FEMTO: Ultrafast Tunable X-Ray Source

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Motivation: Why femtosecond $\simeq$ 1 Angstrom X-Rays ?

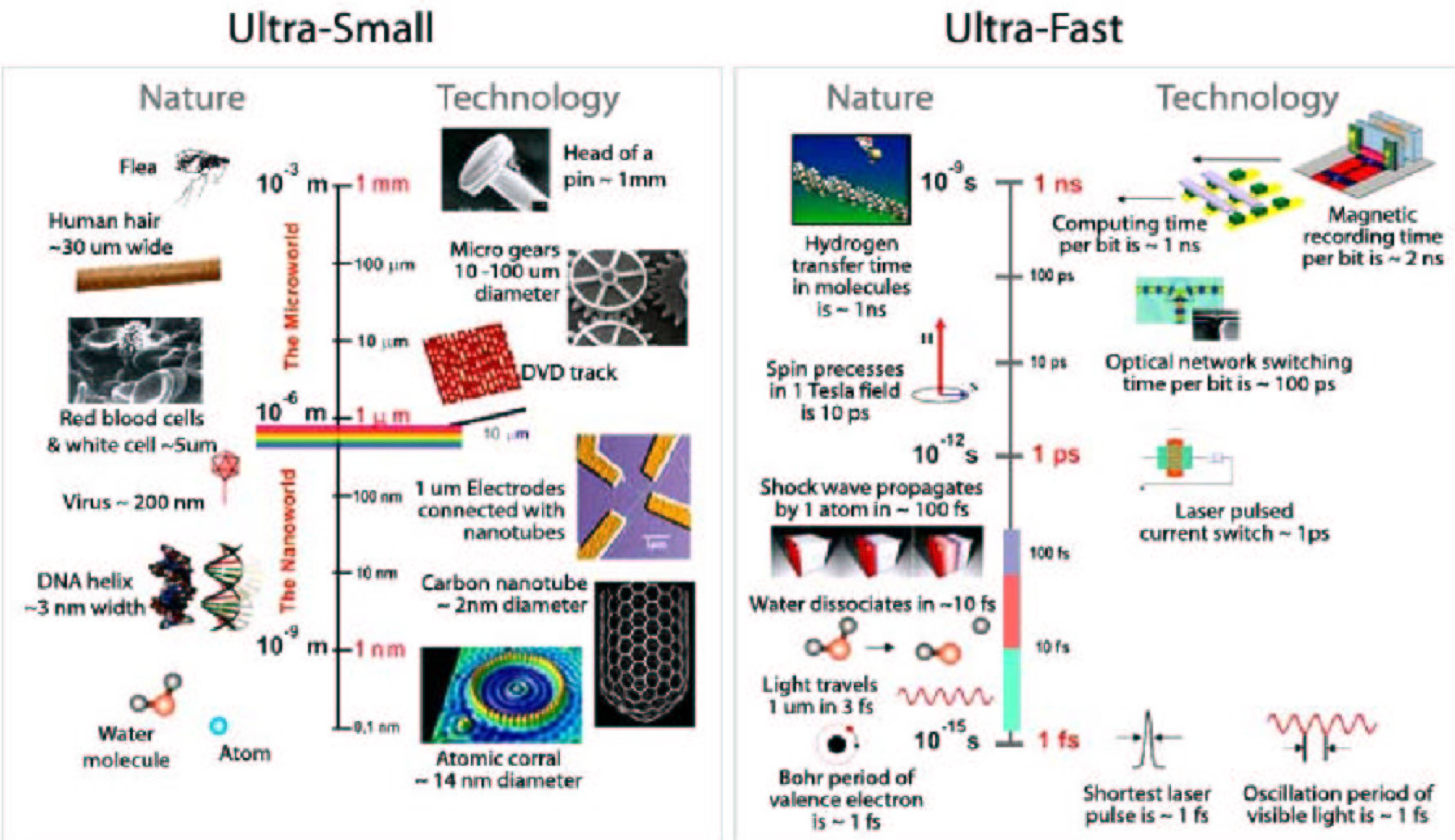
- time scale of chemical reactions: fs
- X-ray: wavelength of atomic scale
- fs-X-ray pulse → “4D imaging with atomic resolution”

- ultrafast chemistry & biology:
  - ❑ conformational changes
  - ❑ electron transfers in molecules
- phase transitions in material science

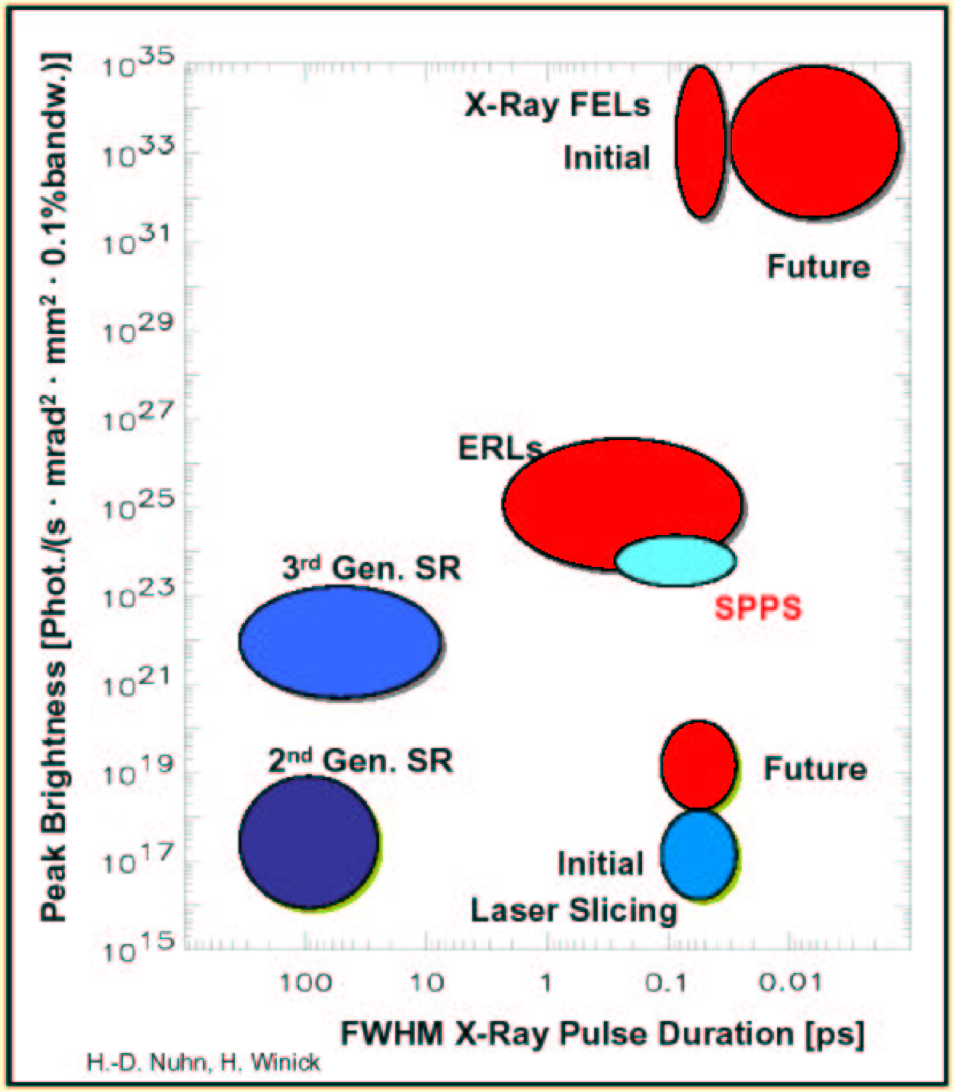
Motivation: Why femtosecond $\simeq$ 1 Angstrom X-Rays ?

- advantage of photons:
  - ➢ photons as **atomic probes** both in *space* and *time*
    - ❑ energy of 15 keV corresponds to wavelength of 0.8 Å
    - ❑ pulses can be on scale of attoseconds (1 as = 10^{-18} s)
    - ❑ atomic scale of space = a_0 = 0.53 Å (Bohr radius)
    - ❑ atomic scale of time ≈ $2\pi a_0/v_0$ ≈ 150 as ($v_0 \approx c/137$)
  - ➢ X-ray photons can penetrate matter well beyond surface

Why do we need fs 1 Å x-rays ?

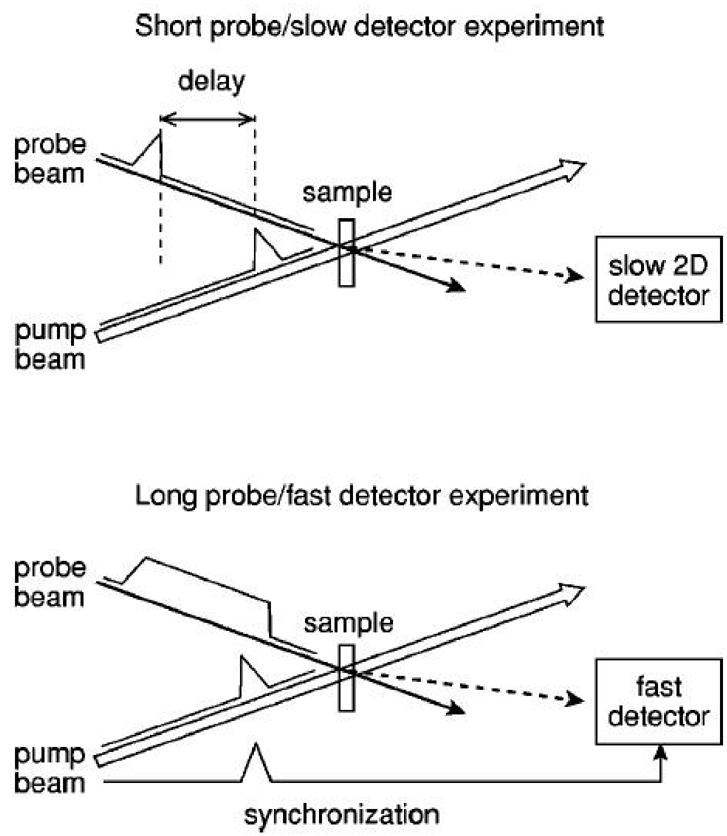


Ultrashort X-Ray Sources



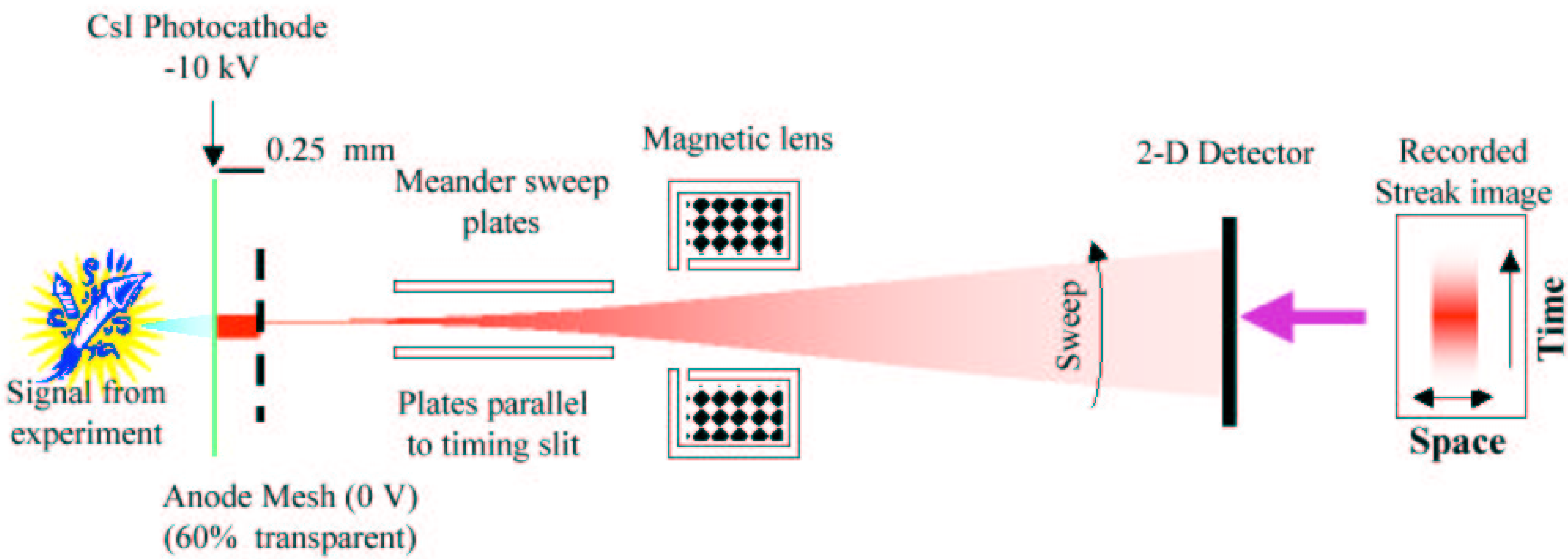
Ultrafast x-ray sources will probe space and time with atomic resolution.

Pump-Probe Experiments: Ultrashort Bunches vs. Ultrafast Detectors



Fastest X-Ray Detector (Streak Camera) limited to $\simeq$ 1 ps (multi-shot)
$\Leftrightarrow$ Develop Accelerator based Short-Pulse X-Ray Facility using Lasers & Undulators
$\Leftrightarrow$ Strategy: use inherently synchronized 100 fs Pump/Probe Pulses

Ultrashort X-Ray Detector: Streak Camera



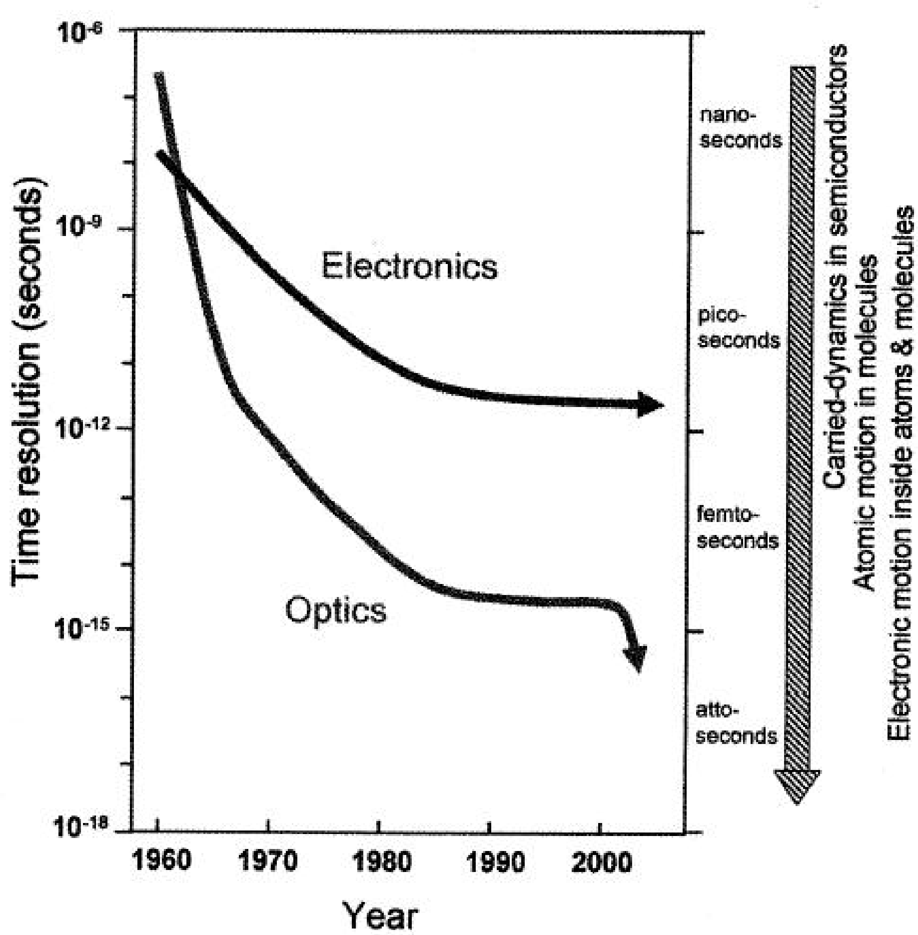
Single shot resolution: $\sim$ 0.5 ps

Jitter limited resolution: $\sim$ 2 ps (5000 shots)

