



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: S112*

*NSRRC: Tel (03)5780281 ext. 7114 lab. 3165*

*Fax(03)5783813*

*Email:yylee@nsrrc.org.tw*

*101 Hsin-Ann Road, Hsinchu Science Park,  
Hsinchu 300092, 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- **bunches selection**
- 
- Semiconductor EUV Lithography

# Time-resolved experiments

---

## Fundamental parameters in physical world

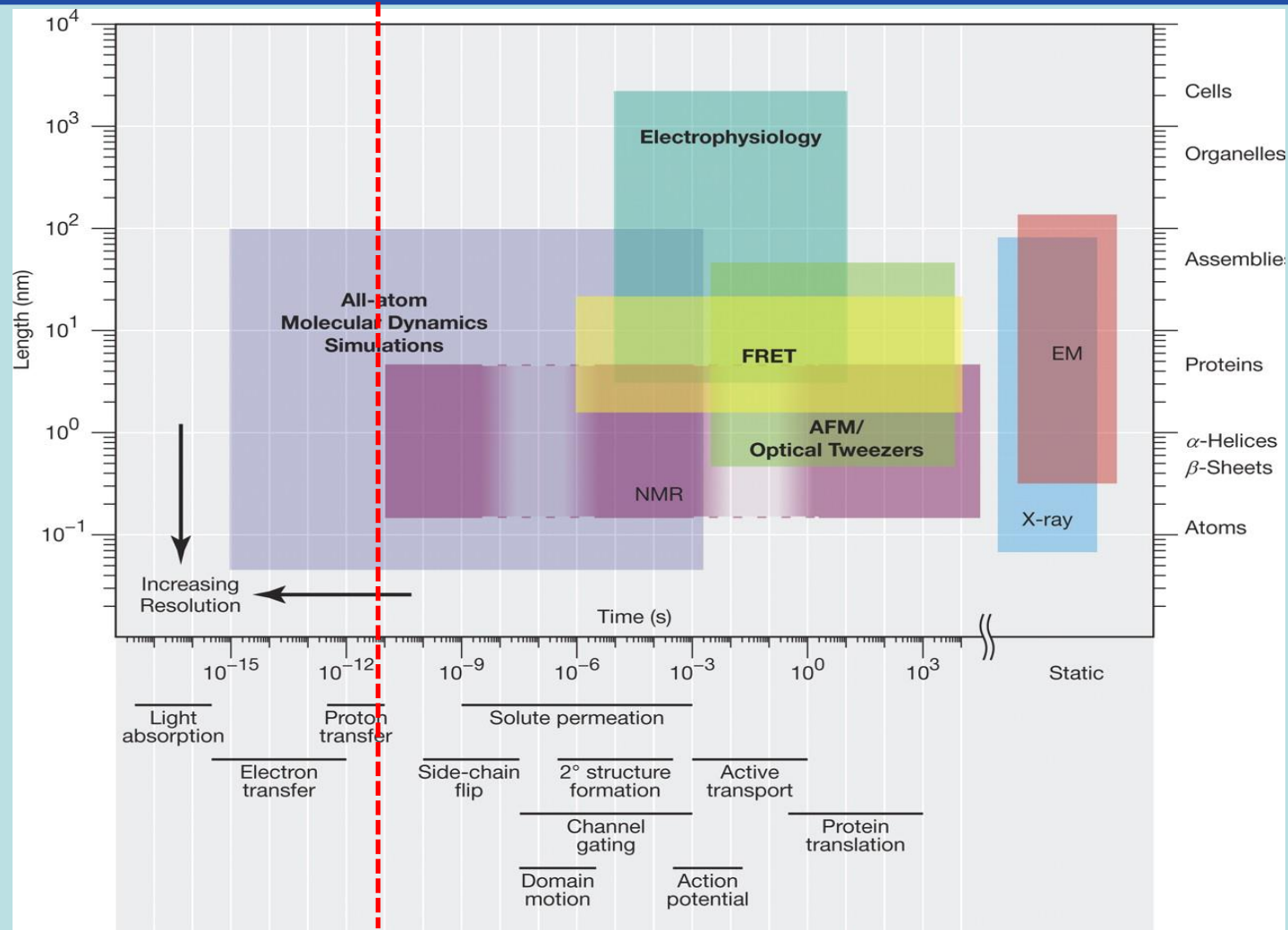
- **Energy, Momentum, Position, Time**
- Time can be applied to all the techniques, in principle
- Some time-resolved experiments take advantage of the pulsed nature of the synchrotron radiation

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.**

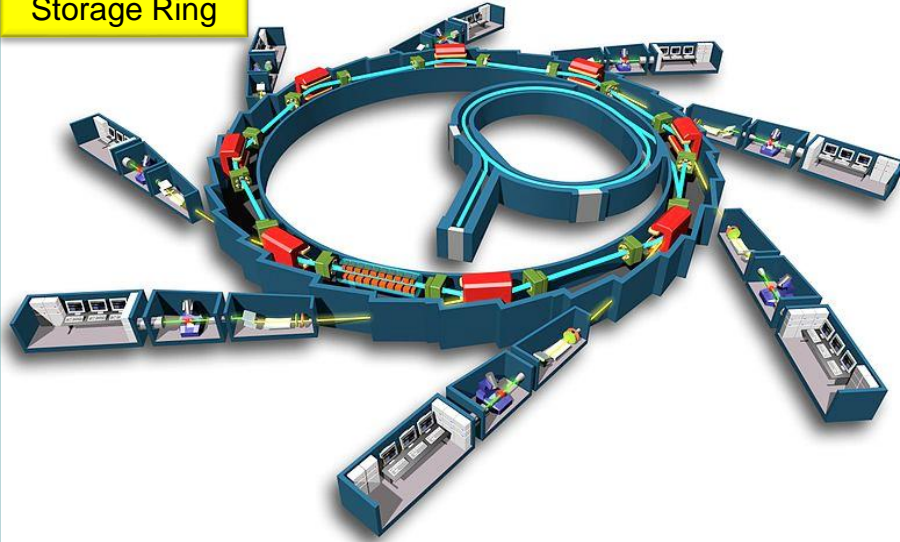
# Spatial and Temporal Resolution for Phenomena



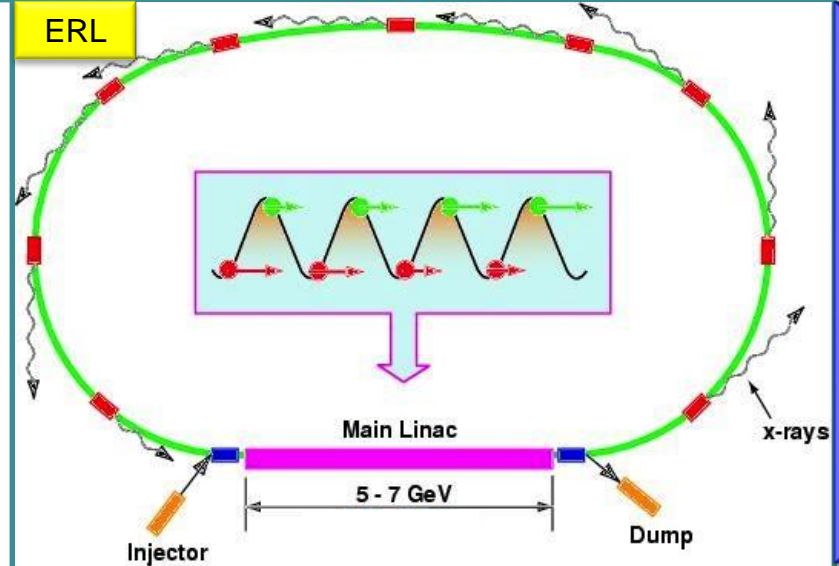
**Spatiotemporal resolution of various biophysical techniques. Techniques capable of yielding data on single molecules (as opposed to only on ensembles) are shown in bold. The timescales of some fundamental molecular processes, as well as composite physiological processes, are indicated below the abscissa.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

# Light Sources: Storage Ring, ERL, & FEL

Storage Ring

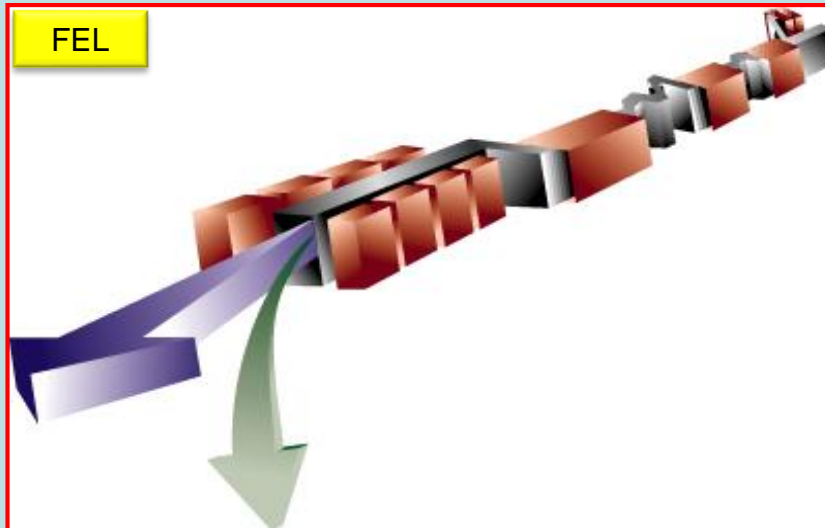


ERL



Spontaneous Emission

FEL



Stimulated Emission

# Light Source Pulse Structures

**Ring-based  
Storage ring**



**ERL**



**FEL**

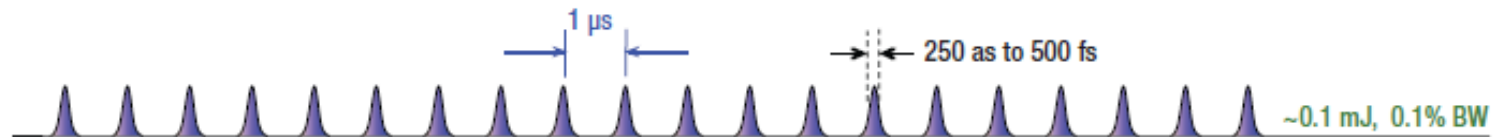
**Burst Mode  
( $\sim 10$  Hz)**



**Pulsed  
( $\sim \text{k}$  Hz)**



**CW  
( $\sim \text{M}$  Hz)**



# 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- $\alpha$  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  $10^{-3}\sim 10^{-6}$
- Chopper required for over exposure and avoiding sample damage

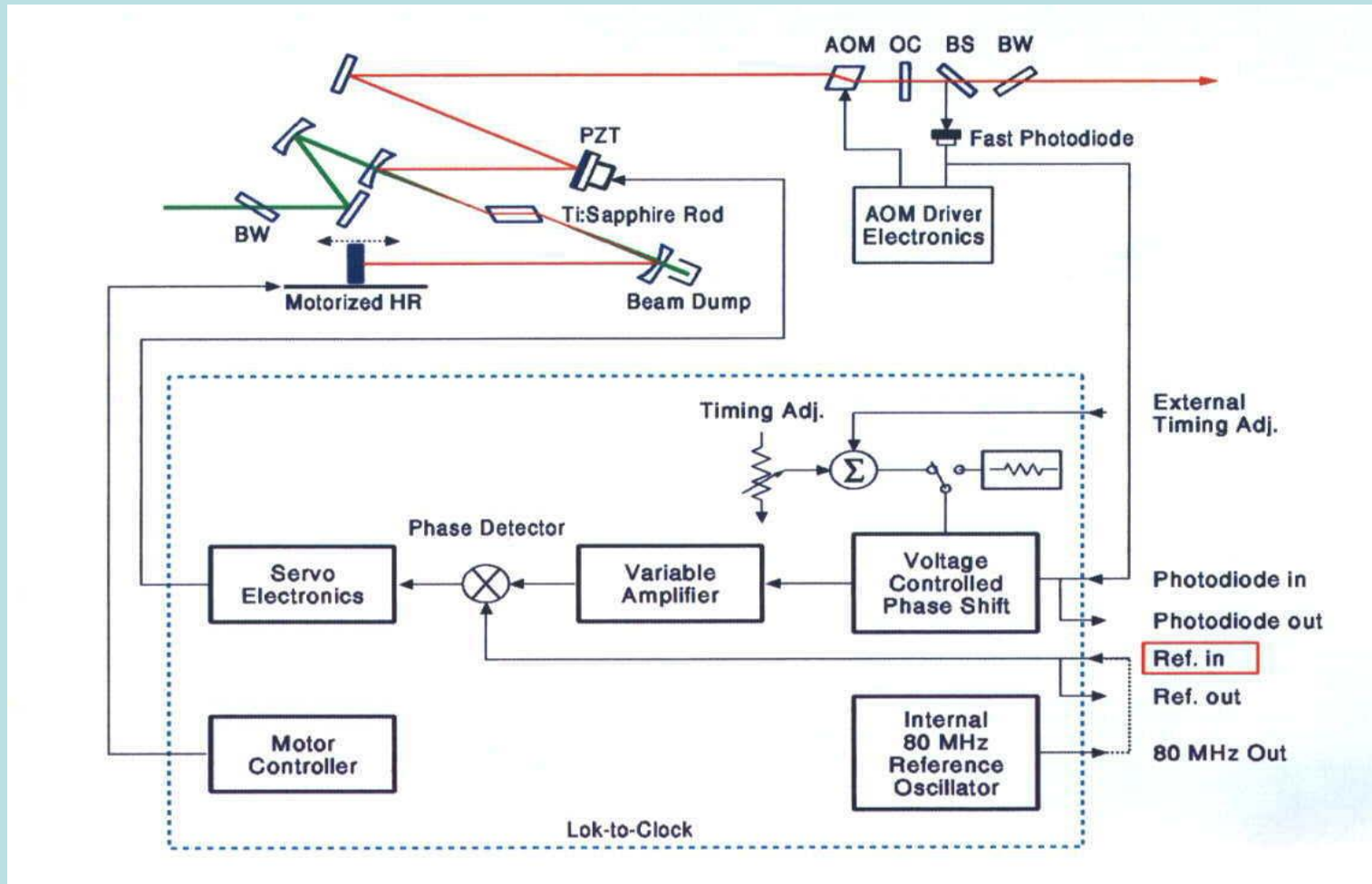
### ☑ Time-resolution is limited by the X-ray pulses duration at NSRRC currently.

