

Science with next-generation soft X-ray sources

EXPERIMENTAL METHODS

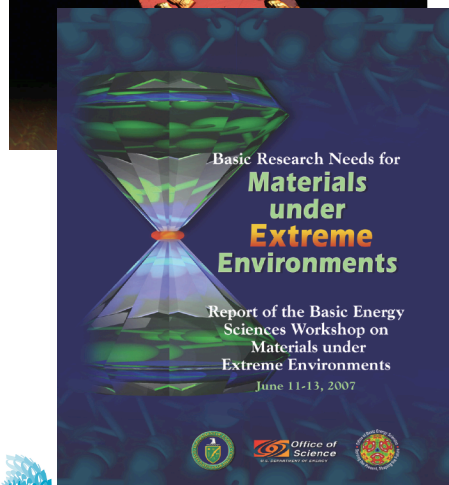
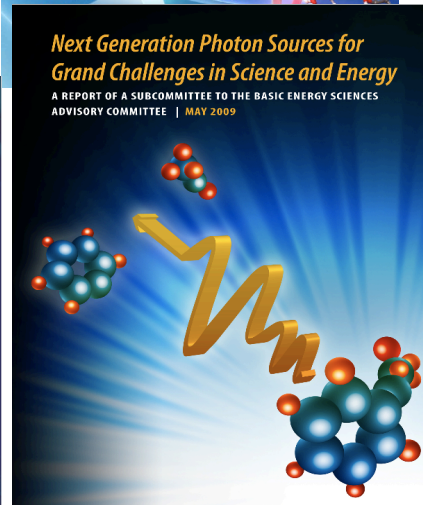
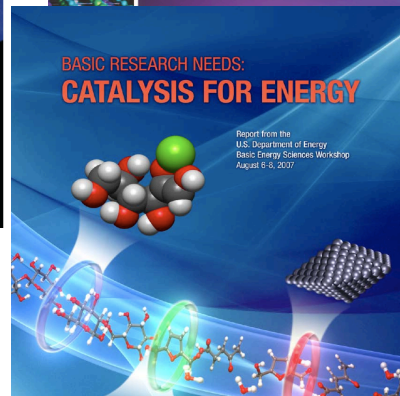
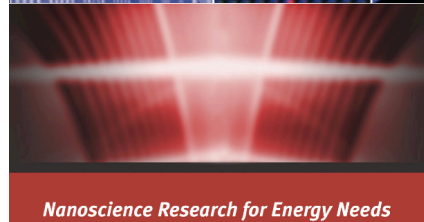
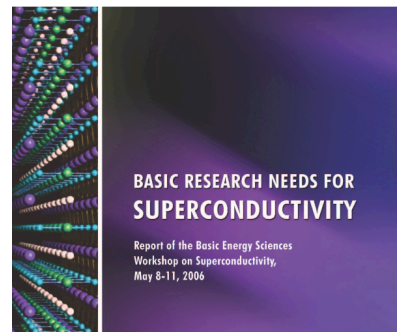
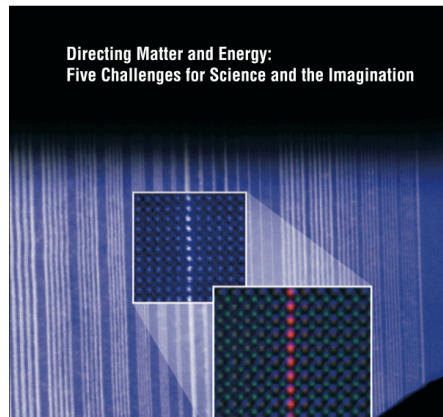
Fulvio Parmigiani

University of Trieste (Italy)

and Sincrotrone Trieste



Vision of the science with future light sources



**CONTROL OF COMPLEX MATERIALS
AND CHEMICAL PROCESSES**

**REAL TIME EVOLUTION
OF CHEMICAL REACTIONS, MOTION
OF ELECTRONS AND SPIN**

**IMAGING AND SPECTROSCOPY
OF INDIVIDUAL NANO-OBJECTS**

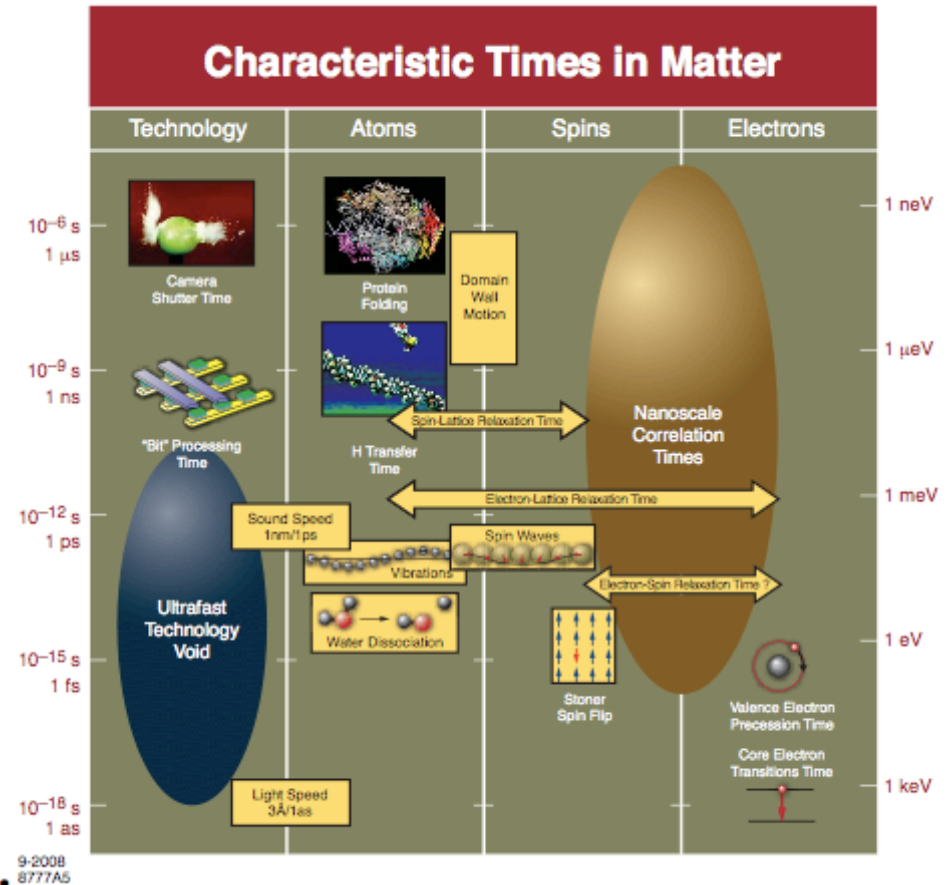
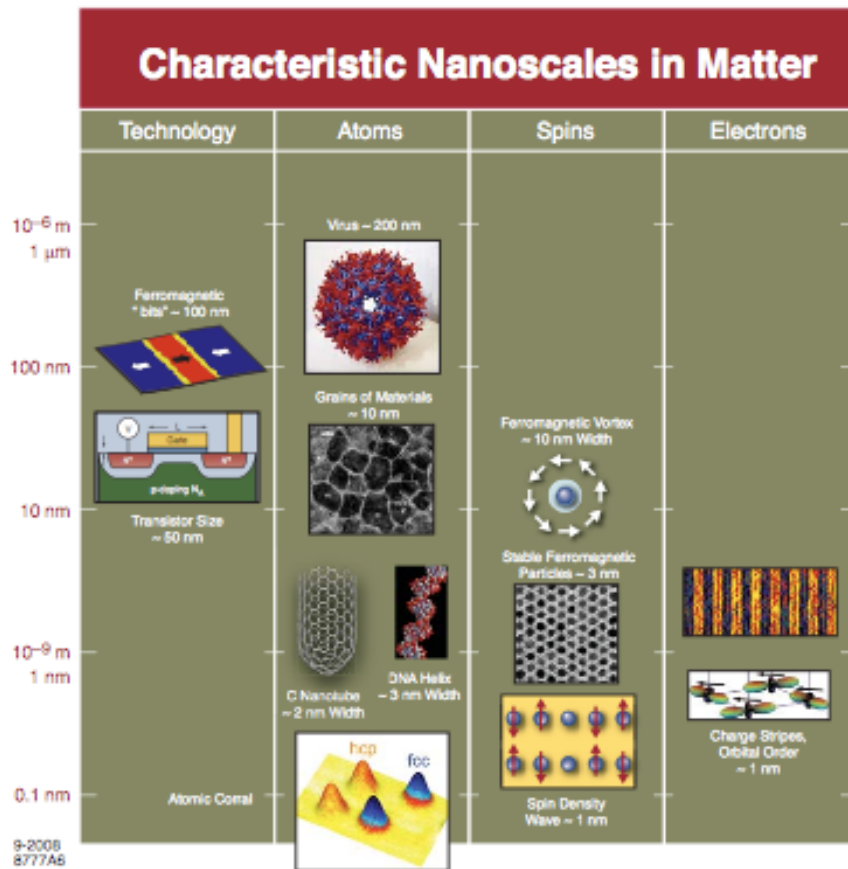
**STATISTICAL LAWS OF COMPLEX
SYSTEMS**

**SIMULTANEOUS ULTRASHORT
AND ULTRAFAST MEASUREMENTS**

Questions

- Can we solve the problem of HTSC ?
Can we understand the coexistence of SC and ferromagnetism?
- Can we make imaging resolution with an information content better than STEM of living matter ?
- Can we make material with a photovoltaic efficiency as in the natural process?
- How small and how fast can we make the magnetic recording devices?
- Can we observe a catalytic process under real operating conditions ?
- Can we fill the gap between the atomic and condensed matter properties ?
- How far can we push our capability to observe the matter under ultra-extreme conditions?

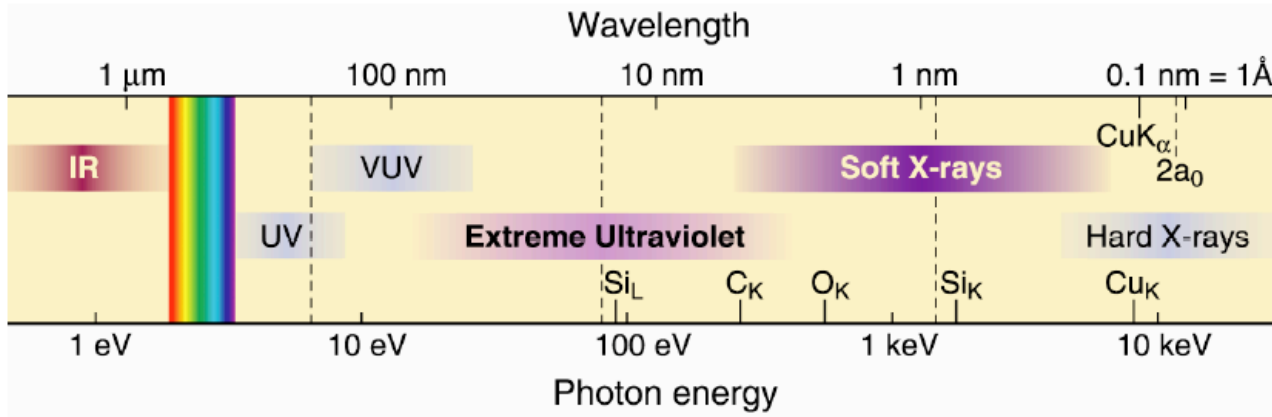
Characteristics



To set the path for probing the matter with the length, time and energy resolution required for unexploring critical and exotic phenomena:
nm, fs (as), and sub-meV.