But: detection quantum efficiency is typically low

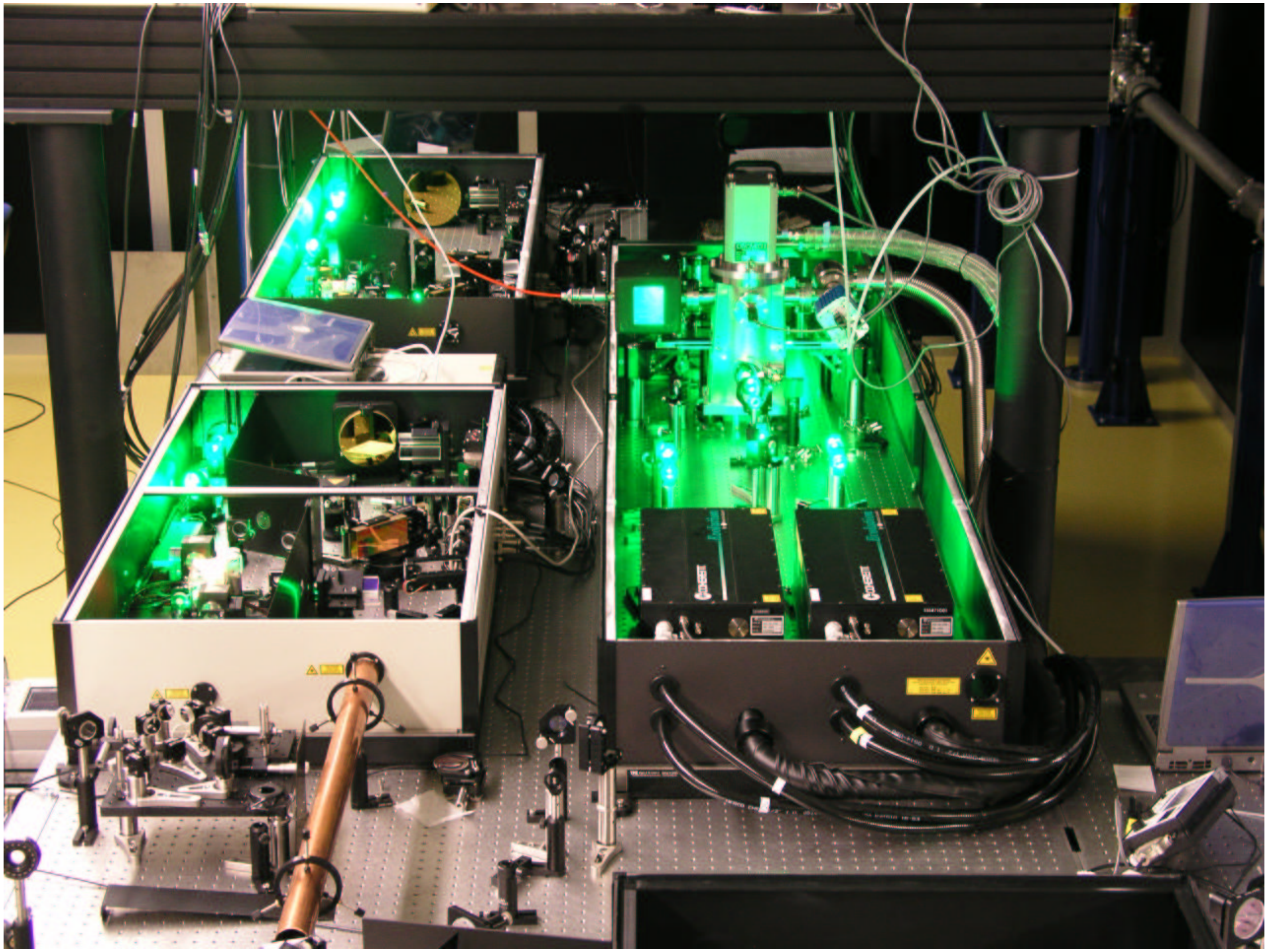
(R. Falcone, LBL)

Ultrafast Optics vs. Electronics



⇒ **Strategy: Use Optical fs-Lasers to Generate sub-ps Electron Bunches**

Installed fs Laser System to Generate Laser/X-Ray Pump/Probe Pulses



The FEMTO Project at SLS

- generation of femtosecond hard x-ray 5 – 18 keV
- planned time-resolved x-ray experiments:
  - x-ray absorption on condensed phase chemical systems
  - order – disorder phase transitions in condensed matter
- R & D for x-ray FEL:
  - laser - electron interaction
  - laser - accelerator synchronization
  - diagnostic of femtosecond x-ray pulses

X-Ray Source Characteristics

• Relativistic e-beam:	γ= E[MeV]/0.511 >> 10^2
• X-ray angular cone:	$\theta \sim 1/\gamma \leq$ 0.2 mrad
• X-ray energy:	0.1 - 20 keV
• Source point stability:	$\leq$ 1 μm, $\leq$ 1 μrad
• Undulator: [$\leftrightarrow$ spatial $\oplus$ temporal coherence]	high brightness
• Tunability: [$\rightarrow$ flexible lin./circ. pol.]	energy $\oplus$ polarization
• High harmonics:	suppression $\leftrightarrow$ operation
• Laser/e-beam interaction: [no Compton scattering, etc.]	resonant
• Short X-ray bunches:	50 ps $\rightarrow$ 100 fs
• Synchronization: [pump/probe: laser(100 fs)/X-ray(100 fs)]	"natural"
• XFEL - pulses: [$\rightarrow$ laser seeding $\leftrightarrow$ temp. coherence]	transform limited harmonic generation

Short Bunches: Slicing or Compression

	slicing	compression
e-beam	storage ring:	linac/FEL:
	'slicing' source 100 - 300 fs	chirped pulse compression 10 - 100 fs
X-rays	"Bragg-switch" $\sim$ 1 ns $<$ 100 fs ?	asym. crystals chirped pulse compression $<$ 100 fs
	monochromator $\oplus$ chirped pulse $<$ 10 fs	

Our Strategy to Generate sub-ps X-Rays:

- An electron (charge) moving at speed of light will emit electromagnetic radiation during acceleration: To produce sub-ps x-rays, we first have to produce sub-ps electron pulses.

⇒ Use electromagnetic fields (laser or rf-cavities) to modulate the energy of relativistic electrons with pulse length 10 - 100 ps [depending on the accelerator: linear accelerator (linac) or storage ring].

We use a 50 fs optical laser to modulate the energy of 100 ps electron pulses in a storage ring.

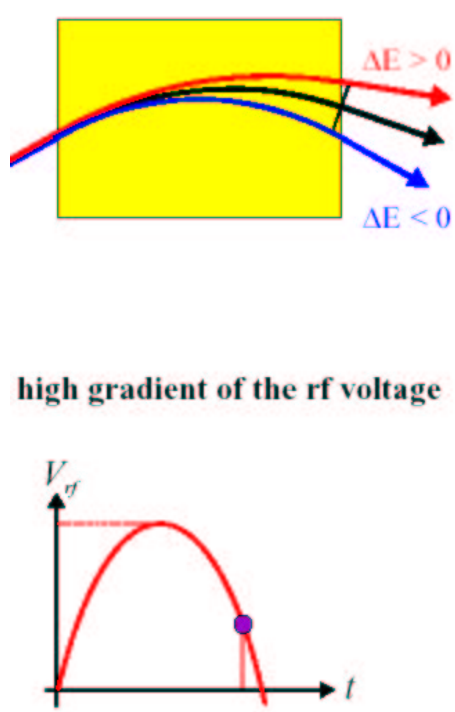
⇒ Use dispersion provided by static magnetic fields to slice, or compress, or bunch the electron pulses.

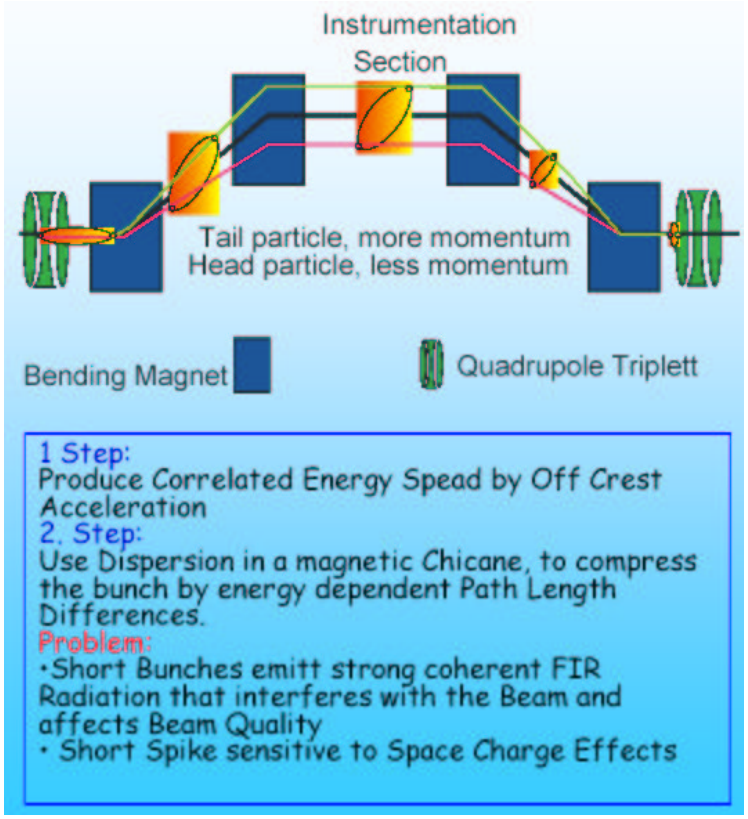
We use angular dispersion to slice the electron beam in a storage ring (2.4 GeV).
[↔ linac: pulse compression; FEL mechanism: bunching]

- Use short period, small gap magnetic undulator to generate hard x-rays (3-18 keV).

- Use and develop technology suitable for a Free Electron Laser (FEL) user facility in the future.

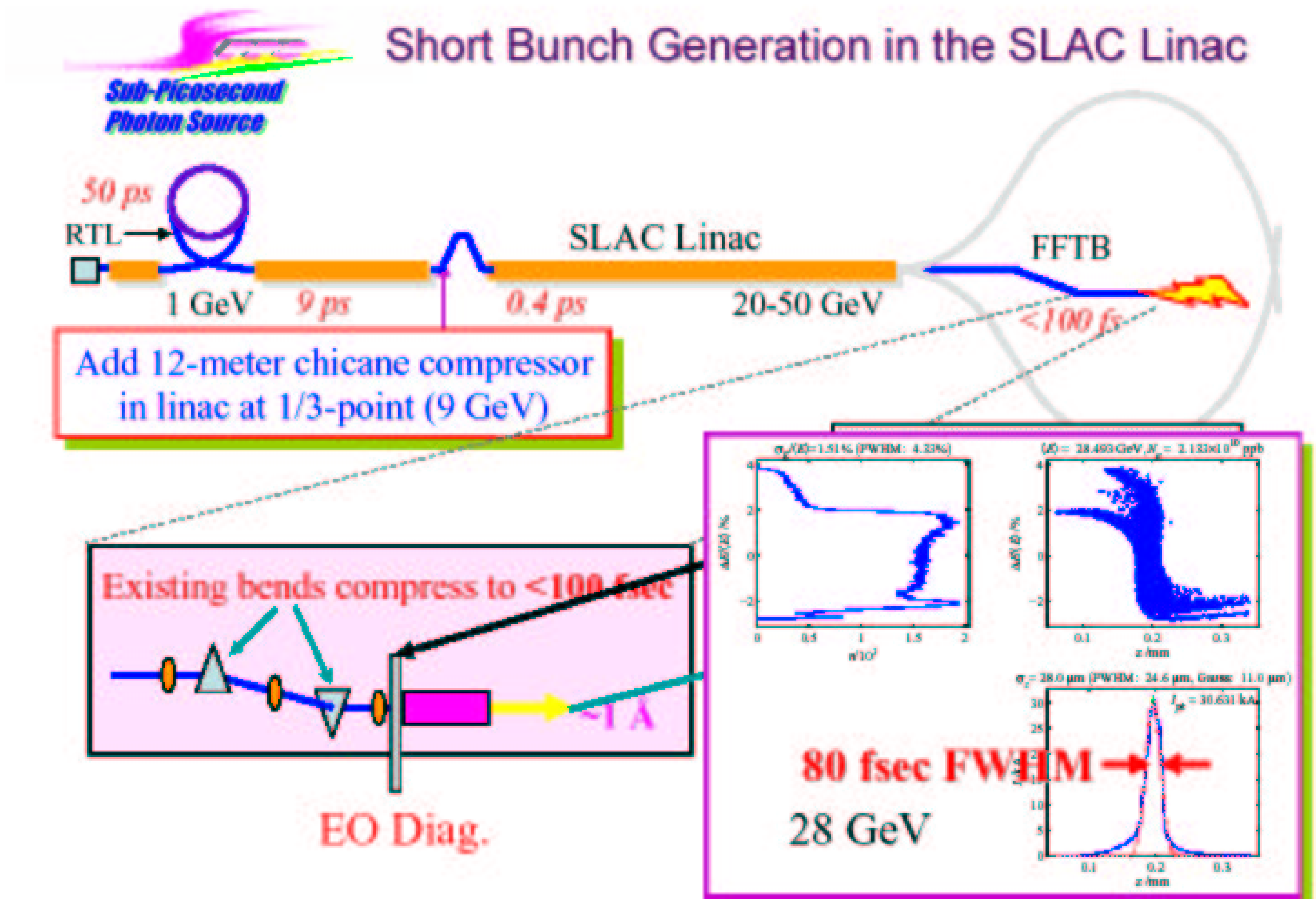
LINAC: Electron Bunch Compression (2 ps $\rightarrow$ 100 fs



