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)

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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

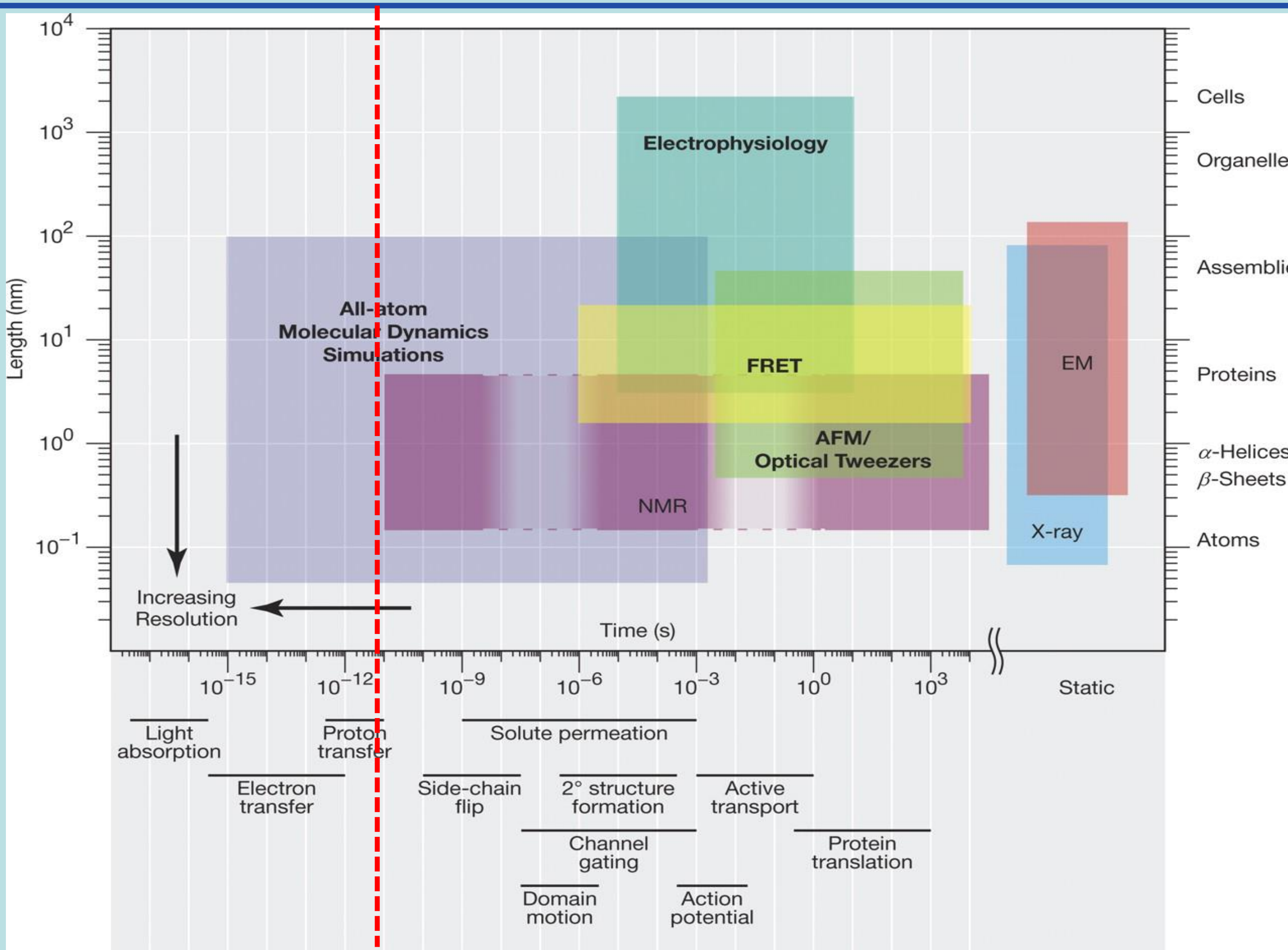
- **Energy, Momentum, Position, Time**
- Time can be applied to all the techniques, in principle
- Some time-resolved experiments take advantage of the pulsed nature of the synchrotron radiation

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.**

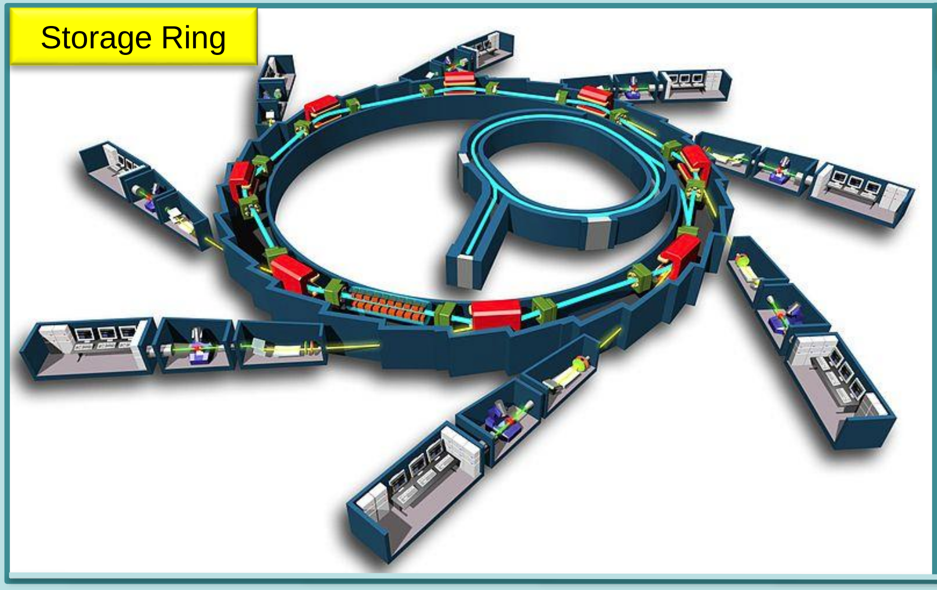
Spatial and Temporal Resolution for Phenomena

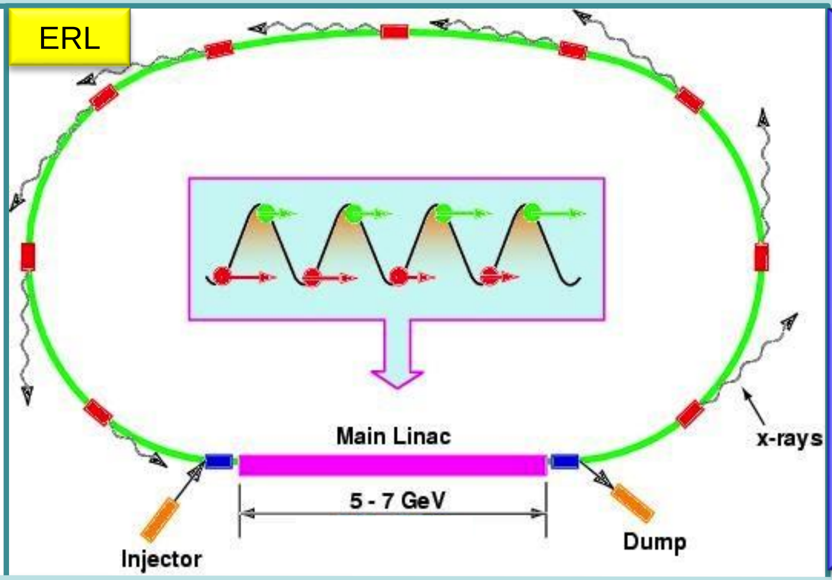


Spatiotemporal resolution of various biophysical techniques. Techniques capable of yielding data on single molecules (as opposed to only on ensembles) are shown in bold. The timescales of some fundamental molecular processes, as well as composite physiological processes, are indicated below the abscissa. 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.

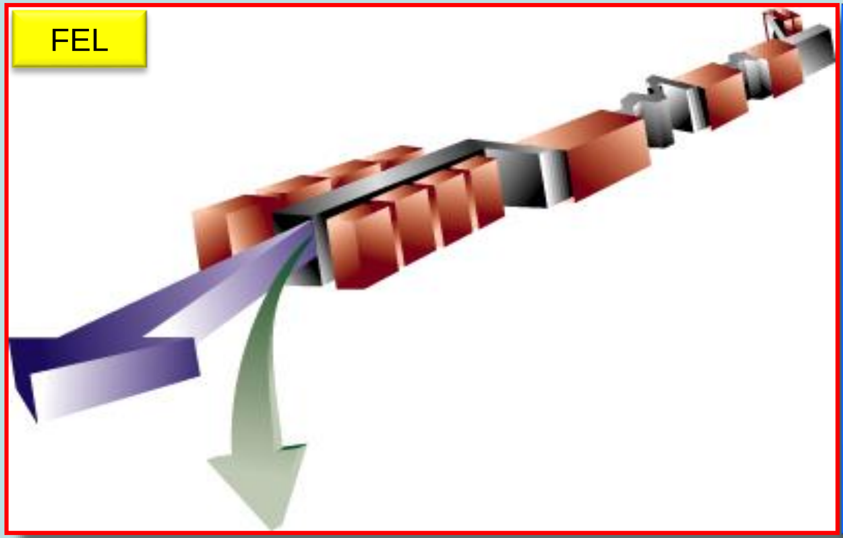
R. O. Dror et al, JG Physiology **135**, 555 (2010)