SR INTRODUCTION

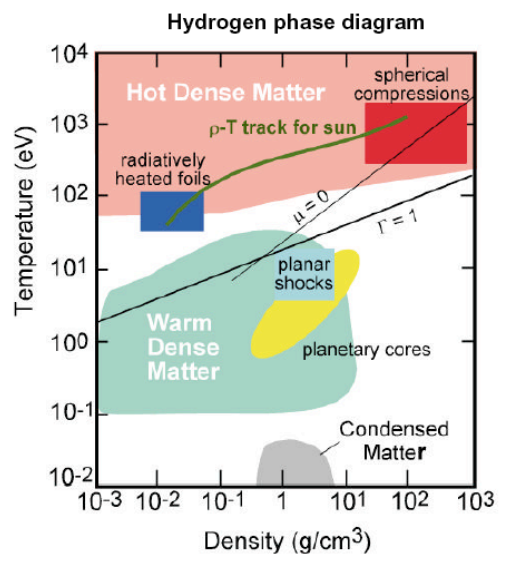
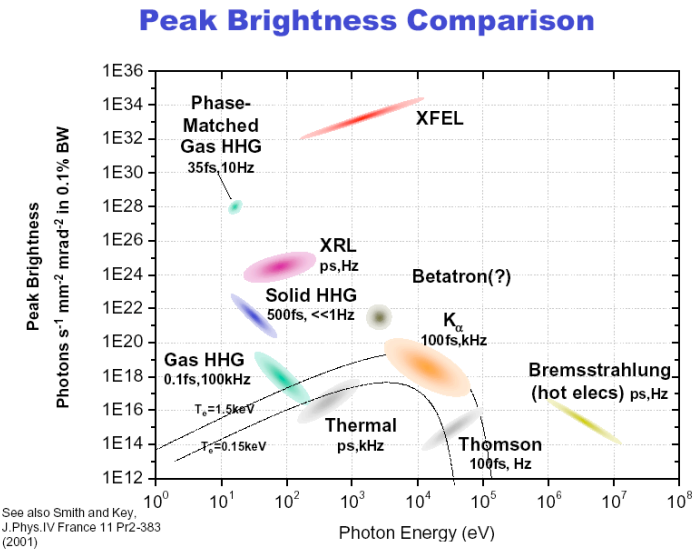
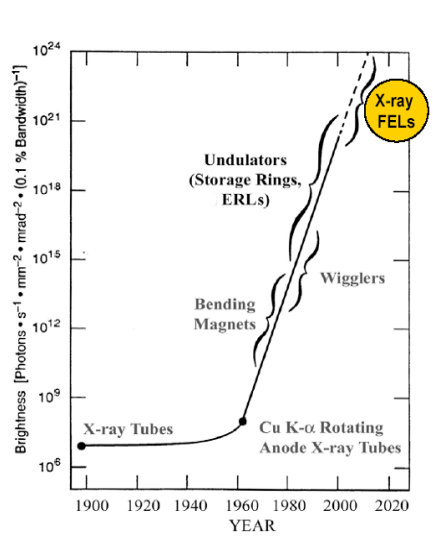


$$\hbar\omega \cdot \lambda = hc = 1239.842 \text{ eV nm}$$

Electronic Structure and Bonding
- where are the **electrons** -

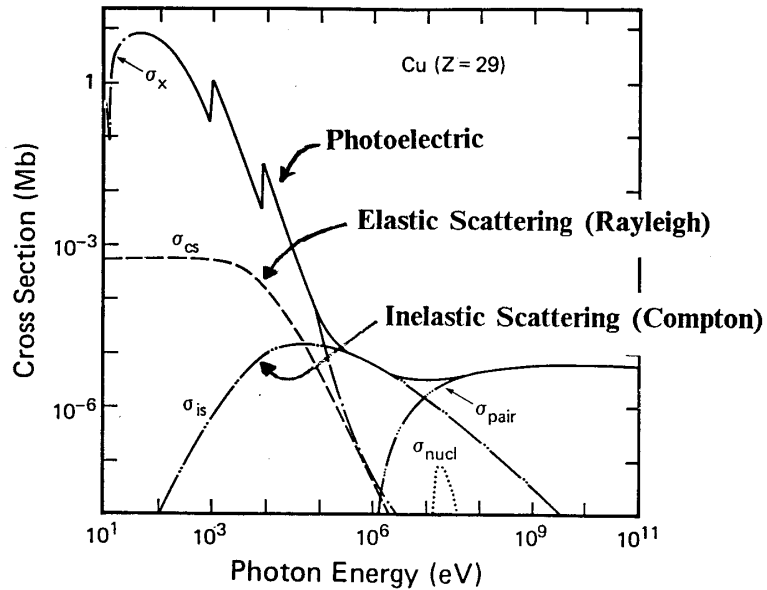
Magnetic Structure and Properties
- where are the **spins** -

Atomic and Molecular Structure
- where are the **atoms** -



Photon-matter interactions

Photon interaction with electrons



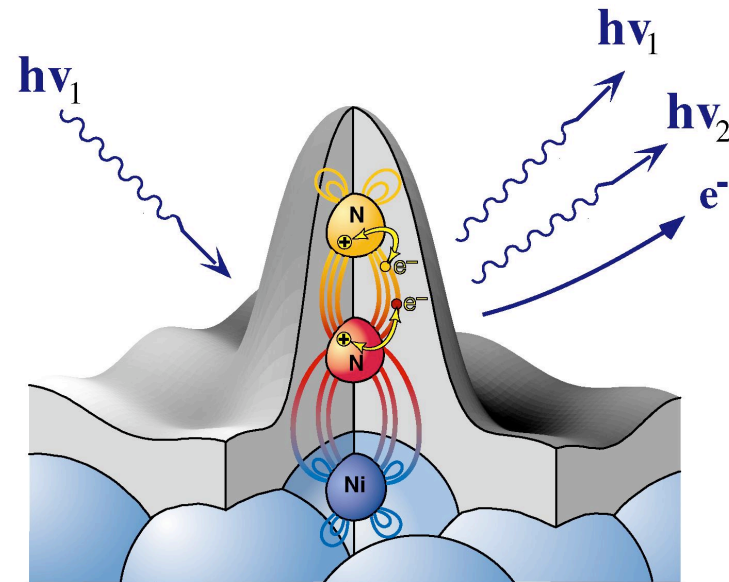
Elastic Scattering

Free electron: **Thomson Scattering**
 Bound Electron: **Rayleigh Scattering**

Inelastic Scattering

Quasi-free electron: **Compton Scattering**

Photon Interaction with Matter



Photon-in

Absorption

Photon-in \longrightarrow Electron-out

Linear: **Electron Photoemission**

Non-Linear: **Multi-photon processes**

Photon-in \longrightarrow Photon-out

Elastic Scattering: **Diffusion and Diffraction**

Inelastic Scattering: **Brillouin and Raman (phononic and electronic), Fluorescence, Resonant Inelastic Scattering**

Scattering and Spectroscopy: Methods

•X-ray Diffraction

Hard X-rays (structure)

Soft X-rays (ordering)

•Photoelectron Spectroscopy (PES)

Core level electron spectroscopy

Micro- and nano-PES

PhotoElectron Emission Microscopy (PEEM)

Angle Resolved PES (ARPES)

Resonant photoemission

Photoelectron Diffraction

•X-ray Absorption Spectroscopy (XAS)

Near Edge X-ray Absorption Spectroscopy (NEXAFS)

Extended X-ray Absorption Fine Structure (EXAFS)

X-ray Magnetic Circular Dichroism (XMCD)

X-ray Magnetic Linear Dichroism (XMLD)

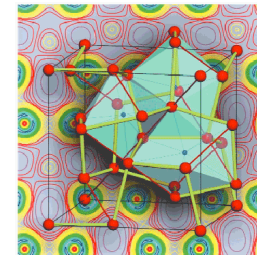
•X-ray Emission Spectroscopy (XES)

Resonant Inelastic X-ray Scattering (RIXS)

•Soft X-ray Elastic Scattering

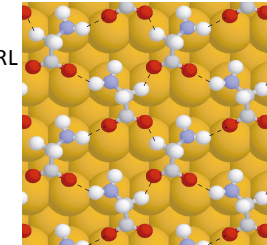
Imaging

Speckle

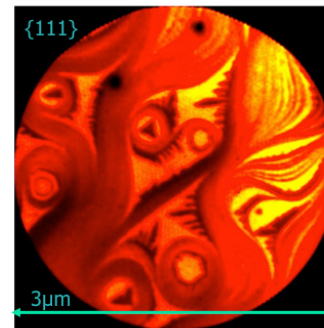


Representation of Li structure at 44 Gpa pressure

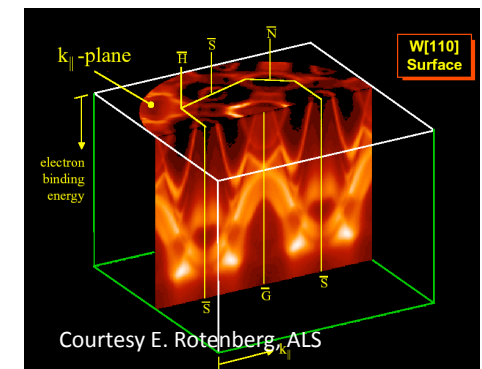
Courtesy, A. Nilsson, SSRL



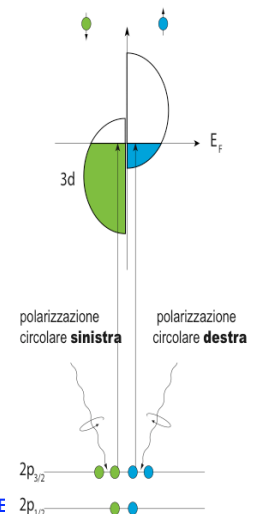
Two dimensional structure of glycine adsorbed on Cu(110)



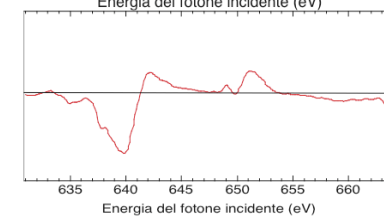
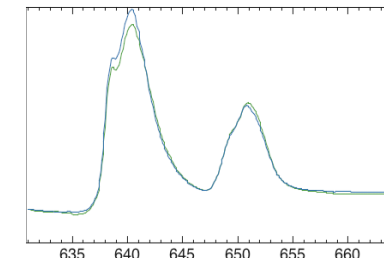
Courtesy Nanospectroscopy, Elettra



Courtesy E. Rotenberg, ALS

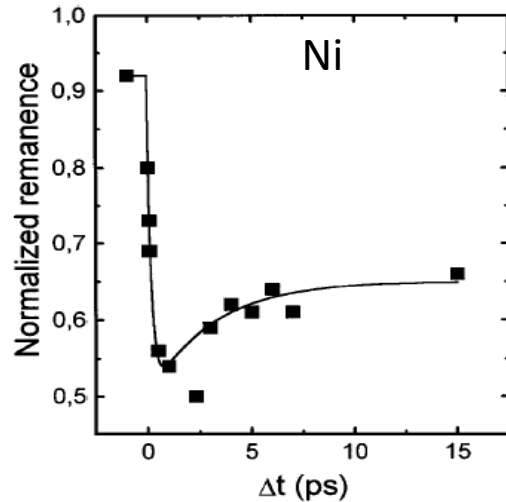


intensità [unità arbitrarie]



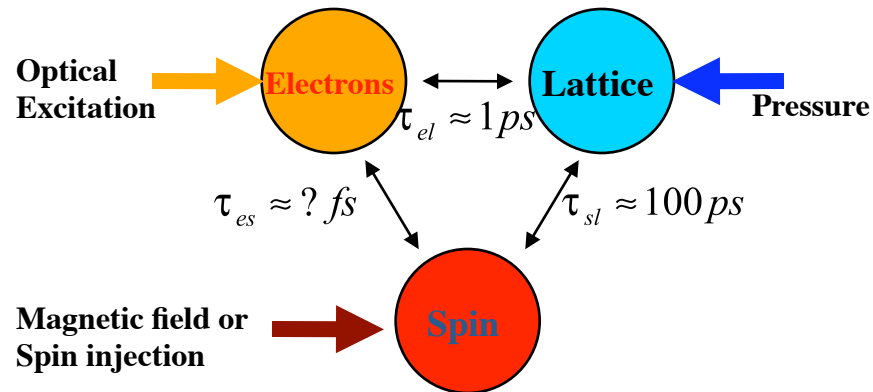
Probing the Properties of Magnetic Materials

