

Achieve the Time-resolved Experiments Using Pulsed X-ray Source Based on Synchrotron Radiation

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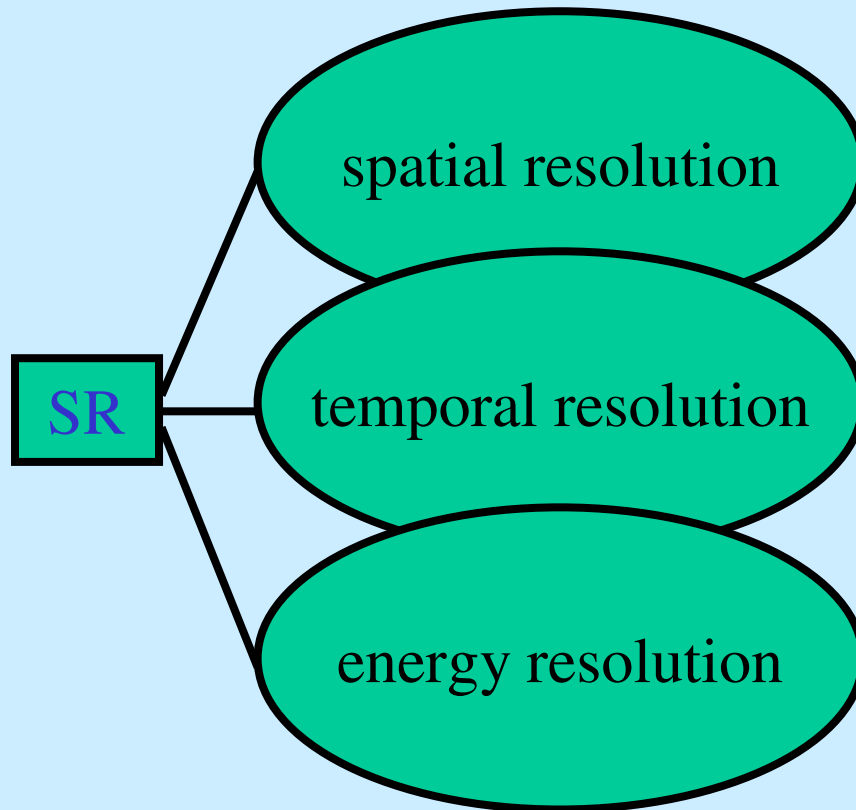
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Outline

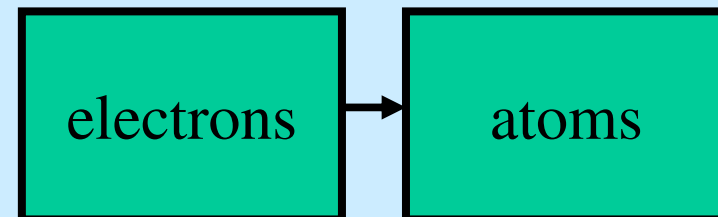
- Timing system for synchrotron radiation
- Generation of pulsed X-rays
- Time-resolved VUV spectroscopy at BSRF
- Laser pump-X-ray probe XAFS experiment
- Some concerns for pump probe experiments

Why time-resolved experiment



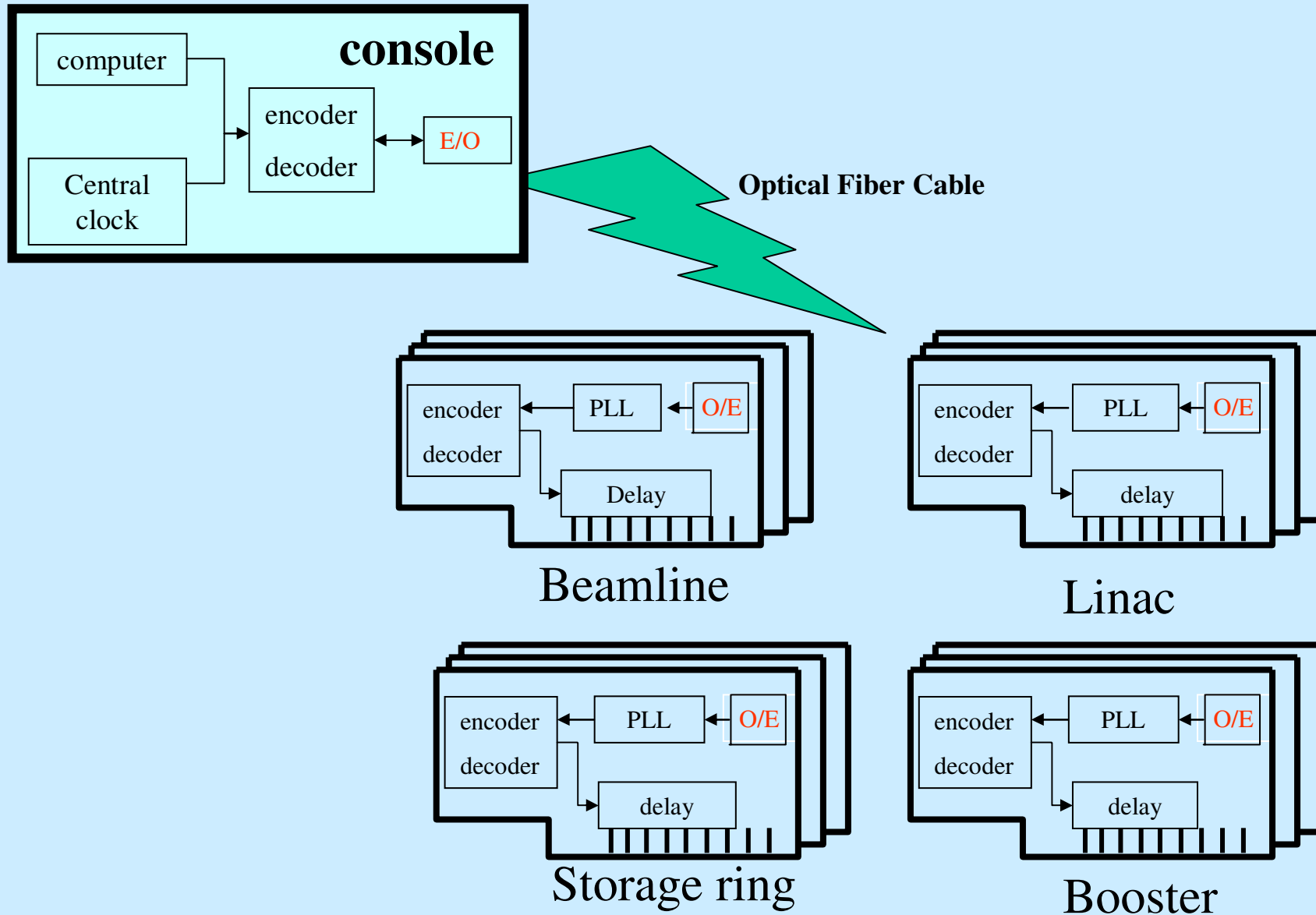
fast scan, fast data acquisition, pump-probe technique

- electronic lifetime spectra of excited states, metastable state
- ultrafast chemical reactions
- ultrafast phase transitions
- surface dynamics
- ultrafast biological processes