Light Sources: Storage Ring, ERL, & FEL



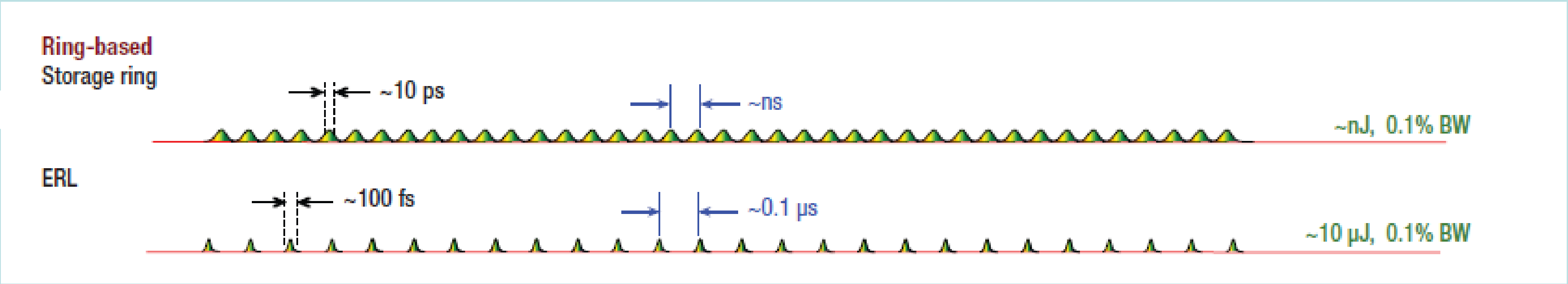


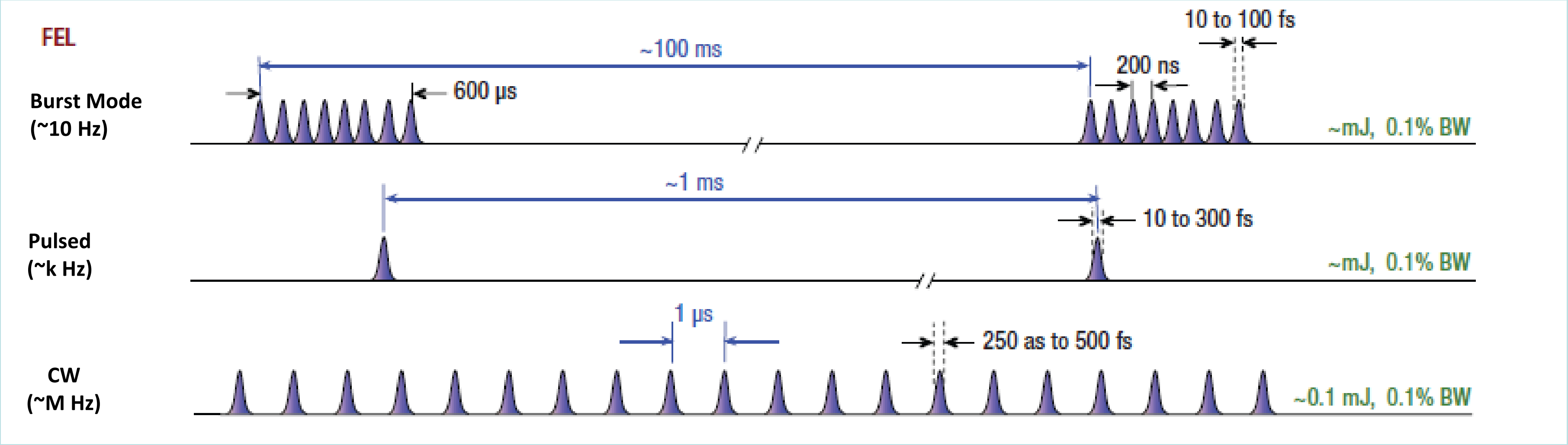
Spontaneous Emission



Stimulated Emission

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

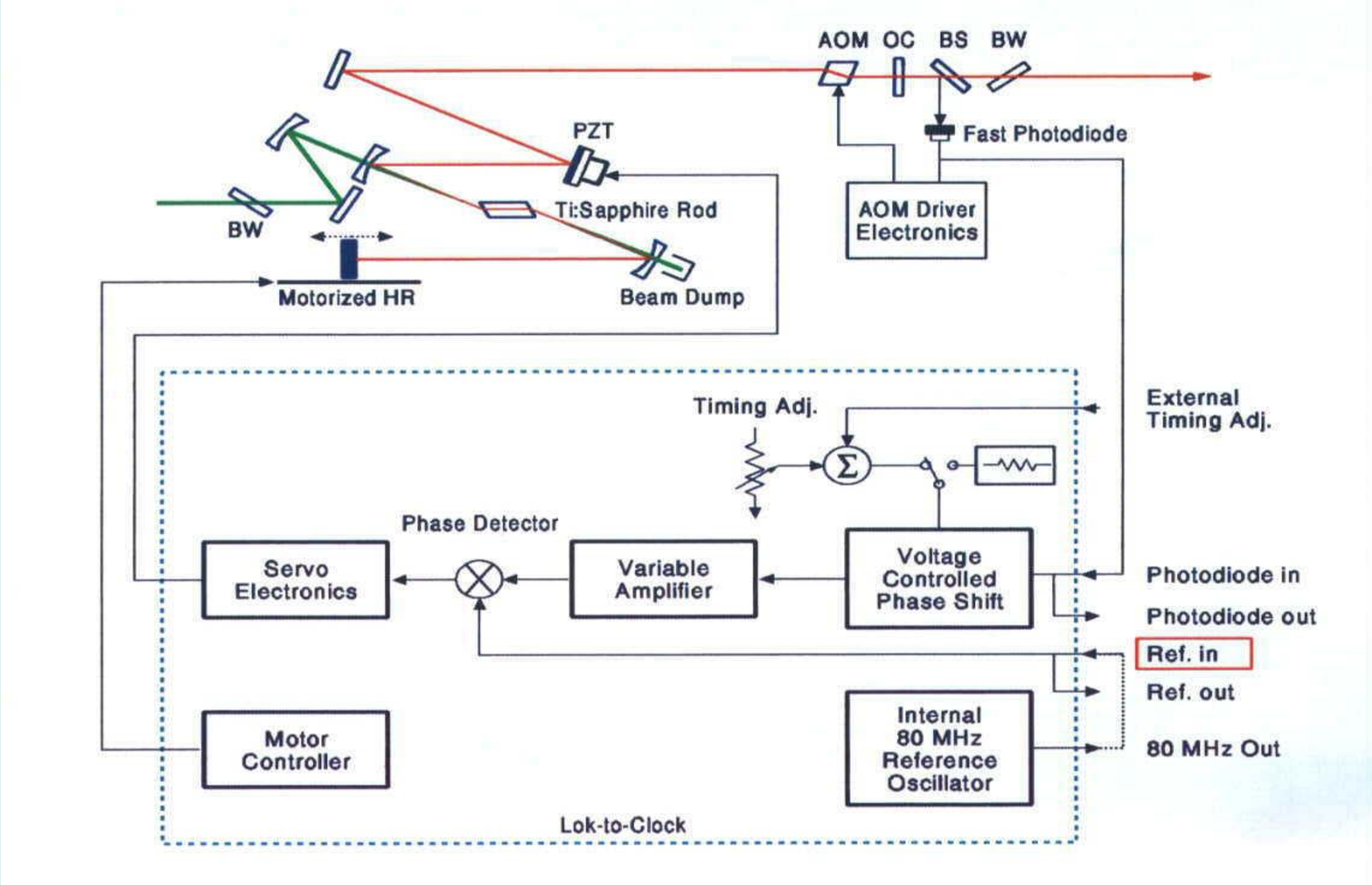
☑ **Time-resolution is limited by the X-ray pulses duration at NSRRC currently.**

1. Temporal width of a synchrotron X-ray pulse: 20-40 ps FWHM
2. Low-α 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

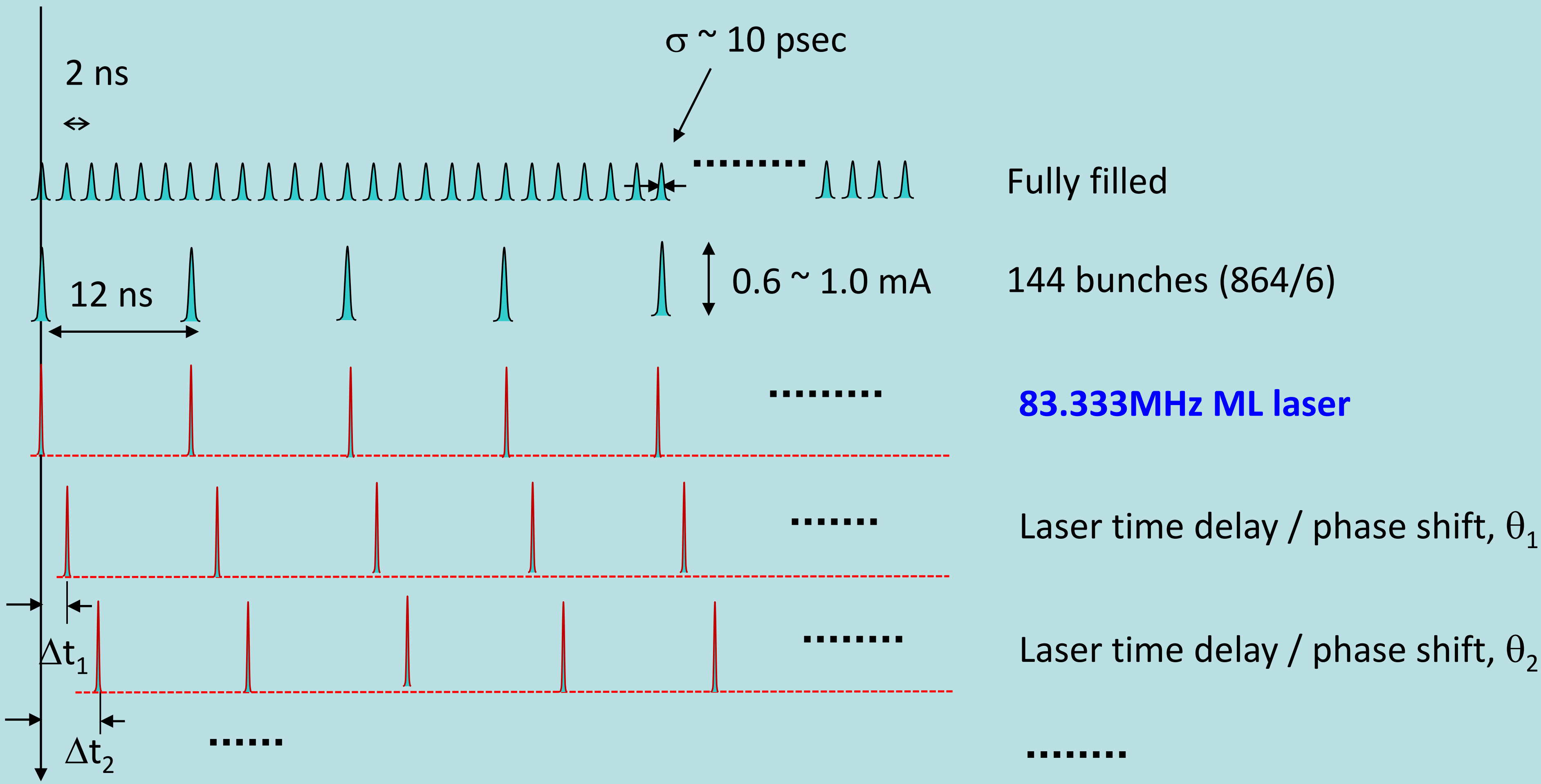


Cavity modes: $\lambda_n = 2L/n$ $\quad\quad$ $\Delta f = c/2L$ *----repetition rate*

Pump-probe experimental scheme I

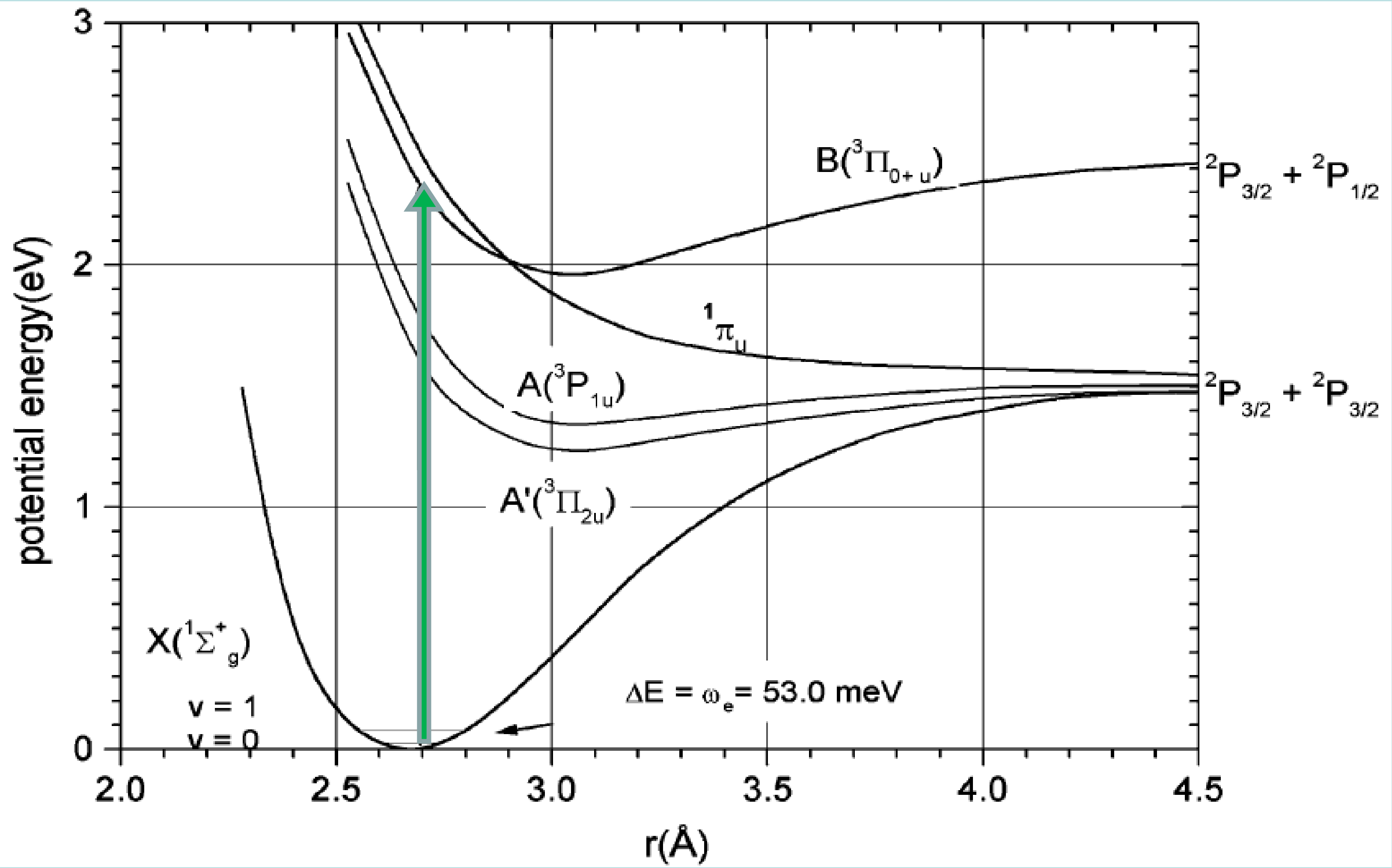
Low peak power, High repetition rate

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 $I_2^* \rightarrow I_2$

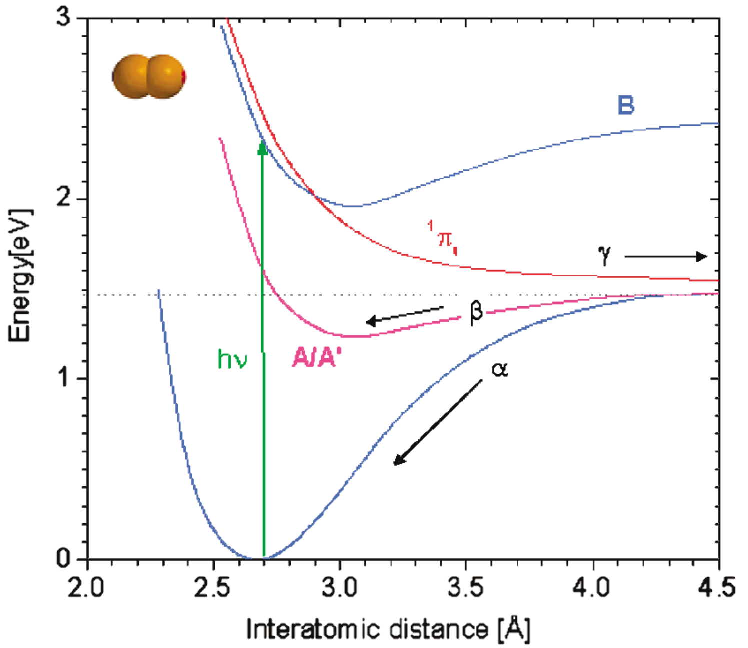


FIG. 1 (color). Low lying electronic energy surfaces of I_2: the states X, A/A', and B are attractive, whereas the state $^1\pi_u$ is repulsive. The processes α, β, and γ denote vibrational cooling along the X potential, geminate recombination through the states A/A', and nongeminate recombination, respectively.