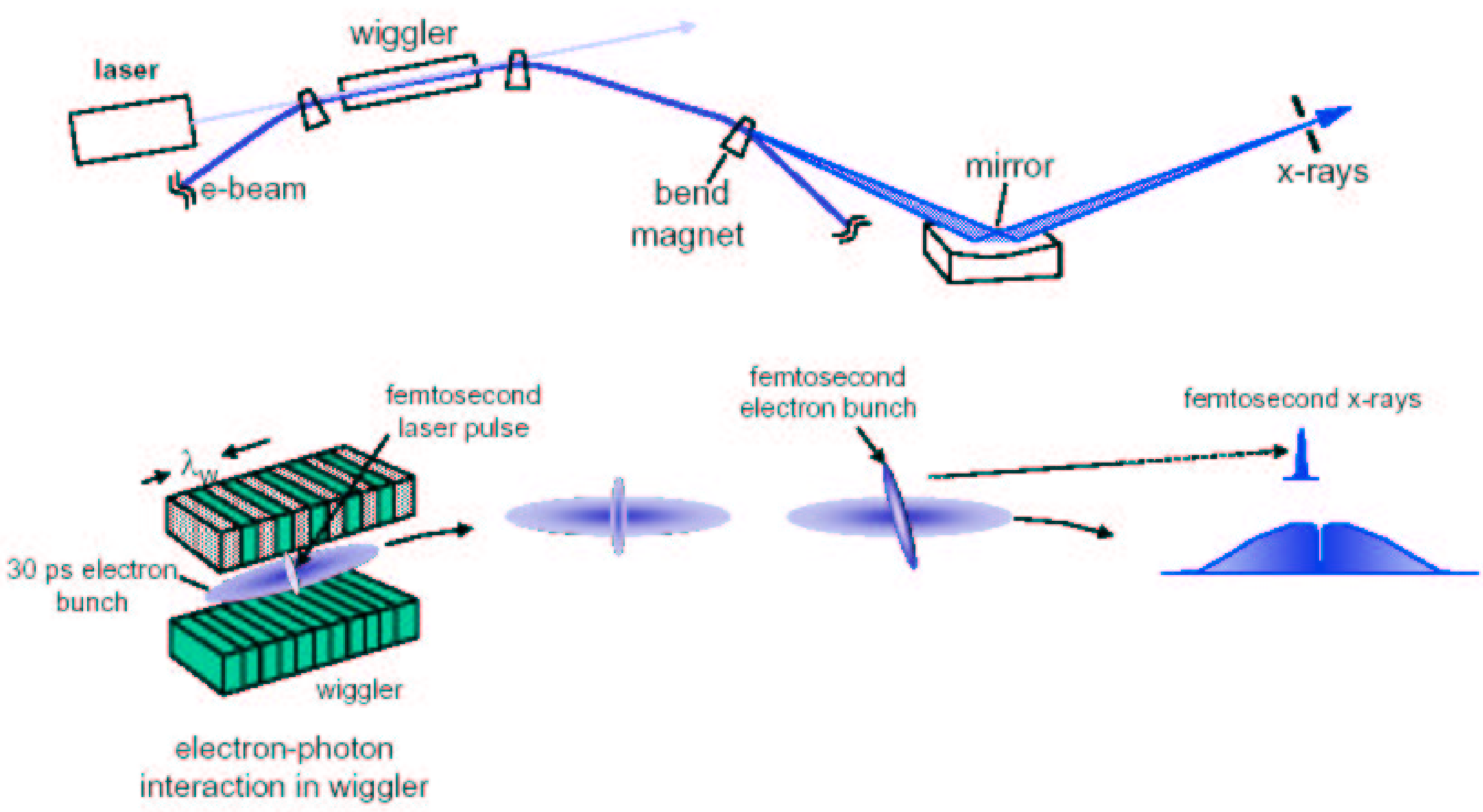


The rf voltage is phased such that particles ahead (behind) the bunch center are accelerated (decelarated) following a longer (shorter) path than the reference particles in the center of the bunch $\Leftrightarrow$ magnetic chicane $\Rightarrow$ pulse compression.

SLAC Linac: $\leq$ 100 fs Electron Bunches Demonstrated Using Compression



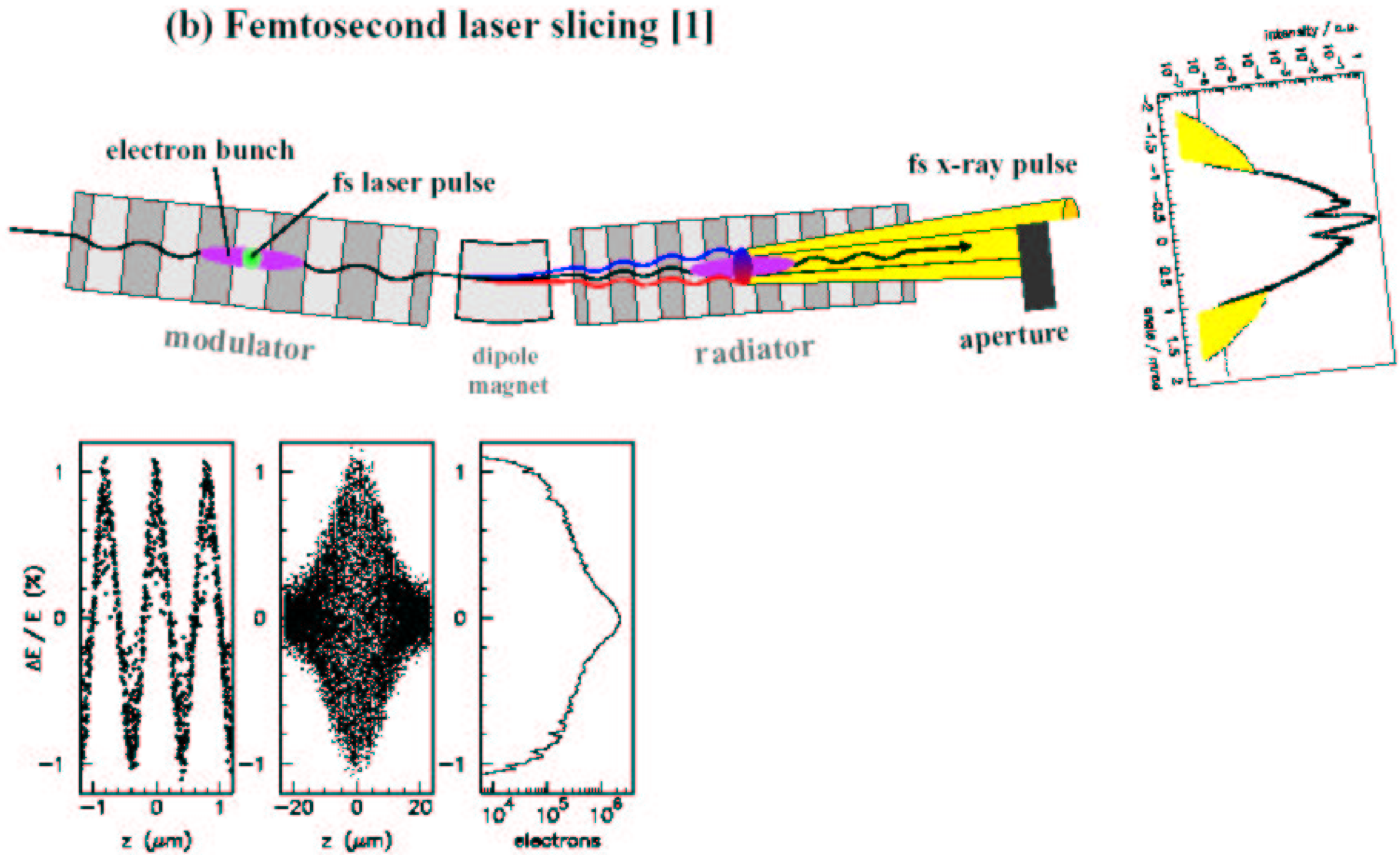
Storage Ring: Electron Pulse Slicing (100 ps → 100 fs



Zholents and Zolotorev, *Phys. Rev. Lett.*, 76, 916,(1996).

Schoenlein et al., *Science*, 287, (2000)

Bunch Slicing: fs-Laser for Energy Modulation



[1] A. A. Zholents, M. S. Zoloterev, PRL 76 (1996), 912.

Radiation by Moving Charges

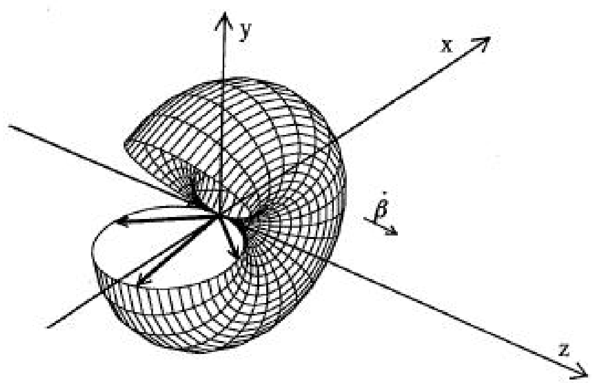
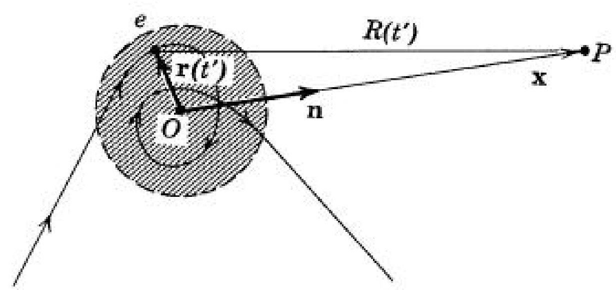


Fig. 7.2. Radiation pattern in the particle frame of reference or for nonrelativistic particles in the laboratory system



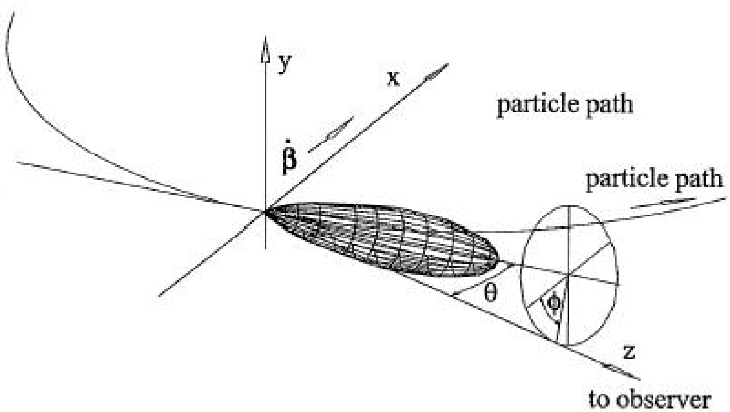
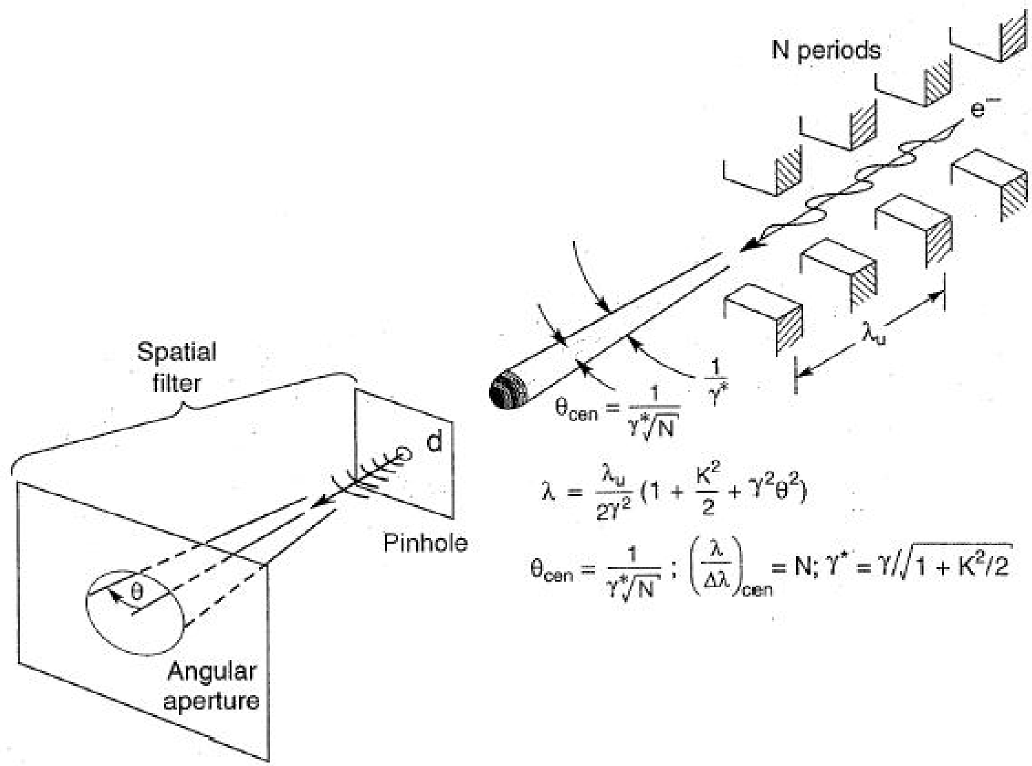


Fig. 7.3. Radiation geometry in the laboratory frame of reference for highly relativistic particles

$$\frac{d^2 I}{d\omega\, d\Omega} = \frac{e^2\omega^2}{4\pi^2 c}\left|\int_{-\infty}^{\infty} \mathbf{n}\times(\mathbf{n}\times\boldsymbol{\beta})\, e^{i\omega(t-\mathbf{n}\cdot\mathbf{r}(t)/c)}\, dt\right|^2$$

Undulator Radiation



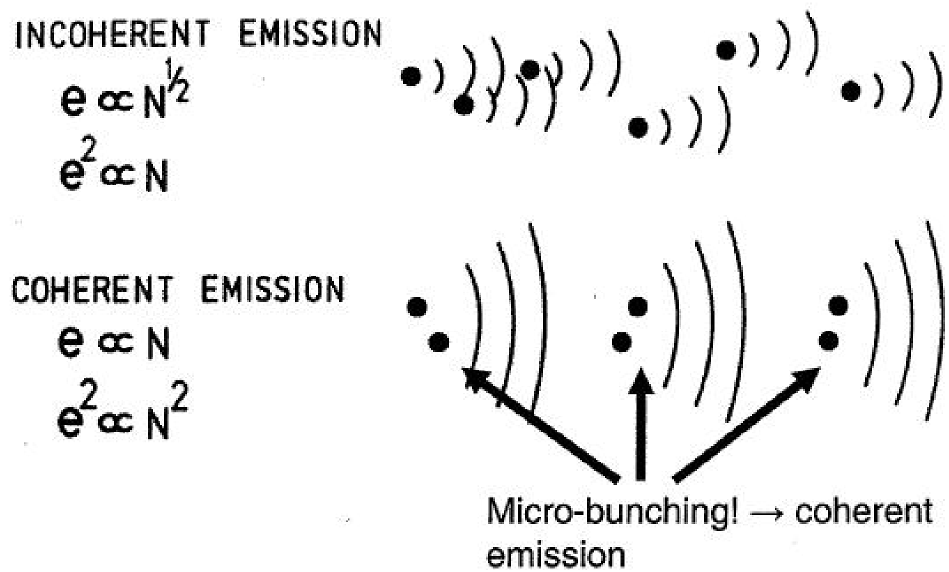
In an undulator , K is moderate ($K \leq 1$) and radiation from different periods interfere coherently $\Rightarrow$ sharp peaks at harmonics of the fundmental (n=1).

The angular distribution of the nth harmonic is concentrated in a narrow cone.