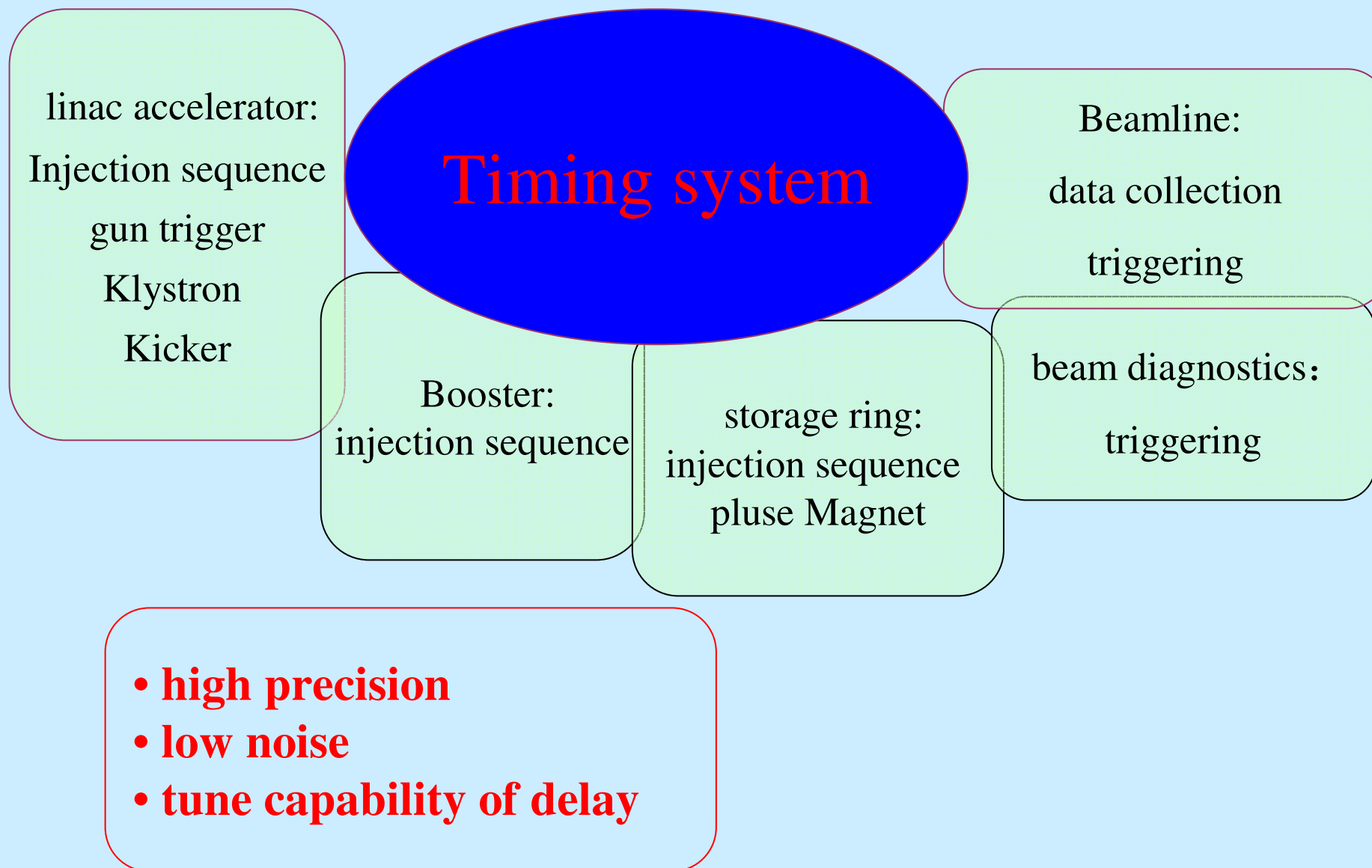


visible-ultraviolet-X-ray

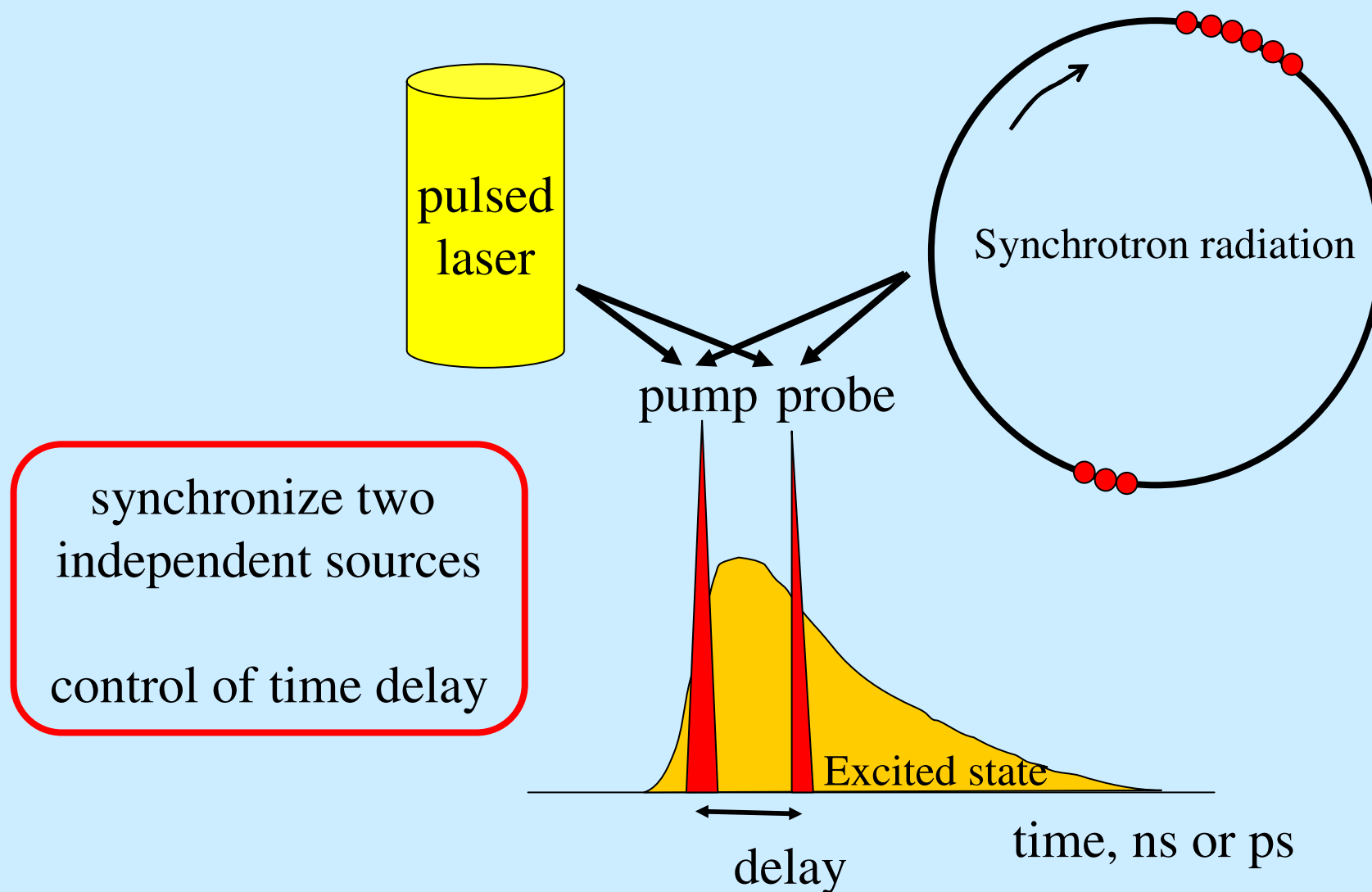
Timing system in synchrotron radiation



Control of timing system



Pulsed light source in time-resolved experiment



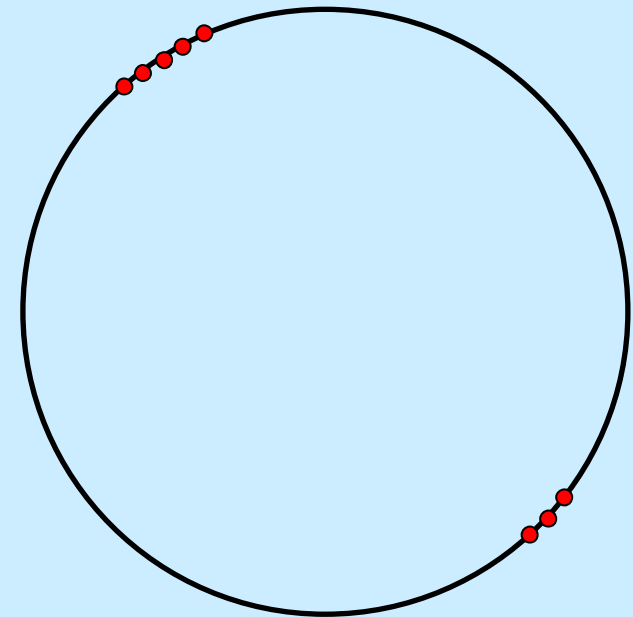
Fill mode at storage ring

BSRF

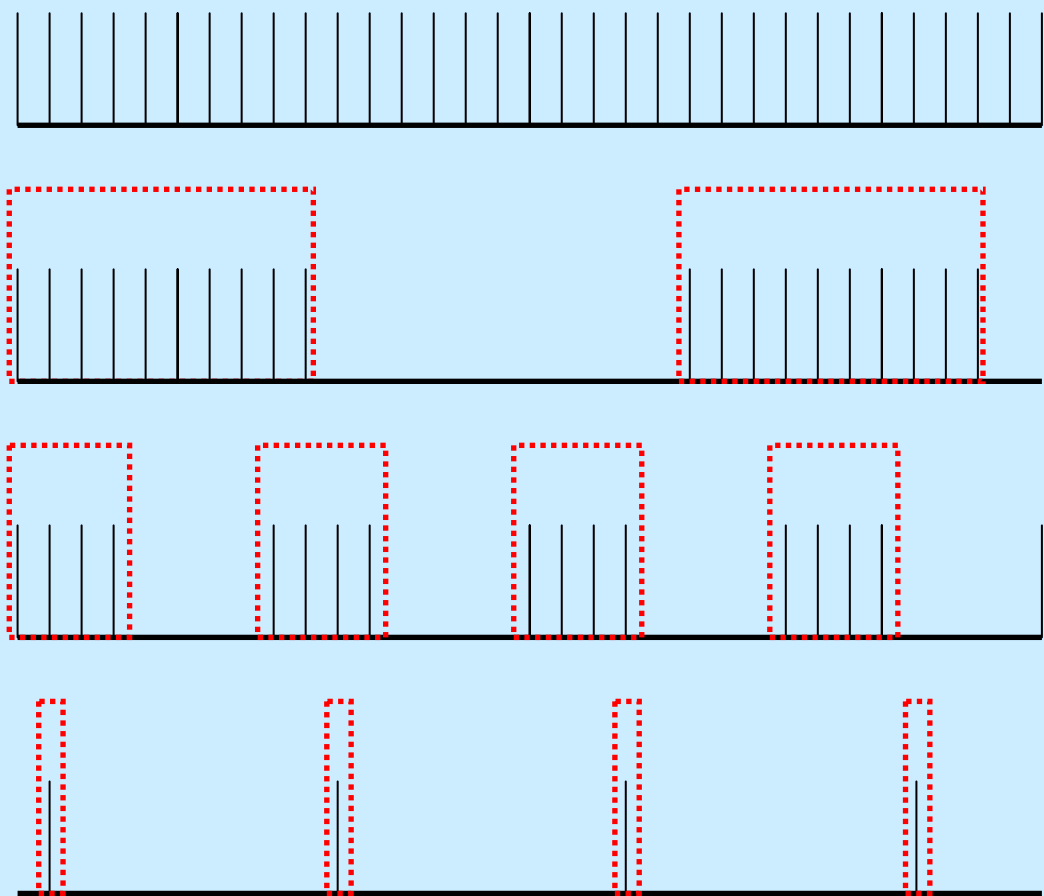
- RF 200MHz (500MHz for BEPCII)
- Multi-bunch fill : 100 bunches, 160-260ps in width (50ps for BEPCII)
- Single-bunch fill: period 800ns, 160ps in width