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



# Synchronization Mechanism: Lock-to-clock

- (1)  $2.5\text{MHz} \times 32 \longrightarrow 80\text{ MHz}(12.5\text{ns})$        $500\text{ MHz} / 6 = 83.33\text{ MHz}, 12\text{ ns}$   
 (2)  $500\text{MHz} / 25 \times 4$



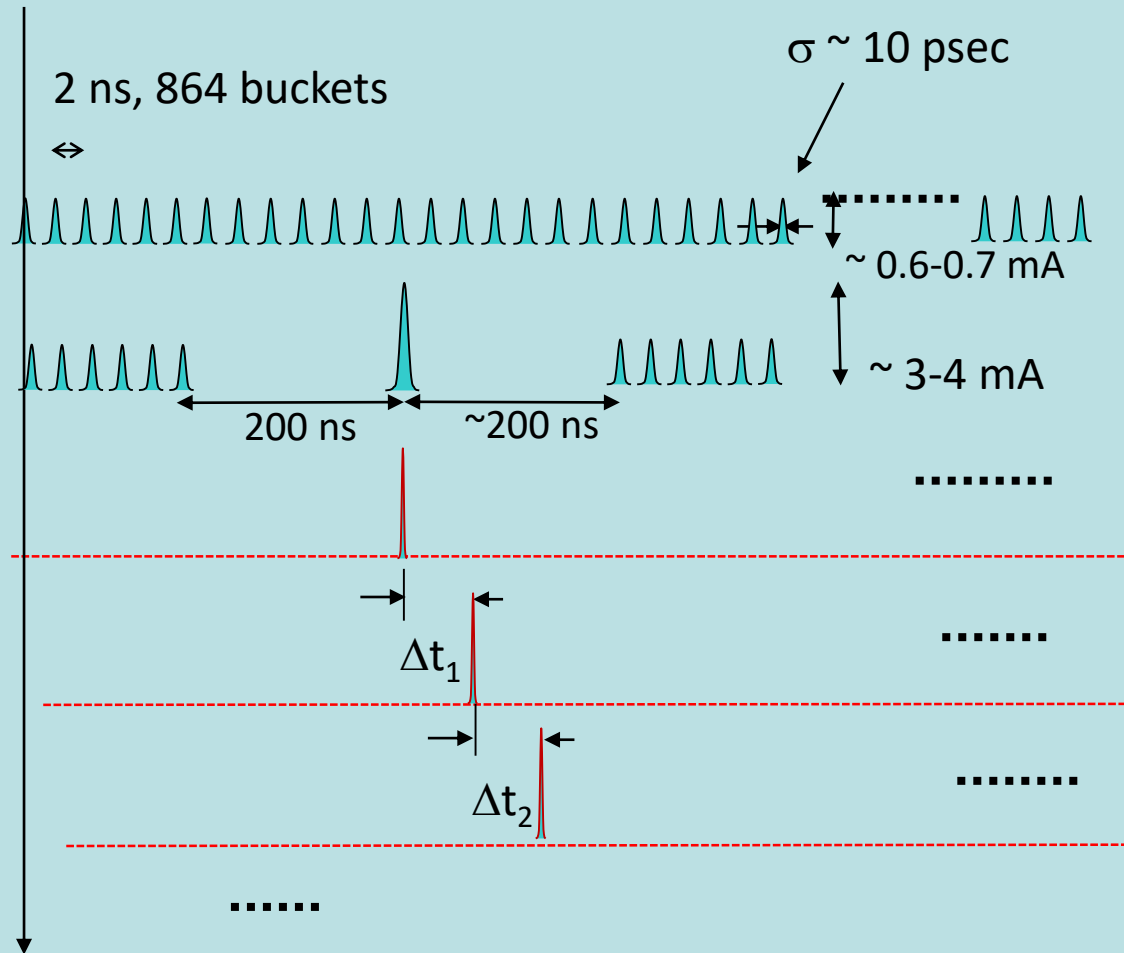
Cavity modes:  $\lambda_n = 2L/n$

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



# Single bunch in Hybrid mode with laser Osc.

( 578.7 kHz,  $\text{TPS}_{\text{R}}=518.4\text{m}$  )



Fully filled

Single bunch in hybrid mode  
(Dep. on # of empty buckets)

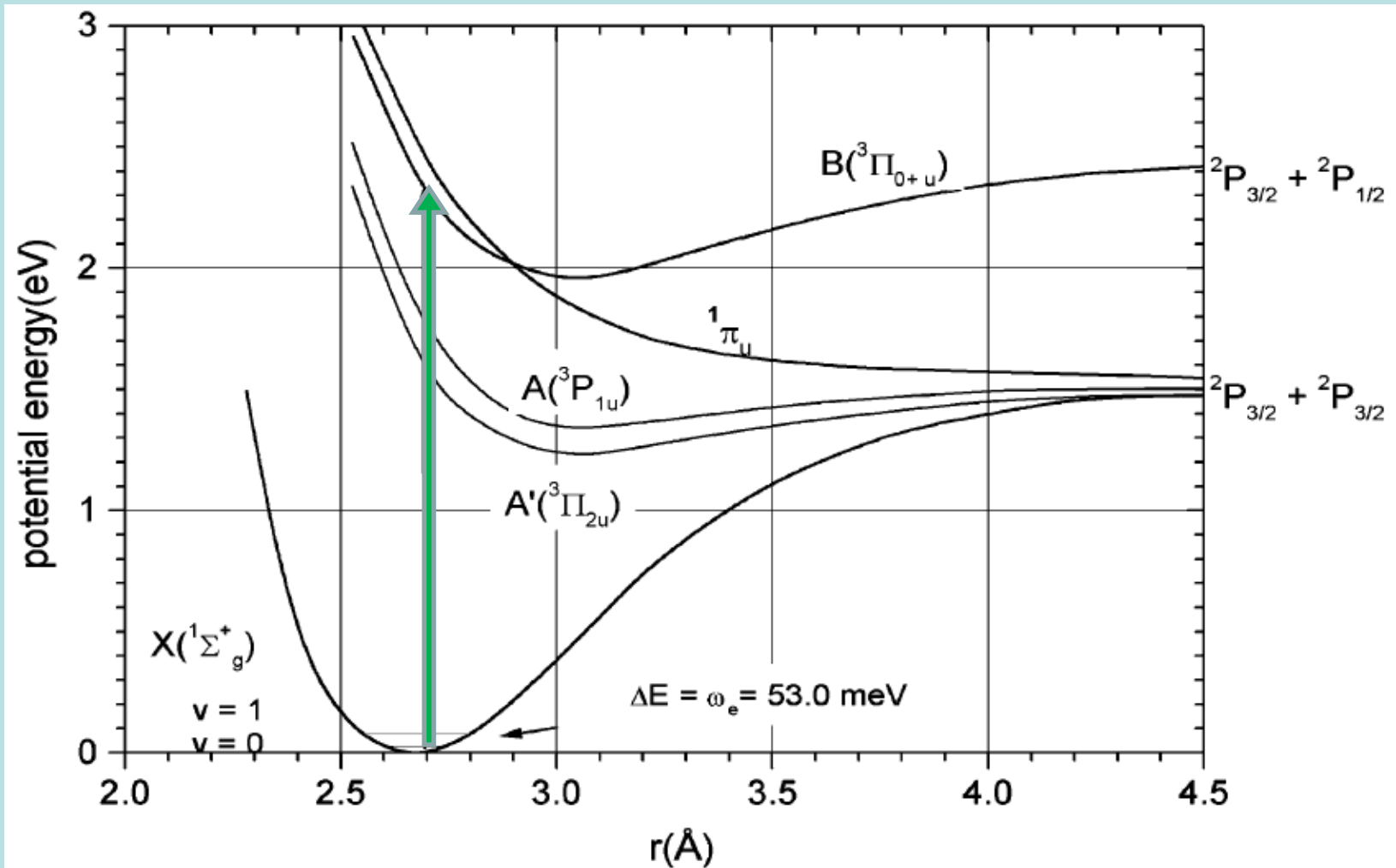
578.7 kHz ML laser

Laser time delay / phase shift,  $\theta_1$

Laser time delay / phase shift,  $\theta_2$

Reversed for laser pump - X-ray probe up to time span 200ns (Dep. on # of empty buckets)

# 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 $CCl_4$

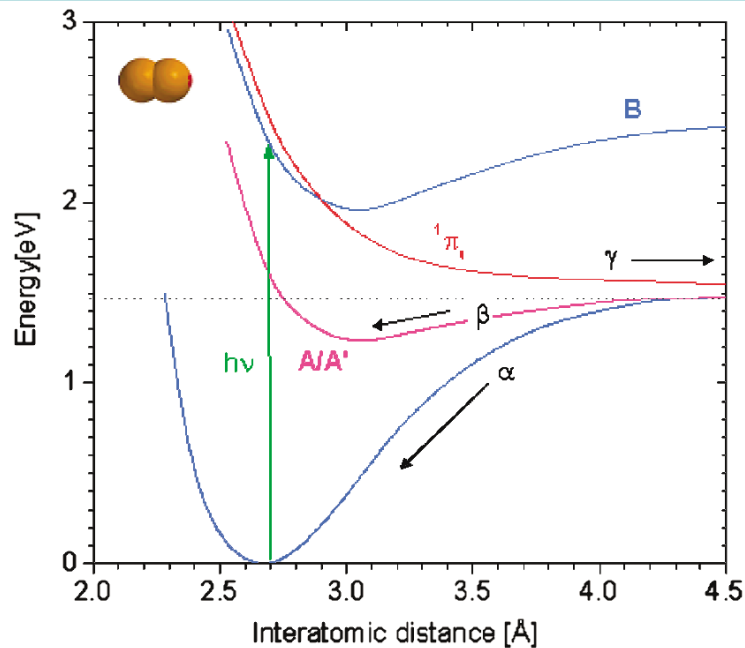


FIG. 1 (color). Low lying electronic energy surfaces of  $I_2$ : the states  $X$ ,  $A/A'$ , and  $B$  are attractive, whereas the state  $1\pi_u$  is repulsive. The processes  $\alpha$ ,  $\beta$ , and  $\gamma$  denote vibrational cooling along the  $X$  potential, geminate recombination through the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

## Solution molecules

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

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

----monitor atomic motions in liquids

Diffacted signal depends on **scattering wave vector,  $q$**  and **time delay,  $\tau$** .