Incoherent vs Coherent Radiation

incoherent emission amplitude *e* from **random walk**

(intensity ~ amplitude²)

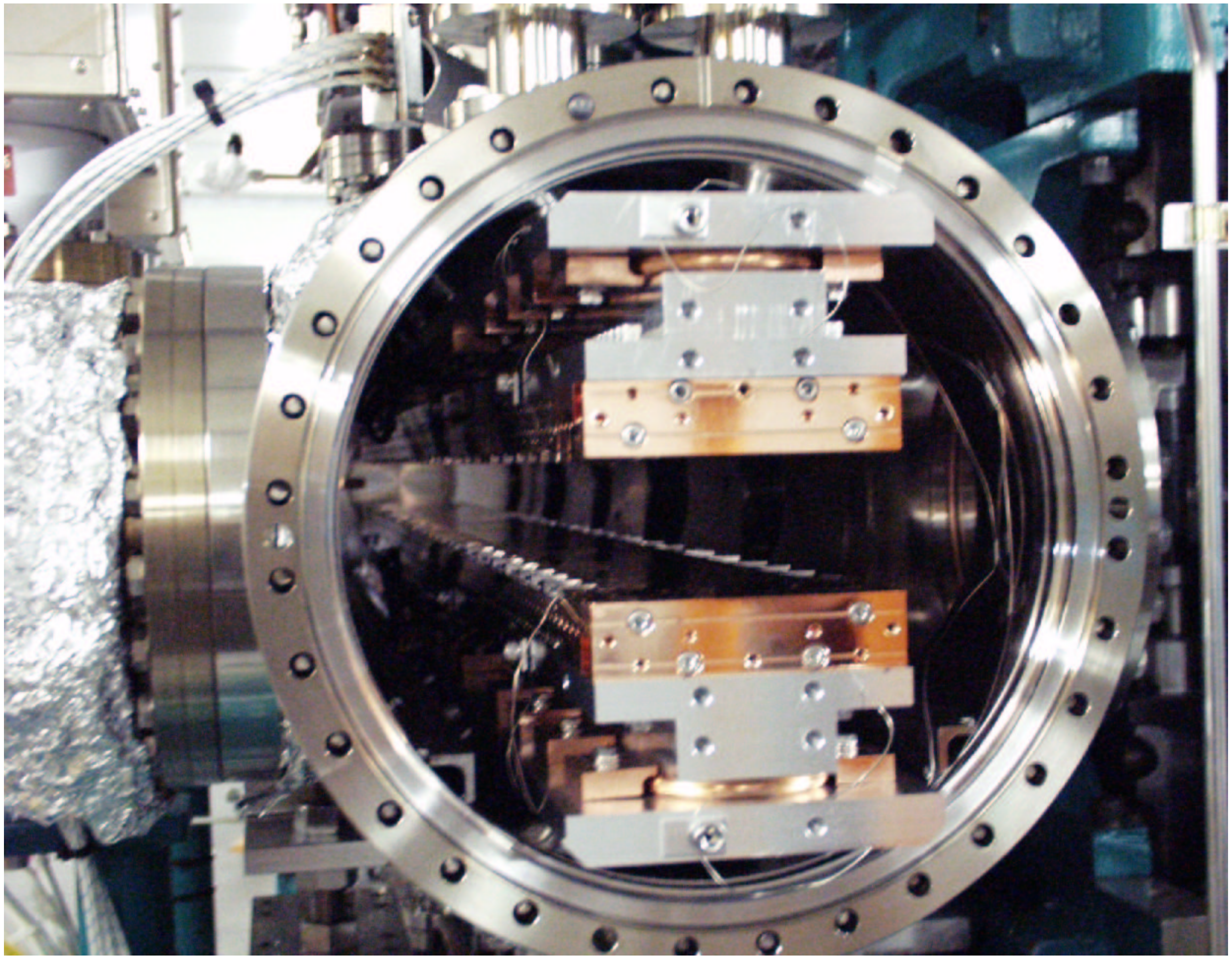


Incoherent Limit: $f(\omega)=0 \Rightarrow I \sim N$

Coherent limit: $f(\omega)=1 \Rightarrow I \sim N^2$ **($N = 10^{10}$ (= 1 nCb))**

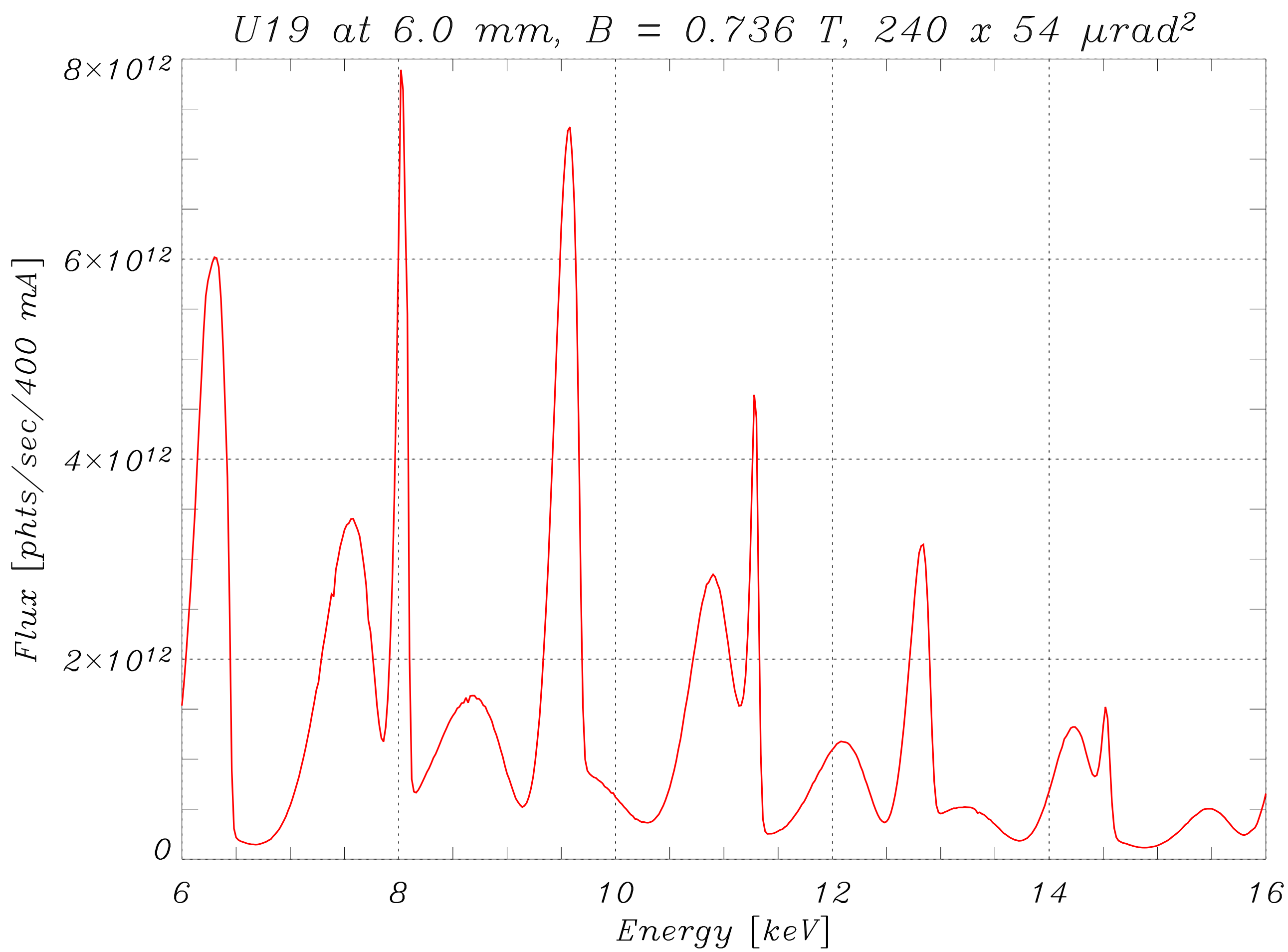
Coherent radiation observed if electron bunch length $\leq$ radiation wavelength

Undulator: Periodic Magnet Arrays Used to Generate High Flux X-Rays



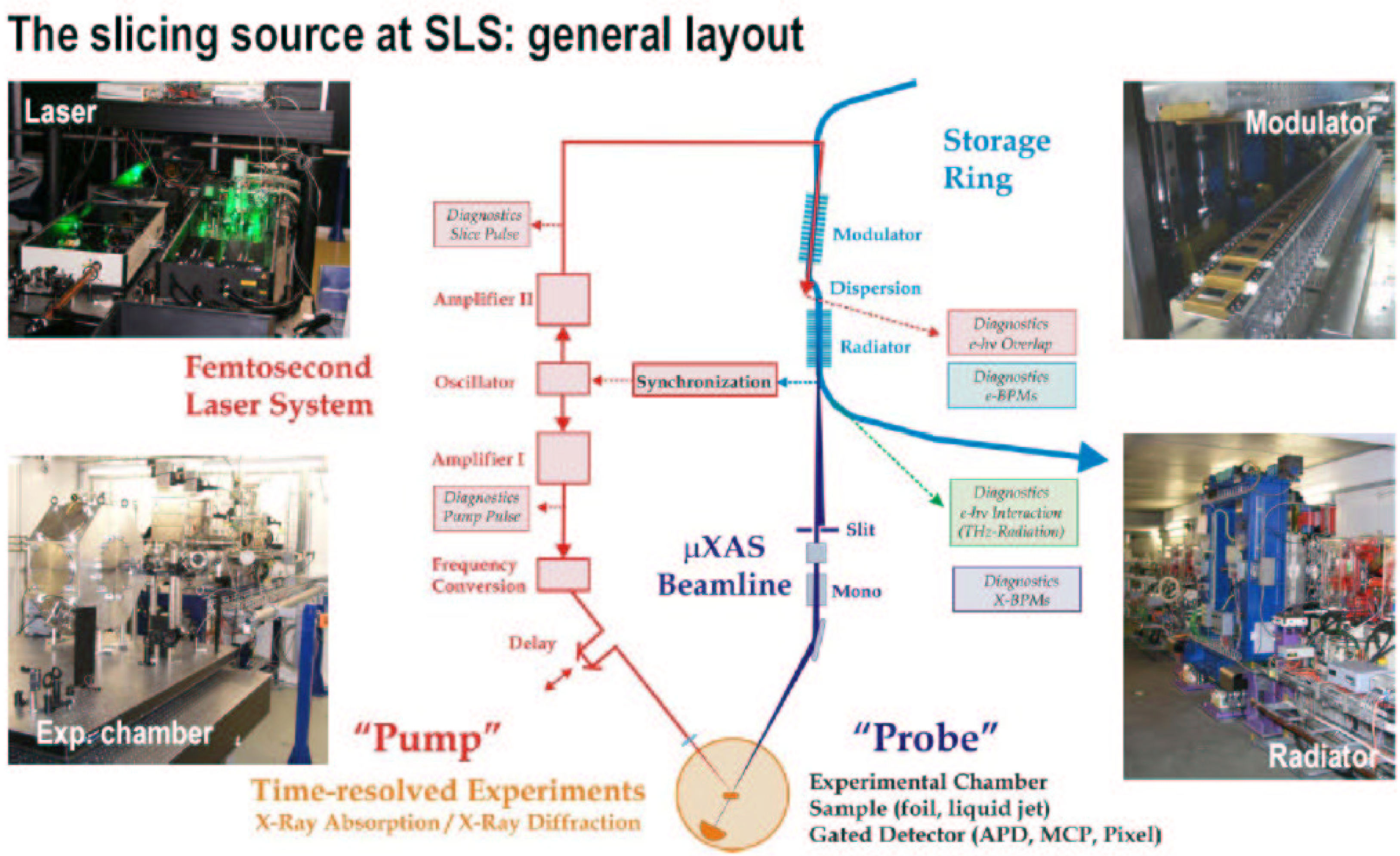
In-vacuum undulator: magnets installed inside ultra-high vacuum to reach 4-5 mm gaps

Measured Undulator Spectrum at SLS Beamline



Undulator line spectrum: constructive interference of periodically emitted radiation

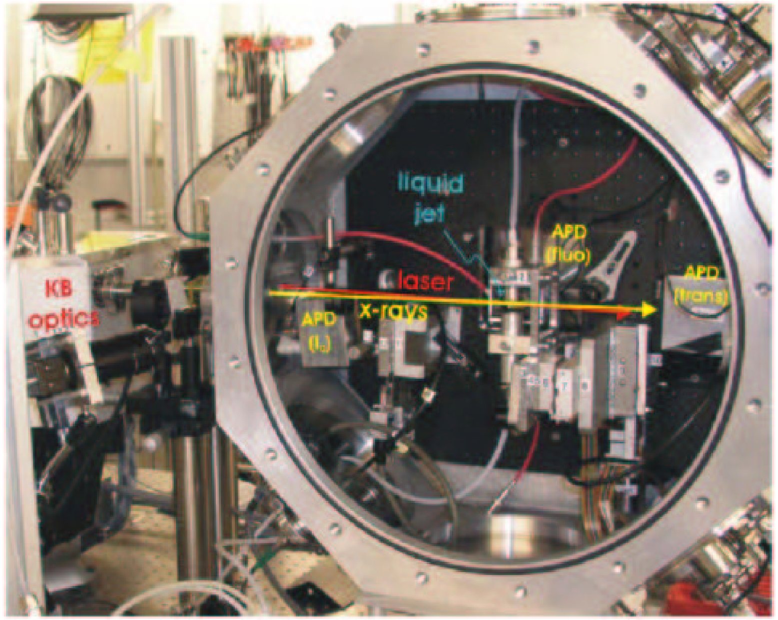
FEMTO: Instrinsically Synchronized Laser/X-Ray Pump/Probe Beams



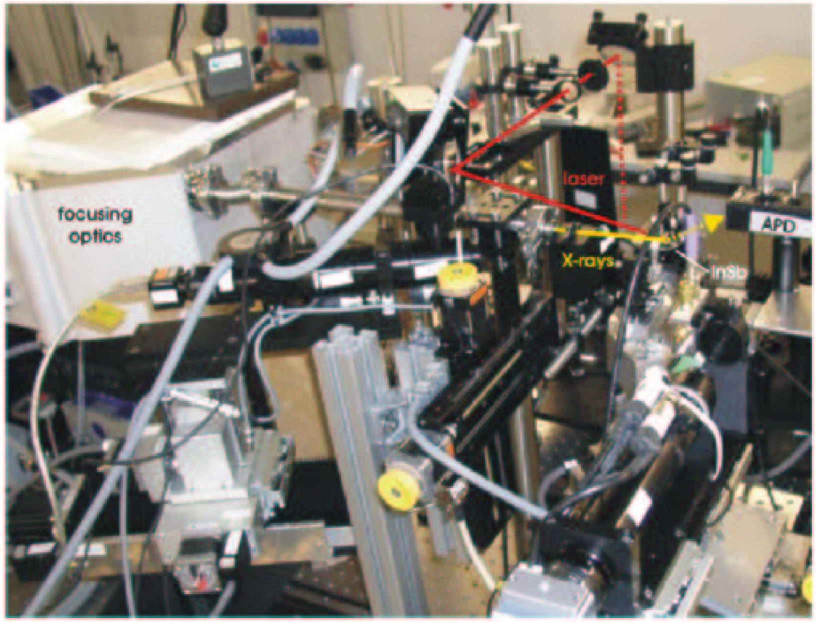
Laser/X-Ray Pump-Probe Experiments (Liquid Jet or Solid Sample in Air)