Magneto Optical Kerr Effect in the Visible

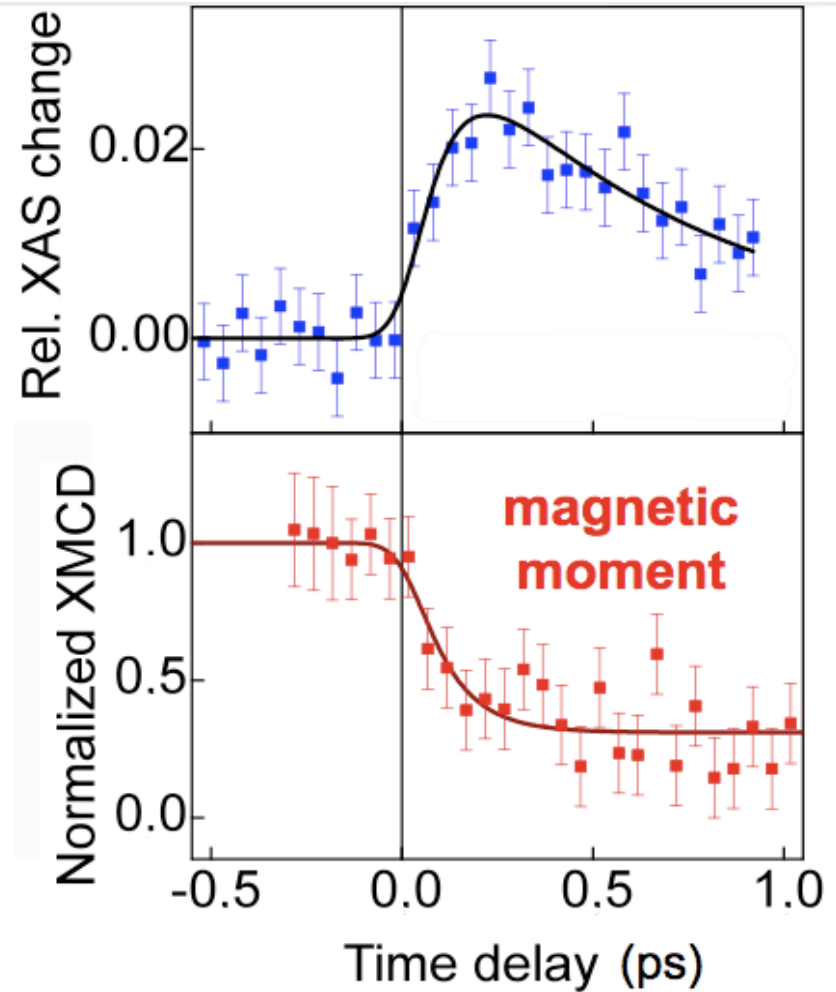


Beaurepaire et al., PRL 76, 4250 (1996)

Non-Equilibrium Magnetization Dynamics:
transfer of energy and angular momentum



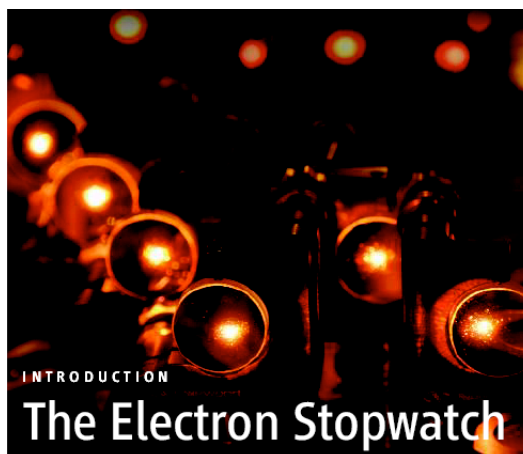
Magnetism: From Fundamentals to Nanoscale Dynamics," J. Stohr & H.C. Siegmann



Stamm et al., Nature Mat. 6, 740(2007)

The 10⁻¹⁸ s Challenges

SPECIAL SECTION



Attosecond Spectroscopy

CONTENTS

Reviews

- 766 The Future of Attosecond Spectroscopy
P. H. Bucksbaum
- 769 Attosecond Control and Measurement: Lightwave Electronics
E. Goulielmakis et al.
- 775 Harnessing Attosecond Science in the Quest for Coherent X-rays
H. Kapteyn et al.

THE PRECISION ATTAINABLE IN TIMING EVENTS ONCE DEPENDED ON HOW fast a human being could press the button on a stopwatch. More recently, pulsed laser sources have taken the place of those hand-held devices for measuring the fastest phenomena. The technology for tracking the time scale of nuclear motion in free molecules and solids was limited by the duration of a single cycle of visible light: approximately 0.00000000000001 second, or 1 femtosecond. Electrons move even faster than that, and for a long time, scientists could only watch their rearrangements as an indistinct blur. Over the past several years, however, laser technology has crossed the threshold into the attosecond regime (a thousandth of a femtosecond). This series of three Reviews highlights the methods underlying this advance and the scientific prospects they have enabled.

Bucksbaum (p. 766) lays out the essential physics of high harmonic generation, a technique whereby an intense laser field pulls an atomic electron away from the nucleus like a loaded slingshot and then sends it careening back, giving rise to the emission of an attosecond light pulse. The Review also describes in general terms what events such light pulses can be used to track, ranging from electron rearrangements in chemical bonding to conduction dynamics in metallic solids.

Goulielmakis *et al.* (p. 769) take a more in-depth look at the laser techniques that create and detect attosecond pulses. Their Review also details the prospects not only of passively probing electron motions, but of actively manipulating and controlling them.

In keeping with the uncertainty principle, compressing a light pulse's duration must also broaden its spectral bandwidth. Thus, attosecond pulses extend into the x-ray region of the electromagnetic spectrum. Kapteyn *et al.* (p. 775) describe efforts to harness this feature of the technology in diffraction and imaging experiments, which would otherwise depend on much more elaborate x-ray generation apparatus.

Optical technology continues to evolve. It seems that just as events at the atomic scale are at last observed with precision, they bring into view a new series of blurs, previously unappreciated. Then the quest begins for an even faster stopwatch.

—IAN OSBORNE AND JAKE YESTON

Harnessing Attosecond Science in the Quest for Coherent X-rays

Henry Kapteyn, Oren Cohen, Ivan Christov, Margaret Murnane*

Modern laser technology has revolutionized the sensitivity and precision of spectroscopy by providing coherent light in a spectrum spanning the infrared, visible, and ultraviolet wavelength regimes. However, the generation of shorter-wavelength coherent pulses in the x-ray region has proven much more challenging. The recent emergence of high harmonic generation techniques opens the door to this possibility. Here we review the new science that is enabled by an ability to manipulate and control electrons on attosecond time scales, ranging from new tabletop sources of coherent x-rays to an ability to follow complex electron dynamics in molecules and materials. We also explore the implications of these advances for the future of molecular structural characterization schemes that currently rely so heavily on scattering from incoherent x-ray sources.

Soft X-ray–Driven Femtosecond Molecular Dynamics

Etienne Gagnon,¹ Predrag Ranitovic,² Xiao-Min Tong,³ C. L. Cocke,² Margaret M. Murnane,¹ Henry C. Kapteyn,¹ Arvinder S. Sandhu^{1*}

Attosecond electron wave packet interferometry

T. REMETTER¹, P. JOHNSON¹, J. MAURITSSON², K. VARJÚ¹, Y. NI³, F. LÉPINE³, E. GUSTAFSSON¹, M. KLING³, J. KHAN³, R. LÓPEZ-MARTENS⁴, K. J. SCHAFFER², M. J. J. VRAKKING³ AND A. L'HUILLIER^{1*}

¹Department of Physics, Lund University, PO Box 118, SE-221 00 Lund, Sweden

²Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803-4001, USA

³FOM-Institute AMOLF, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands

⁴Laboratoire d'Optique Appliquée, Ecole Nationale Supérieure des Techniques Avancées (ENSTA) - Ecole Polytechnique CNRS UMR 7639, 91761 Palaiseau Cedex, France

*e-mail: anne.lhuillier@fysik.lth.se

Science



SR General Concept

PHYSICAL REVIEW

VOLUME 75, NUMBER 12

JUNE 15, 1949

On the Classical Radiation of Accelerated Electrons

JULIAN SCHWINGER

Harvard University, Cambridge, Massachusetts

(Received March 8, 1949)

This paper is concerned with the properties of the radiation from a high energy accelerated electron, as recently observed in the General Electric synchrotron. An elementary derivation of the total rate of radiation is first presented, based on Larmor's formula for a slowly moving electron, and arguments of relativistic invariance. We then construct an expression for the instantaneous power radiated by an electron moving along an arbitrary, prescribed path. By casting this result into various forms, one obtains the angular distribution, the spectral distribution, or the combined angular and spectral distributions of the radiation. The method is based on an examination of the rate at which the electron irreversibly transfers energy to the electromagnetic field, as determined by half the difference of retarded and advanced electric field intensities. Formulas are obtained for an arbitrary charge-current distribution and then specialized to a point charge. The total radiated power and its angular distribution are obtained for an arbitrary trajectory. It is found that the direc-

tion of motion is a strongly preferred direction of emission at high energies. The spectral distribution of the radiation depends upon the detailed motion over a time interval large compared to the period of the radiation. However, the narrow cone of radiation generated by an energetic electron indicates that only a small part of the trajectory is effective in producing radiation observed in a given direction, which also implies that very high frequencies are emitted. Accordingly, we evaluate the spectral and angular distributions of the high frequency radiation by an energetic electron, in their dependence upon the parameters characterizing the instantaneous orbit. The average spectral distribution, as observed in the synchrotron measurements, is obtained by averaging the electron energy over an acceleration cycle. The entire spectrum emitted by an electron moving with constant speed in a circular path is also discussed. Finally, it is observed that quantum effects will modify the classical results here obtained only at extraordinarily large energies.



SR General Concept

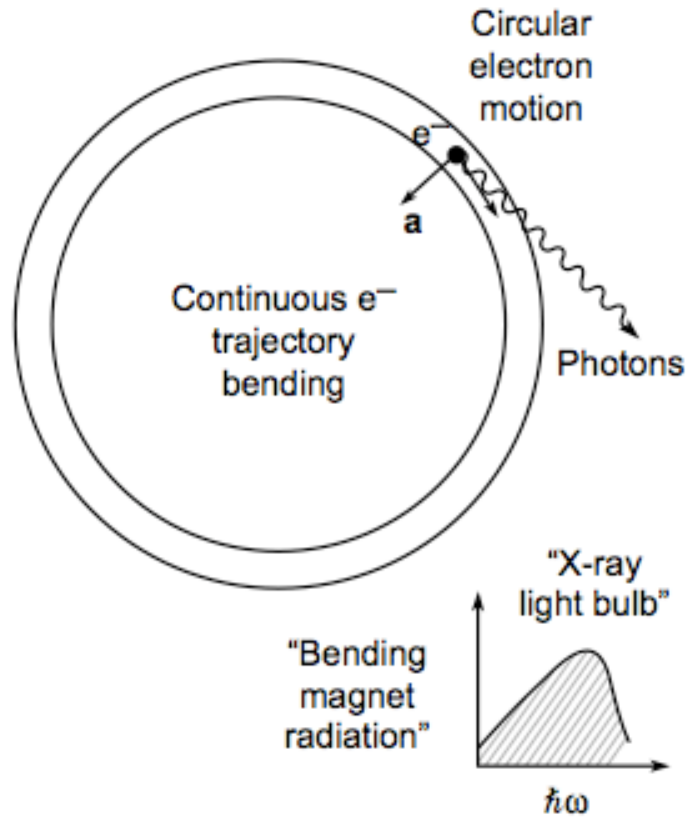


JOINT US-CERN-JAPAN-RUSSIA SCHOOL on Particle Accelerators "SYNCHROTRON RADIATION & FREE ELECTRON LASERS" *Erice 6-15 April, 2011*