APS

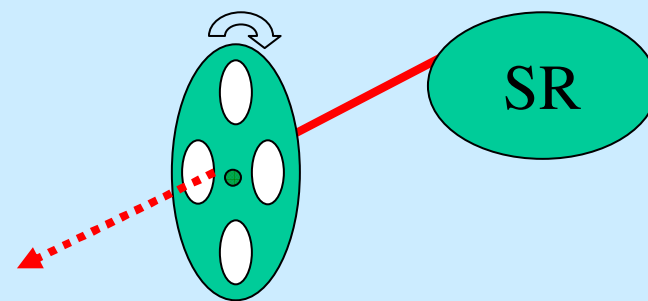
- RF 352MHz
- Routine: non-Top up operation 23 singlets
- Routine: Top up operation 23 singlets, continuous top-up with refill
- Special operating mode (SOM)
 - hybrid fill $1+8\times 7$
 - hybrid fill $3+8\times 7$



Synchrotron radiation : pulsed X-ray source



Rotating shutter

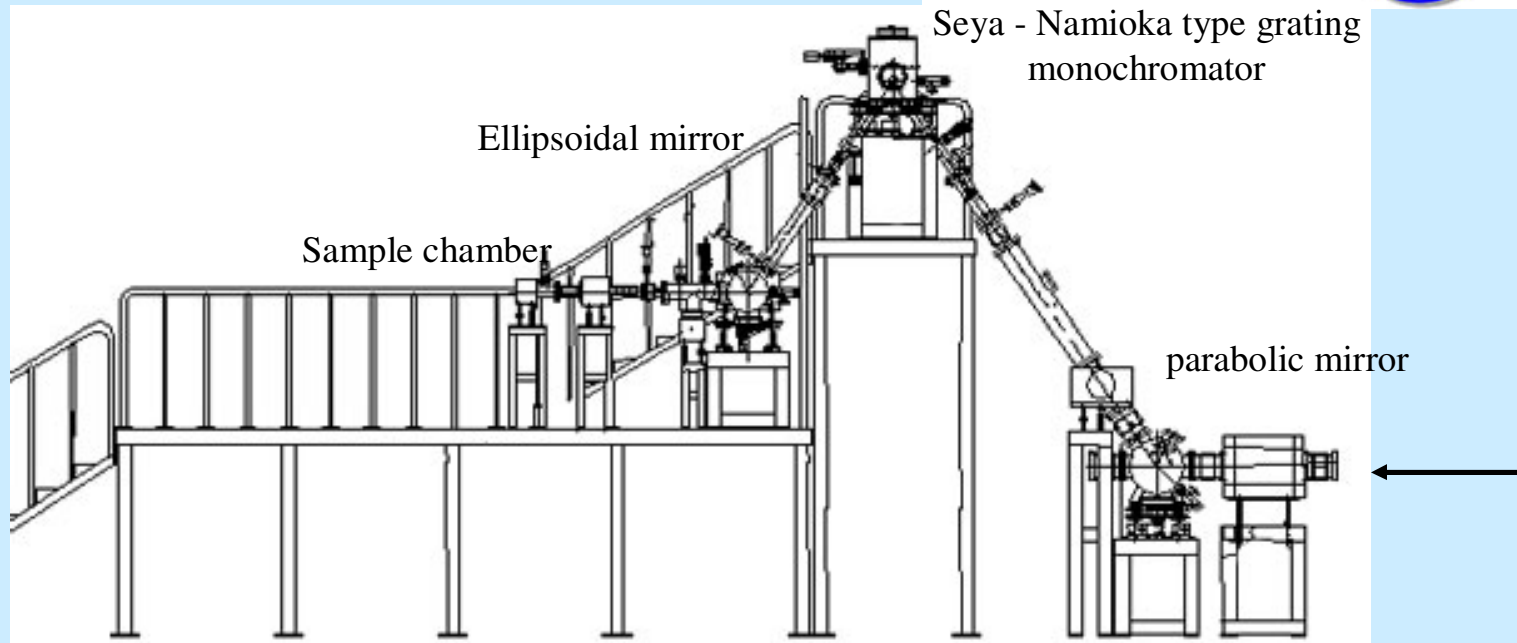
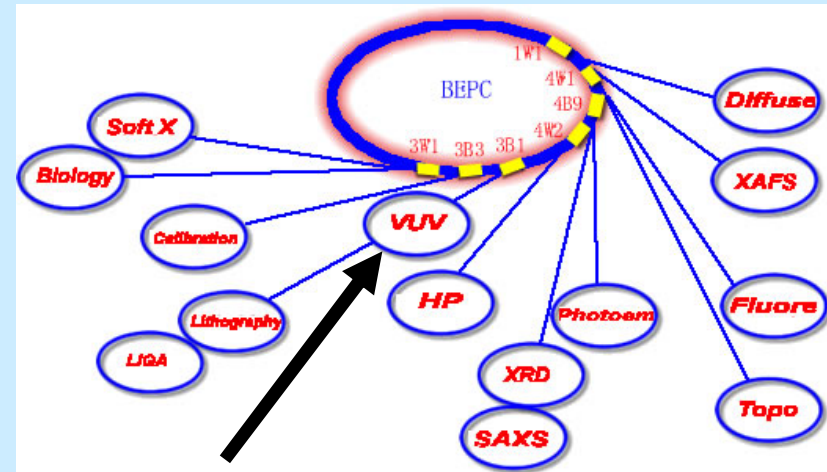


Sliced X-ray pulses

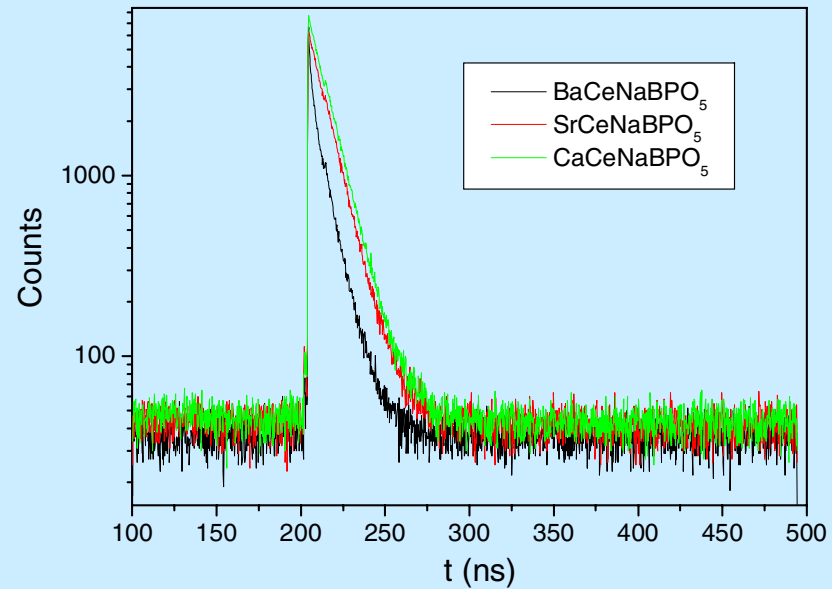
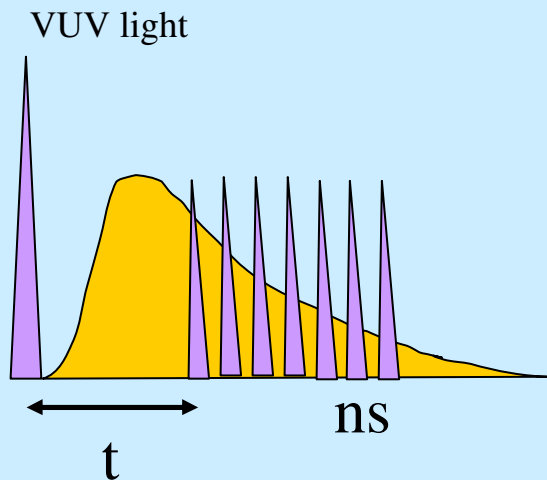
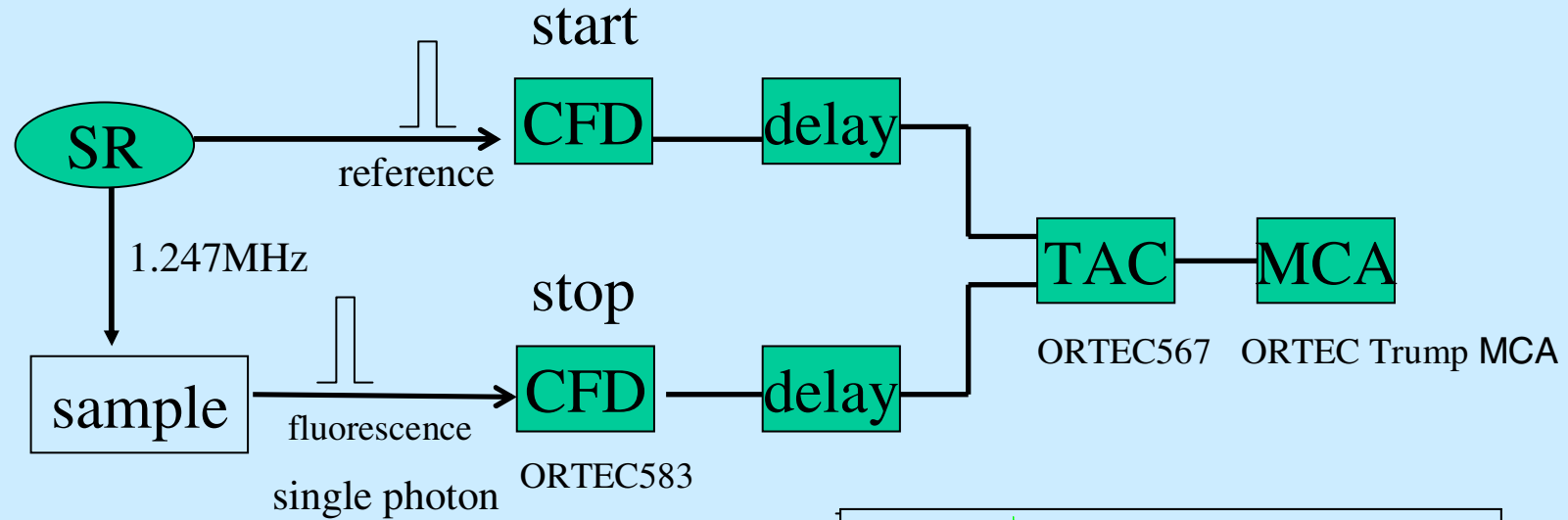
Synchronized with single bunch