Existing setups for time-resolved experiments

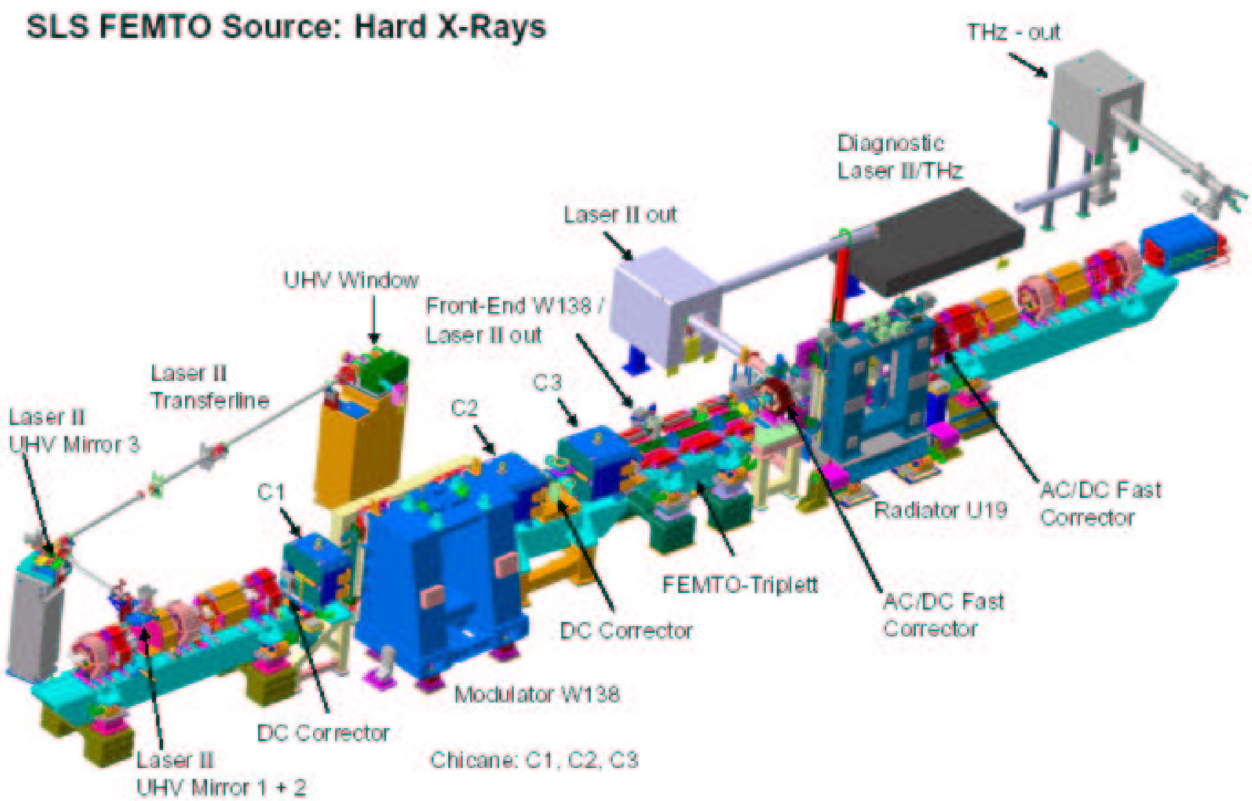
Photochemical transients (absorption, fluorescence)



Transient structure in solids (diffraction, reflection, absorption, fluorescence)



SLS FEMTO: Tunable sub-ps Hard X-Ray (3-10 keV) Source

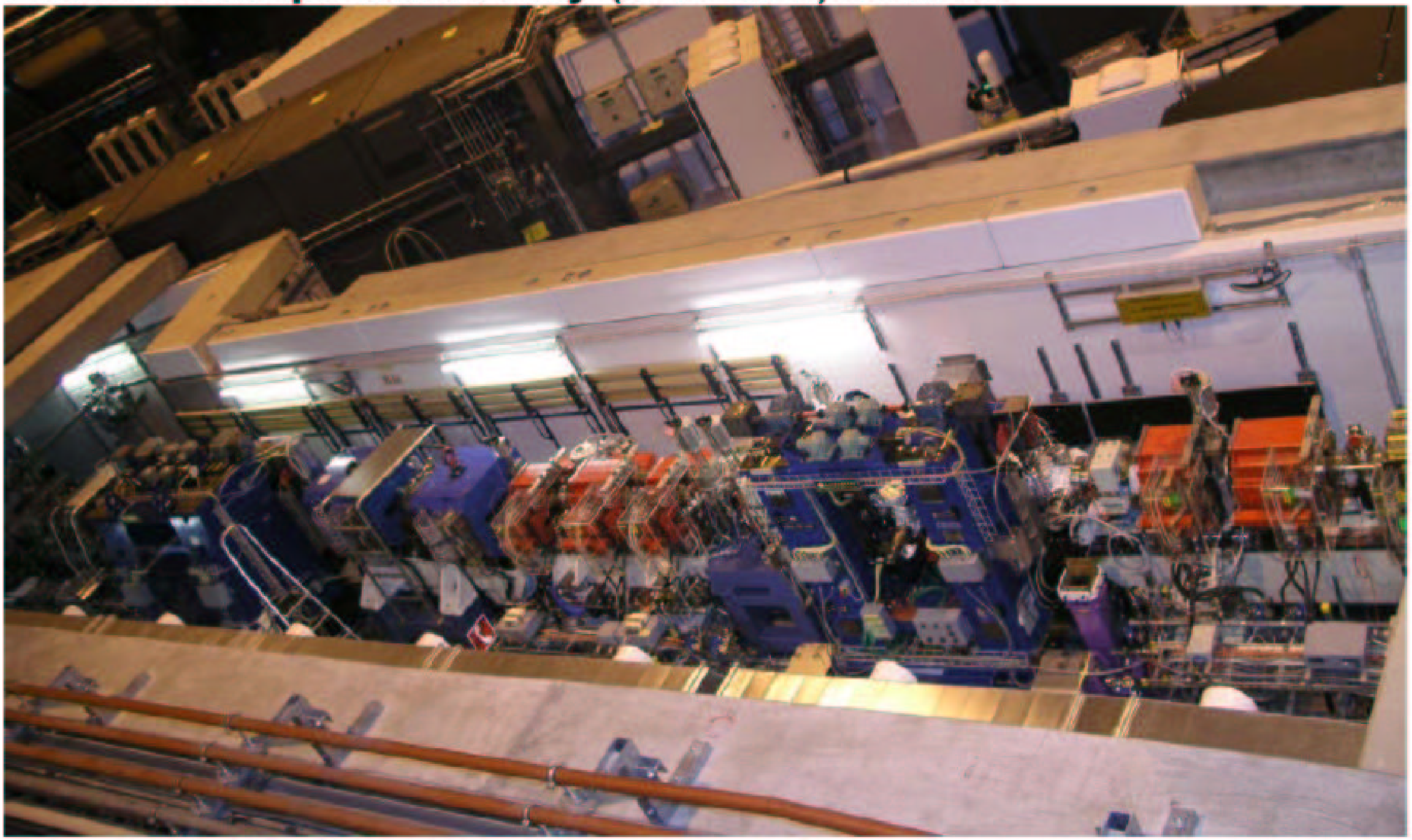


Principle: Electron/Laser Interaction ⇒ Pulse Separation ⇒ sub-ps X-Rays

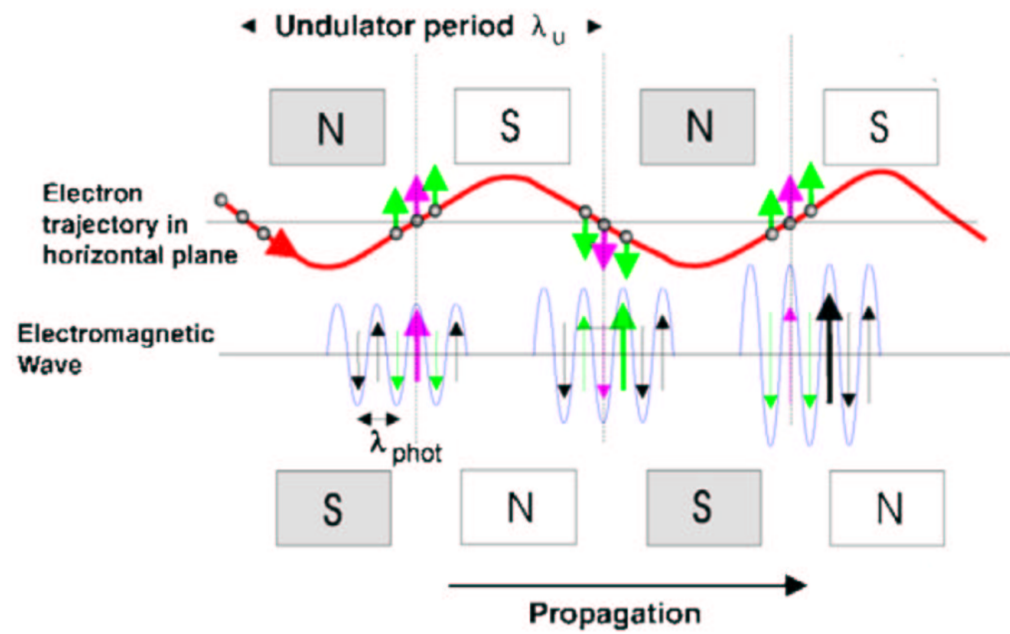
Modulator (Wiggler)	Angular Dispersion (Chicane Magnets)	Radiator (Undulator)

FEMTO Source Installed at the SLS Storage Ring (Length 13 m)

Tunable sub-ps hard X-ray (3-18 keV) source



Modulator: Resonance Condition for Energy Transfer from Laser to Electron

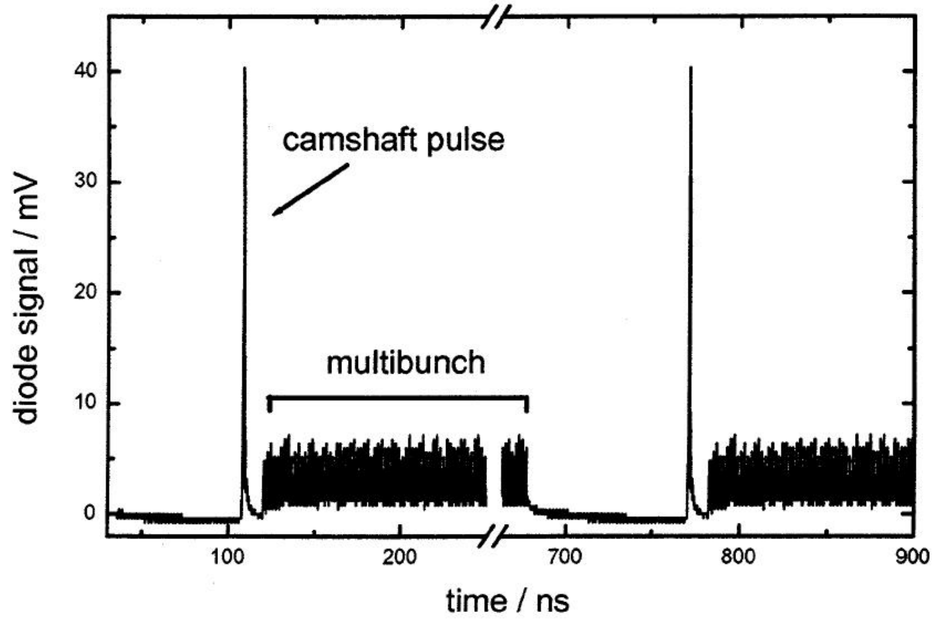


transverse polarized optical field $\vec{E}$ $\Rightarrow$ transverse electron velocity needed
$\Rightarrow$ vertical periodic magnetic field $\vec{B}$ needed: use undulator or wiggler !

energy exchange: $$m\,c^2\,\frac{d\gamma}{dt} \simeq -e \cdot \frac{B_0 \cdot E_0}{\gamma}\,\sin(\Phi_0)$$

Φ_0 is a random phase: determines the initial position of the electron relative to the optical wave $\Rightarrow$ energy is only modulated !

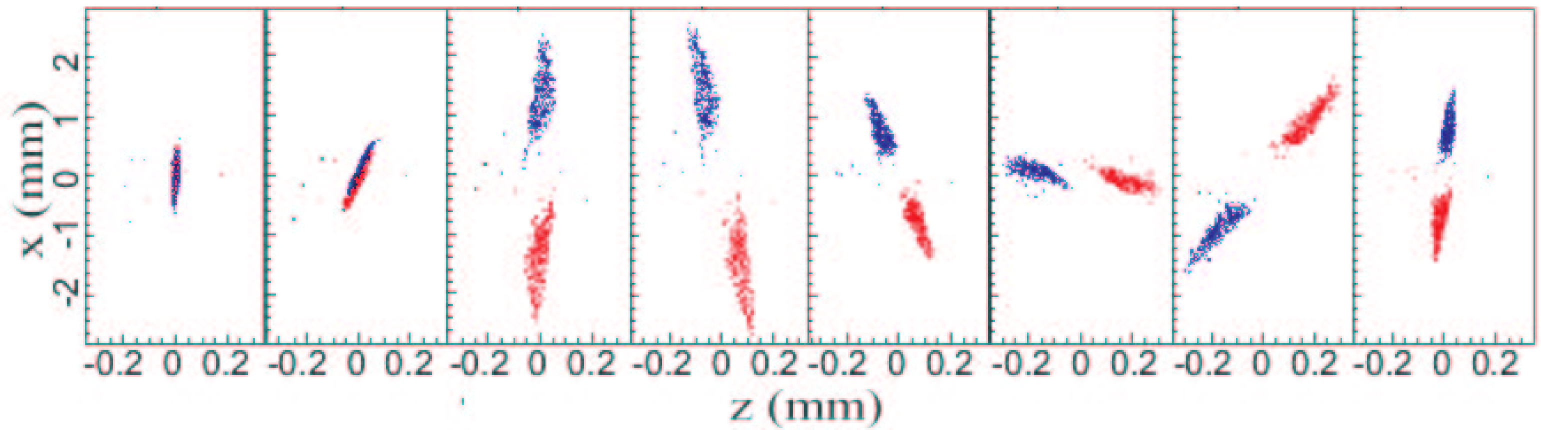
FEMTO: Timing and Synchronization



Storage ring rf frequency: 500 MHz (↔ 2 ns pulse spacing in multibunch train);
Single camshaft pulse sitting in a 100-200 ns gap is hit by the laser;
Bunch revolution frequency: 1 MHz;

Laser oscillator: 100 MHz (5th subharmonic of 500 MHz rf frequency);
Oscillator cavity length modulated in feedback loop using 500 MHz rf reference;
Laser amplifier rep. rate (slicing): 1 kHz;
Relative timing controlled by electronic phase shifter of 500 MHz rf reference;

Diagnostic for Laser/e-Beam Interaction: Coherent THz-Radiation



Sliced electrons ($\simeq$ 100 fs) and dip in the bunch emit coherent radiation ($\propto N_e^2$); Radiation in the THz-range corresponds to the length of the sliced electron bunch;

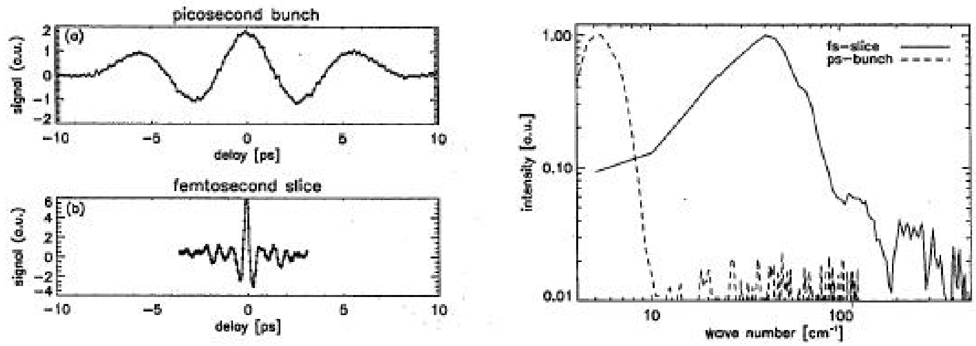
[100 μm $\simeq$ 300 fs $\simeq$ 100 cm^{-1} $\simeq$ 3 THz]

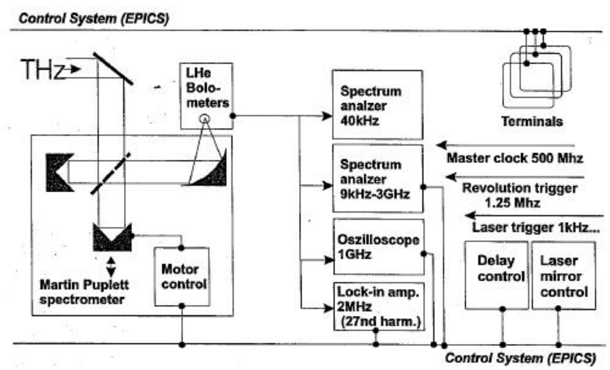
Direct sliced bunch length measurement: interferogram measured with Michelson interferometer (Martin-Puplett spectrometer) $\rightarrow$ bunch form factor obtained by fourier transform; [detector: 1 MHz, InSb-bolometer, 4.2 K]