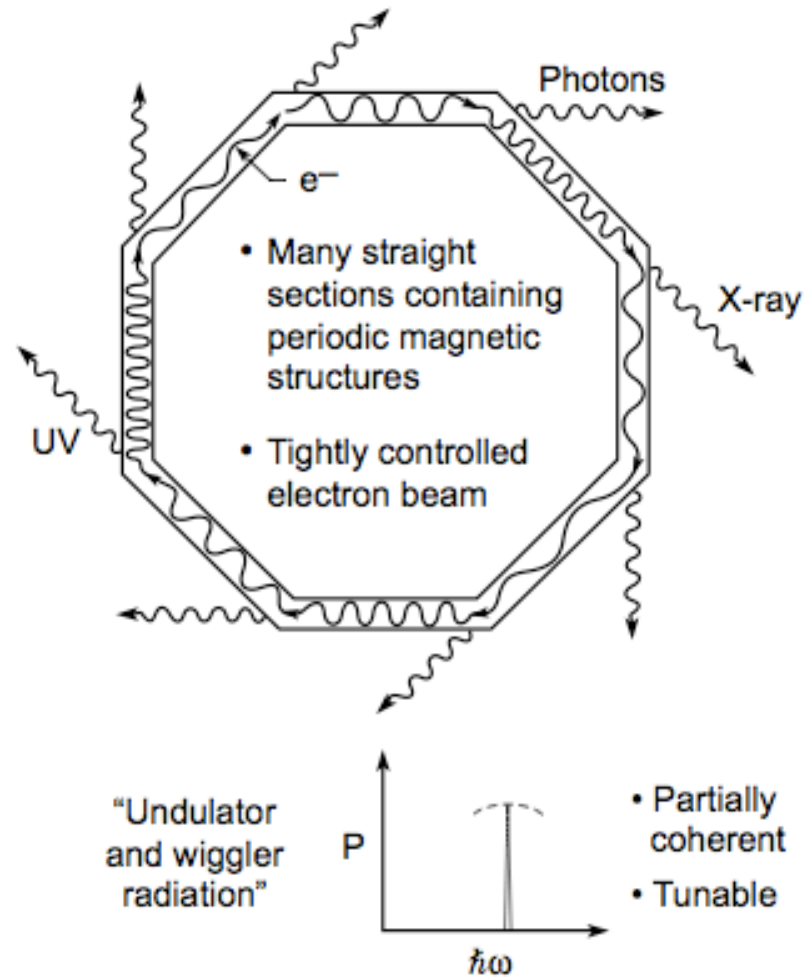


SR General Concept

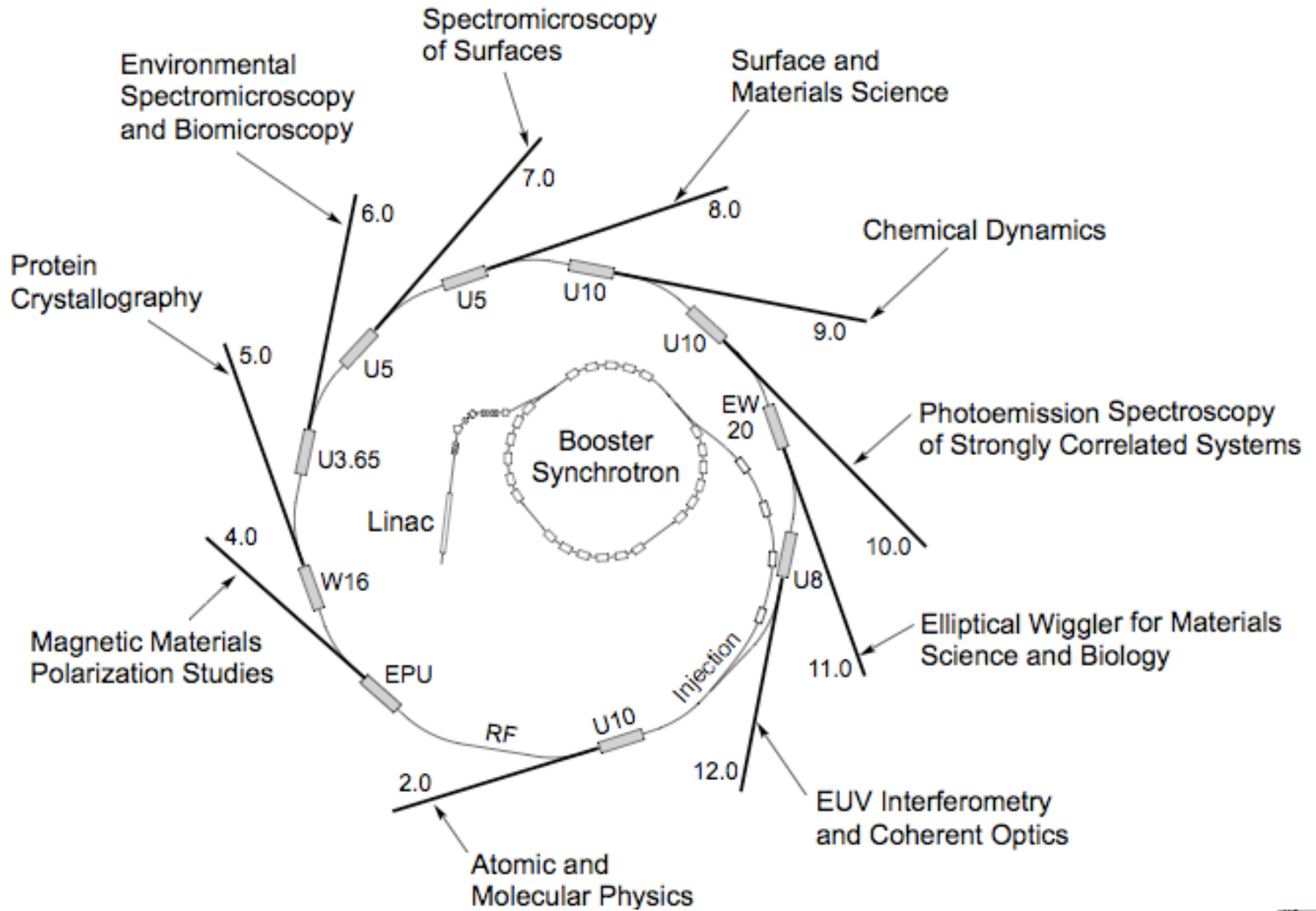
Older Synchrotron Radiation Facility



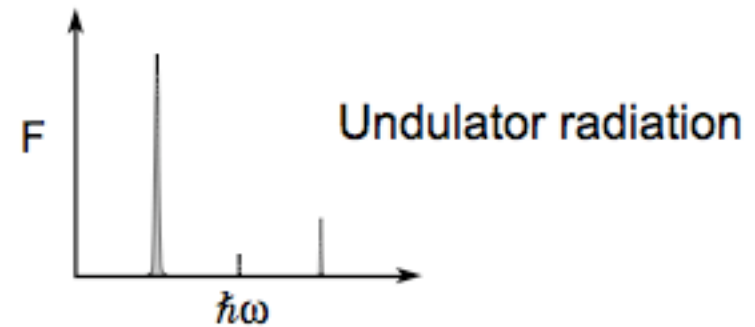
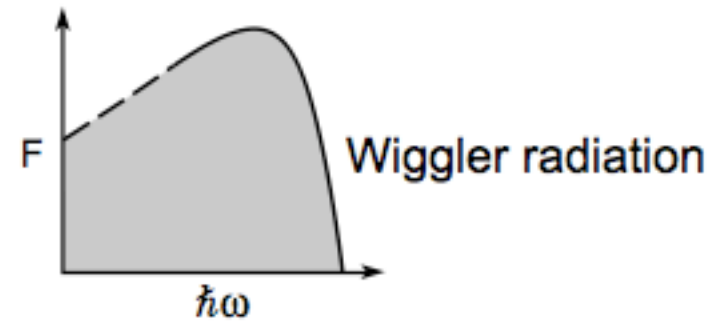
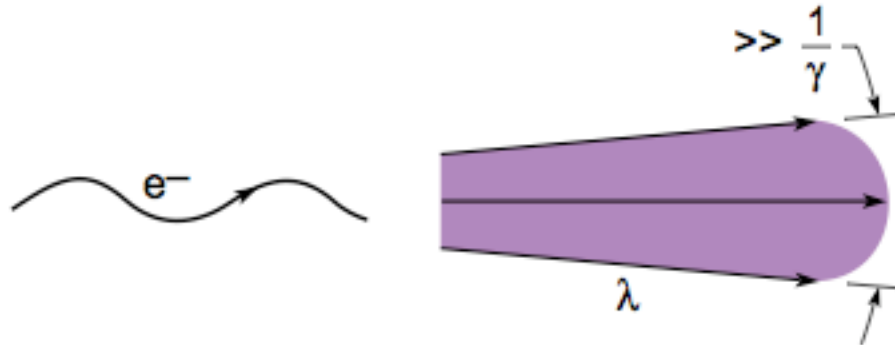
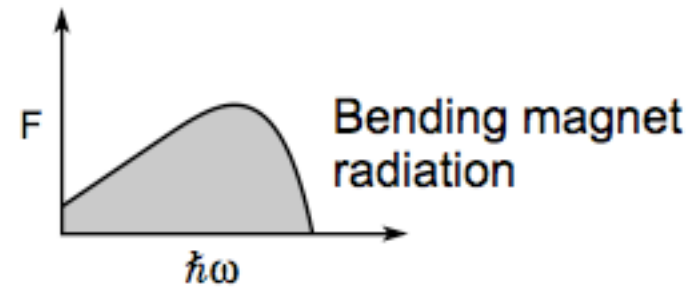
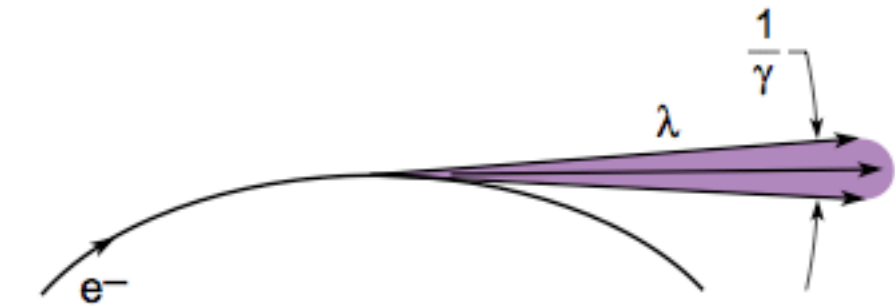
Modern Synchrotron Radiation Facility



SR General Concept



SR General Concept



Future light sources

Third generation x-ray sources

Storage ring

$$\epsilon \sim E^2/R$$

$$\tau_{lifetime} \gg \tau_{relaxation}$$

bunch charge 1nC

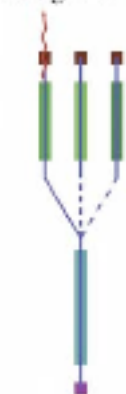


- Many experiments
- Ready tunability
- High flux
- ps pulses

Fourth generation x-ray sources

LINAC source
(=> FEL)

bunch charge ≤ 1 nC



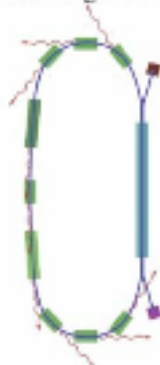
- Extremely high peak brilliance
- Full spatial coherence
- Ultrashort (fs) pulses
- Temporal coherence with seeding in future
- pulse rep. Rate 10^2 to 10^6 Hz
- Few experiments

Energy-Recovery LINAC

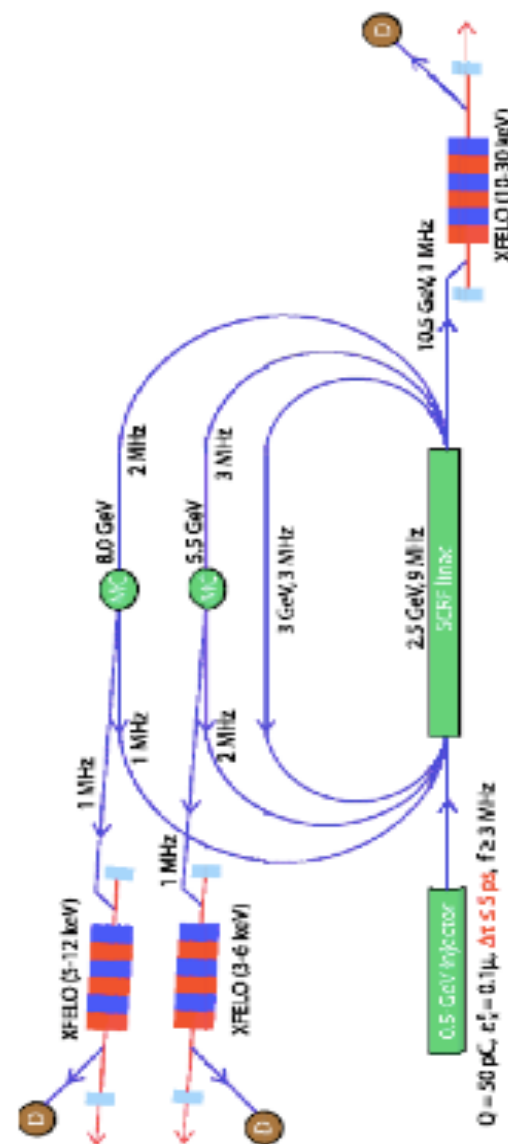
$$\epsilon \sim I/E$$

$$\tau_{lifetime} \ll \tau_{relaxation}$$

bunch charge < 100 pC



- High average brilliance
- Full spatial coherence
- Many experiments
- Ready tunability
- High flux
- Flexible pulse characteristics
- fs to ps pulse lengths
- 10^6 pulses/s



Scheme showing the combination of a superconducting linear accelerator with several return arcs and various X-FEL oscillators (from K.J. Kim).