Single photon timed-resolved spectroscopy at beamline 3B1B, BSRF

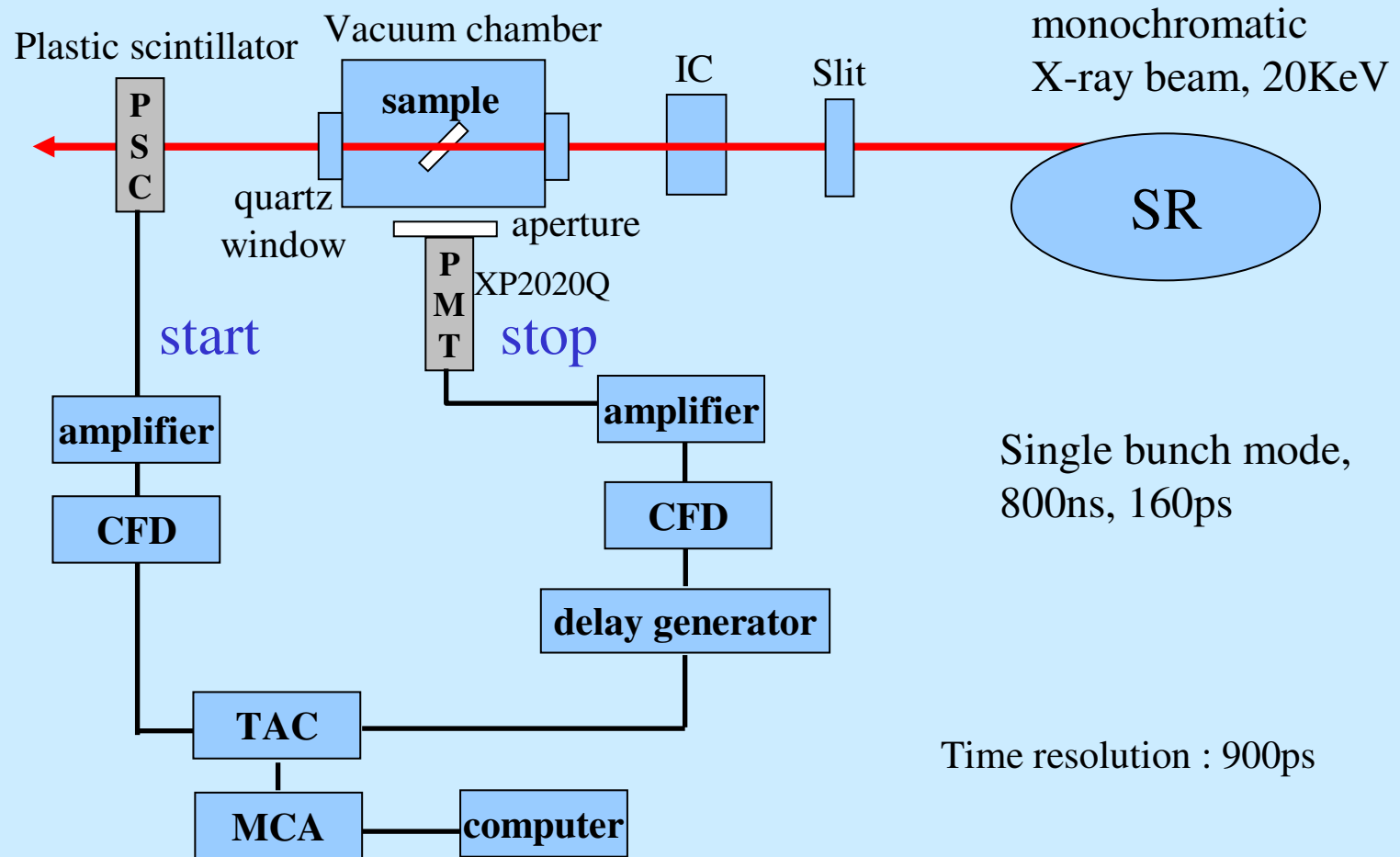
Wavelength Range: 100 nm - 550 nm
Resolution: 10^{-3}
Photon flux: 10^9 photons/sec
Beam size: 3 mm (H)*1 mm (V)



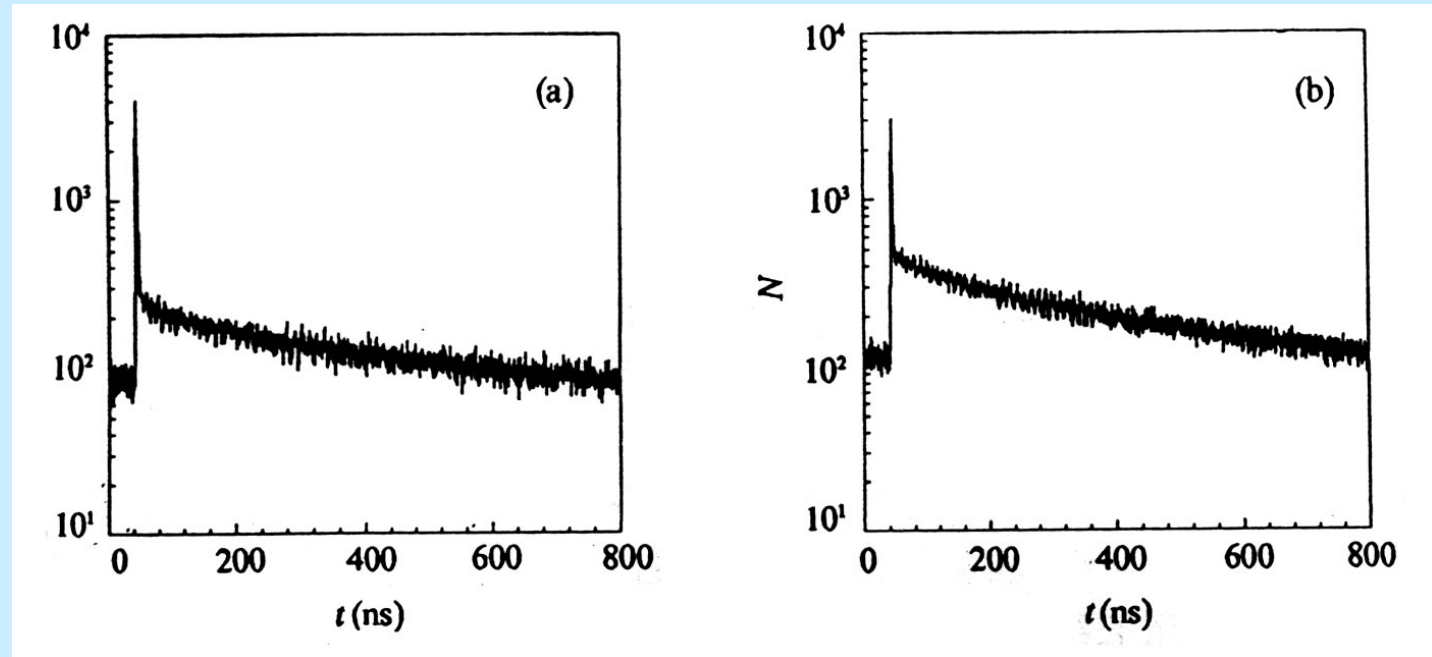
Time correlated single photon counting



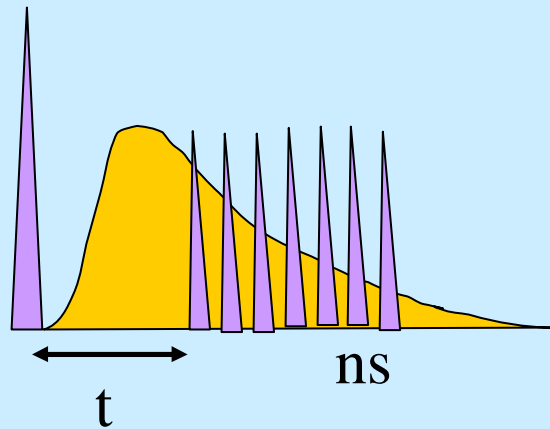
A time-resolved experiment performed at beamline, 4W1B