Sub-ps Electron Bunch Length Measurement: Coherent SR ⊗ Interferometer

FIR-Interferograms of a 1.2 ps rms bunch in low alpha vs. femtoslice

Corresponding FIR spectra





pulse energy: > 0.25 nJ
peak power estimate: > 400 W
max. E- field: >30 kV/cm
conversion efficiency at 1mJ laser power: 0.25×10^{-6}

Compares well to Laser based sources (optical rectification)

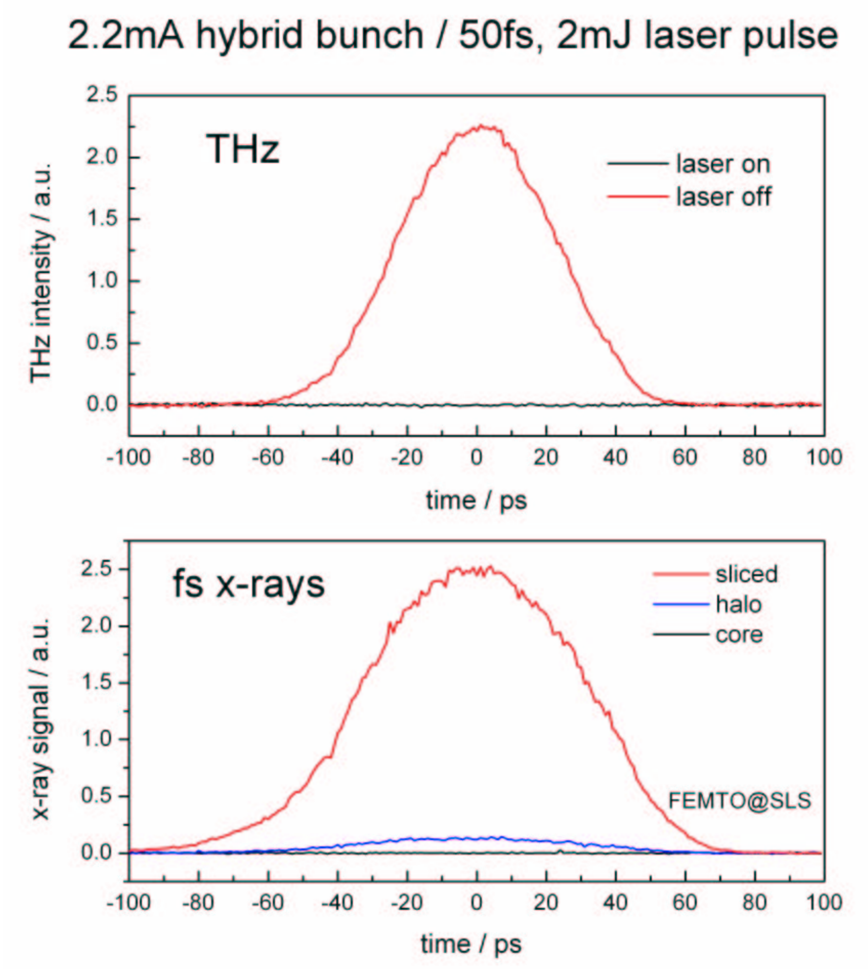
Coherent synchrotron radiation (CSR) emitted by a ps- and sub-ps electron bunch.

Bunch length (form factor) obtained by fourier transform of the interferogram measured with a Martin-Puplett spectrometer (Michelson Interferometer).

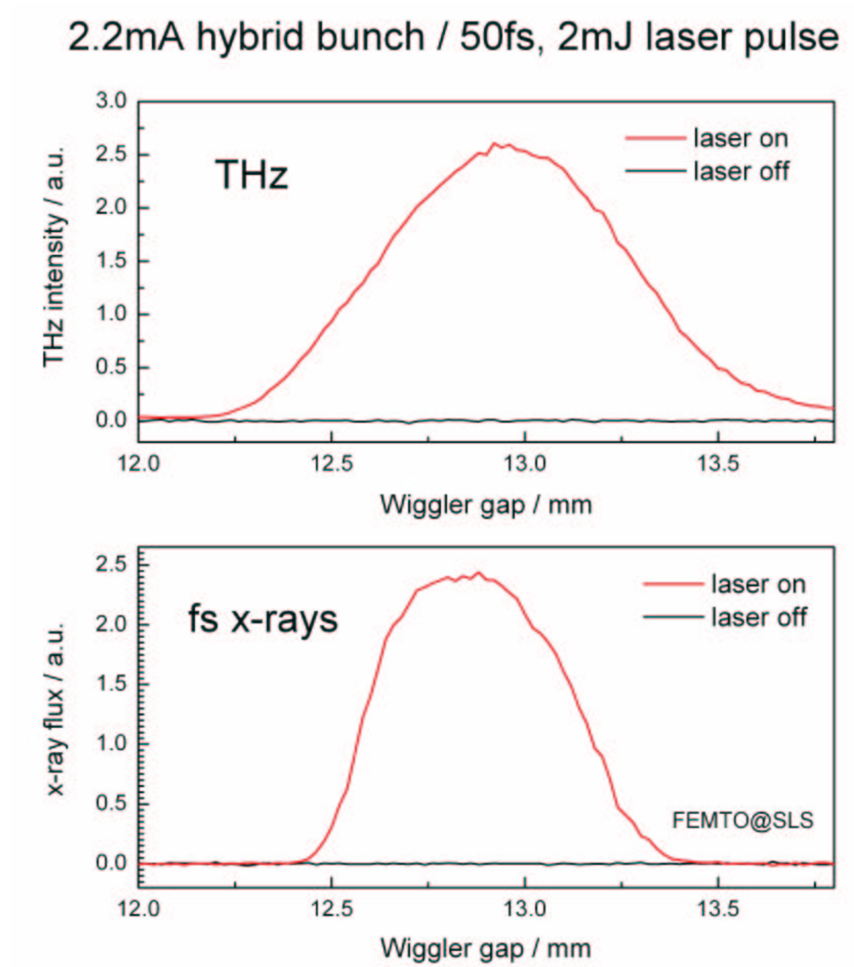
[K. Holldack et al., Phys. Rev. ST AB 8 (2005) 040704.]

Laser-Electron Interaction: Tuning for Optimum Energy Exchange

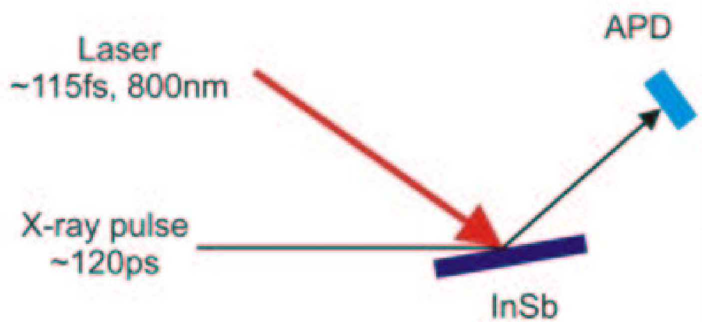
Time Overlap

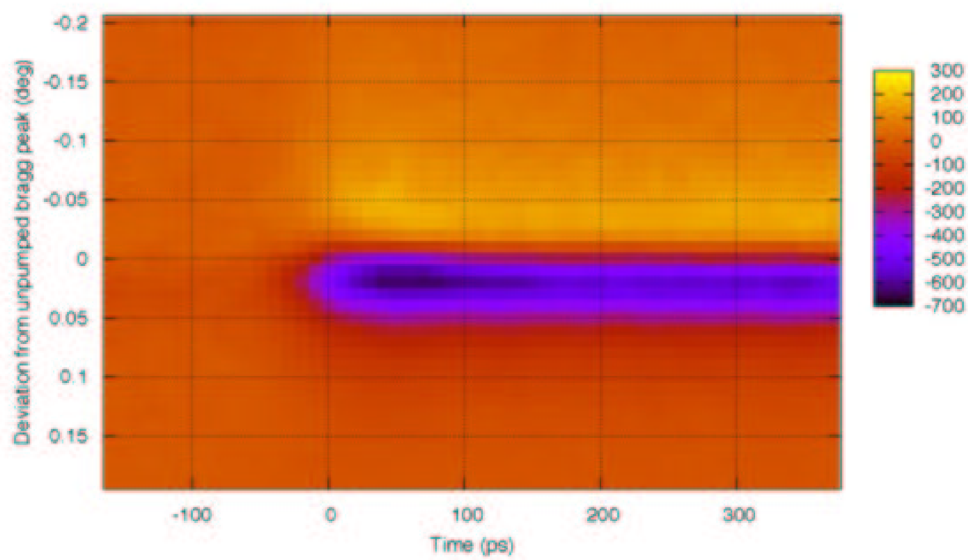


Resonant Energy



Ultrafast X-Ray: Timing Diagnostics

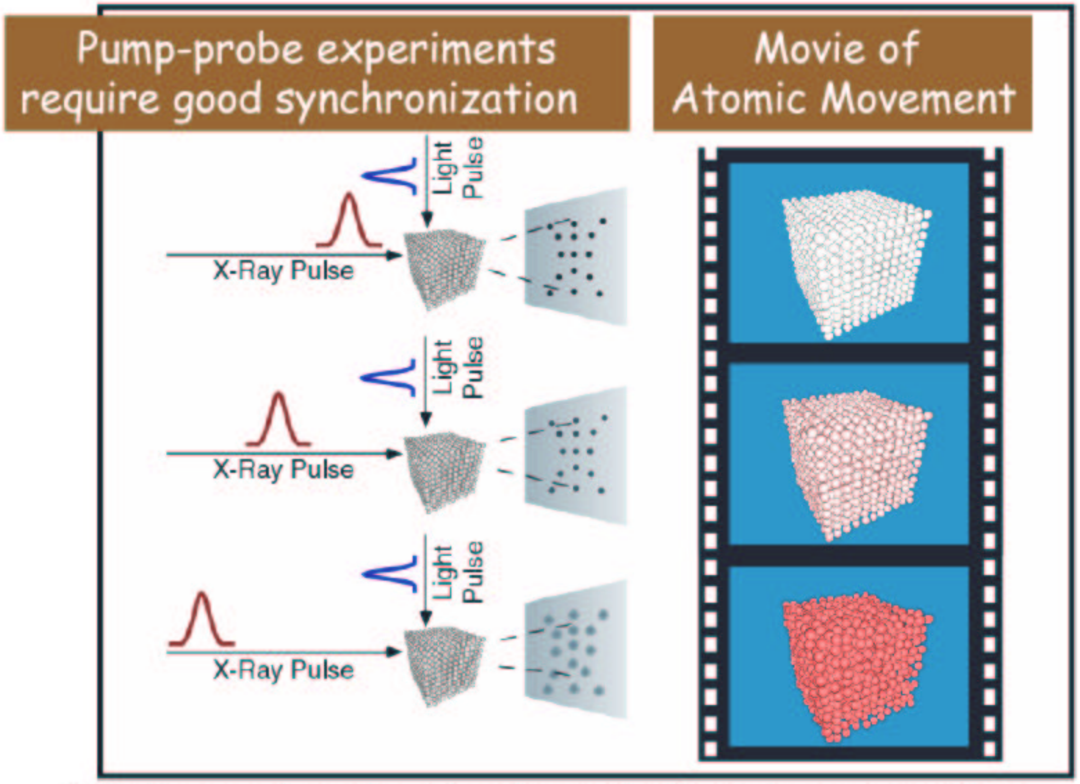




Ultrafast X-ray diffraction: ps strain wave dynamics in a surface-heated InSb crystal. The laser strikes the surface a 0 ps, resulting in the formation of an expansion layer over the x-ray probe depth.

Example: Lattice Oscillations (Phonons)

Viewing femtosecond dynamics:
Pump-Probe Spectroscopy



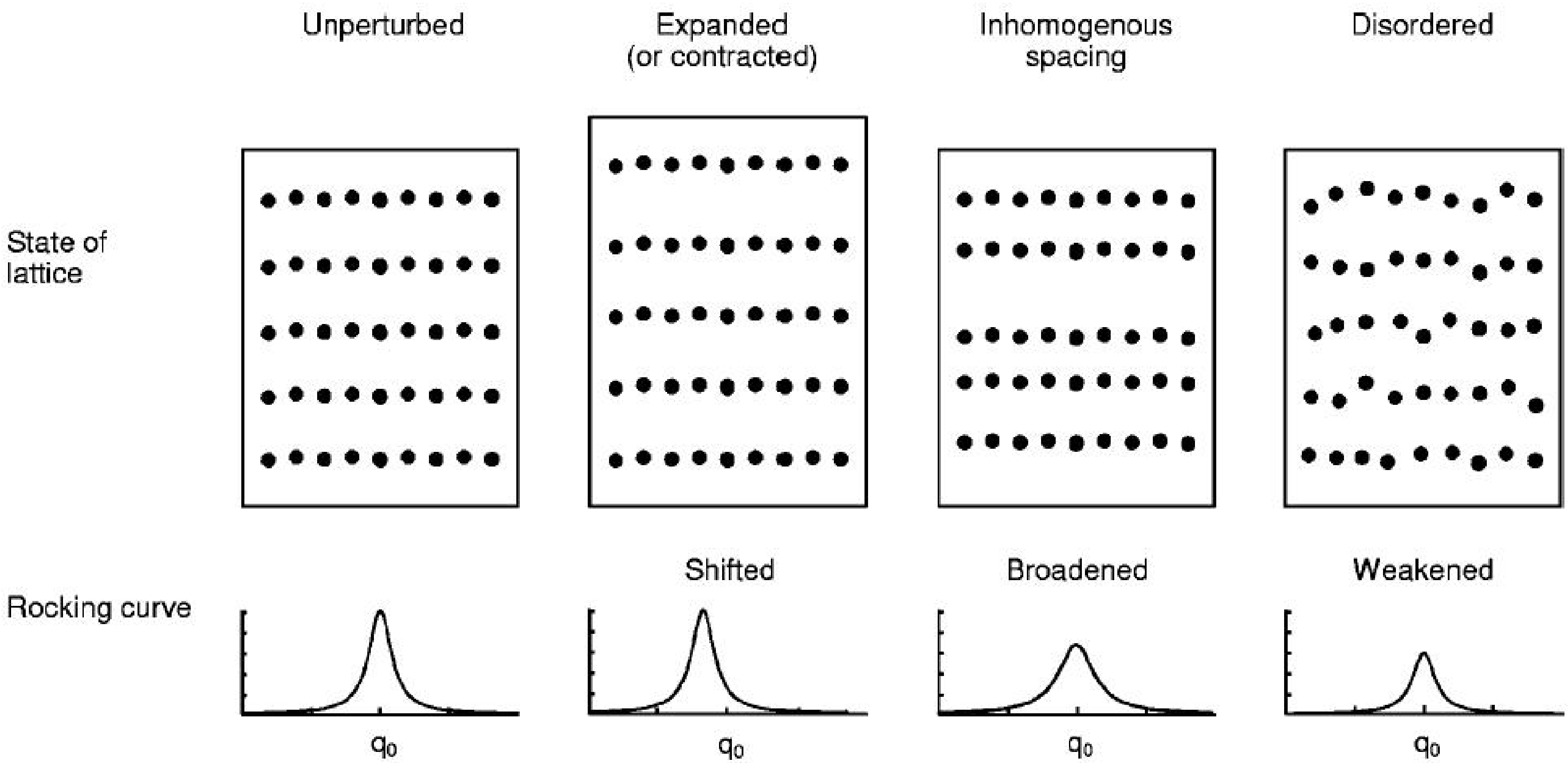
Stuff moves in 0.01-10 ps

100 fs synchronization is equivalent to controlling the arrival of the electron bunch to 1 part in 100 million of its total travel.