ANL-08/39

BNL-81895-2008

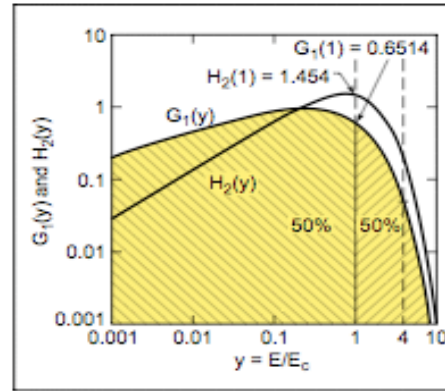
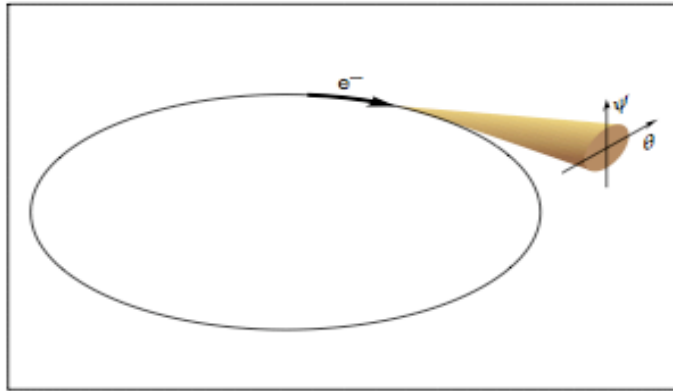
LBNL-1090E-2009

SLAC-R-917

Science and Technology
of Future Light Sources



Bending Magnet Radiation



y	$G_1(y)$	$H_2(y)$
0.0010	2.131×10^{-1}	2.910×10^{-2}
0.0100	4.450×10^{-1}	1.348×10^{-1}
0.1000	8.182×10^{-1}	6.025×10^{-1}
0.3000	9.177×10^{-1}	1.111×10^0
0.5000	8.708×10^{-1}	1.356×10^0
0.7000	7.879×10^{-1}	1.458×10^0
1.000	6.514×10^{-1}	1.454×10^0
3.000	1.286×10^{-1}	5.195×10^{-1}
5.000	2.125×10^{-2}	1.131×10^{-1}
7.000	3.308×10^{-3}	2.107×10^{-2}
10.00	1.922×10^{-4}	1.478×10^{-3}

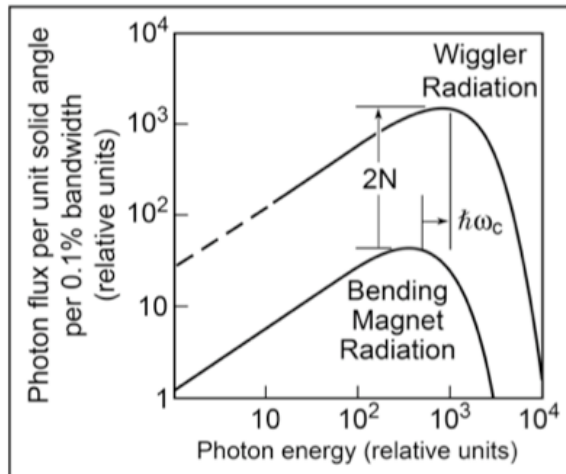
$$E_c = \hbar\omega_c = \frac{3e\hbar B\gamma^2}{2m} \quad (5.7a)$$

$$E_c(\text{keV}) = 0.6650 E_e^2(\text{GeV}) B(\text{T}) \quad (5.7b)$$

$$\gamma = \frac{E_e}{mc^2} = 1957 E_e(\text{GeV}) \quad (5.5)$$

$$\left. \frac{d^3 F_B}{d\theta d\psi d\omega/\omega} \right|_{\psi=0} = 1.33 \times 10^{13} E_e^2(\text{GeV}) I(\text{A}) H_2(E/E_c) \frac{\text{photons/s}}{\text{mrad}^2 \cdot (0.1\% \text{ BW})} \quad (5.6)$$

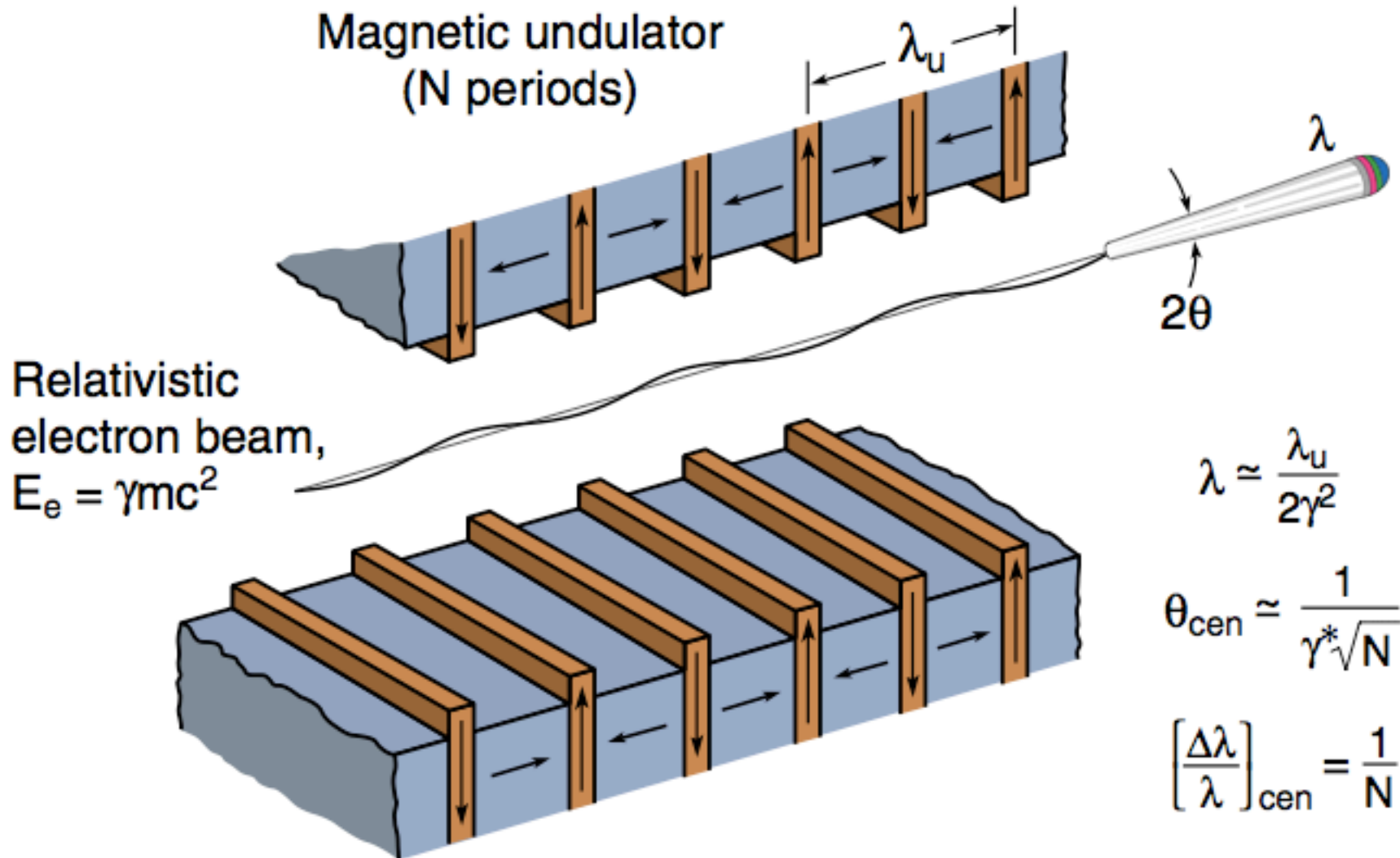
$$\frac{d^2 F_B}{d\theta d\omega/\omega} = 2.46 \times 10^{13} E_e(\text{GeV}) I(\text{A}) G_1(E/E_c) \frac{\text{photons/s}}{\text{mrad} \cdot (0.1\% \text{ BW})} \quad (5.8)$$



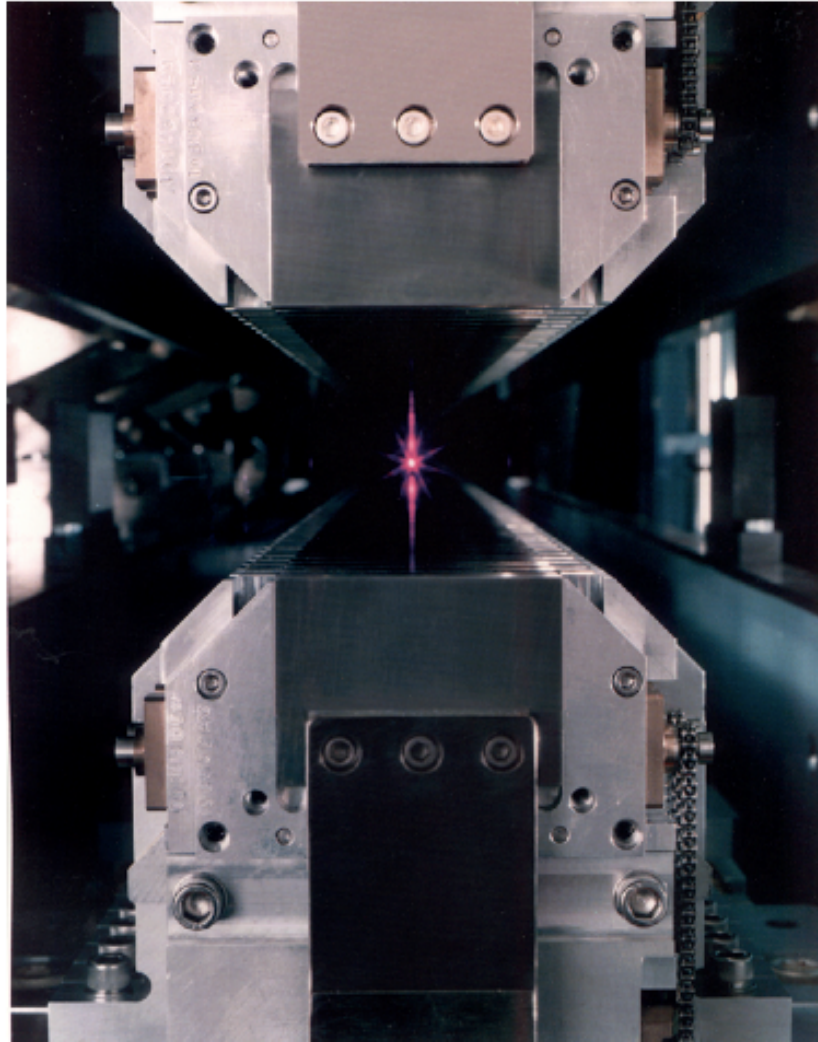
$F_B = \text{Integrated Photon Flux}$

Problem: Calculate the differential flux per unit angle and unit frequency for $\gamma(E/E_c) = 5$, $I = 0.5 \text{ A}$ and $E_e = 3 \text{ GeV}$

