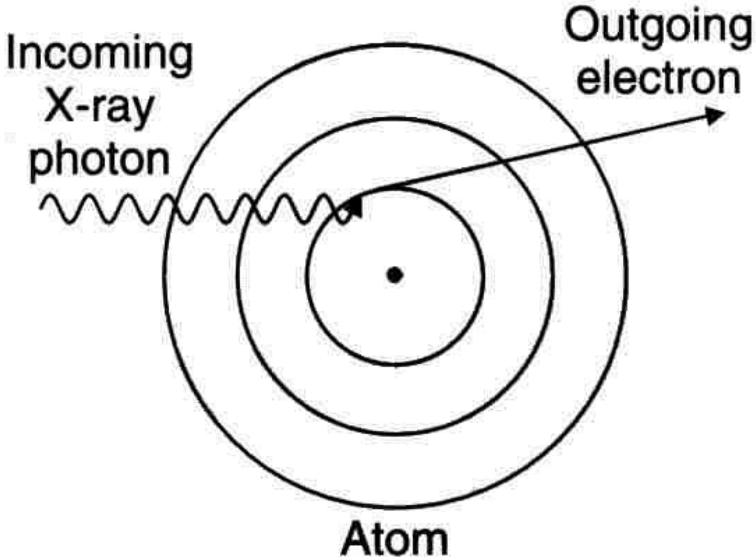
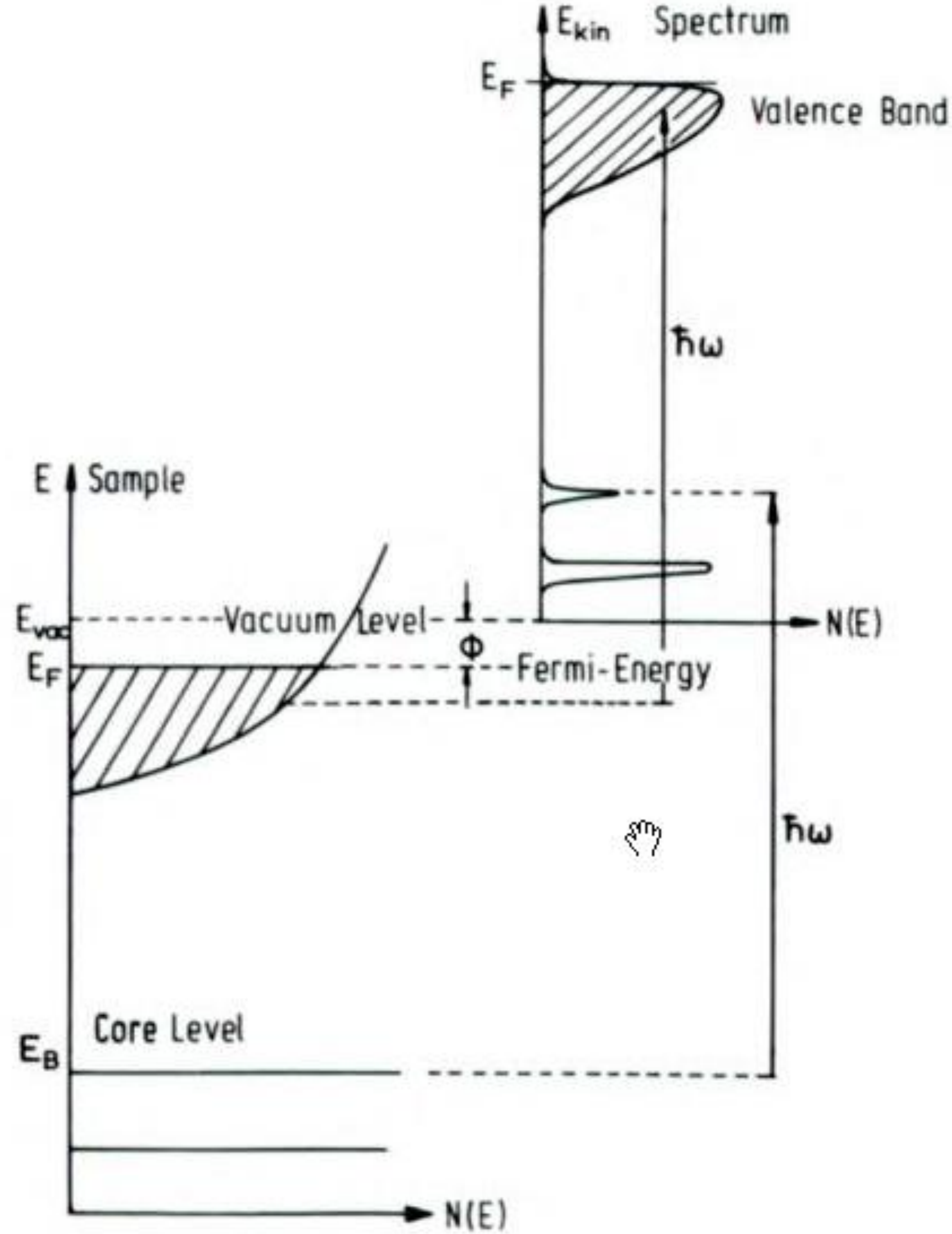
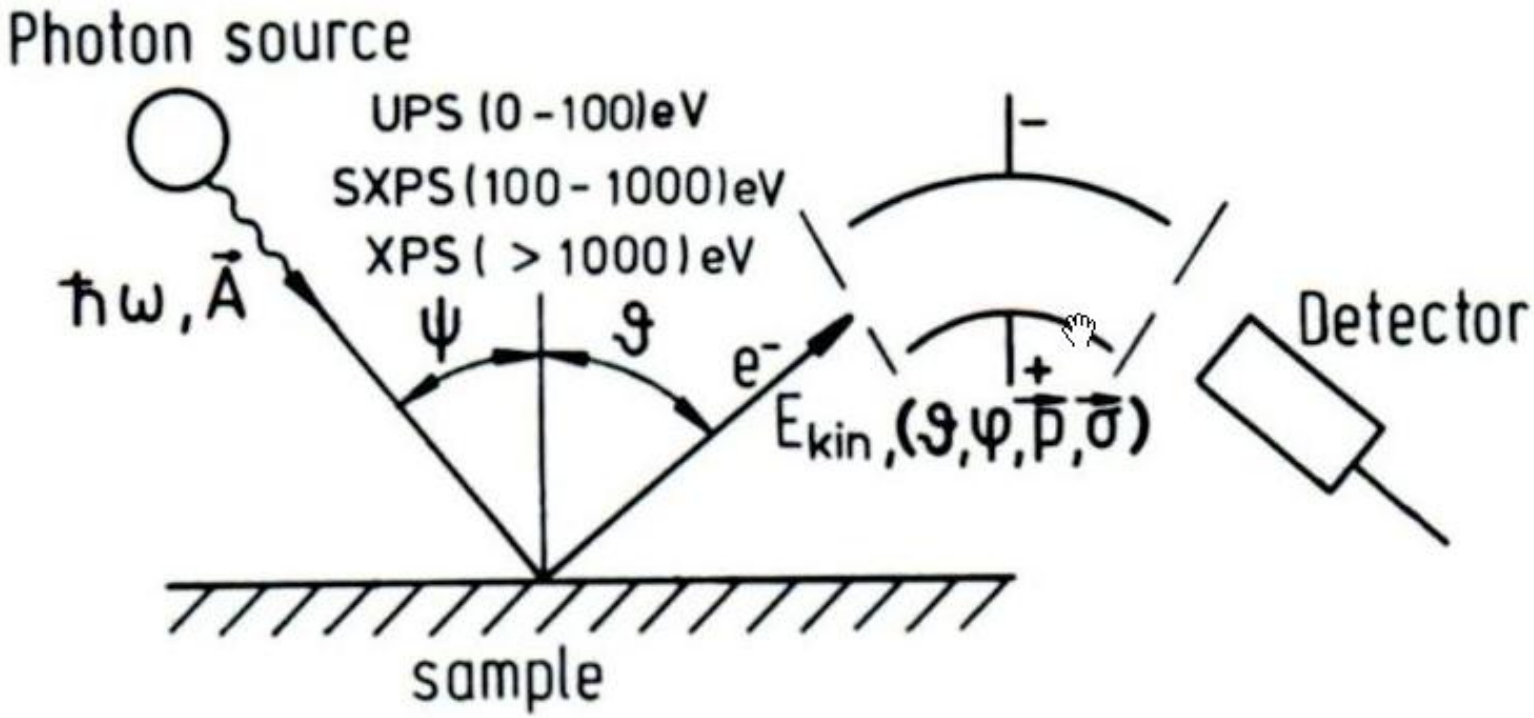


$$E_{\mathrm{kin}} = \hbar\omega - \phi - E_{\mathrm{b}}$$





$\vec{A}$: vector potential of incident light

ψ : incident angle

ϑ : polar angle of emission

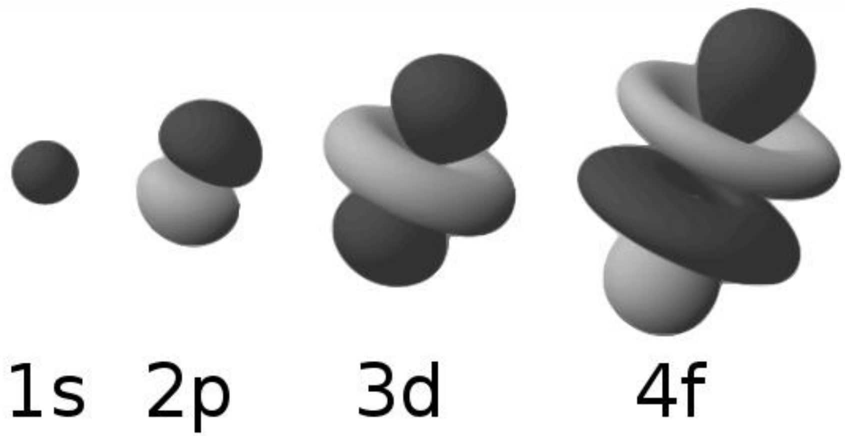
φ : azimuthal angle of emission

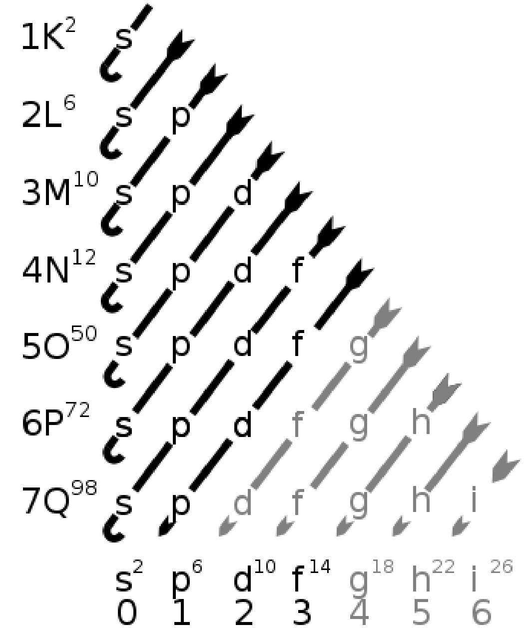
E_{kin} : kinetic energy of emitted electron

$\vec{p} = \hbar\vec{k} = \hbar\dfrac{2\pi}{\lambda}\vec{e}_k$: momentum of emitted electron

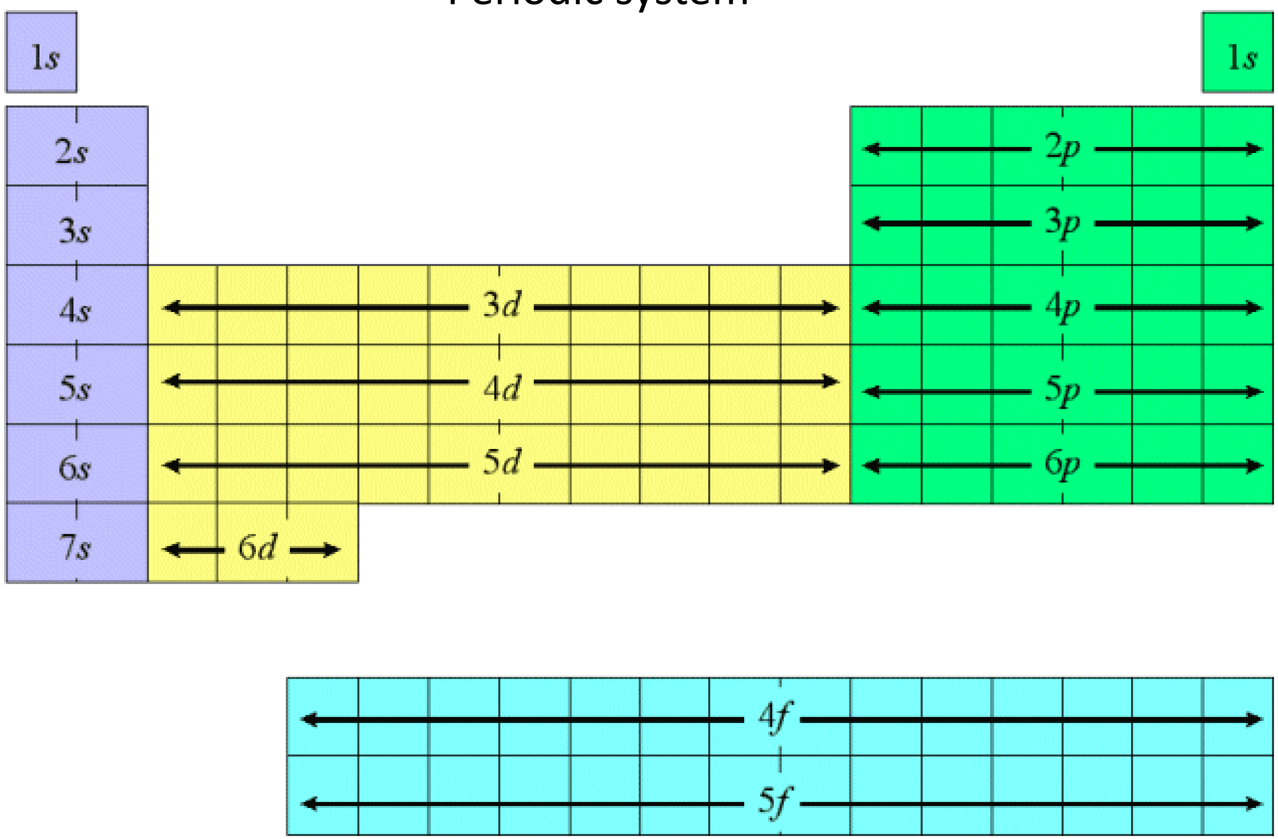
$\vec{\sigma}$: spin of emitted electron

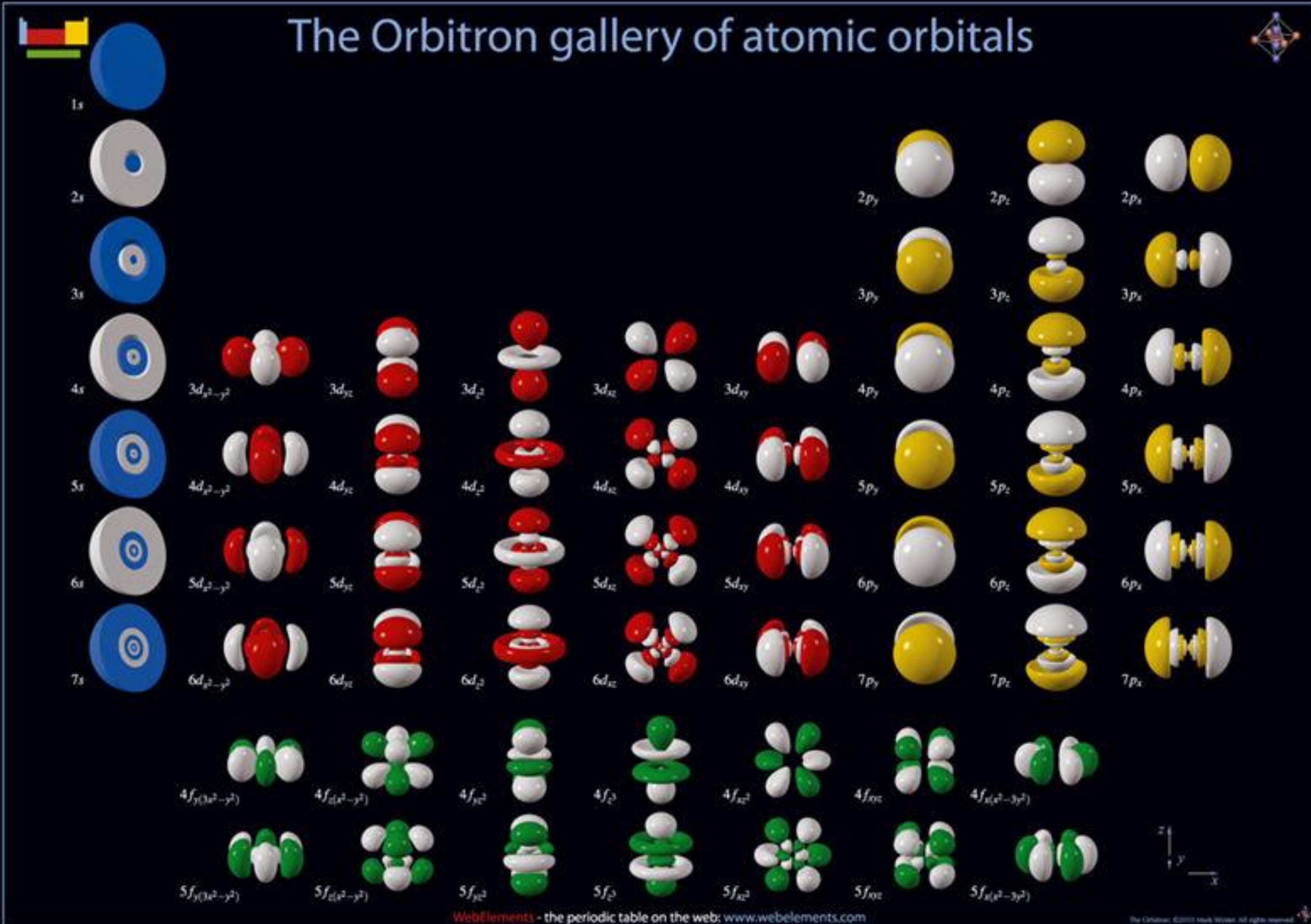
Nobel price in physics 1981 for Siegbahn (shared with Bloembergen and Schawlow)
“for his contribution to the development of high-resolution electron spectroscopy".