Example: Crystal Lattice Vibrations

- **X-ray diffraction from the plane of atoms in the crystal can be used to take snapshots of the atomic motion at precise intervals after excitation has taken place.**
- **For stroboscopic measurements (many shots), the timing jitter between the pump (laser) and probe (x-ray) must be less than the pulse duration.**
- **X-ray probe pulse has to be short enough, just as a flashbulb freezes motion.**
- **The structural question is very simple: What is the spacing between atomic layers ?**
- **The timing is very difficult: To capture the motion, the x-ray pulses must be much less than 1 ps in duration.**

Transient Crystal Lattice Distortions: Effect on Rocking Curve



Rocking curve: **intensity of the diffracted beam vs. θ when the crystal is rotated ("rocked") through the Bragg angle θ_B.**

Why Does Light Excite Lattice Vibrations ?

Two possible mechanisms exist:

- **Impulsive (Raman) scattering:**

  A very short pulse of light literally kicks the lattice, sending it into motion.

  Momentum transfer depends on the strength of the pulse, but is typically small. (dominant excitation mechanism for optical phonons in transparent media)

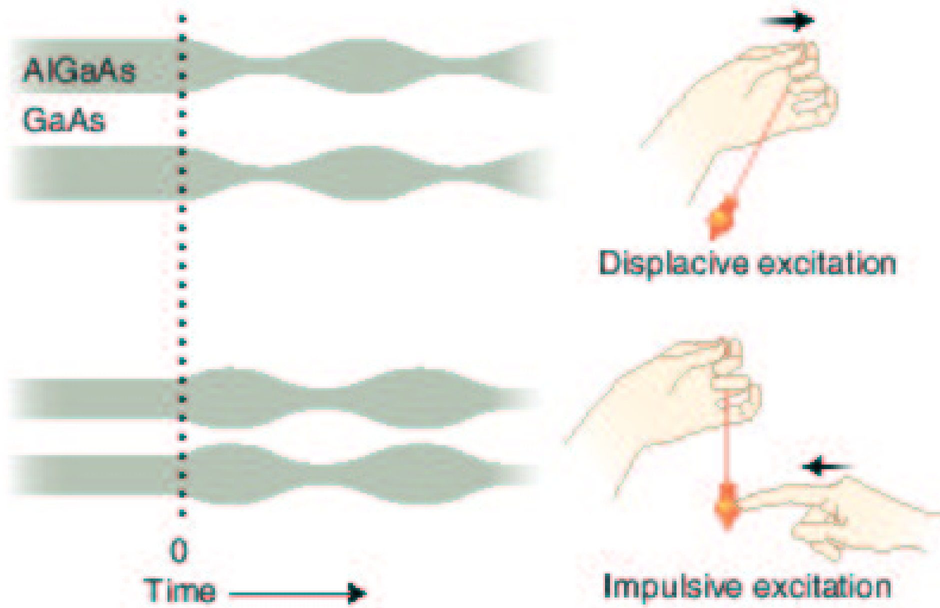
- **Displacive excitation:**

  Requires optical absorption (800 nm) in opaque media. The absorbed light excites electron-hole pairs. This excitation can change the equilibrium distances between atoms. Instantaneous strain is created that relaxes via expansion or contraction of the material.

- **Example: (I) Bismuth, (II) GaAs/AlGaAs (quantum well)**

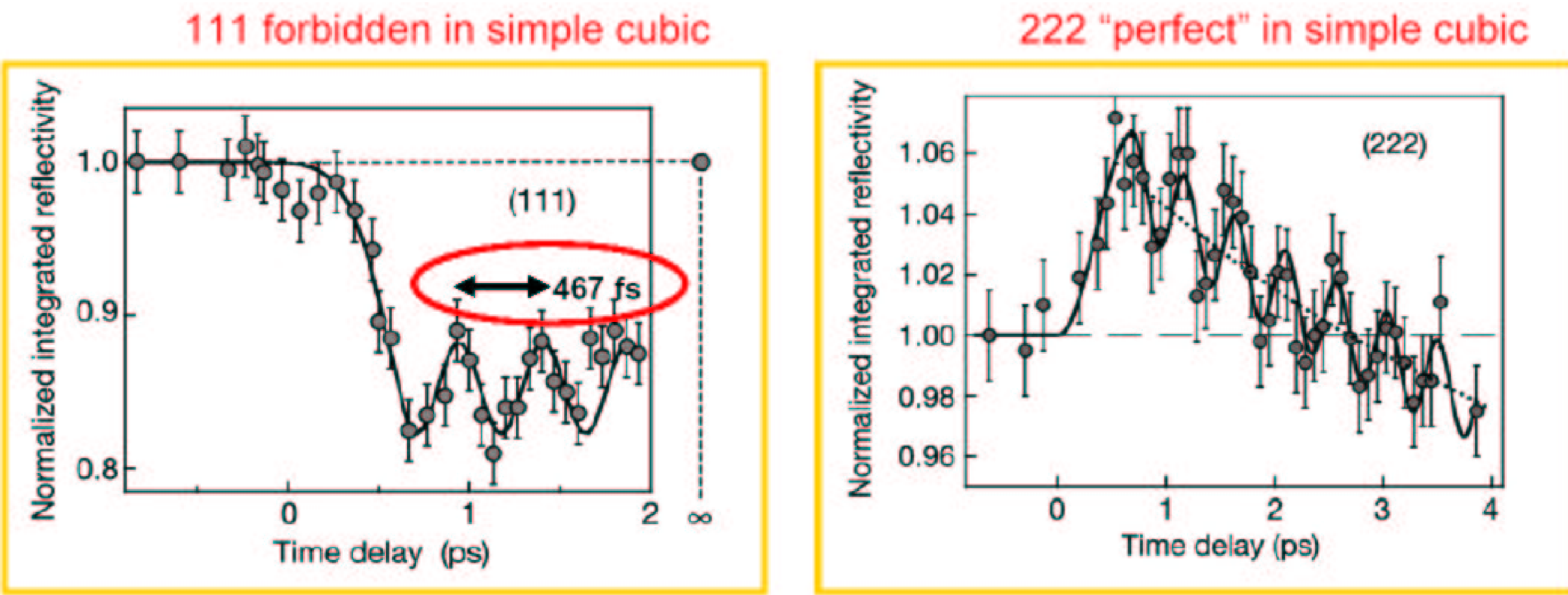
$\leftrightarrow$ **FEMTO commissioning experiment: measure lattice vibrations (phonons) to verify sub-ps x-ray pulse length.**

How Do Crystals Oscillate ?



How crystals oscillate. (**Left**) Displacive and impulsive excitations can be distinguished by the phase of the oscillations. The distortions are greatly exaggerated in these drawings to show that displacive excitations oscillate as cos(ωt), whereas impulsive excitations oscillate as sin(ωt). (**Right**) A pendulum can be used to demonstrate the two excitation mechanisms.

Example (I): Lattice Vibrations in Bismuth

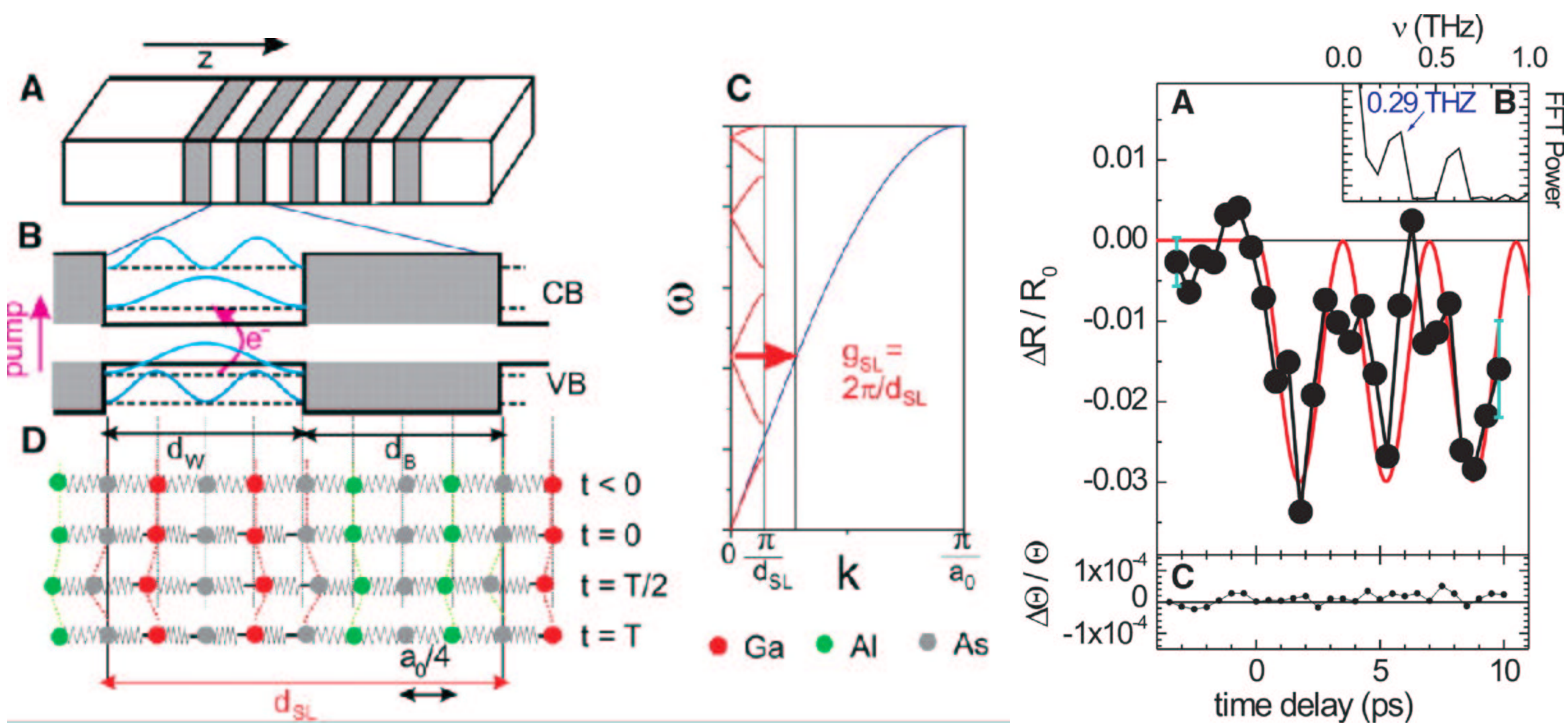


•Measure large displacements: 15–20 pm

Displacive excitation is observed: instantaneous contraction of the lattice due to electron-hole pairs excited by fs optical laser.

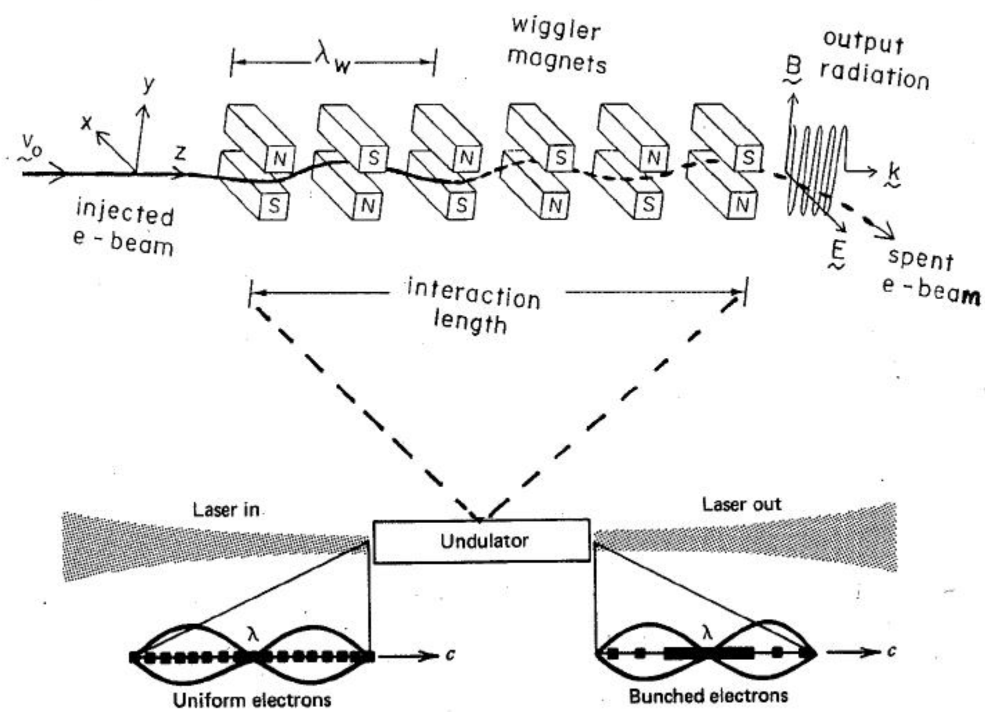
[Sokolowski-Tinten et al., Nature 422 (2003)]

Example (II): Superlattice Phonons in GaAs/AlGaAs



[M. Bargheer et al., Science 306 (2004)]

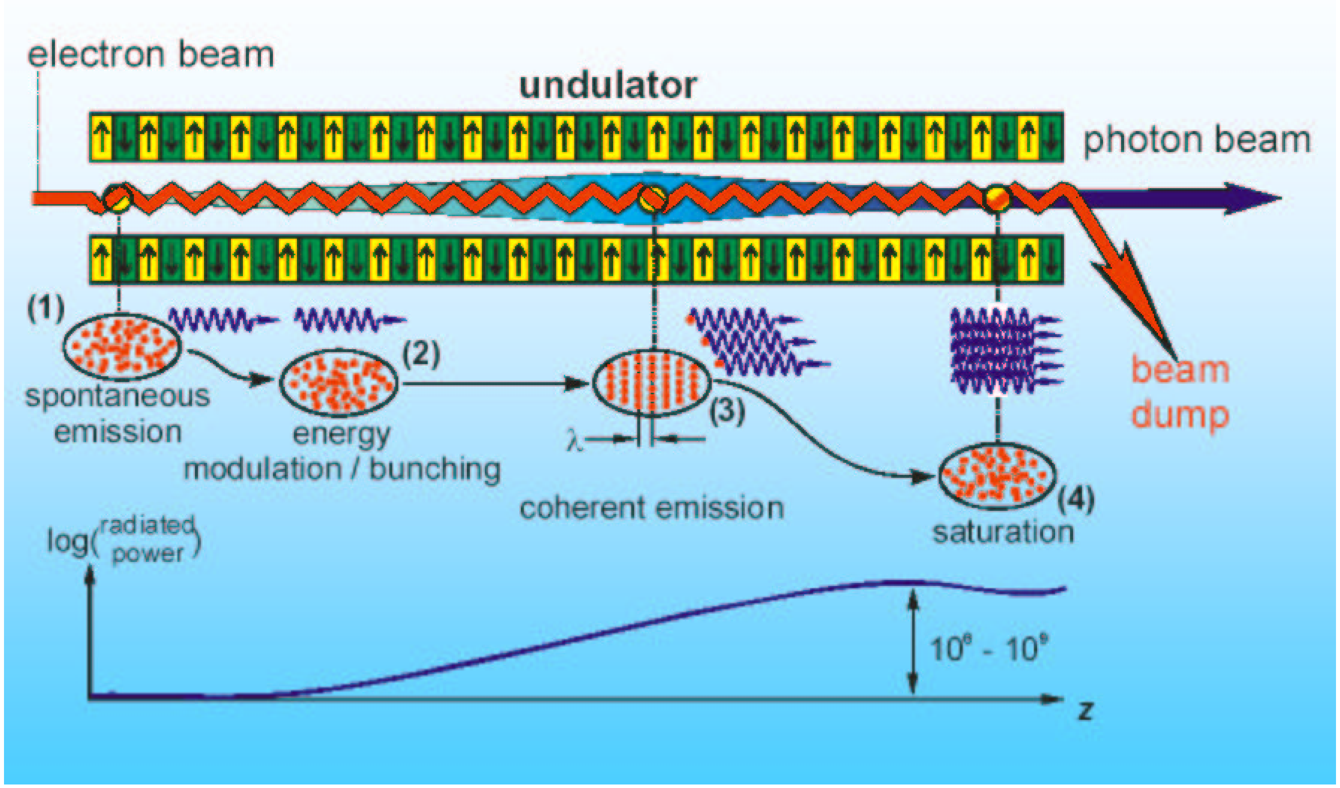
Electron Beam Bunching: Beating of Radiation and Undulator Field



Due to the $\vec{v}$ x $\vec{B}_R$-term in the force equation the electron interacts both with the undulator and radiation field leading to an periodic axial force.