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

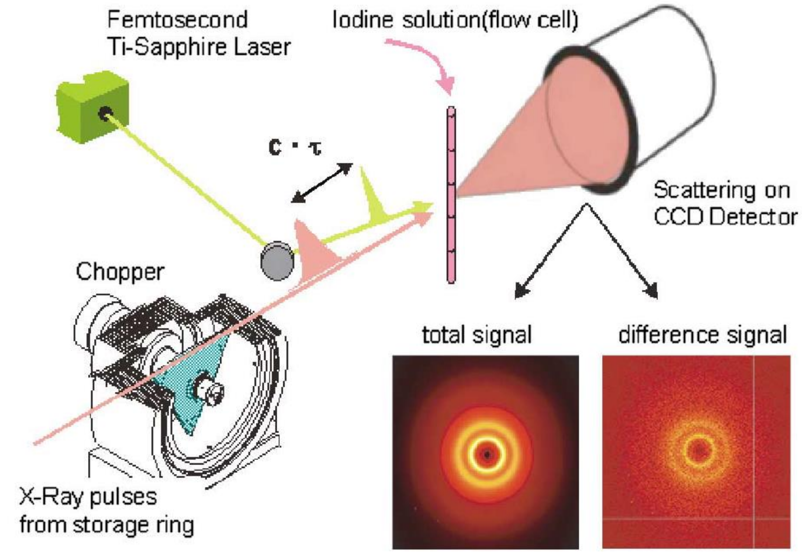


FIG. 2 (color). Experimental setup: the x-ray pulses are generated by an undulator. The spectrum is centered at 0.67 Å (18.5 keV) and its bandwidth width is $d\lambda/\lambda = 0.03$. The flux on the sample is 5×10^8 per pulse and the pulse length is 150 ps. The solution is excited by 150 fs laser pulses 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

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

Magnetic bottle electron time-of-flight spectrometer

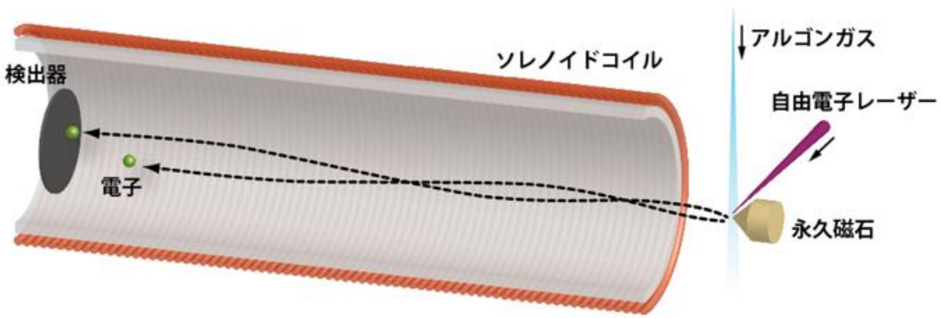
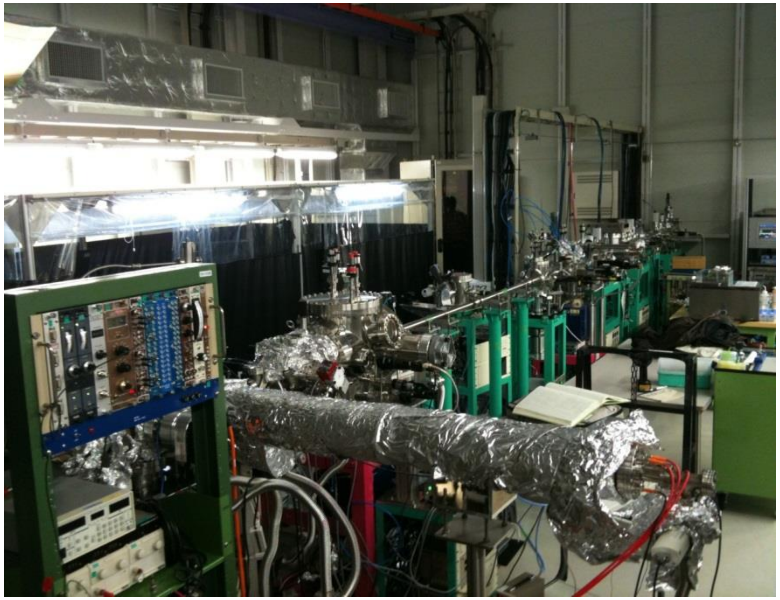


図１　磁気ボトル型光電子分光器

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.

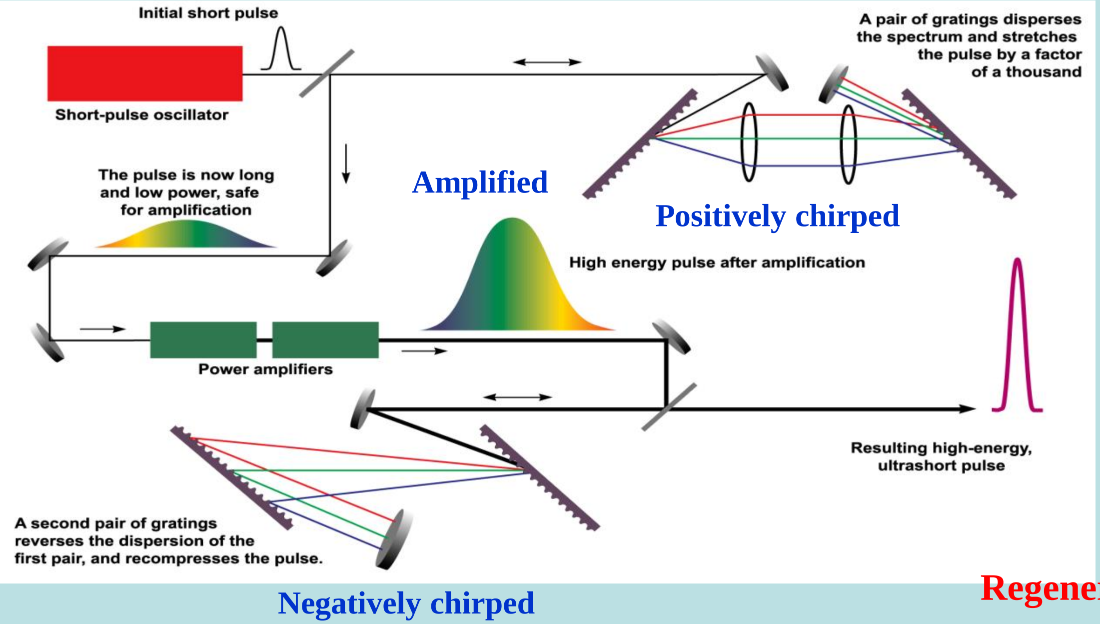
CM Tseng et al./NCTU

Pump-probe experimental scheme II

High peak power, low repetition rate

Mode-locked Laser System

Ti:Sapphire regenerative amplifier



	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

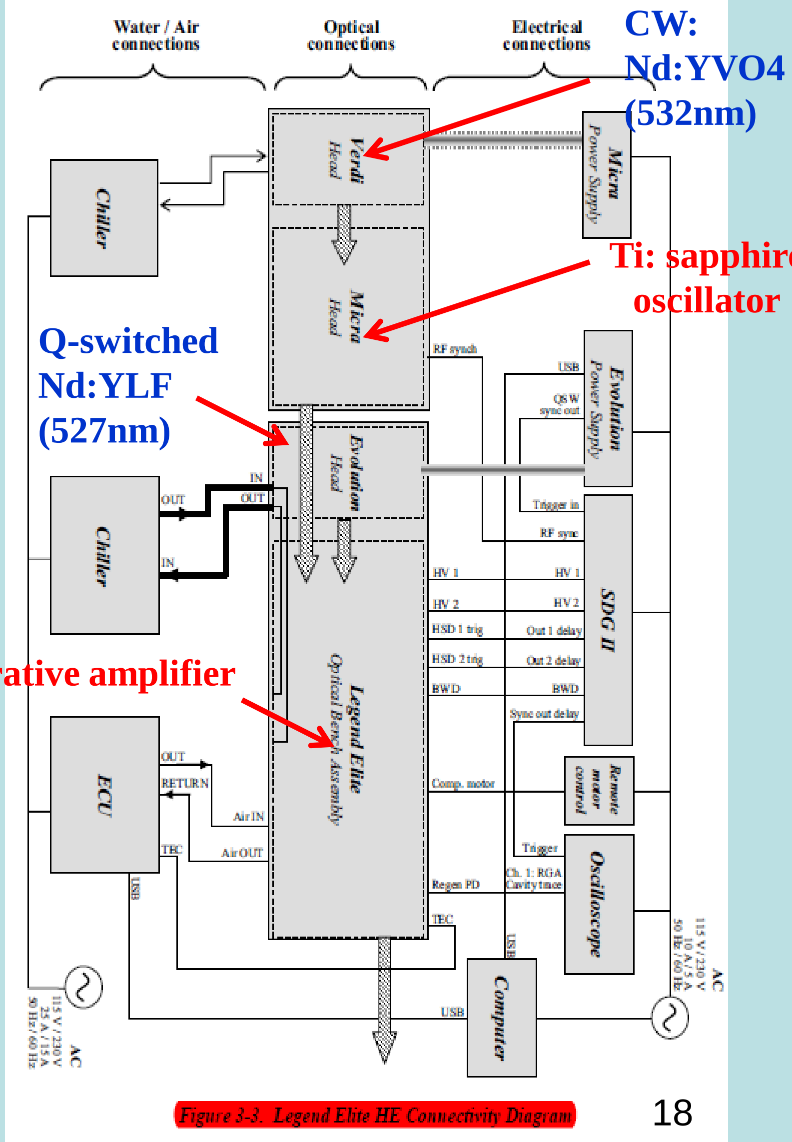
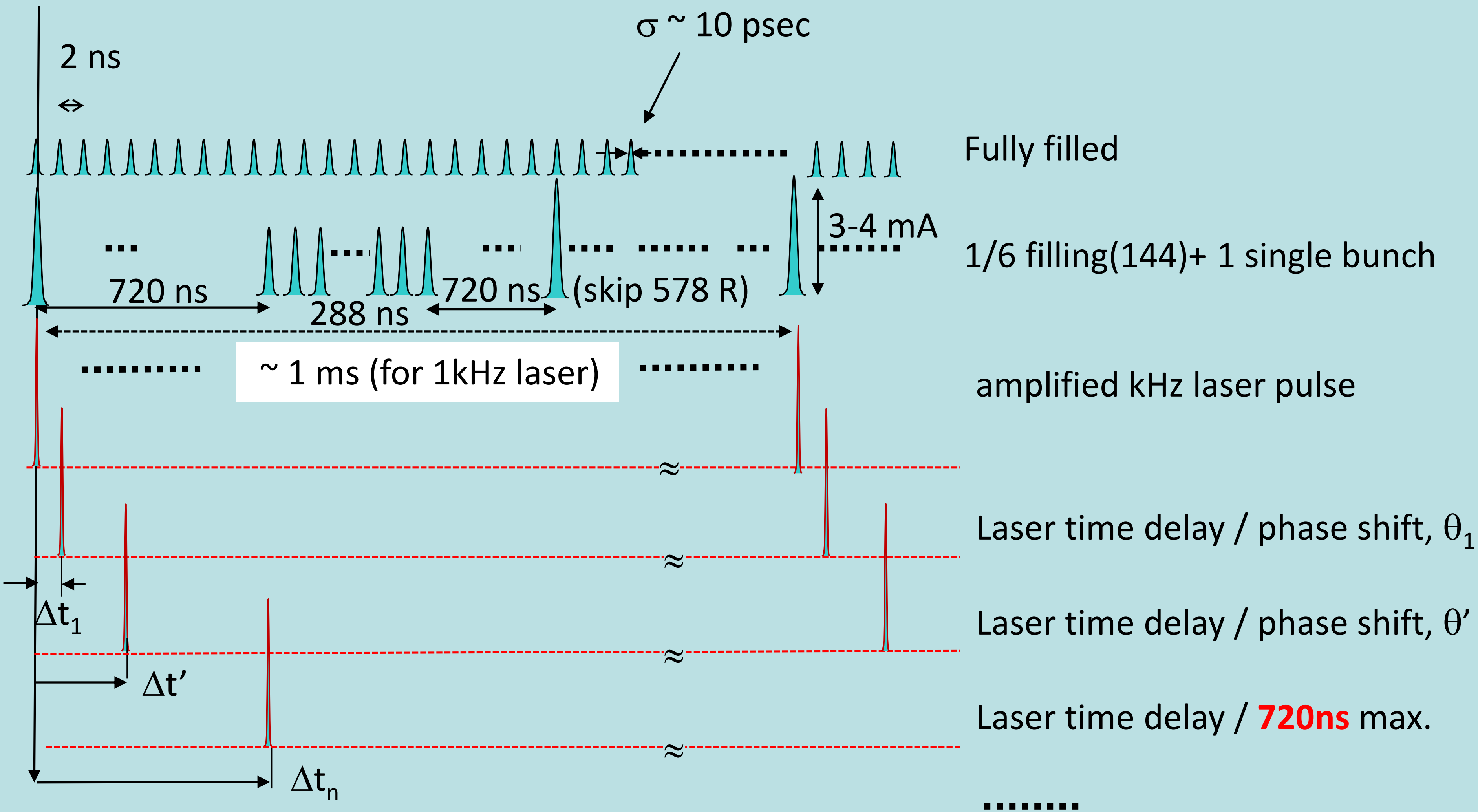