Fluorescence lifetime spectra of BaF₂ crystal doped with Ce³⁺

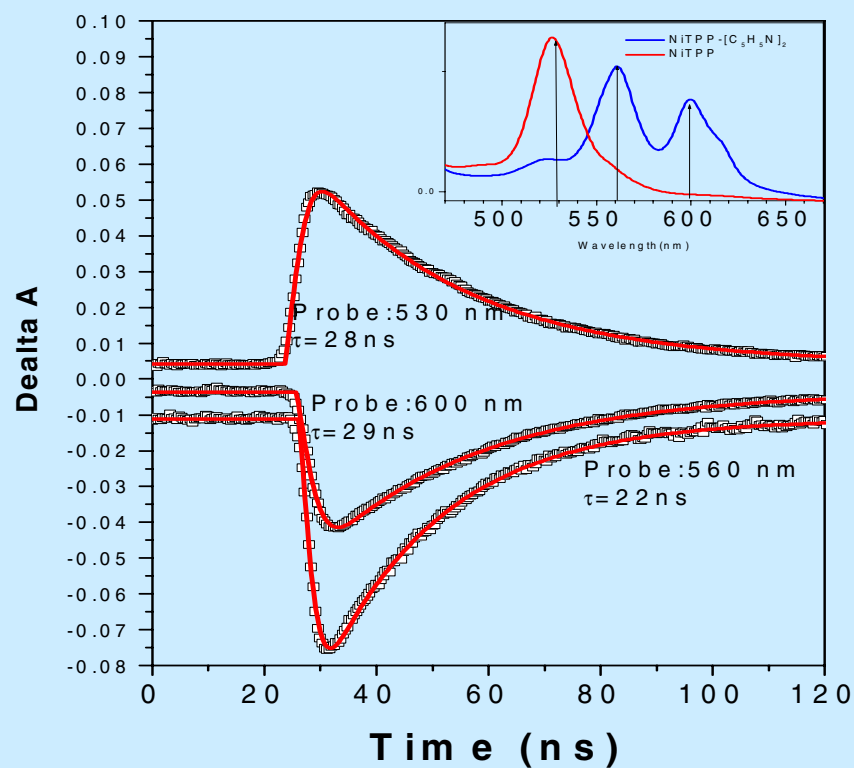
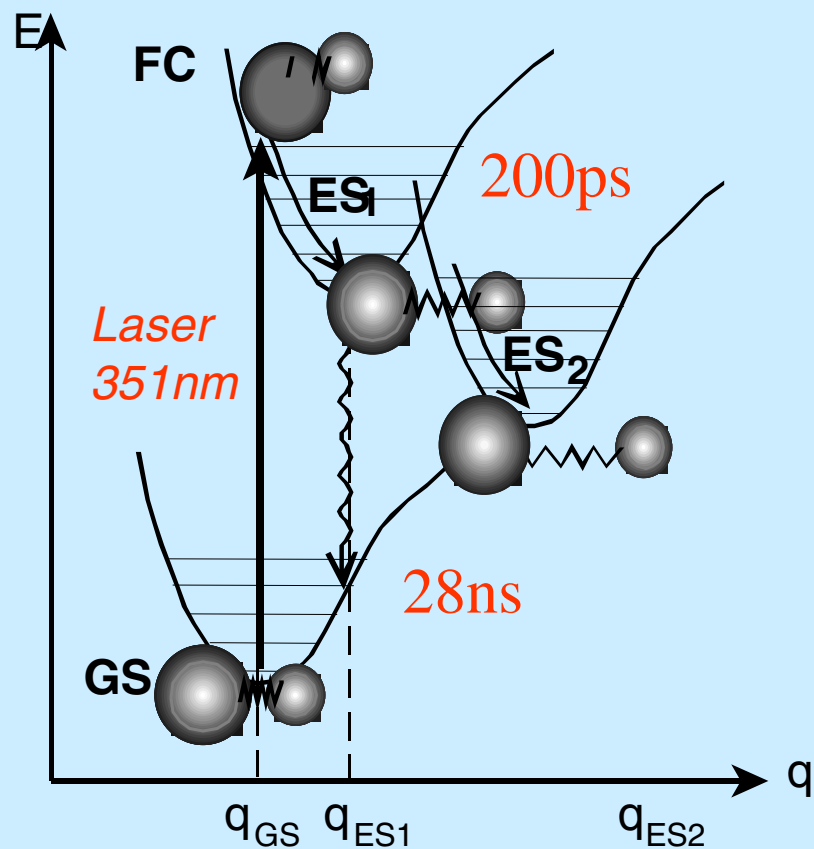


X-ray, 20KeV



(a) 0.56mol%, (b) 1.64mol%

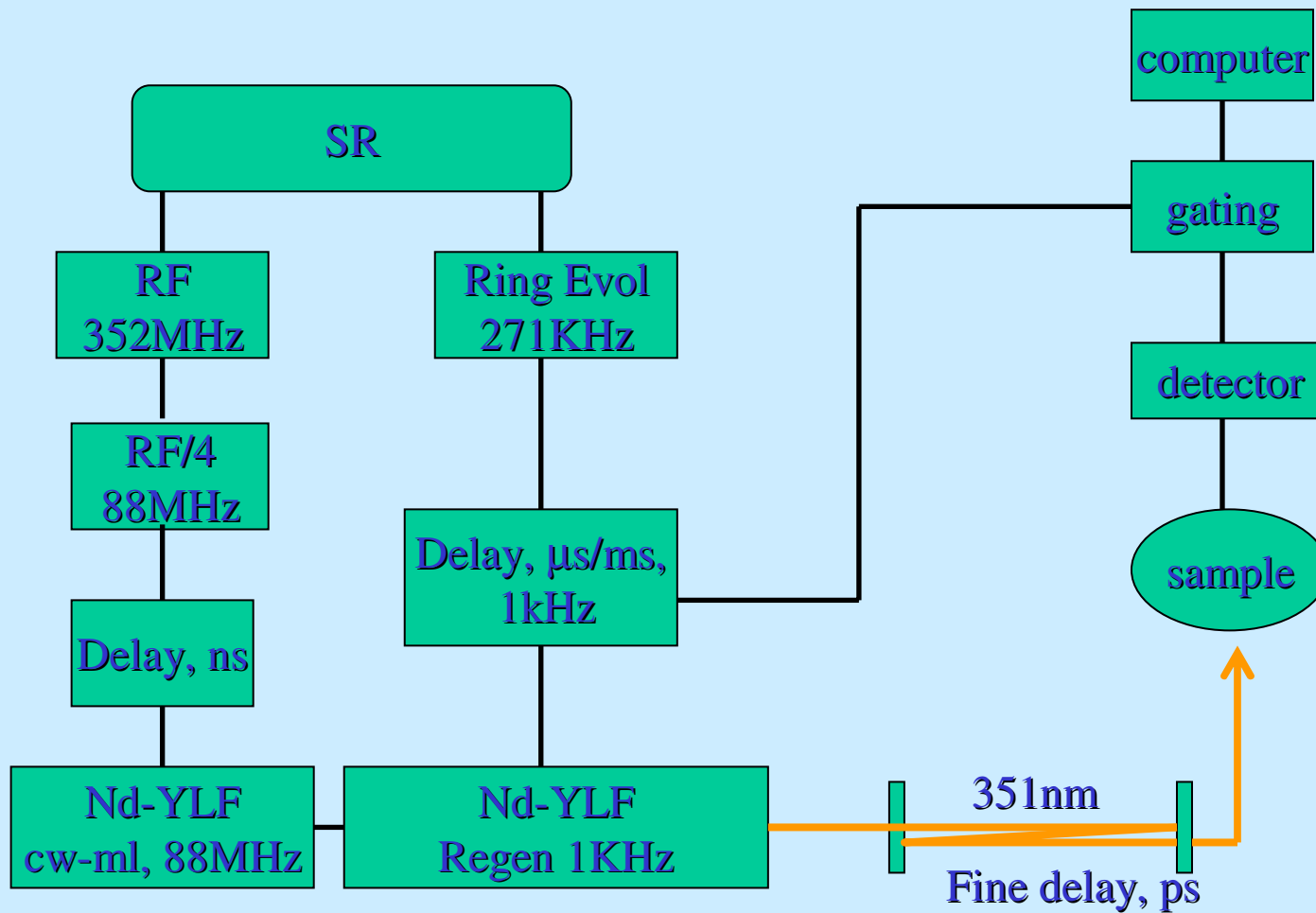
Probe the excited state induced by light in molecules



Time evolution of optical absorption change monitored at characteristic wavelengths for unchelated (530nm) and bi-axial chelated (560 and 600 nm) NiTPP in piperidine solution using 351 nm pulse as the pump and a xenon lamp as the probe.

