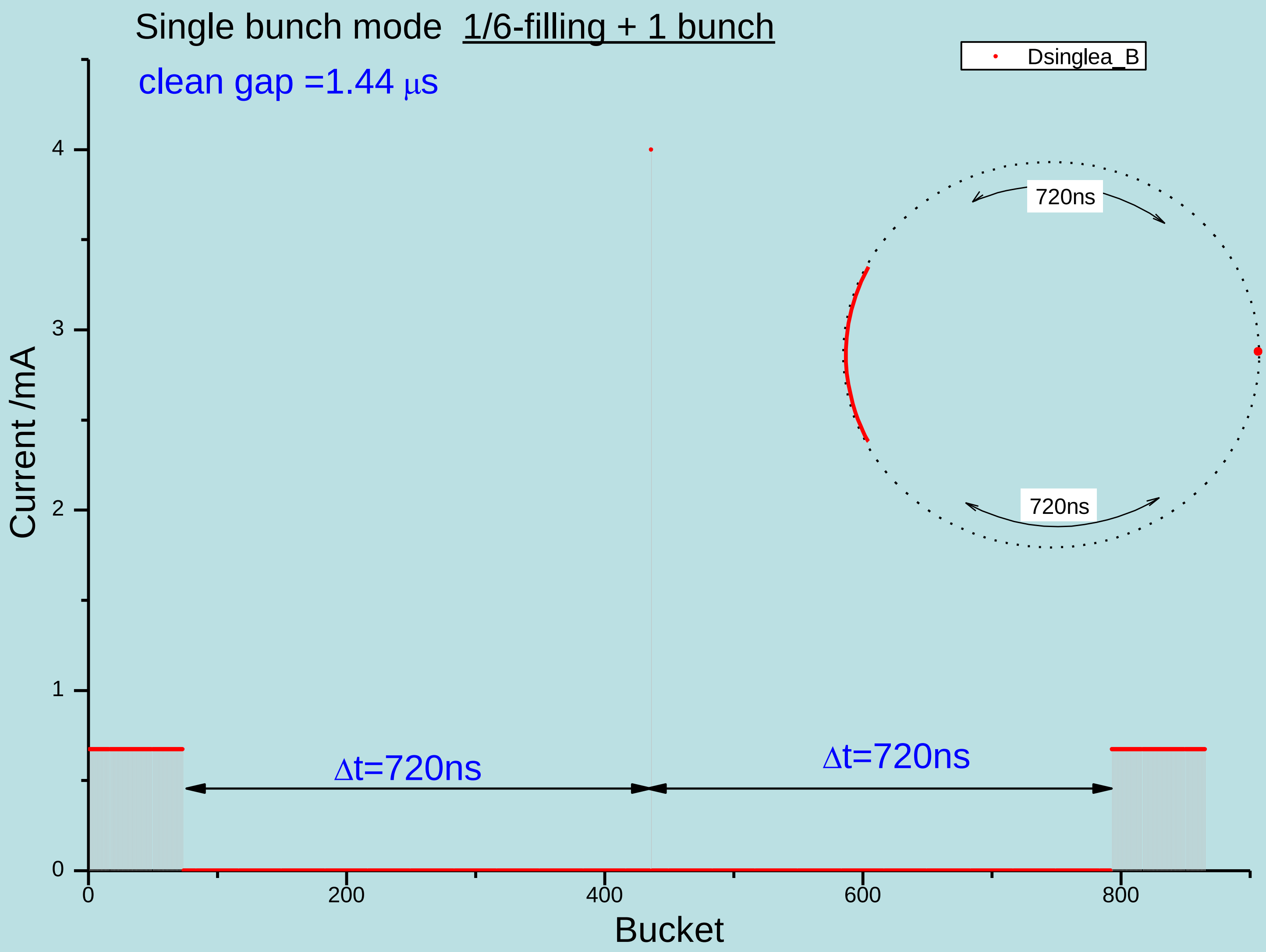
Bunch number: 864
Multi-bunch current: ~ 500 mA (dependent on available RF power)
Multi-bunch bunch current: < 1 mA/bunch
Single-bunch (isolated bunch?) current: 2 ~ 3 mA
Hybrid mode (camshaft mode)

TPS Storage Ring: 1.728 nC/bunch = 1 mA/bunch
TLS Storage Ring: 0.4 nC/bunch = 1 mA/bunch