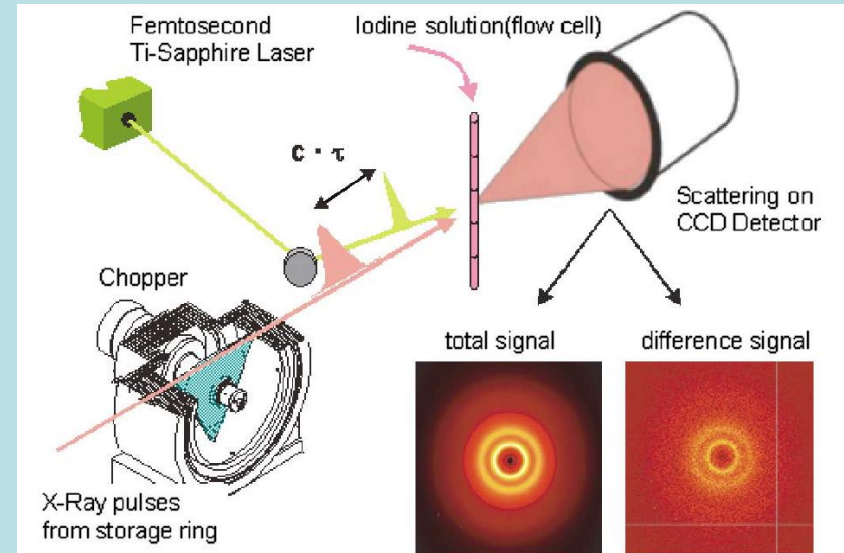
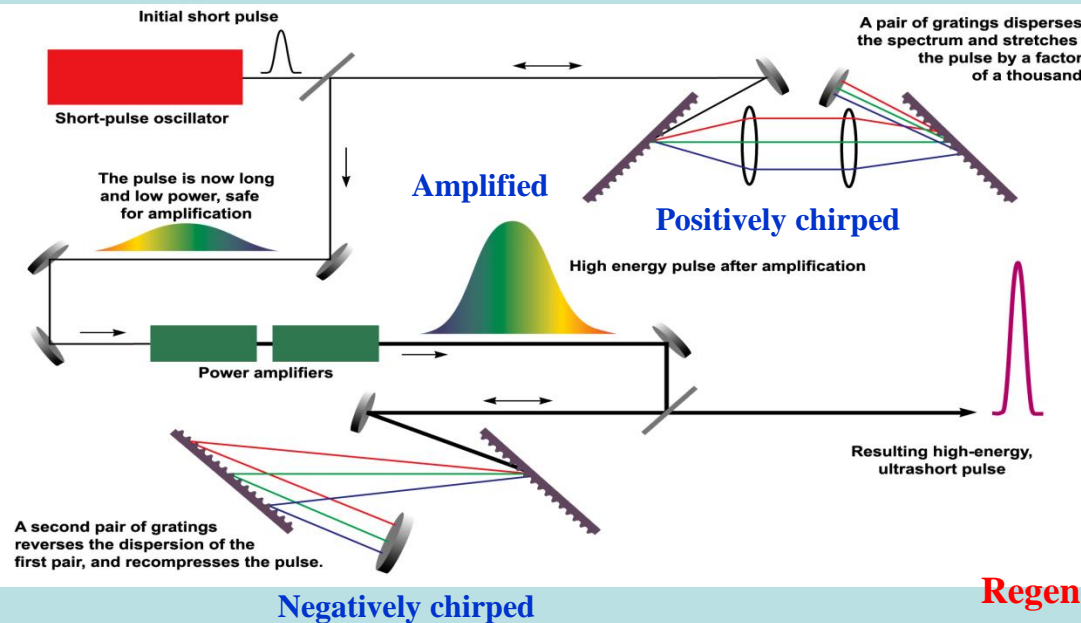


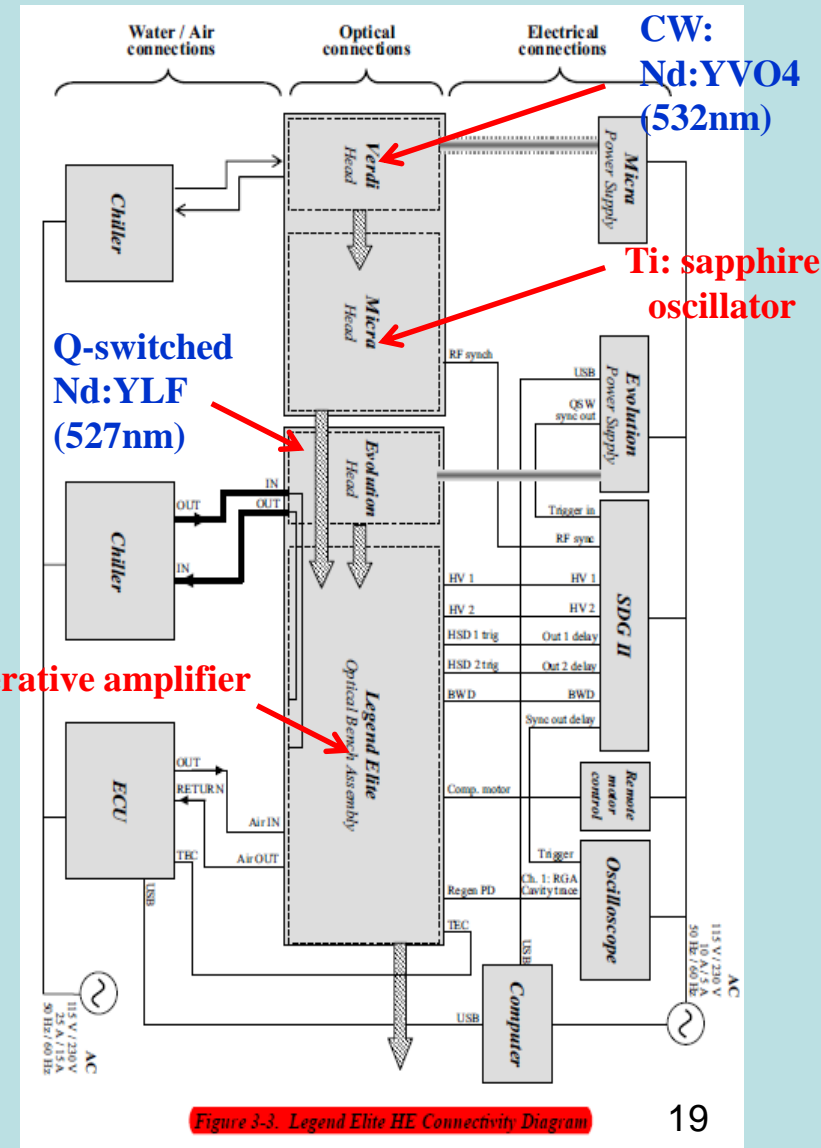
FIG. 2 (color). Experimental setup: the x-ray pulses are generated by an undulator. The spectrum is centered at  $0.67 \text{ \AA}$  ( $18.5 \text{ keV}$ ) and its bandwidth width is  $d\lambda/\lambda = 0.03$ . The flux on the sample is  $5 \times 10^8$  per pulse and the pulse length is  $150 \text{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