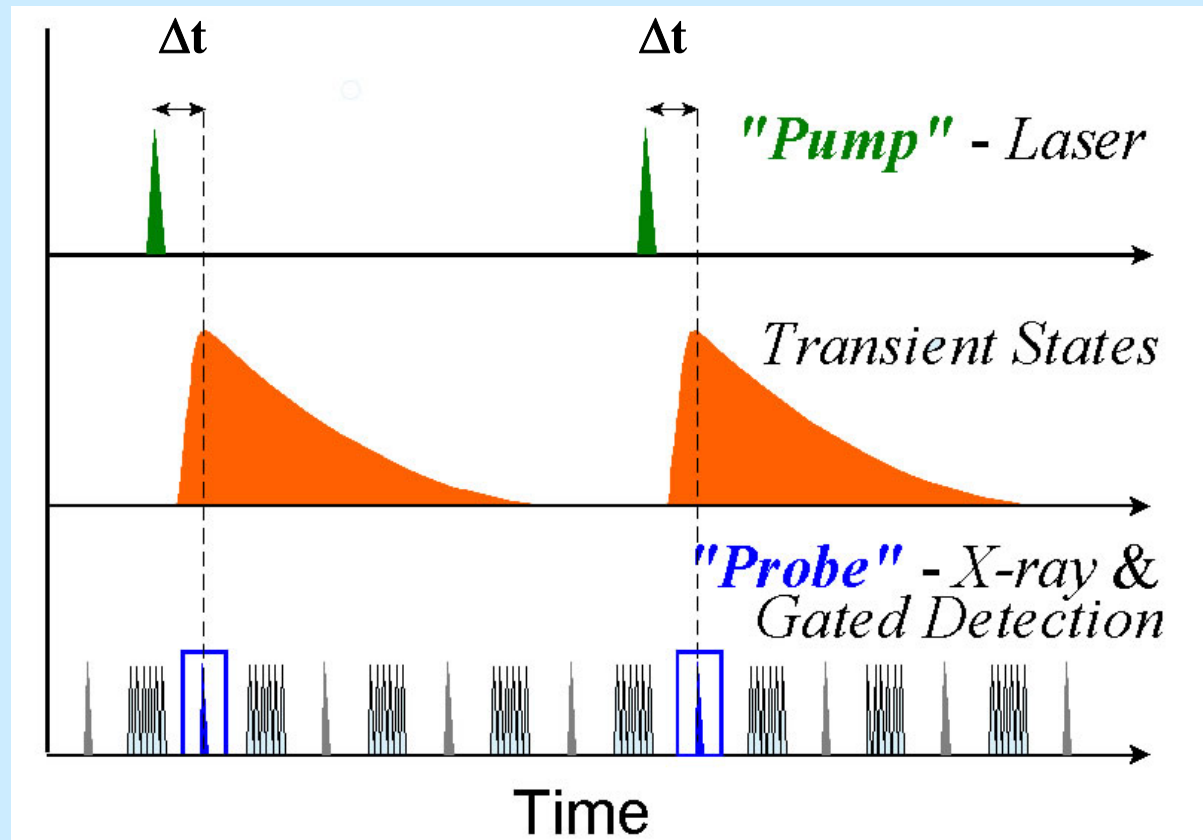
Timing-system for laser pump and X-ray probe XAFS at APS

ID11D, BESSRC-CAT



Guy Jennings, et al. Rev. Sci. Inst. 73, 2002, 362

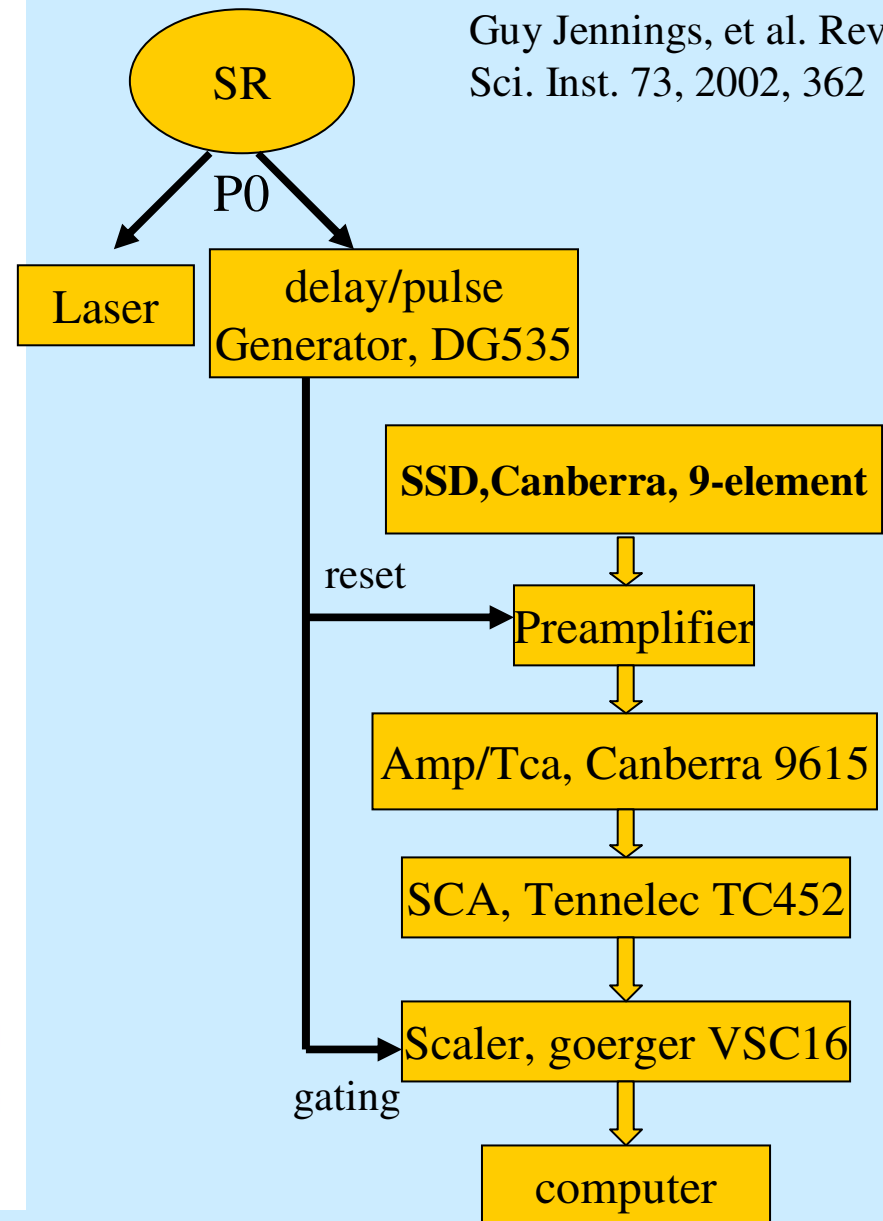
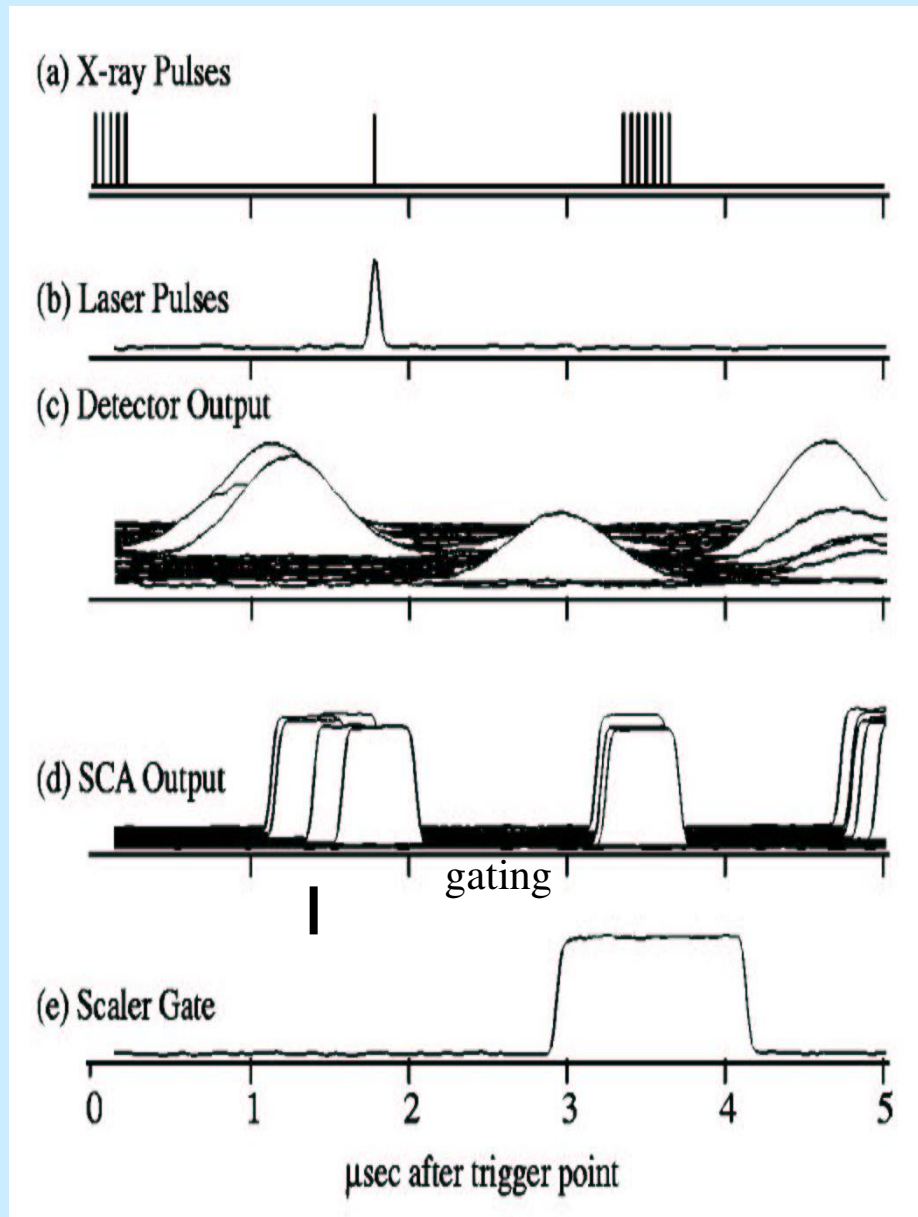
Time-resolved laser pump-X-ray probe techniques