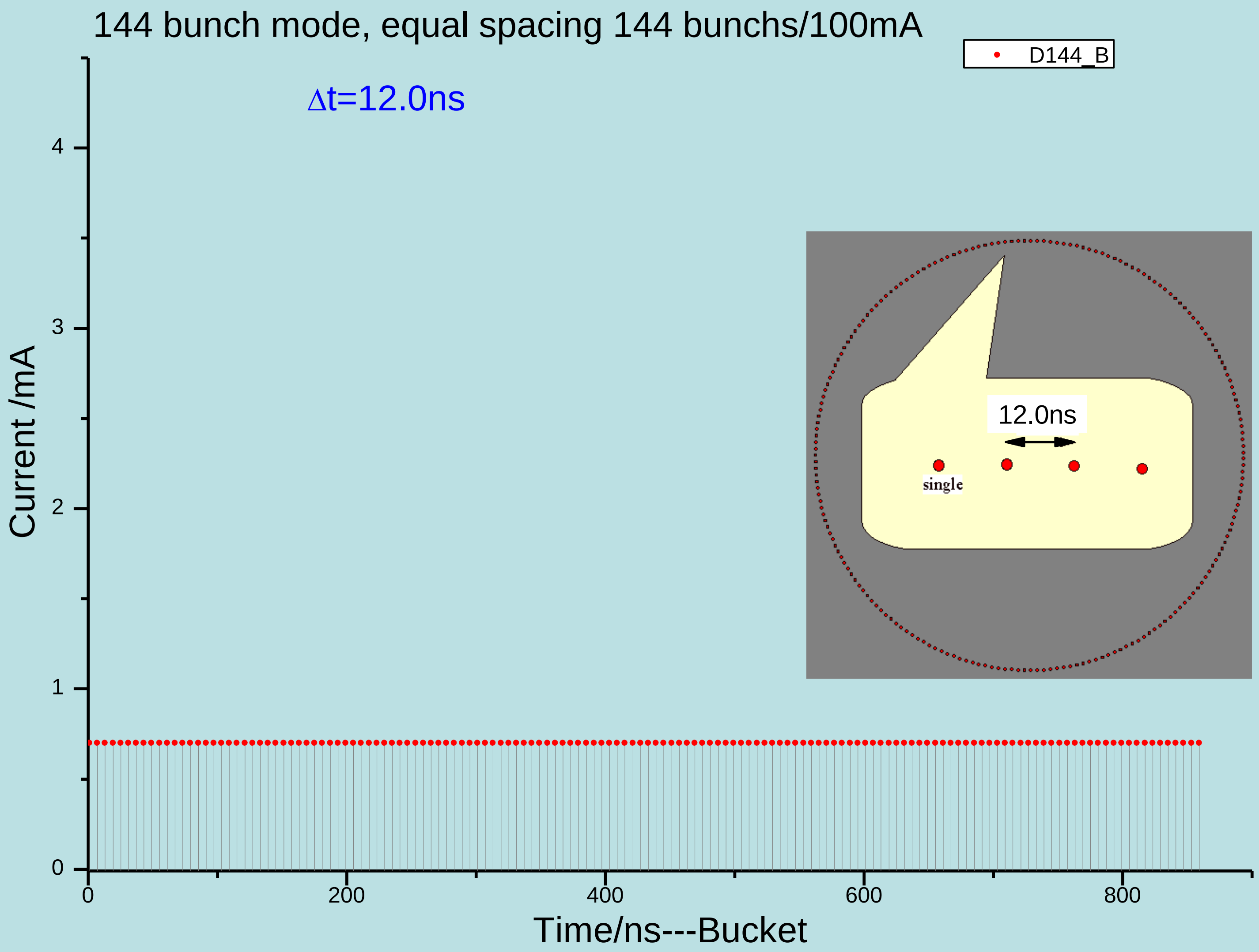
Camshaft mode, Hybrid mode, …



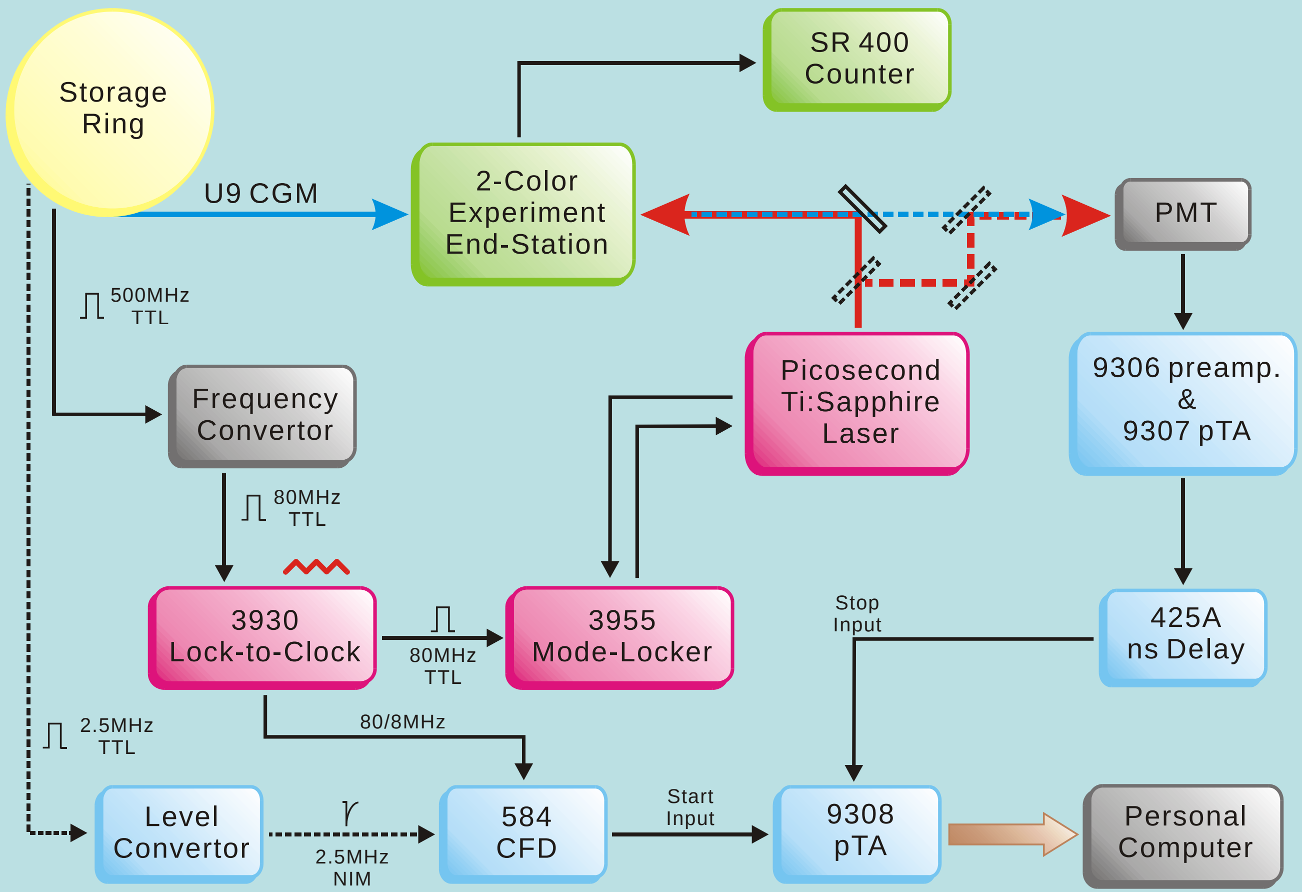
Single bunch-mode : 1/6-filling(144) + 1 bunch /TPS



144-bunch mode : every 6 bucket, 12ns spacing /TPS

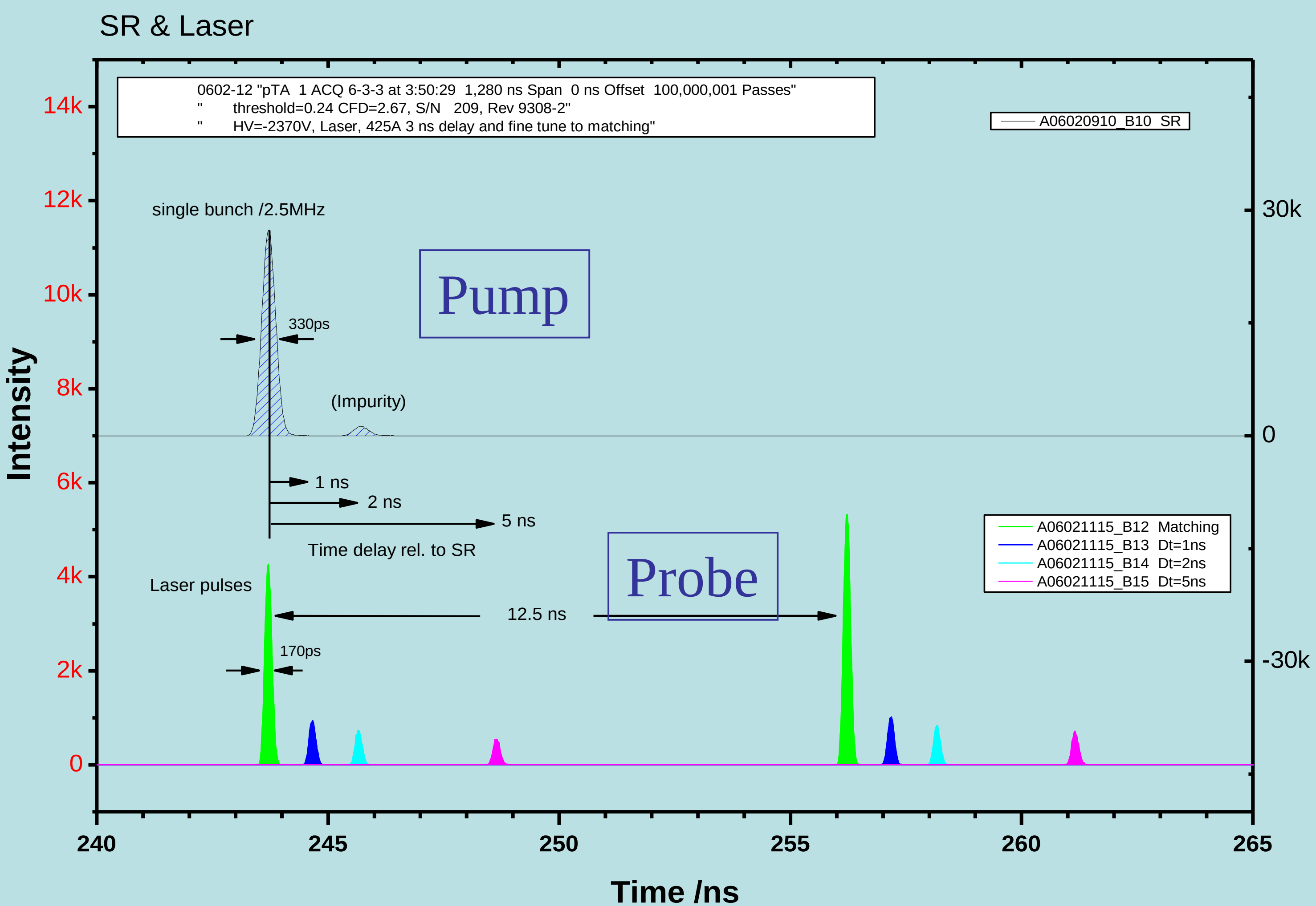


Setup for timing control and measurement system at TLS



Block diagram of pico-second timing measurement system. The trigger to the mode-locked laser is linked to a frequency convertor which divide the master oscillator of the rf cavity in the storage ring of TLS. The timing jitter between the synchronized laser and SR systems is less than 50 ps.

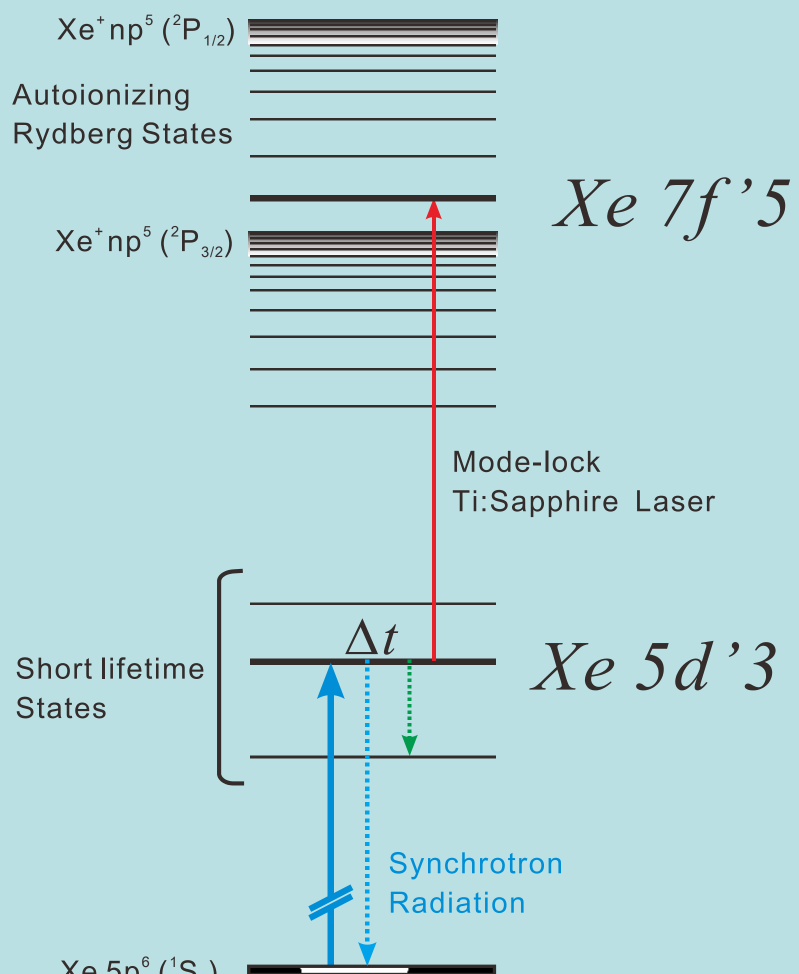
Pump-Probe scheme of SR and laser combination

