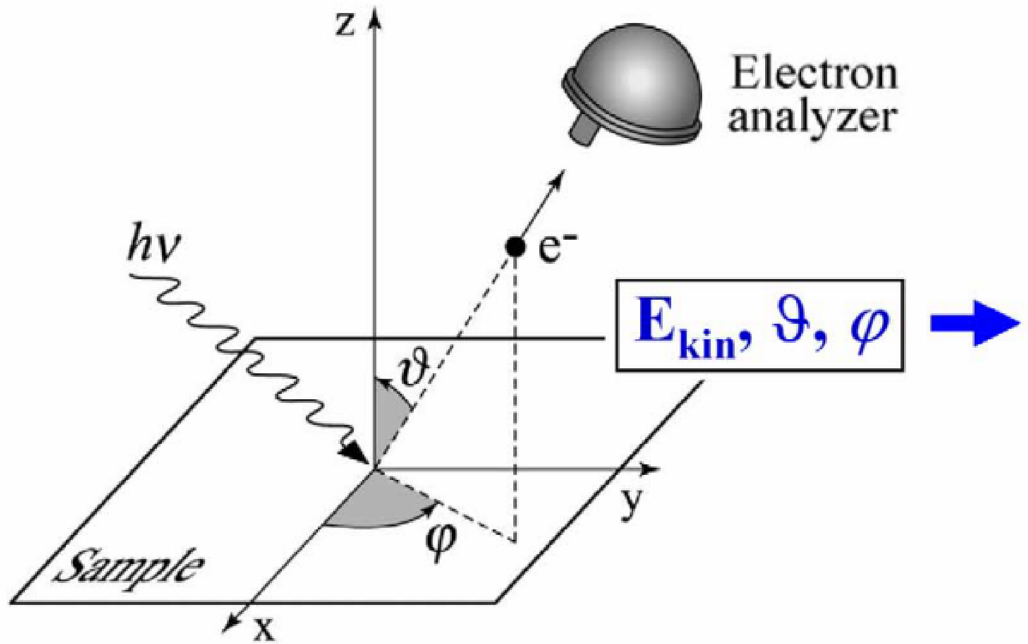
ARPES principle



$$\mathbf{K} = \mathbf{p}/\hbar = \sqrt{2m\boldsymbol{E_{kin}}}/\hbar$$

$$K_x = \frac{1}{\hbar}\sqrt{2mE_{kin}}\sin\vartheta\cos\varphi$$

$$K_y = \frac{1}{\hbar}\sqrt{2mE_{kin}}\sin\vartheta\sin\varphi$$

$$K_z = \frac{1}{\hbar}\sqrt{2mE_{kin}}\cos\vartheta$$

Vacuum

E_{kin}

$\vec{K}$

Conservation laws

$$E_{kin} = h\nu - |E_B| - \Phi$$

$$\vec{K}_{||} = \vec{k}_{||} + \cancel{\vec{k}_{photon}}$$

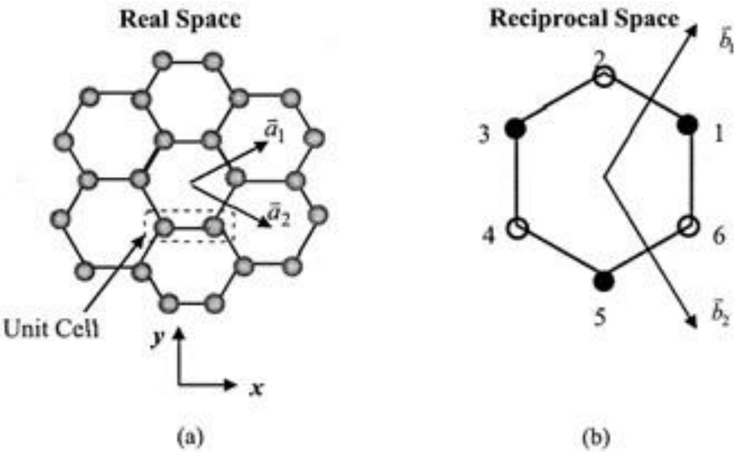
Solid

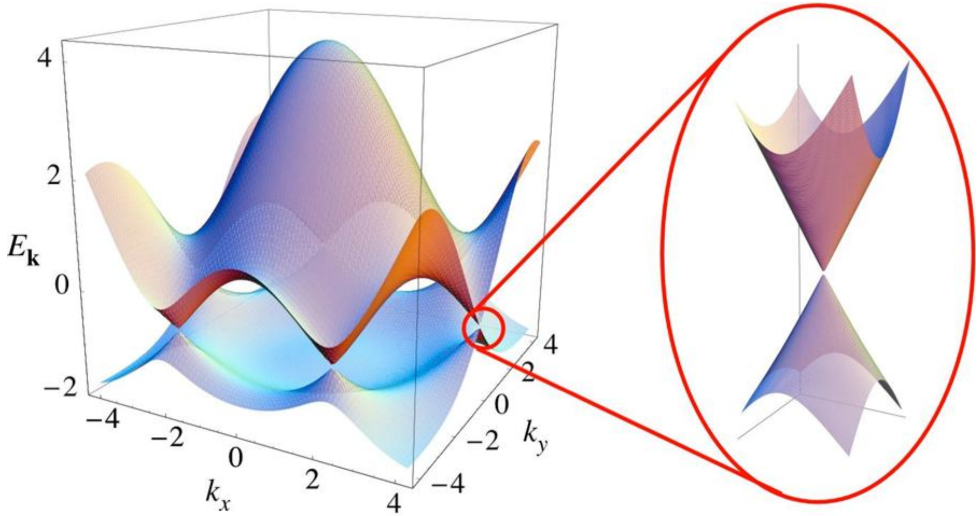
E_B

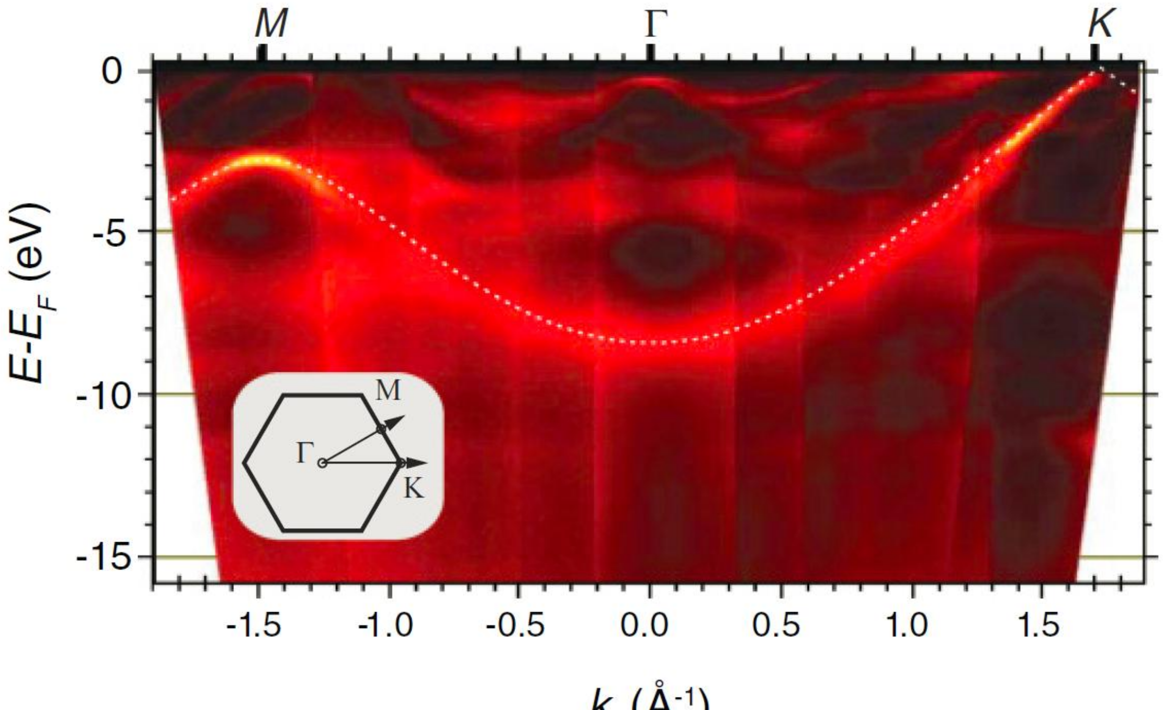
$\vec{k}_{||}$

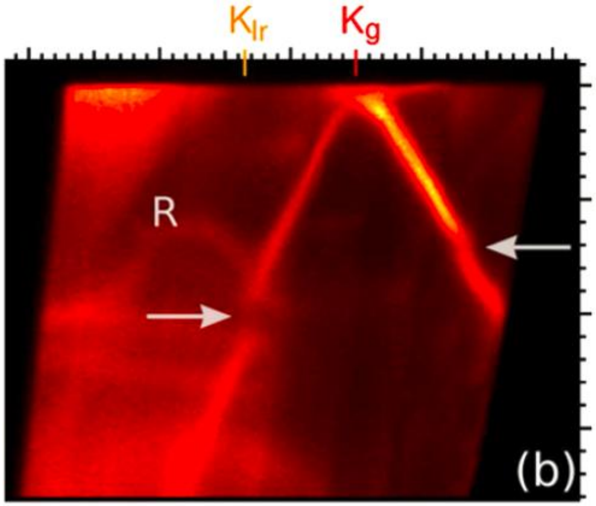
conservation of parallel momentum

ARPES on graphene









Inverse photoemission spectroscopy

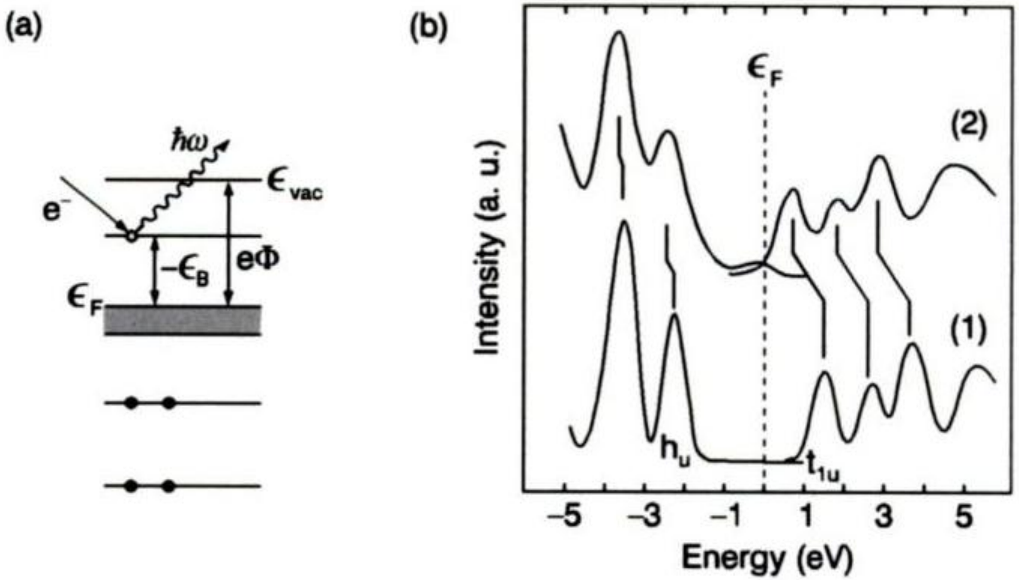
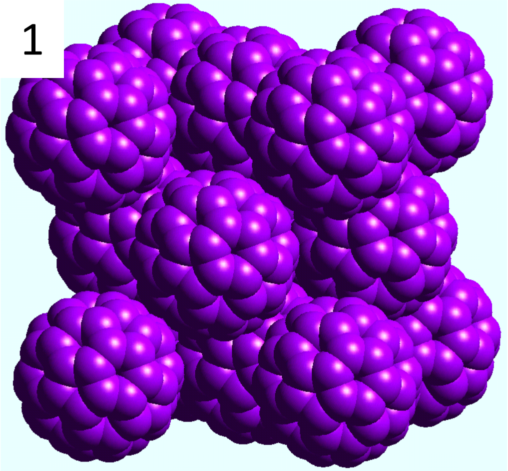
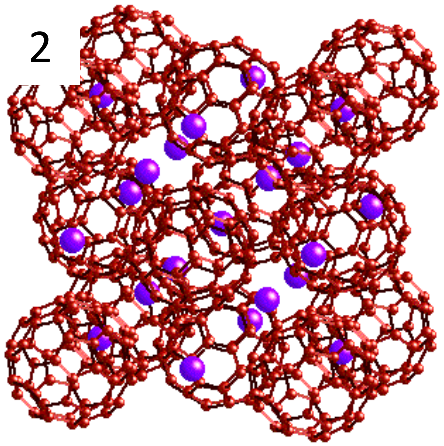


Fig. 12.19. Electronic levels and recombination processes for inverse photoemission (**a**) and photoemission and inverse photoemission for C_{60} (1) and K_3C_{60} (2) (**b**); The Mullikan symbols h_u and t_{1u} label the symmetry of the bands; (b) after [12.15].

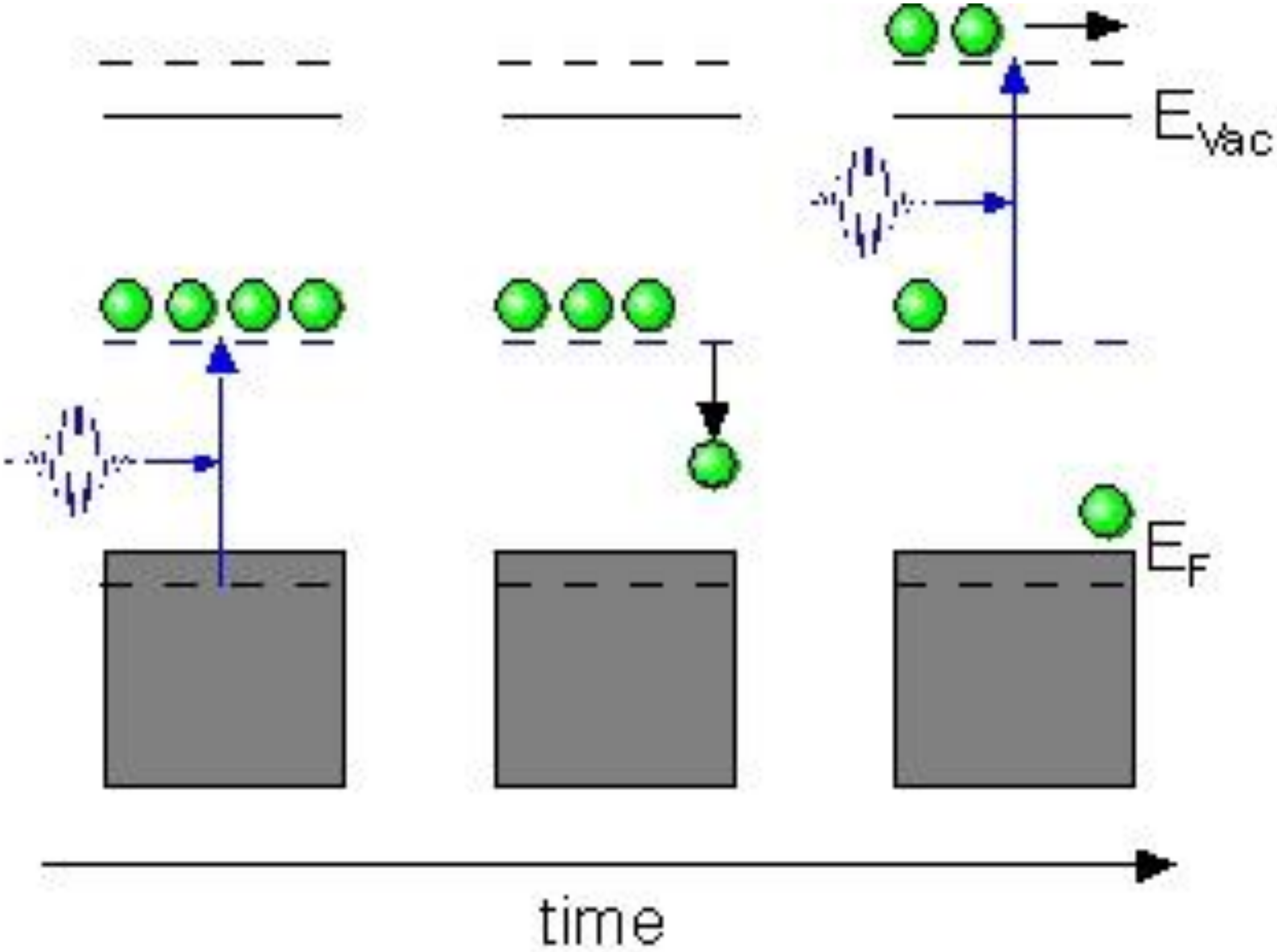
1



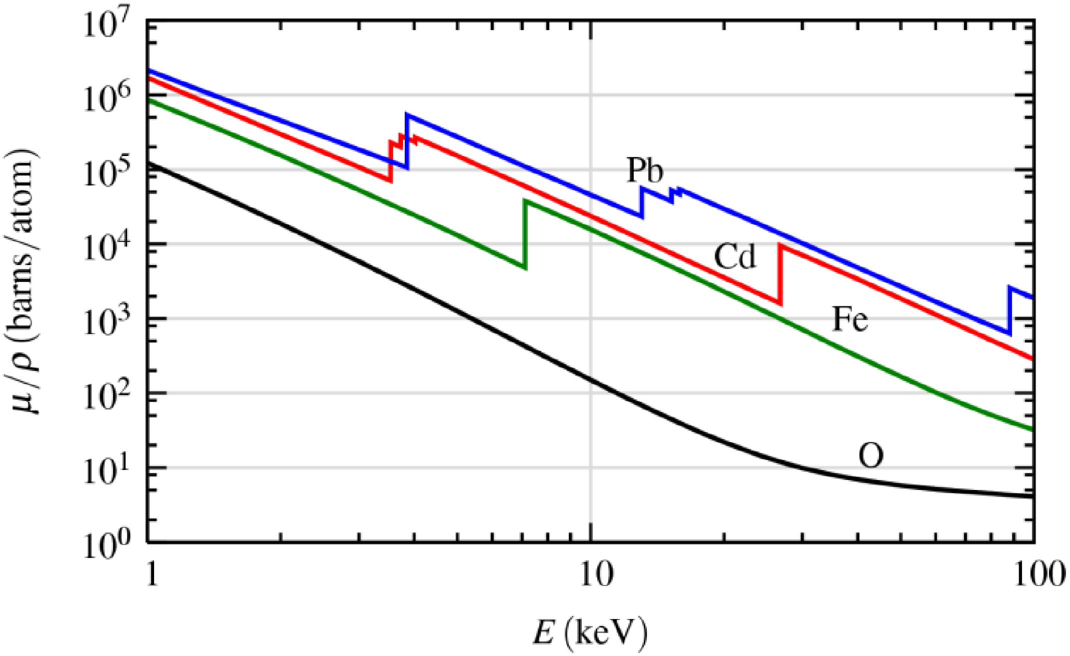
2



Two-photon photoemission (2PPE)