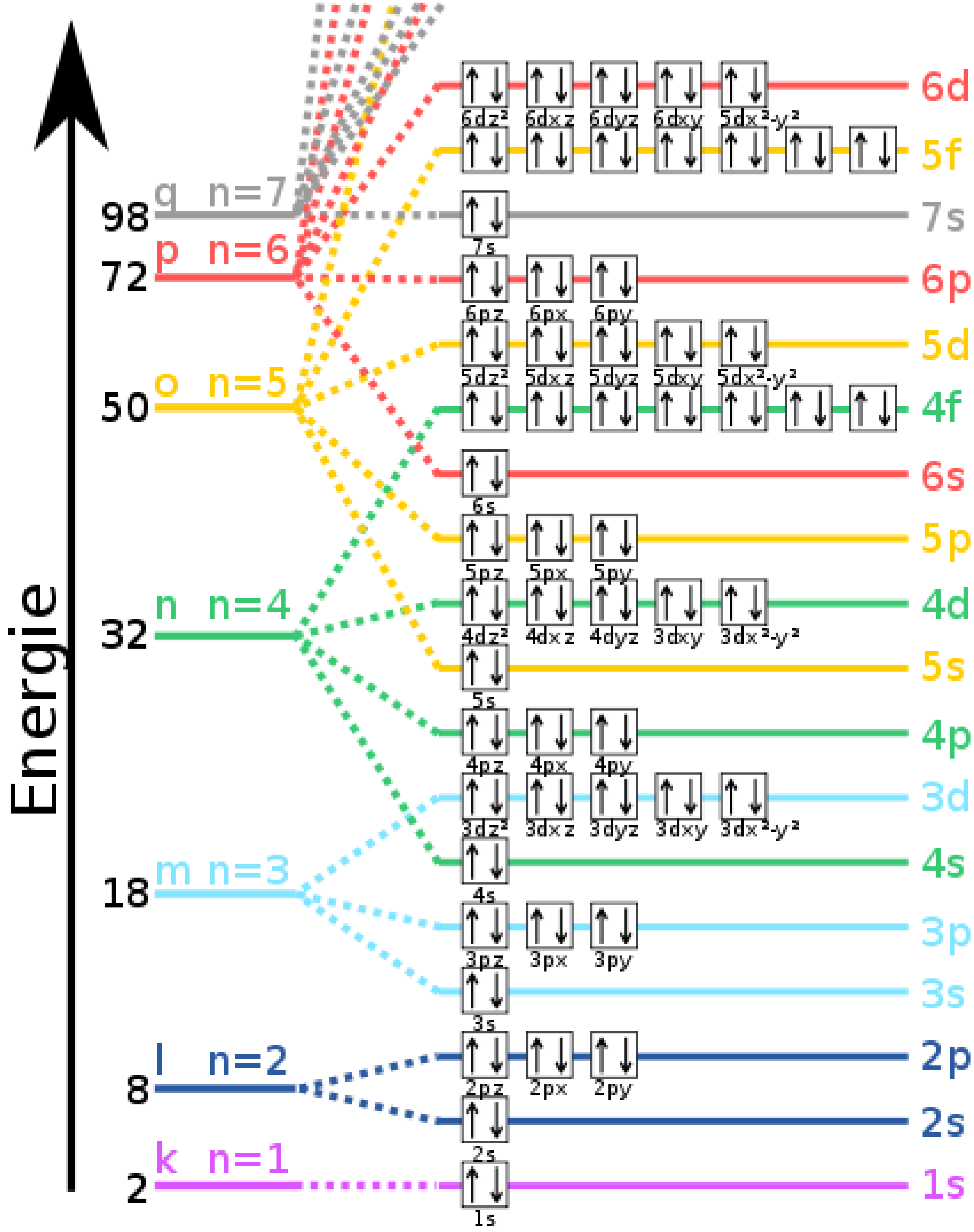




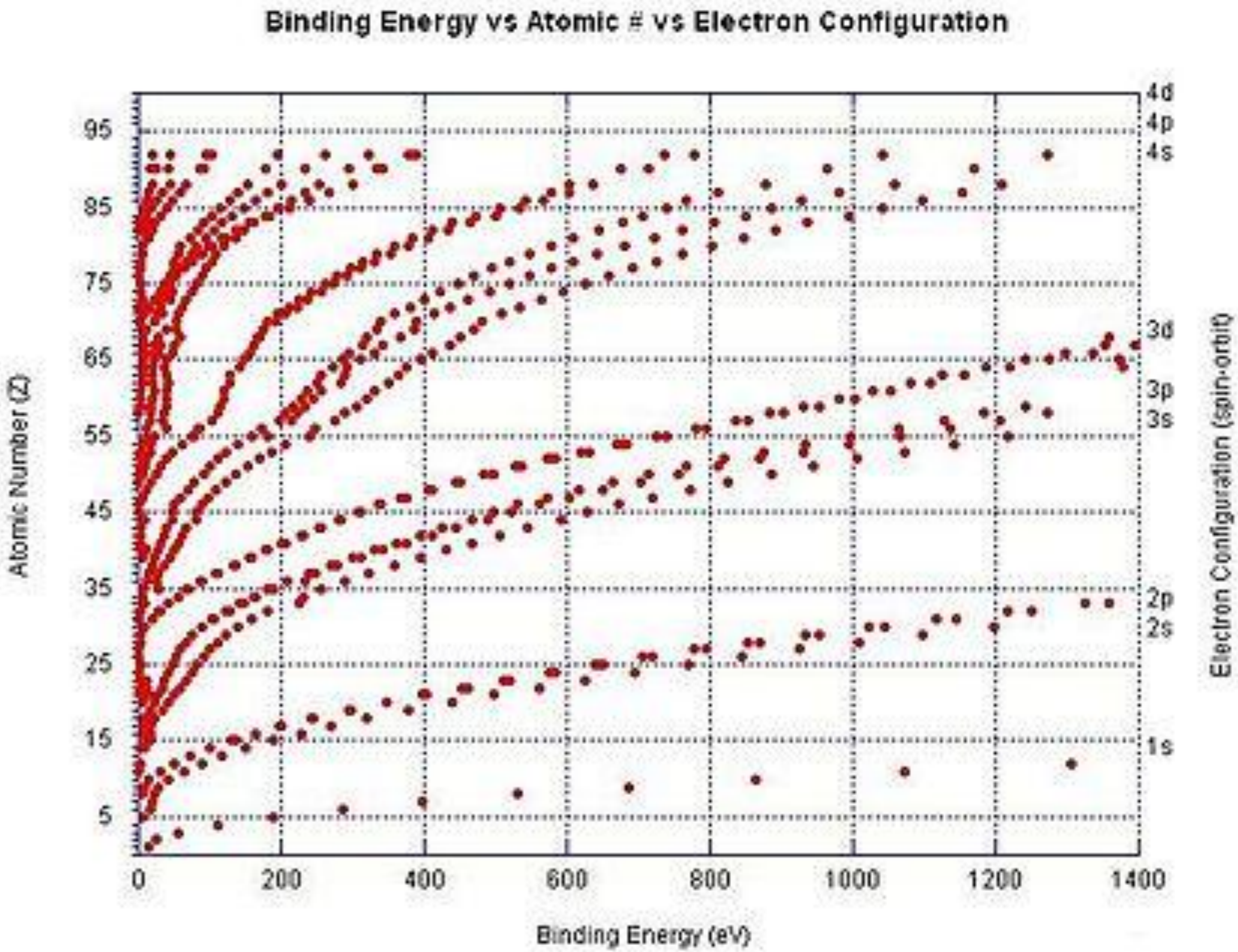
Periodic system




The Orbitron gallery of atomic orbitals
1s
2s
3s
4s
5s
6s
7s
2p_y 2p_z 2p_x
3p_y 3p_z 3p_x
3$d_{x^2-y^2}$ 3d_{yz} 3d_{z^2} 3d_{xz} 3d_{xy} 4p_y 4p_z 4p_x
4$d_{x^2-y^2}$ 4d_{yz} 4d_{z^2} 4d_{xz} 4d_{xy} 5p_y 5p_z 5p_x
5$d_{x^2-y^2}$ 5d_{yz} 5d_{z^2} 5d_{xz} 5d_{xy} 6p_y 6p_z 6p_x
6$d_{x^2-y^2}$ 6d_{yz} 6d_{z^2} 6d_{xz} 6d_{xy} 7p_y 7p_z 7p_x
4$f_{y(3x^2-y^2)}$ 4$f_{z(x^2-y^2)}$ 4f_{yz^2} 4f_{z^3} 4f_{xz^2} 4f_{xyz} 4$f_{x(x^2-3y^2)}$
5$f_{y(3x^2-y^2)}$ 5$f_{z(x^2-y^2)}$ 5f_{yz^2} 5f_{z^3} 5f_{xz^2} 5f_{xyz} 5$f_{x(x^2-3y^2)}$
z
y
x
WebElements - the periodic table on the web: www.webelements.com

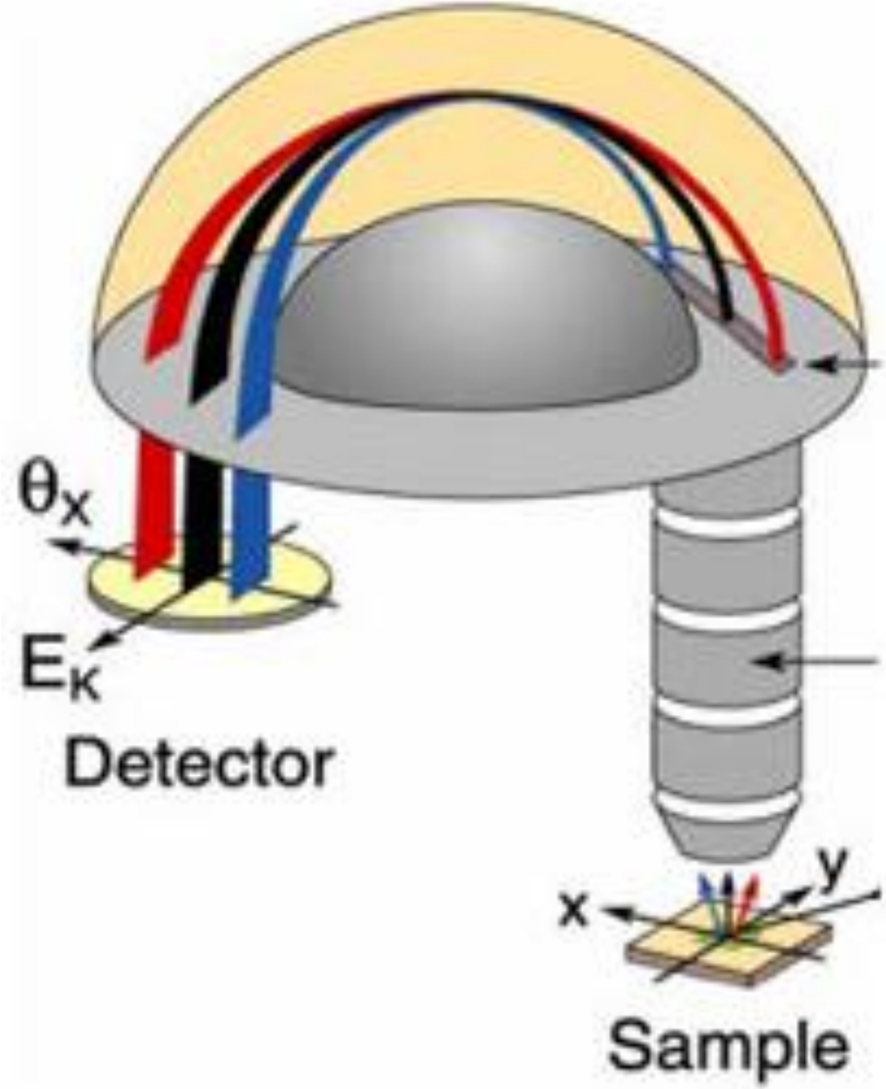


Energie
98
q n=7
72
p n=6
50
o n=5
32
n n=4
18
m n=3
8
l n=2
2
k n=1
6dz² 6dxz 6dyz 6dxy 5dx²-y²
6d
5f
7s
7s
6pz 6px 6py
6p
5dz² 5dxz 5dyz 5dxy 5dx²-y²
5d
4f
6s
6s
5pz 5px 5py
5p
4dz² 4dxz 4dyz 3dxy 3dx²-y²
4d
5s
5s
4pz 4px 4py
4p
3dz² 3dxz 3dyz 3dxy 3dx²-y²
3d
4s
4s
3pz 3px 3py
3p
3s
3s
2pz 2px 2py
2p
2s
2s
1s
1s

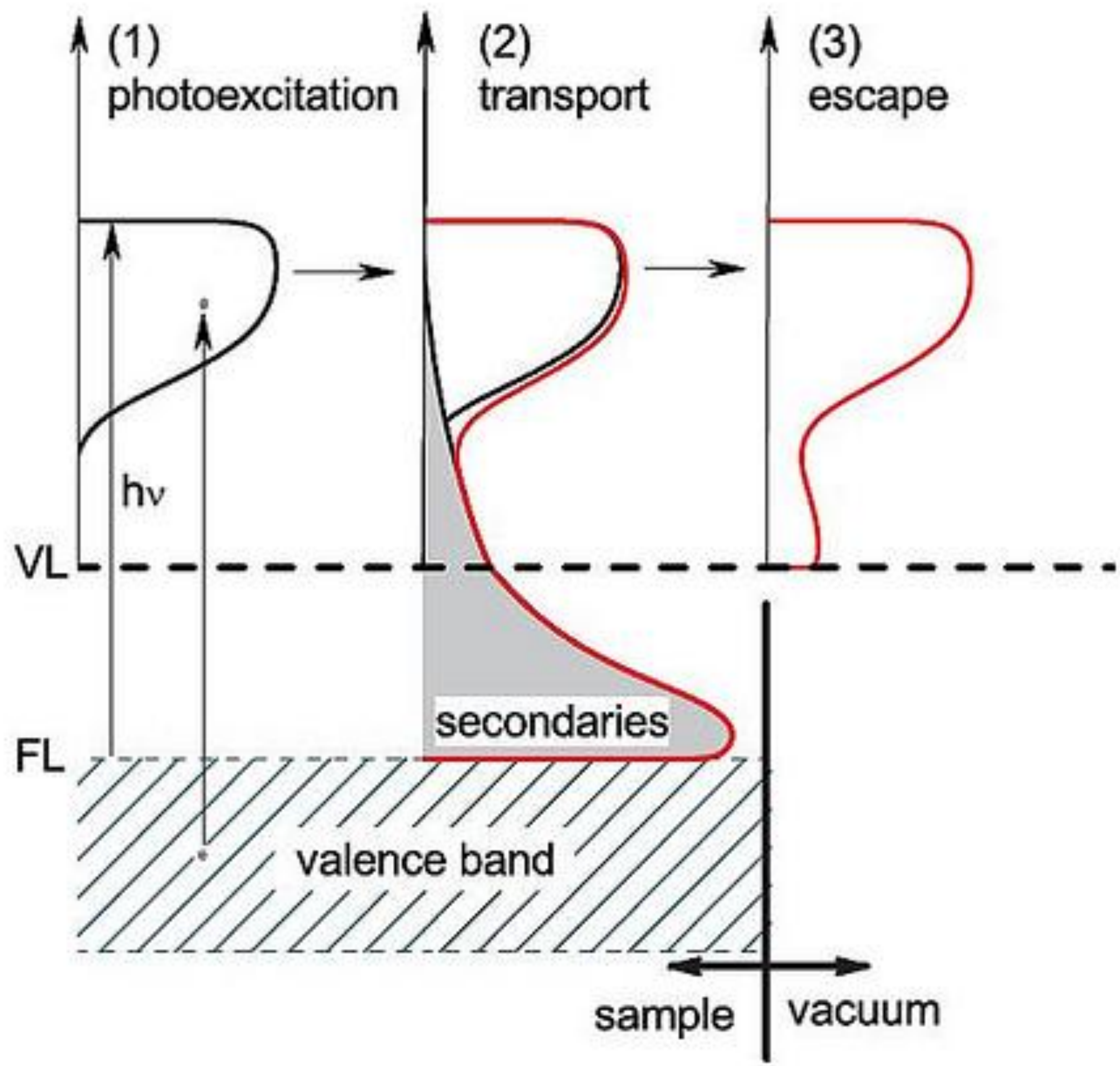


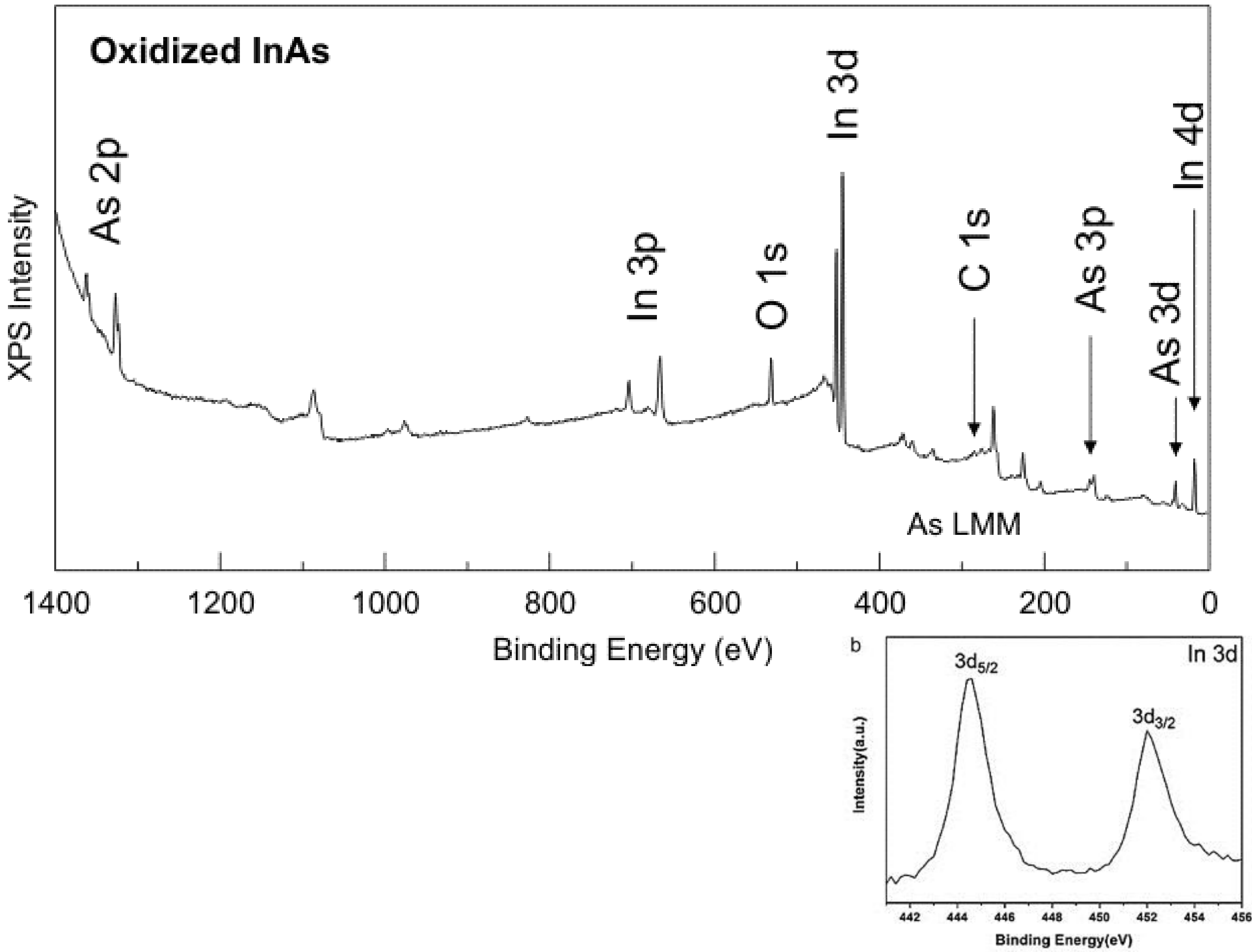
Binding Energy vs Atomic # vs Electron Configuration
Atomic Number (Z)
95
85
75
65
55
45
35
25
15
5
0
200
400
600
800
1000
1200
1400
Binding Energy (eV)
4d
4p
4s
3d
3p
3s
2p
2s
1s
Electron Configuration (spin-orbit)

hemispherical energy analyzer



Three-step model



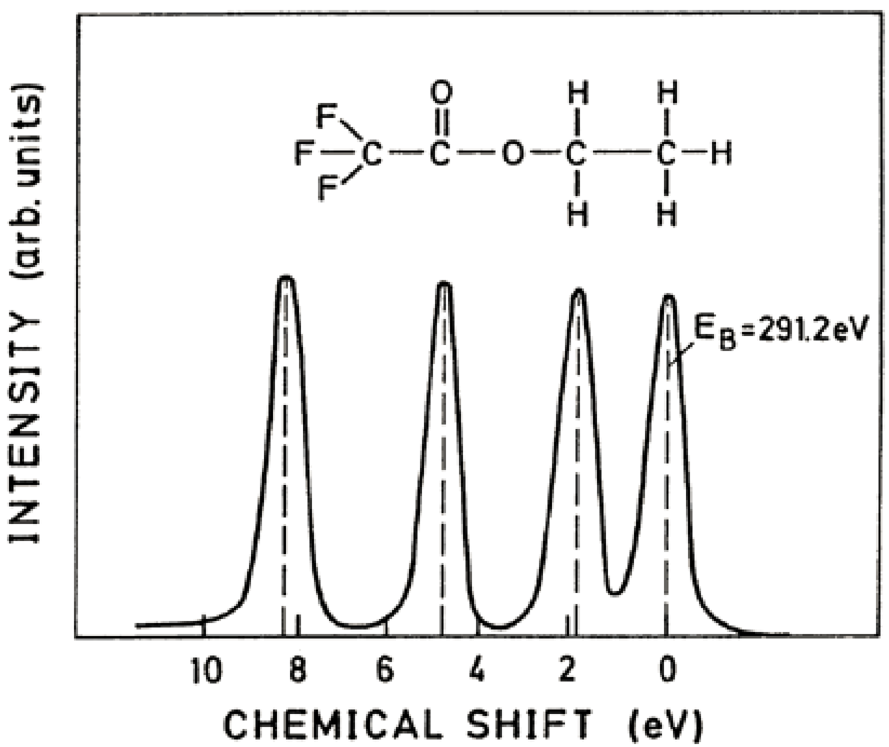


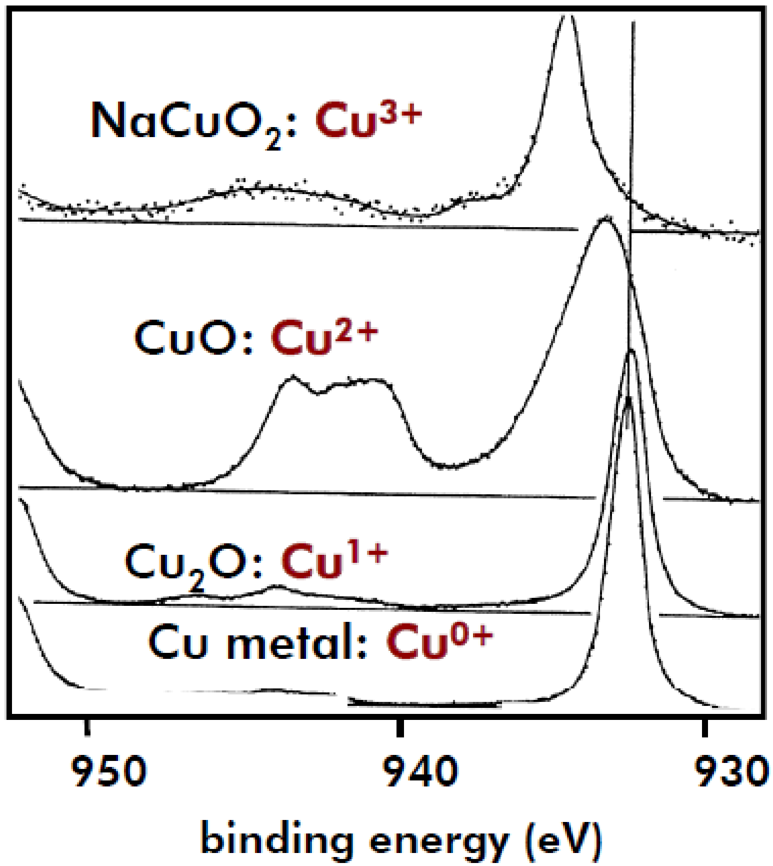
Oxidized InAs
XPS Intensity
As 2p
In 3p
O 1s
In 3d
C 1s
As 3p
As 3d
In 4d
As LMM
1400
1200
1000
800
600
400
200
0
Binding Energy (eV)
b
In 3d
$3d_{5/2}$
$3d_{3/2}$
Intensity(a.u.)
442
444
446
448
450
452
454
456
Binding Energy(eV)