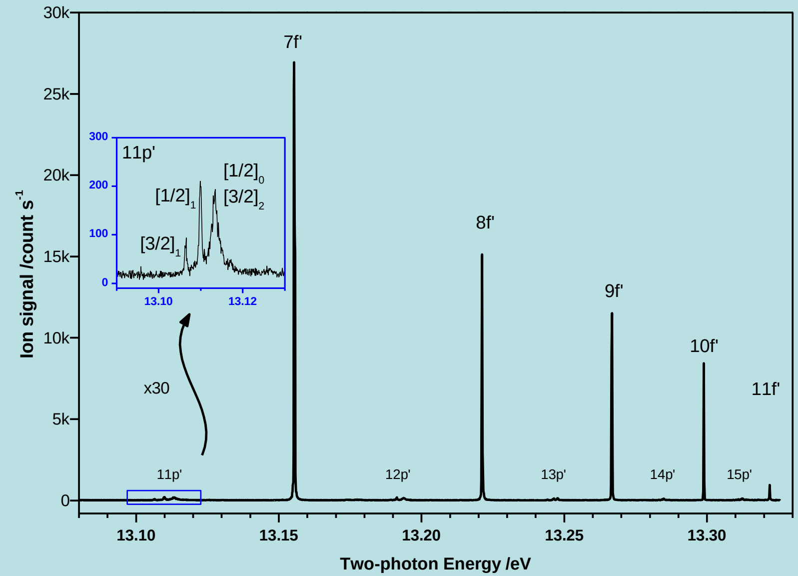


Hybrid bunch modes to extend the clean gap

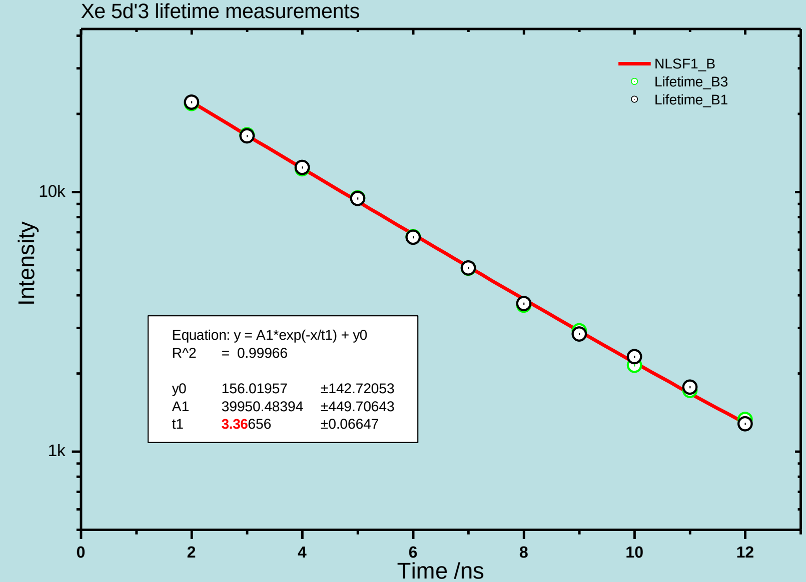
Two-color Pump-Probe Experiments:

Atomic Xe Spectroscopy and lifetime measurement





Xe 5d'3 lifetime measurements

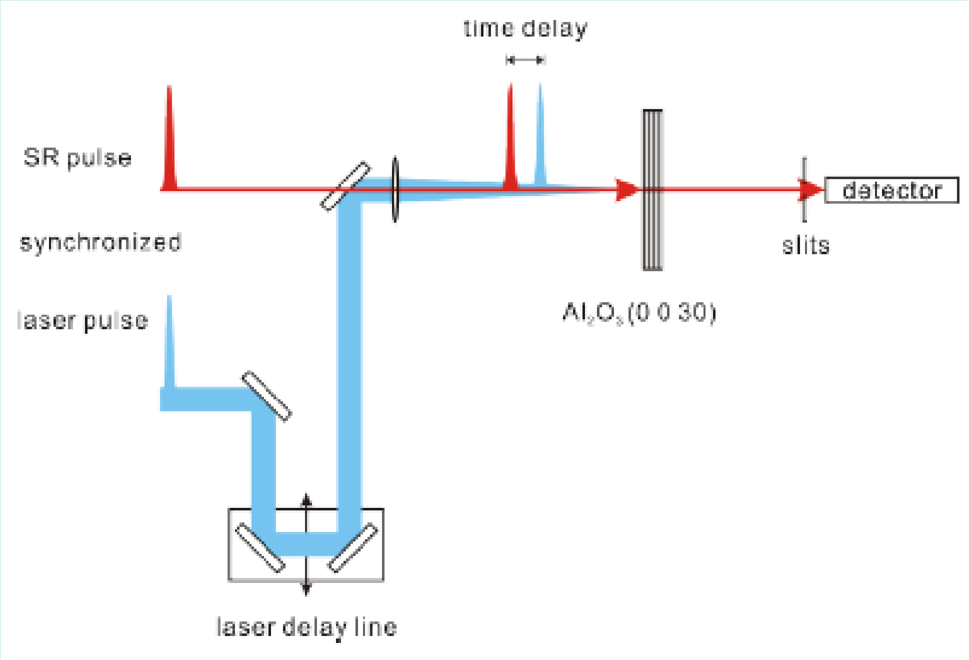


Y.-Y. Lee *et al, J. Elec.Spect.Rel.Phen.*,144-147,29(2005)

TPS 09A Temporally Coherent XRD

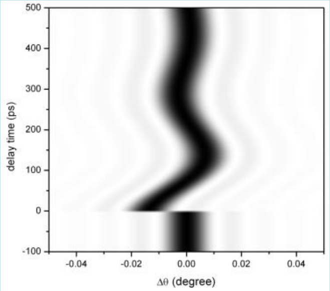
---Lattice dynamics study by ultrafast X-ray diffraction

Probing optical properties of x-ray resonators



The excited phonon phenomena cause to reflectivity oscillations under the back diffraction condition with sapphire (0 0 30) for Hard X-ray(14.315)

Simulations of 400nm Ge(111) Film



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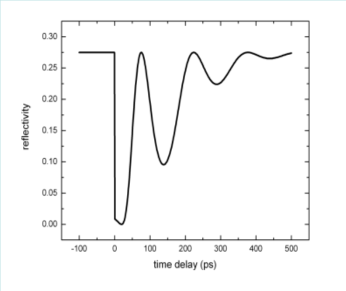
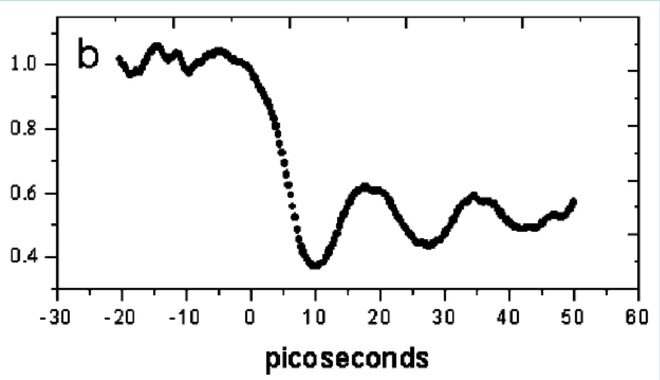




Diffraction from coherent phonons, A.M. Lindenberg, PRL 84, 111(2000)

- Ultrafast laser pulses induced stress and lattice vibration vary with time which can be determined with X-ray pump-prob experiments
- Each vibration mode corresponds to characteristic profile and frequency
- Laser pulse induces lattice dynamic which dependent on the laser profile and fluence

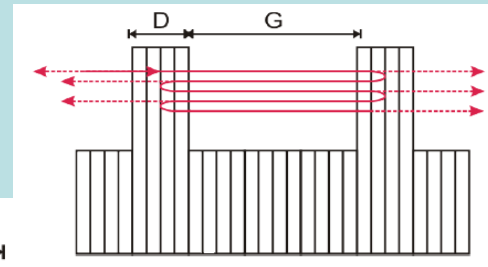
TPS 09A Temporally Coherent XRD

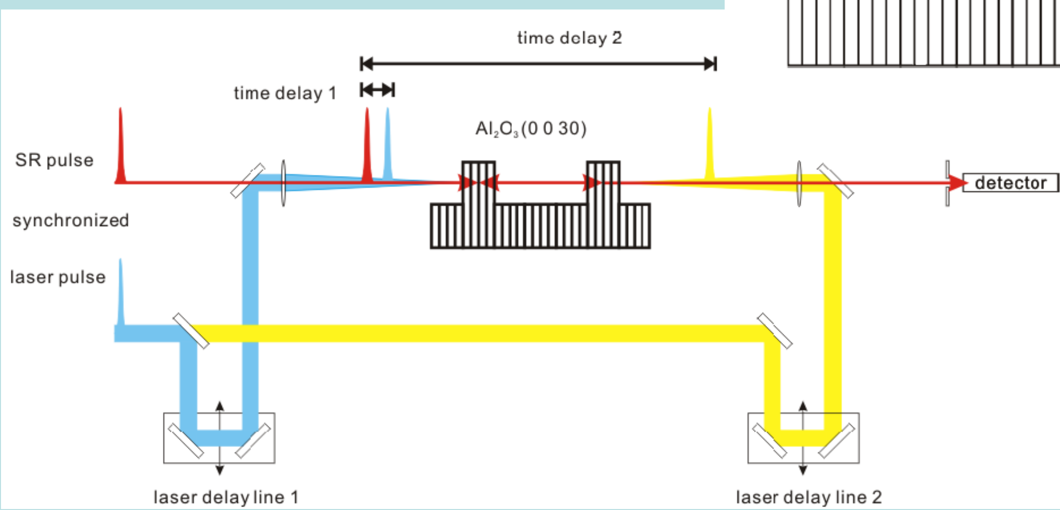
Potential Experiments

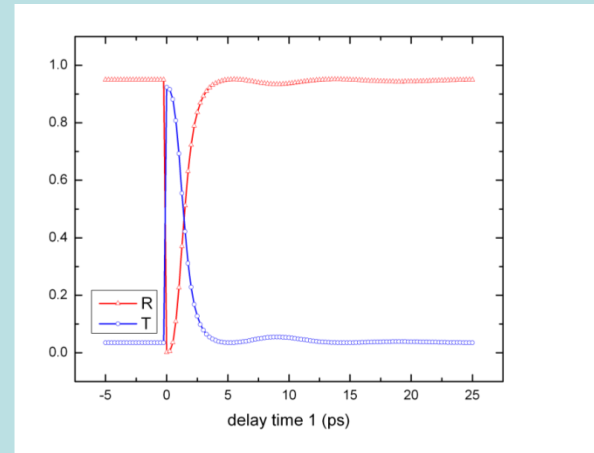
Study phonon phenomena with Al_2O_3, GaAs, Ge, …….