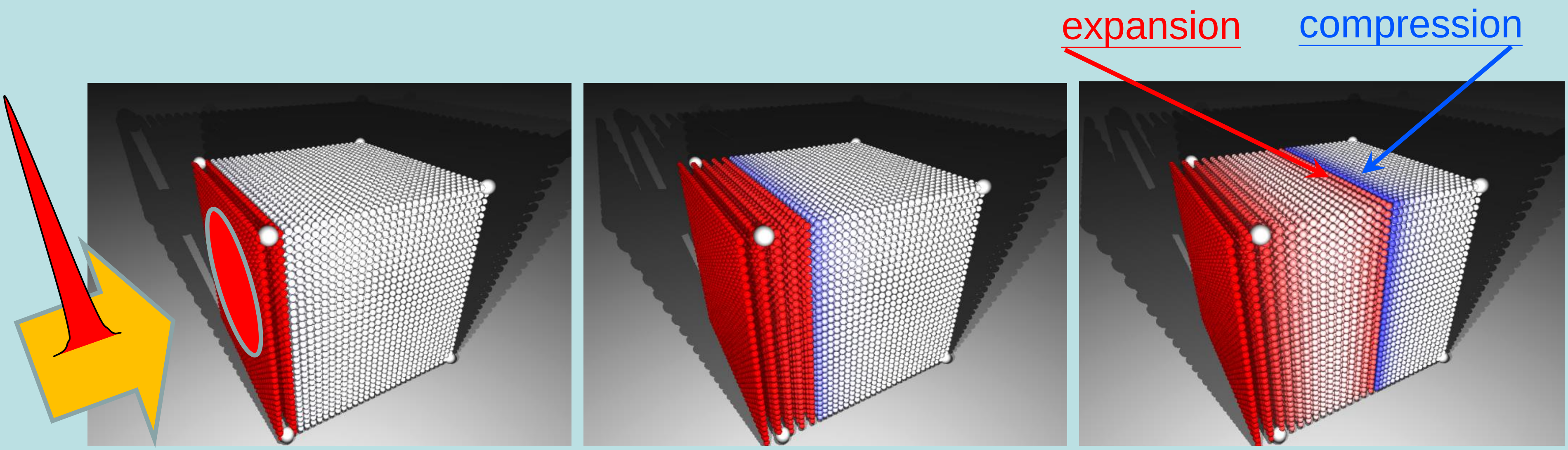
Figure 3-3. Legend Elite HE Connectivity Diagram

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



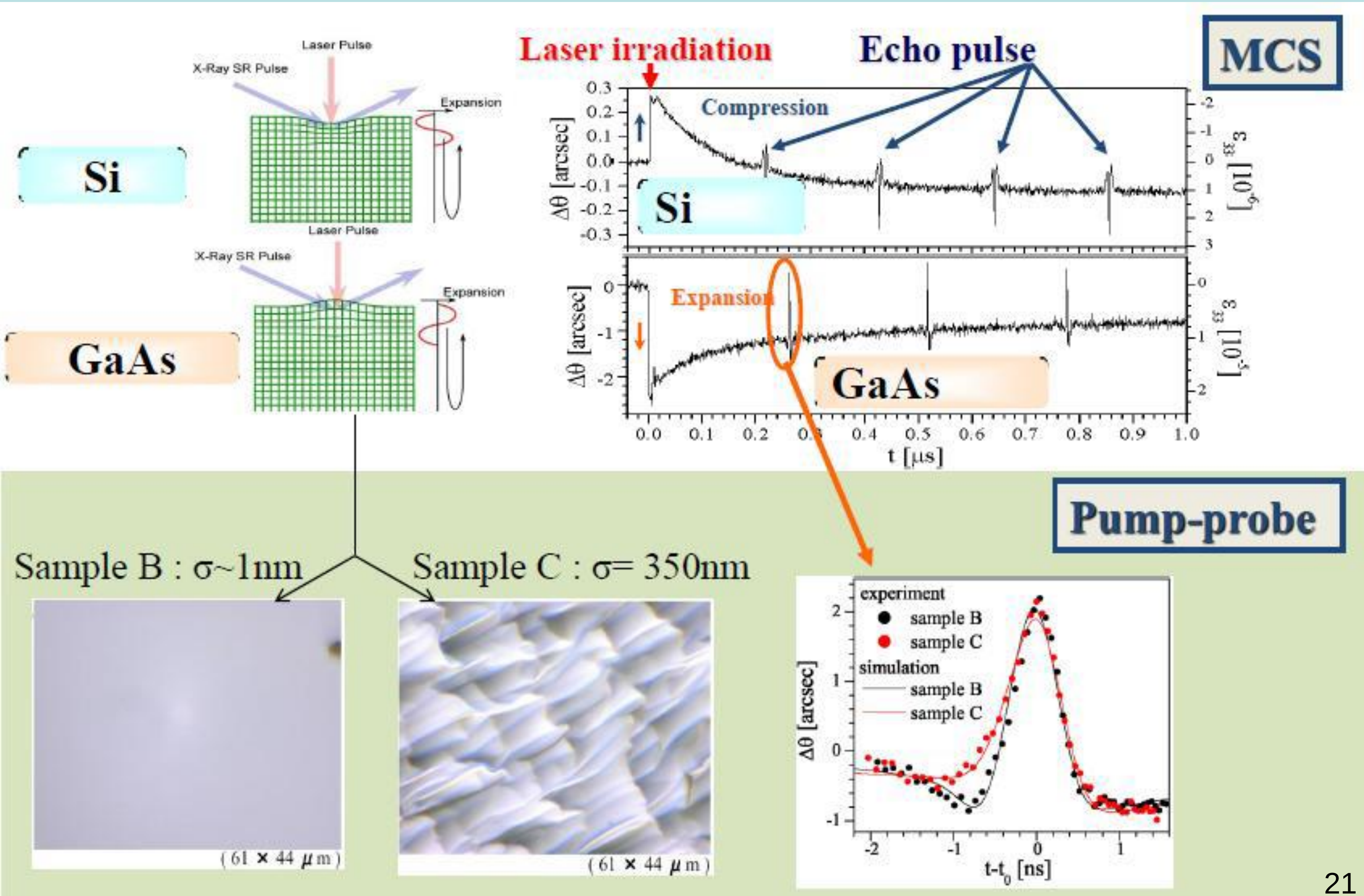
a) **Electrons are excited→ electronic stress due to deformation**
b) **Electrons heat lattice**
c) **Thermal stress**