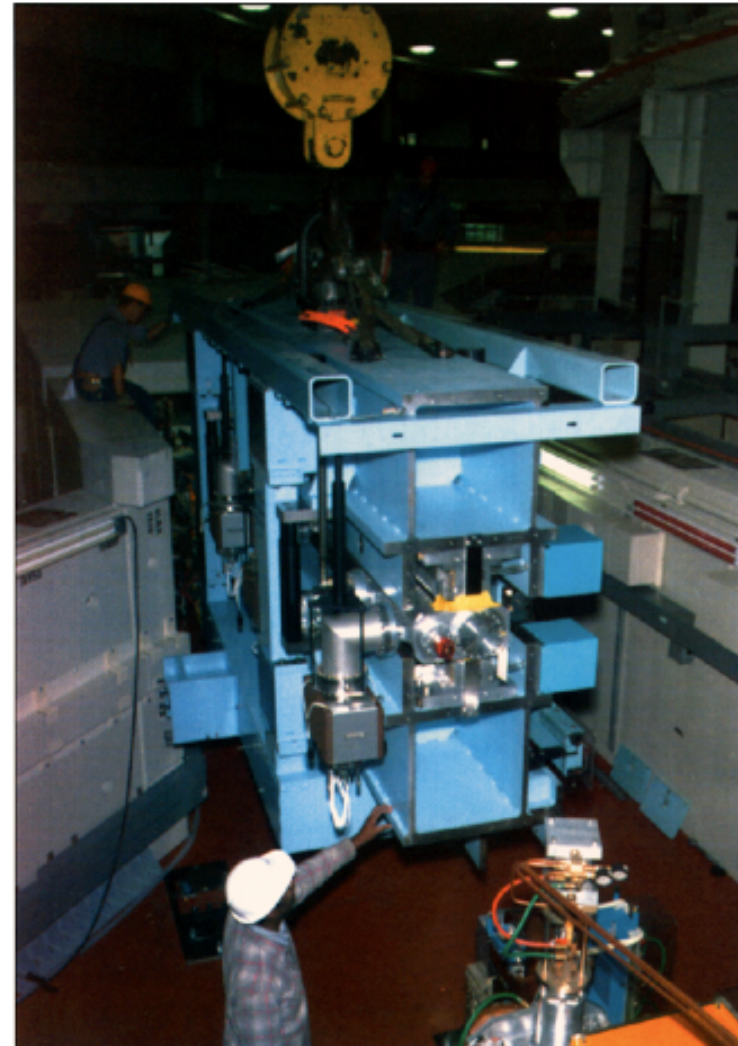
SR Undulators 1



SR Undulators 2



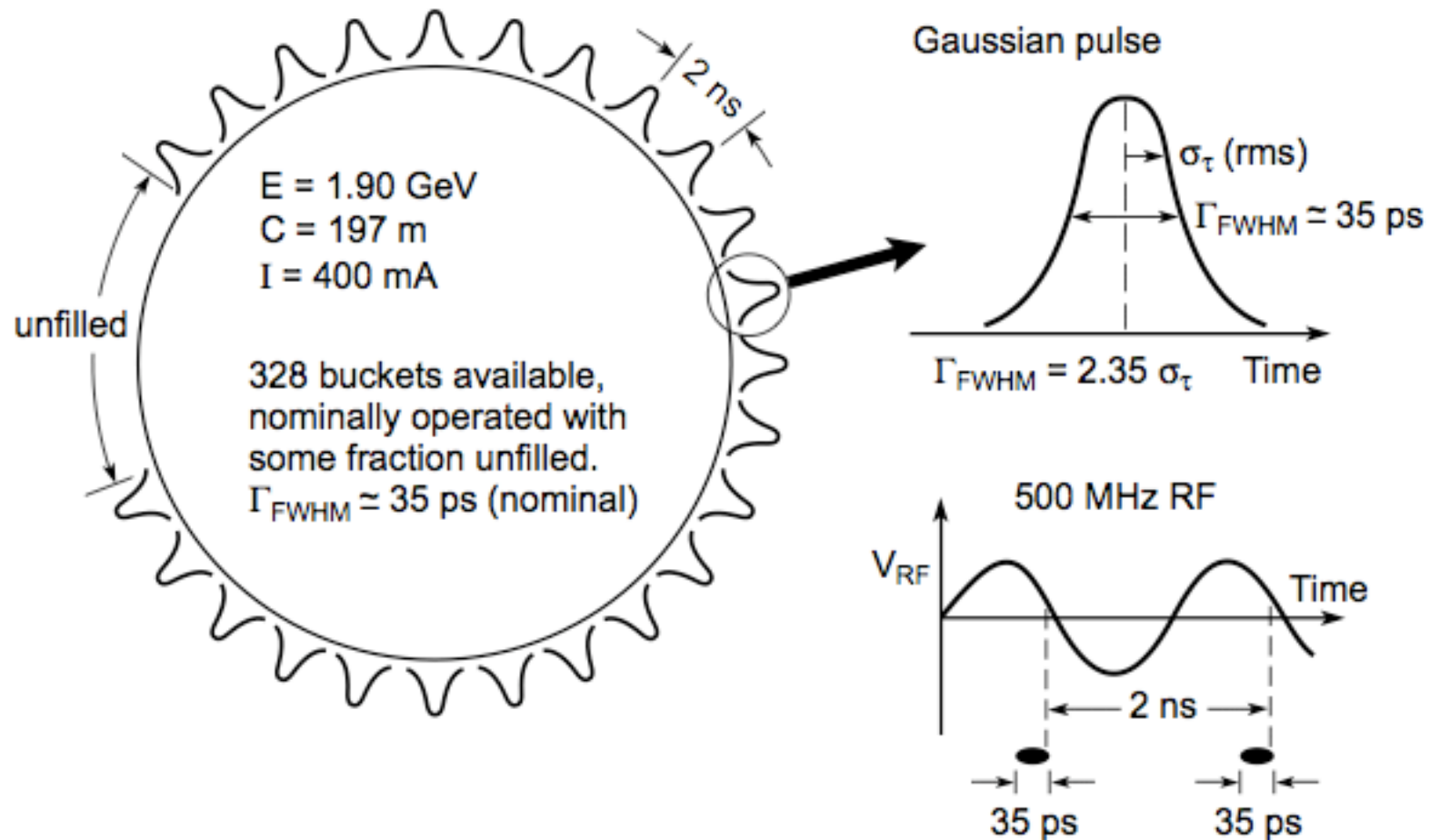
ALS U5 undulator, beamline 7.0, $N = 89$, $\lambda_U = 50$ mm



ALS Beamline 9.0 (May 1994), $N = 55$, $\lambda_U = 80$ mm

SR Electron bunches

The axial electric field within the RF cavity, used to replenish lost (radiated) energy, forms a potential well “bucket” system that forces electrons into axial electron “bunches”. This leads to a time structure in the emitted radiation.

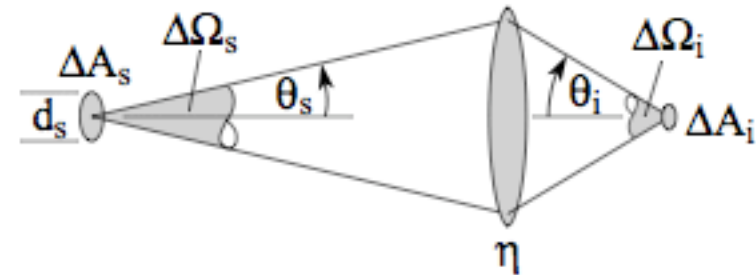


Brightness 1

Brightness is defined as radiated power per unit area and per unit solid angle at the source:

$$B = \frac{\Delta P}{\Delta A \cdot \Delta \Omega} \quad (5.57)$$

Brightness is a conserved quantity in perfect optical systems, and thus is useful in designing beamlines and synchrotron radiation experiments which involve focusing to small areas.

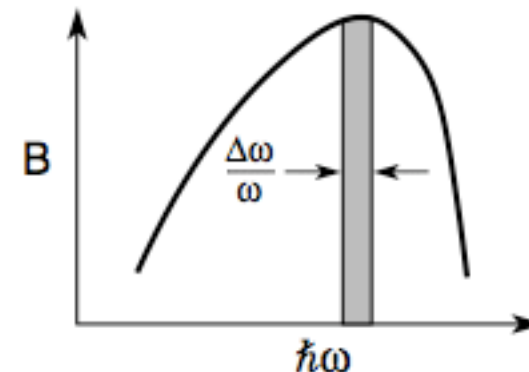


Perfect optical system:

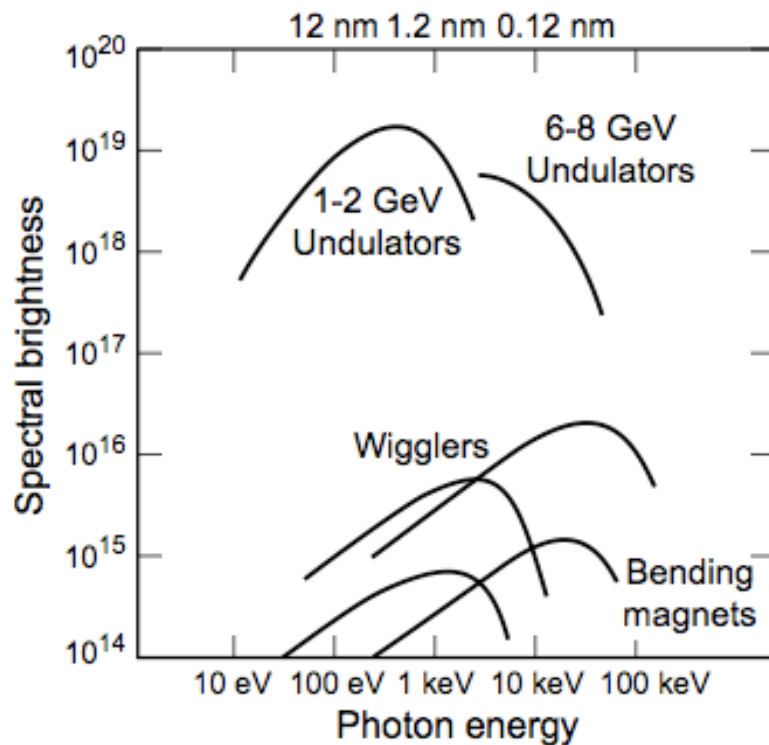
$$\Delta A_s \cdot \Delta \Omega_s = \Delta A_i \cdot \Delta \Omega_i ; \eta = 100\%$$

Spectral brightness is that portion of the brightness lying within a relative spectral bandwidth $\Delta \omega / \omega$:

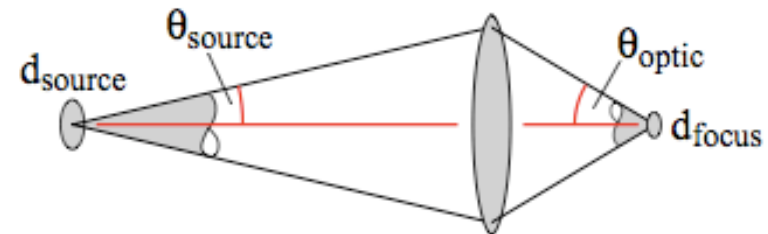
$$B_{\Delta \omega / \omega} = \frac{\Delta P}{\Delta A \cdot \Delta \Omega \cdot \Delta \omega / \omega} \quad (5.58)$$



Brightness 2



- Brightness is conserved (in lossless optical systems)



$$d_{\text{source}} \cdot \theta_{\text{source}} = d_{\text{focus}} \cdot \theta_{\text{optic}}$$

Smaller
after focus

Large in a
focusing optic

- Starting with many photons in a small source area and solid angle, permits high photon flux in an even smaller area

Brightness 3

$$\text{Brilliance} = \frac{\Phi}{\sigma_x \sigma_y \sigma'_x \sigma'_y BW}$$

where:

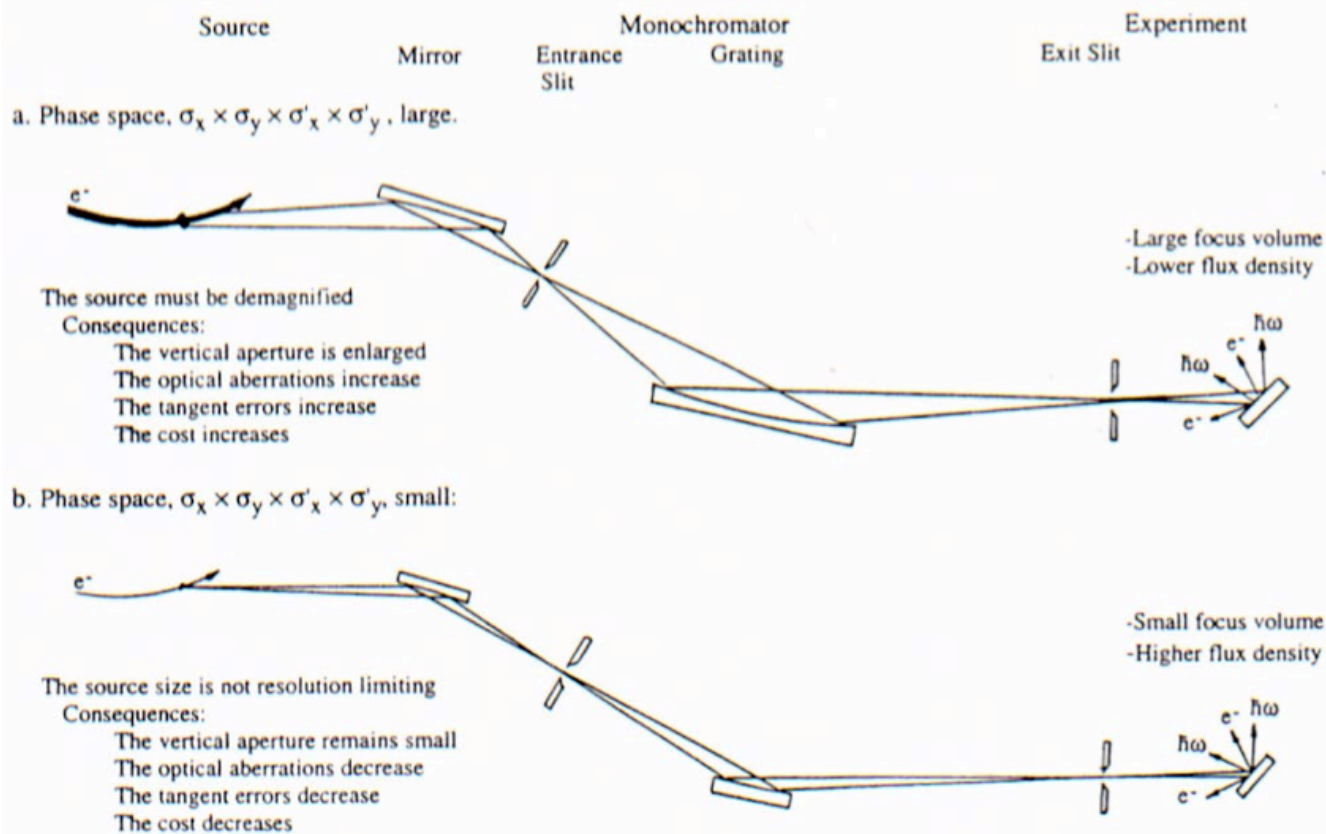
Φ is the photon flux

$\sigma_{x,y}$ are the source sizes

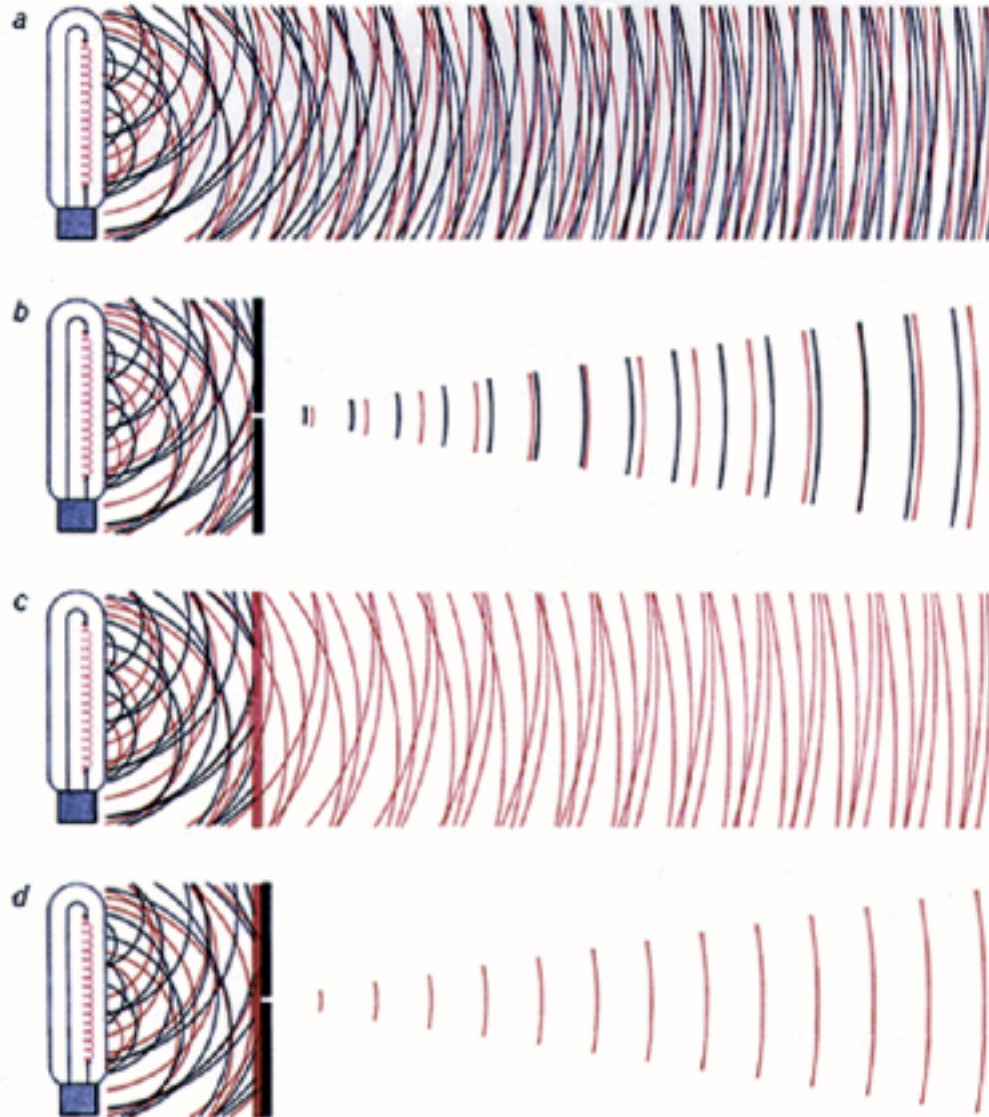
$\sigma'_{x,y}$ are the source divergences

BW is the bandwidth

Figure 1.2.1: The Practical Meaning of Brilliance



Coherence 1



Courtesy of A. Schawlow, Stanford.

Coherence 2

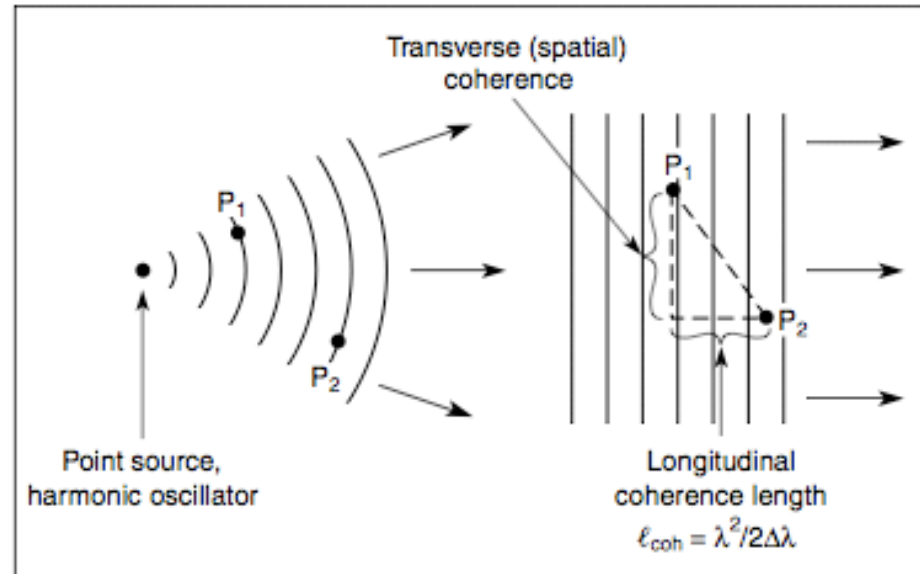
Mutual coherence factor

$$\Gamma_{12}(\tau) \equiv \langle E_1(t + \tau) E_2^*(t) \rangle \quad (8.1)$$

Normalize degree of spatial coherence
(complex coherence factor)

$$\mu_{12} = \frac{\langle E_1(t) E_2^*(t) \rangle}{\sqrt{\langle |E_1|^2 \rangle} \sqrt{\langle |E_2|^2 \rangle}} \quad (8.12)$$

A high degree of coherence ($\mu \rightarrow 1$) implies an ability to form a high contrast interference (fringe) pattern. A low degree of coherence ($\mu \rightarrow 0$) implies an absence of interference, except with great care. In general radiation is partially coherent.



Longitudinal (temporal) coherence length

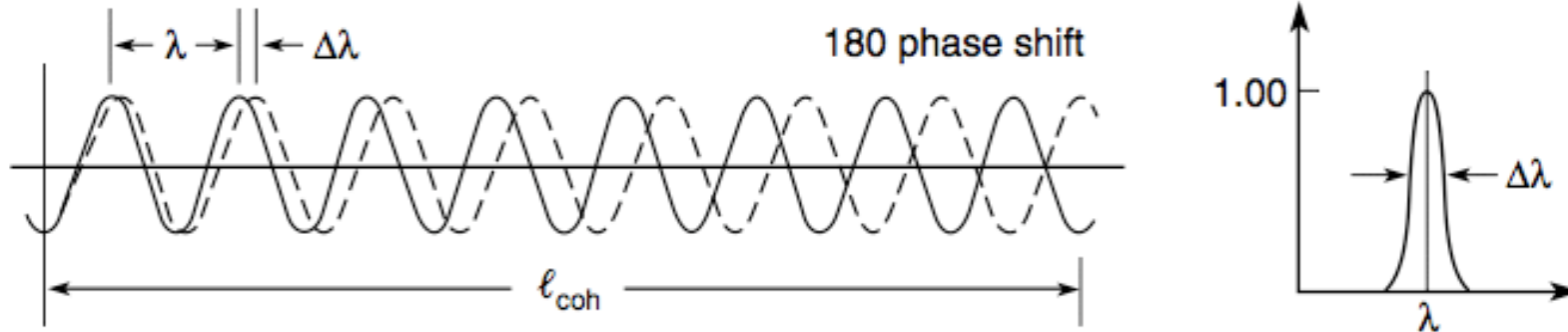
$$l_{\text{coh}} = \frac{\lambda^2}{2 \Delta\lambda} \quad (8.3)$$

Full spatial (transverse) coherence

$$d \cdot \theta = \lambda / 2\pi \quad (8.5)$$

See any text book of OPTICS

Coherence 3



Define a coherence length ℓ_{coh} as the distance of propagation over which radiation of spectral width $\Delta\lambda$ becomes 180° out of phase. For a wavelength λ propagating through N cycles

$$\ell_{\text{coh}} = N\lambda$$

and for a wavelength $\lambda + \Delta\lambda$, a half cycle less $(N - \frac{1}{2})$

$$\ell_{\text{coh}} = (N - \frac{1}{2})(\lambda + \Delta\lambda)$$

Equating the two

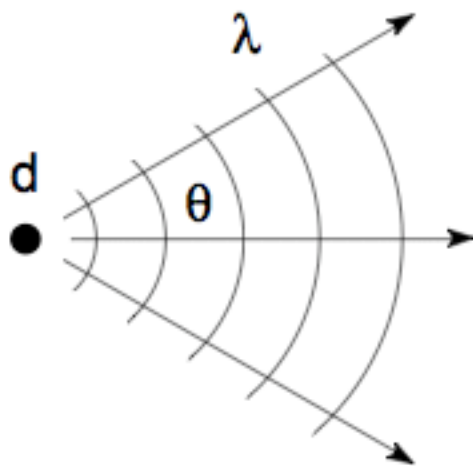
$$N = \lambda/2\Delta\lambda$$

so that

$$\boxed{\ell_{\text{coh}} = \frac{\lambda^2}{2 \Delta\lambda}} \quad (8.3)$$

Coherence 4

- Associate spatial coherence with a spherical wavefront.
- A spherical wavefront implies a point source.
- How small is a “point source”?