X-ray adsorption spectroscopy



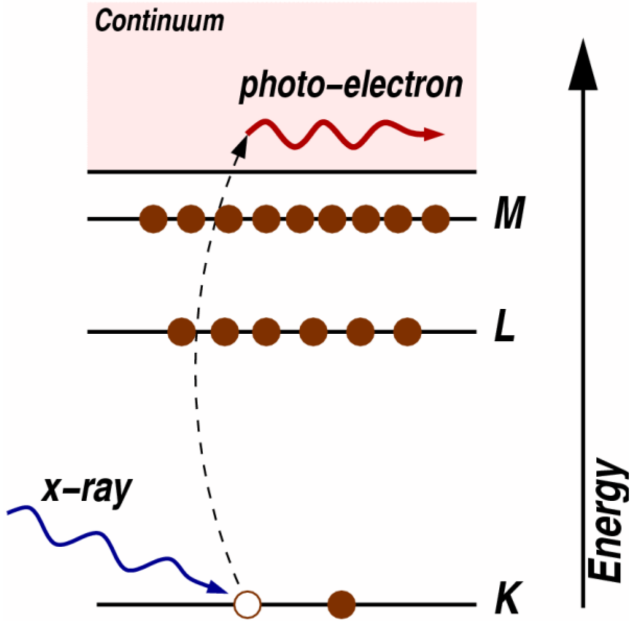


Figure 2.3: The absorption cross-section μ/ρ for several elements over the x-ray energy range of 1 to 100 keV. Notice that there are at least 5 orders of magnitude in variation in μ/ρ, and that in addition to the strong energy dependence, there are also sharp rises corresponding to the core-level binding energies of the atoms.

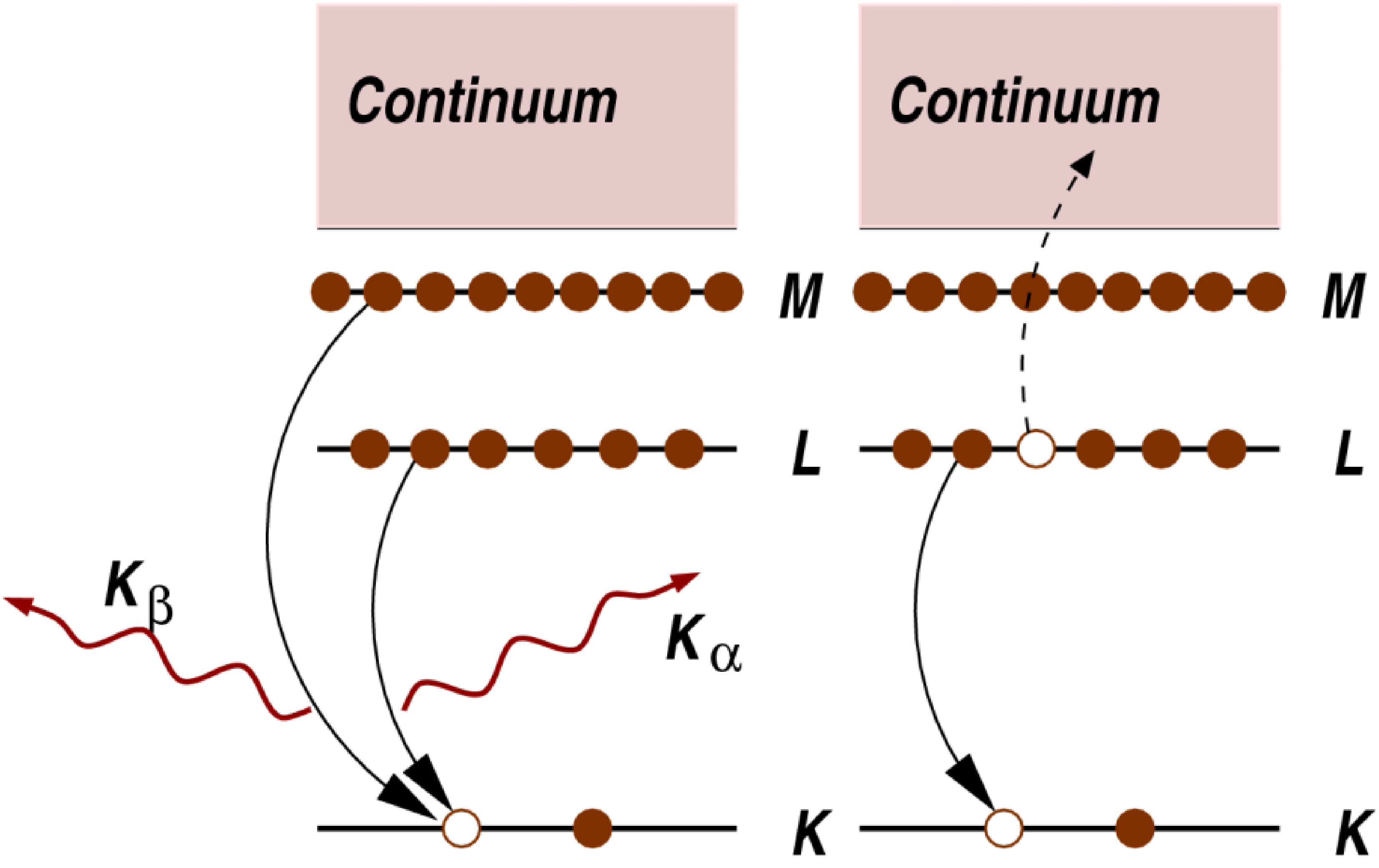
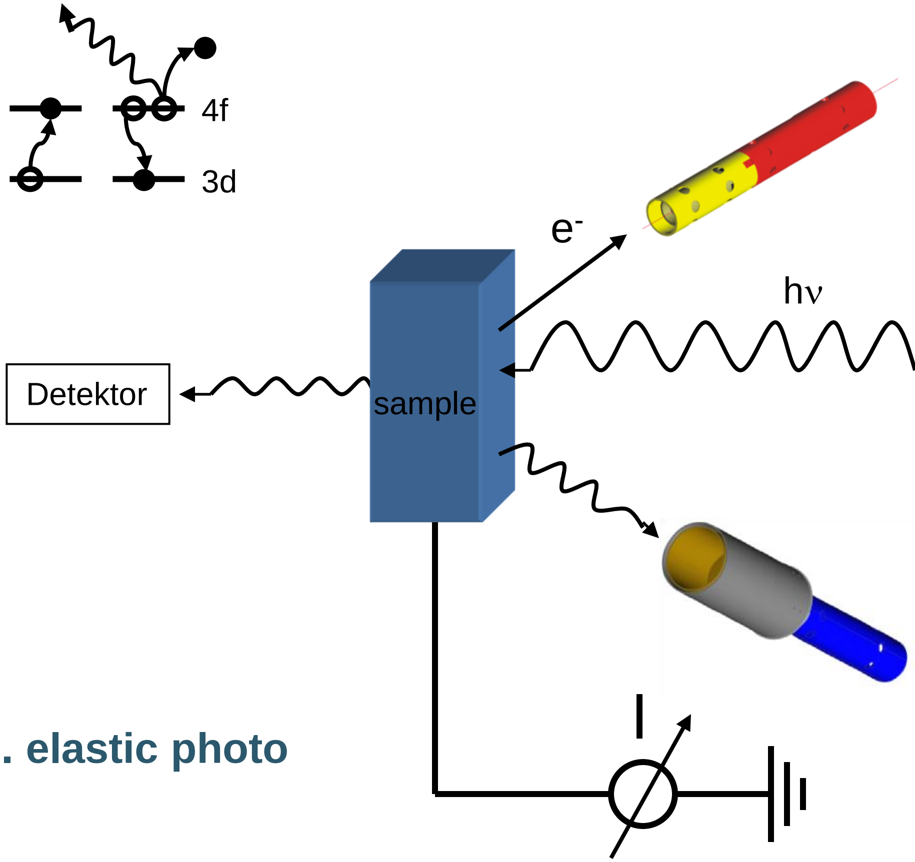


Figure 2.5: Decay of the excited state: x-ray fluorescence (left) and the Auger effect (right). In both cases, the probability of emission (x-ray or electron) is directly proportional to the absorption probability.

XAS experimentally



- Transmission mode: $I(h\nu)=I_0\, e^{-\mu z}$
- **Fluorescence Yield** **(bulk sensitive, but often saturation problems)**
- **Total Yield (TY):**

  All (in-) elastic photoelectrons
  ✸Probing depth: 40Å to 100Å
  ✸good signal to noise ratio (I~100 pA)
- **Partial Electron Yield (PEY):**

  only photo electrons with $E_{kin} \geq E_{threshold}$, i.e. elastic photo electrons (ca. 5% of TY-signal)
  ✸probing depth: ~15Å (surface)

All methods can be measured simultaniously to get more information

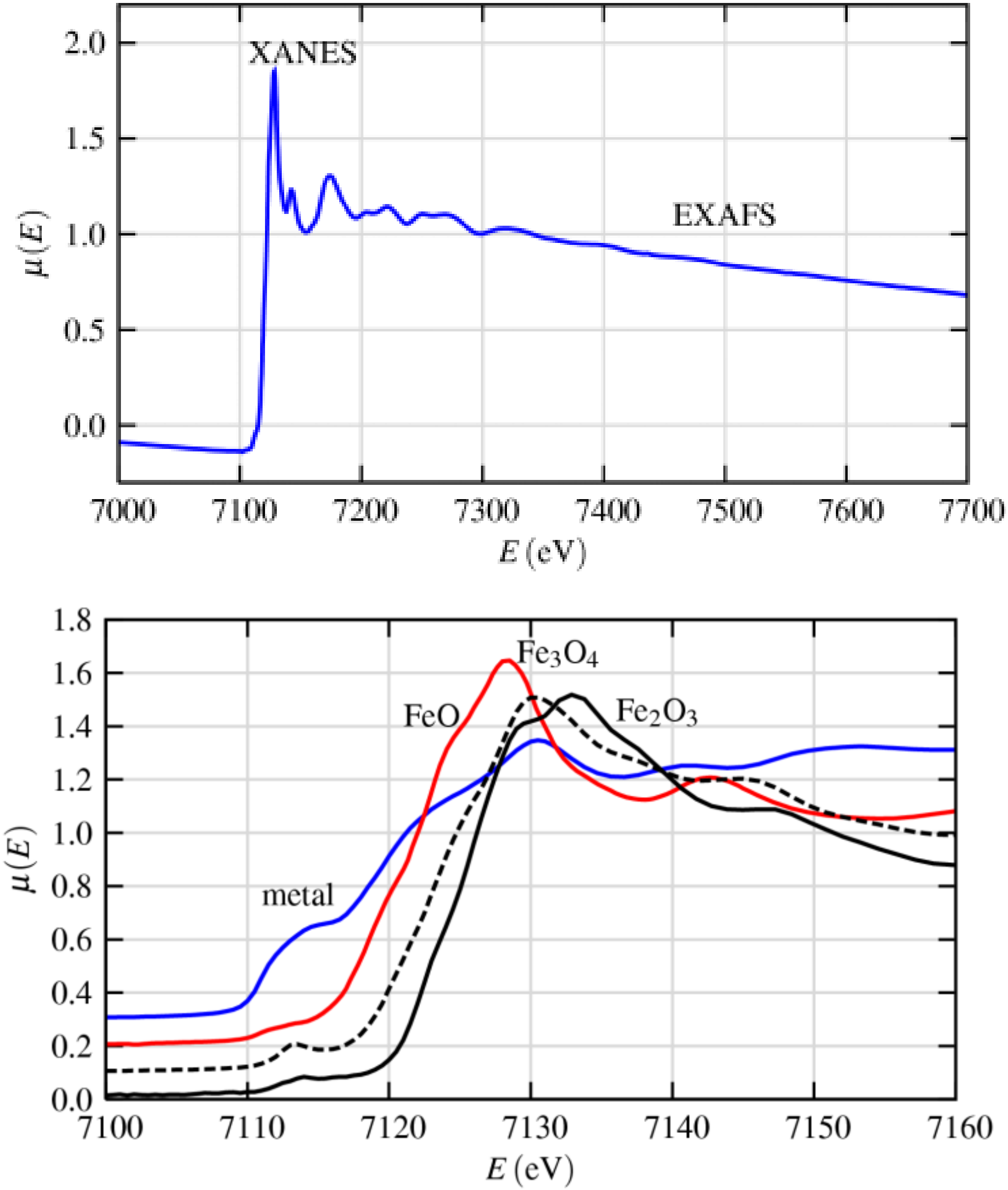
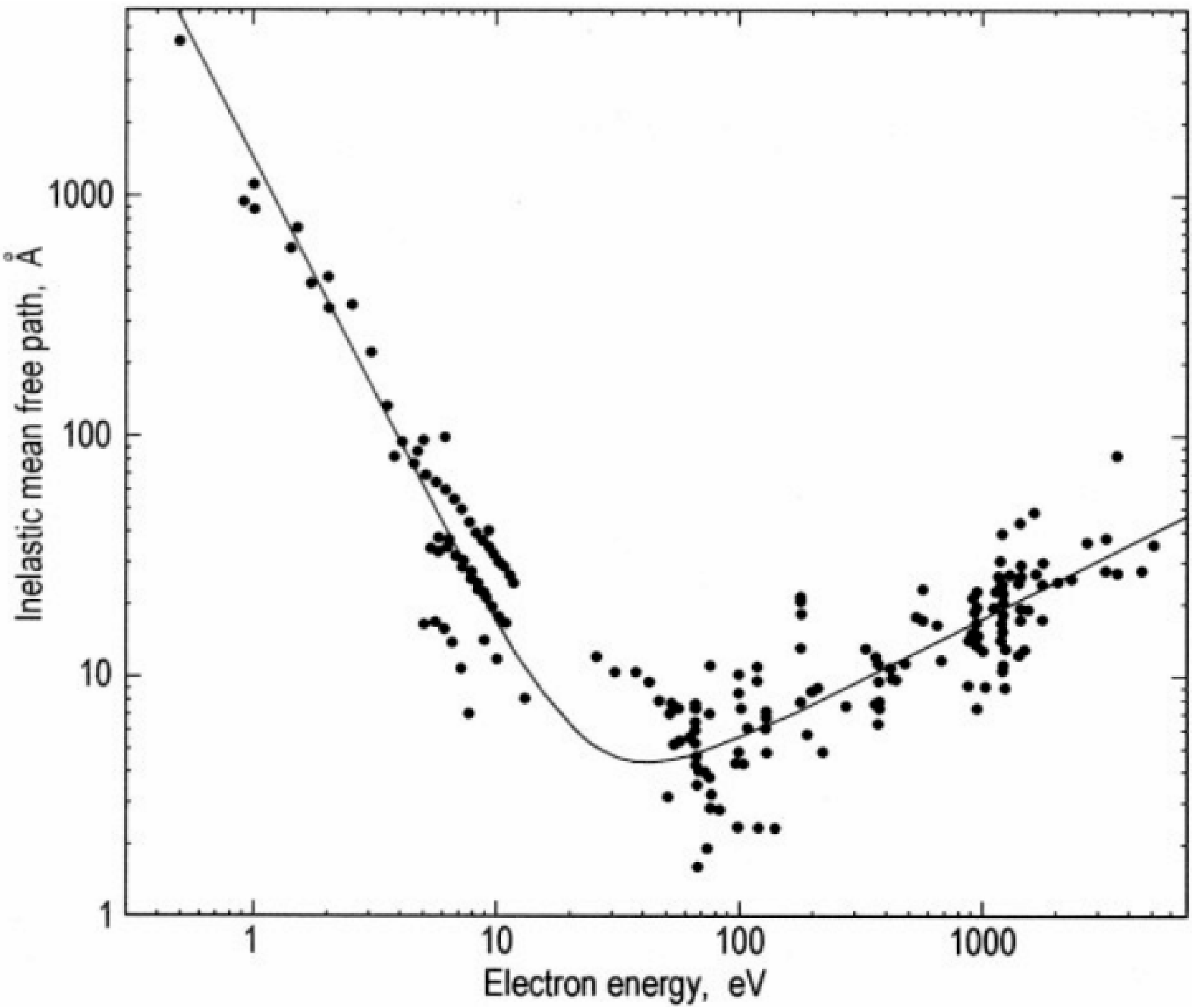
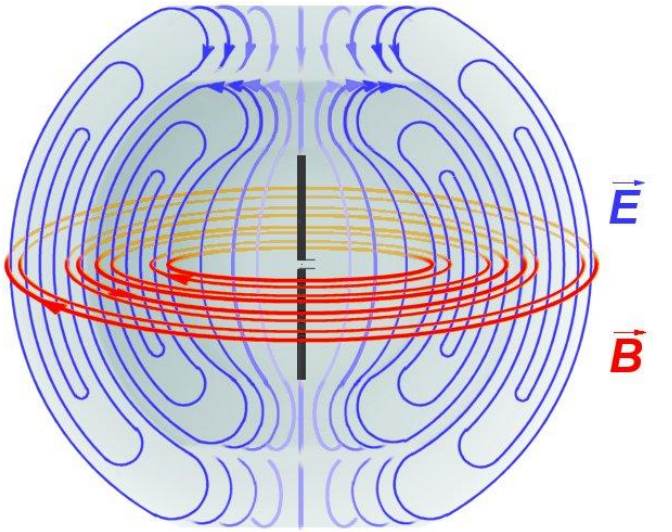