Chemical shift in XPS

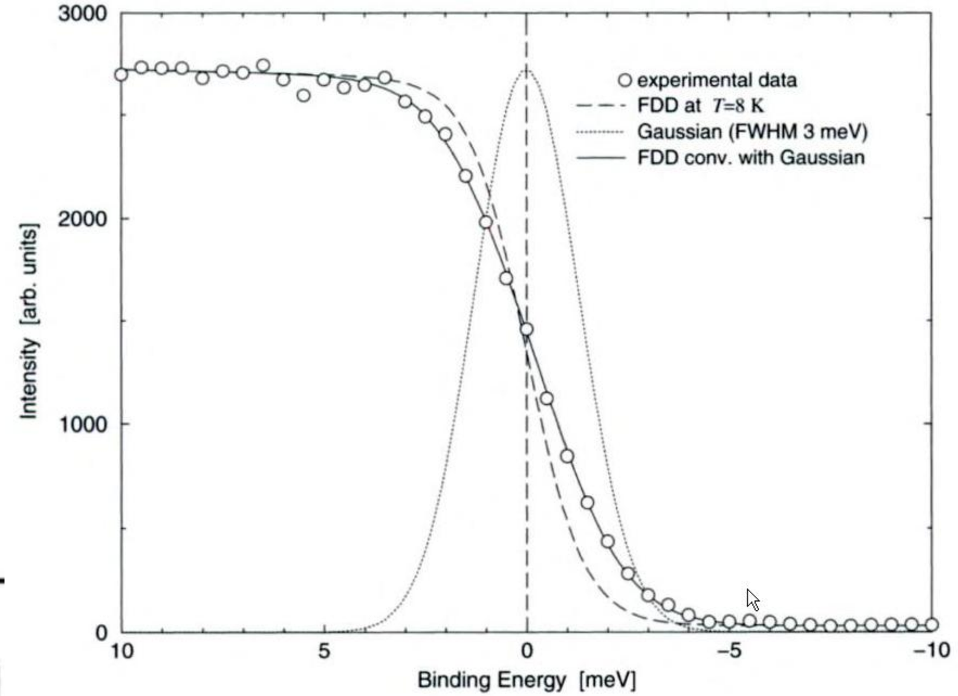
Example:

C 1s XPS signal in ethylfluoroace

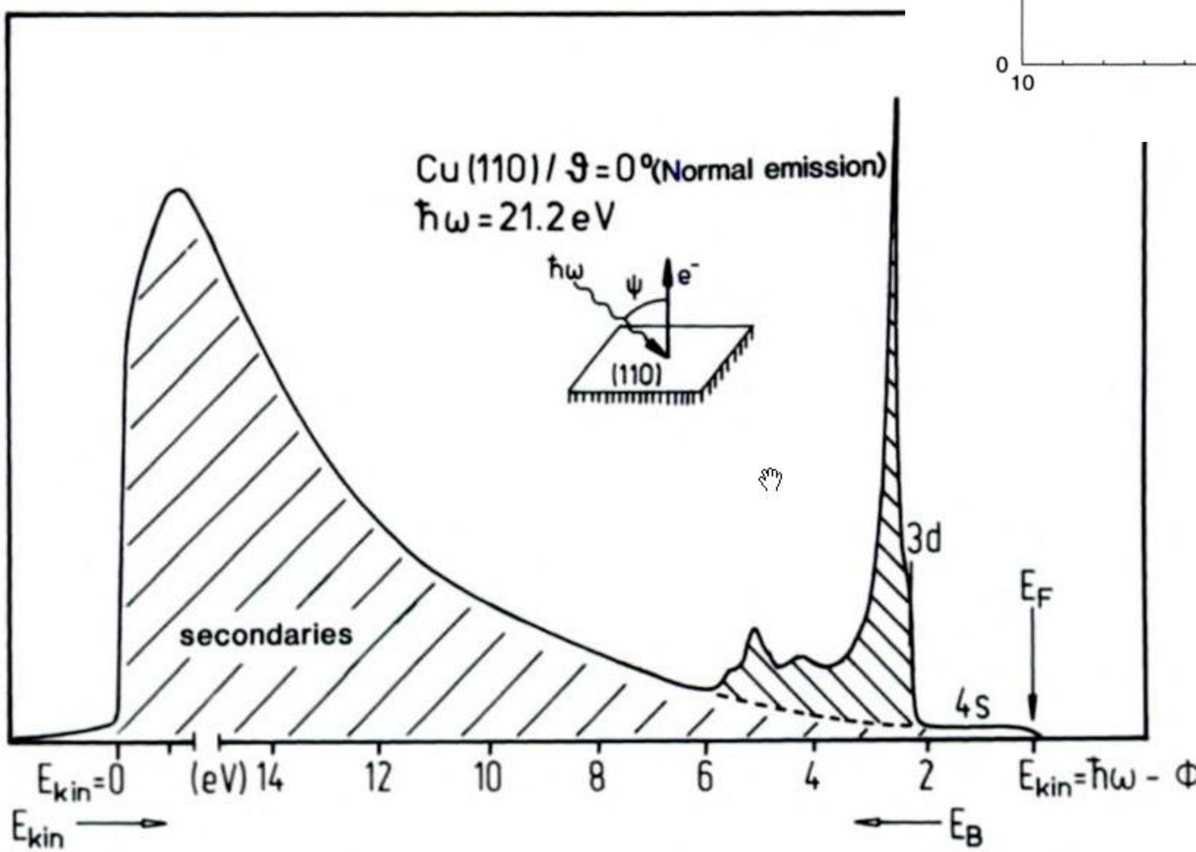




UPS

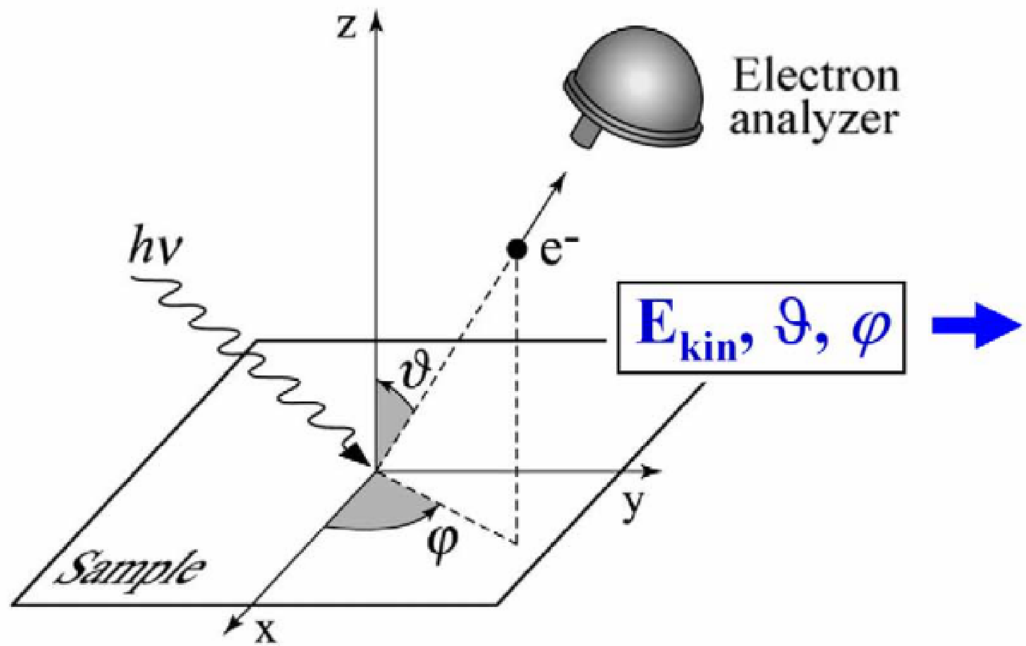


experimental data
FDD at T=8 K
Gaussian (FWHM 3 meV)
FDD conv. with Gaussian
Intensity [arb. units]
Binding Energy [meV]
3000
2000
1000
0
10
5
0
−5
−10



Cu (110) / ϑ = 0°(Normal emission)
ħω = 21.2 eV
ħω
ψ
e⁻
(110)
secondaries
3d
4s
E_F
E_kin=0
(eV) 14
12
10
8
6
4
2
E_kin=ħω - Φ
E_kin
E_B

ARPES principle



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

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

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

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

Vacuum

E_{kin}

$\vec{K}$

Conservation laws

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

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

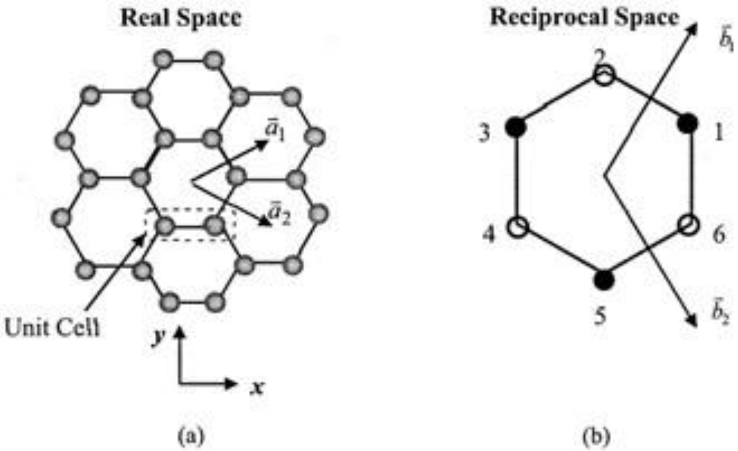
Solid

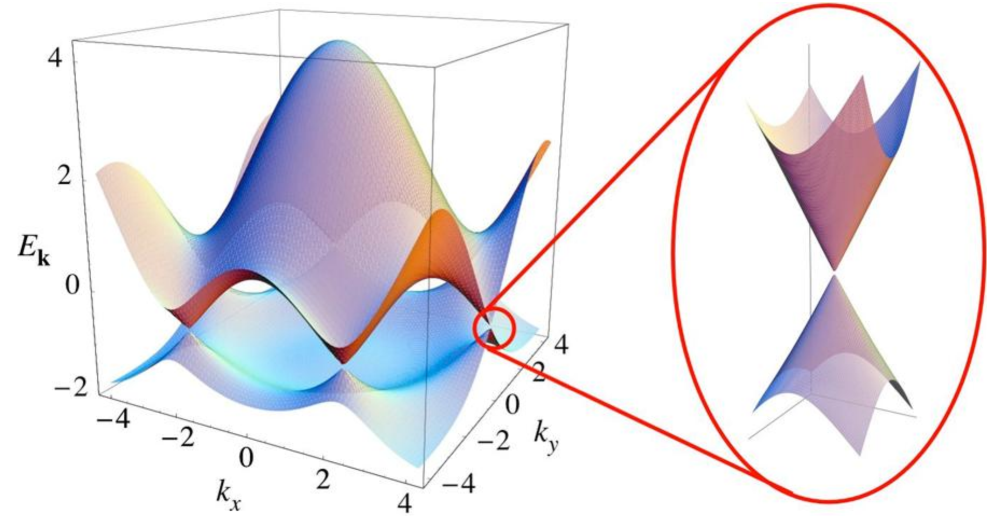
E_B

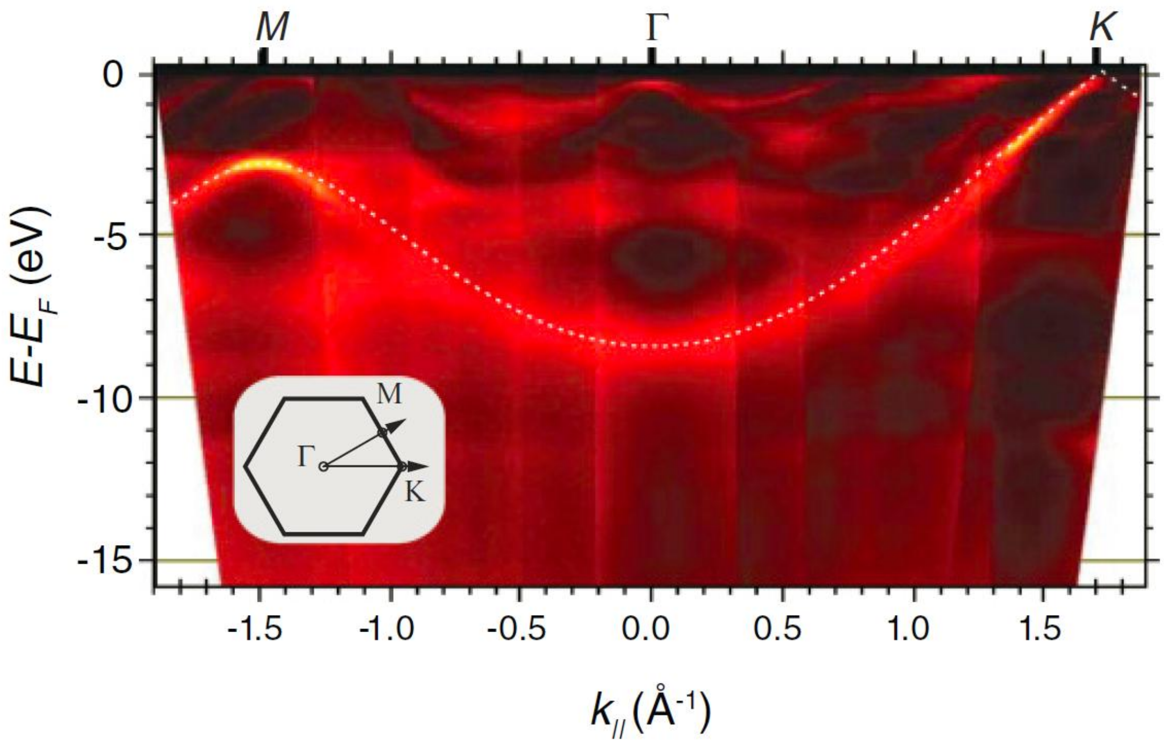
$\vec{k}_{||}$

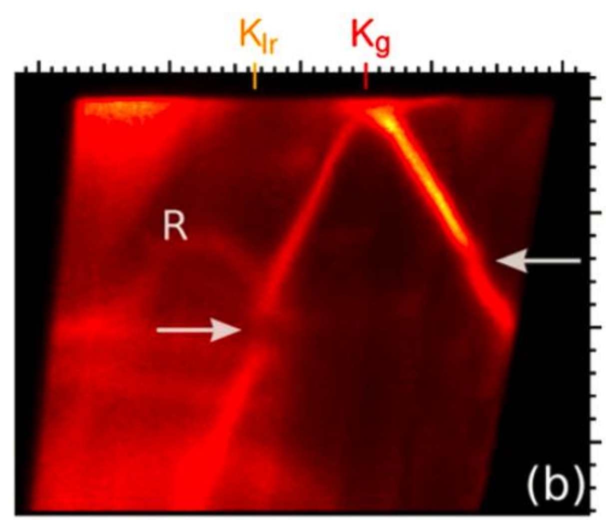
conservation of parallel momentum

ARPES on graphene









Inverse photoemission spectroscopy

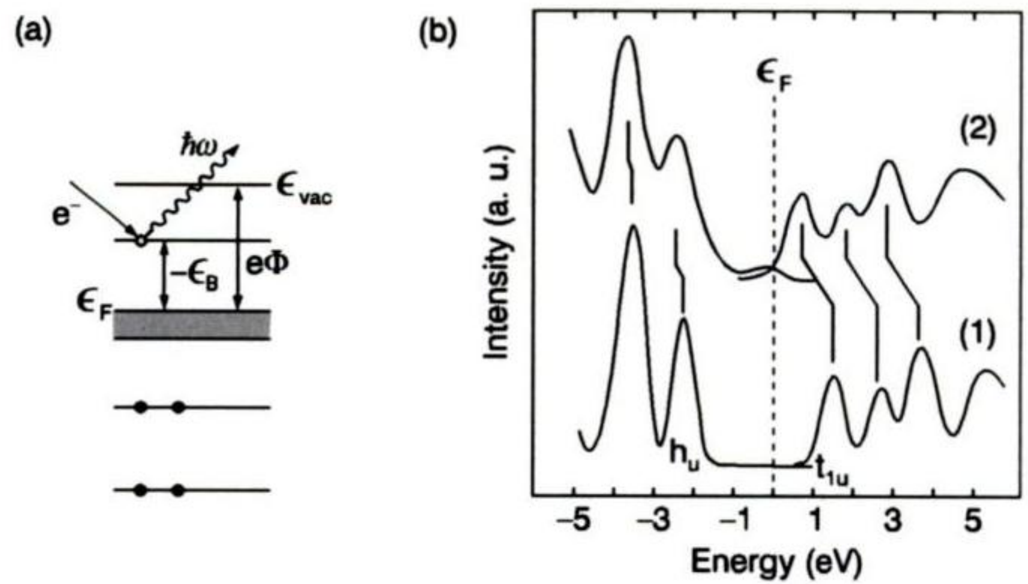
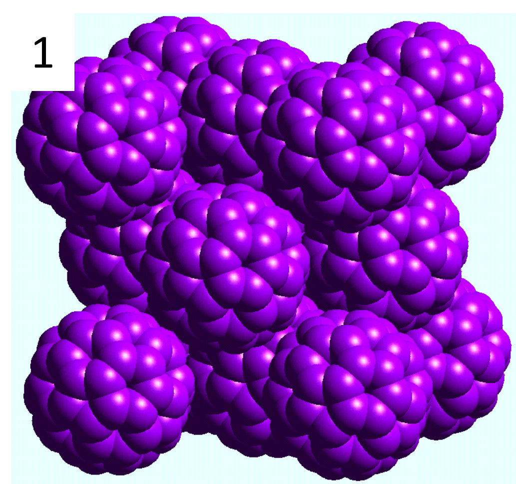
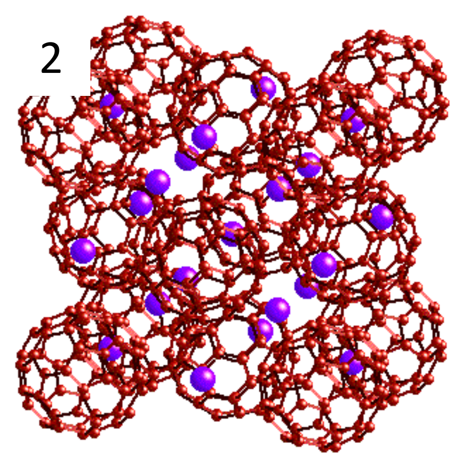


Fig. 12.19. Electronic levels and recombination processes for inverse photoemission (**a**) and photoemission and inverse photoemission for C_{60} (1) and K_3C_{60} (2) (**b**); The Mullikan symbols h_u and t_{1u} label the symmetry of the bands; (b) after [12.15].