Electron beam bunching occurs on the length scale of the radiation wavelength. The bunching depends on E_0 (radiation field), B_0 (undulator field) and the phase Φ_0.

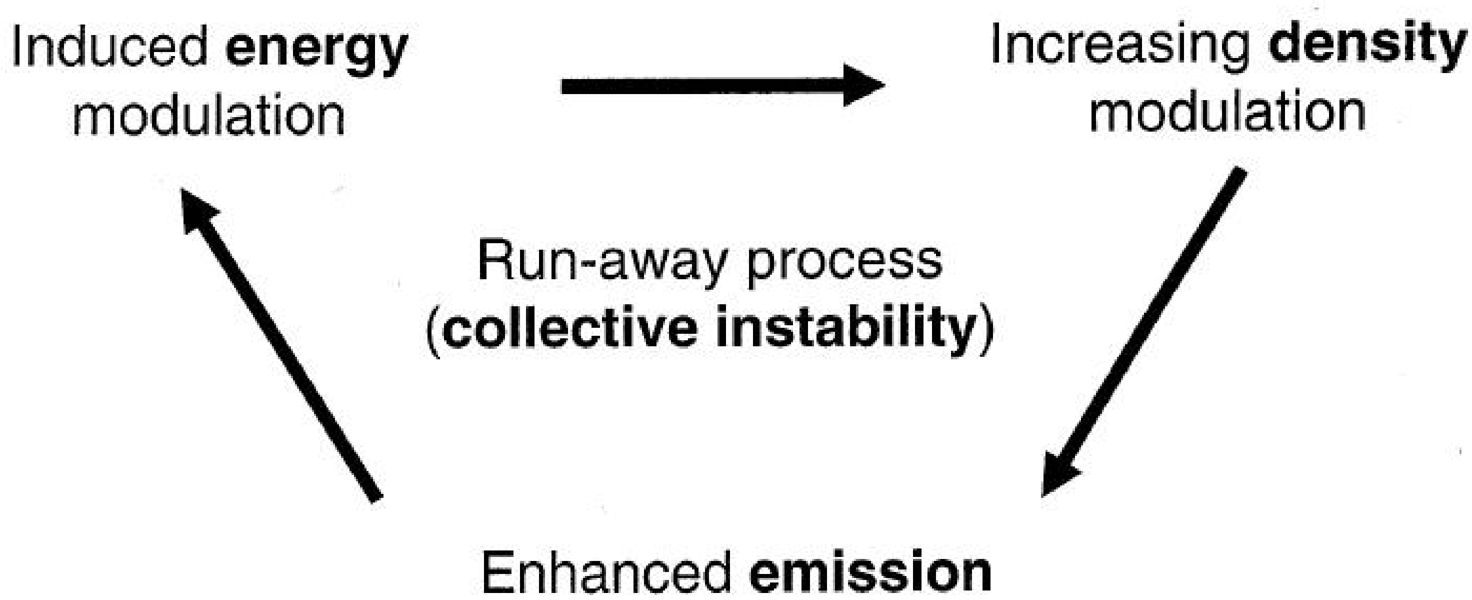
Principle: SASE Free Electron Laser



SASE (Self-Amplified Spontaneous Emission): phase Φ_0 is not 'locked'.
[radiation field: spontaneous radiation (i.e. FEL starting from noise)].

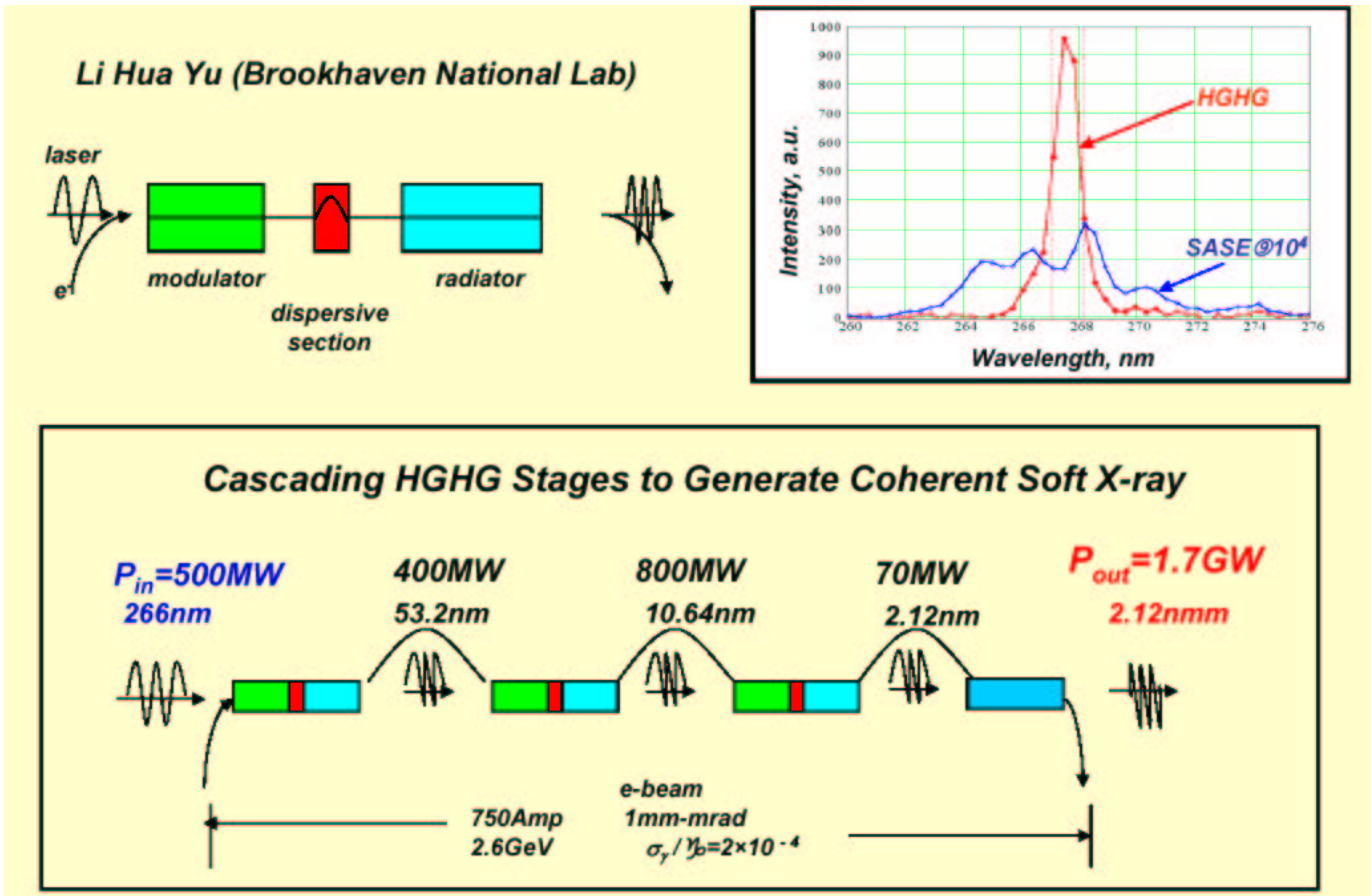
⇔ Seeded FEL amplifier: phase Φ_0 is 'locked'.
[radiation field: coherent (laser) field].

FEL Principle



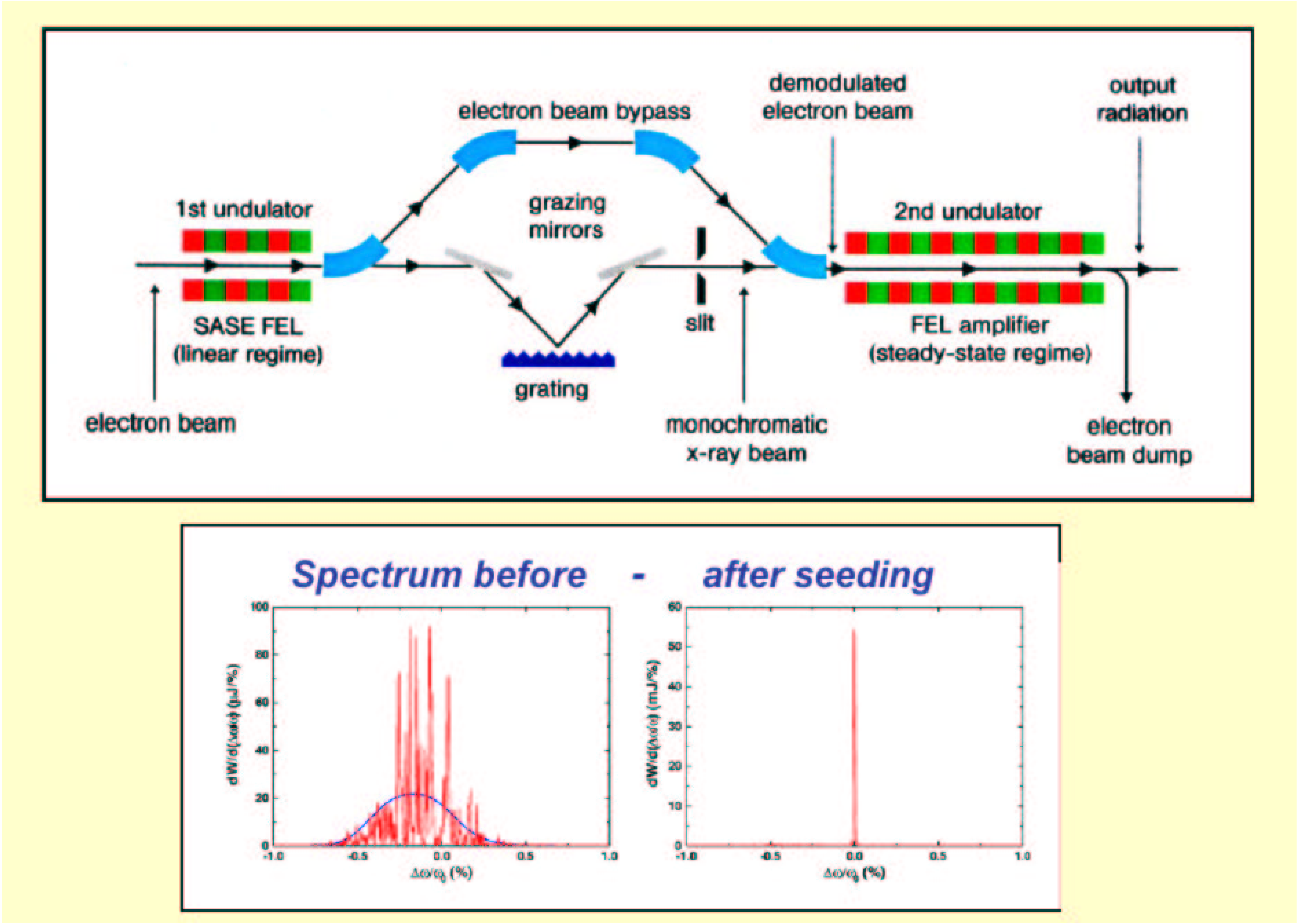
The FEL process **saturates** when maximum density modulation (bunching) is achieved.

HGHG-FEL: Laser Seeding ⊕ High Gain Harmonic Generation

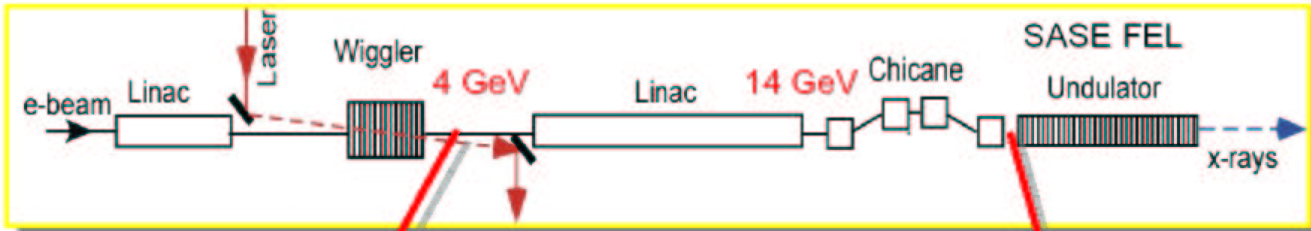


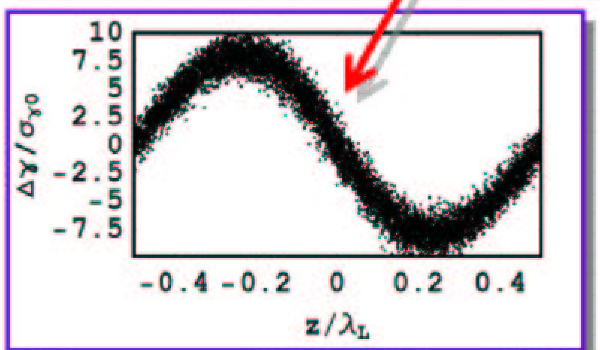
[proof-of-principle experiment: L.H. Yu et al., Science 289 (2000).]

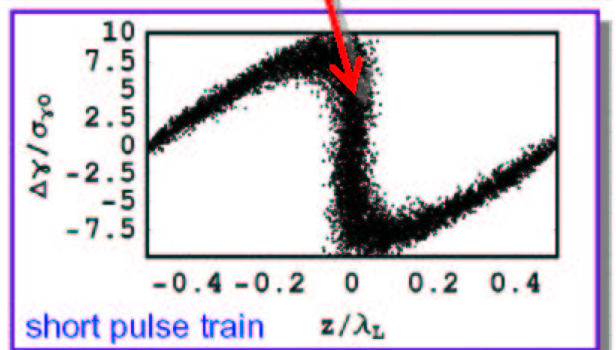
Self-Seeding: proposed for European XFEL (DESY)



Sub-fs FEL X-Ray Pulses: Energy Modulation ⊕ Pulse Compression



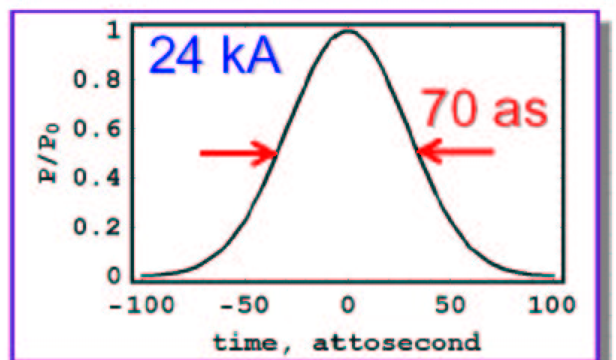




800-nm modulation (few GW)

Allows synchronization between laser pulse and x-ray pulse

E-SASE (applied to *LCLS*)



A. Zholents and G. Penn, Phys. Rev. ST AB 8(2005)050704