Figure 2.6: XAFS $\mu(E)$ for FeO. On top, the measured XAFS spectrum is shown with the XANES and EXAFS regions identified. On the bottom, $\mu(E)$

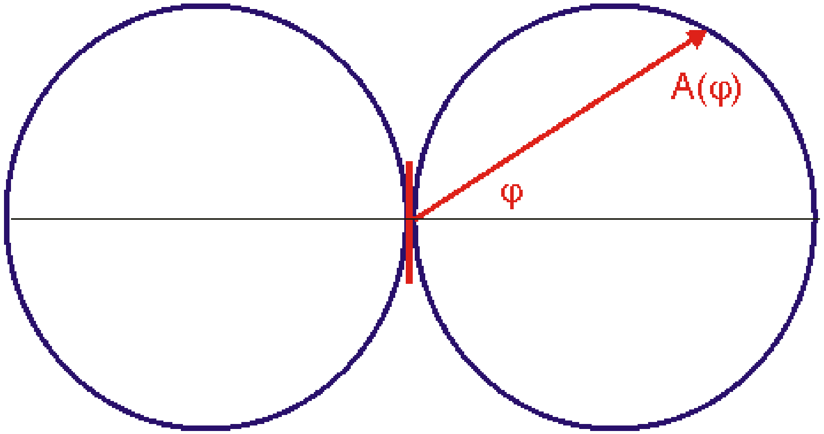
The Universal Curve for the Electron Mean Free Path



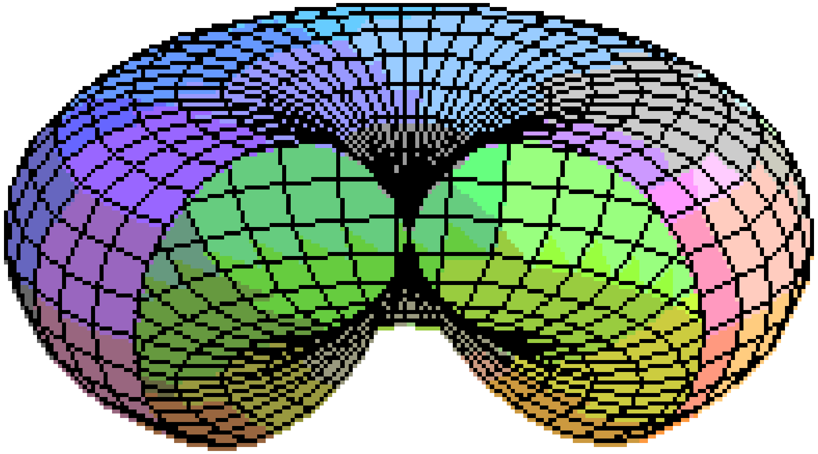
Dipole radiation



Dipole antenna (harmonic oscillation of charge) with induced E- and B-field



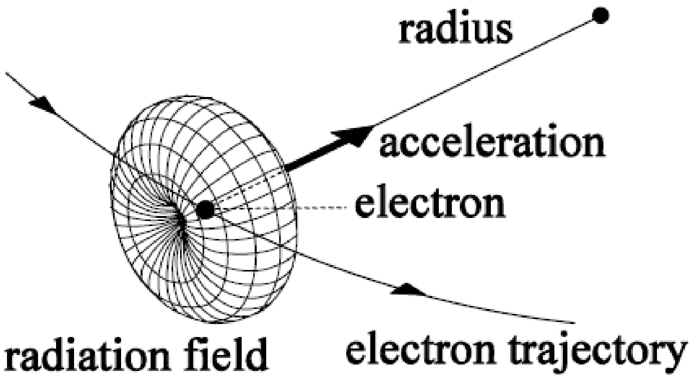
Emission characteristics (A=intensity)



3D-view

Electrons on circular orbit

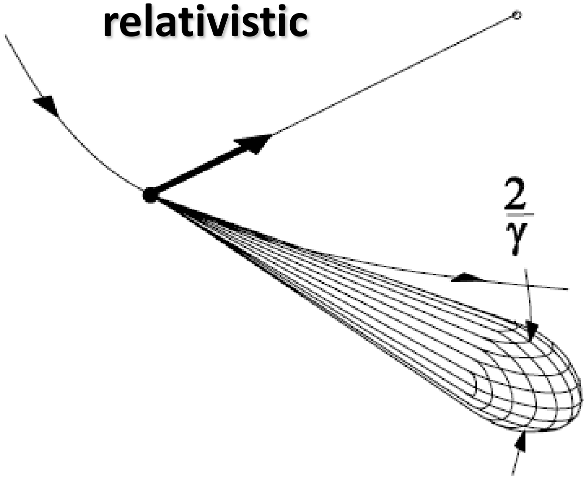
nonrelativistic



Radiation Power *P*

$$P = \frac{2}{3}\frac{e^2 c}{R^2}\beta^4 \underbrace{\left(\frac{E}{m_0 c^2}\right)}_{\gamma}{}^4$$

relativistic



opening angle *θ*

nonrelativistic:

$\rightarrow v << c \rightarrow \beta << 1$

⇒ Radiation power is very small and emitted in all directions

E = particle energy
R = radius of curvature
m_0 = particle mass

$$\beta = \frac{v}{c};\ \gamma = \frac{E}{m_0 c^2}$$

Relativistic:

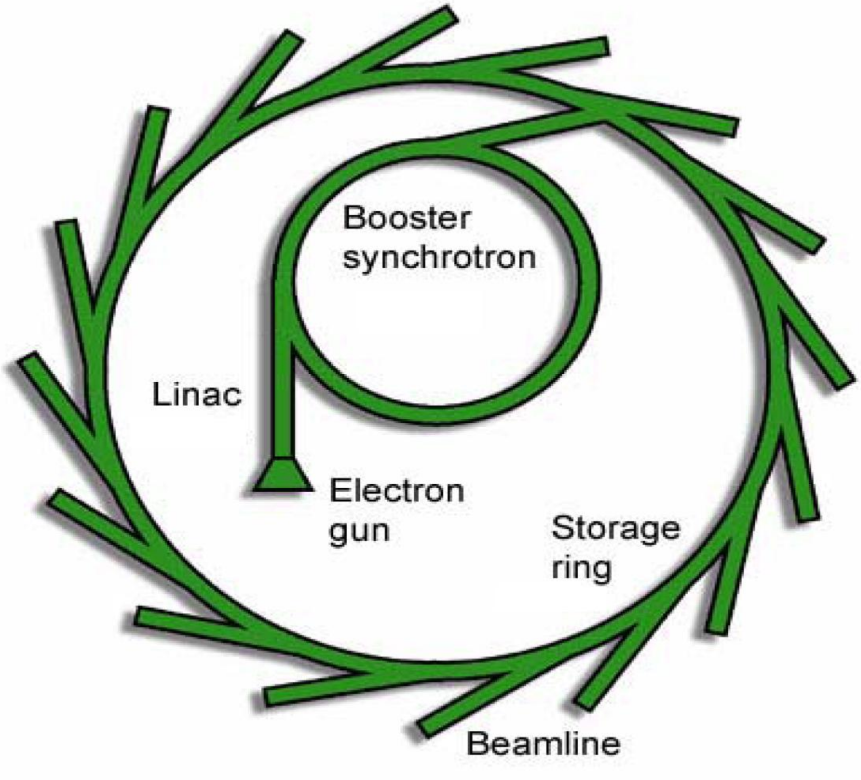
$\rightarrow v \approx c \rightarrow \beta \approx 1$

$$P = \frac{2}{3}\frac{e^2 c}{R^2}\gamma^4$$

⇒ extremely high radiation power, emitted in a sharp forward cone!

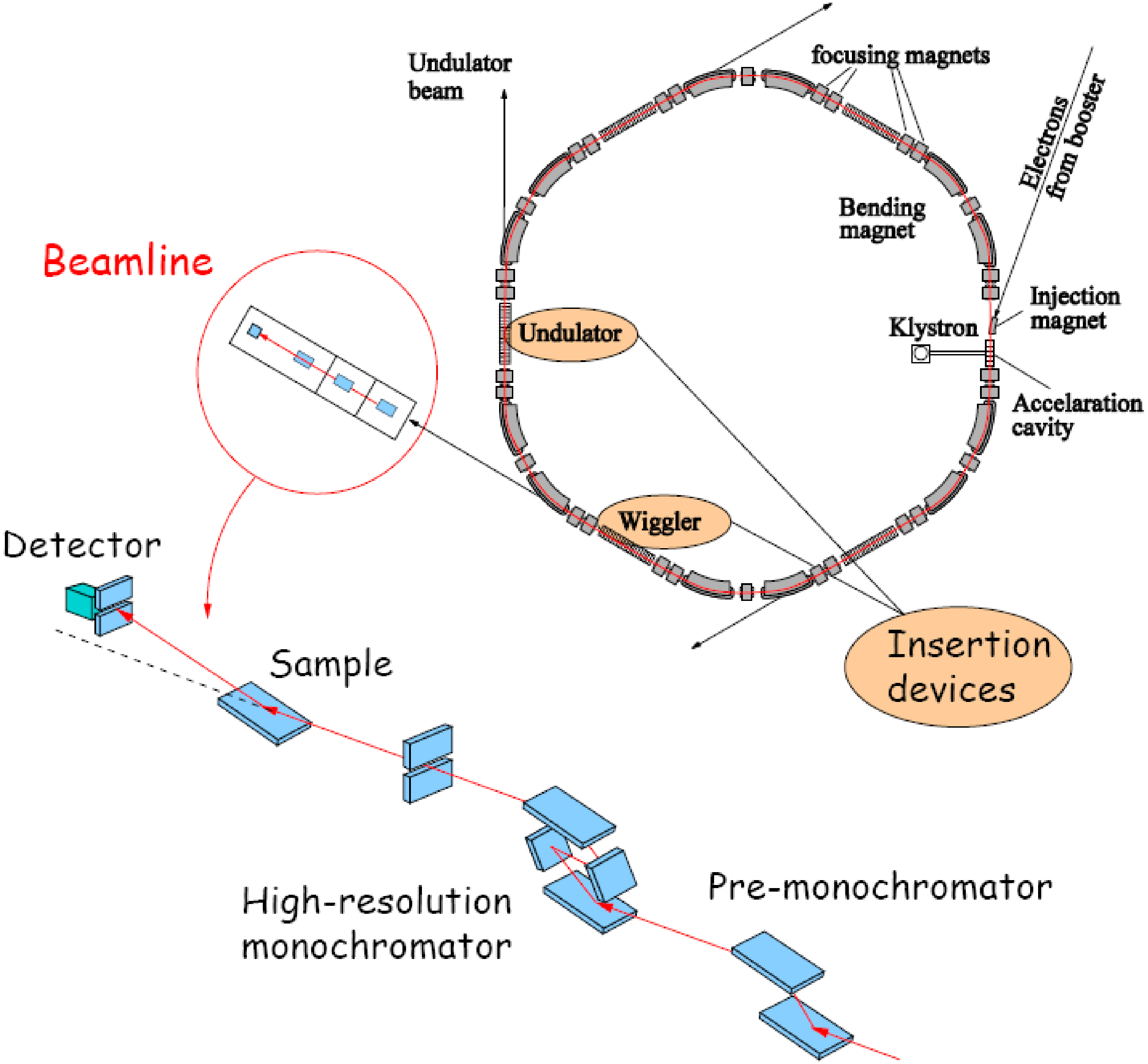
Generation of Synchrotron Radiation

1. emission of electrons by an electron gun
2. acceleration in a linear accelerator (LINAC)
3. transmission to a circular accelerator (booster synchrotron) to reach the required energy level (e.g. E = 6 GeV at ESRF)
   → relativistic electrons
4. injection of high energy electrons into a large storage ring (circumfence e.g. 844 m at ESRF) where they circulate in vacuum at a constant energy for many hours



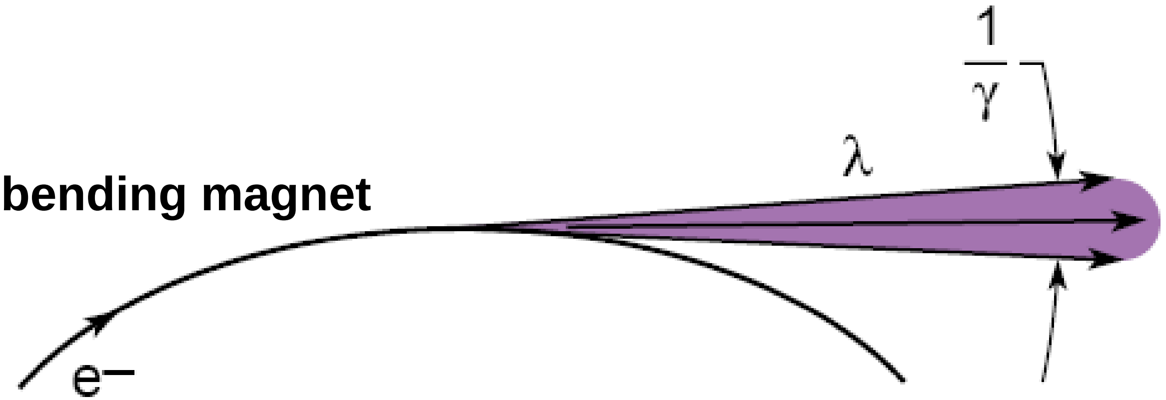
Velocity of relativistic electrons (6 GeV) *v* is only 107 cm/s slower than the velocity of light

Storage rings and beamlines



Angular distribution (relativistic)

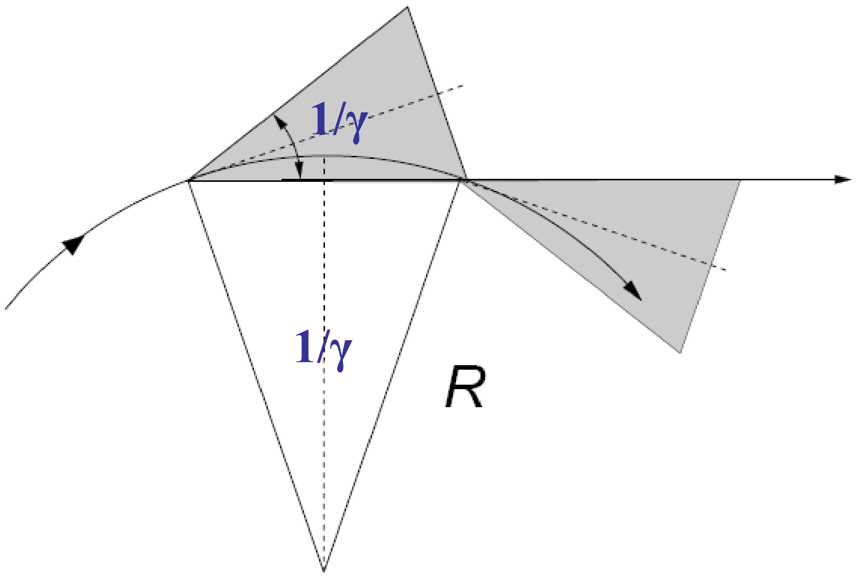
Radiation from bending magnet



<u>Example:</u>
E = 6 GeV, v is only 107 cm/s slower than the velocity of light ($c \cong 3 \times 10^{10}$ cm/s)
$\gamma = E/mc^2 \cong 1820$
$\theta \approx 8 \times 10^{-5}$ rad (0.08 mrad)
The emitted radiation is a sharp cone with an opening angle $\theta \approx 0.08$ mrad