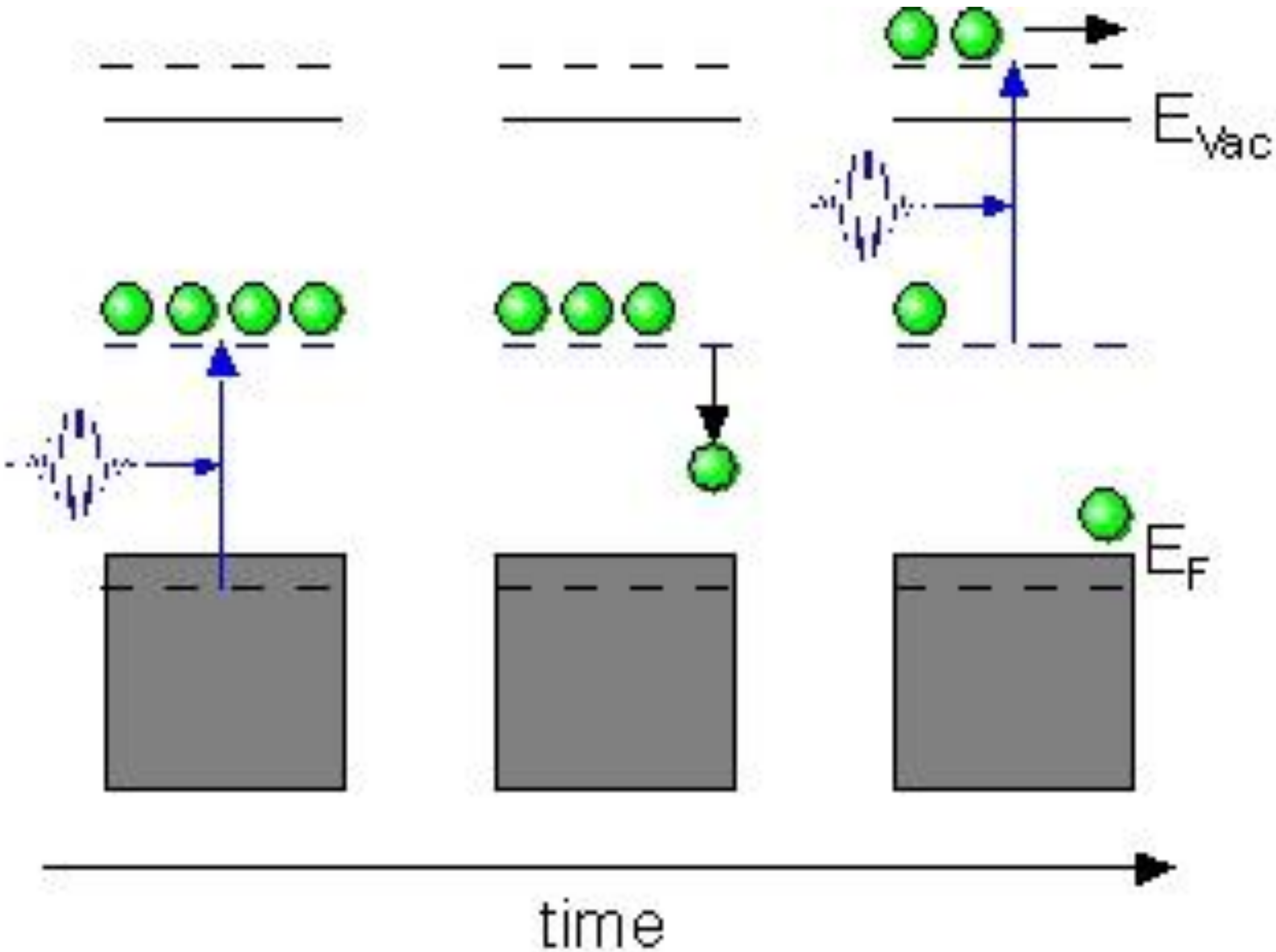
1



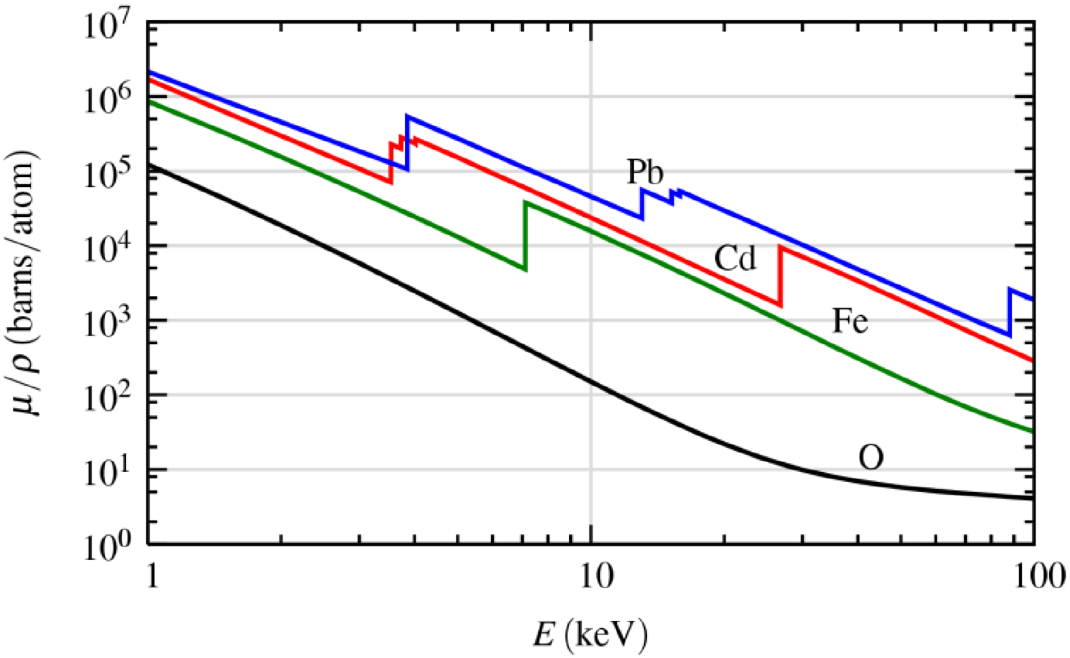
2



Two-photon photoemission (2PPE)



X-ray adsorption spectroscopy



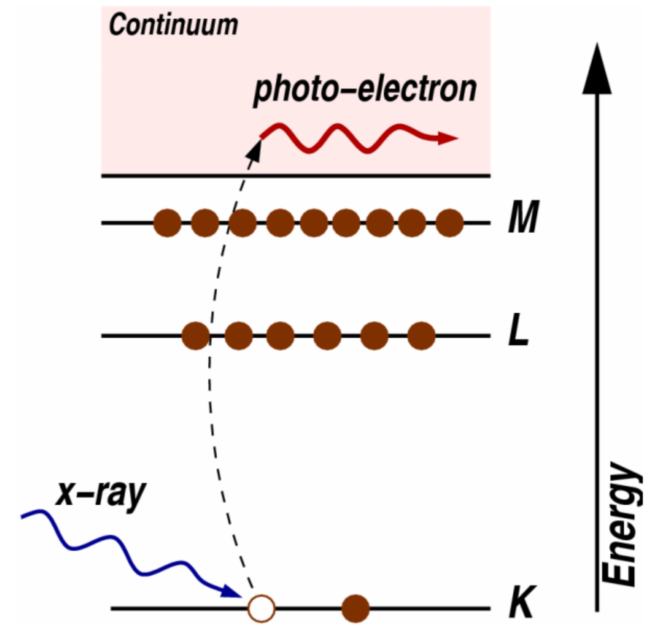


Figure 2.3: The absorption cross-section μ/ρ for several elements over the x-ray energy range of 1 to 100 keV. Notice that there are at least 5 orders of magnitude in variation in μ/ρ, and that in addition to the strong energy dependence, there are also sharp rises corresponding to the core-level binding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

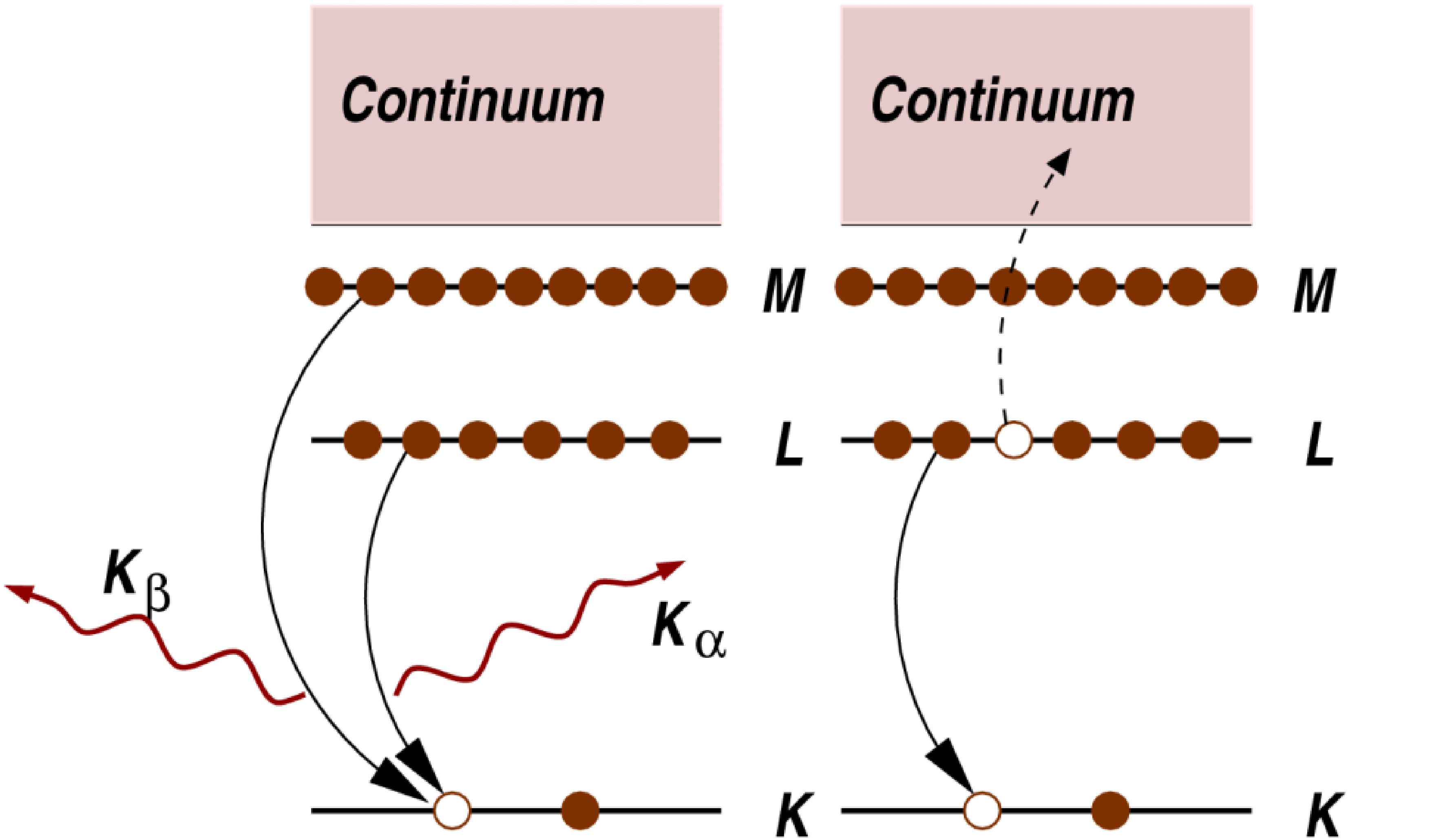


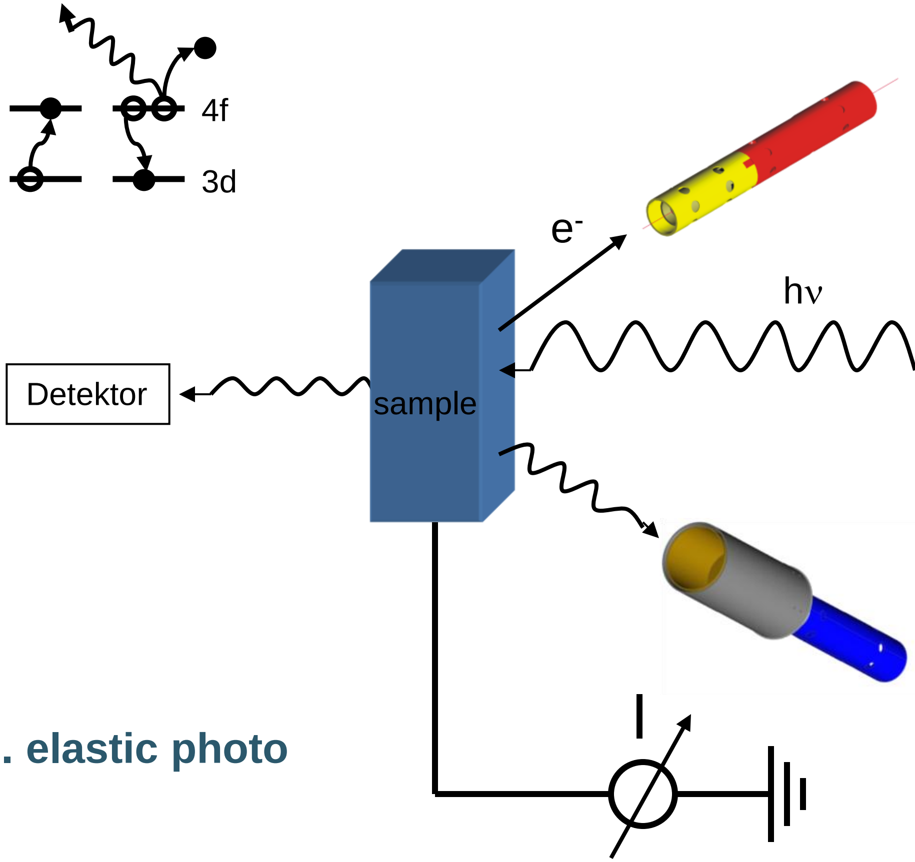
Figure 2.5: Decay of the excited state: x-ray fluorescence (left) and the Auger effect (right). In both cases, the probability of emission (x-ray or electron) is directly proportional to the absorption probability.

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

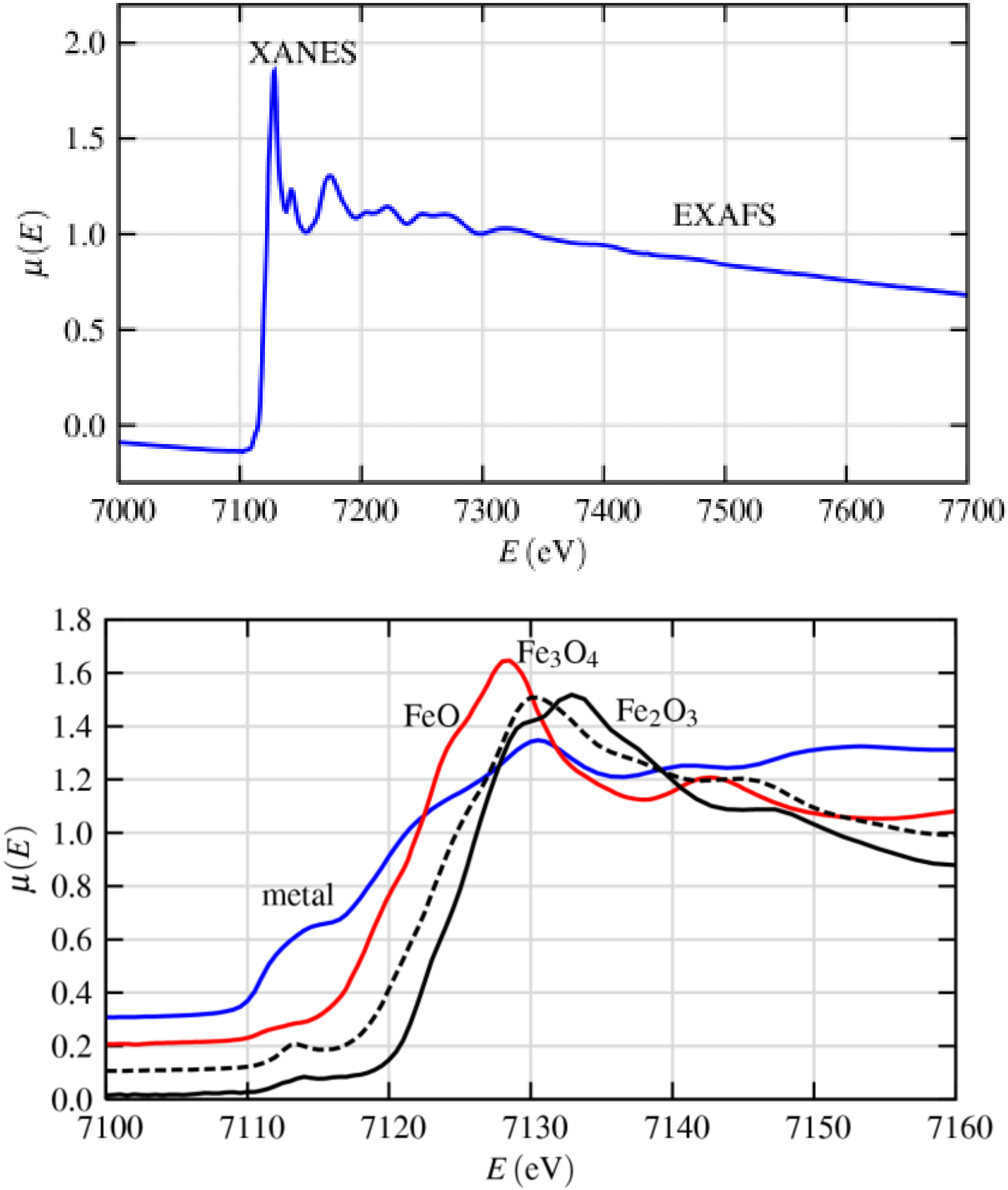
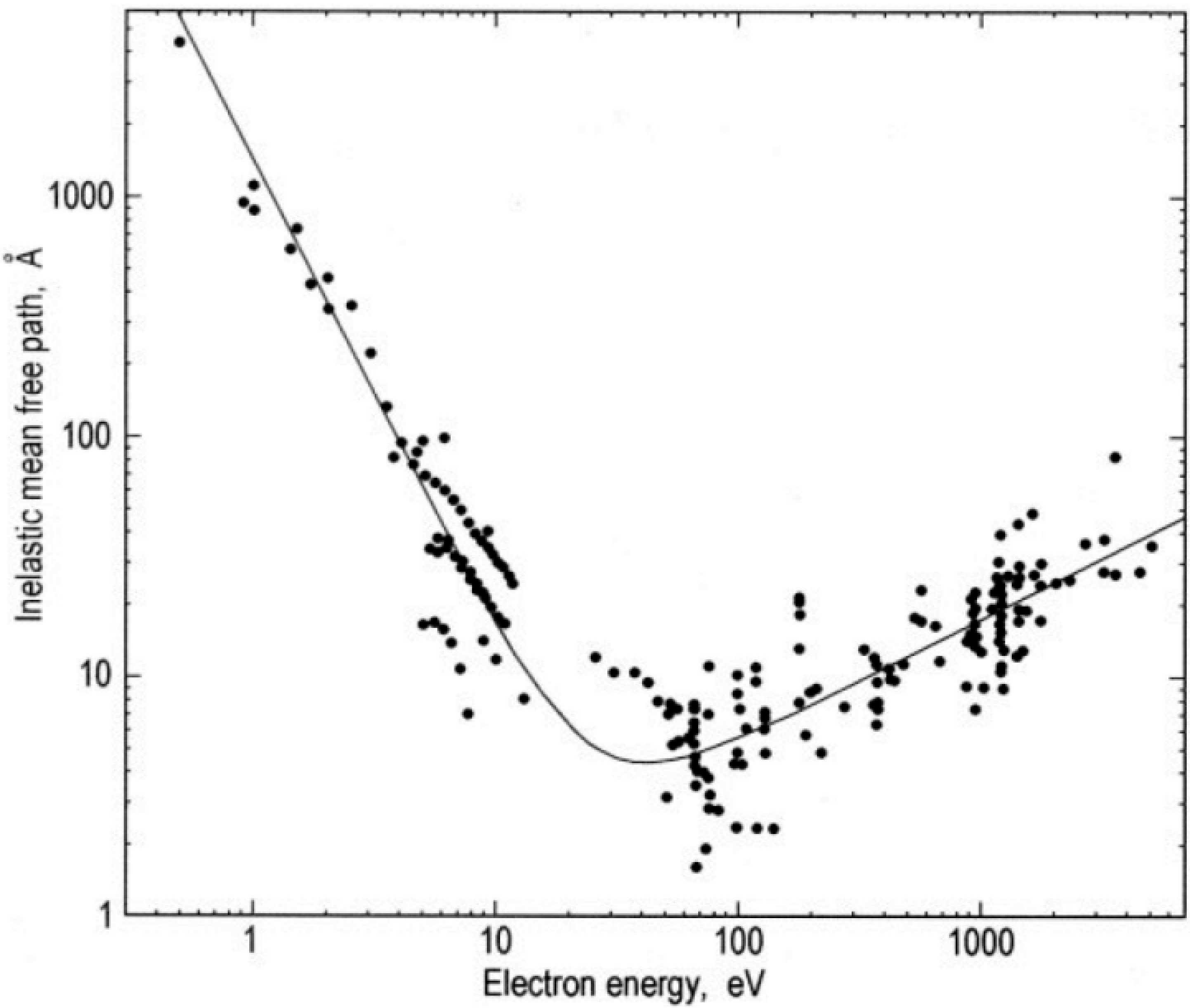
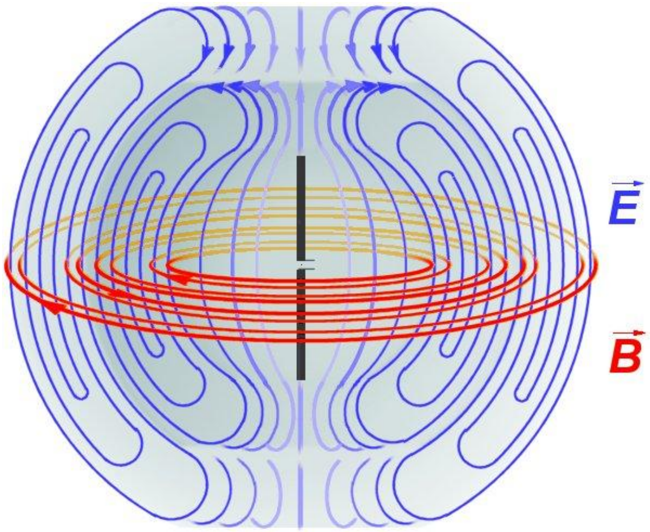