# Mode-locked Laser System

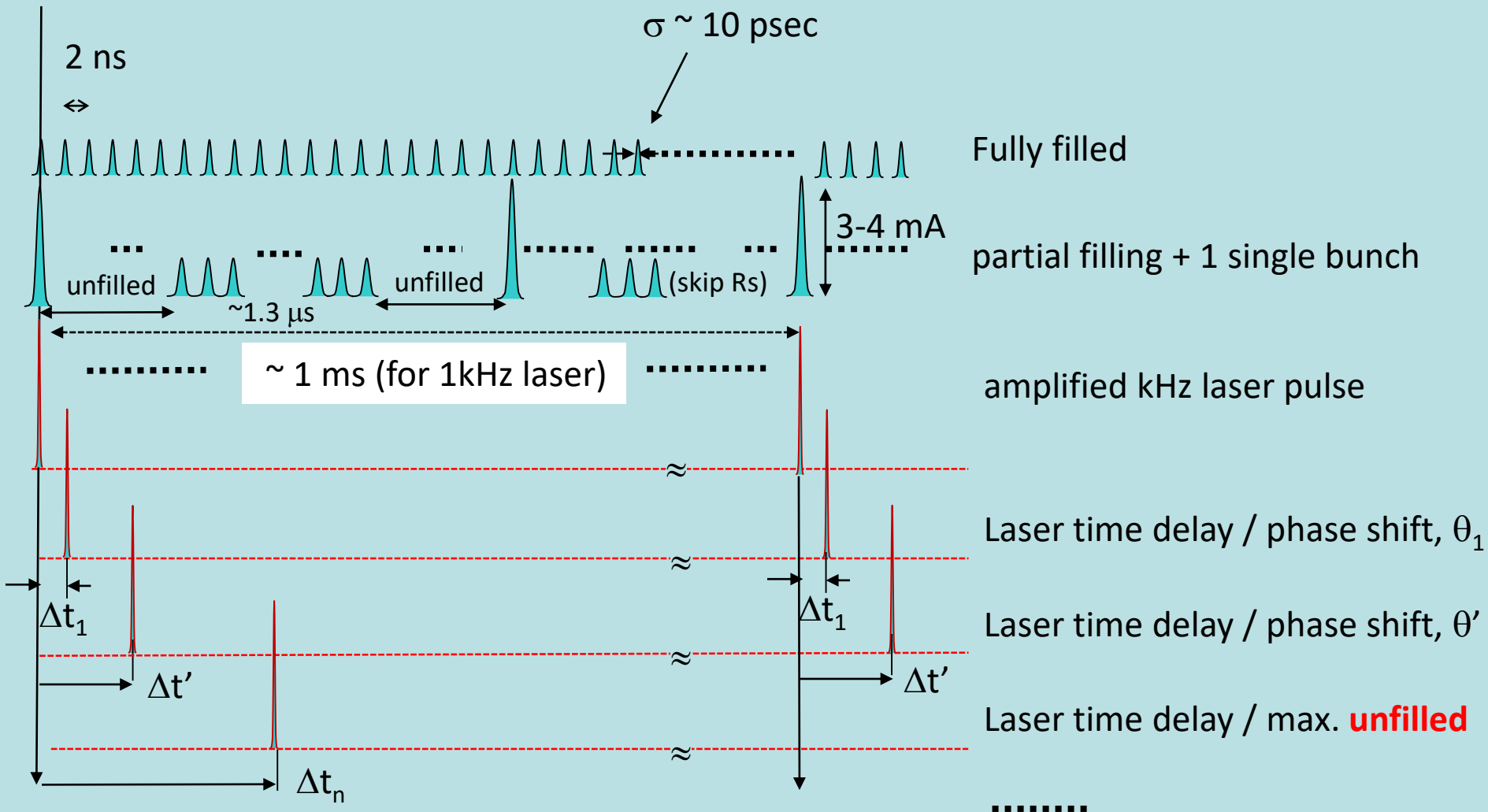
## Ti:Sapphire regenerative amplifier



	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	$1.15 \times 10^{11}$ W

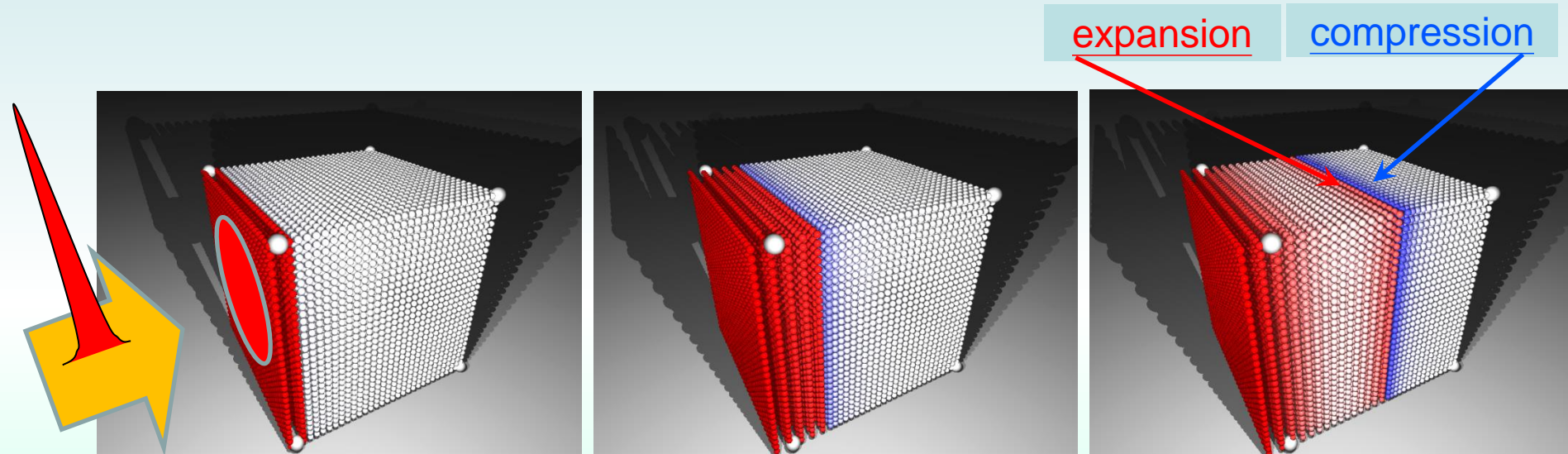


# Gated Single bunch with amplified laser pulse



# Laser excitation of a solid (below damage)

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



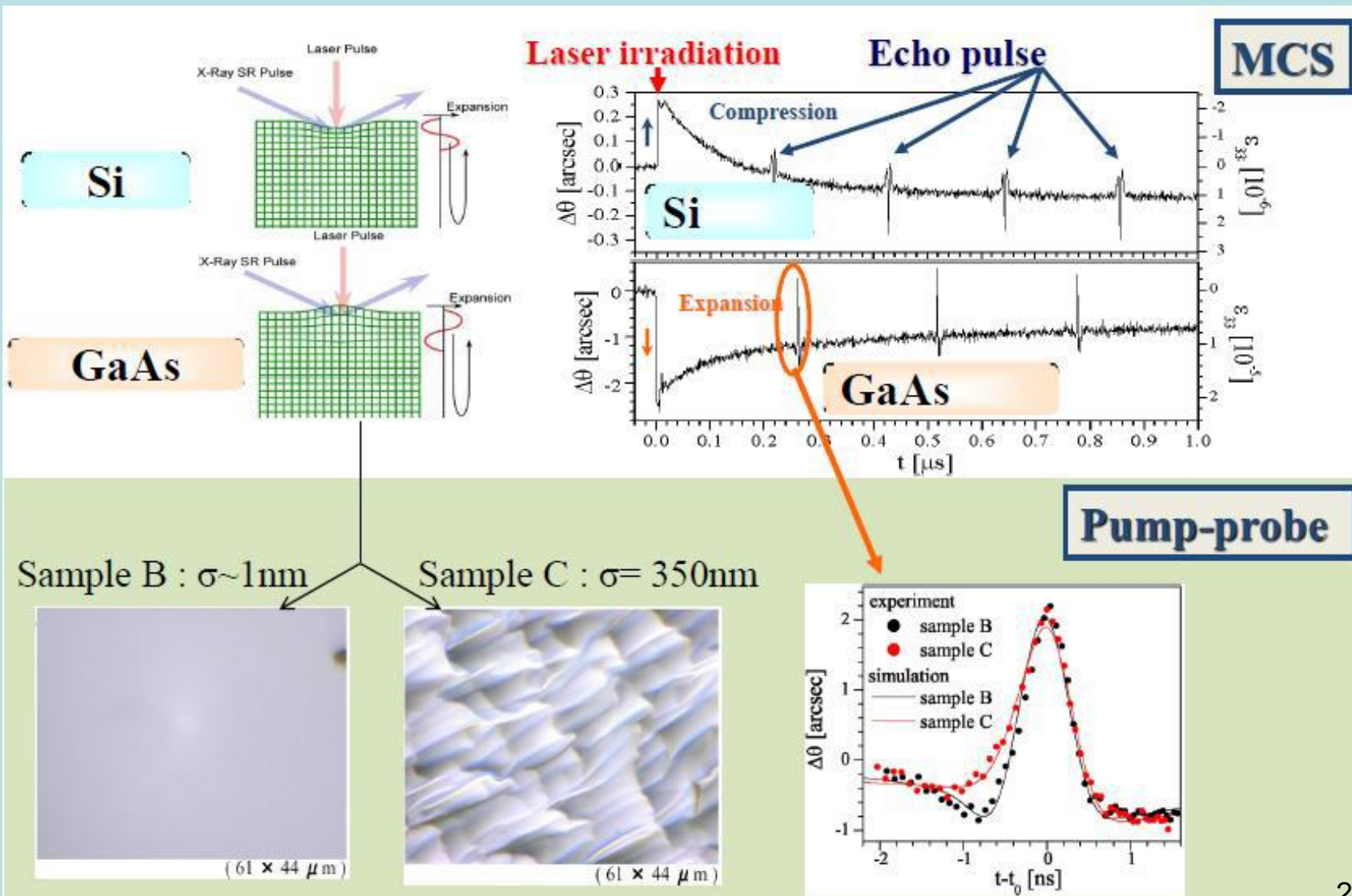
- a) Electrons are excited → electronic stress due to deformation
- b) Electrons heat lattice
- c) Thermal stress

Stress relaxes by  
lattice expansion

Expansion triggers acoustic  
wave  
(coherent phonons)  
--due to Newton's 3<sup>rd</sup> law

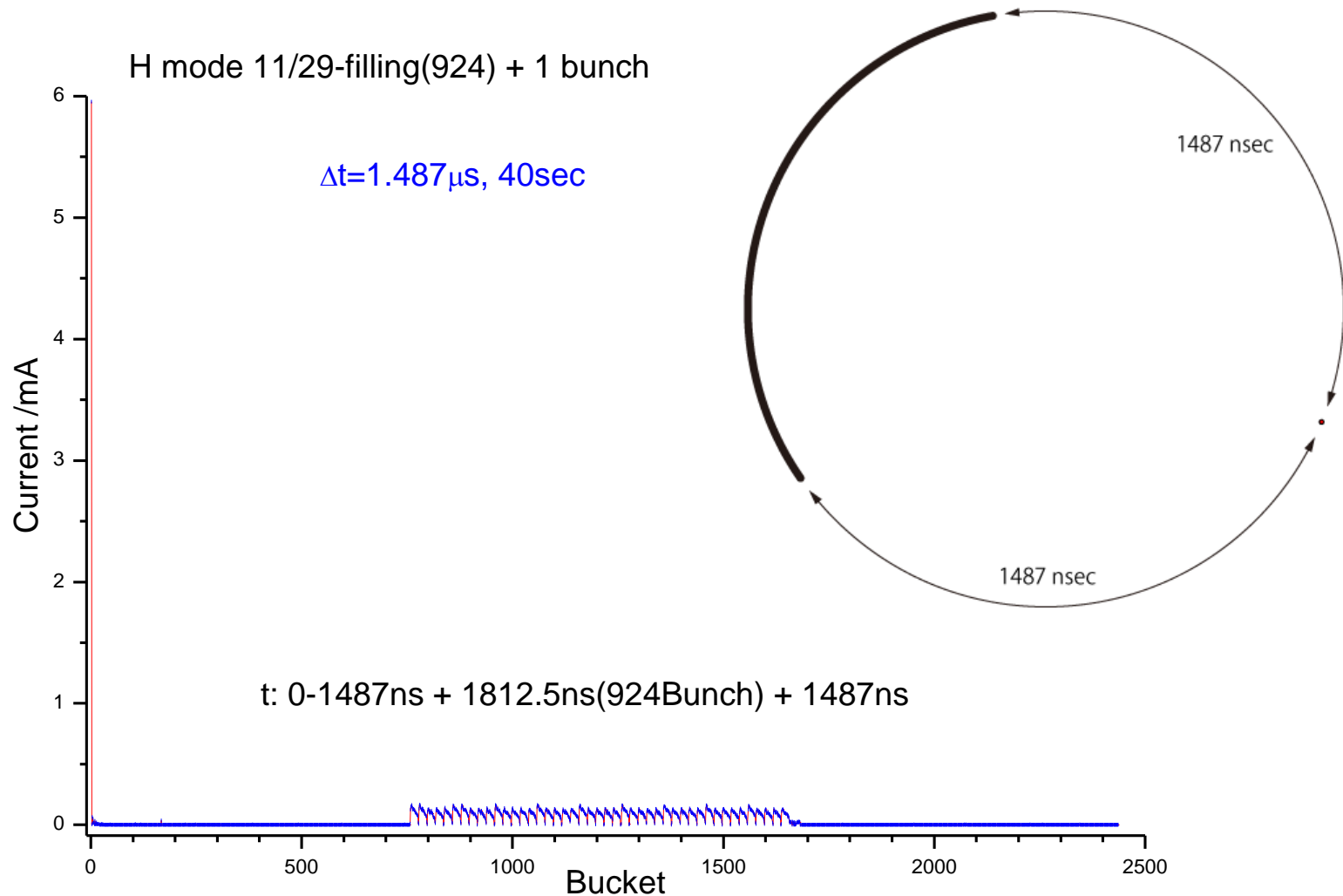


# Time-resolved XRD





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



# TPS Capability

Bunch number: 864

Multi-bunch current:  $\sim 500$  mA (dependent on available RF power)

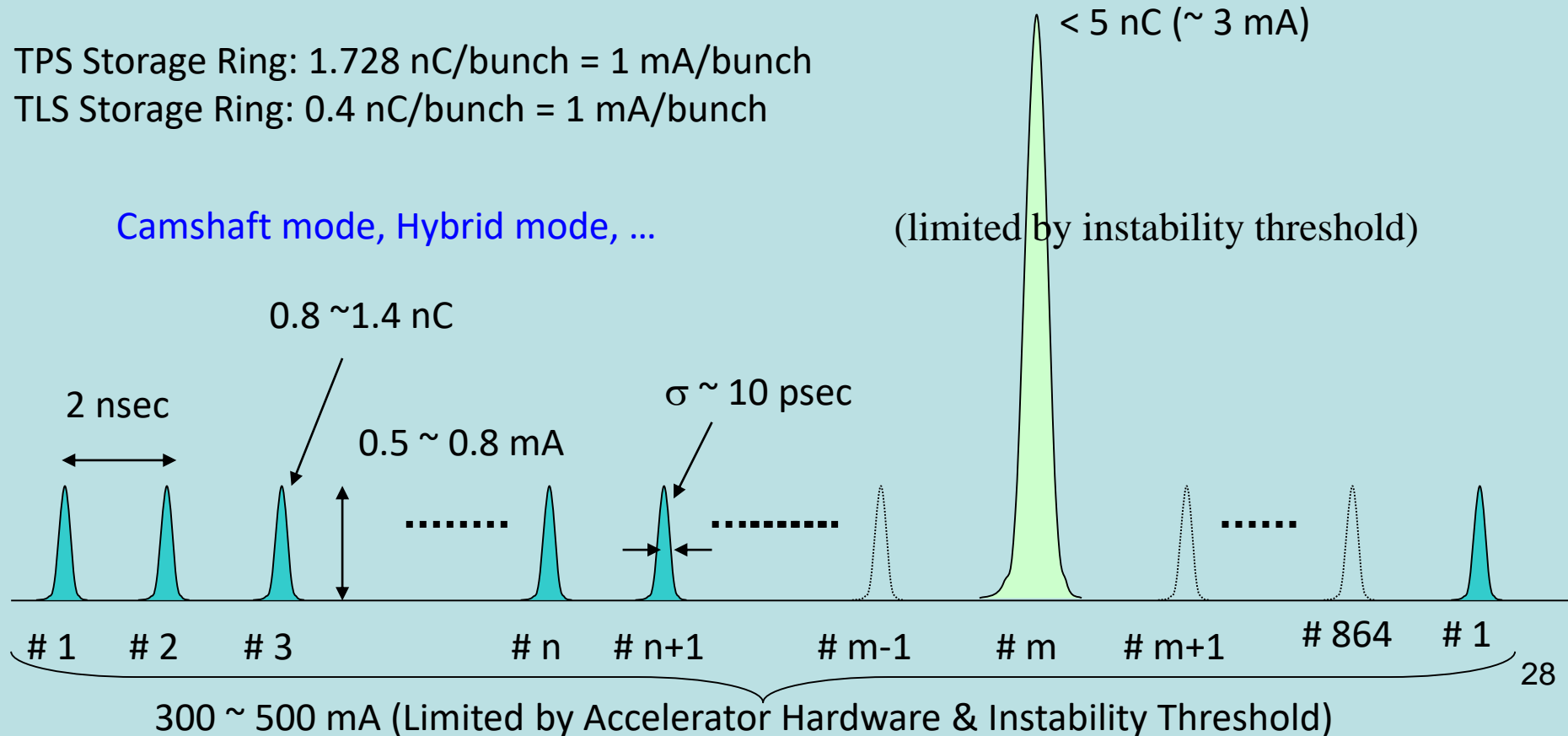
Multi-bunch bunch current:  $< 1$  mA/bunch

Single-bunch (isolated bunch?) current:  $2 \sim 3$  mA

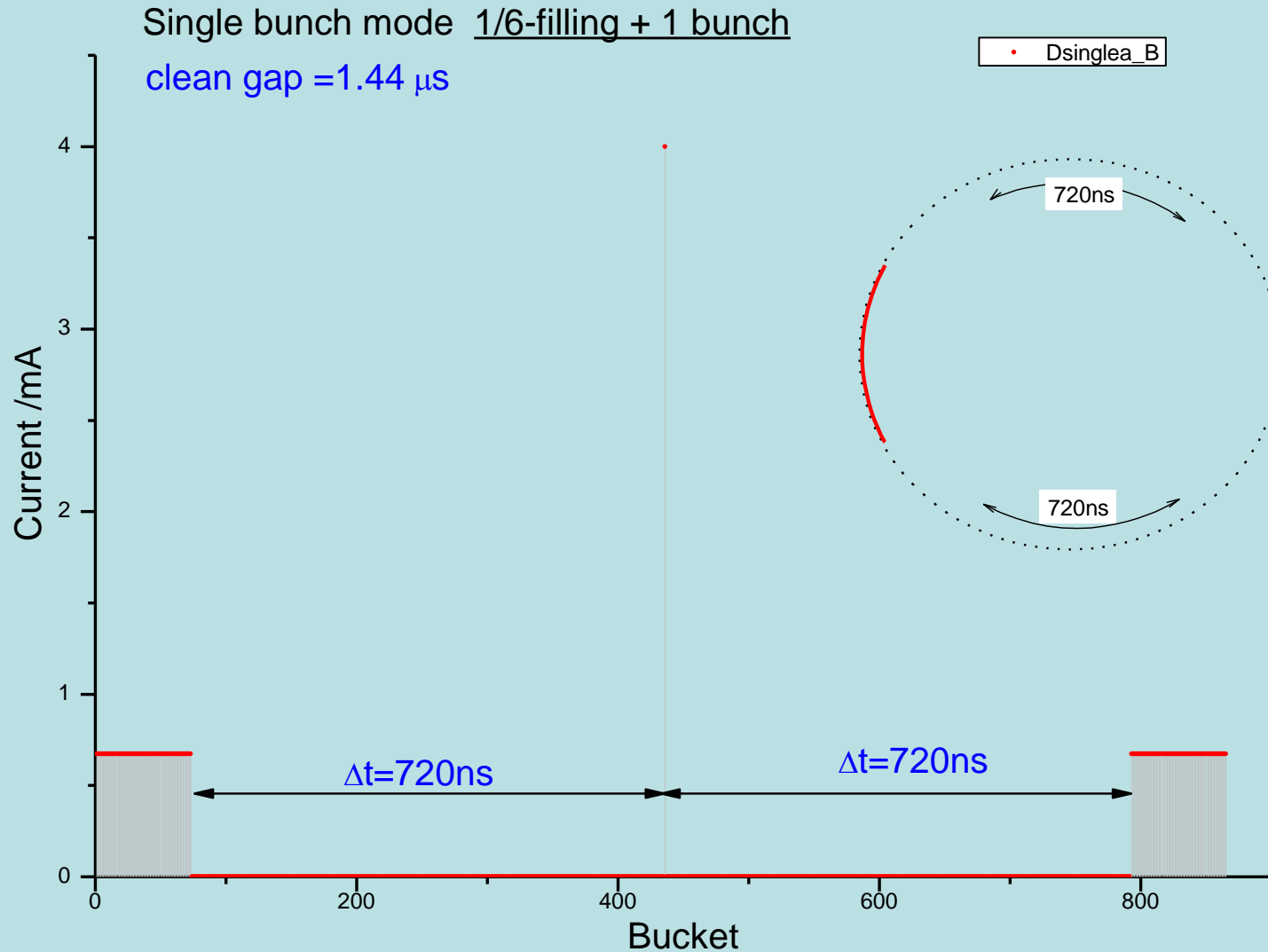
Hybrid mode (camshaft mode)