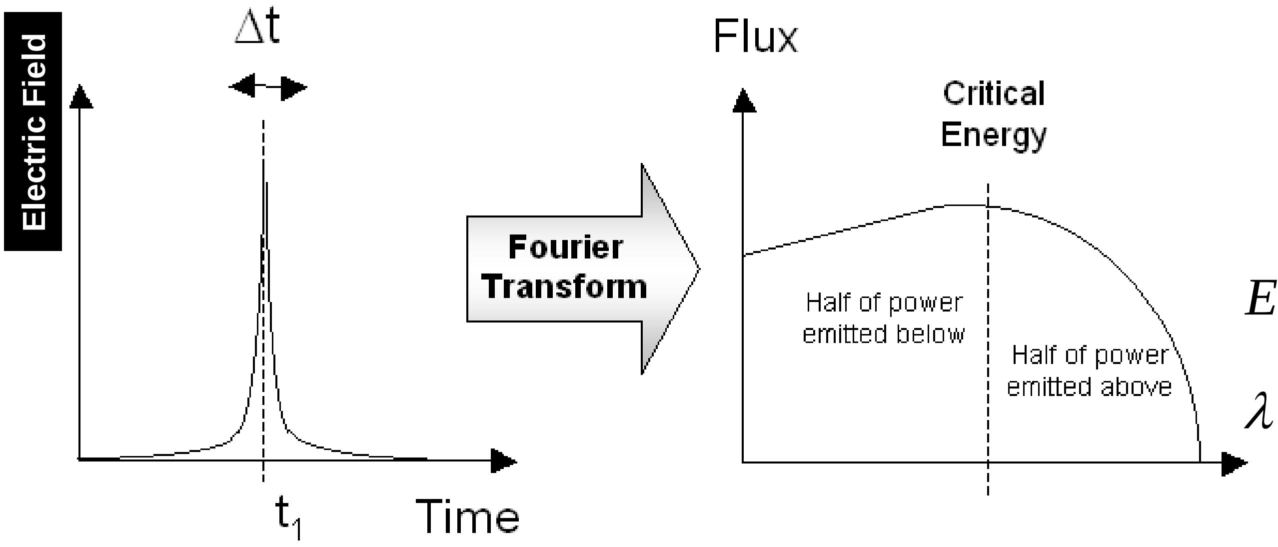
⇒ Excellent collimation!
⇒ in a distance of 50 m from the source, one obtains a spot of only ~ 4 mm!

Pulse duration and energy spectrum



Duration of radiation flash (single electron):

$$\Delta t = \frac{4R}{3c\gamma^{3}}$$



$$E_c(keV) = 0.665\,B(T)\,E(GeV)^{2}$$

$$\lambda_c = 5.59 \cdot \frac{R}{E^{3}}$$

broad energy spectrum!

Charaterize the properties of a Synchrotron Radiation source

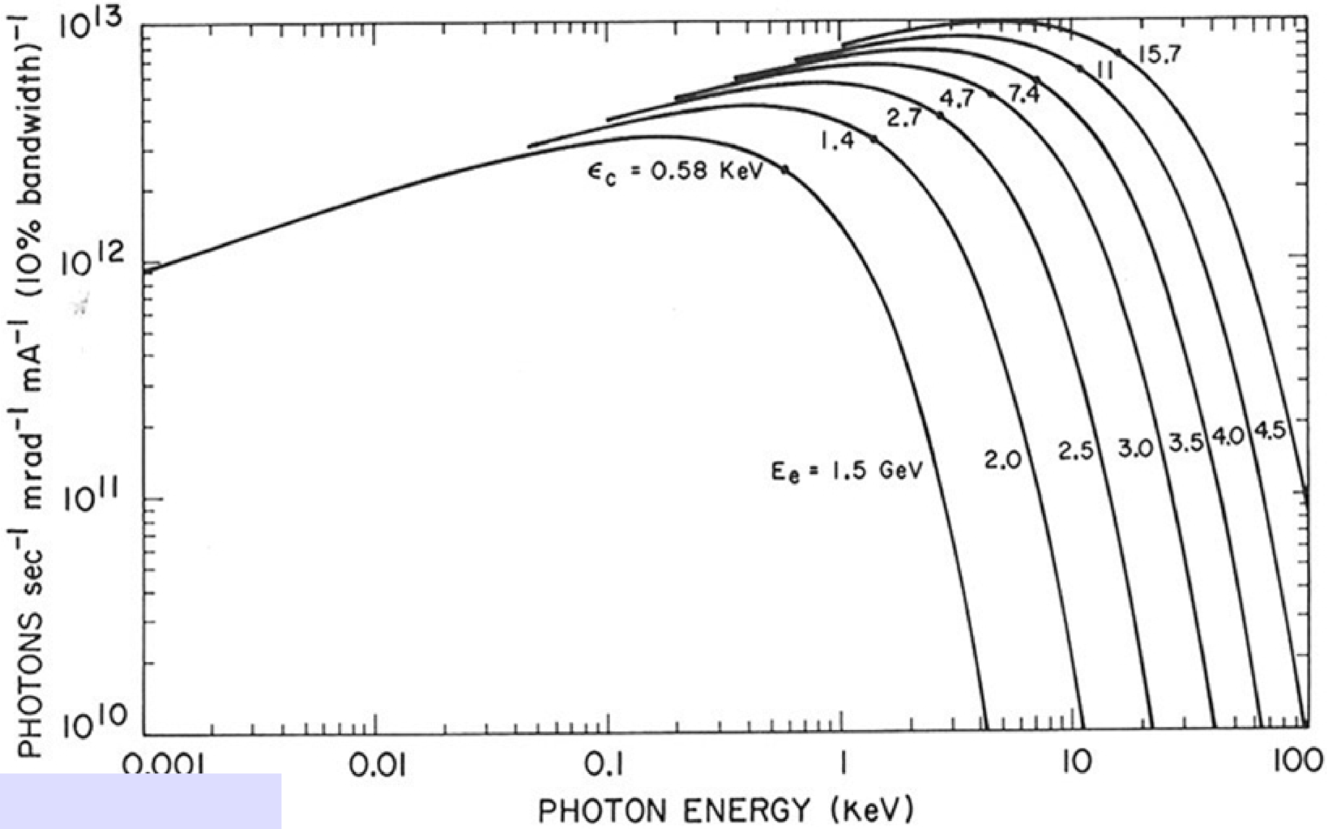
$$\text{Total flux} \equiv \frac{\text{Photons}}{\text{s}}$$

$$\text{Spectral flux} = \frac{\text{Total flux}}{0.1\%\text{bandwidth}} \quad \left[\frac{\text{Photons/s}}{0.1\%\text{bandwidth}}\right]$$

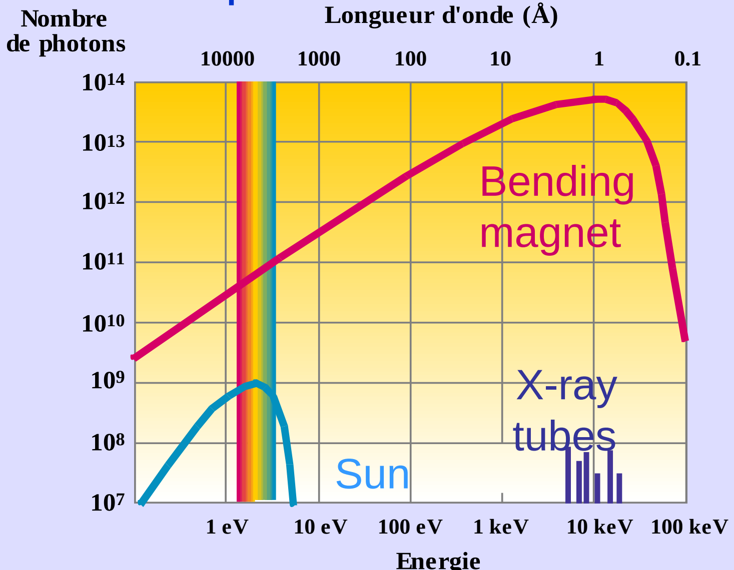
$$\text{Brightness} = \frac{\text{Total flux}}{\text{solid angle} \cdot 0.1\%\text{bandwidth}} \quad \left[\frac{\text{Photons/s}}{\text{mrad}^2 \cdot 0.1\%\text{bandwidth}}\right]$$

$$\boxed{\text{Brilliance} = \frac{\text{Total flux}}{\text{solid angle} \cdot \text{source area} \cdot 0.1\%\text{bandwidth}} \quad \left[\frac{\text{Photons/s}}{\text{mrad}^2 \cdot \text{mm}^2 \cdot 0.1\%\text{bandwidth}}\right]}$$

Brilliance is the figure of merit for the design of new Synchrotron Radiation sources

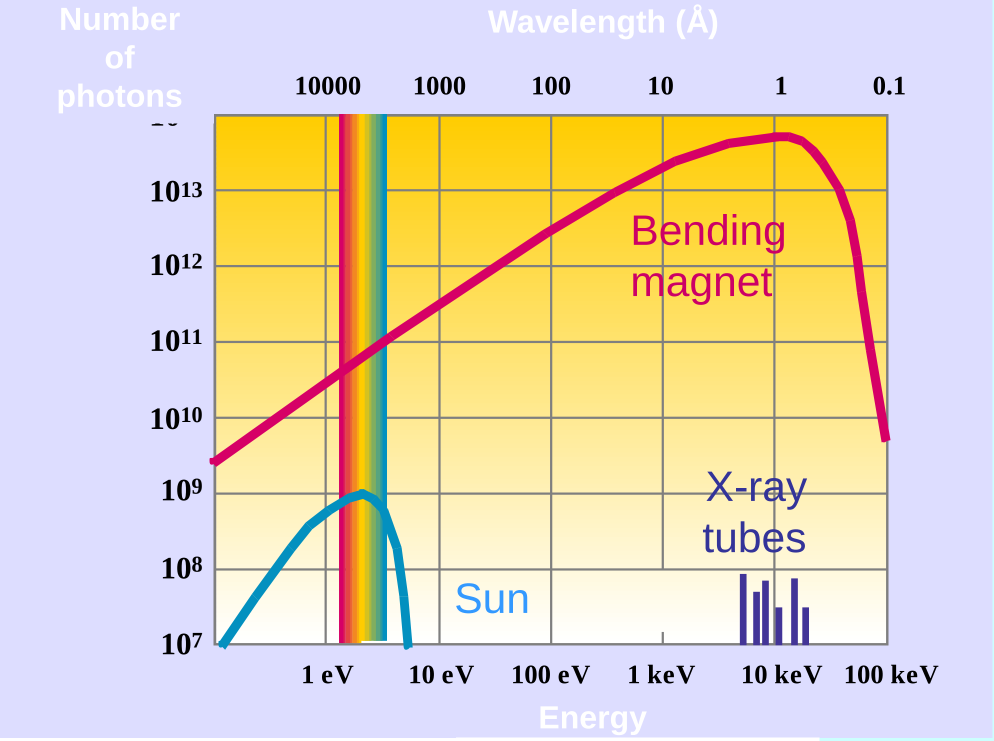


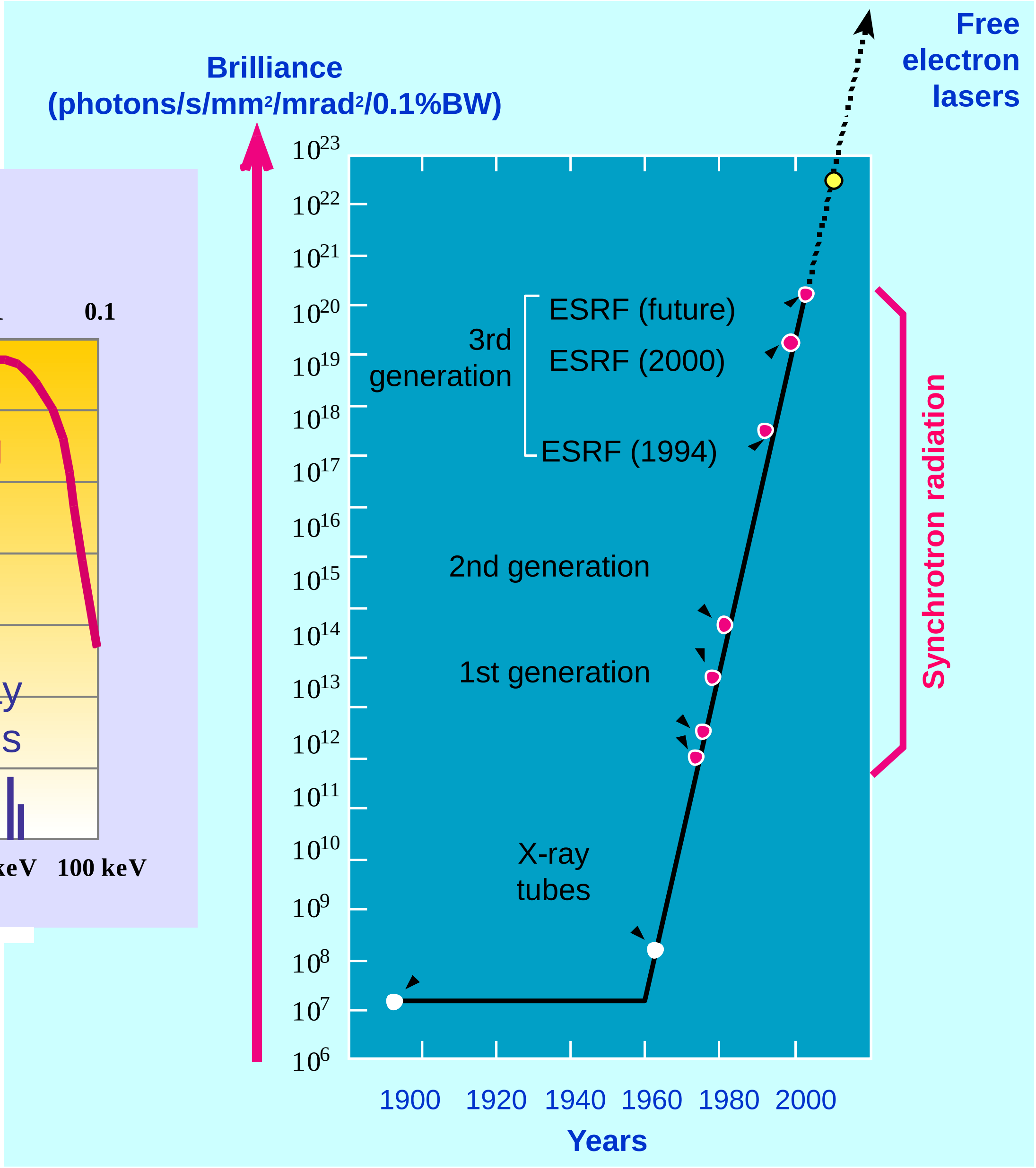
Emission spectrum



Extremely high intensity, broad energy range

Emission spectrum

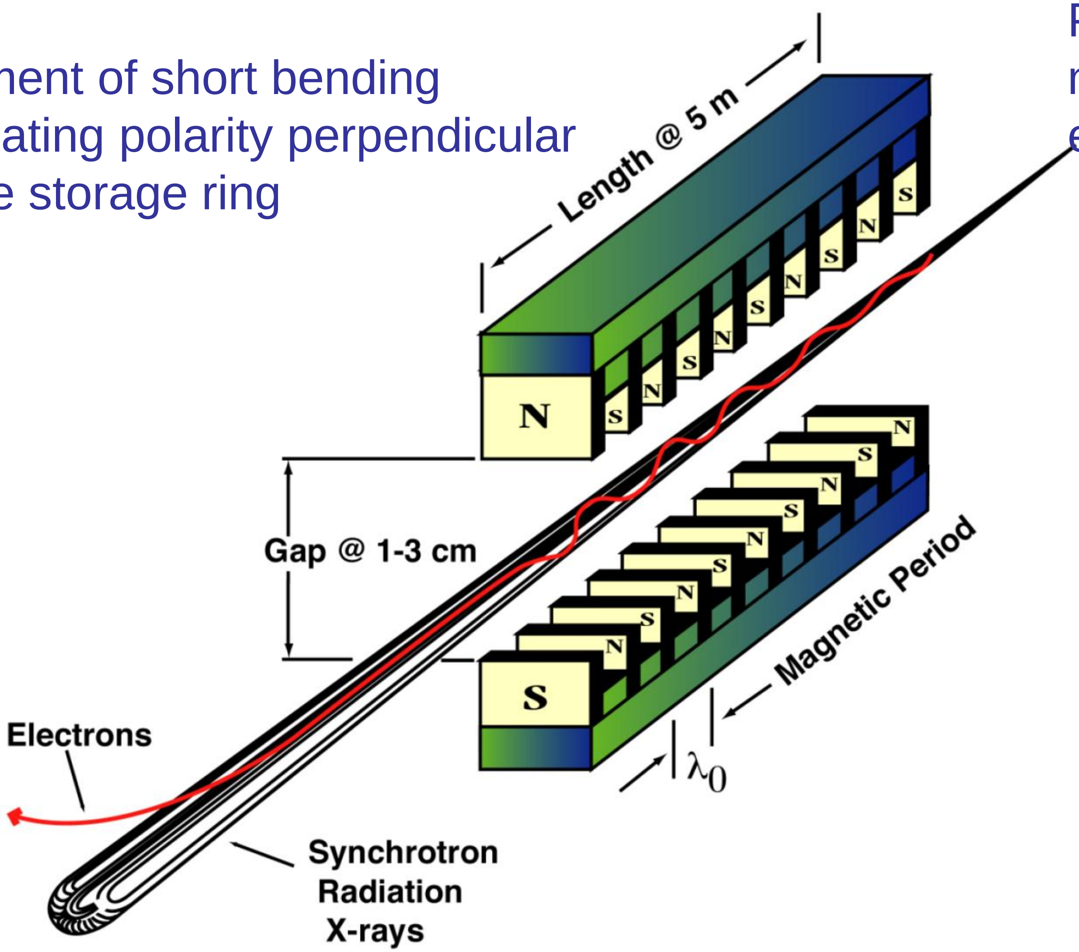




Magnetic wigglers and undulator (N periods)