Stress relaxes by lattice expansion

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

C. W. Siders et al.

Time-resolved XRD

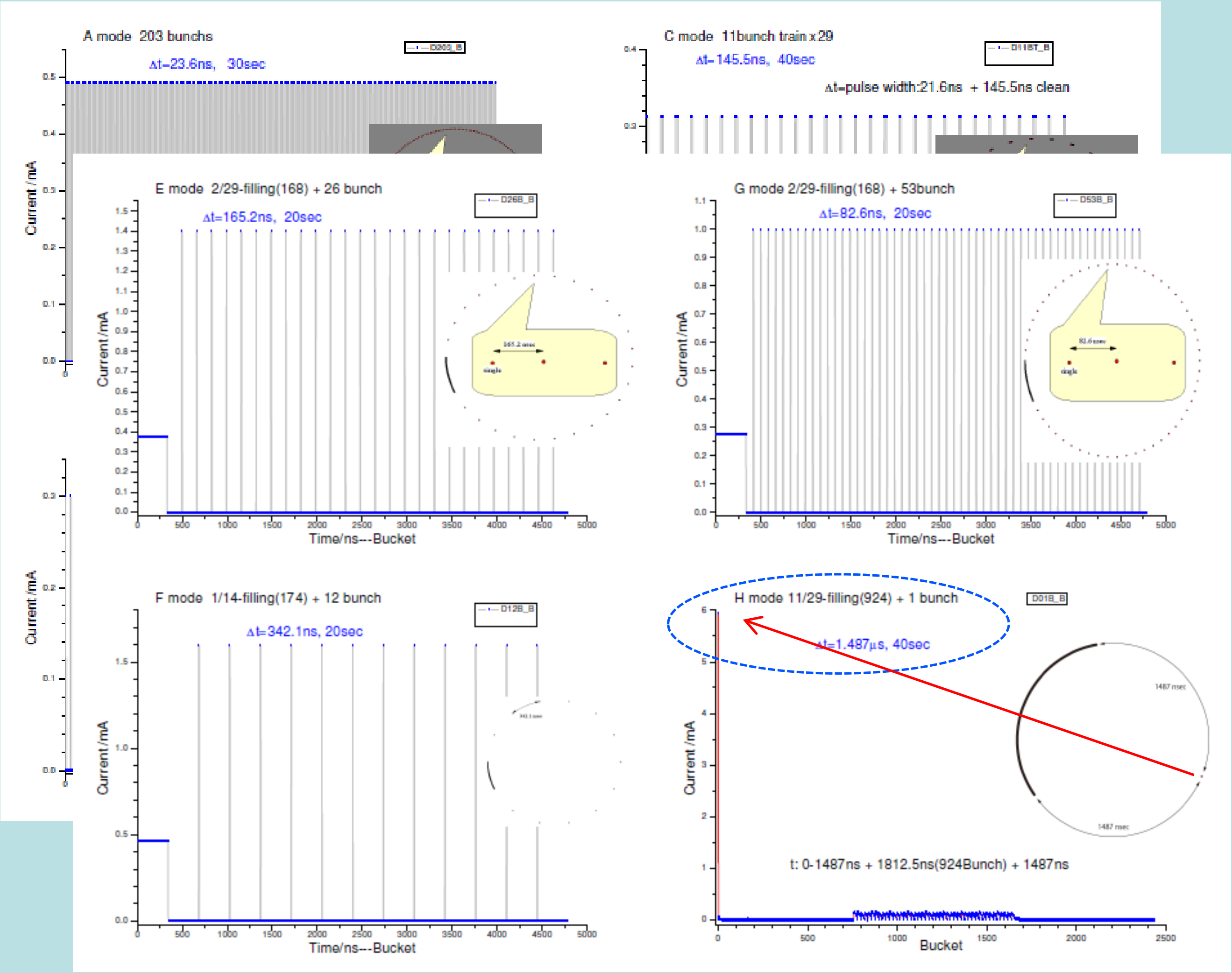


Y. Hayashi, Y. Tanaka, T. Kirimura, N. Tsukuda, E. Kuramoto, T. Ishikawa: Phys. Rev. Lett. 96 (2006) 115505.

SPring-8 experimental facility

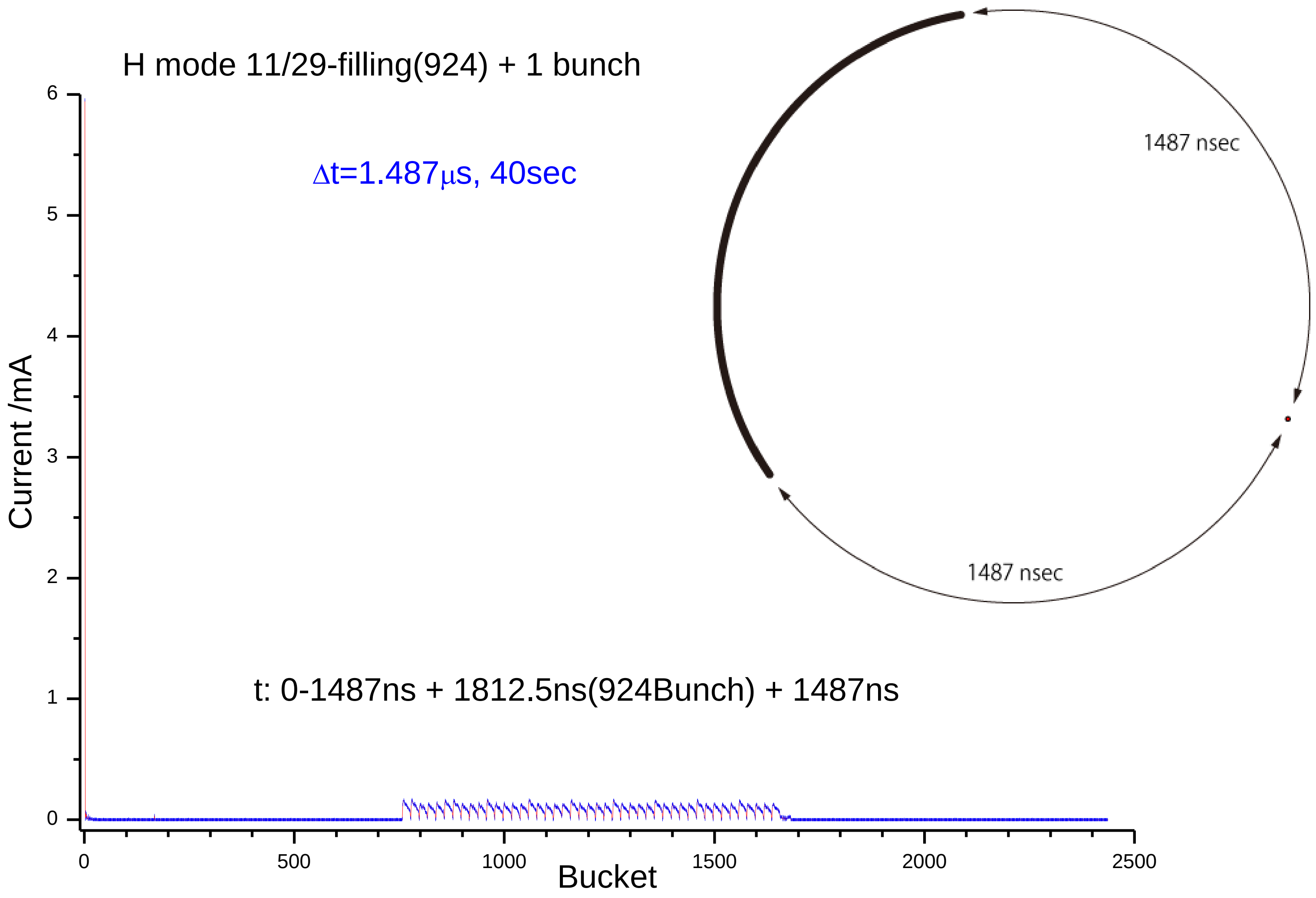
Spring-8 Several Bunch Modes

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



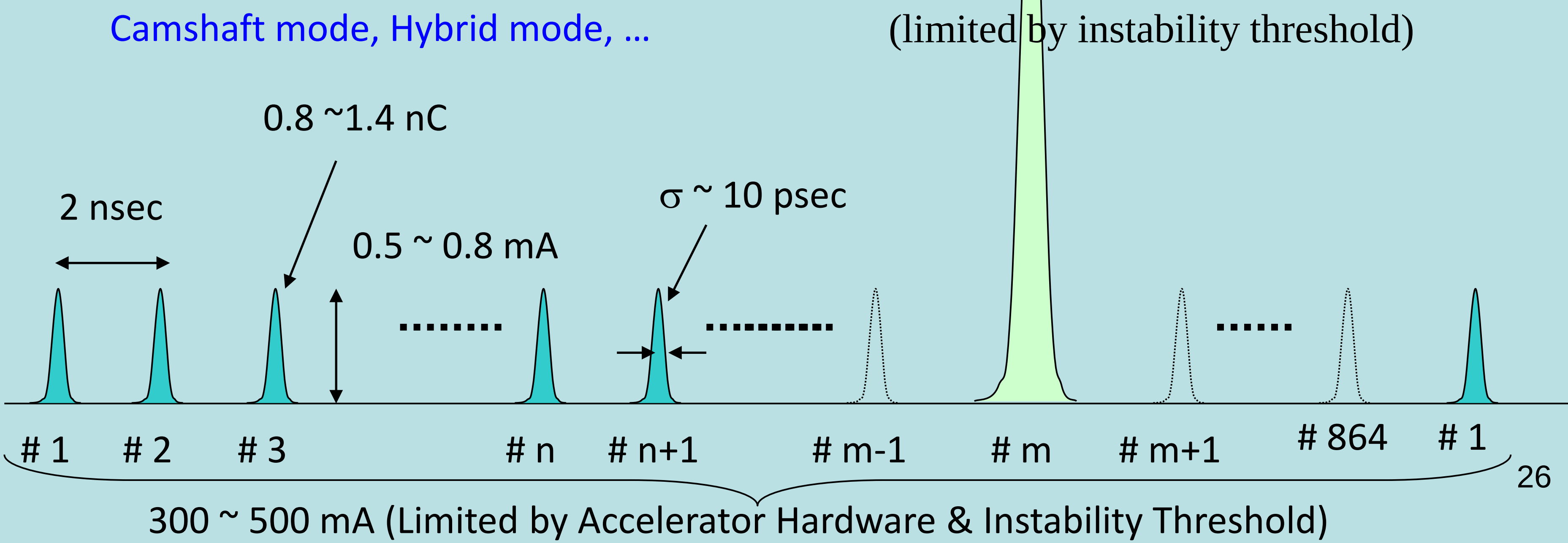
TPS Experimental Capability

TPS Capability

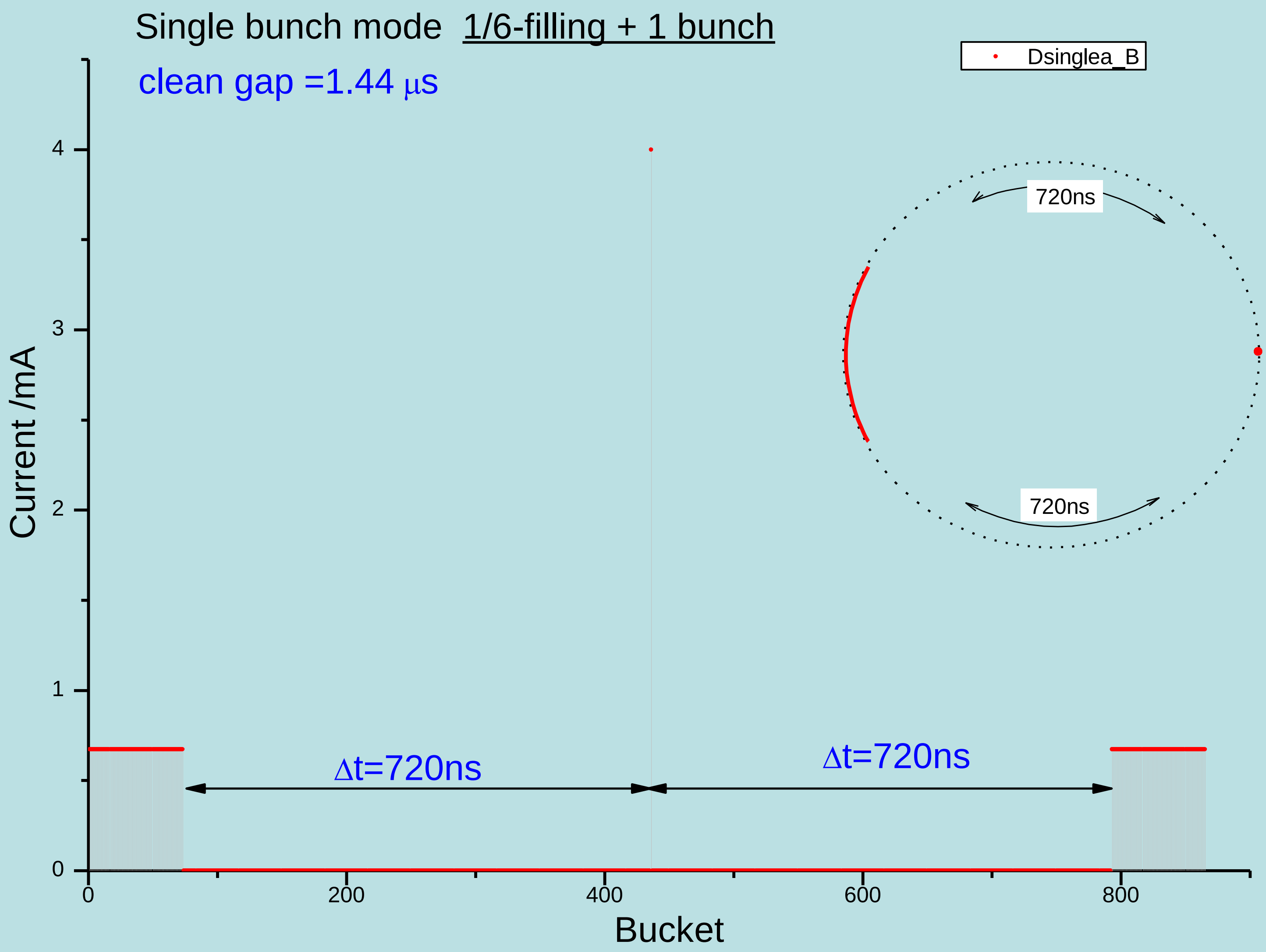
Bunch number: 864
Multi-bunch current: ~ 500 mA (dependent on available RF power)
Multi-bunch bunch current: < 1 mA/bunch
Single-bunch (isolated bunch?) current: 2 ~ 3 mA
Hybrid mode (camshaft mode)