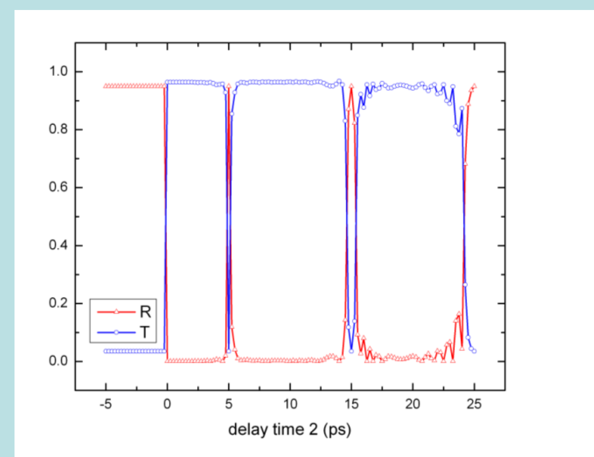
Hard X-ray Q-switching cavity

--Study of optical properties of the excitation and nonlinearity of materials filled in an x-ray cavity









- **Adjust Td1 and Td 2 to control the timing of reflectivity gates of front and rear plates.**
- **Under the condition D= 120 um and D+G=600 um, round trip propagating time closes to 4 ps. The timing different of delay time 1 and 2 corresponds to the number of cycles of X-ray pulse in cavity.**
- **Laser profiles and fluences are dominated the reflectivity profiles varying with time**

Y.Y. Chang, S.L. Chang *et al.*,Opt. Exp. 18, 7886(2010)

Required Timing control System

Build up the timing control system from ps-ms

1. **Trigger and clock delay units**
   - high precision, 1ps
   - ultra-wide range time delay, >μs

2. **Fast Choppers/Shutters**, <300ns, jitter < 5ns, atm.
   - X-ray attenuation
     avoiding sample over exposure and damage
   - encoded and phase lock loop controlled
   - small long-term jitter
   - single-bunch extraction, kHz

Support users to do time-resolved experiments.

Trigger and Clock delay module

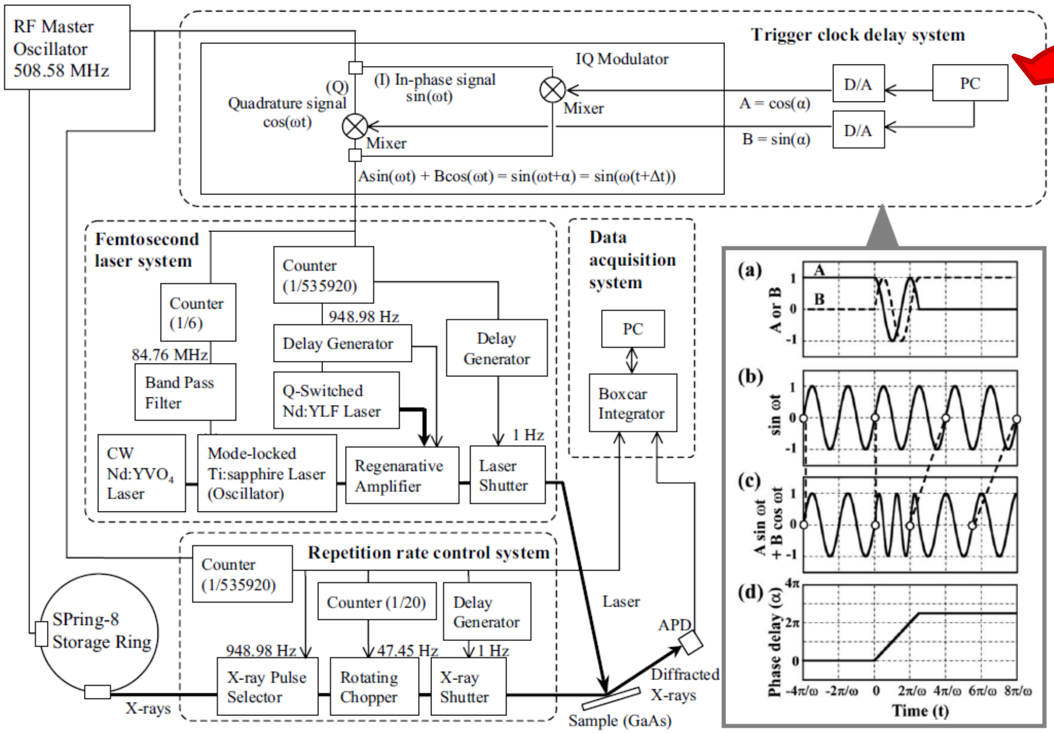


FIG. 1. Block diagram of the trigger clock delay system, the femtosecond laser system, the repetition rate control system, and the data acquisition system. The inset gives the schematic graphs to explain the principle of the continuous phase change at the trigger clock delay system. When the control voltages (a) are applied from the D/A converters, a rf input signal (b) to the IQ modulator is converted into the output signal (c) with the phase delay of α in (d). The open circles and the broken lines between them in (b) and (c) show the certain phases to find the phase shift by larger than 2π.

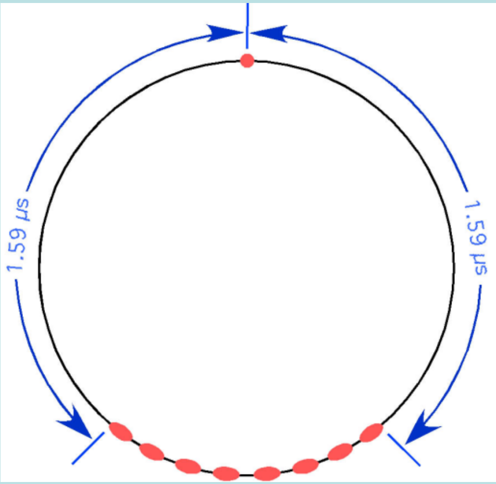
Trigger and Clock delay units/*SPring-8*

- In phase quardrature modulator and a synchronous counter
- Delay time nearly infinite amount
- Precision in ±8.4 ps

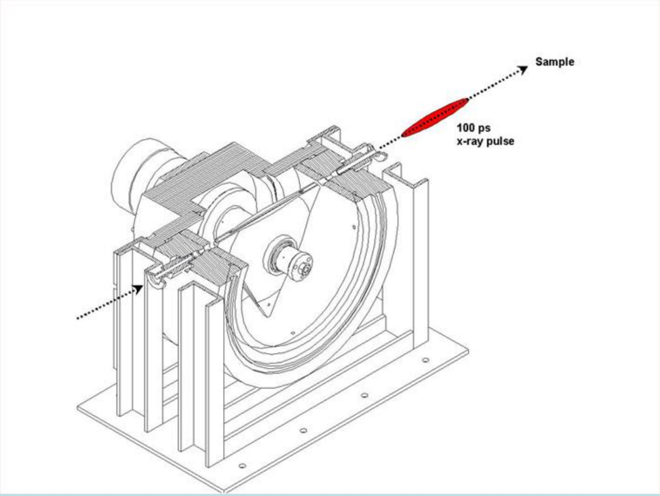
Y. Fukuyama *etal*, RSI 79, 45107 (2008)

X-ray Choppers

X-ray choppers can select x-ray pulses from synchrotron pulse train
1. Define temporal resolution
2. Limit heat load
3. Reduce noise



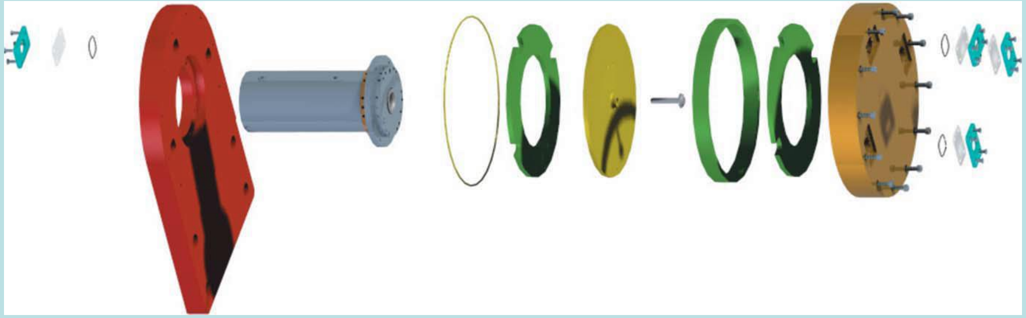
Mechanically Rotating
1. ESRF/Julich: adjustable time window, white beam
2. APS highspeed: fixed window, mono or pink beam
3. Philip Coppens' rotating chopper(SUNY,Buffalo)