Principle:
periodic arrangement of short bending magnets of alternating polarity perpendicular to the plane of the storage ring

Permanent magnetic materials e.g. Nd-Fe-B



⇒ force the electrons to oscillate („wiggle“) perpendicular to their direction of motion

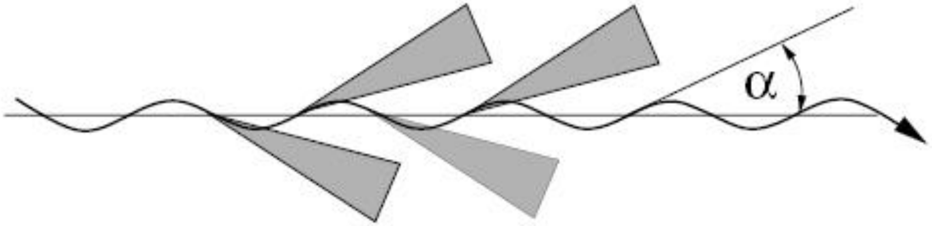
⇒ Radiation is emitted during each individual wiggle

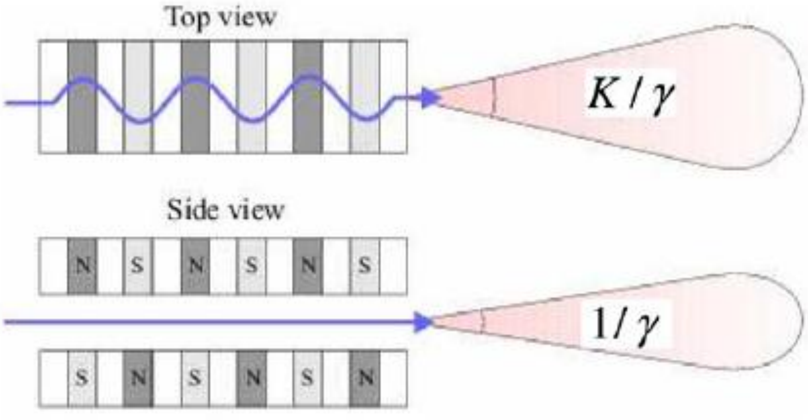
⇒ increase of the intensity

wiggler and undulator

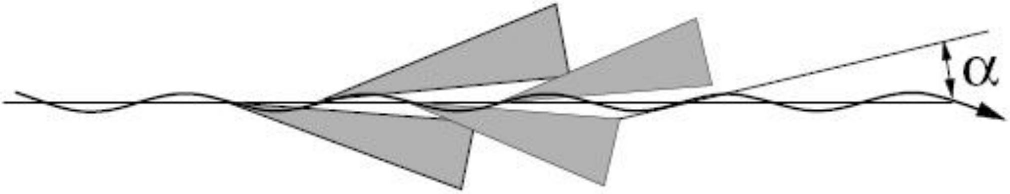
$$K := \alpha \cdot \gamma = \frac{e\, B_0\, \lambda_0}{2\pi\, m_e\, c}$$

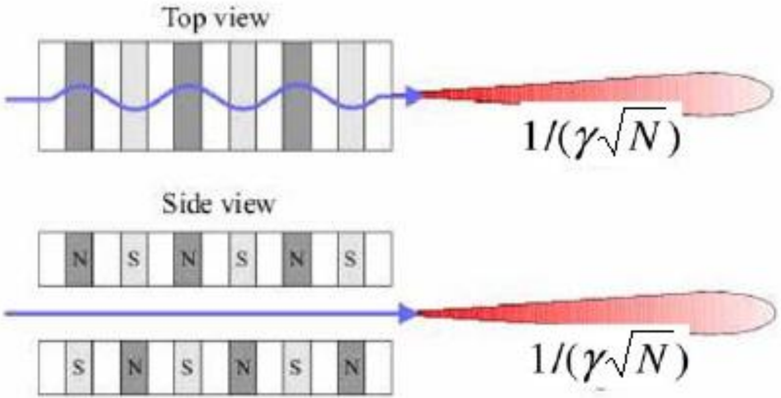
Wiggler regime: α > 1/γ





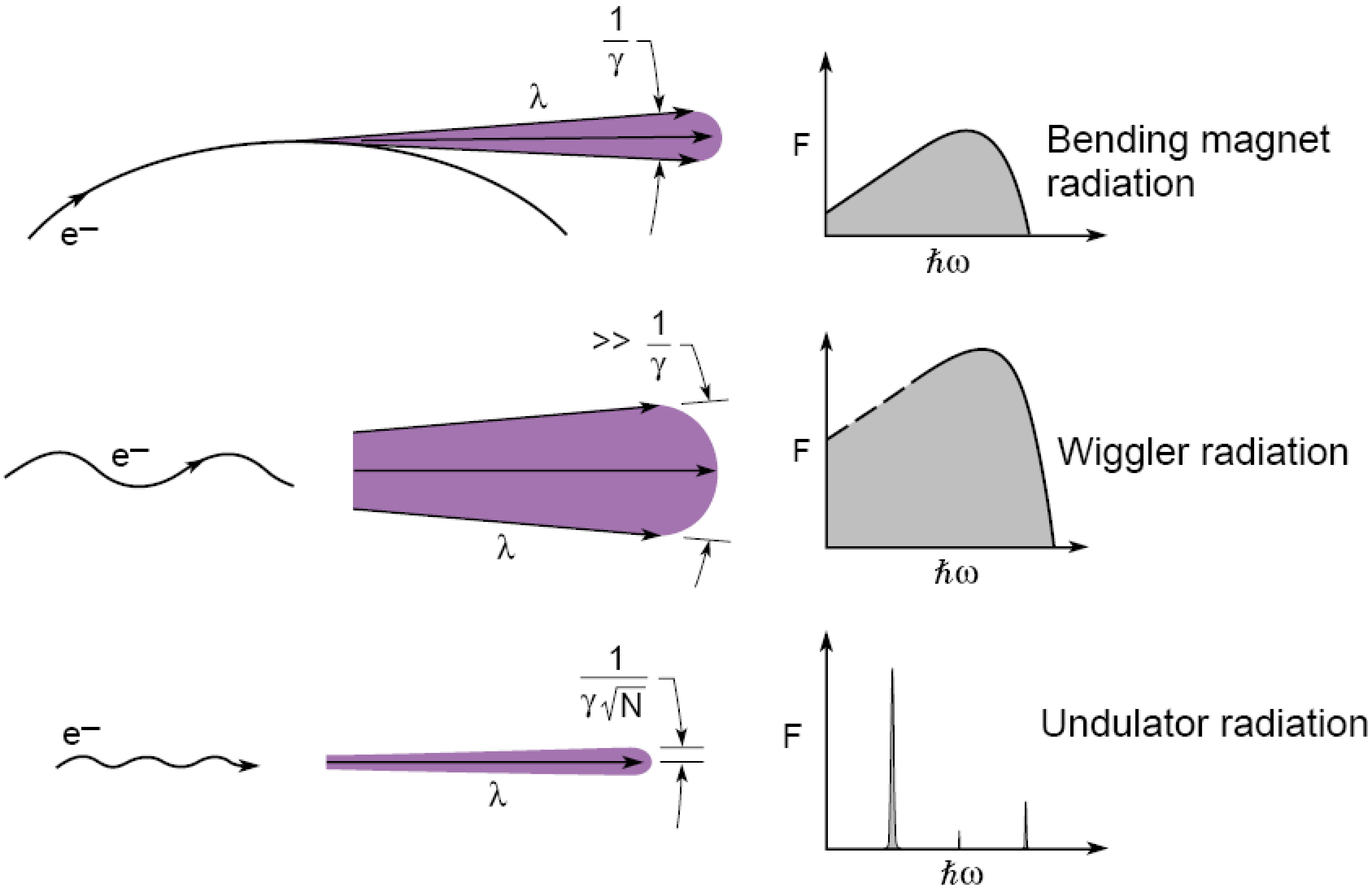
Undulator regime: α ~ 1/γ



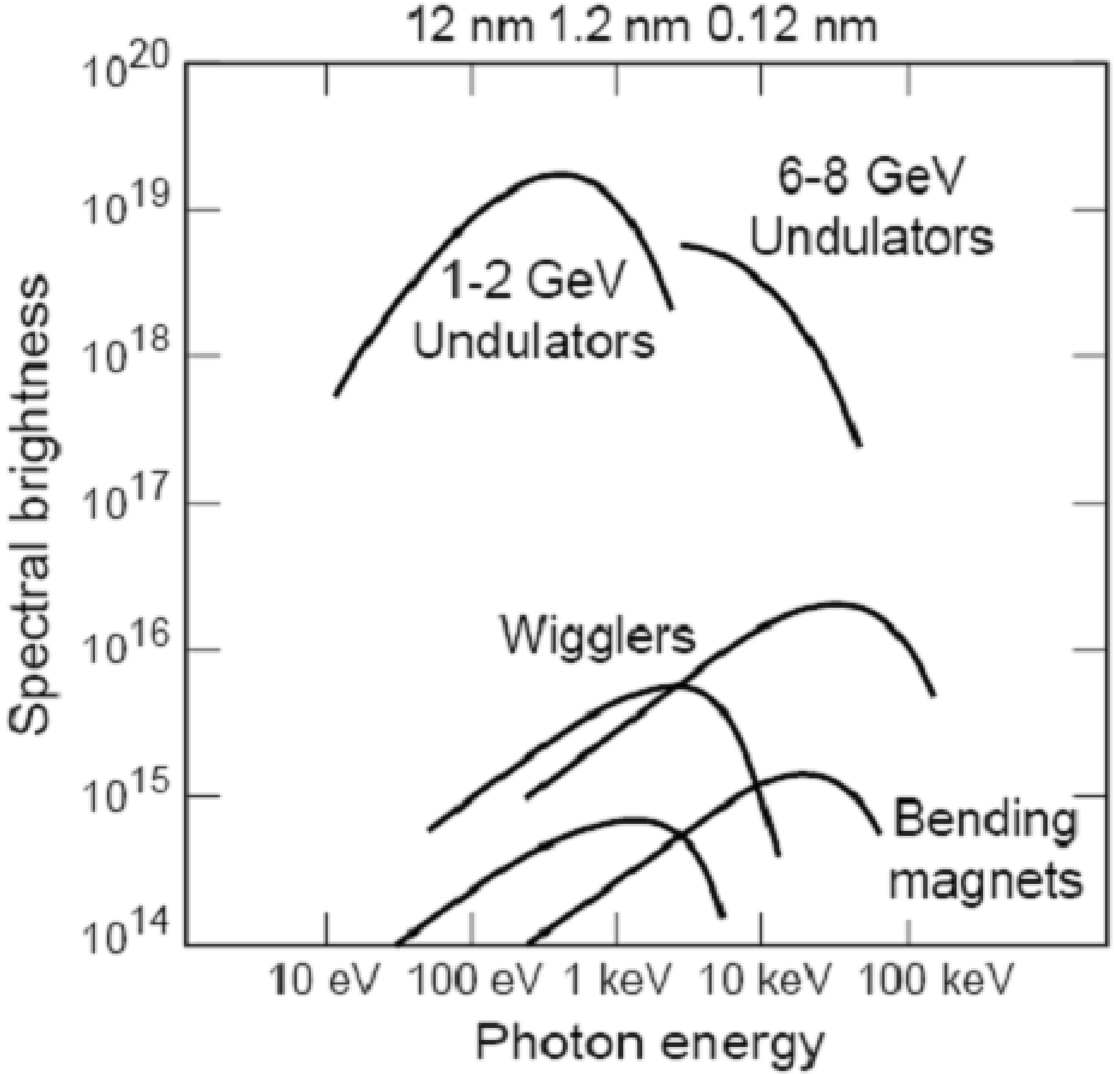


In the undulator regime the radiation cones overlap and the wave trains can interfere

Forms of Synchrotron Radiation

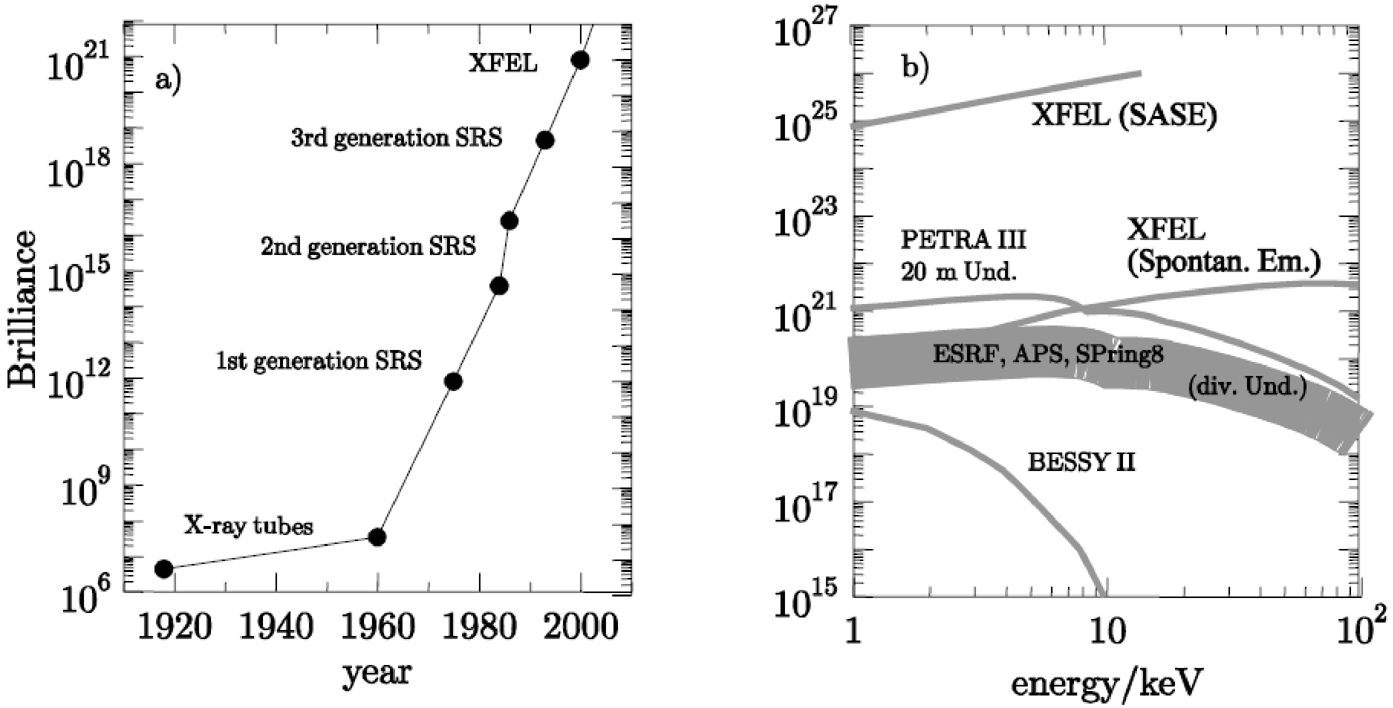


Spectral Brightness



Examples of Wigglers and Undulators

Evolution of Brilliance



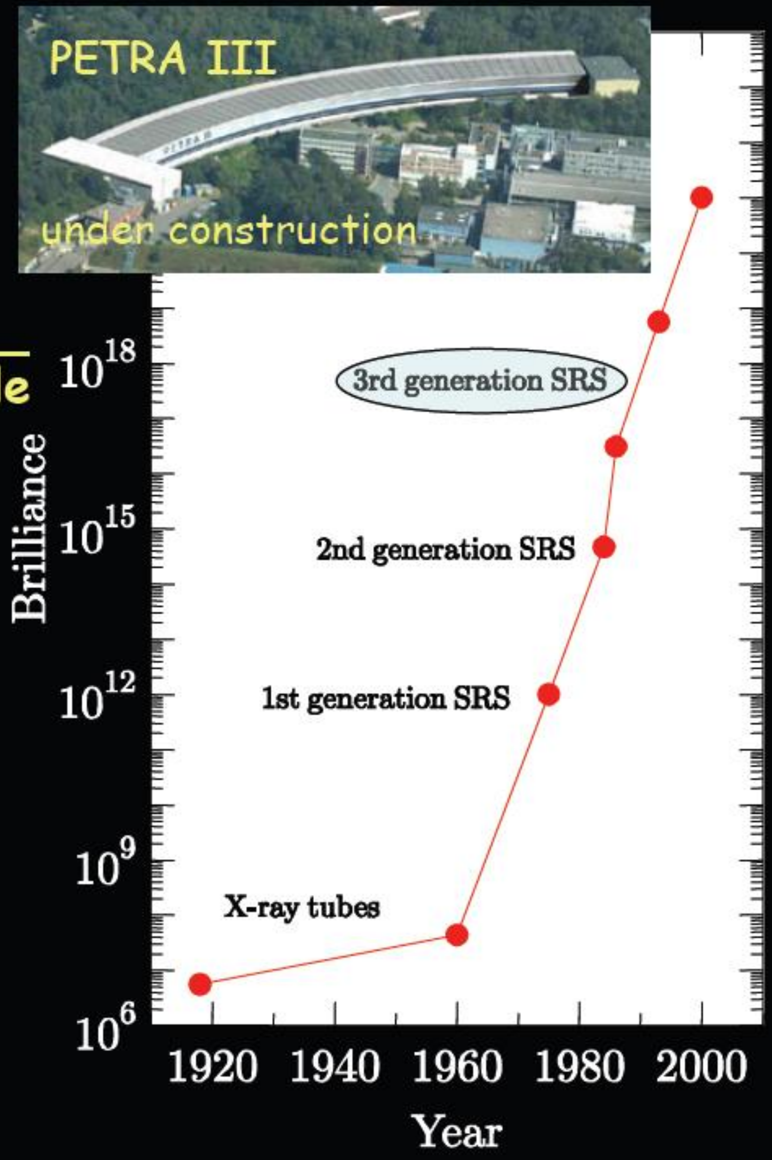
1st generation: Exploitation of the light from the bending magnets of e+/e- colliders originally built for elementary particle physics