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

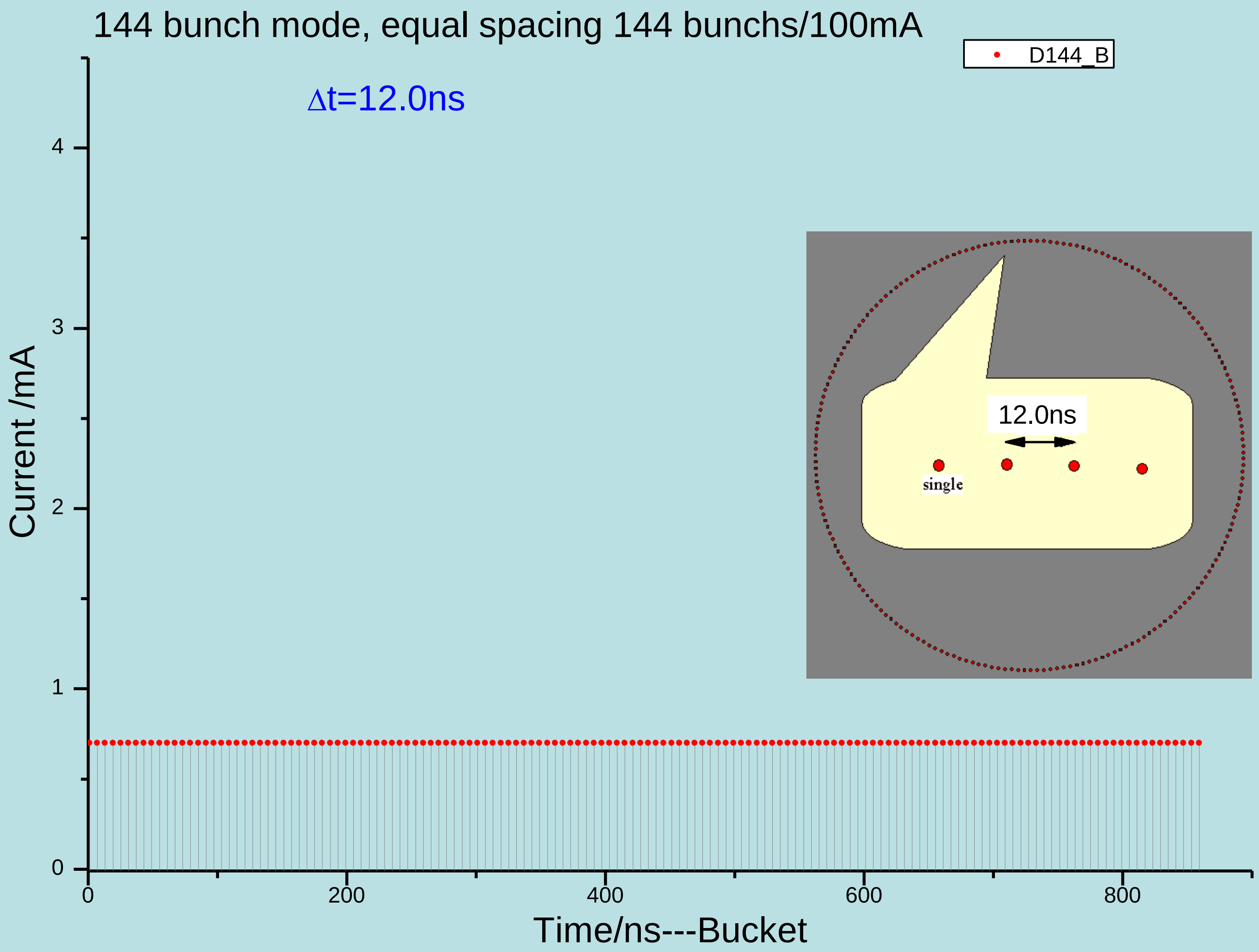
Camshaft mode, Hybrid mode, …



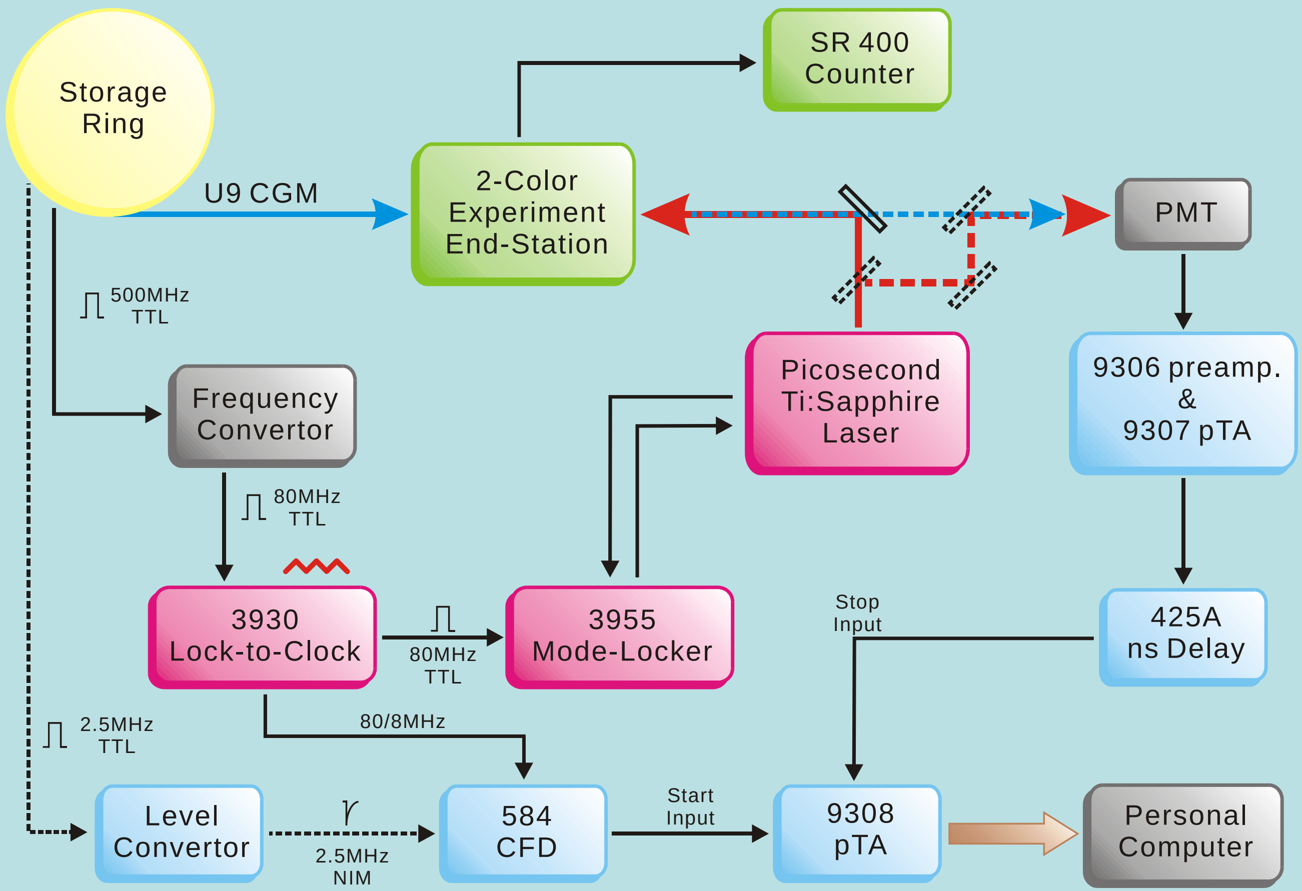
Single bunch-mode : 1/6-filling(144) + 1 bunch /TPS



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

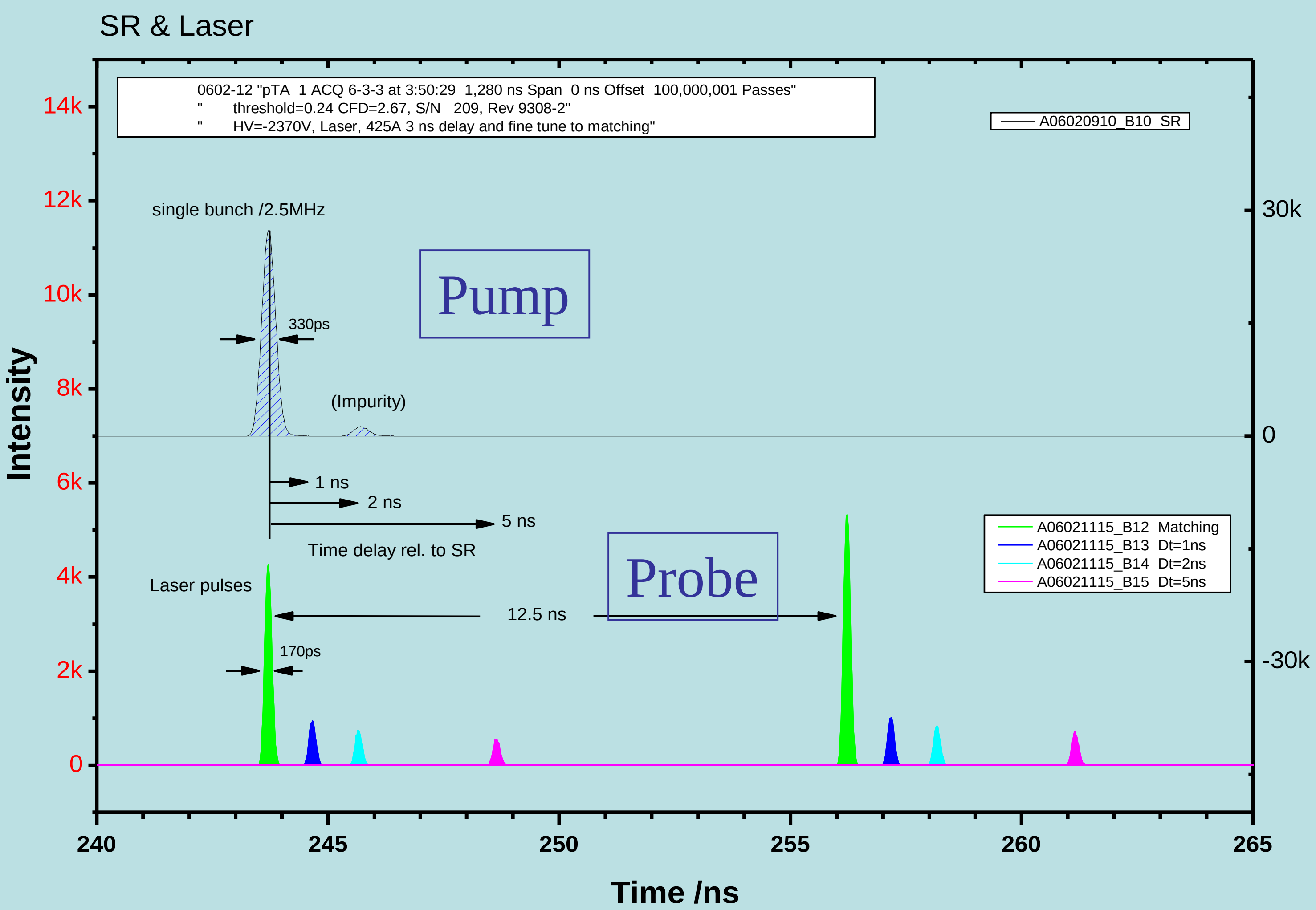


Setup for timing control and measurement system



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. The timing jitter between the synchronized laser and SR systems is less than 50 ps.

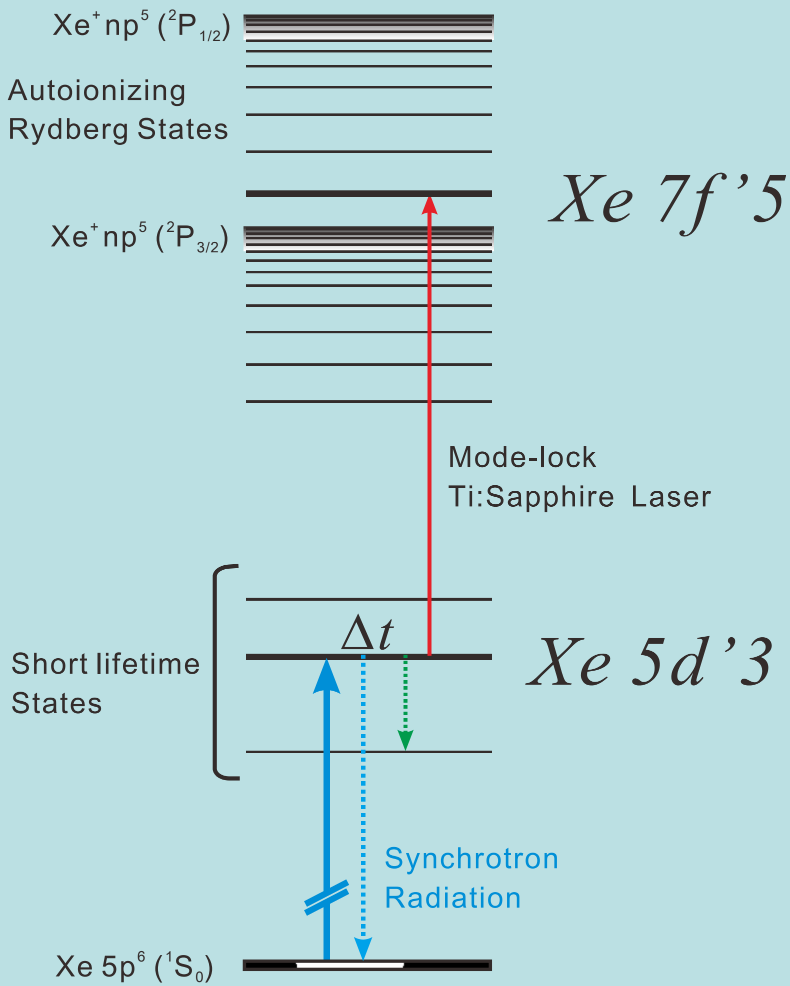
Pump-Probe scheme of SR and laser combination