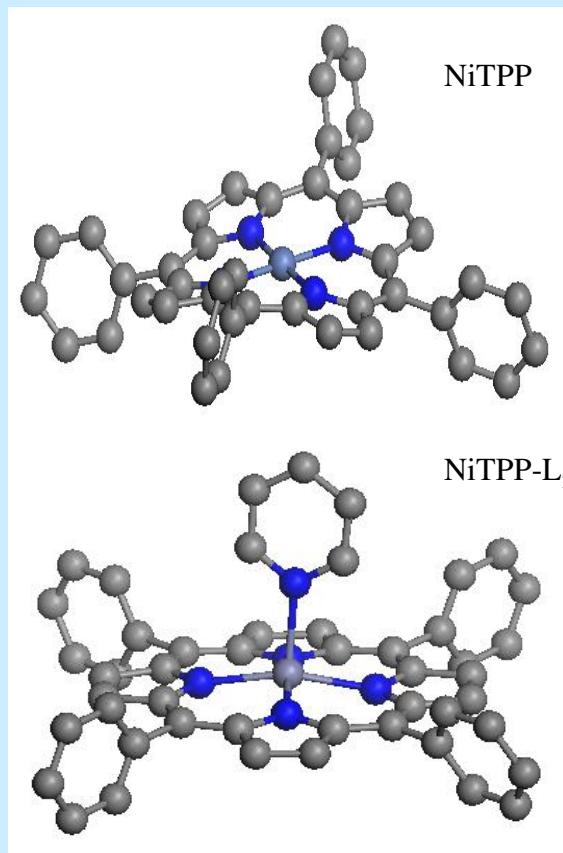
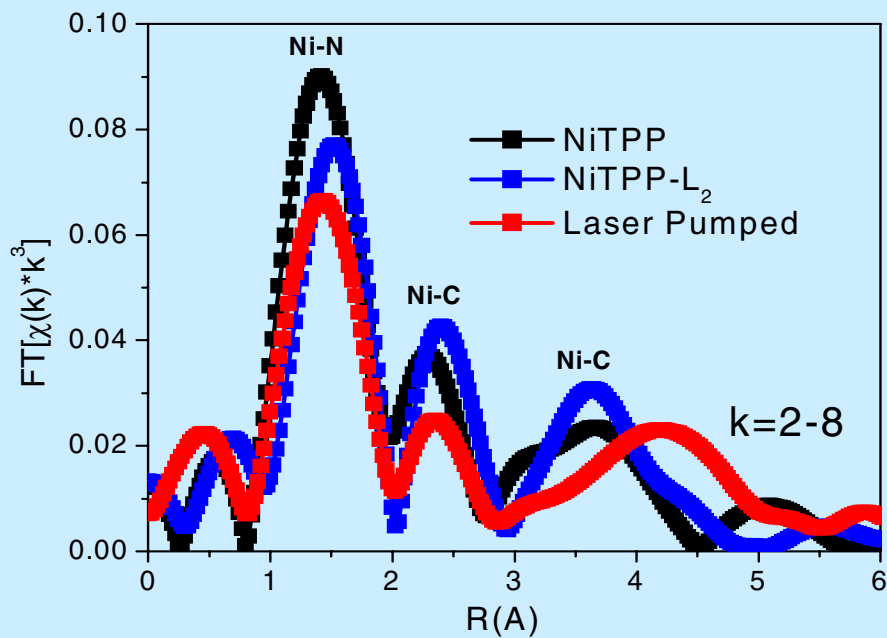
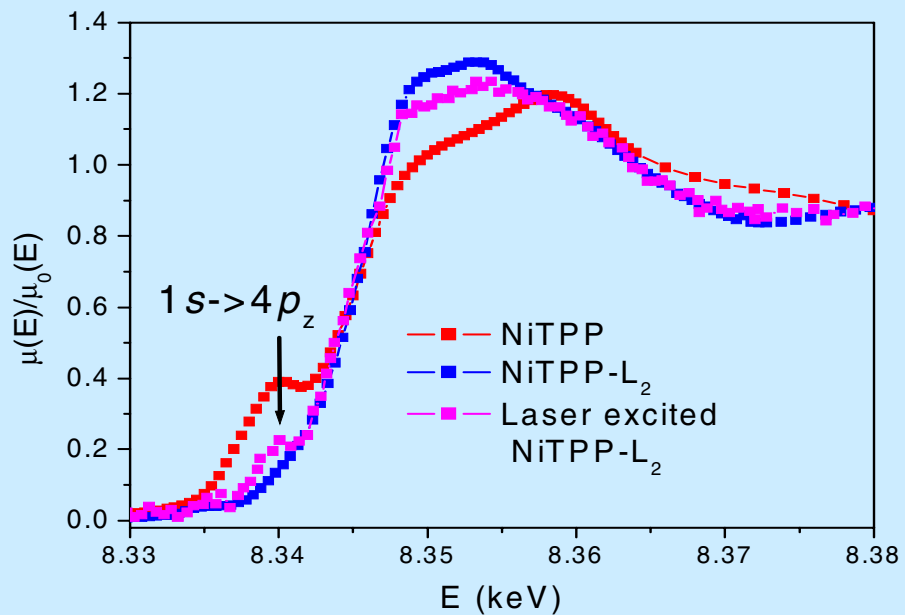


Time sequence of the laser pump/X-ray probe experiment, which requires synchronization of laser pulse, X-ray pulses and gated detection, Δt .

Readout electronics

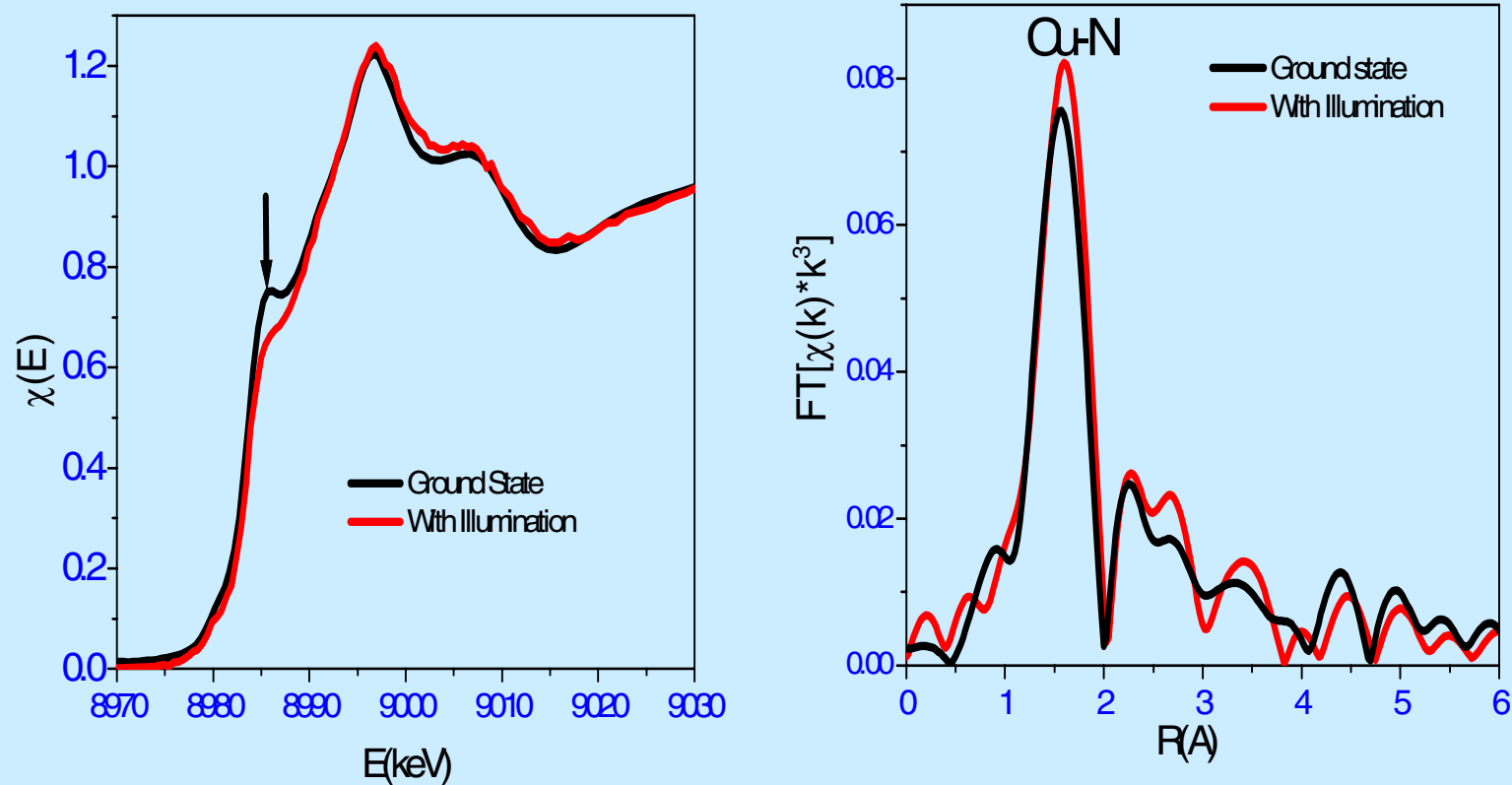
Guy Jennings, et al. Rev. Sci. Inst. 73, 2002, 362





L.X.Chen, et al. Science, 2001, 292, 262

[Cu(dmp)₂](BArF) in toluene with and without light excitation



XANES spectra and Fourier transformed spectra of [Cu^I(dmp)₂](BArF) in toluene with and without light excitation.

L.X.Chen, et al. J.Am.Chem.Soc.2002, 124, 10861

Some concerns in pump-probe experiments

Pump-probe

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graph TD; A((Pump-probe)) --- B[synchronization]; A --- C[time resolution]; A --- D[sample damage due to high flux!]; A --- E[laser and X-rays make differences!]; A --- F[high flux signal to noise ratio]; B --- B1[• high frequency X-ray]; B --- B2[• low repeated laser]; B --- B3[• low response detector]; E --- E1[Laser : very surface for solid, valence and free electron]; E --- E2[X-rays : bulk, inner shell];
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synchronization

- high frequency X-ray
- low repeated laser
- low response detector

time resolution

sample damage
due to high flux!