Figure 2.6: XAFS $\mu(E)$ for FeO. On top, the measured XAFS spectrum is shown with the XANES and EXAFS regions identified. On the bottom, $\mu(E)$

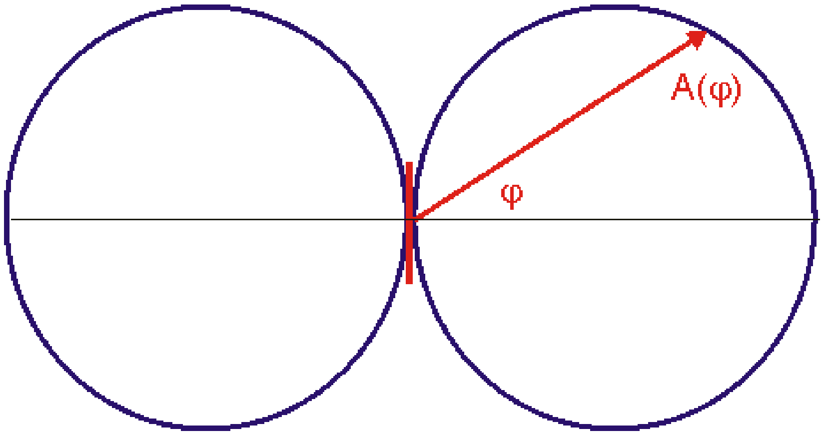
The Universal Curve for the Electron Mean Free Path



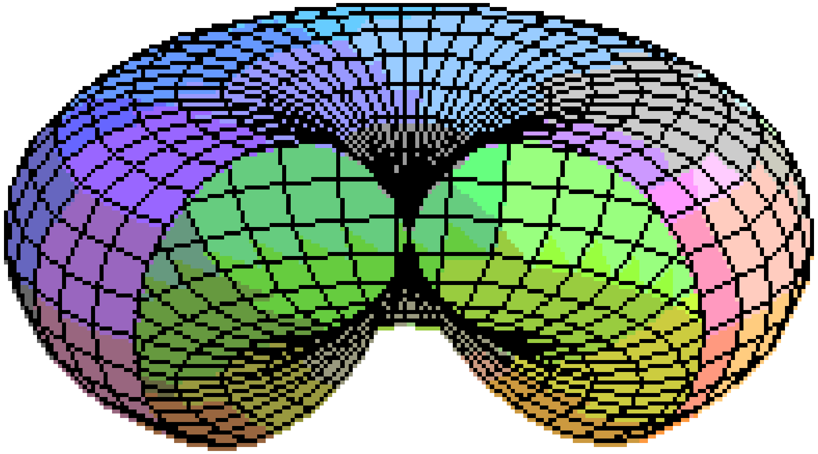
Dipole radiation



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



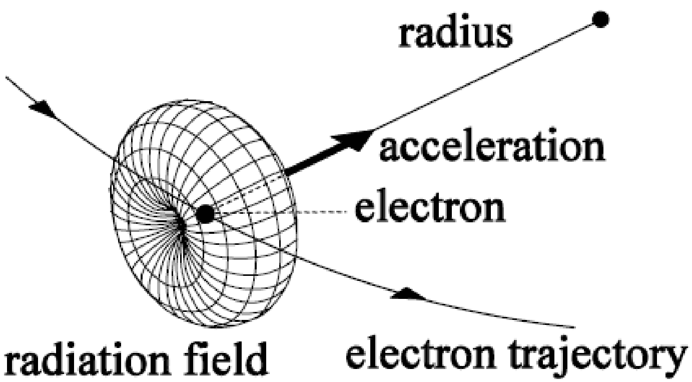
Emission characteristics (A=intensity)



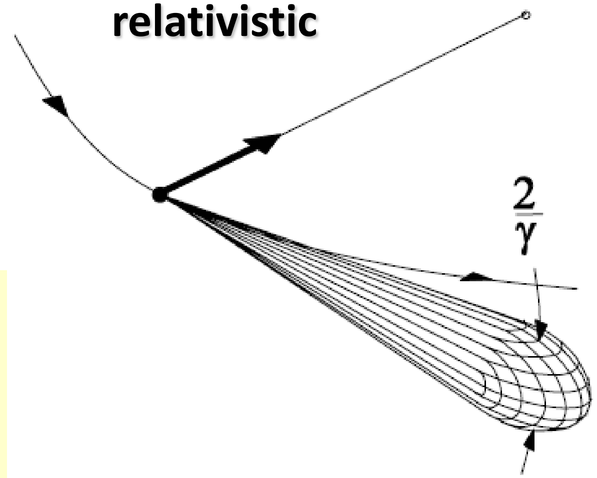
3D-view

Electrons on circular orbit

nonrelativistic



relativistic



opening angle ***θ***

Radiation Power *P*

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

nonrelativistic:

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

$\Rightarrow$ **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:

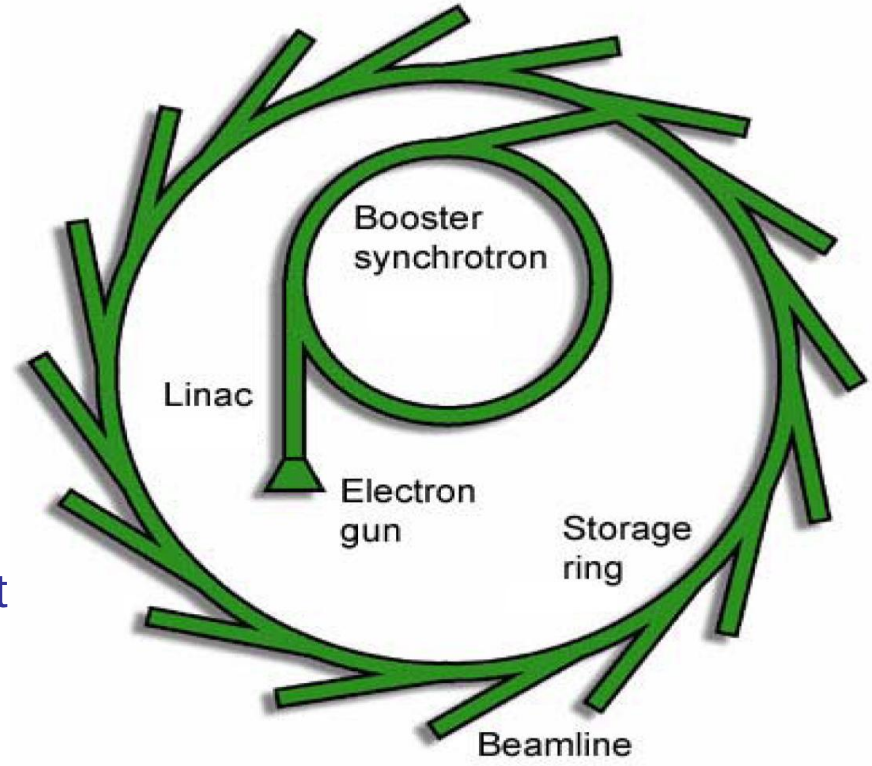
$\rightarrow v \approx c \rightarrow \beta \approx 1$

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

$\Rightarrow$ **extremely high radiation power, emitted in a sharp forward cone!**

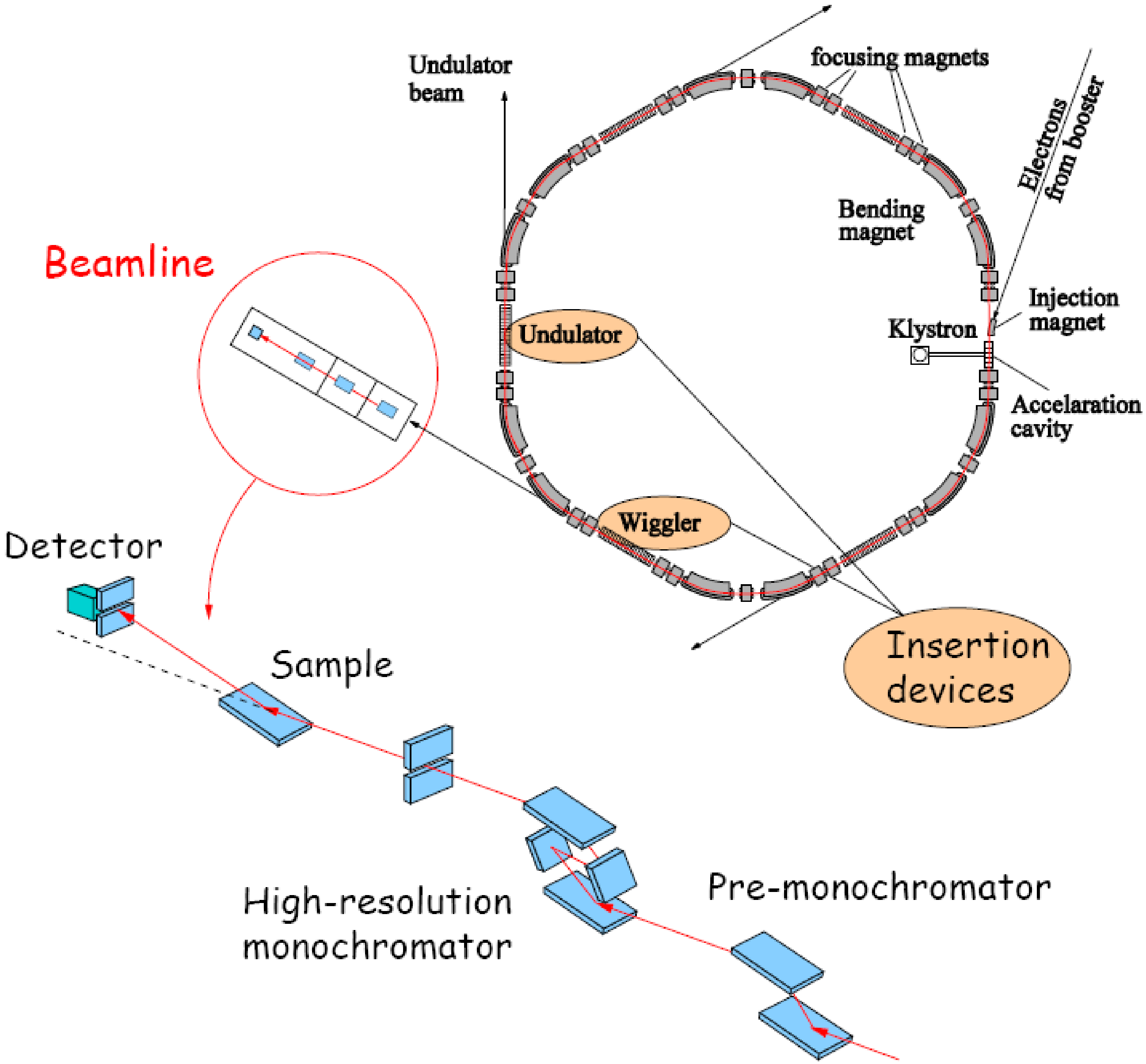
Generation of Synchrotron Radiation

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)
   $\rightarrow$ 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



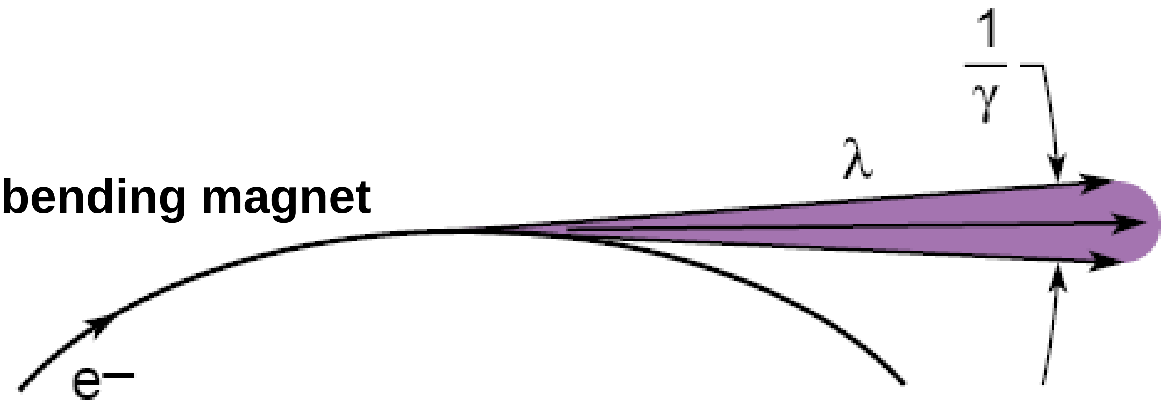
Velocity of relativistic electrons (6 GeV) *v* is only 107 cm/s slower than the velocity of light

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)

$\gamma = E/mc^2 \cong 1820$

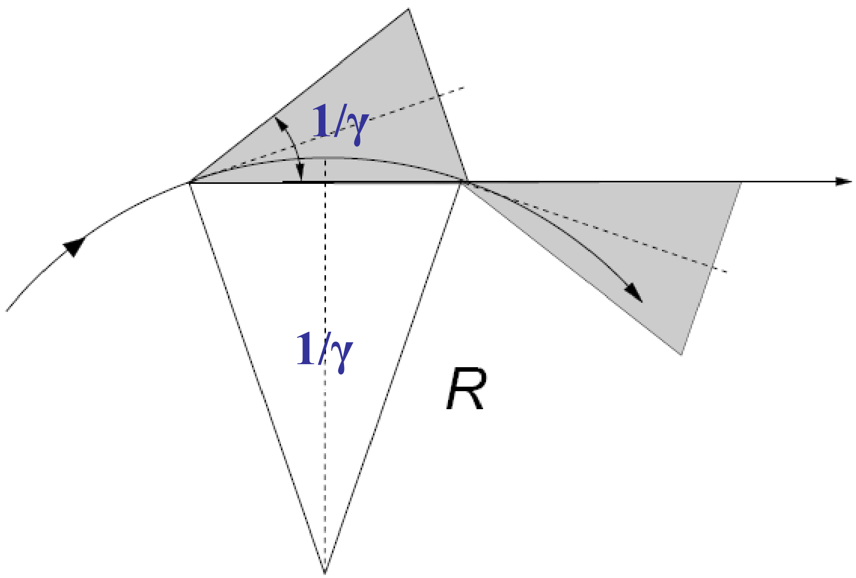
$\theta \approx 8 \times 10^{-5}$ rad (0.08 mrad)

The emitted radiation is a sharp cone with an opening angle $\theta \approx 0.08$ mrad

$\Rightarrow$ Excellent collimation!

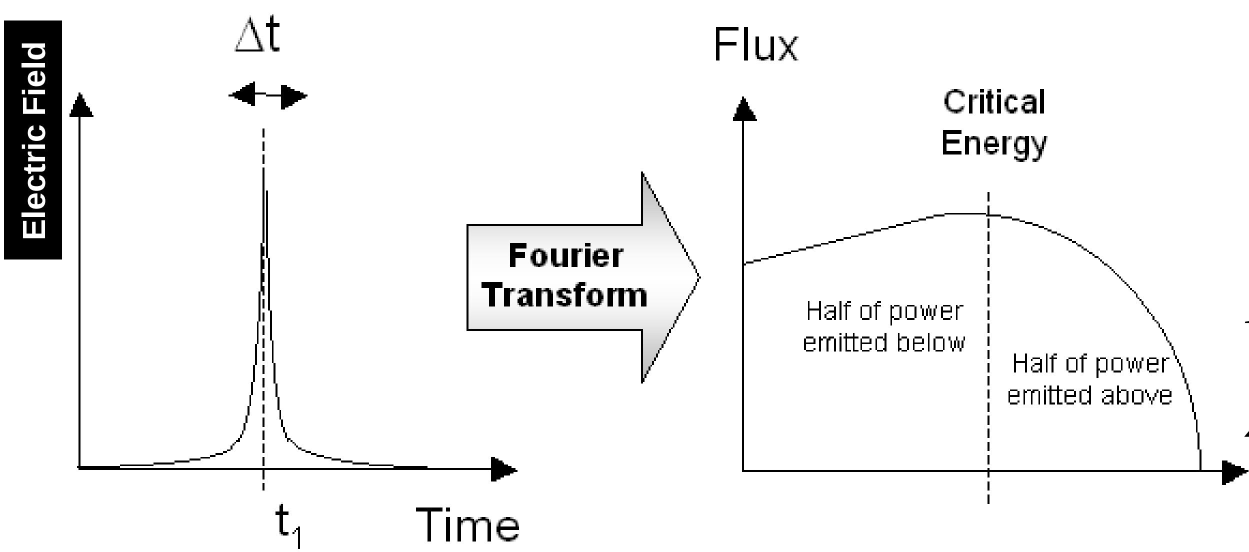
$\Rightarrow$ in a distance of 50 m from the source, one obtains a spot of only ~ 4 mm!