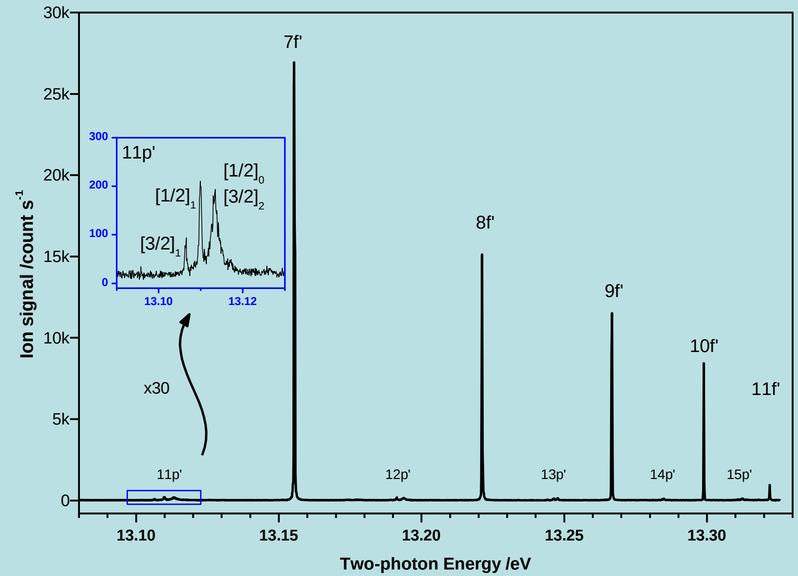


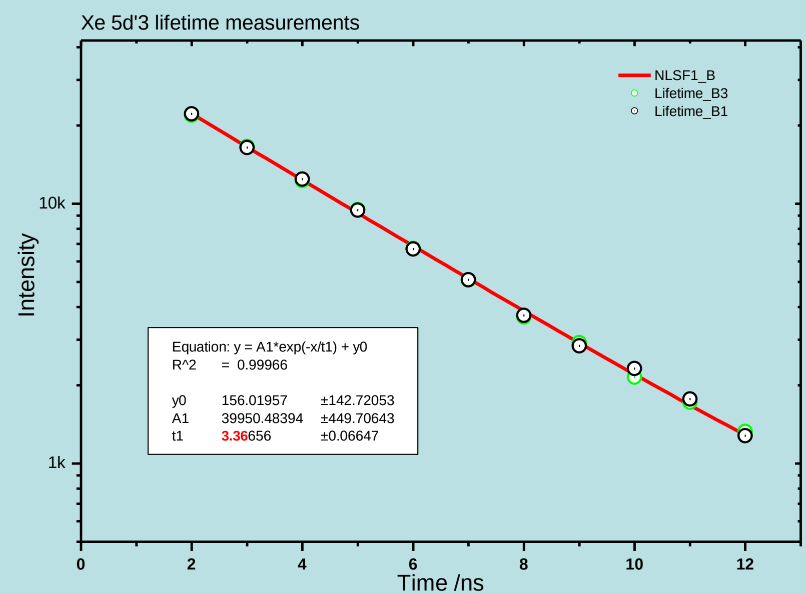
Hybrid bunch modes to extend the clean gap

Two-color Pump-Probe Experiments:

Atomic Xe Spectroscopy and lifetime measurement







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

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

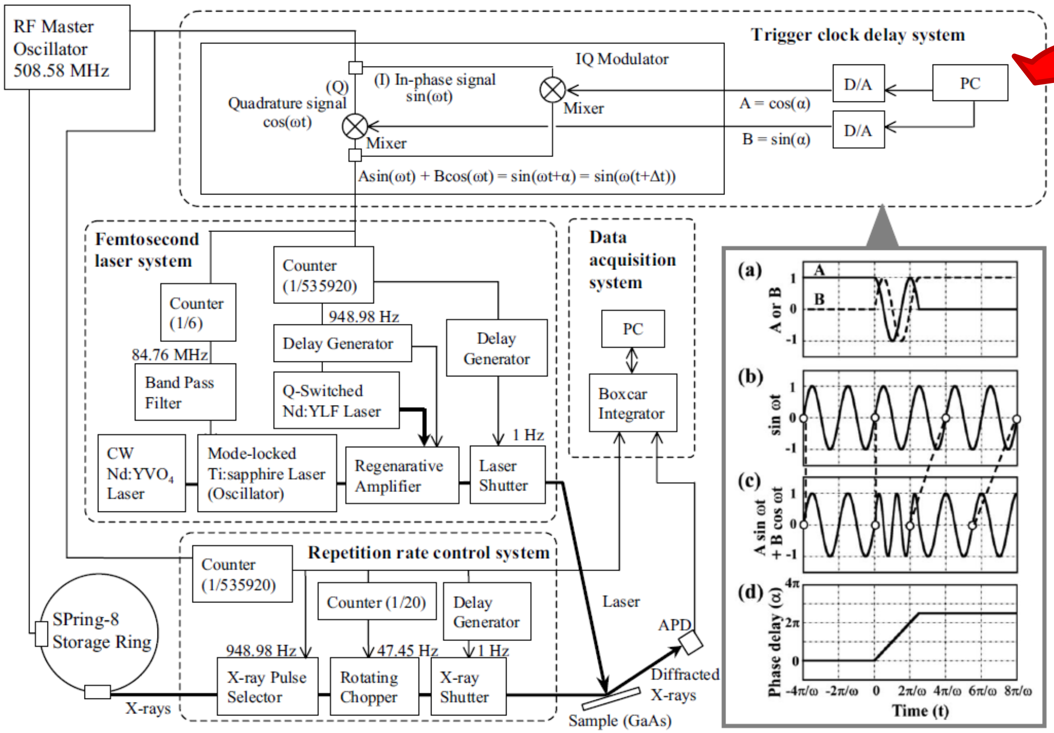


FIG. 1. Block diagram of the trigger clock delay system, the femtosecond laser system, the repetition rate control system, and the data acquisition system. The inset gives the schematic graphs to explain the principle of the continuous phase change at the trigger clock delay system. When the control 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.

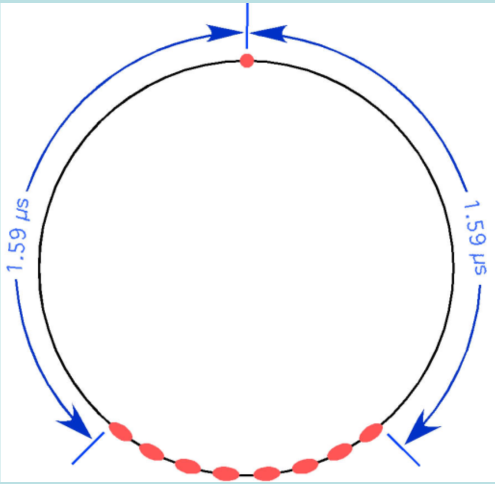
Trigger and Clock delay units/*SPring-8*

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

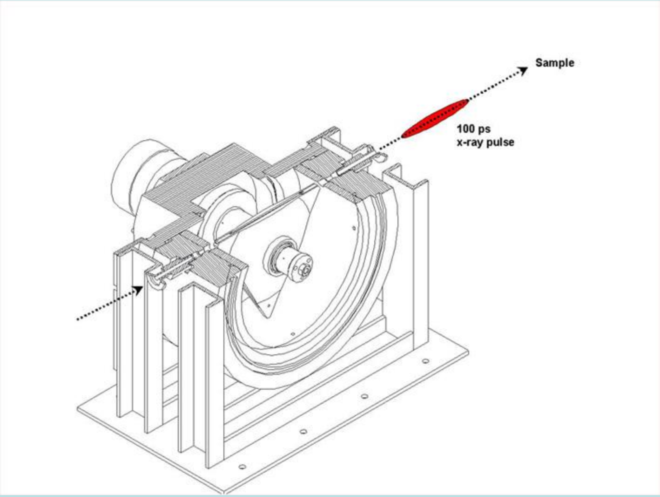
Y. Fukuyama *etal*, RSI 79, 45107 (2008)

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)

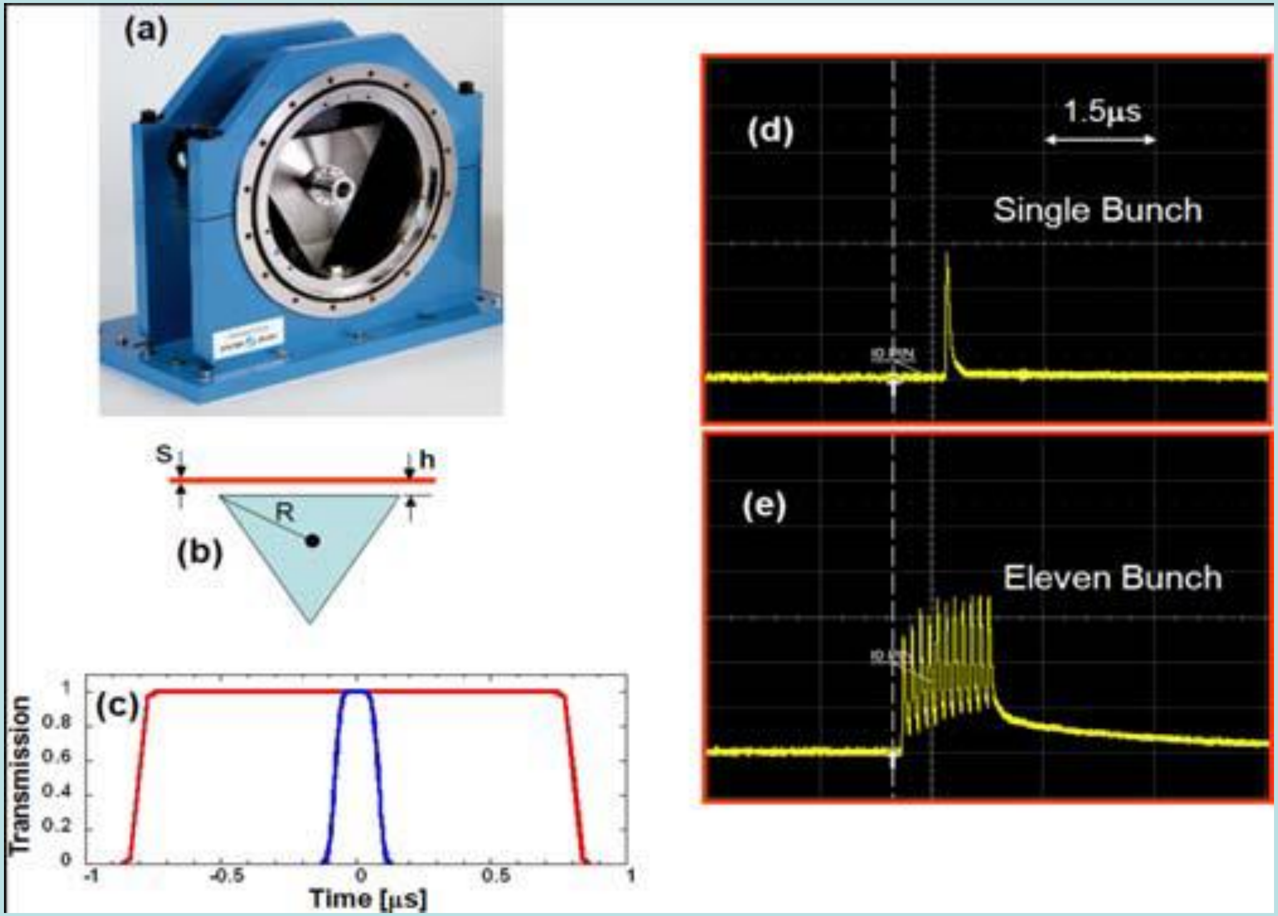


Julich rotating chopper
– In-vacuum triangular Ti rotor with beam tunnel
– maximum radius 96.8 mm, tunnel length 165 mm
– Magnetic bearing running from 10 to 900 Hz
– Mono and white beam compatible to 40 keV