TPS Storage Ring:  $1.728$  nC/bunch =  $1$  mA/bunch

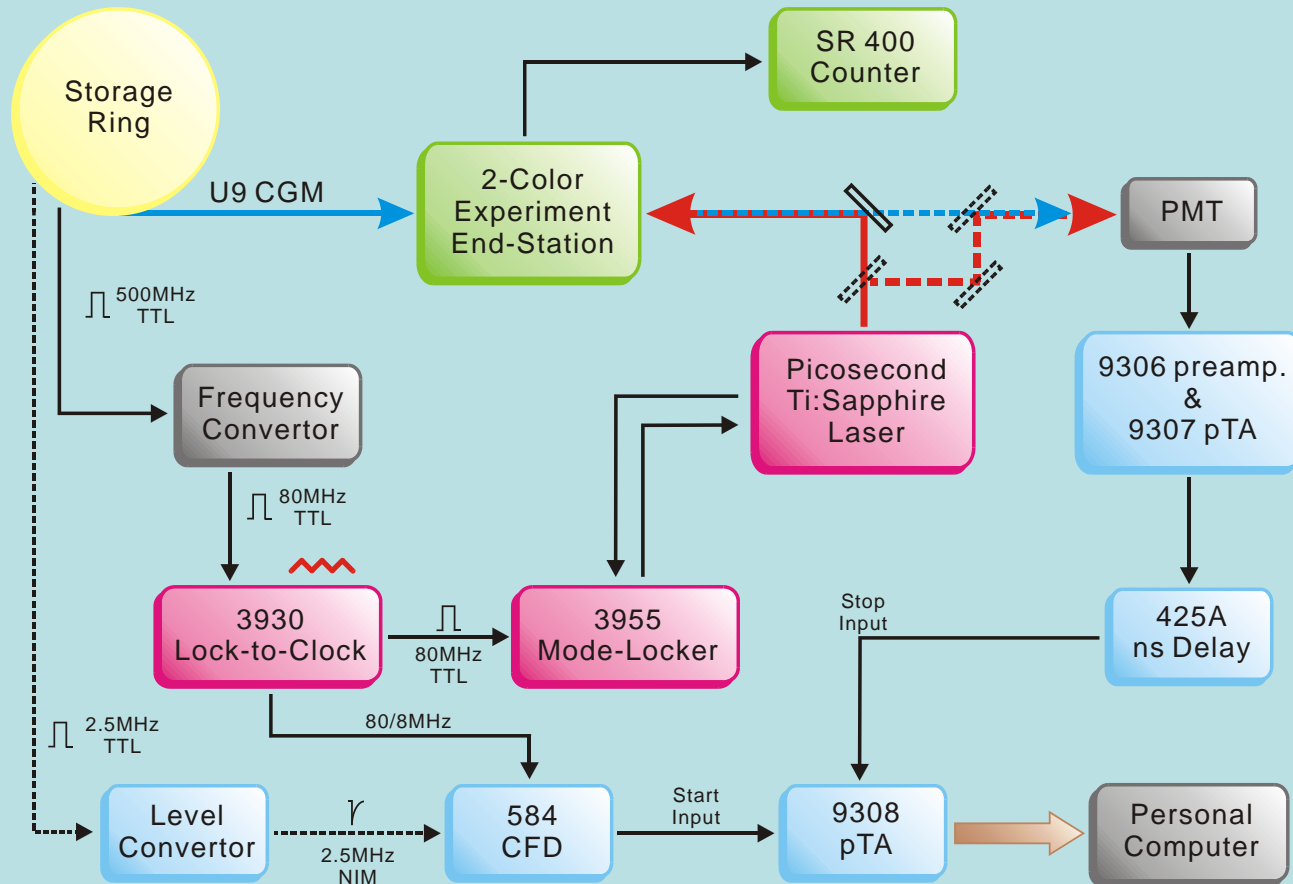
TLS Storage Ring:  $0.4$  nC/bunch =  $1$  mA/bunch



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



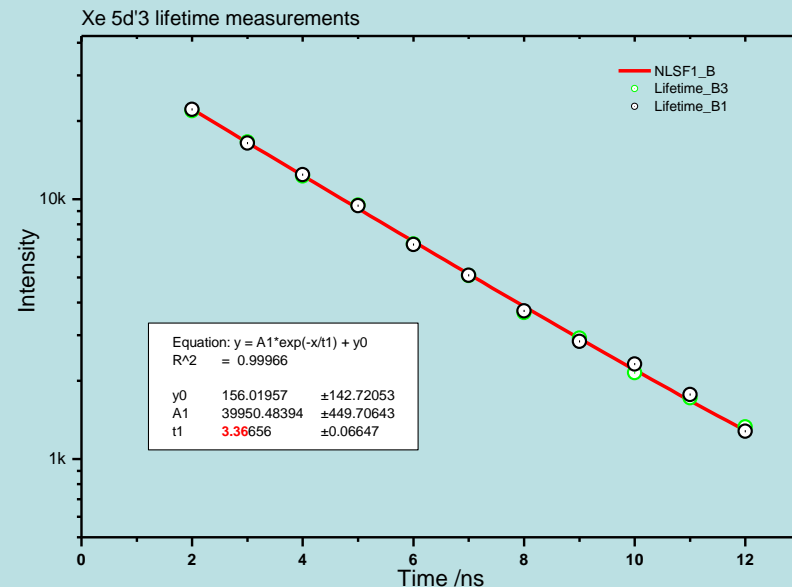
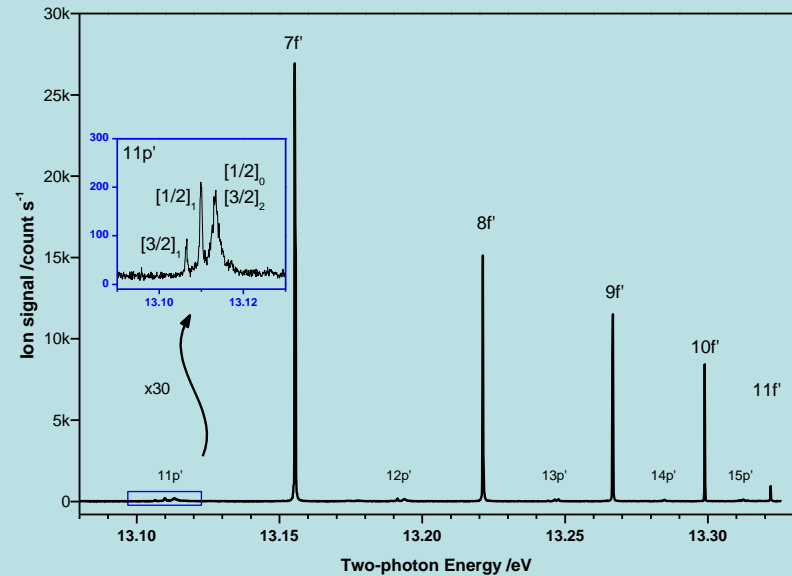
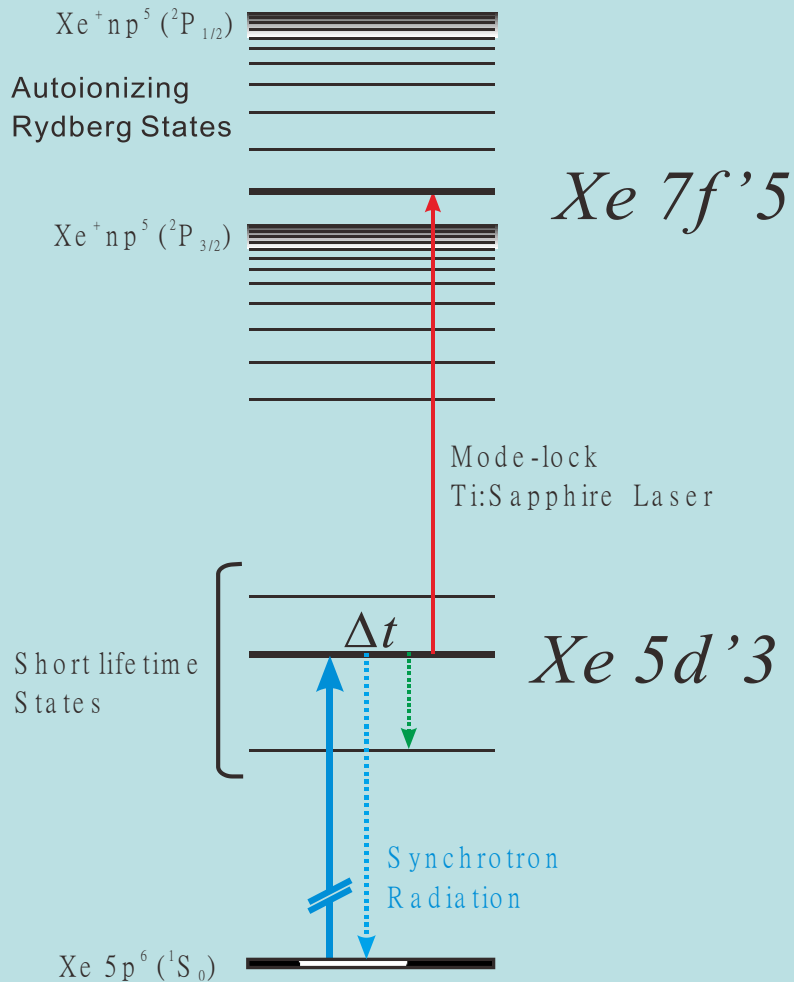
# Setup for timing control and measurement system



Block diagram of pico-second timing measurement system. The trigger to the mode-locked laser is linked to a frequency converter 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.

# Two-color Pump-Probe Experiments:

## Atomic Xe Spectroscopy and lifetime measurement



# 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,  $>\mu\text{s}$

## 2. Fast Choppers/Shutters, $<300\text{ns}$ , jitter $< 5\text{ns}$ , 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

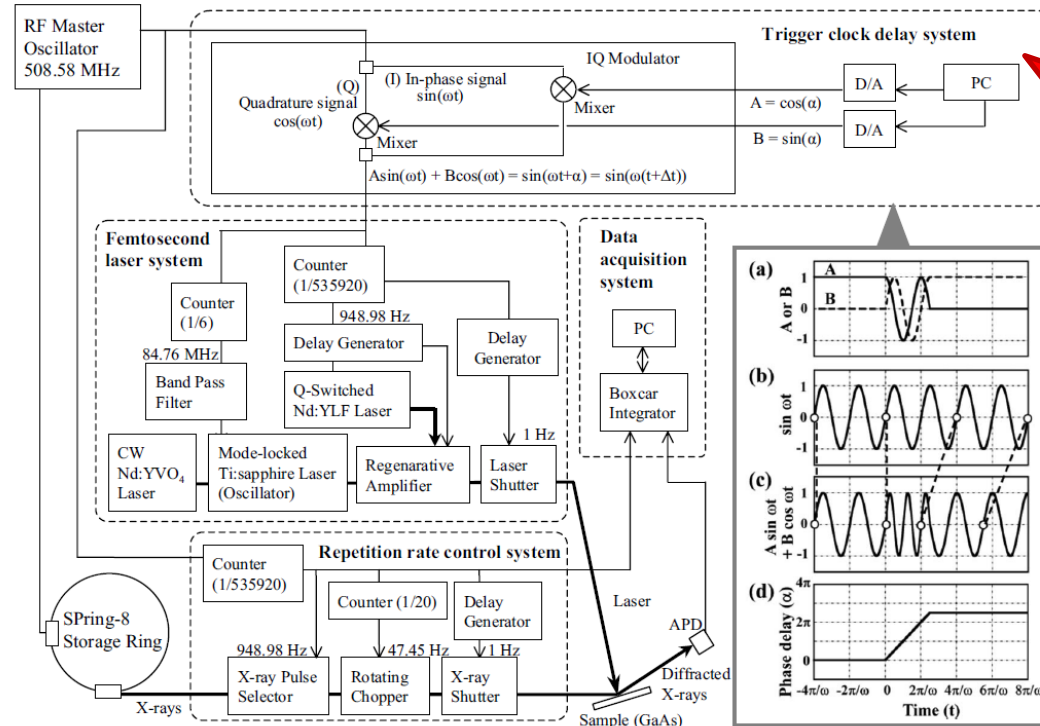


FIG. 1. Block diagram of the trigger clock delay system, the femtosecond laser system, the repetition rate control system, and the data acquisition system. The inset gives the schematic graphs to explain the principle of the continuous phase change at the trigger clock delay system. When the control 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  $\alpha$  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  $2\pi$ .