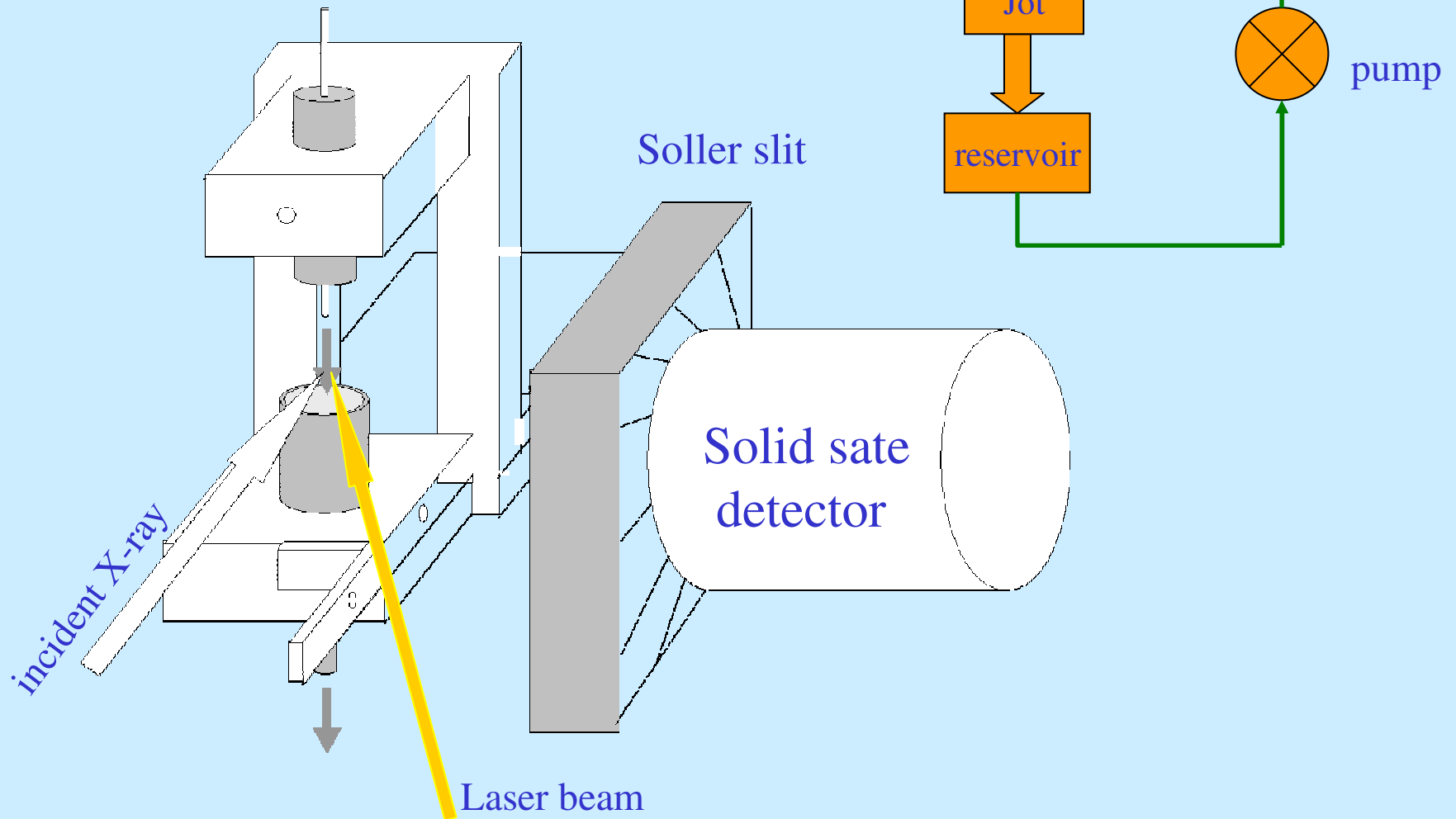
high flux
signal to noise ratio

laser and X-rays make differences!

Laser : very surface for solid,
valence and free electron
X-rays : bulk, inner shell

Jot sample system for pump-probe experiment

Sample damage due to high flux of laser and X-rays!



Why 3d generation synchrotrons in pump-probe experiment

The fluorescence photons from a sample can be estimated by

$$I_f = I_a \times (\Omega/4\pi) \times \eta \times (\mu_k/\mu_T) \times \eta_{\text{Det}}$$

I_f : fluorescence signal, I_a : number of photon absorbed by the sample, Ω : solid angle covered by the detector, η : quantum yield of the fluorescence, μ_k, μ_T : the absorption cross sections of the atoms of interest and of the whole sample, η_{Det} : detector efficiency

For 1mMol Ni-containing sample(NiTPP), 1mm² area and 0.5mm in thickness (15% absorbed), $I_a=4.5 \times 10^7$ photons at 1KHz of pump-probe cycle, $\Omega/4\pi=2.9\%$, $\eta=40\%$ for Ni, the fraction of Ni in sample=0.6%, $\eta_{\text{Det}}=1$

3rd-generation SR
(5.5×10^{11} photons/s)

$I_f = 3100$ photons/s, for a total count of 100000, 35s integration time is required for each point

1st or 2nd-generation SR
(10^9 photons/s)

$I_f = 0.2$ photons/s, 110h is needed!

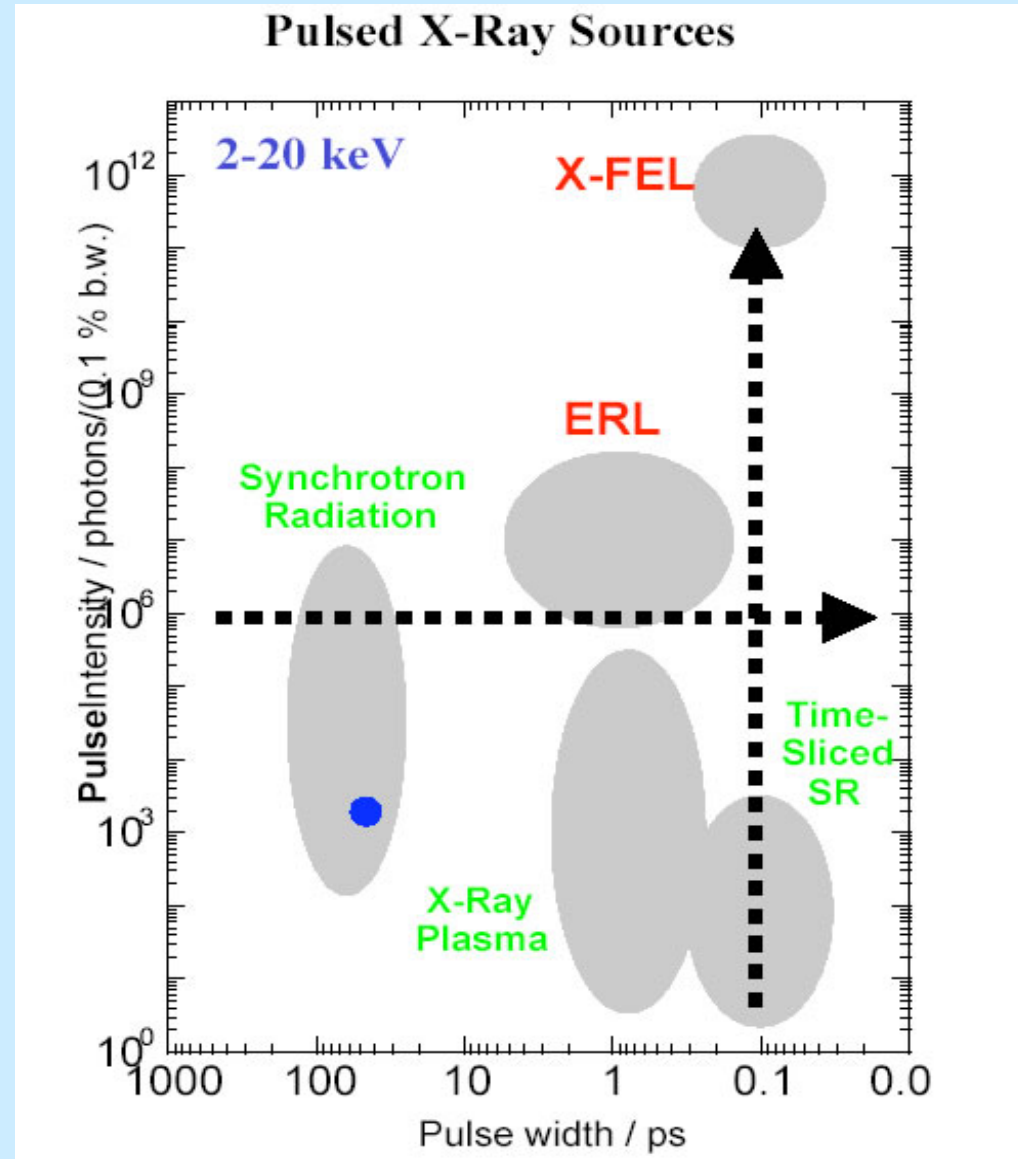
Time Resolution

Time resolution defined
by the width of X-ray pulse

Time scale for atomic
motion, vibrational period
~100fs

Synchrotron radiation
~ 100ps

X-FEL
~ 100fs



Acknowledgements

Y. Tao

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