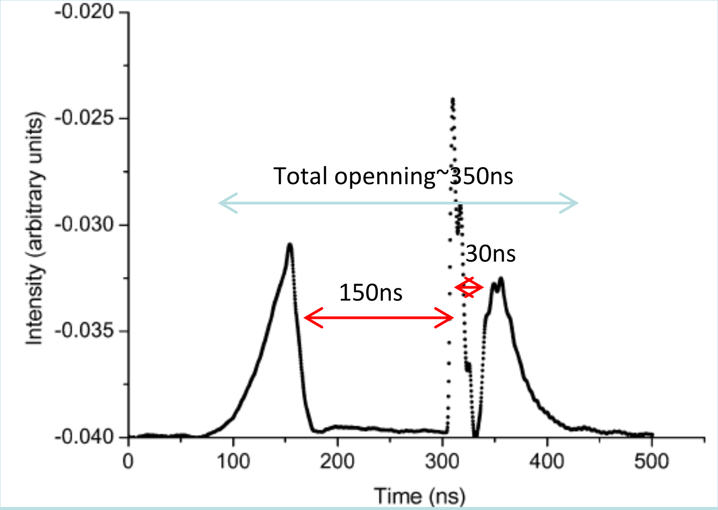
Julich rotating chopper
– In-vacuum triangular Ti rotor with beam tunnel
– maximum radius 96.8 mm, tunnel length 165 mm
– Magnetic bearing running from 10 to 900 Hz
– Mono and white beam compatible to 40 keV

Fast Chopper

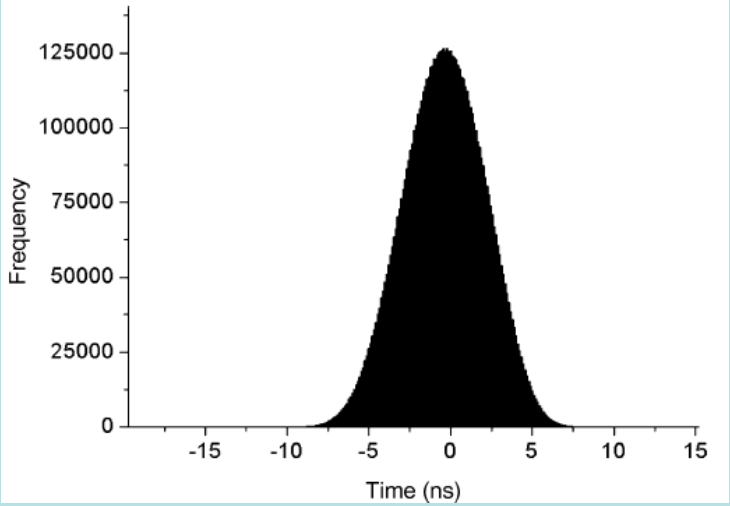
- <u>Wheel</u>,
- <u>Slit</u>,
- <u>Bearing</u>,
- <u>Encoder</u>,
- <u>Phase lock loop controller</u>



- **PSI, DASY/SLS: diameter: 150mm, slit: 50μm, 4mm Cr: ε<10^{-10} for 25keV**
- 500Hz—ratio: 1/4.7M → 350nm, jitter <5ns, 1 atm.
- 1kHz--- ratio:1/9.4M → <150ns, air-bearing, low pressure.
- Multi-slit, >100kHz Xray pulses capability



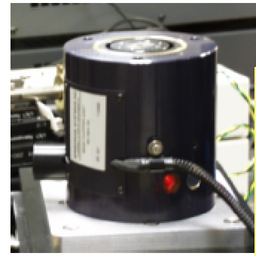
Time-averaged intensity profile of the X-ray signal of the chopped X-ray bunch measured with a fast Si diode

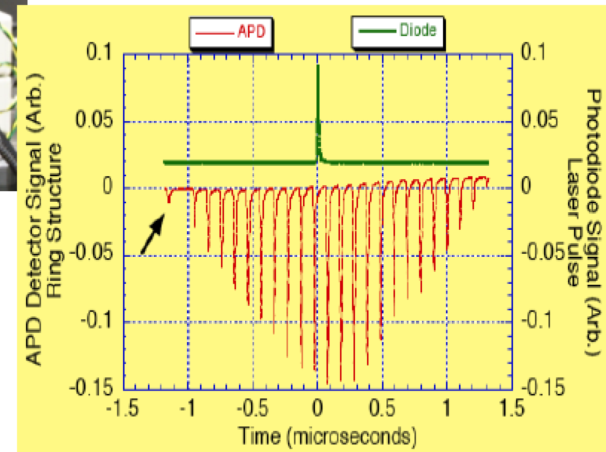


Long-term jitter, 80min, PSI, SLS

--A. Meents etal, JACry, 42, 901, (2009) --PSI, SLS

Compact Rotating Choppers

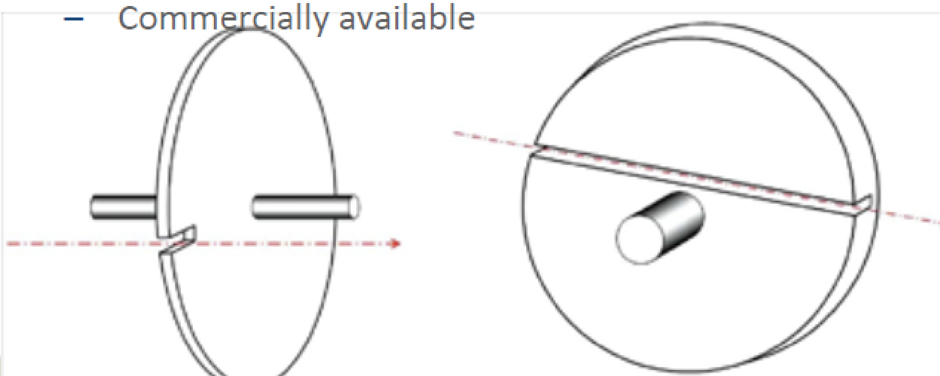


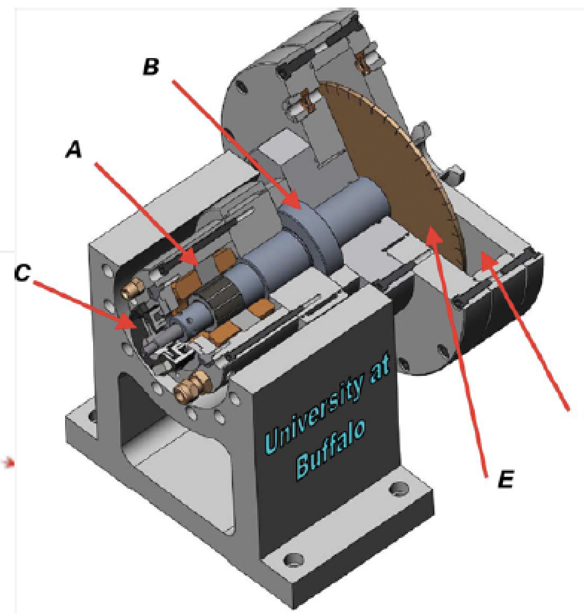


- APS Rotating Chopper
  - In-He disk rotor (R=25 mm) with 0.5x2.29 mm^2 slot
  - Air bearing running at fixed 1331 Hz (< 3 ns jitter)
  - 2.45 µs opening for (singlet) hybrid fill pattern only
  - Mono and white beam compatible to 30 keV
  - Very compact, portable, and inexpensive

McPherson, et al. J. Synchrotron Rad. (2000). 7, 1-4

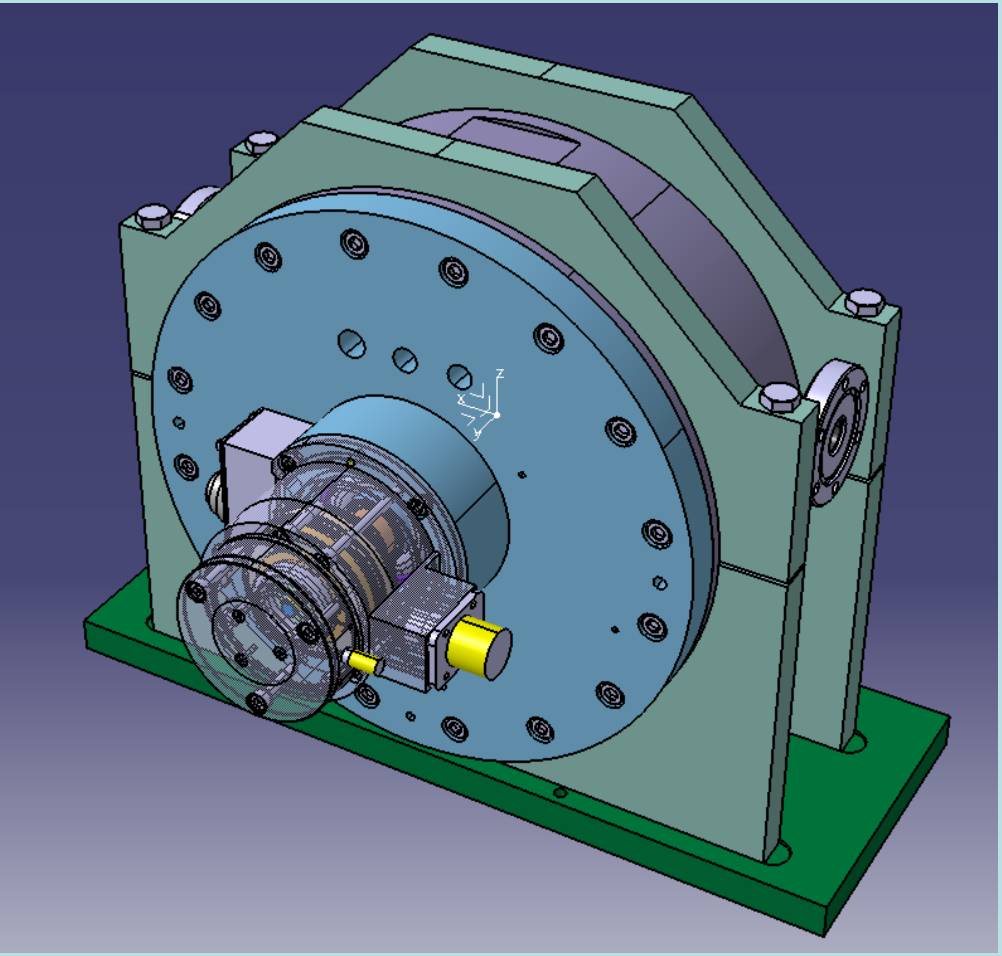
- Rotating Chopper by Phil Coppens
  - Disk rotor (R=70 mm)
  - Air bearing running at fixed 500 Hz (< 3 ns jitter)
  - 1.56 µs opening (350 µm slot)
  - Rotor configuration is very flexible
  - Commercially available