## Trigger and Clock delay units/*S*Spring-8

- In phase quadrature modulator and a synchronous counter
- Delay time nearly infinite amount
- Precision in  $\pm 8.4$  ps



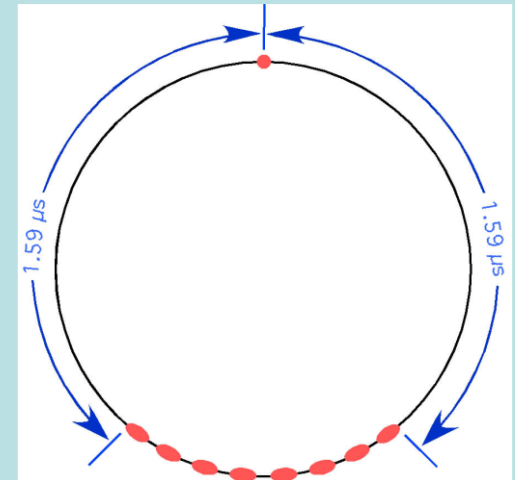
# Hard X-ray Choppers

X-ray choppers can select x-ray pulses from synchrotron pulse train

1. Define temporal resolution
2. Limit heat load
3. Reduce noise

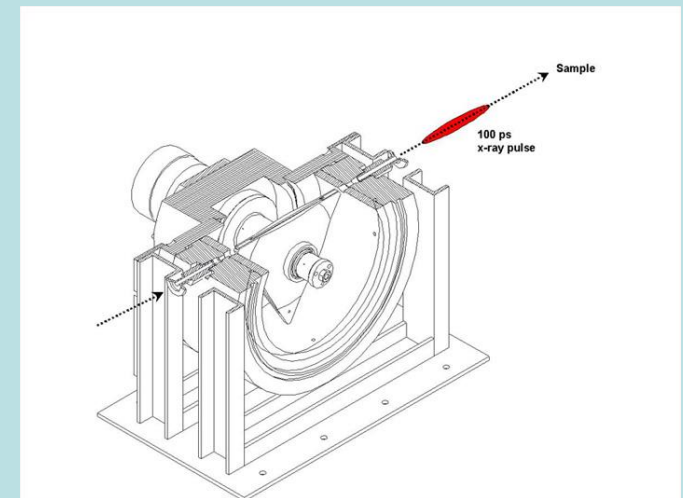
## Mechanically Rotating

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



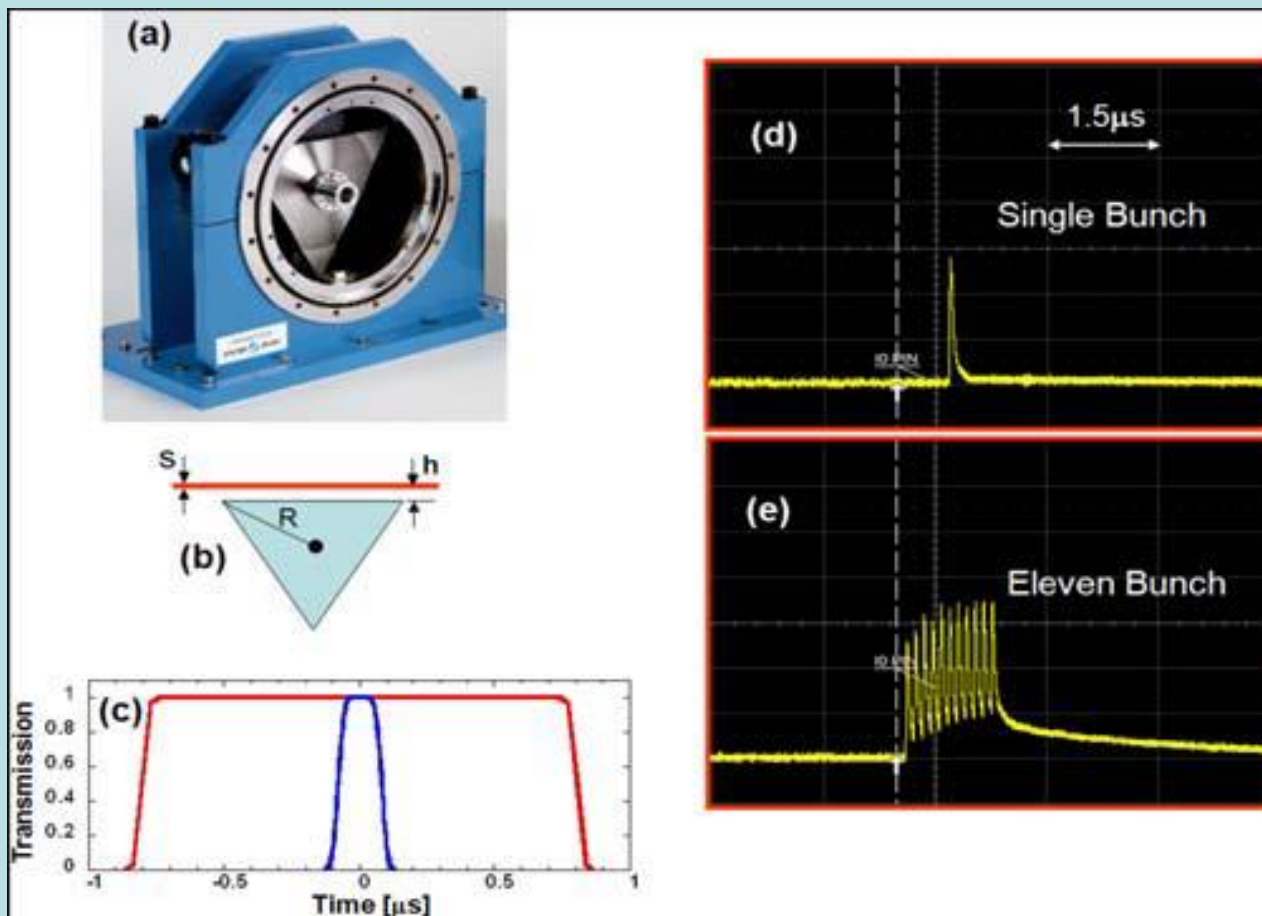
## Julich rotating chopper

- 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



# Chopper/Shutter

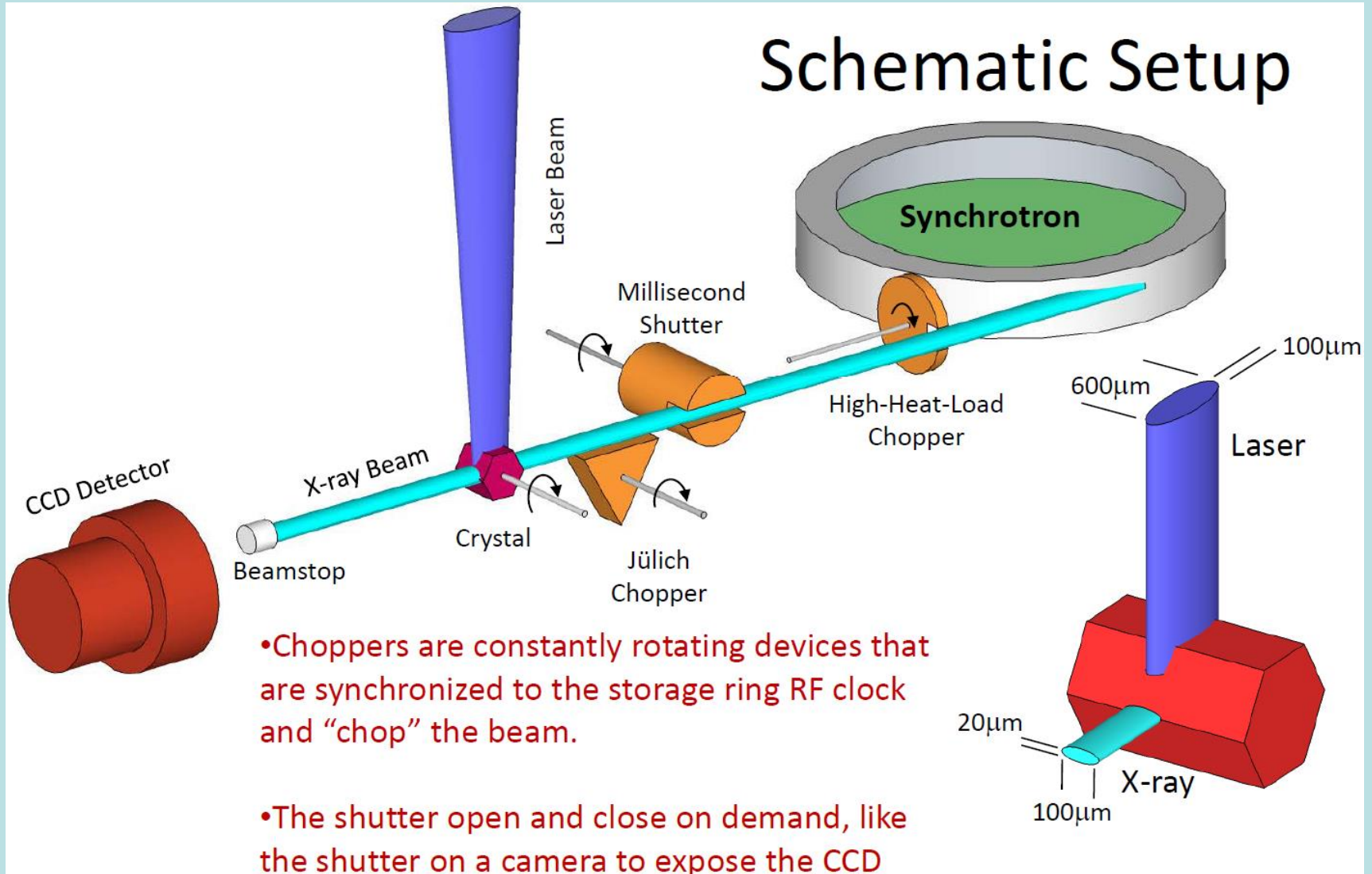
It is based on a directly servomotor-driven triangular rotor with a magnetic bearing and operated in vacuum. Currently its repetition rate is limited to about **900 Hz** with an upgrade option to **2700 Hz**.



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.

# Time-resolved X-ray scattering in ID14. APS

## Schematic Setup



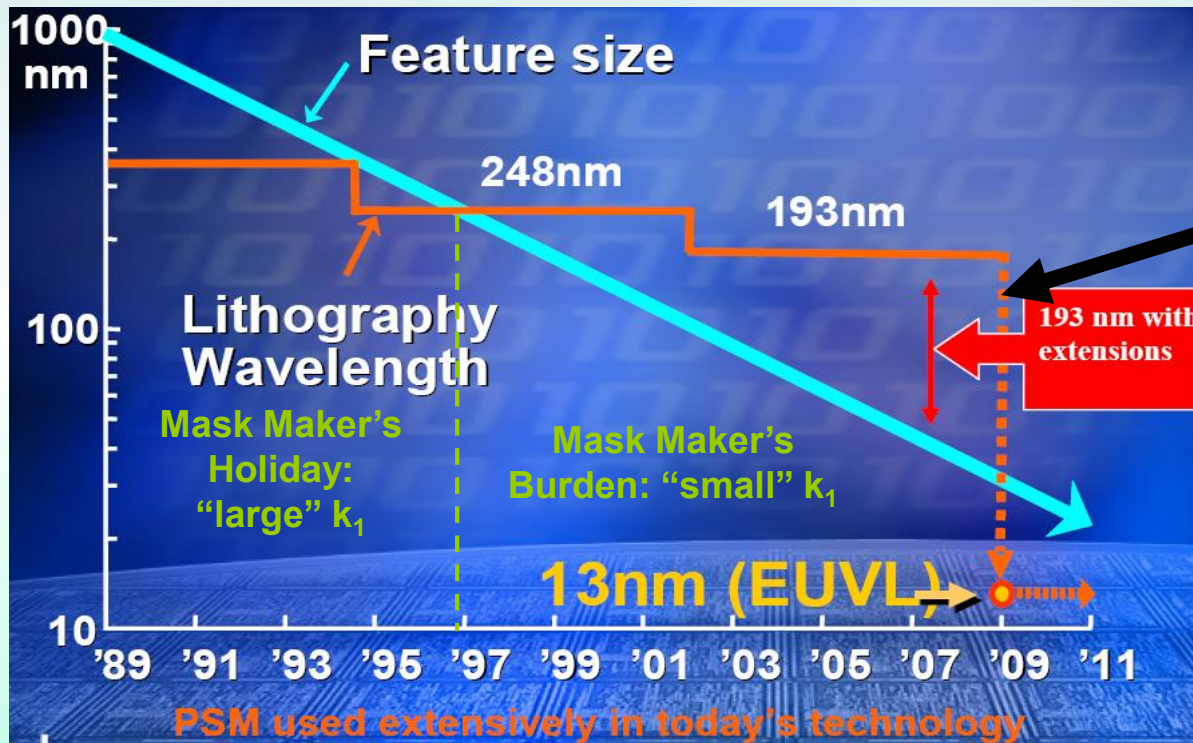
# Semiconductor Lithography

## EVU Lithography

# Semiconductor Lithography

“Transitions to EUV is a big jump”