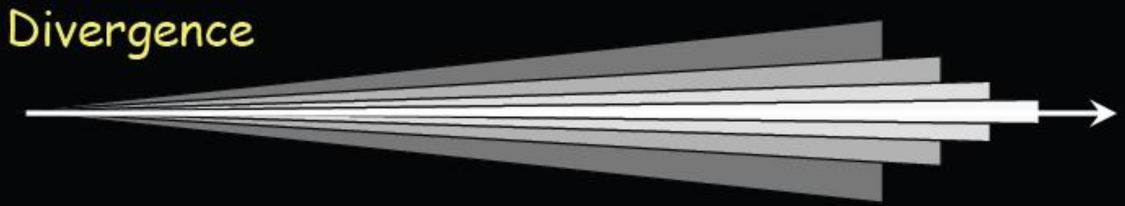
2nd generation: Radiation from bending magnets and introduction of first insertion devices, lower e-beam emittance, optimization of light extraction

3rd generation: dedicated storage rings, very low e-beam emittance, brilliance is figure of merit, mainly undulators, long straight sections

Evolution of Source Brilliance

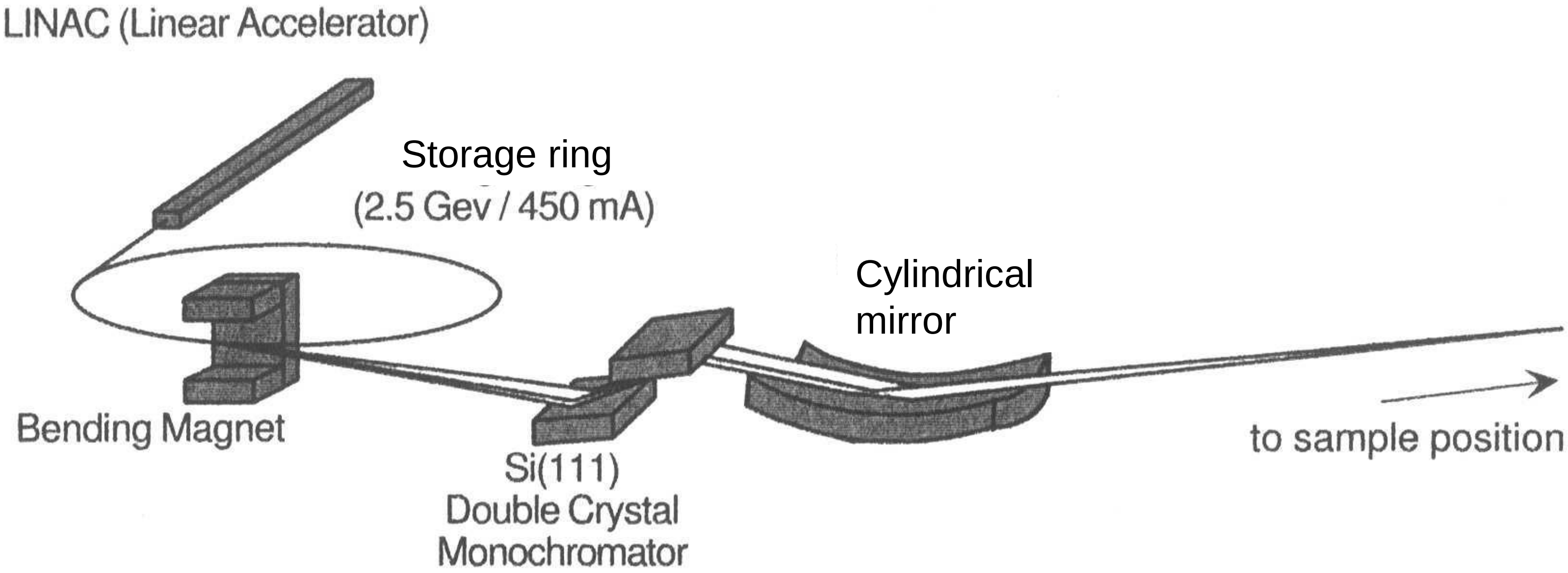
$$\text{Brilliance} = \frac{\text{Spectral flux}}{\text{source area} \times \text{solid angle}}$$



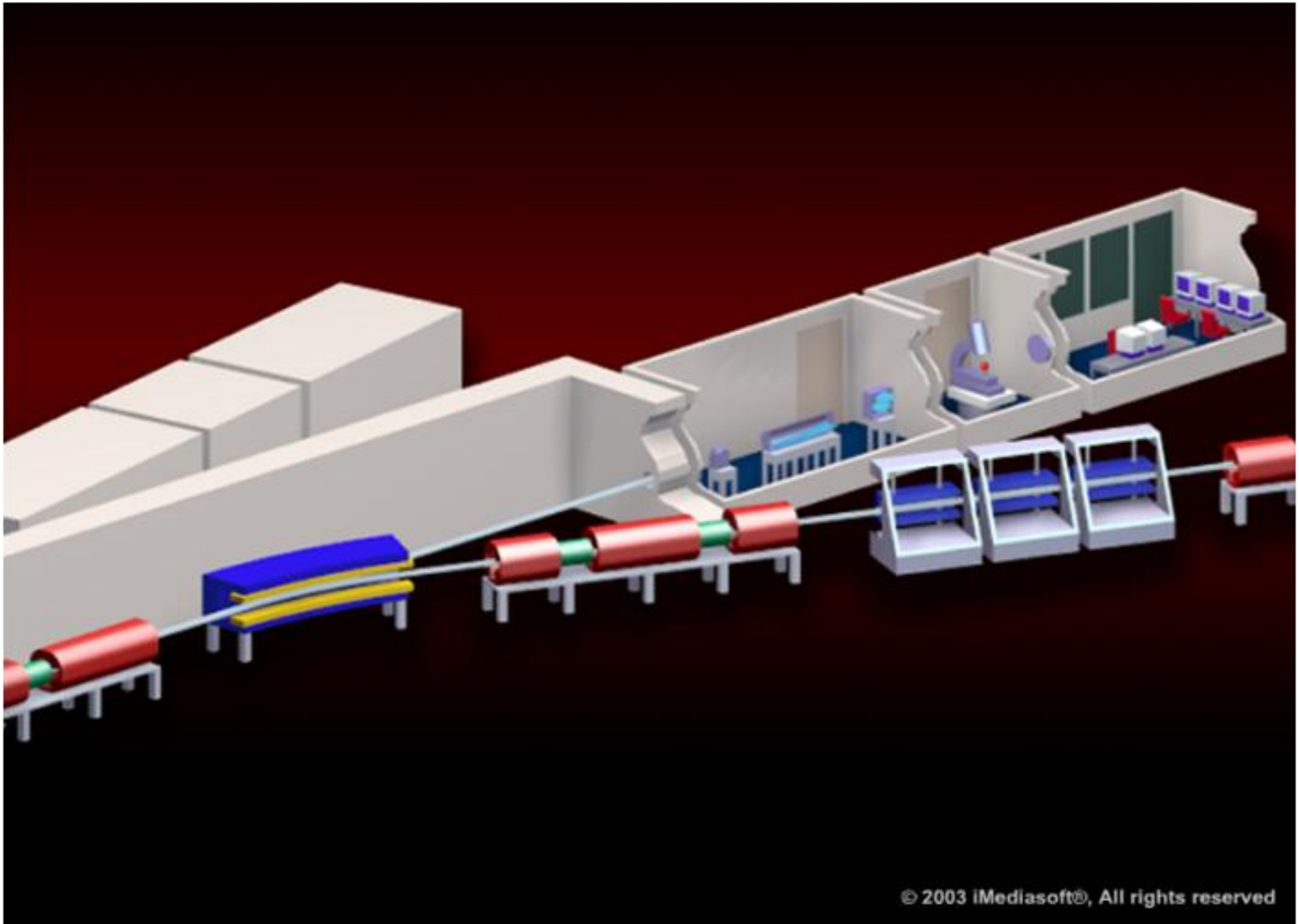


European Synchrotron Radiation Facility (ESRF)

Beamline organization



How does a beamline work?

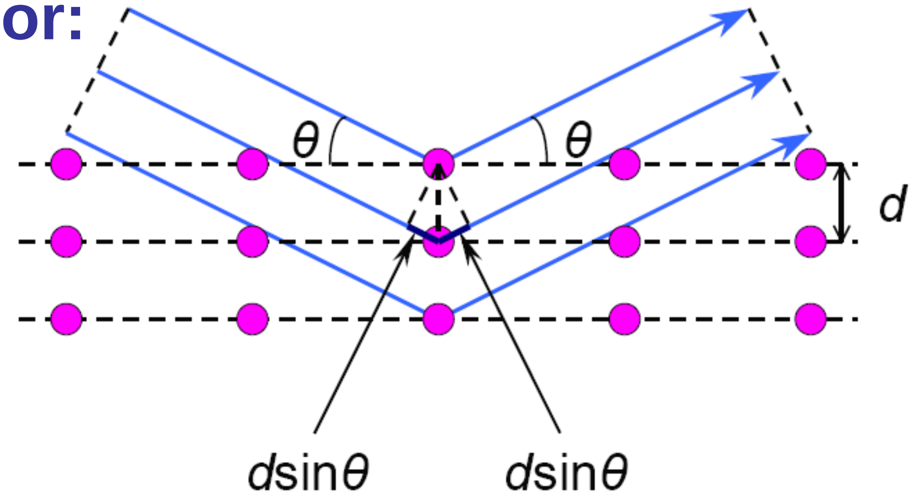


X-ray monochromator using perfect crystal

Basic principle of monochromator:

Bragg reflection from perfect single crystal

$$\boxed{2d_{hkl}\sin\theta = n\lambda}$$



Energy range of standard monochromator

Bragg Reflection

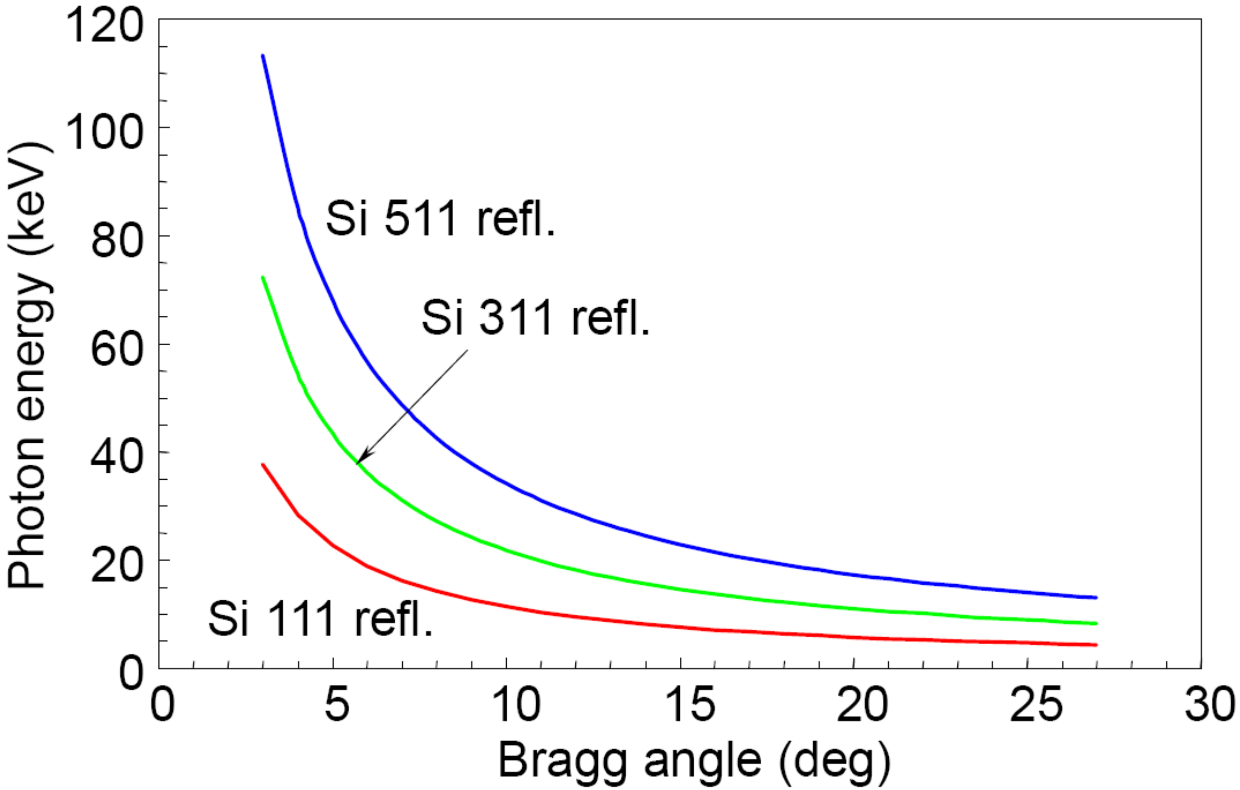
Si 111
Si 311
Si 511
.....

Bragg angles

3~27°

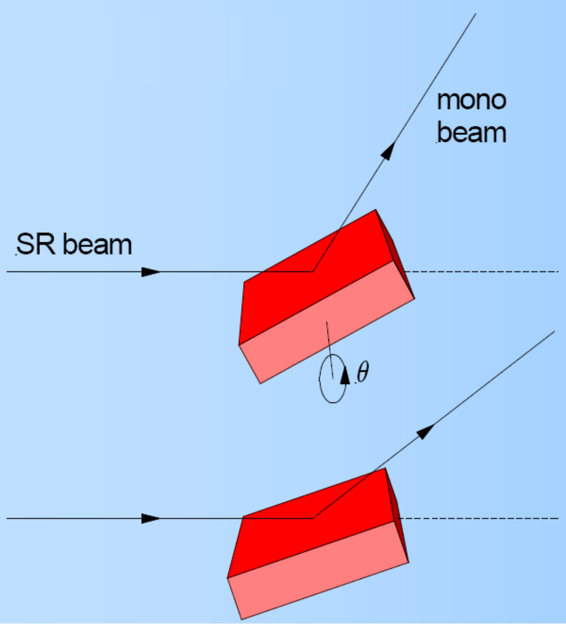
Energy range

4.4~110 keV



Photon energy (wavelength) can be selected by crystal, net planes, and Bragg angle.