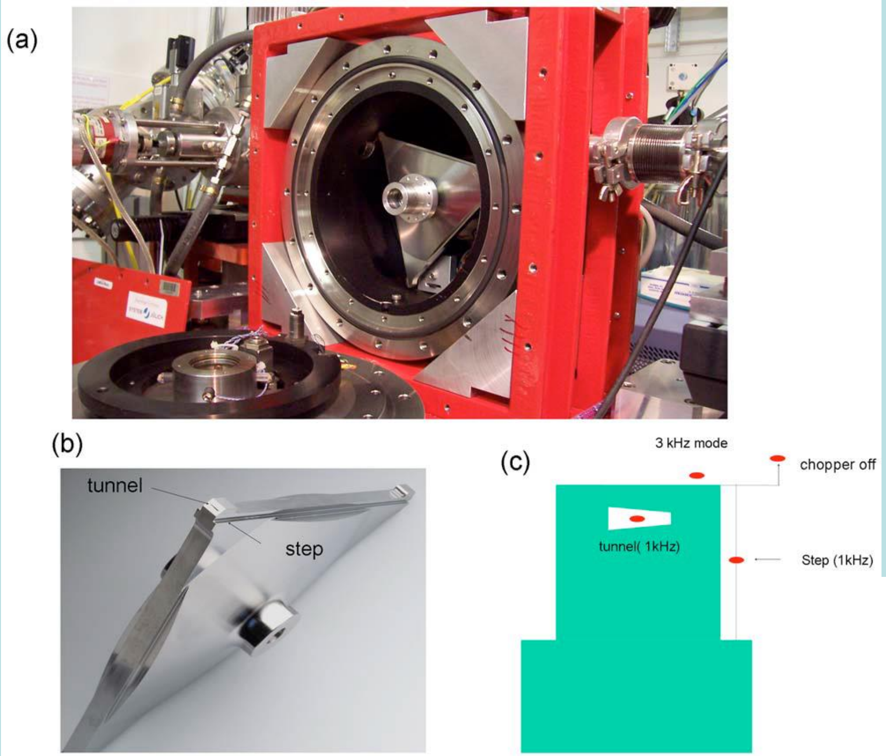


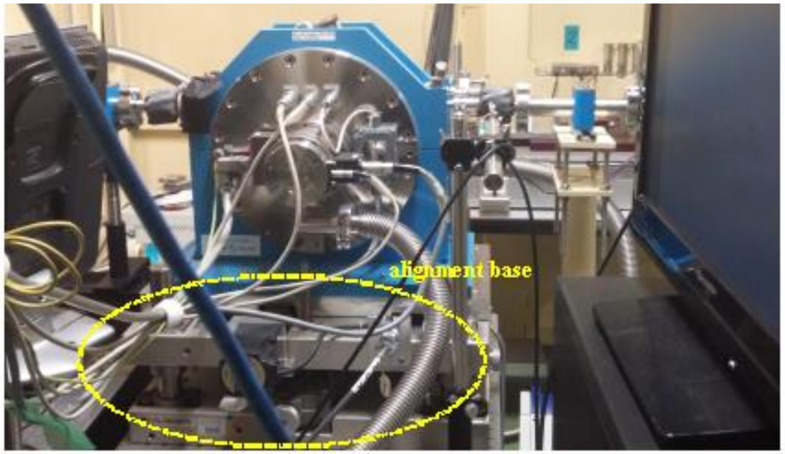


Gembicky, et al. J. Synchrotron Rad. (2005). 12, 665–669

Jülish chopper

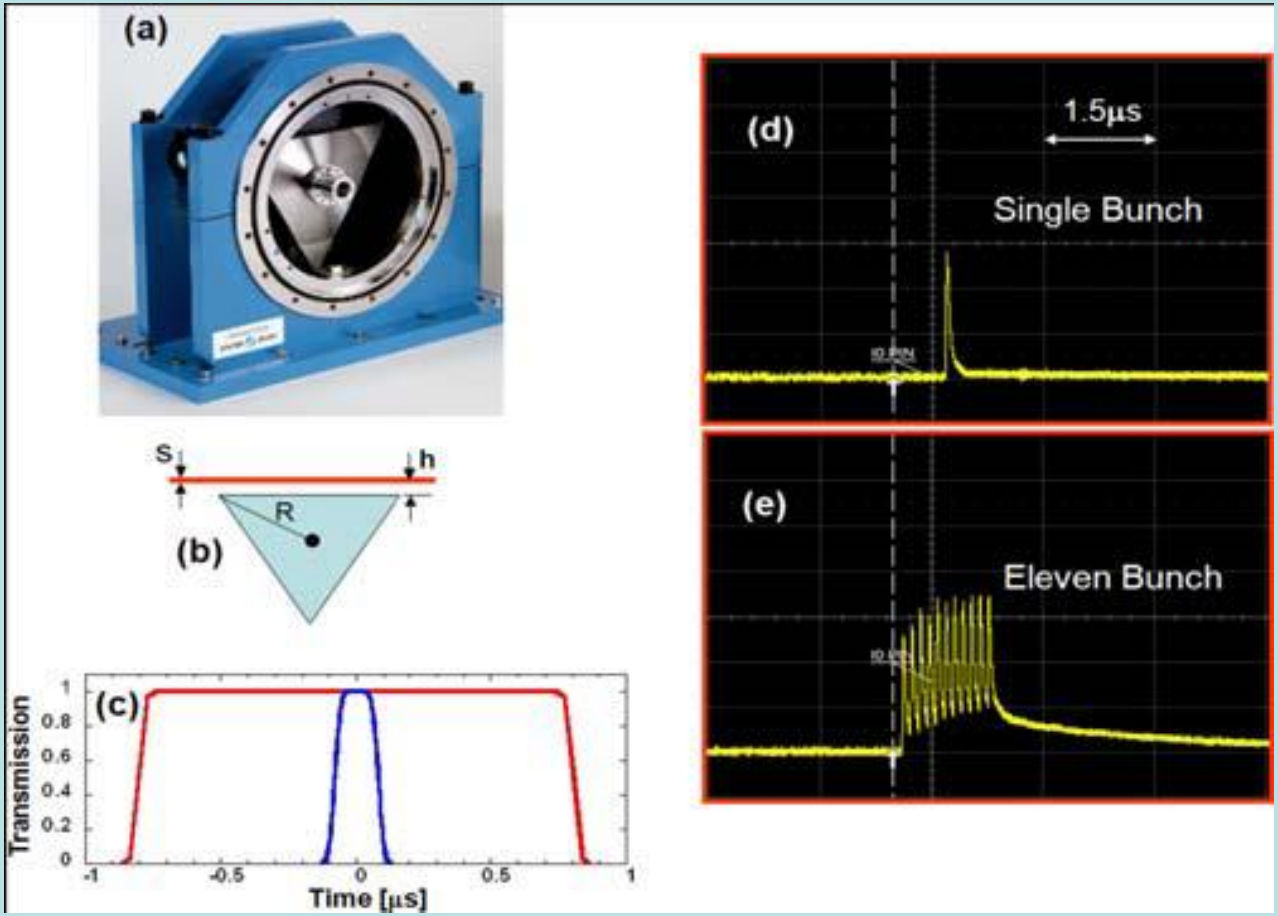






Chopper/Shutter

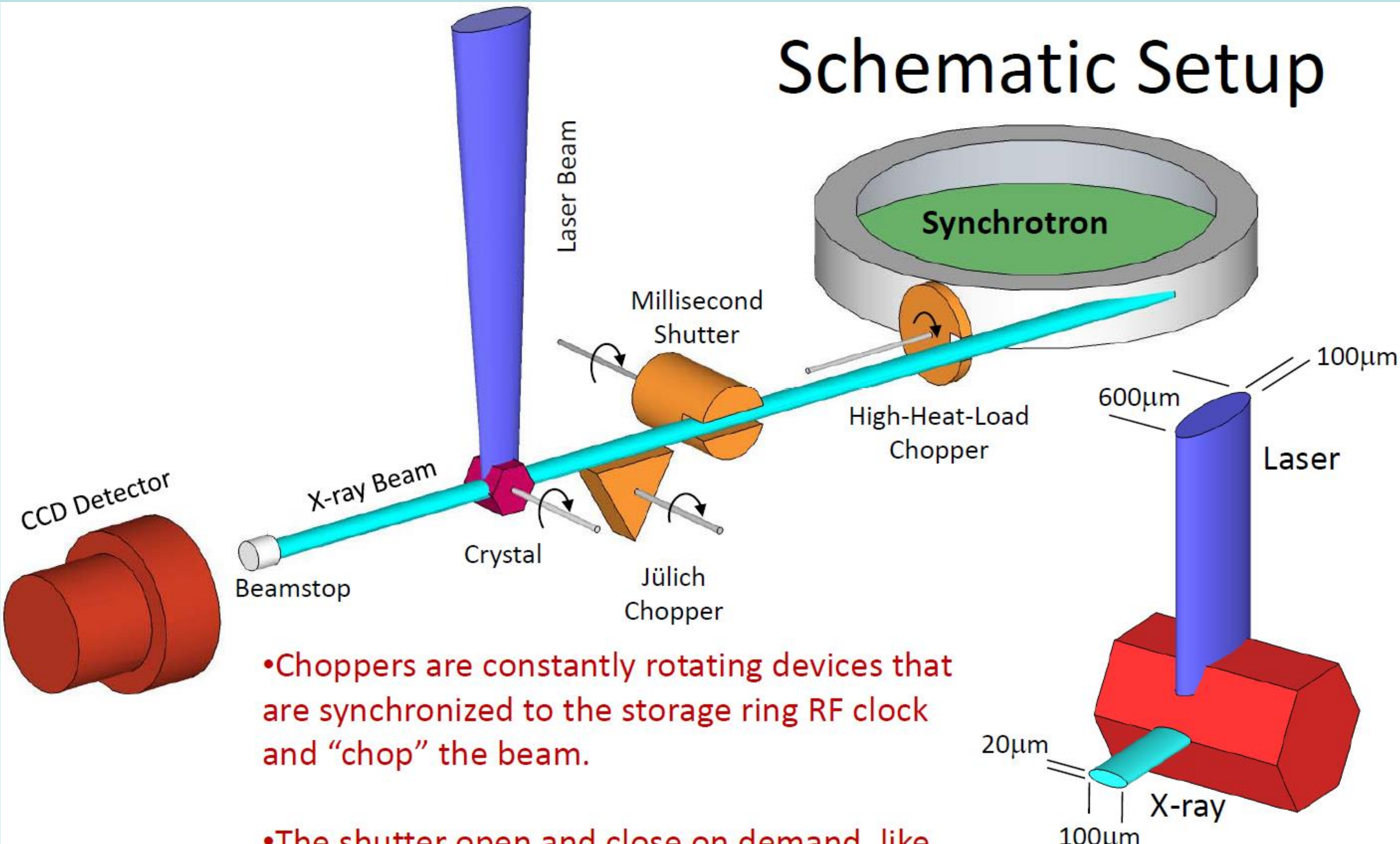
It is based on a directly servomotor-driven triangular rotor with a magnetic bearing and operated in vacuum. Currently its repetition rate is limited to about 900 Hz with an upgrade option to 2700 Hz.



The Jülich chopper. Panel (a) shows a photograph of the chopper with its vacuum flange removed. Panel (b) shows a schematic of its triangular rotor. The open time function, shown in panel (c), has a trapezoidal shape and by changing h, the