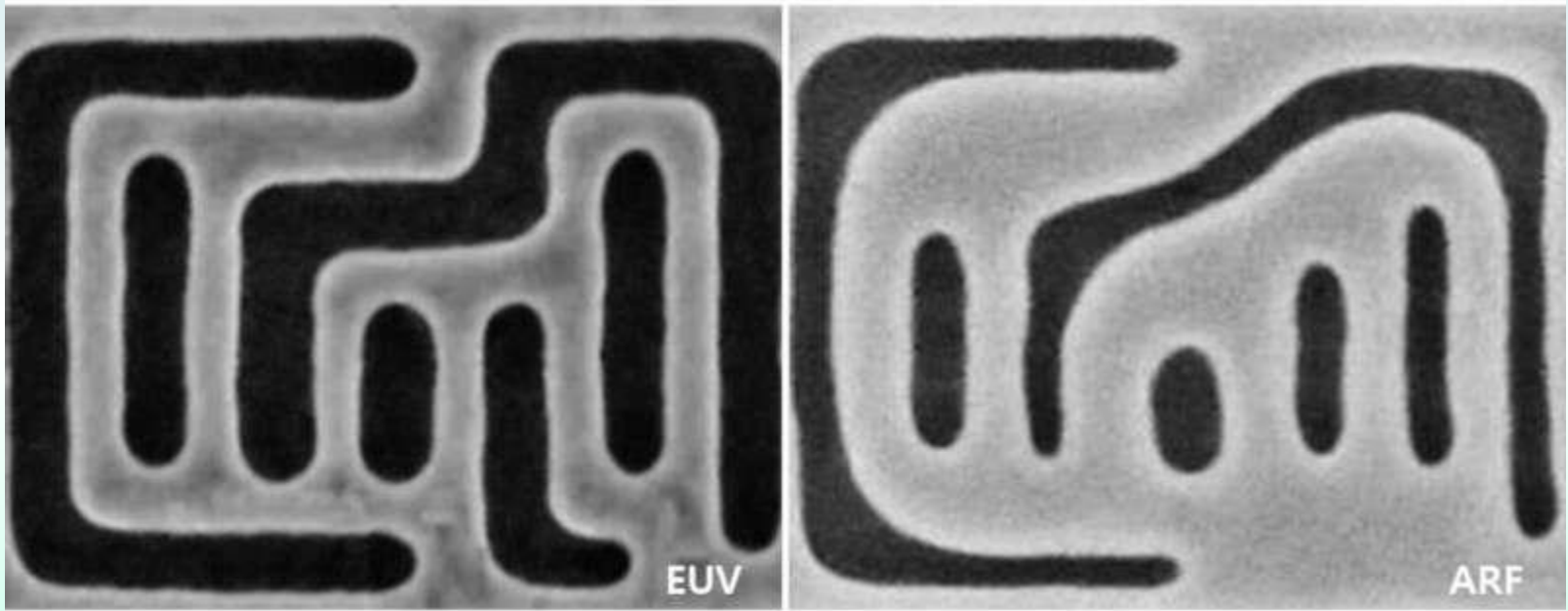
Big jump from 193 to ~13 nm. Before this has about ¼ increase in energy. Now >10x



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

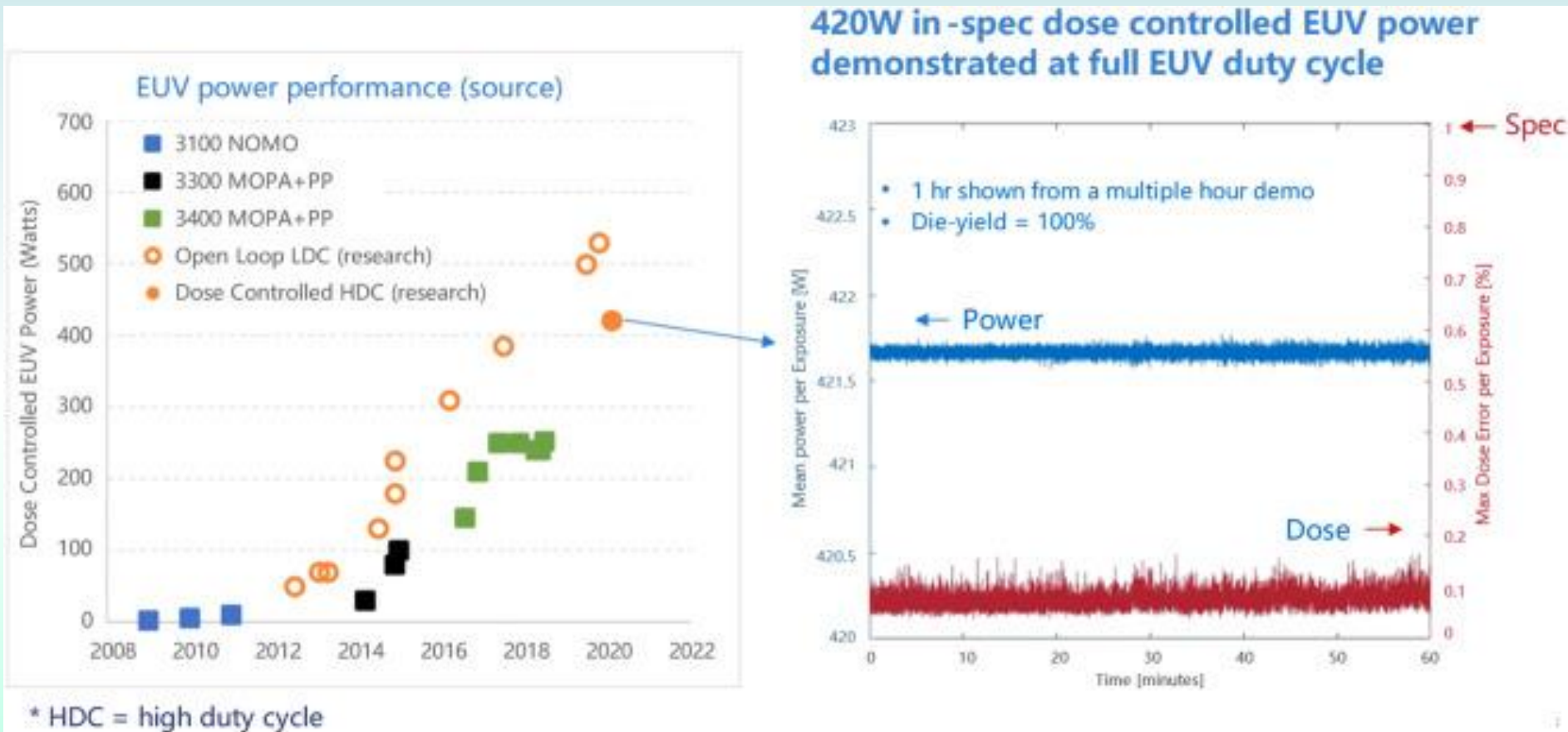
In 2004, it is predicted that EUVL will become mass production tool in 2009. Today it is believed that DUV lithography ( $\lambda=193\text{nm}$ ) with double processing will be used for 32nm generation production, thus delaying the need for EUV.



在最小的節點，*EUV*微影顯然在圖形化功能上超越利用氬氟(*argon-fluoride*，*ARF*)光源的微影。  
(來源：ABI Research)



# Historical EUV power scaling trend using LPP sources.

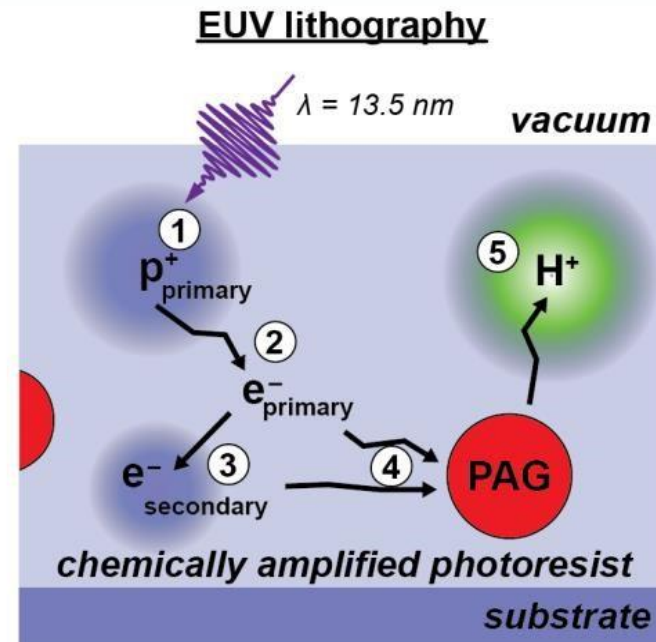
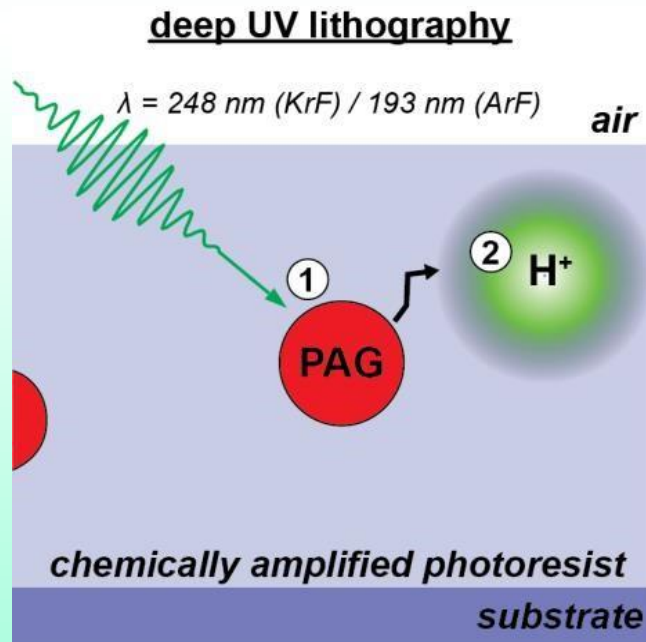
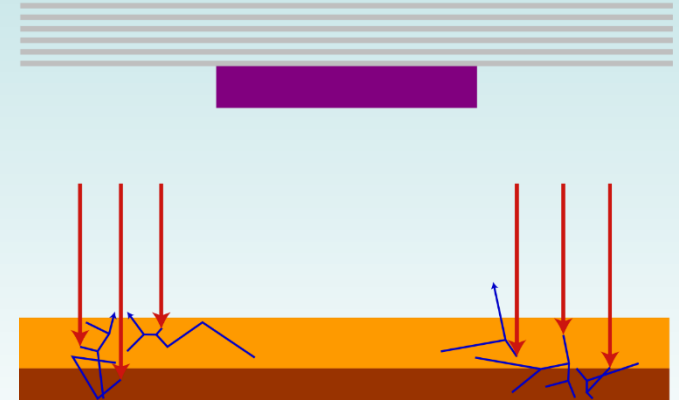


Peter Mayer, David C. Brandt, Igor Fomenkov, Michael Purvis, and Daniel Brown "Laser produced plasma EUV sources for N5 HVM and beyond: performance, availability and technology innovation", Proc. SPIE 11609, Extreme Ultraviolet (EUV) Lithography XII, 1160918 (22 February 2021)