Double crystal monochromator

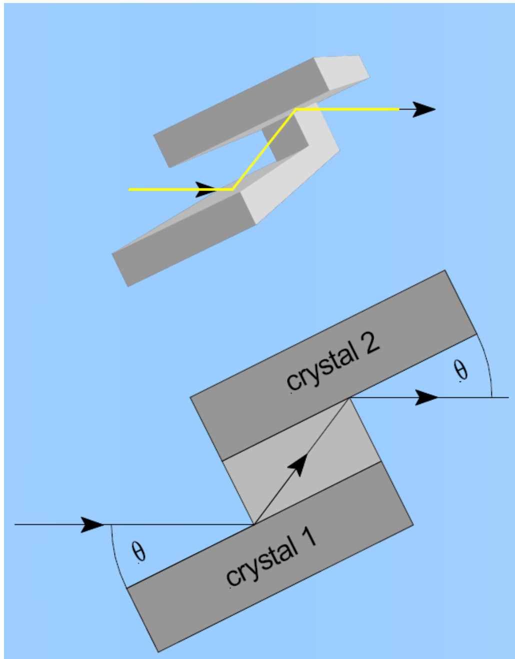


Problems with single crystal monochromators

- the monochromatic beam moves when the energy is changed
- high harmonic content
- big tails

Solution: double crystal design!
Simplest design: cutting a channel for the beam in a silicon block (**channel cut monochromator**)

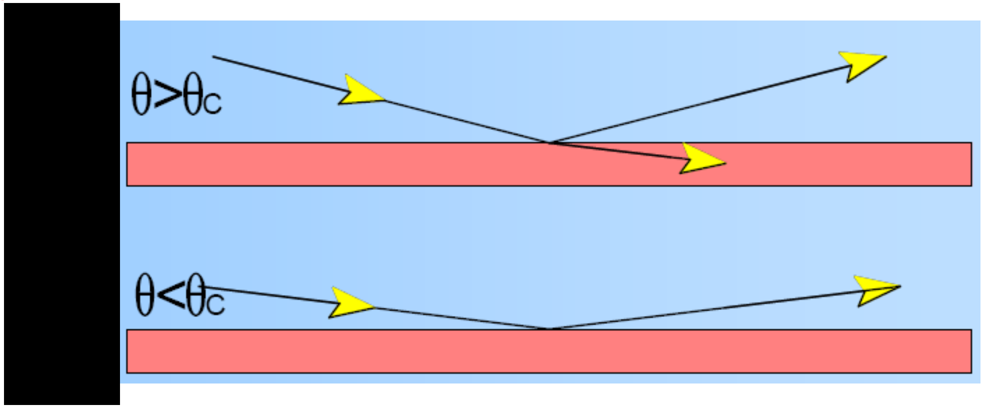
- **Use the same crystals and *d*-spacing for 1st and 2nd crystals**
- **Keep parallel setting**



X-ray Mirrors

reflectivity at grazing angles:

refractive index: $n = 1 - r_0 \rho \lambda^2 / 2\pi - i \mu\lambda / 4\pi$

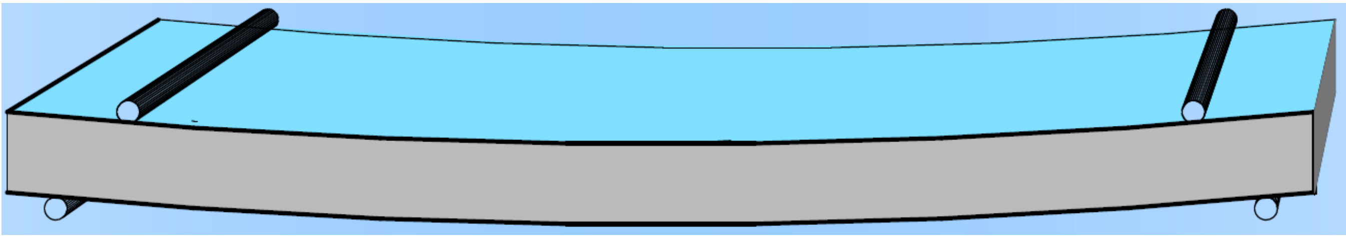


By Snell's law ($n_1 \cos(\theta_1) = n_2 \cos(\theta_2)$ with θ the grazing angle) in the absence of absorption (total reflection), we find total external reflection for angles less than $\theta_c \approx \lambda(r_0 \rho / \pi)^{1/2}$

θ_c typically a few mrad for x-ray mirrors

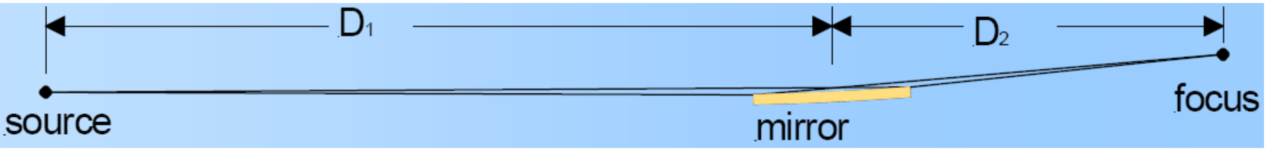
Surface roughness must be considered around critical energy (angle).

Bent mirrors (focusing and collimating)



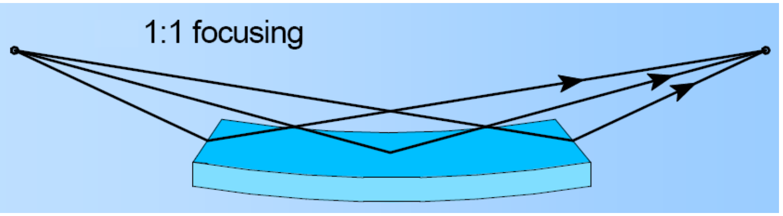
Focusing of the x-ray beam → reflecting surface must have some curvature (achieved e.g. by bending mirror, **mirror focuses in one plane only**!)

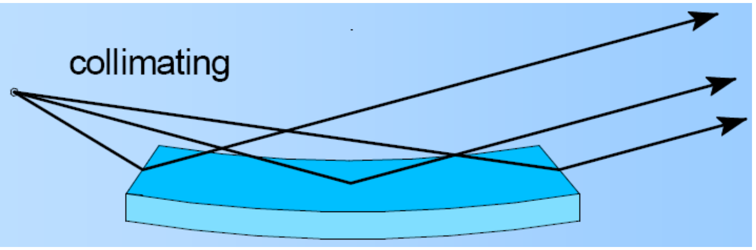
Bending radius *R* (can be ~ 10 km)



$$R = \frac{2D_1D_2}{\theta(D_1 + D_2)}$$

imaging the source in the vertical direction with unity magnefication (1:1 focusing)





improving energy resolution of a following monochromator by production of a parallel beam (collimating)

Free electron laser (FEL)

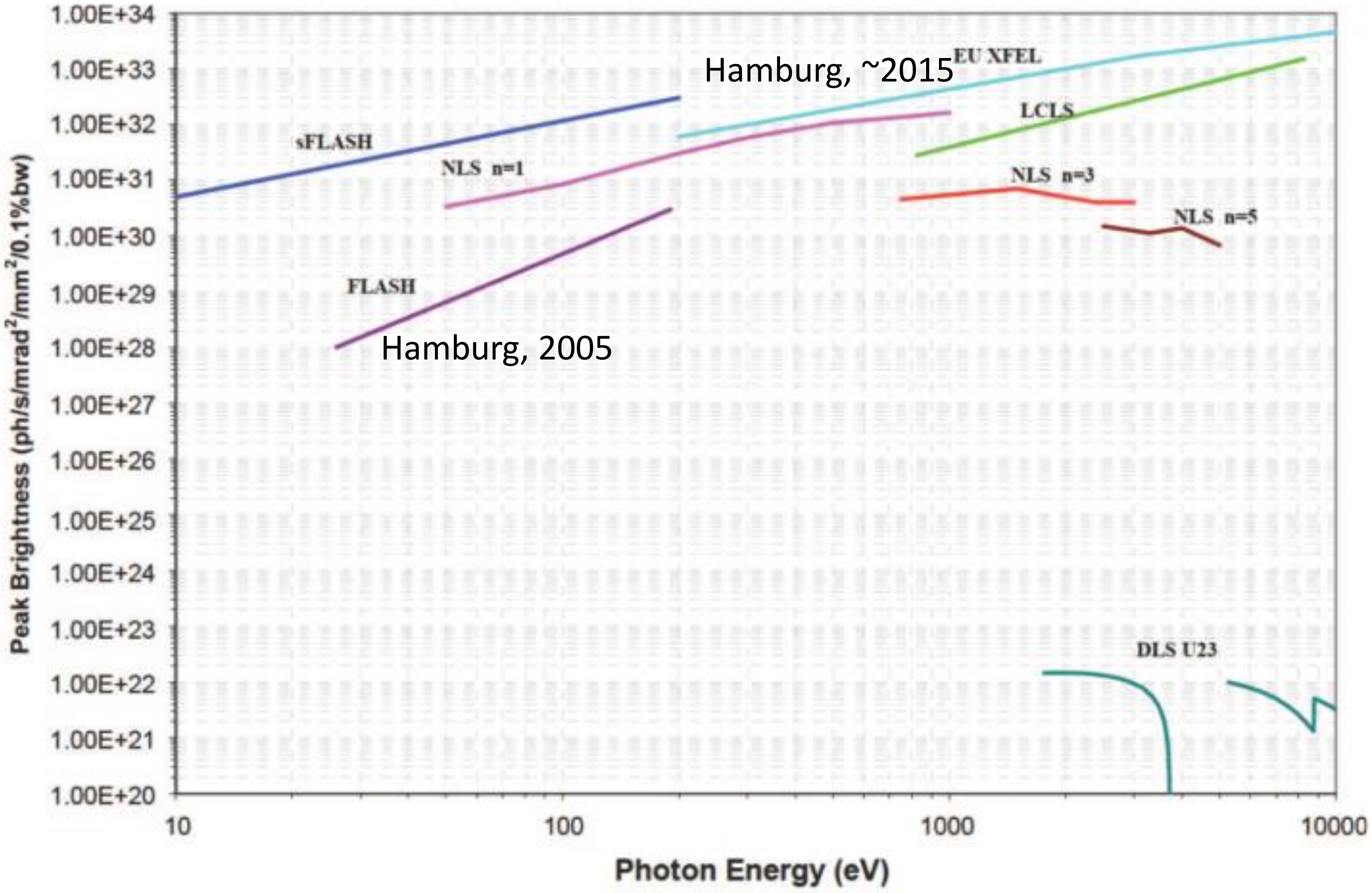


Figure 1. Comparison of several recently commissioned FELs (FLASH and LCLS) and several planned FELs (sFLASH, Euro XFEL, NLS) with a state-of-the-art undulator beamline on the Diamond Light source. The standard definition of brightness is given in photons/unit time/unit solid angle/unit area/normalised bandwidth. Courtesy of STFC, New Light Source Conceptual Design Report (2010) [1].

SASE – spontaneous amplified self-emission

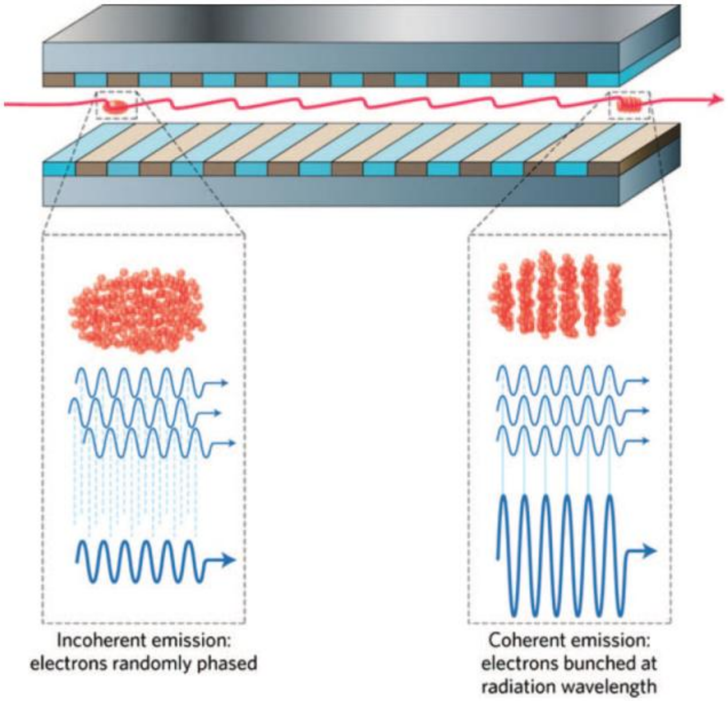
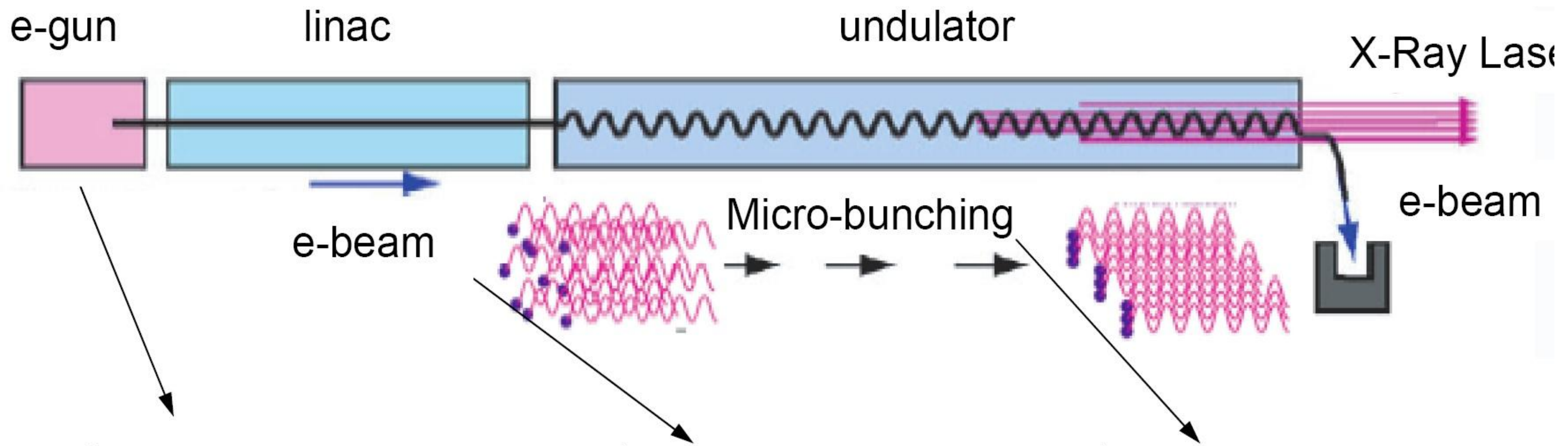


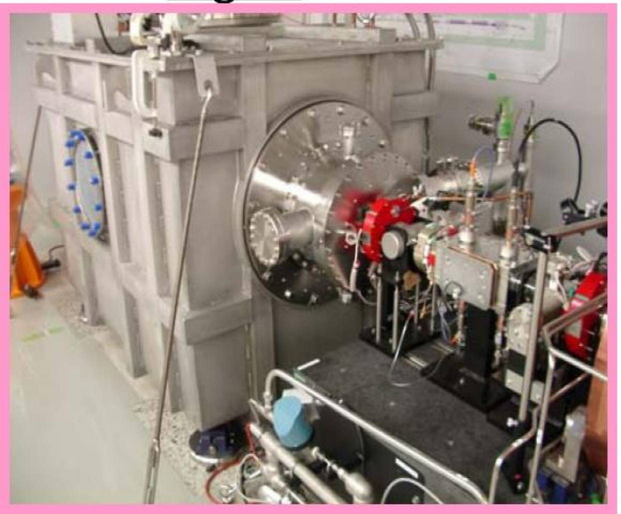
Figure 3. Schematic of SASE operation where the oscillation induced in the electron beam by the periodic magnetic field leads to radiation emission and, at the end of a sufficiently long undulator, self-organisation of the electrons gives rise to coherent X-ray emission. Reprinted by permission from Macmillan Publishers Ltd., Nature Photonics, B.W.J. McNeil and N.R. Thompson, *X-ray free electron lasers*, Nature Photonics, 4 (2010), pp. 814–821, copyright (2010).