distance of the X-ray beam from the rotor edge, the opening time can be set to transmit only one (panel d) or several (panel e) X-ray pulses.

Faraday Discuss,122, 13–26 (2002)

Time-resolved X-ray scattering in ID14. APS



- Choppers are constantly rotating devices that are synchronized to the storage ring RF clock and “chop” the beam.

- The shutter open and close on demand, like the shutter on a camera to expose the CCD

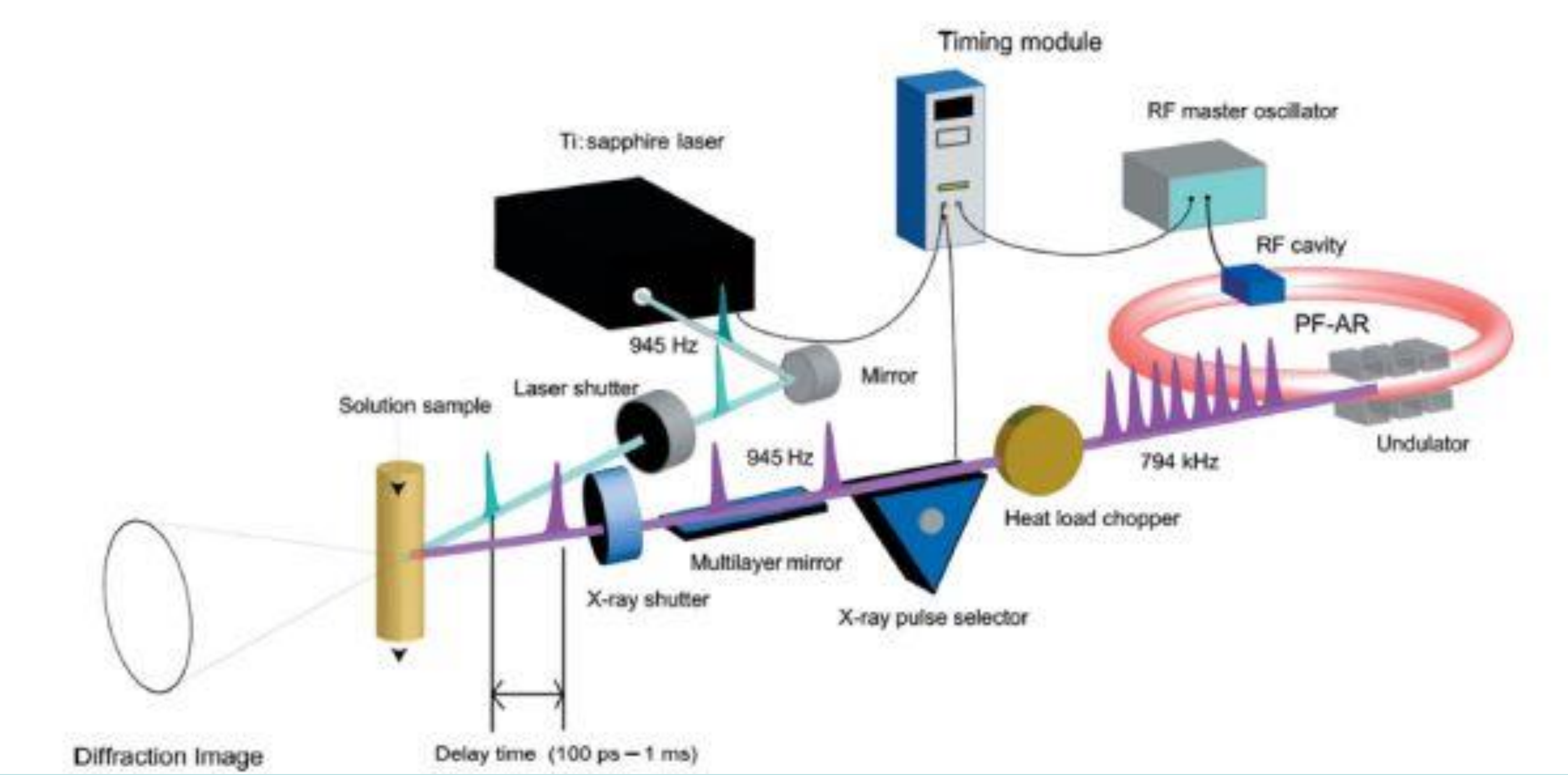
Time-resolved X-ray scattering in ID14 in Advanced Photon Source, USA.

TPS 09A

Temporally Coherent X-ray diffraction

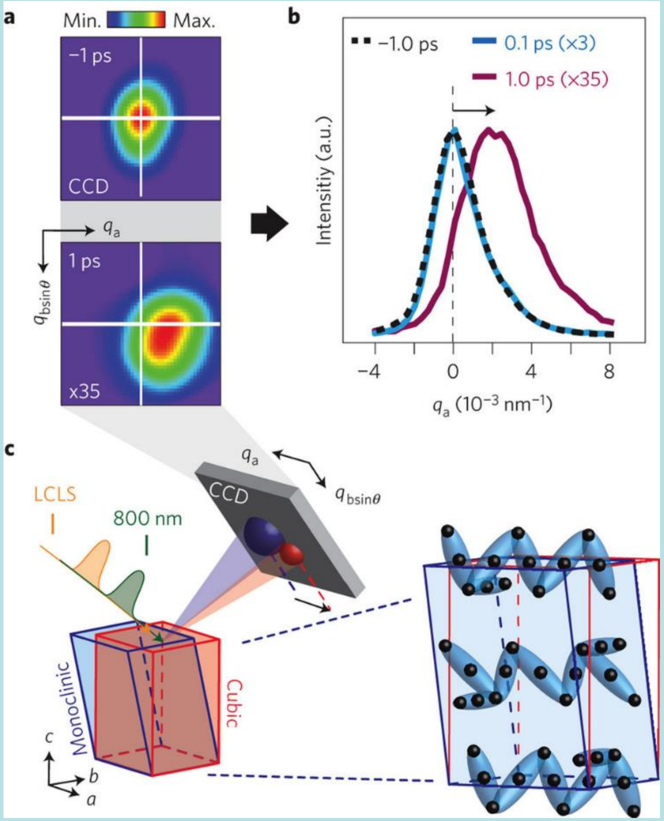
Synchrotron-Based Time-Resolved X-ray Solution Scattering (Liquidography)