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

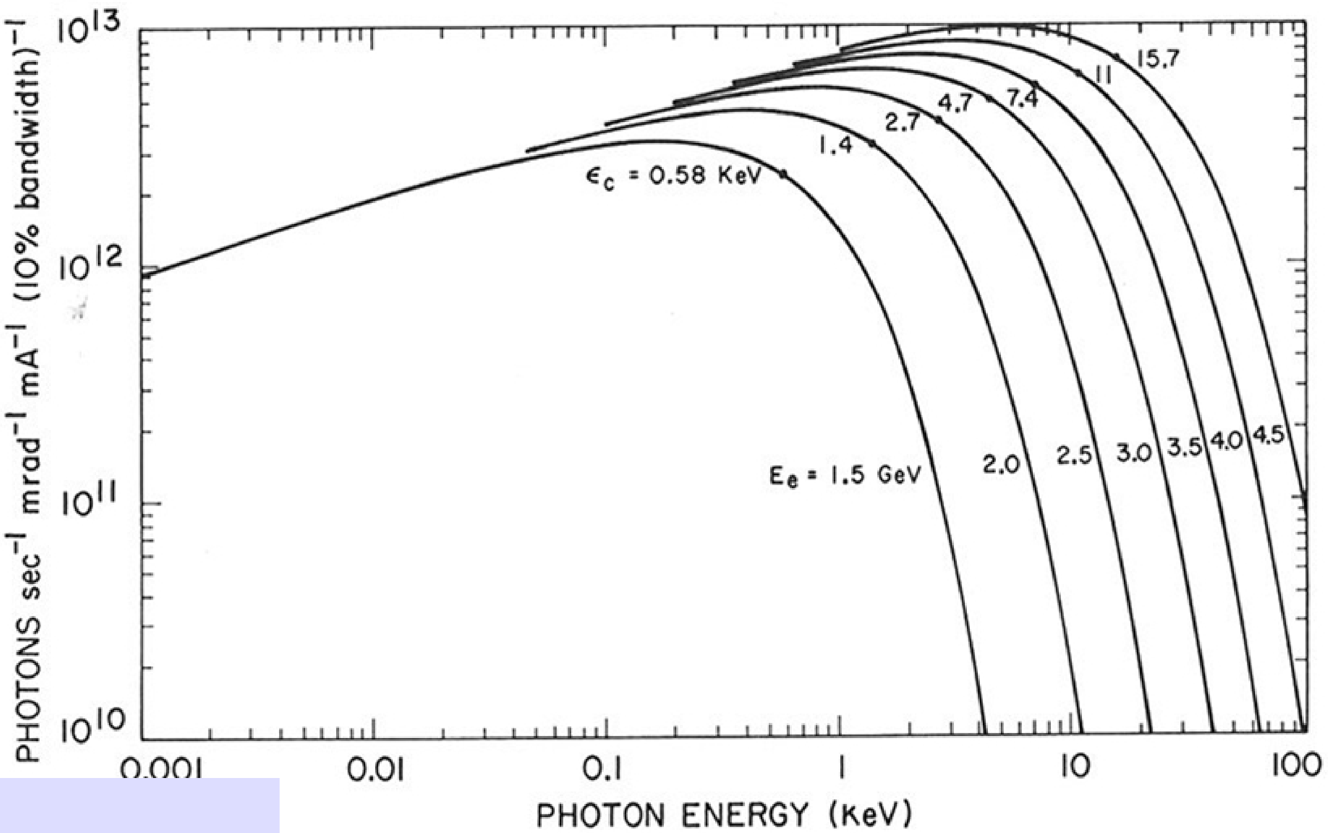
$$\text{Total flux} \equiv \frac{\text{Photons}}{\text{s}}$$

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

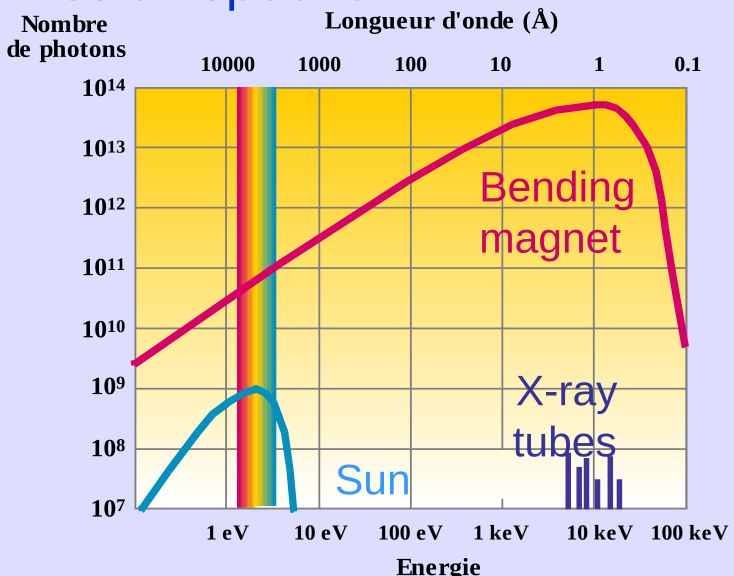
$$\text{Brightness} = \frac{\text{Total flux}}{\text{solid angle} \cdot 0.1\%\text{bandwidth}} \quad \left[\frac{\text{Photons/s}}{\text{mrad}^2 \cdot 0.1\%\text{bandwidth}}\right]$$

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

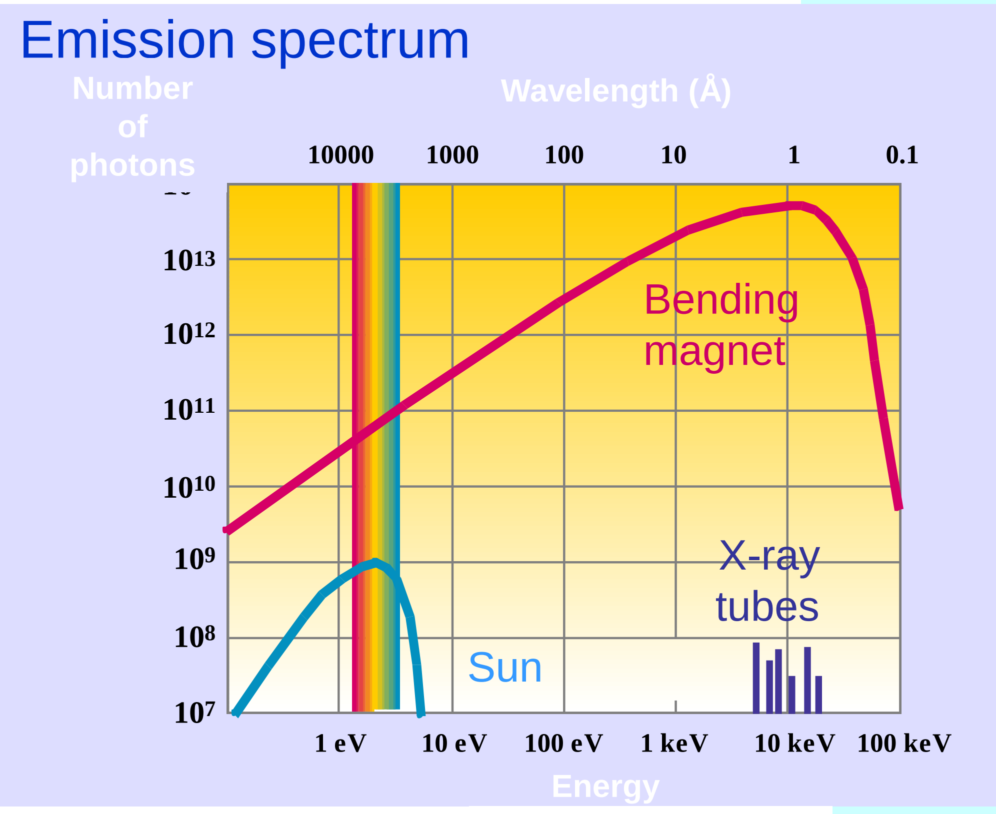
Brilliance is the figure of merit for the design of new Synchrotron Radiation sources

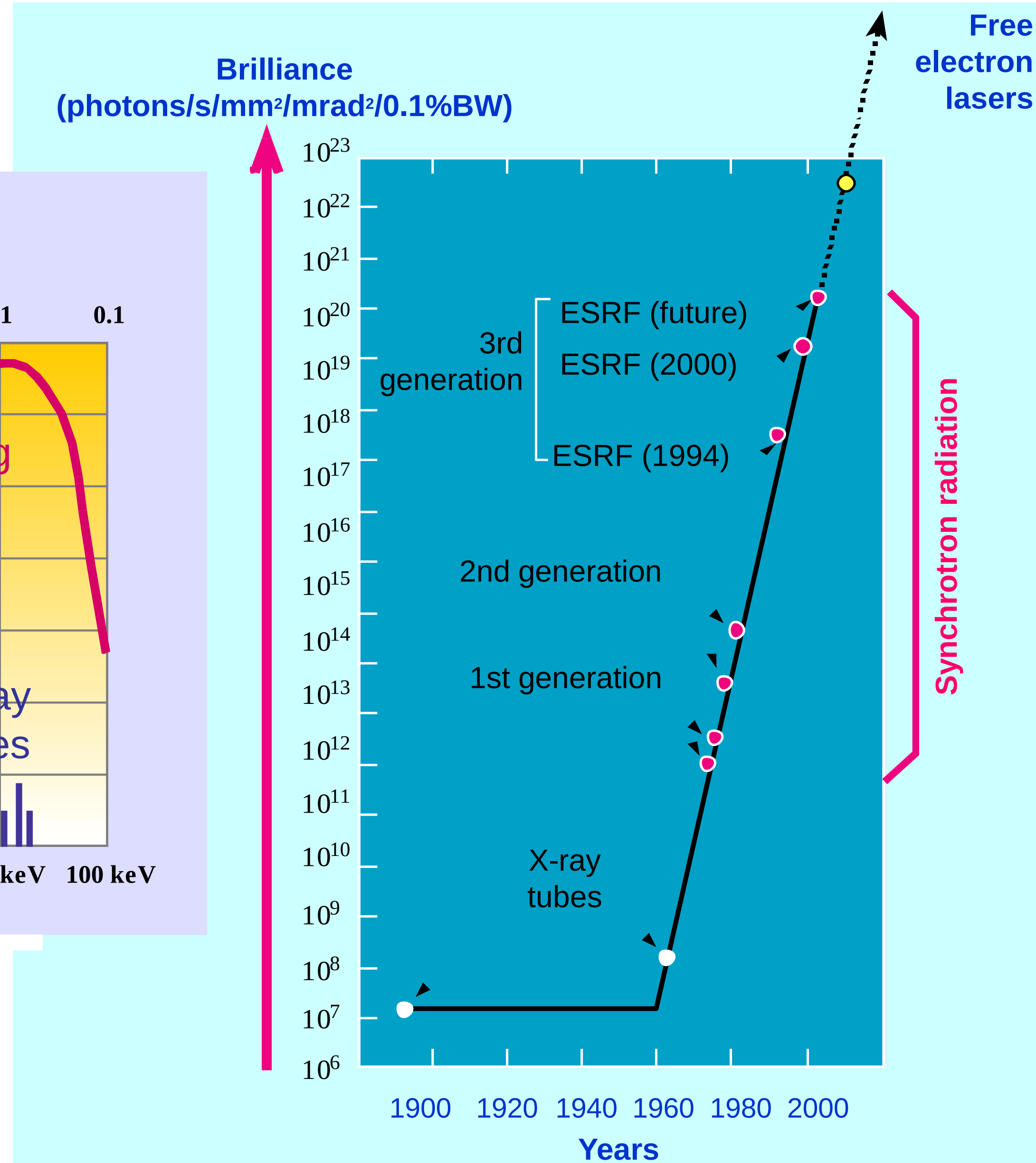


Emission spectrum



Extremely high intensity, broad energy range

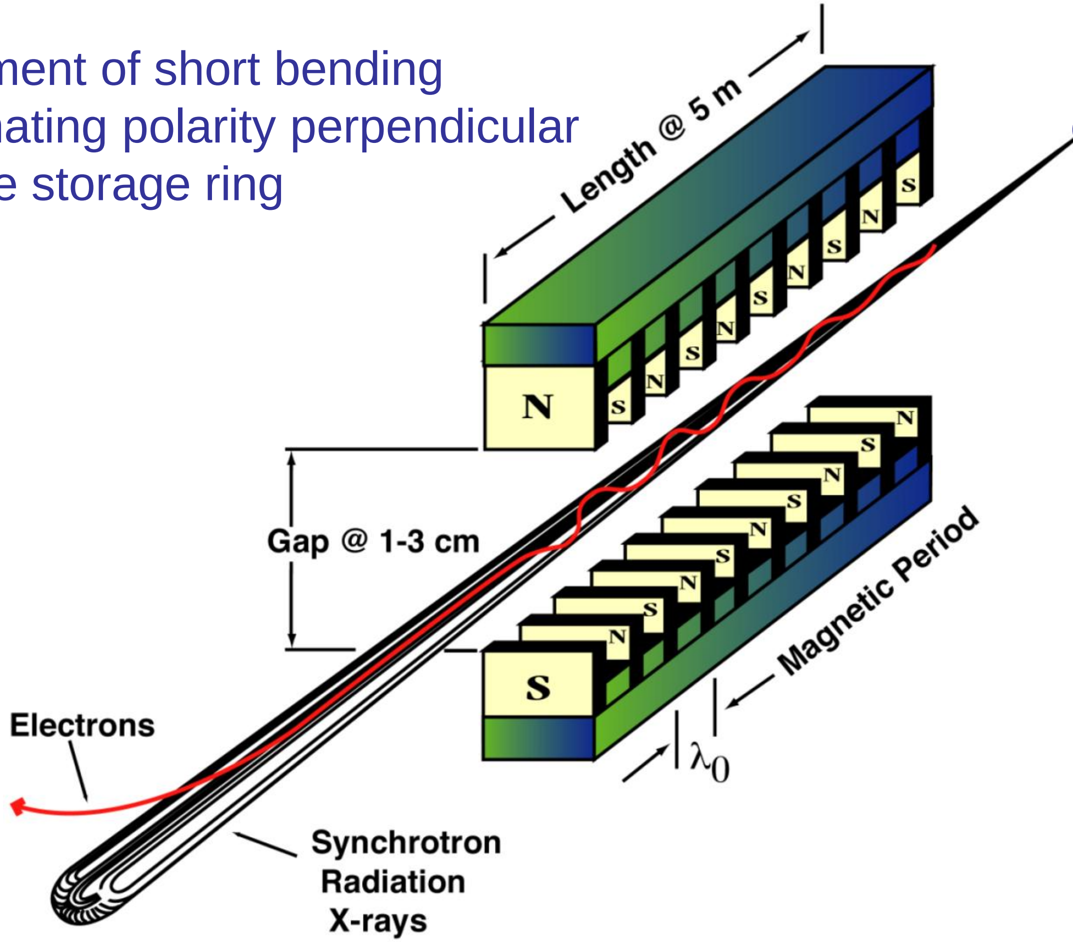




Magnetic wigglers and undulator (N periods)

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



$\Rightarrow$ force the electrons to oscillate („wiggle“) perpendicular to their direction of motion

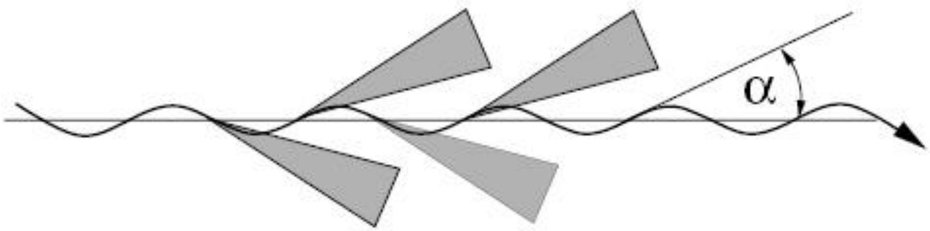
$\Rightarrow$ Radiation is emitted during each individual wiggle

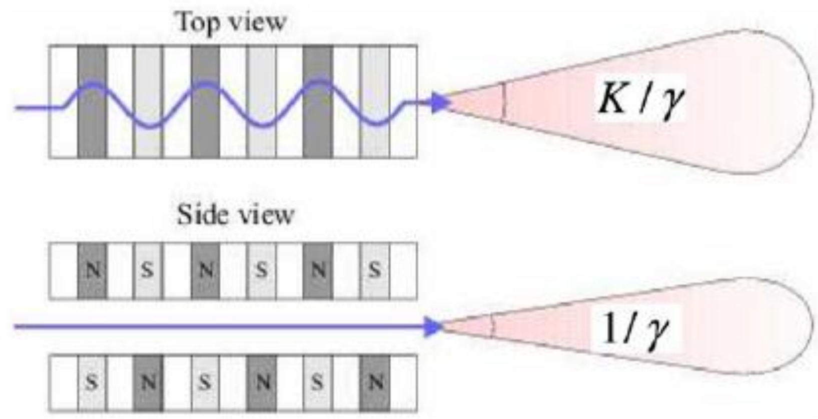
$\Rightarrow$ increase of the intensity

wiggler and undulator

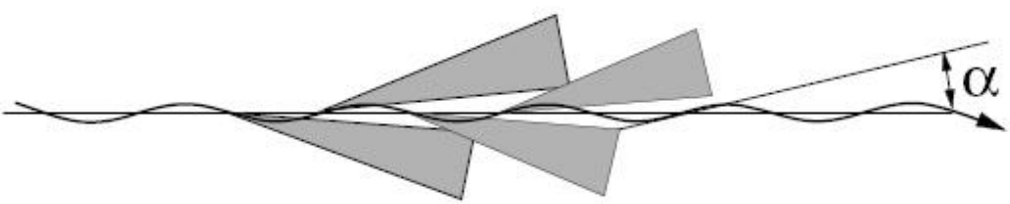
$$K := \alpha \cdot \gamma = \frac{e\,B_0\,\lambda_0}{2\pi\, m_e\, c}$$

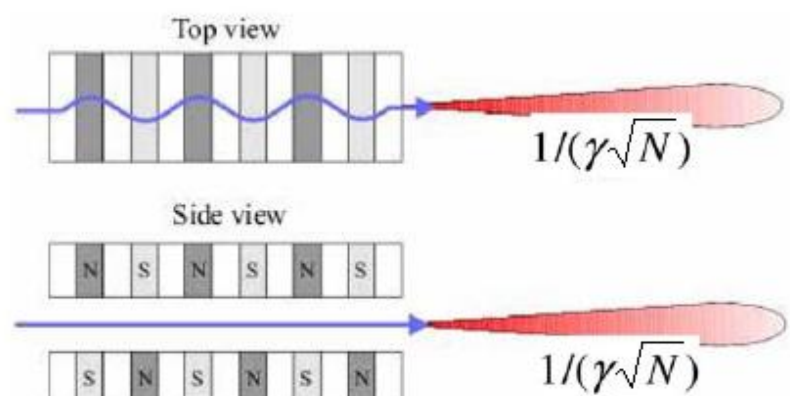
Wiggler regime: α > 1/γ





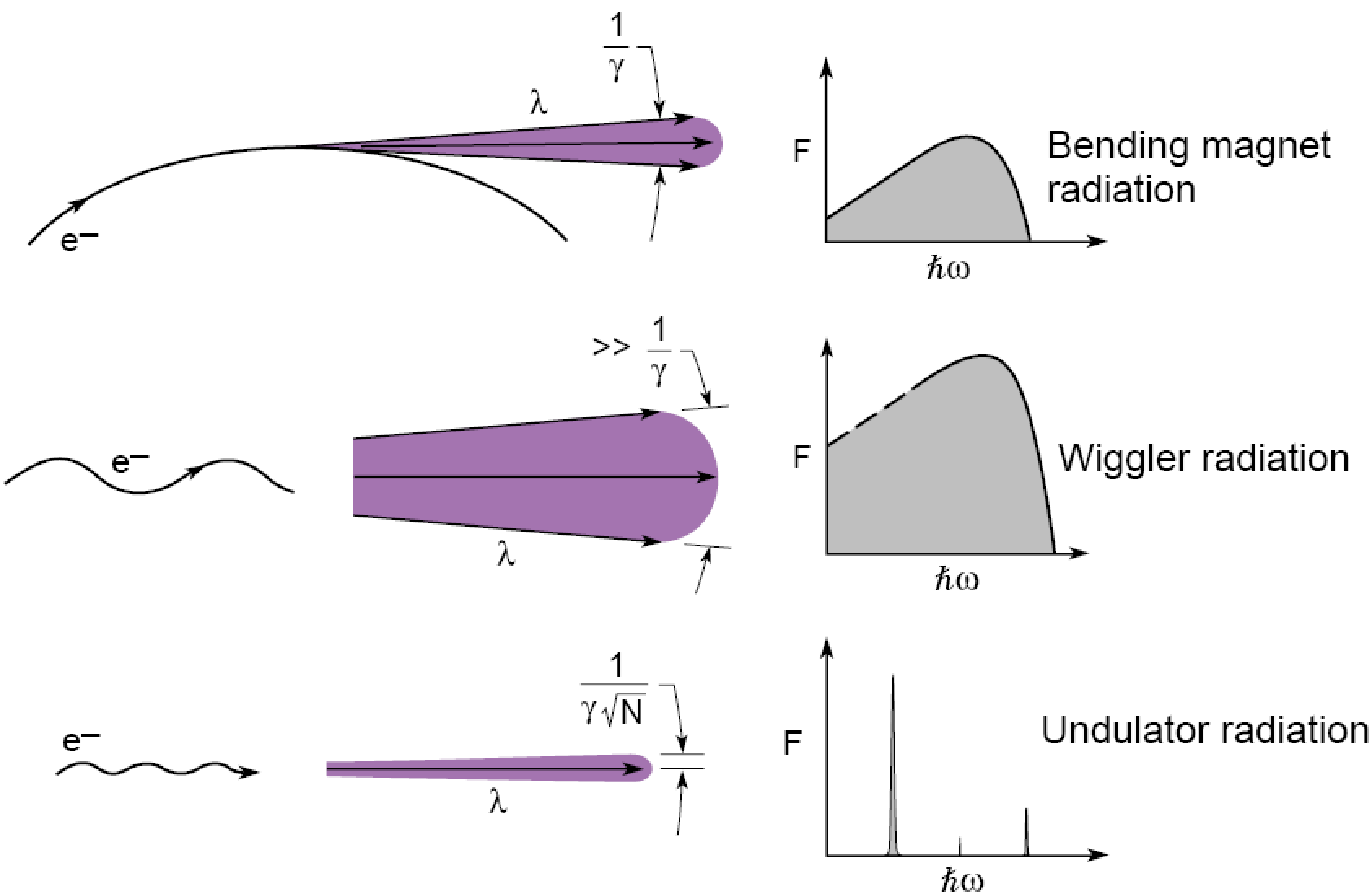
Undulator regime: α ~ 1/γ



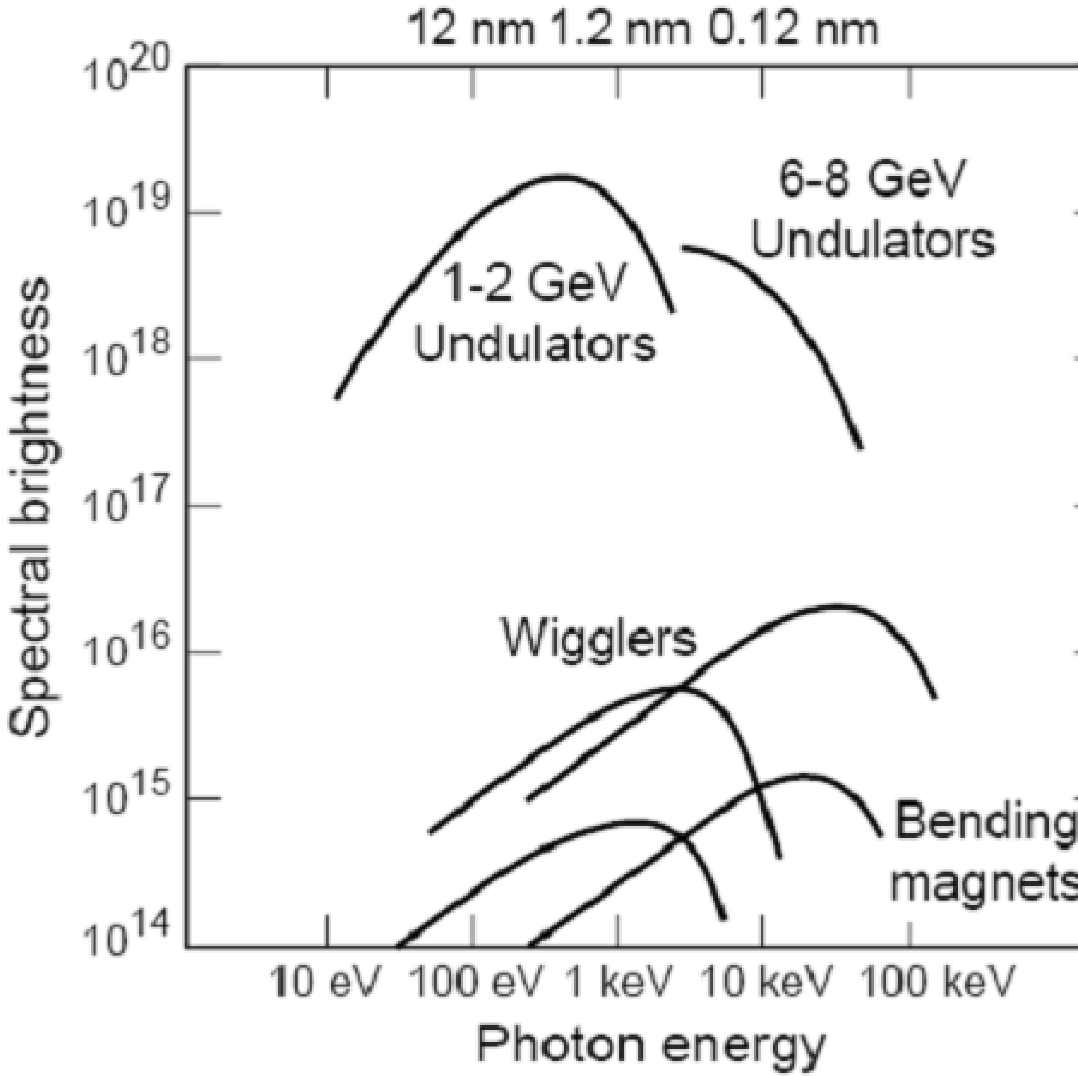


In the undulator regime the radiation cones overlap and the wave trains can interfere