APS

Chopper/Shutter

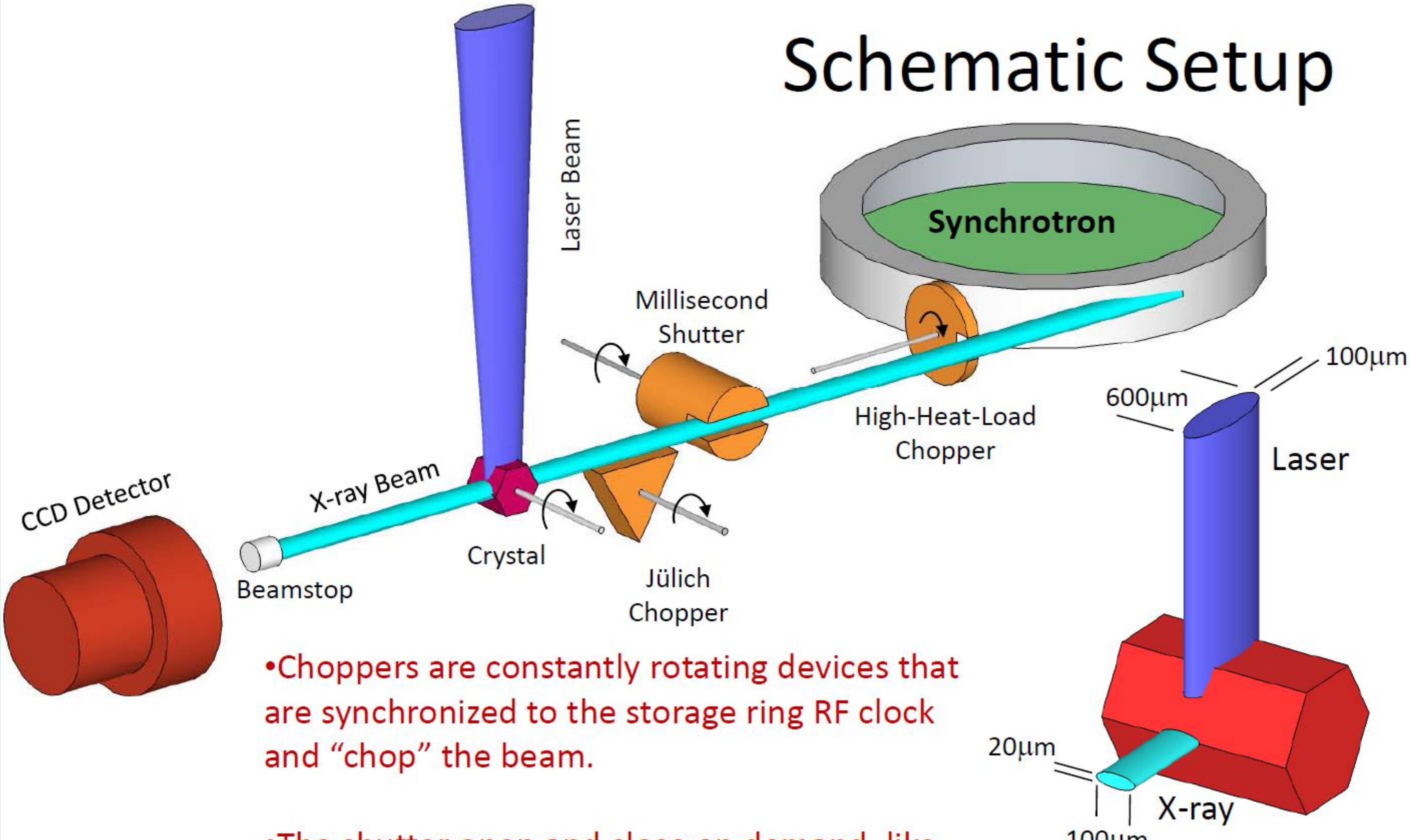
It is based on a directly servomotor-driven triangular rotor with a magnetic bearing and operated in vacuum. Currently its repetition rate is limited to about 900 Hz with an upgrade option to 2700 Hz.



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

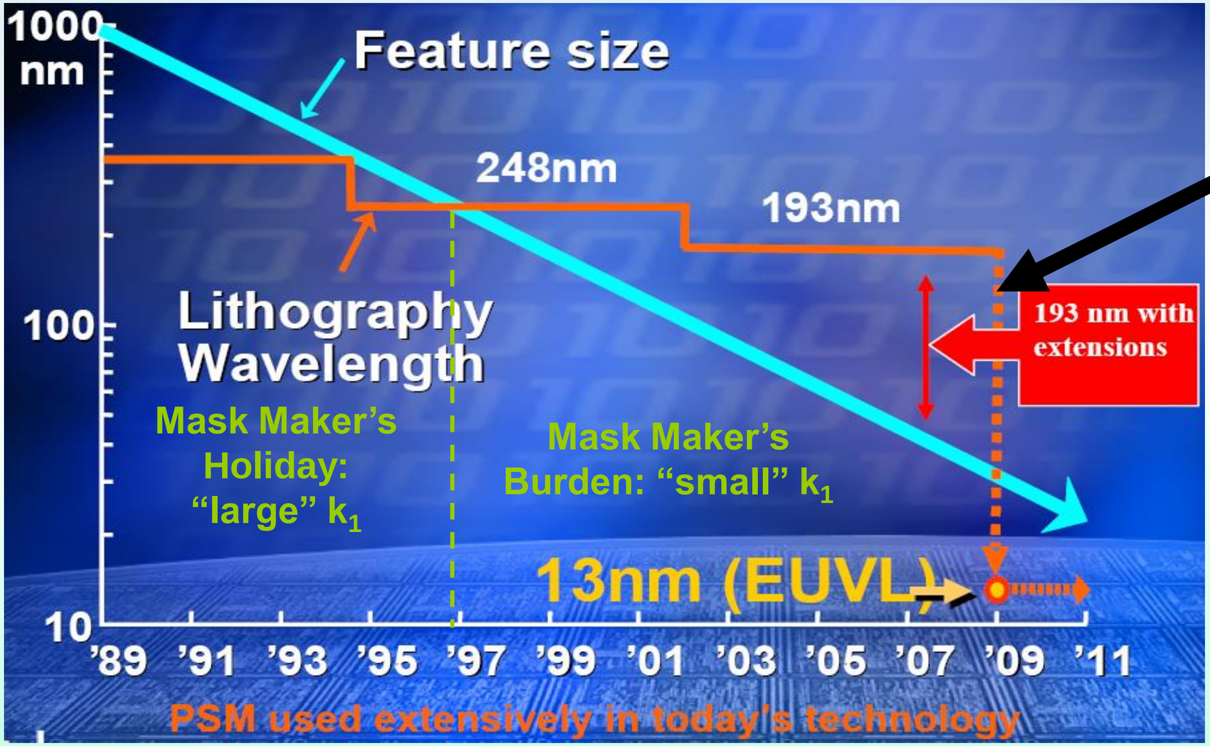
Time-resolved X-ray scattering in ID14 in Advanced Photon Source, USA.

Semiconductor Lithography

Semiconductor Lithography

"Transitions to EUV is a big jump"

Big jump from 193 to ~13 nm. Before this has about ¼ increase in energy. Now >10x



There are only so many "tricks" to increase this gap, and they are very expensive … we must go to a shorter wavelength!

- Phase shift mask Tech.
  (Multiple patterning)
- Immersion lithography

In 2004, it is predicted that EUVL will become mass production tool in 2009. Today it is believed that DUV lithography (λ=193nm) with double processing will be used for 32nm generation production, thus delaying the need for EUV.

EUV 機台三大模組功能--ASML

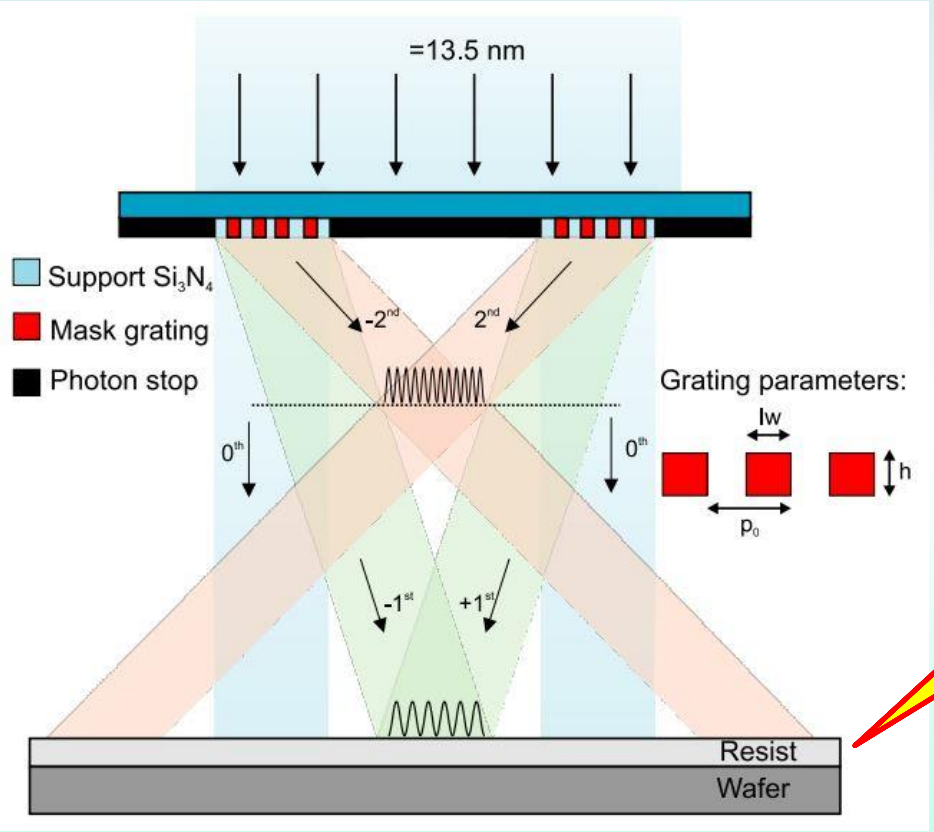
- 照明光學模組—Illumination & Projection
  - 光源模組（Source）
  - 聚光鏡組（Project Lens）

- 光罩模組--Reticle → Patterning
  - 光罩傳輸模組（Reticle Handler）
  - 光罩平台模組（Reticle Stage）

- 晶圓模組--Wafer
  - 晶圓傳送模組（Wafer Handler）
  - 晶圓平台模組（Wafer Stage）Positioning module, alignment

ASML

EUV-Lithography *TLS BL21 Beamline, U9-CGM*



Grating / Mask

Photo-resist

Schematic illustration of the EUV-IL set-up where the second-order interference field is maximized. The grating parameters are illustrated adjacent to the schematics.

Birgit Päivänranta et al, Nanotechnology 22, 375302(2011) SLS,PSI

Photo-resistor contamination

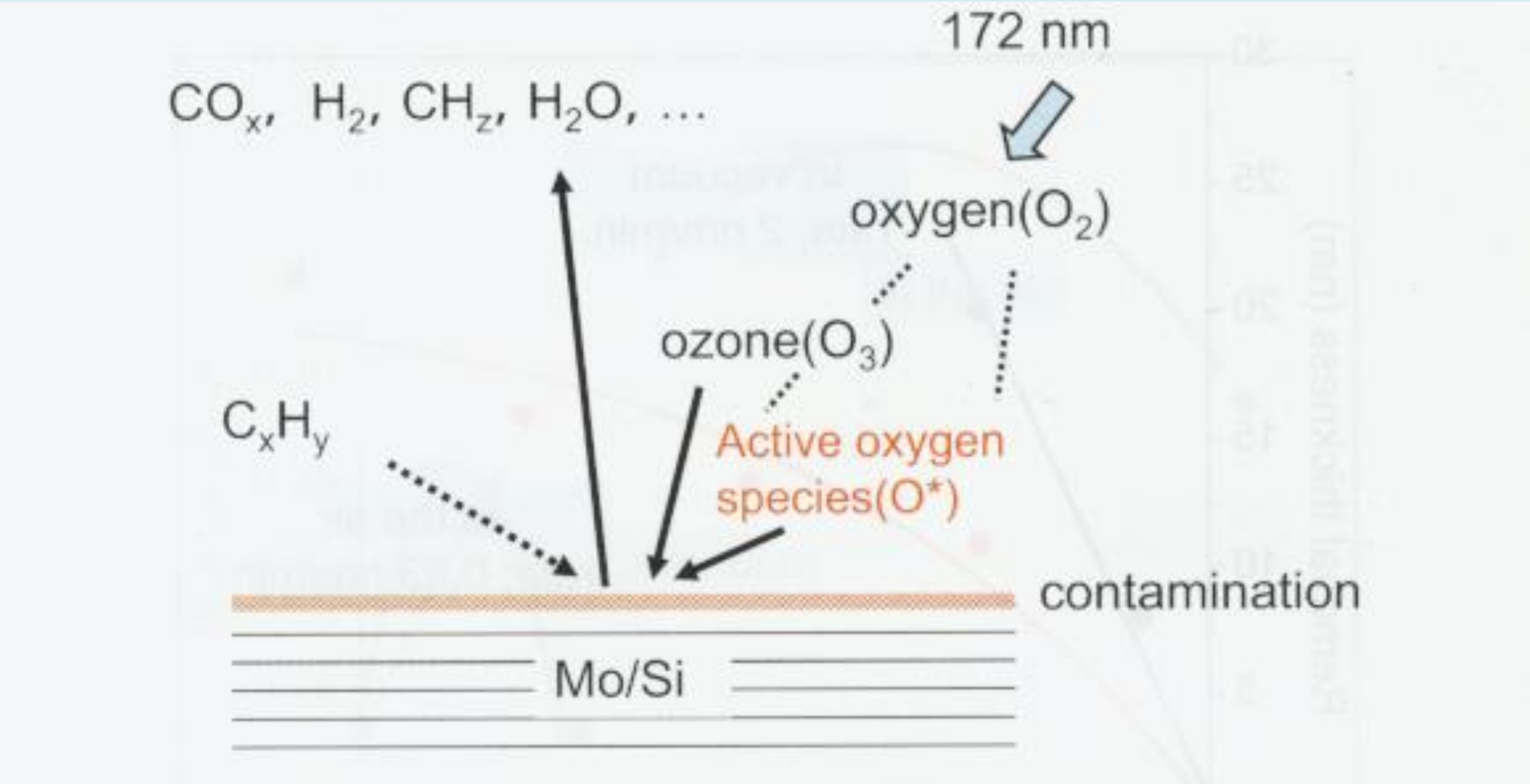


Figure 37. The mechanism of mask cleaning using 172 nm excimer lamp.