From Heisenberg's Uncertainty Principle ($\Delta x \cdot \Delta p \geq \frac{\hbar}{2}$), the smallest source size “d” you can resolve, with wavelength λ and half angle θ , is

$$d \cdot \theta = \frac{\lambda}{2\pi}$$

Coherence 5

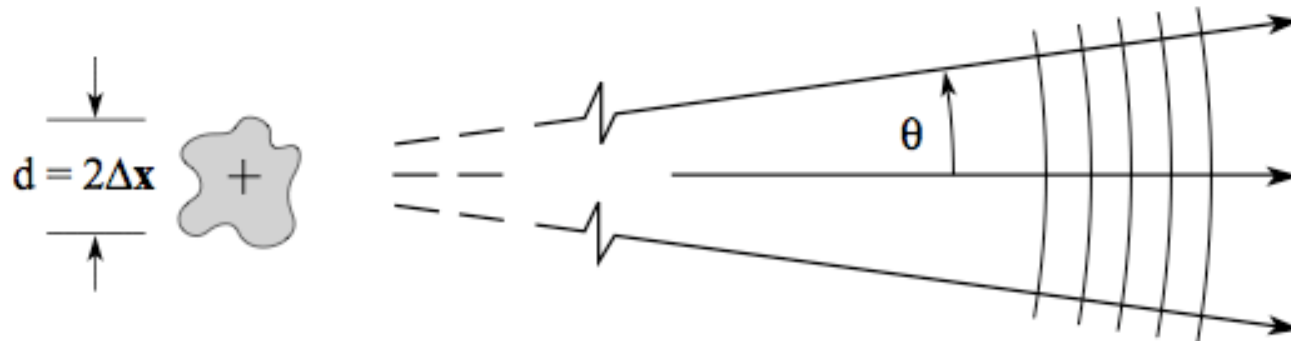
$$\Delta \mathbf{x} \cdot \Delta \mathbf{p} \geq \hbar/2 \quad (8.4)$$

$$\Delta \mathbf{x} \cdot \hbar \Delta \mathbf{k} \geq \hbar/2$$

$$\Delta \mathbf{x} \cdot \mathbf{k} \Delta \theta \geq 1/2$$

$$2\Delta \mathbf{x} \cdot \Delta \theta \geq \lambda/2\pi$$

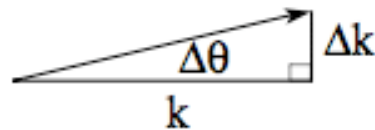
Standard deviations of Gaussian distributed functions
(Tipler, 1978, pp. 174-189)



Note:

$$\Delta \mathbf{p} = \hbar \Delta \mathbf{k}$$

$$\Delta \mathbf{k} = \mathbf{k} \Delta \theta$$



Spherical wavefronts occur
in the limiting case

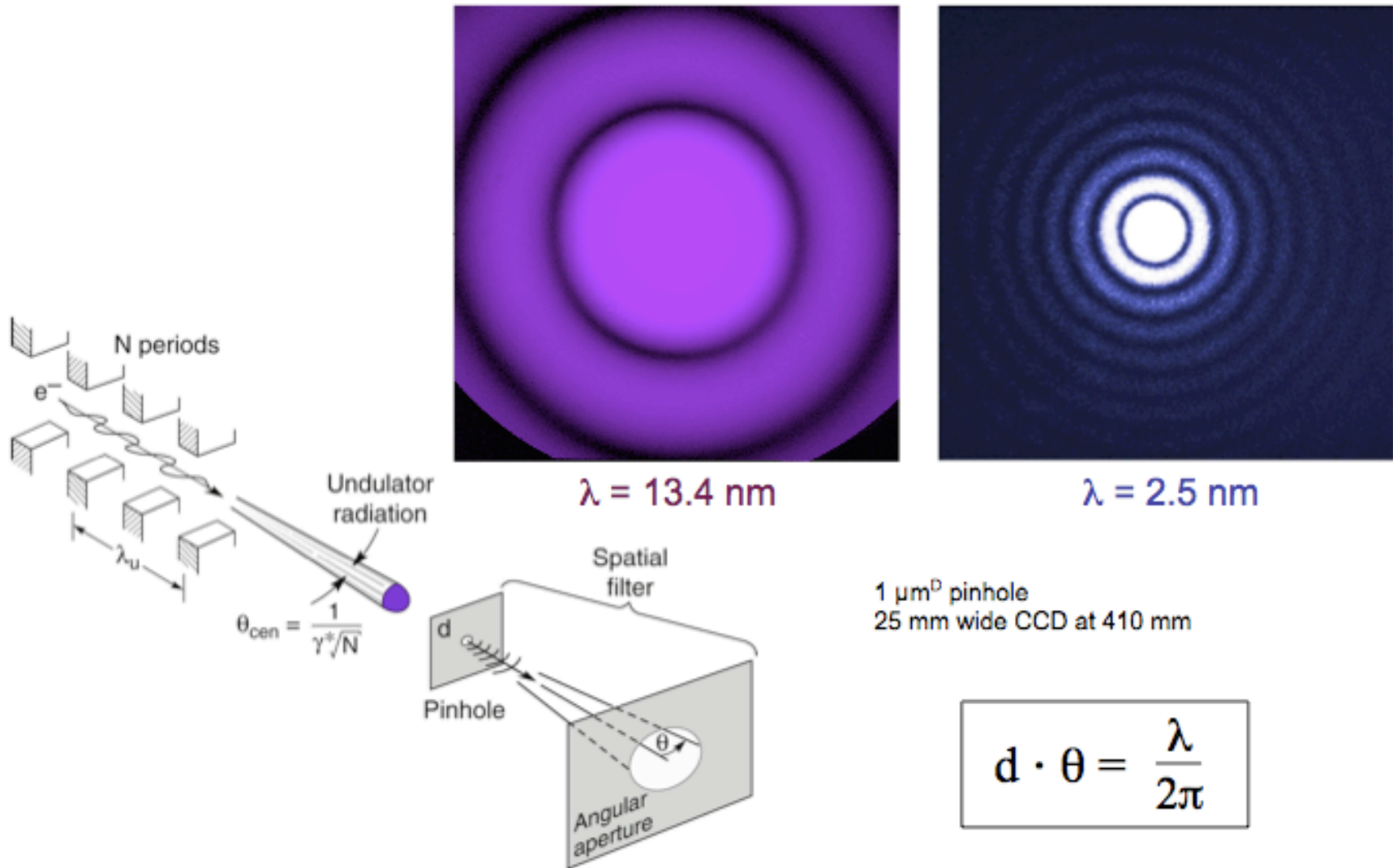
$$\boxed{d \cdot \theta = \lambda/2\pi} \left. \vphantom{\boxed{d \cdot \theta = \lambda/2\pi}} \right\} \frac{1}{\sqrt{e}} \text{ quantities}$$

(spatially coherent)

or

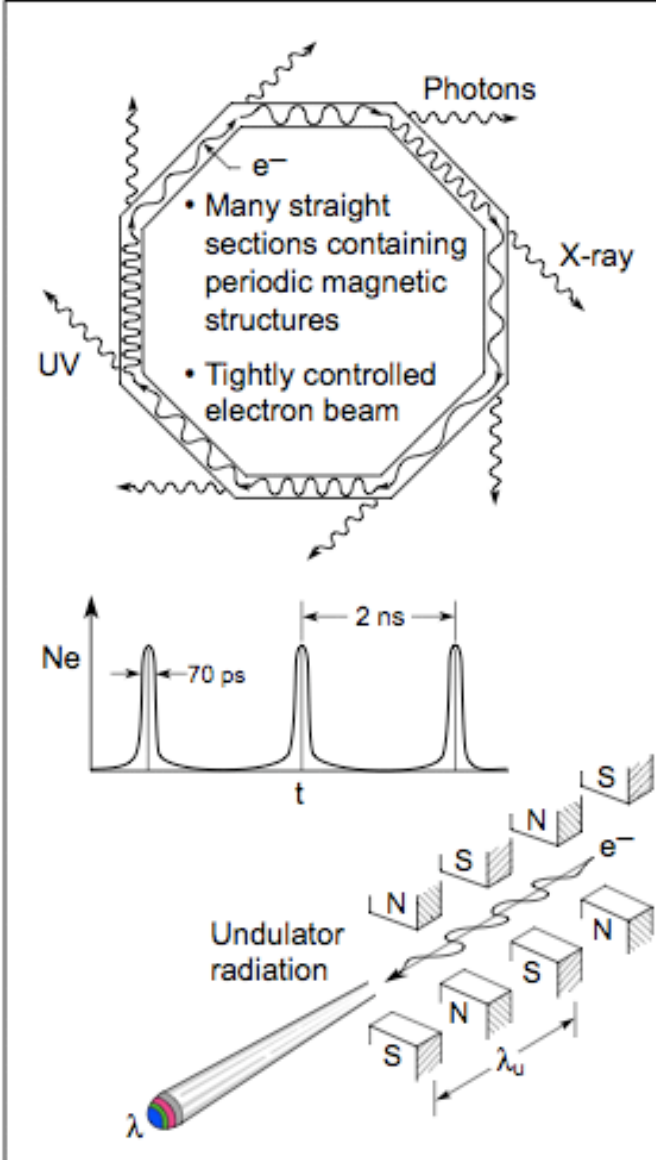
$$(d \cdot 2\theta)_{\text{FWHM}} \approx \lambda/2 \left. \vphantom{(d \cdot 2\theta)_{\text{FWHM}} \approx \lambda/2} \right\} \text{FWHM quantities}$$

Coherence 6



Courtesy of Patrick Naulleau, LBNL / Kris Rosfjord, UCB and LBNL

SR BM-Wiggler-Undulator –Summary 1



Photons

e^-

- Many straight sections containing periodic magnetic structures
- Tightly controlled electron beam

UV

X-ray

Ne

70 ps

2 ns

t

Undulator radiation

e^-

λ_u

λ

Bending Magnet:

$$\hbar\omega_c = \frac{3e\hbar B\gamma^2}{2m} \quad (5.7)$$

Wiggler:

$$\hbar\omega_c = \frac{3e\hbar B\gamma^2}{2m} \quad (5.80)$$

$$n_c = \frac{3K}{4} \left(1 + \frac{K^2}{2}\right) \quad (5.82)$$

$$P_T = \frac{\pi e K^2 \gamma^2 I N}{3\epsilon_0 \lambda_u} \quad (5.85)$$

Undulator:

$$\lambda = \frac{\lambda_u}{2\gamma^2} \left(1 + \frac{K^2}{2} + \gamma^2\theta^2\right) \quad (5.28)$$

$$K = \frac{eB_0\lambda_u}{2\pi mc} \quad (5.18)$$

$$\theta_{cen} = \frac{1}{\gamma^*\sqrt{N}} \quad (5.15)$$

$$\left.\frac{\Delta\lambda}{\lambda}\right|_{cen} = \frac{1}{N} \quad (5.14)$$

$$\bar{P}_{cen} = \frac{\pi e \gamma^2 I}{\epsilon_0 \lambda_u} \frac{K^2}{\left(1 + \frac{K^2}{2}\right)^2} f(K) \quad (5.41)$$

SR BM-Wiggler-Undulator –Summary 2

Bending magnet radiation

- Broad spectrum
- Good photon flux
- No heat load
- Less expensive
- Easier access

Wiggler radiation

- Higher photon energies
- More photon flux
- Expensive magnet structure
- Expensive cooled optics
- Less access

Undulator radiation

- Brighter radiation
- Smaller spot size
- Partial coherence
- Expensive
- Less access

Problem: Describe the main differences in terms of physical mechanisms among A BM a wiggler and an undulator