Forms of Synchrotron Radiation

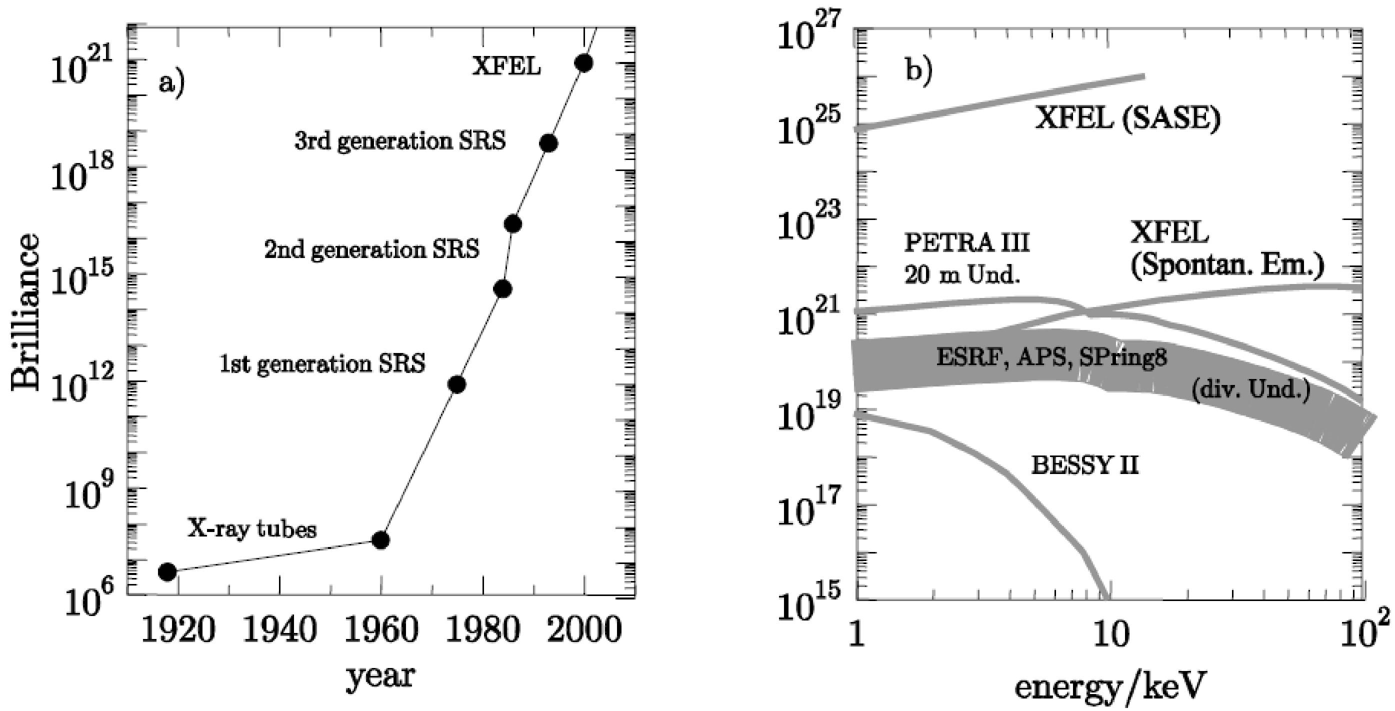


Spectral Brightness



Examples of Wigglers and Undulators

Evolution of Brilliance



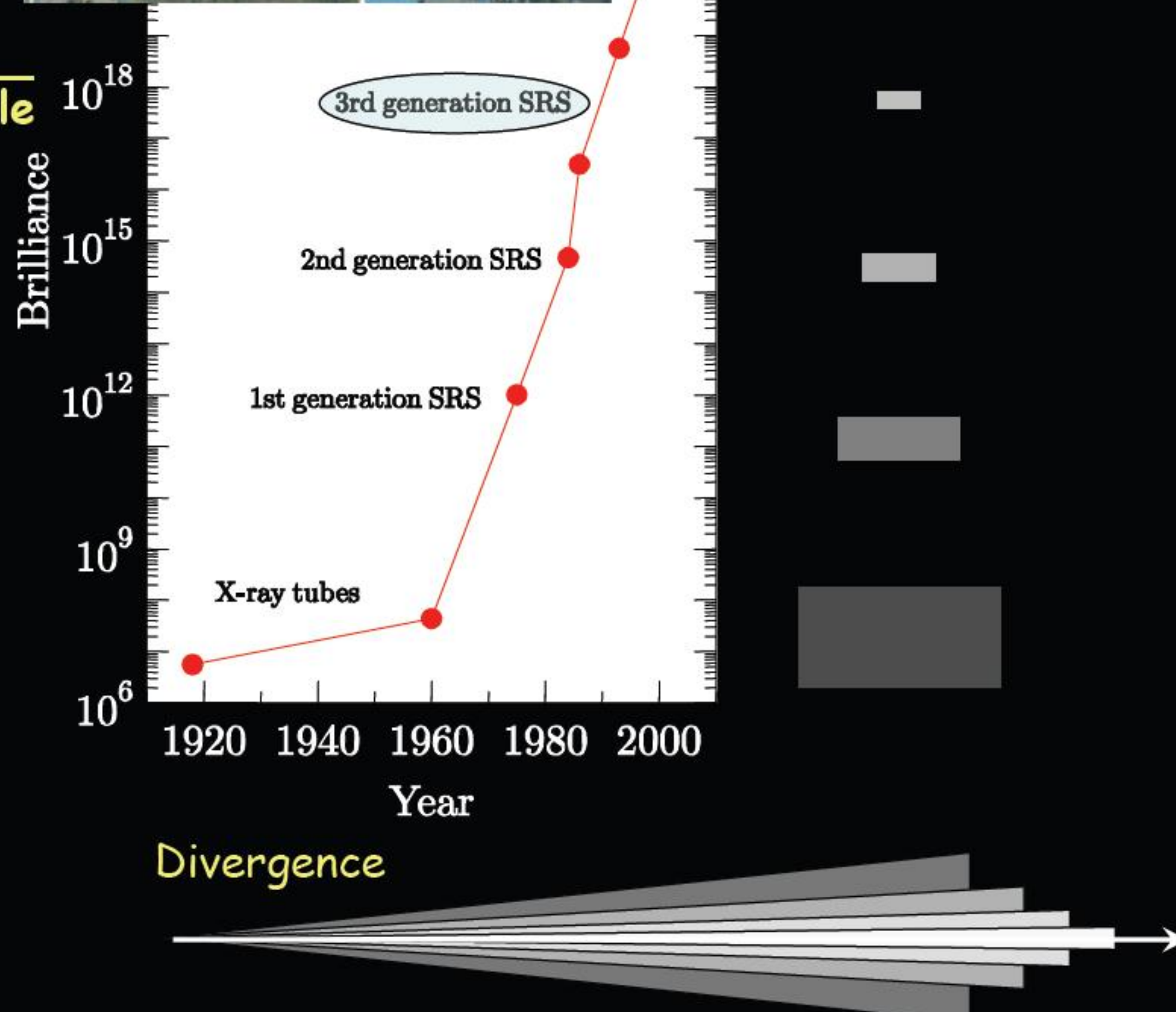
1^{st} generation: Exploitation of the light from the bending magnets of e+/e- colliders originally built for elementary particle physics

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

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

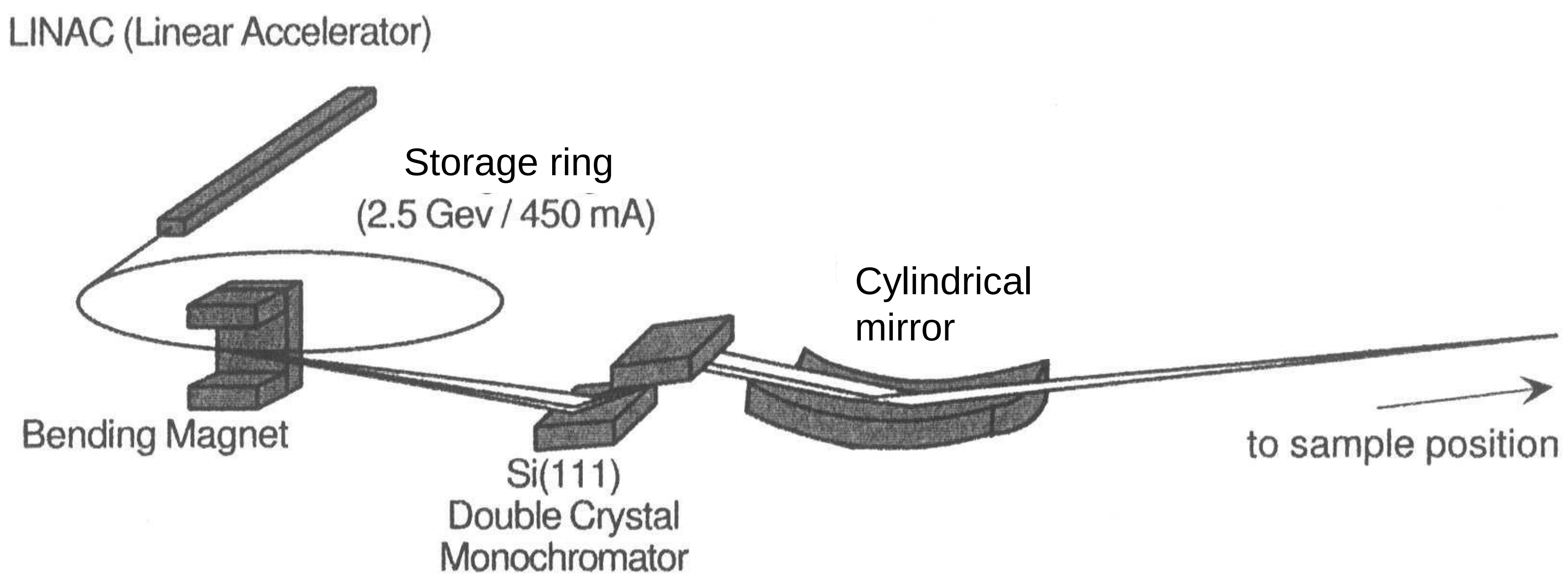
Evolution of Source Brilliance

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

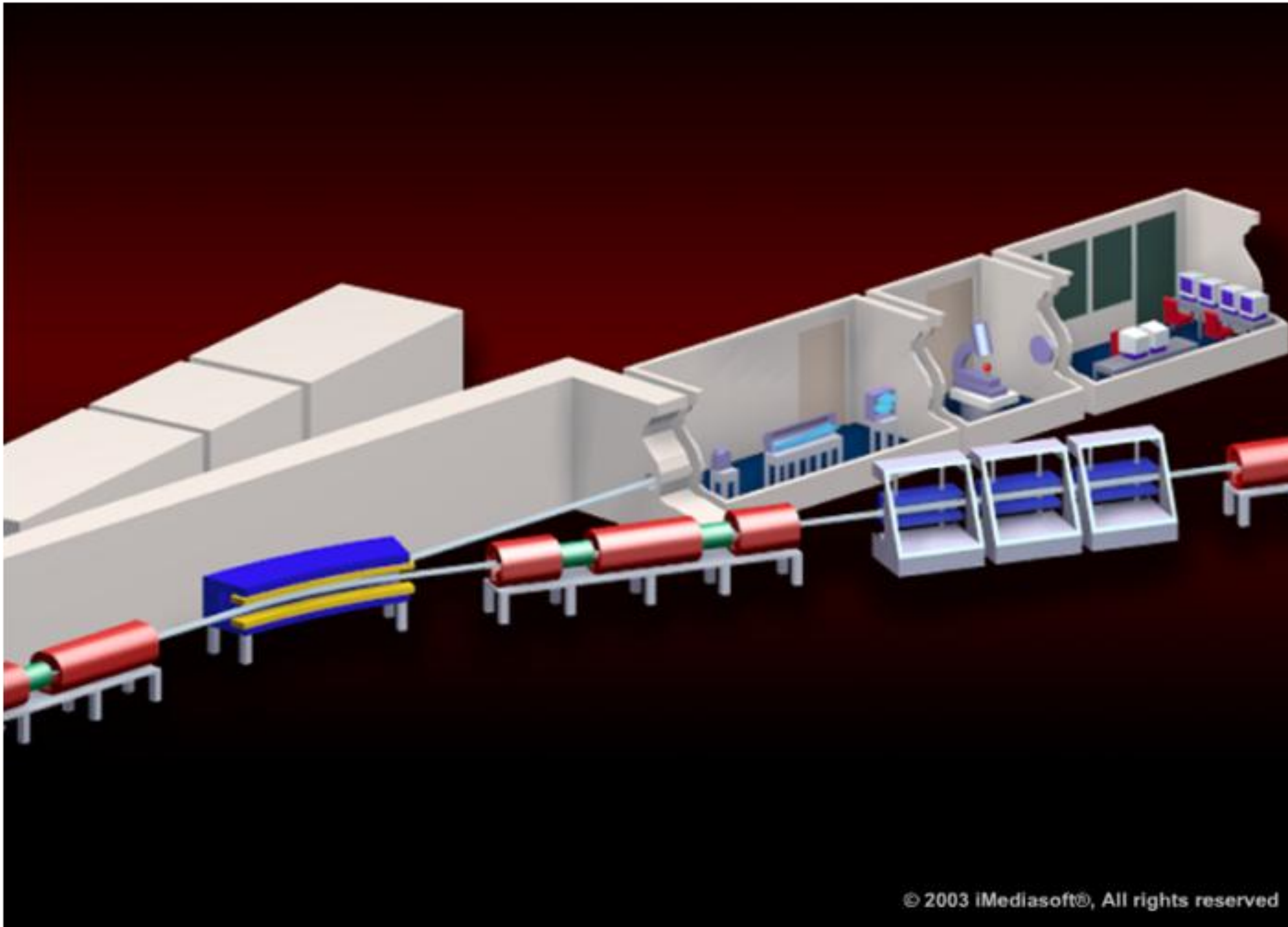


European Synchrotron Radiation Facility (ESRF)

Beamline organization



How does a beamline work?

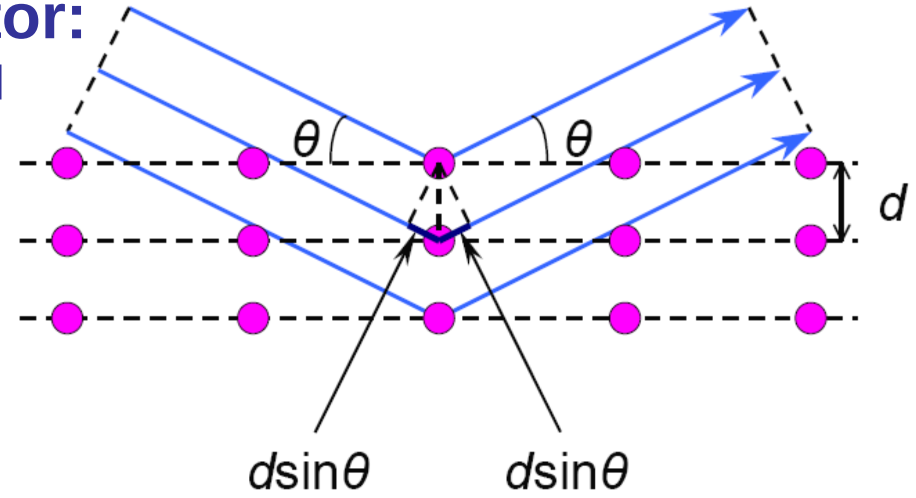


X-ray monochromator using perfect crystal

Basic principle of monochromator:

Bragg reflection from perfect single crystal

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



Energy range of standard monochromator

Bragg Reflection

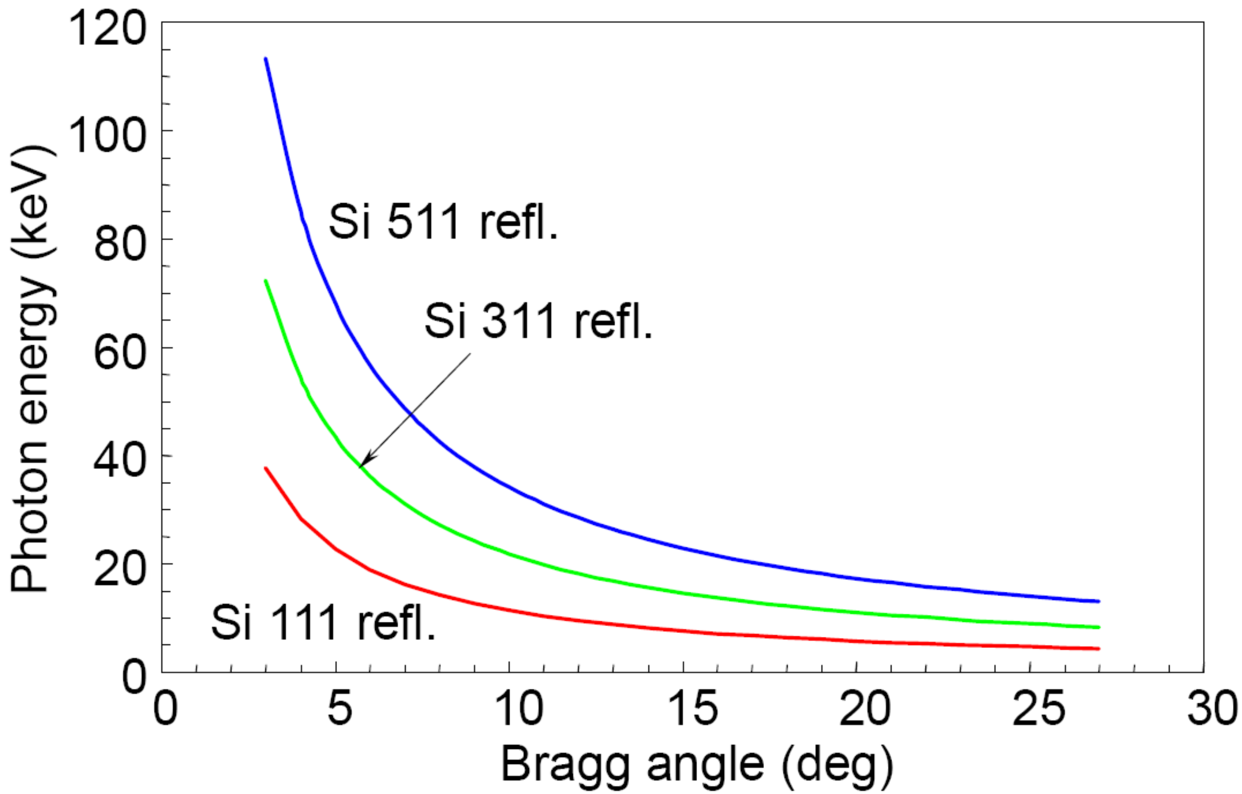
Si 111
Si 311
Si 511
.....