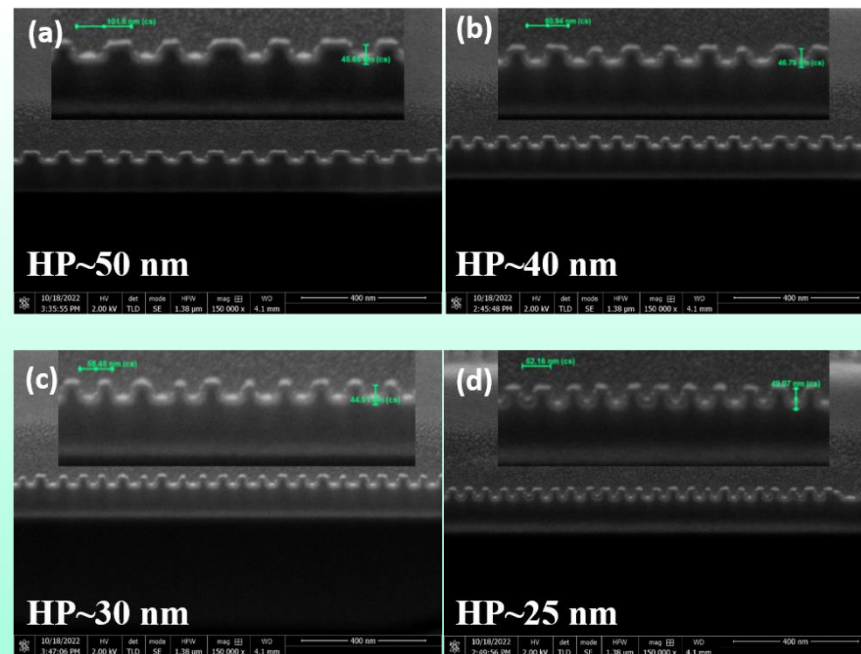
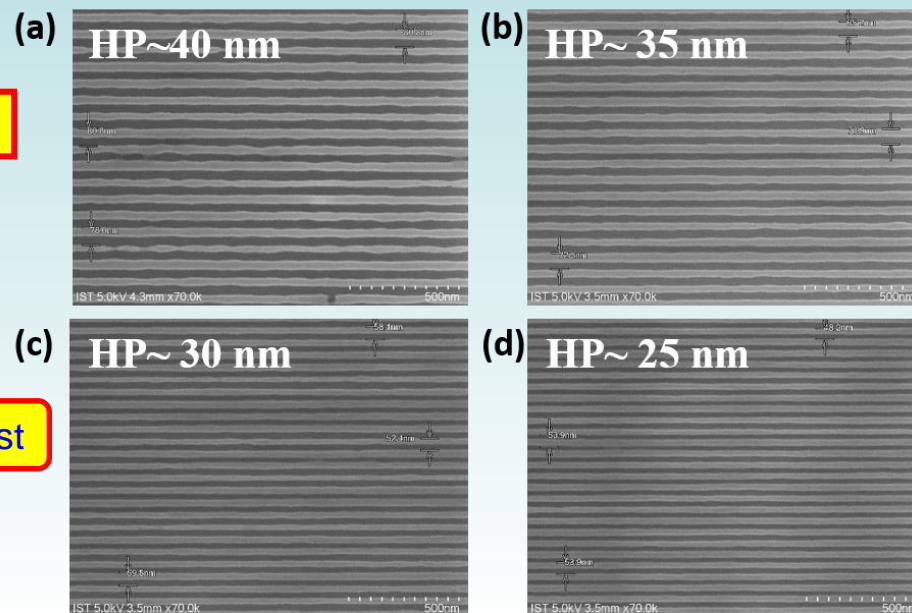
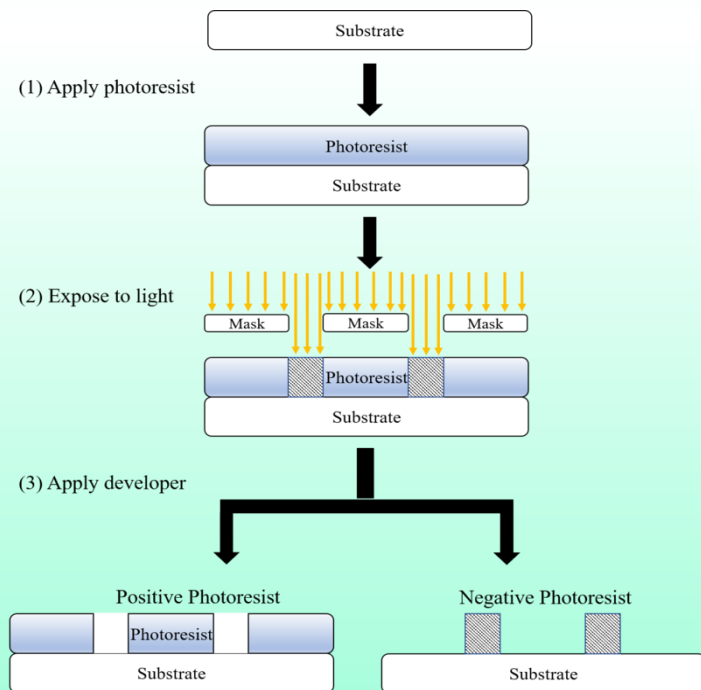
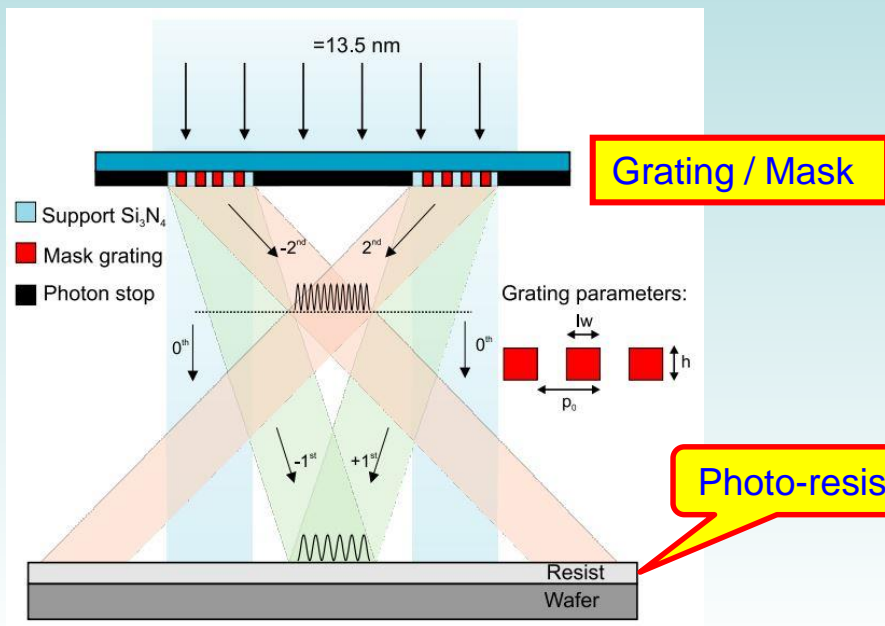


# EUV Photoresist

- Photoacid Generation in EUV lithography is significantly different from DUV
- New chemistries required
- Candidate resists must be studied under EUV illumination before use in manufacturing



Photoacid Generator (PAG)



# Photo-resistor contamination

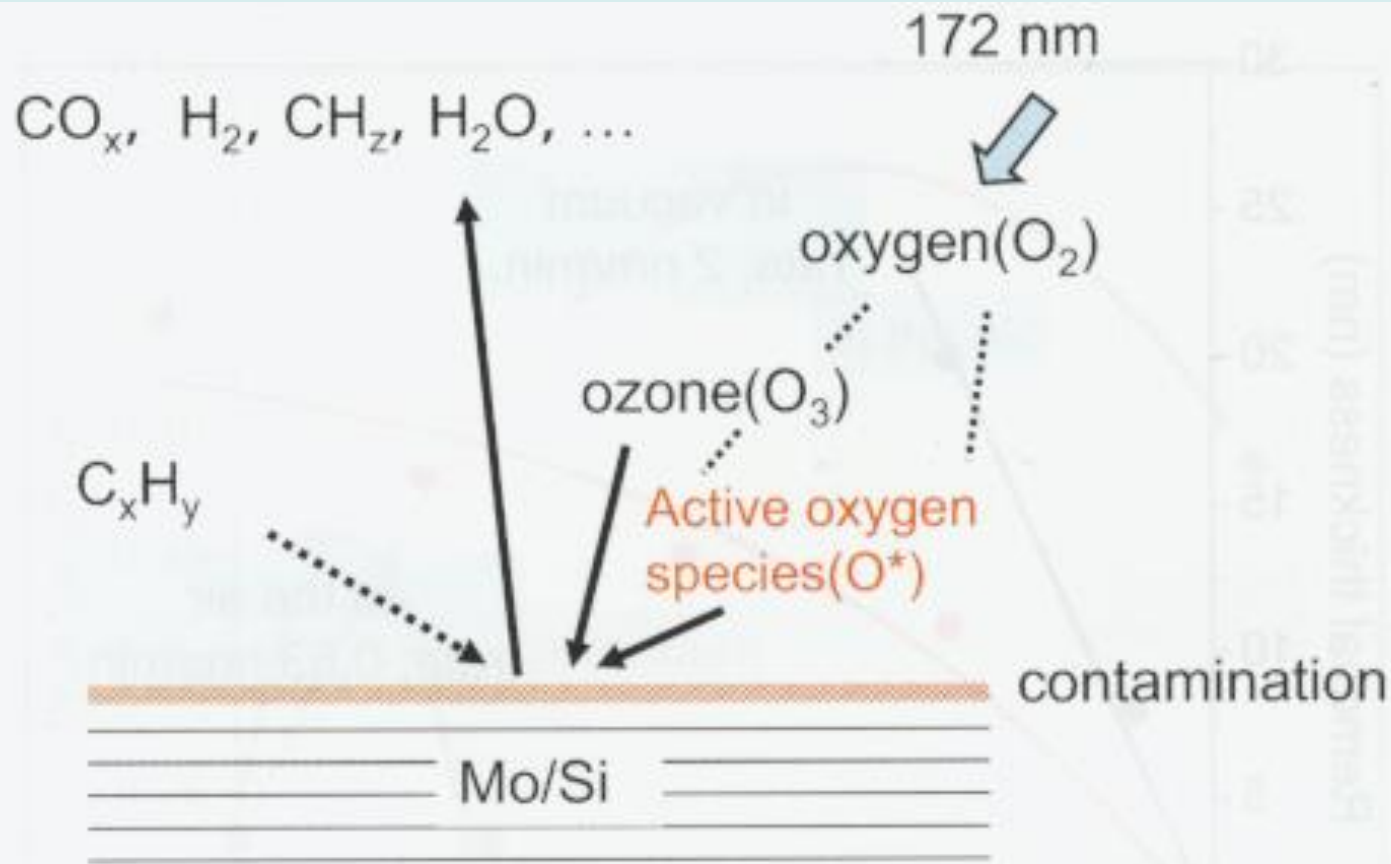
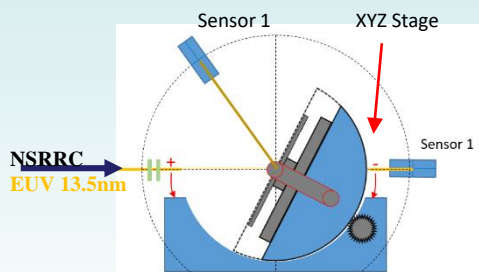


Figure 37. The mechanism of mask cleaning using 172 nm excimer lamp.

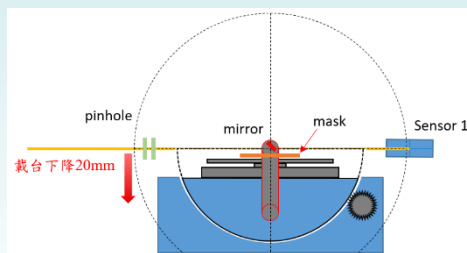
# 檢測平台

## 反射



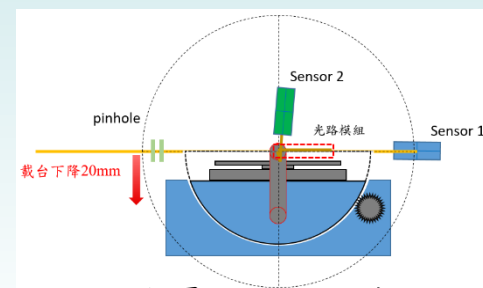
反射率檢測-中央郭倩丞

## 干涉

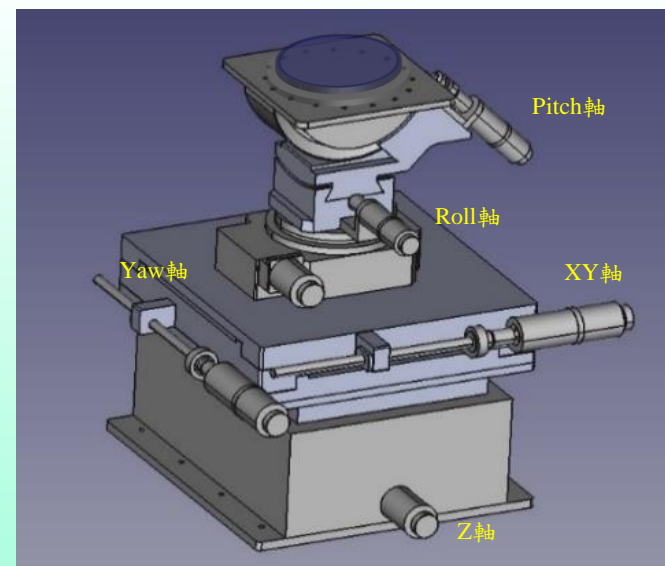
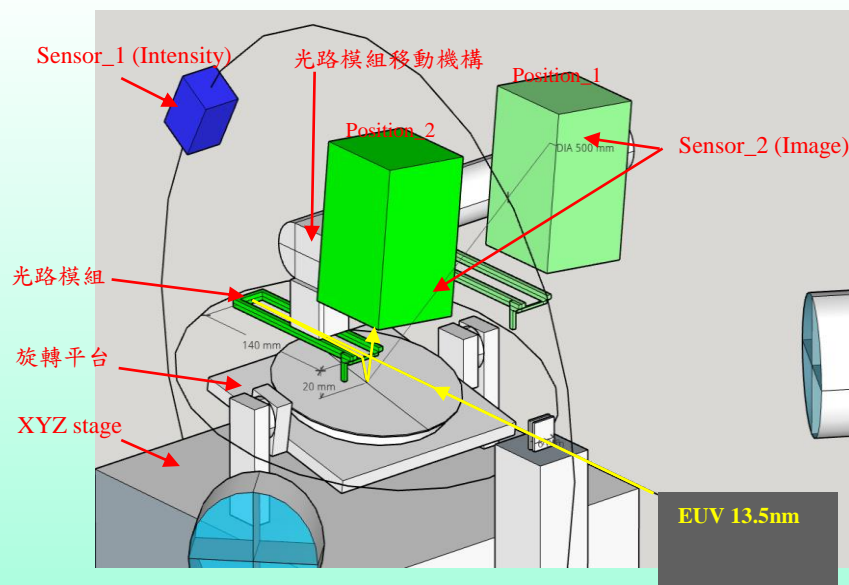


干涉微影-台大蔡坤諭

## 繞射



光罩檢測-台大李佳翰



# 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