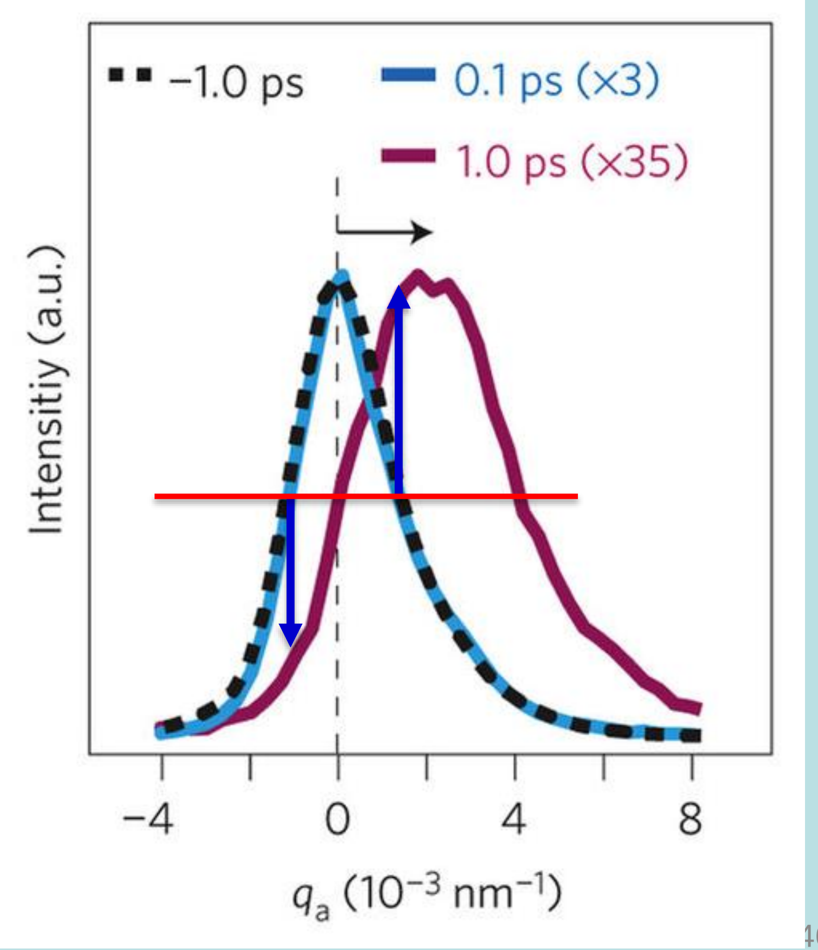
By Shin-ichi Adachi, Jeongho Kim and Hyotcherl Ihee
DOI: 10.5772/8658

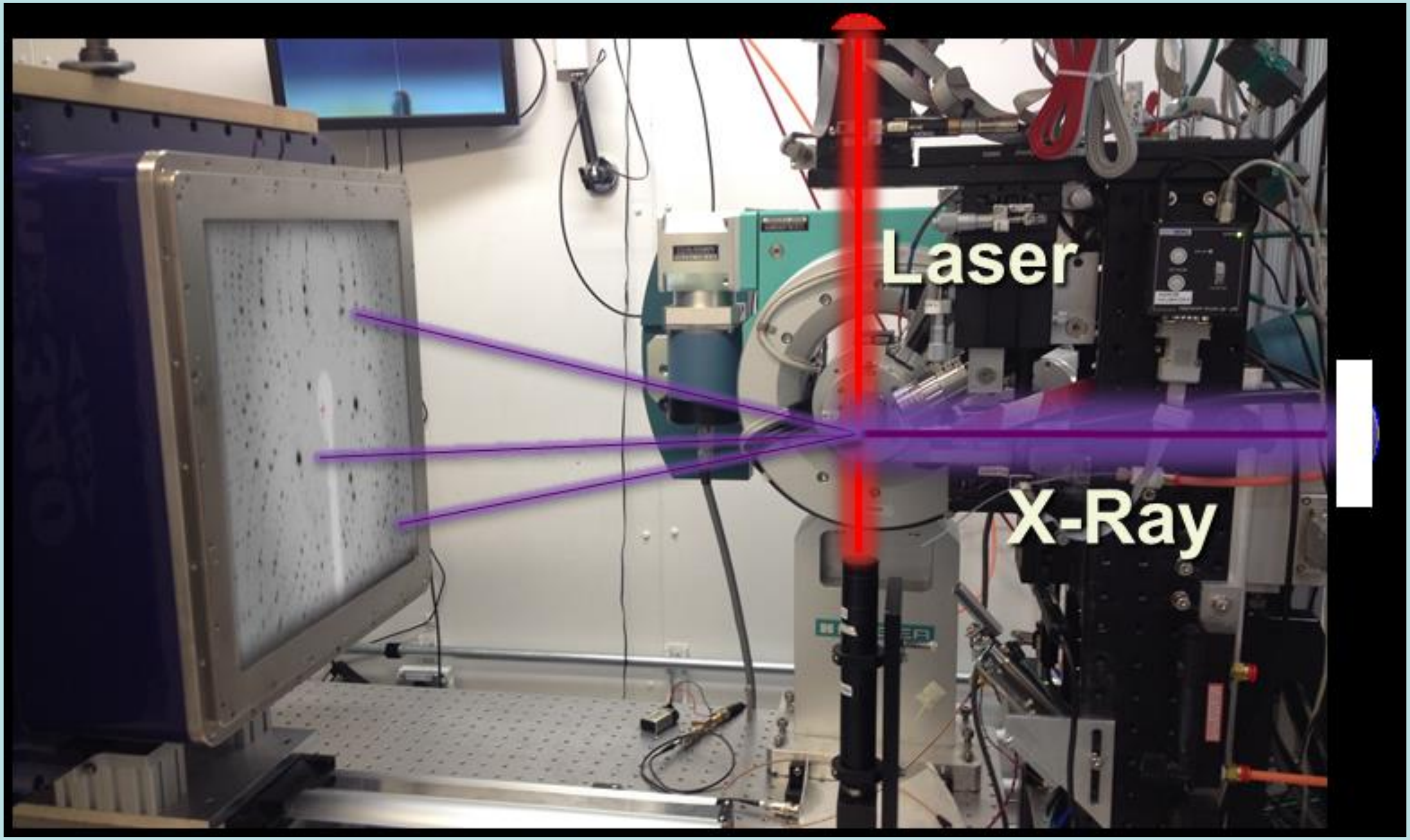


Speed limit of the insulator–metal transition in magnetite

Nature Materials 12, 882–886 (2013)
doi:10.1038/nmat3718

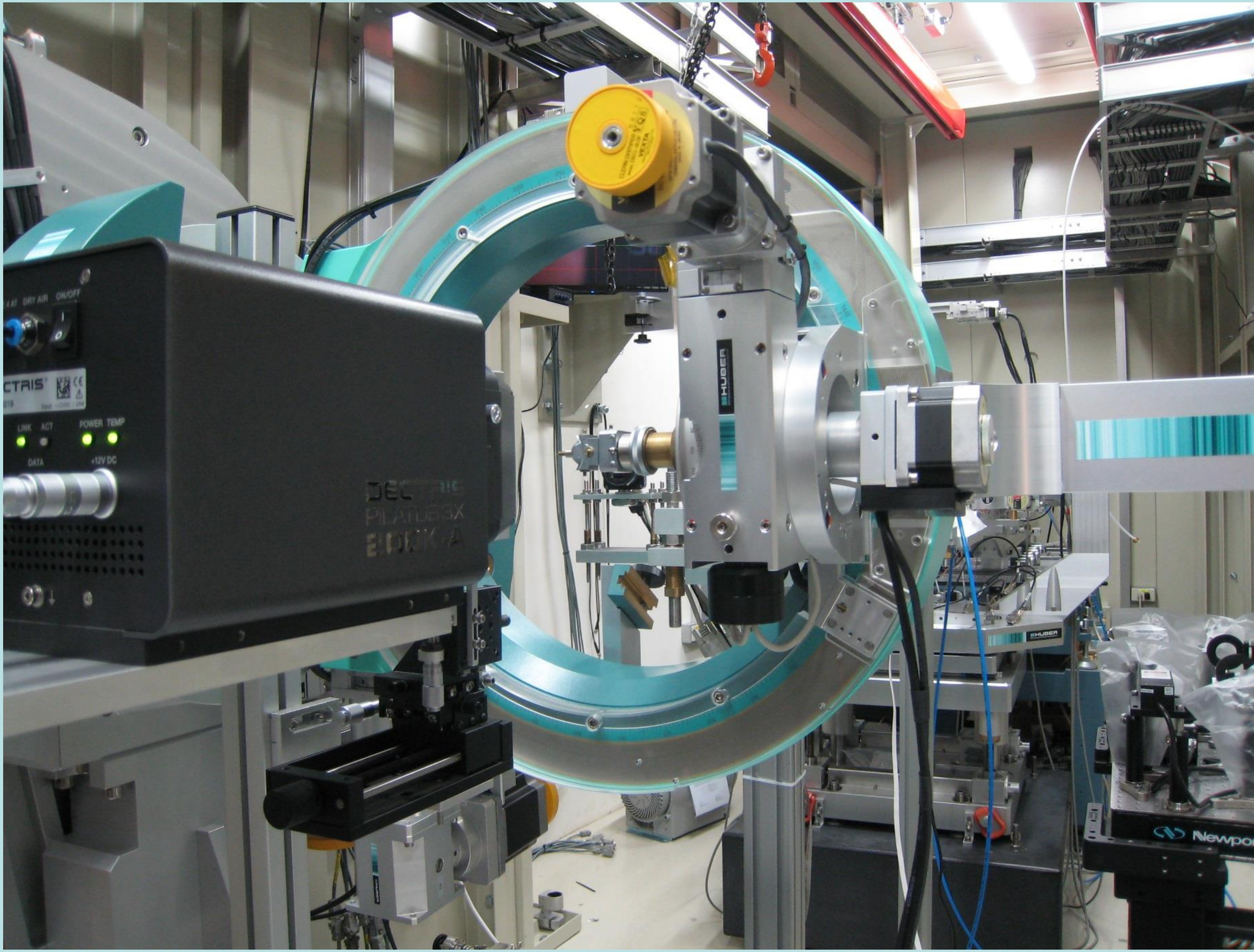






Time-Resolved Research (XSD-TRR) | Advanced Photon Source

Advanced Photon Source - Argonne National Laboratory
14-ID-B is operated as a partnership between BioCARS and XSD and is specialized in ultrafast time resolved techniques such as laser pump high-flux x-ray ...


DRY AIR
ON/OFF
LINK
ACT
POWER
TEMP
DATA
+12V DC
HUBER
Newport