Linac-based Free Electron Laser
Self-Amplified Spontaneous Emission (SASE)

Principle design (SPring-8, Japan):



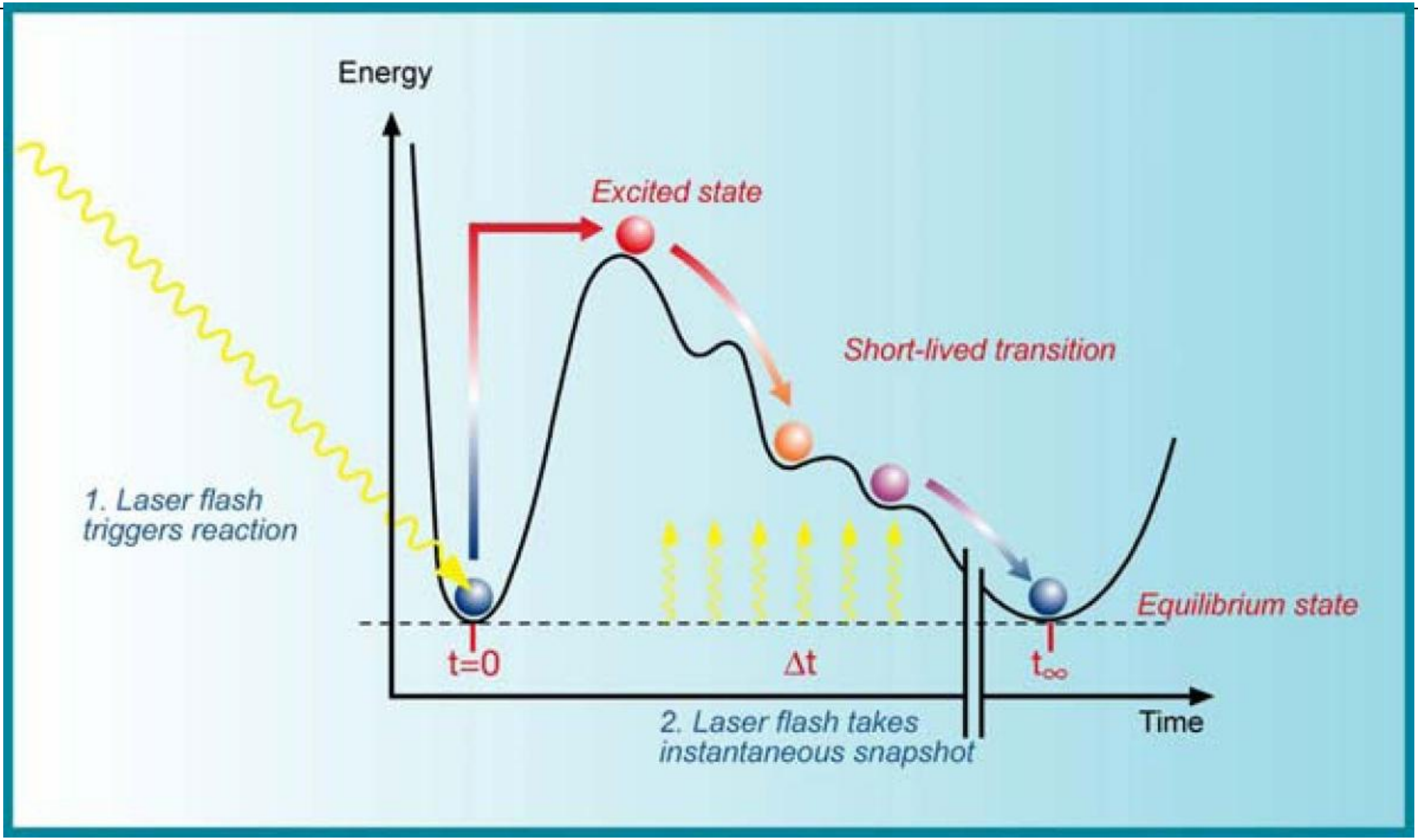
◆ **e-gun** (pulsed)



◆ **linac** (linear accelerator)

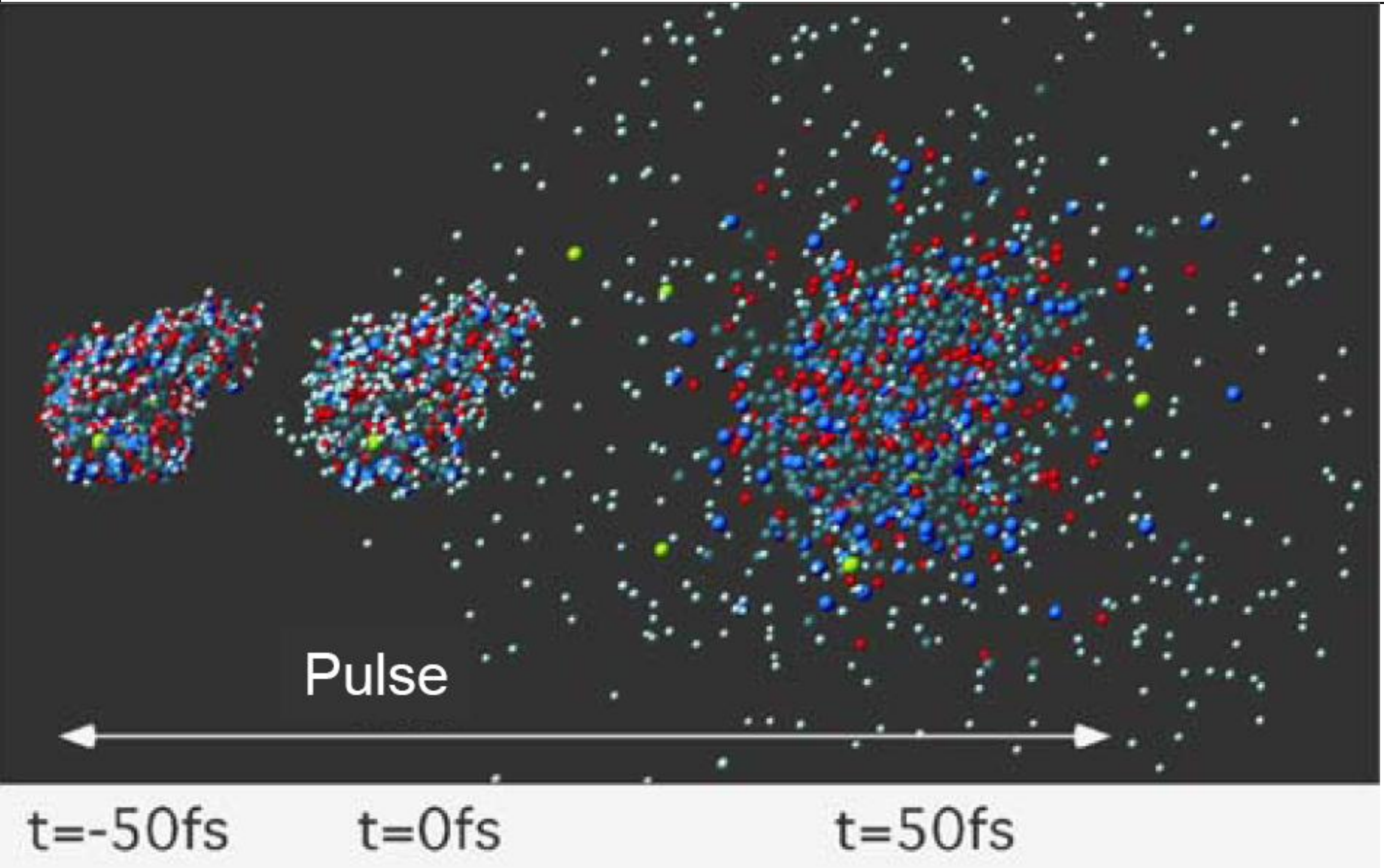
◆ **undulator**

Pump-probe experiment



Snapshots for different times after excitation (“pump-probe experiment”) ⇒ “film” of the reaction

Obstacle: Coulomb-Explosion

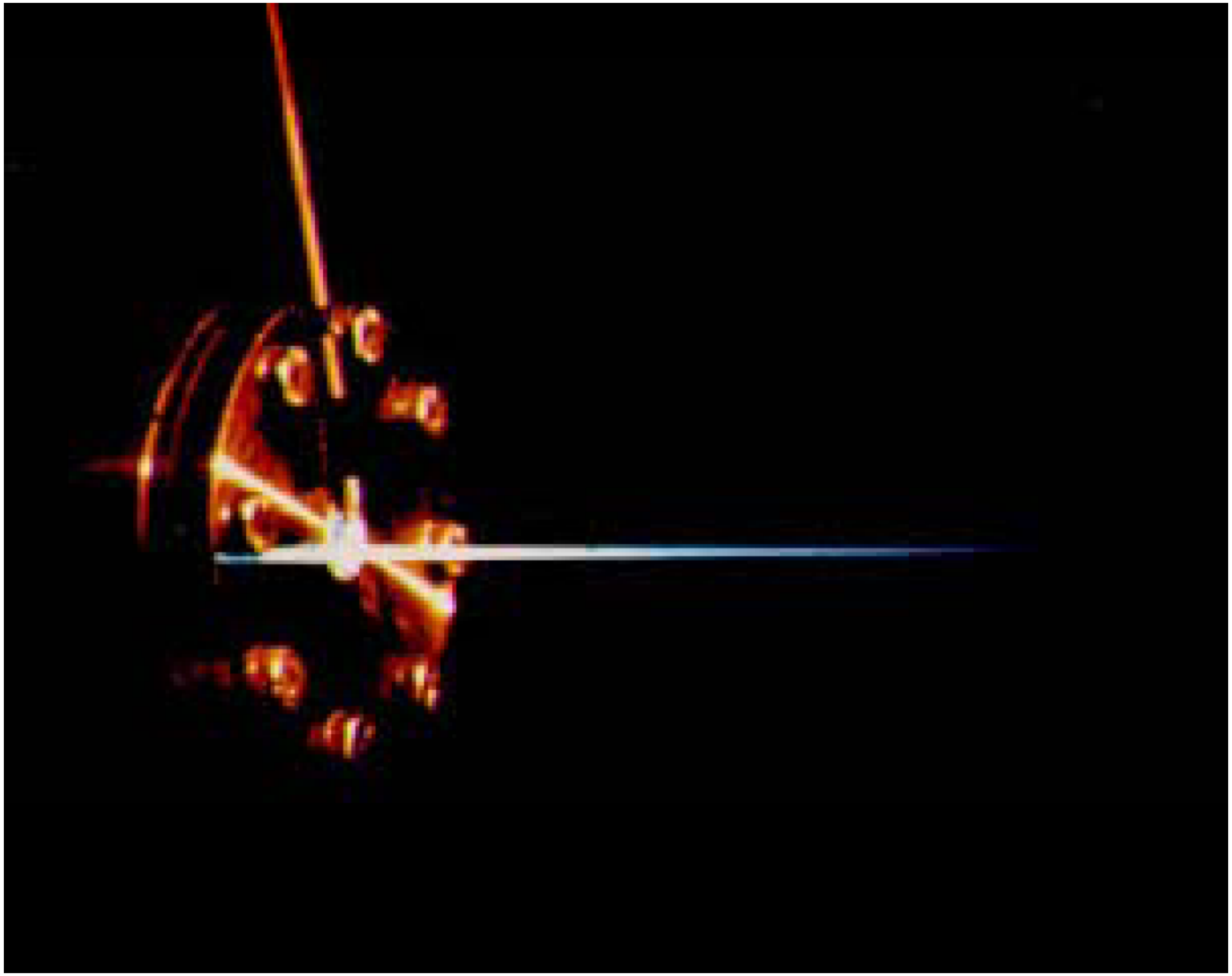


Example:
Lysozyme
white: Hydrogen,
grey: Carbon,
blue: Nitrogen,
red: Oxygen,
yellow: Sulfur

Requirement: **Pulse must be short enough and not to intense, to take picture before molecule disintegrates !**

R. Neutze et al., Nature, August 2000

Accelerator Synchrotron Radiation



This is a focused beam of synchrotron x-rays emerging through a thin window and ionizing the air to give a blue light.

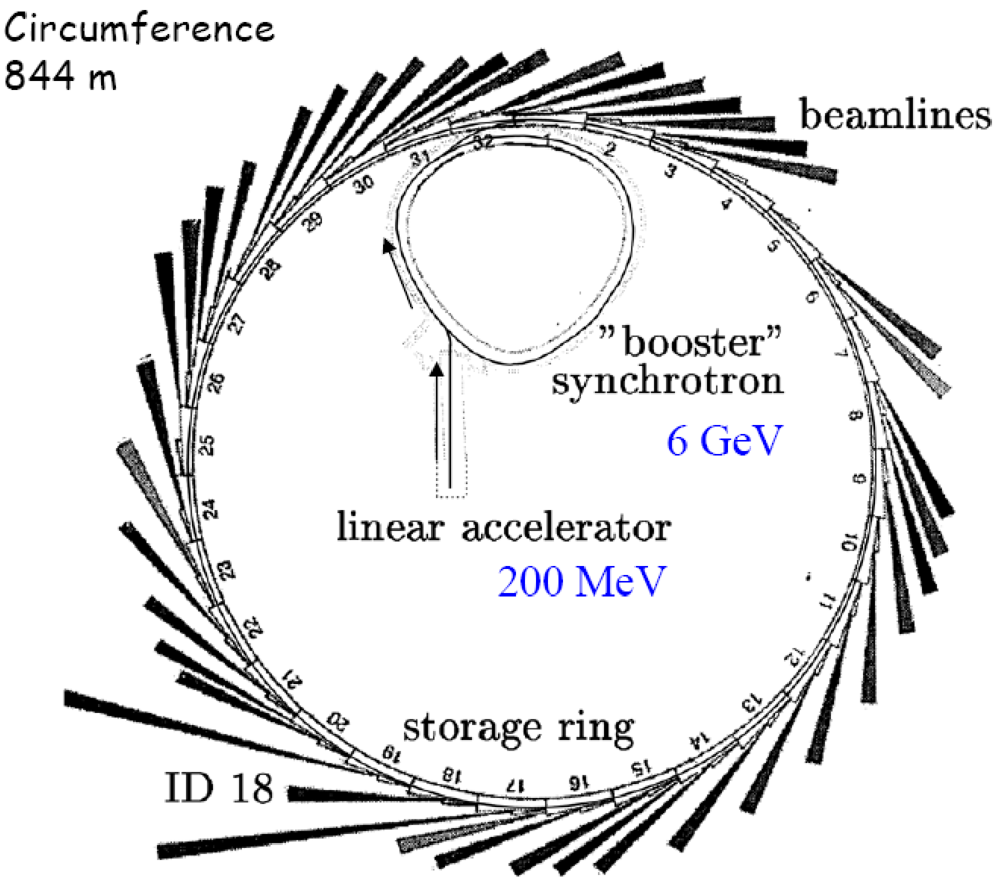
Crab Nebula – an astronomical synchrotron source

The supernova exploded in 1054 AD, and the gas should have cooled by today. But it is still emitting UV and X-rays. Why?

The answer is that very high energy electrons in a weak magnetic field are emitting synchrotron radiation.

Time structure of Synchrotron Radiation I

A close look into the storage ring!



RF-cavities in the ring provide the electric field to accelerate the electrons to compensate for the radiation losses

ν_{RF} = 352 MHz

<u>This means:</u>

992 buckets of stable phase for the electrons

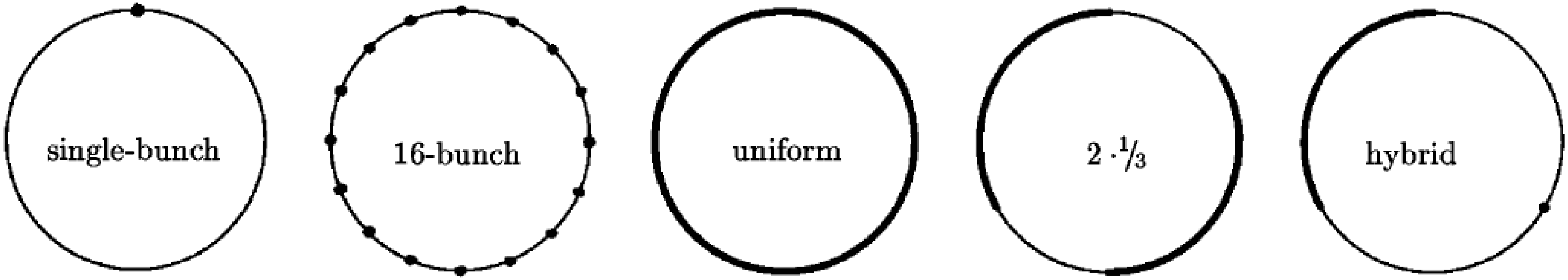
$$N = \nu_{RF} \cdot \frac{L}{c}$$

$$\frac{2.97088 \cdot 10^{11} \frac{m}{s}}{c} = 992$$

A <u>bucket</u> filled with electrons is called a **bunch** (duration 10-100ps).

$$\Delta t = \frac{L}{c} \cdot \frac{1}{N} = 2.84\,ns \quad (\,flashes\,)$$

Time structure of Synchrotron Radiation



$I_{max} = 16\,\mathrm{mA}$
lifetime $= 8\,\mathrm{h}$
$2.81\,\mu\mathrm{s}$ gaps

$I_{max} = 90\,\mathrm{mA}$
lifetime $= 10\,\mathrm{h}$
$176\,\mathrm{ns}$ gaps

$I_{max} = 200\,\mathrm{mA}$
lifetime $= 60\,\mathrm{h}$
$2.839\,\mathrm{ns}$ gaps

$I_{max} = 200\,\mathrm{mA}$
lifetime $= 55\,\mathrm{h}$
$2.839\,\mathrm{ns}$ & $0.94\,\mu\mathrm{s}$

$I_{max} = 193 + 7\,\mathrm{mA}$
lifetime $= 40\,/\,7\,\mathrm{h}$
$2.839\,\mathrm{ns}$ & $0.47\,\mu\mathrm{s}$

By selecting well defined time structure
→ **Time resolved measurements** (e.g. dynamic processes in Biology, chemical bonding, magnetism and Mössbauer spectroscopy with Synchrotron Radiation
→**Mode of operation depends on the type of experiment**