Bragg angles

3~27°

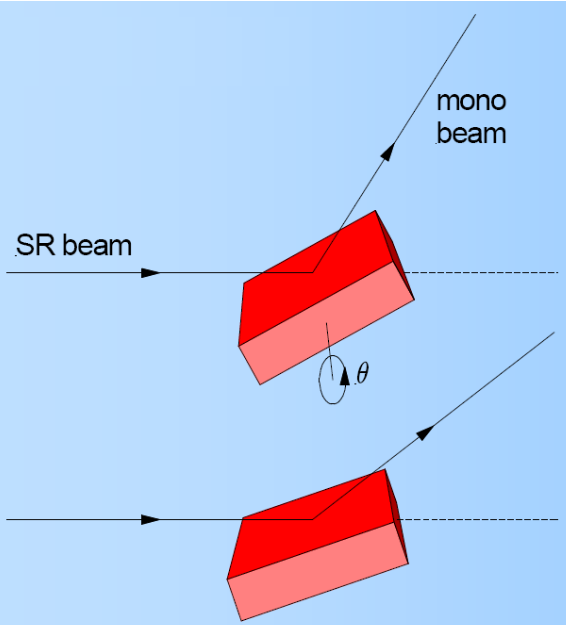
Energy range

4.4~110 keV



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

Double crystal monochromator

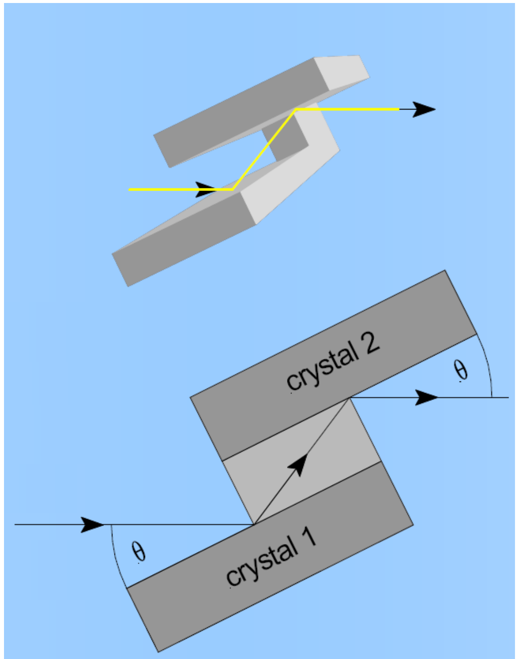


Problems with single crystal monochromators

- 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**)

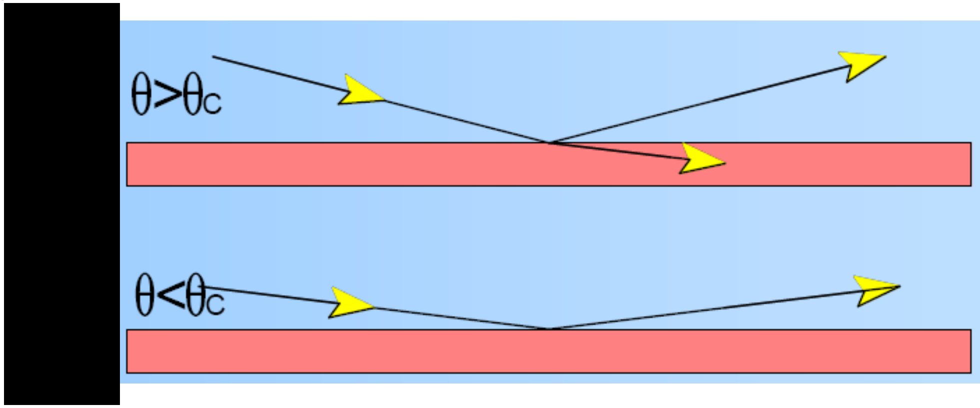
- **Use the same crystals and *d*-spacing for 1st and 2nd crystals**
- **Keep parallel setting**



X-ray Mirrors

reflectivity at grazing angles:

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

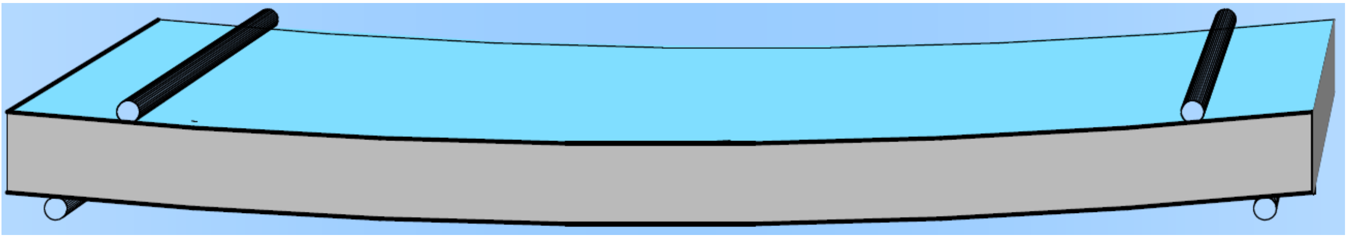


By Snell's law ($n_1 \cos(\theta_1) = n_2 \cos(\theta_2)$) with θ the grazing angle) in the absence of absorption (total reflection), we find total external reflection for angles less than $\theta_c \approx \lambda(r_0\,\rho / \pi)^{1/2}$

θ_c typically a few mrad for x-ray mirrors

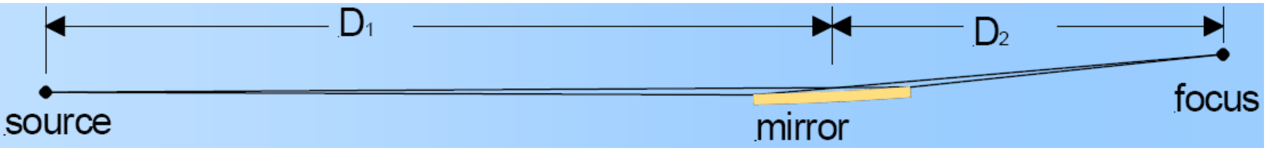
Surface roughness must be considered around critical energy (angle).

Bent mirrors (focusing and collimating)



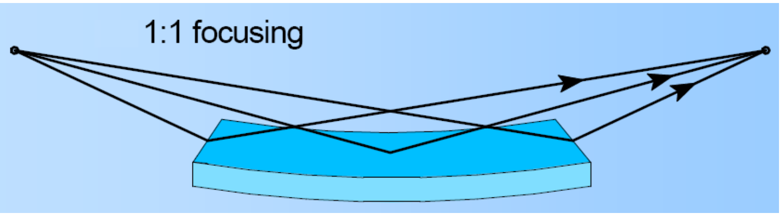
Focusing of the x-ray beam → reflecting surface must have some curvature (achieved e.g. by bending mirror, **mirror focuses in one plane only**!)

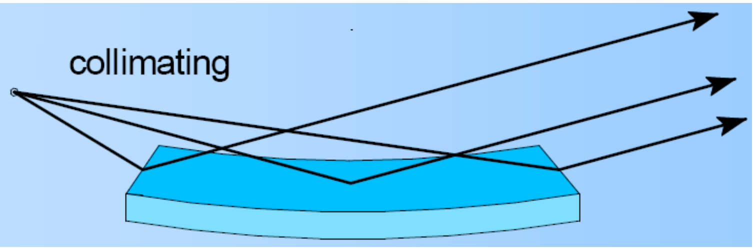
Bending radius R (can be ~ 10 km)



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

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)

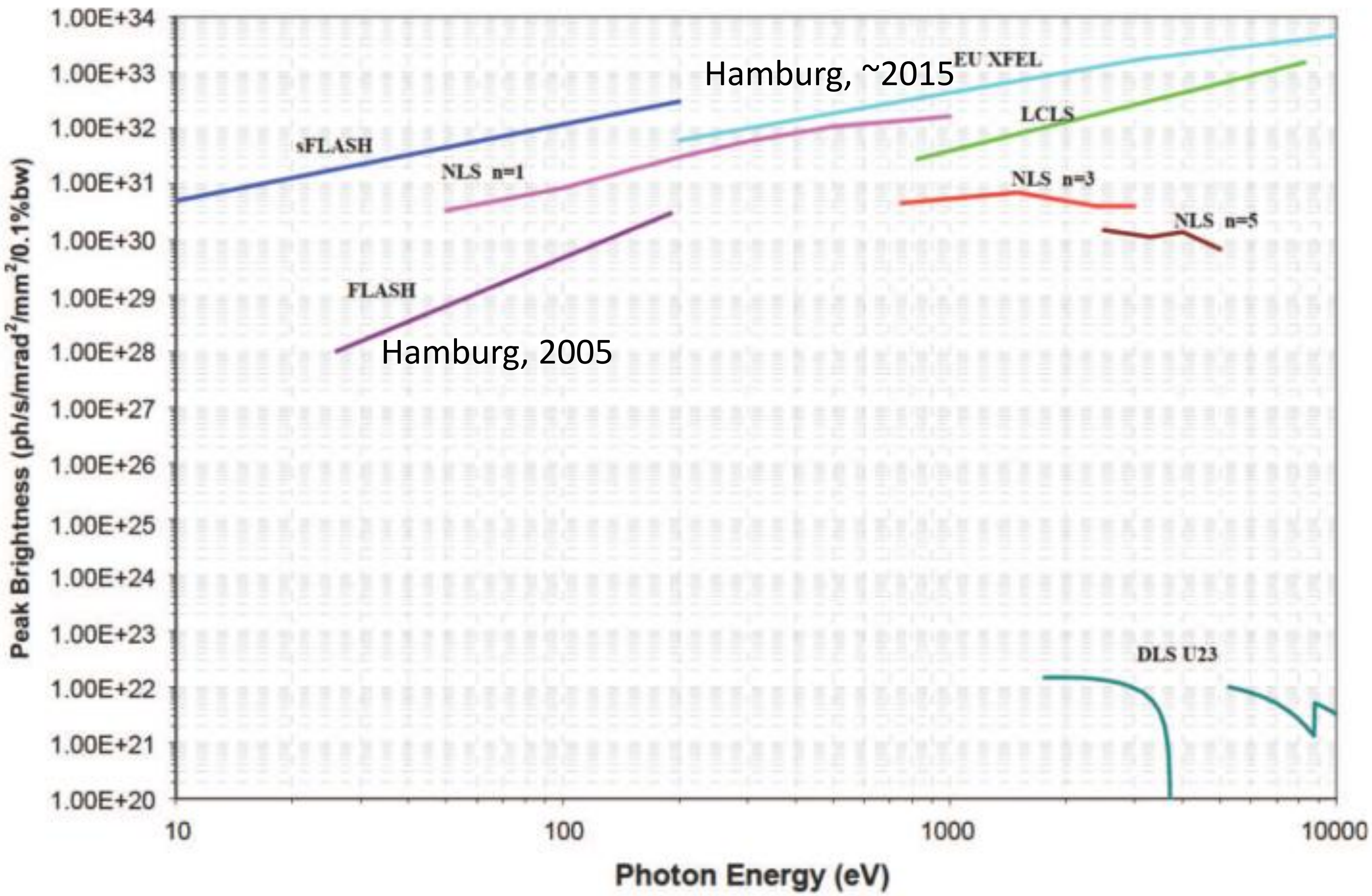


Figure 1. Comparison of several recently commissioned FELs (FLASH and LCLS) and several planned FELs (sFLASH, Euro XFEL, NLS) with a state-of-the-art undulator beamline on the Diamond Light source. The standard definition of brightness is given in photons/unit time/unit solid angle/unit area/normalised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

SASE – spontaneous amplified self-emission

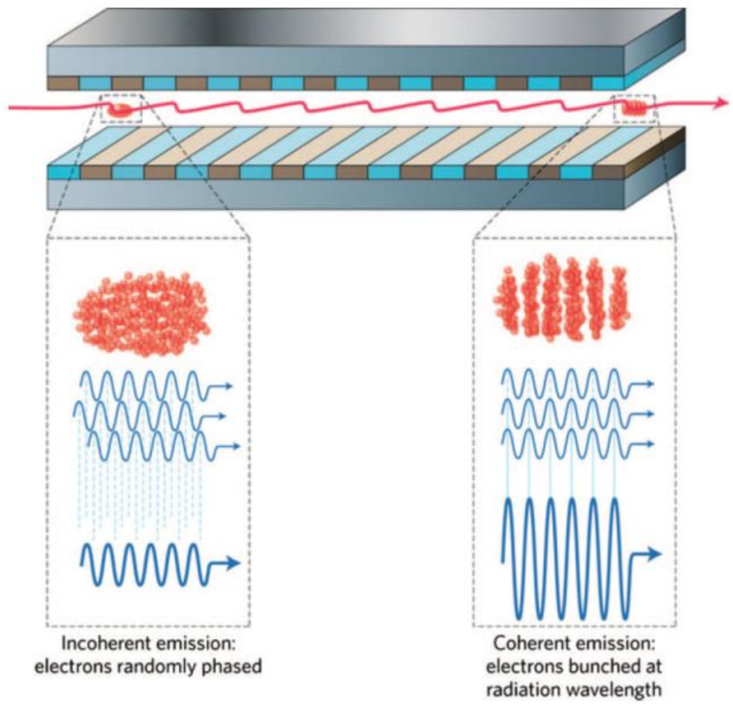
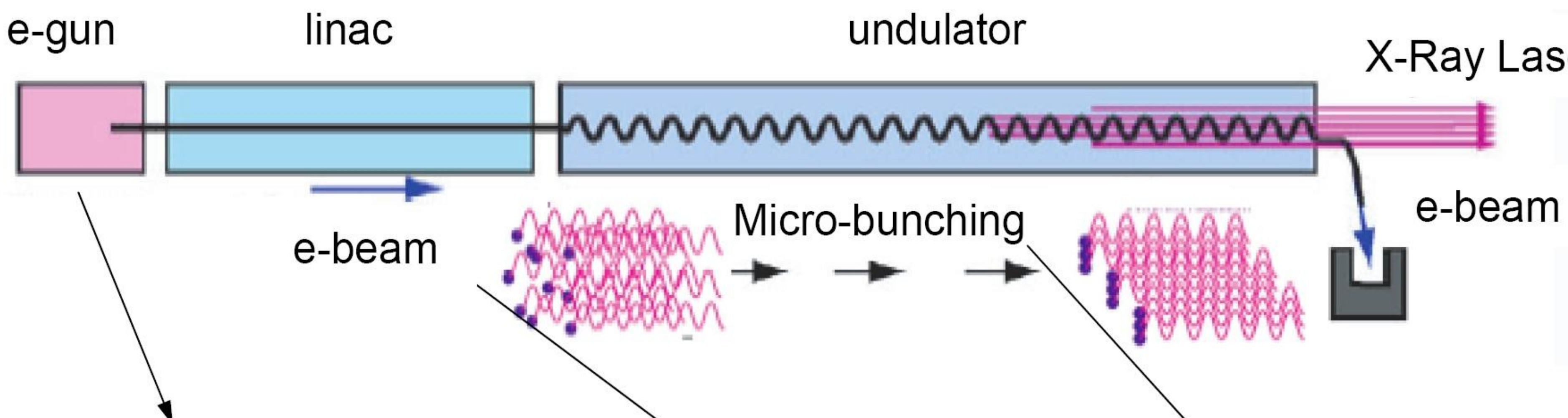


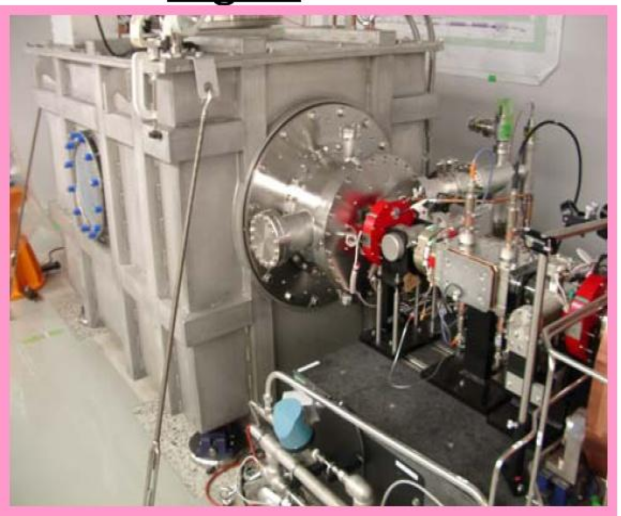
Figure 3. Schematic of SASE operation where the oscillation induced in the electron beam by the periodic magnetic field leads to radiation emission and, at the end of a sufficiently long undulator, self-organisation of the electrons gives rise to coherent X-ray emission. Reprinted by permission from 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)

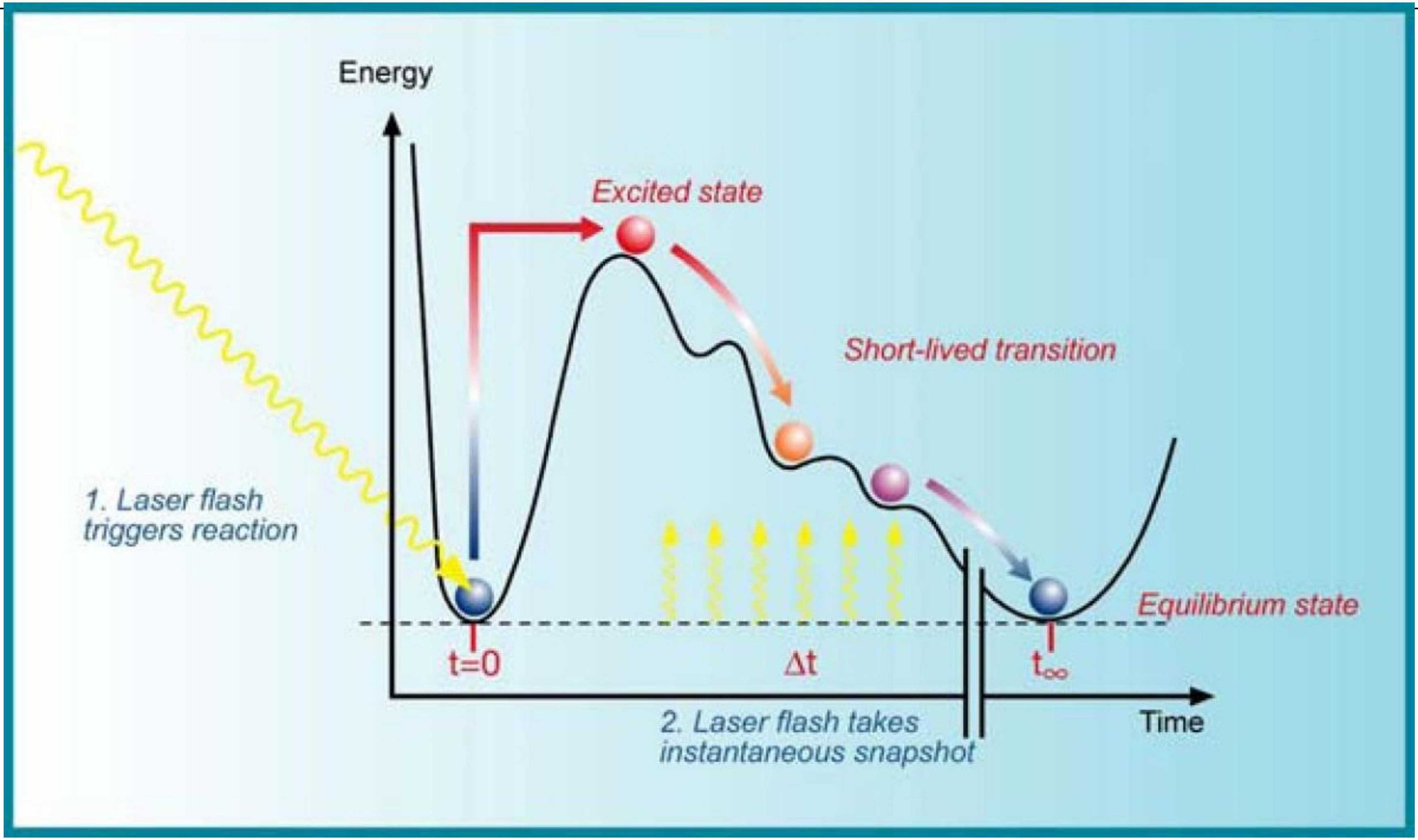
Principle design (SPring-8, Japan):



◆ e-gun (pulsed) ◆ linac (linear accelerator) ◆ undulator

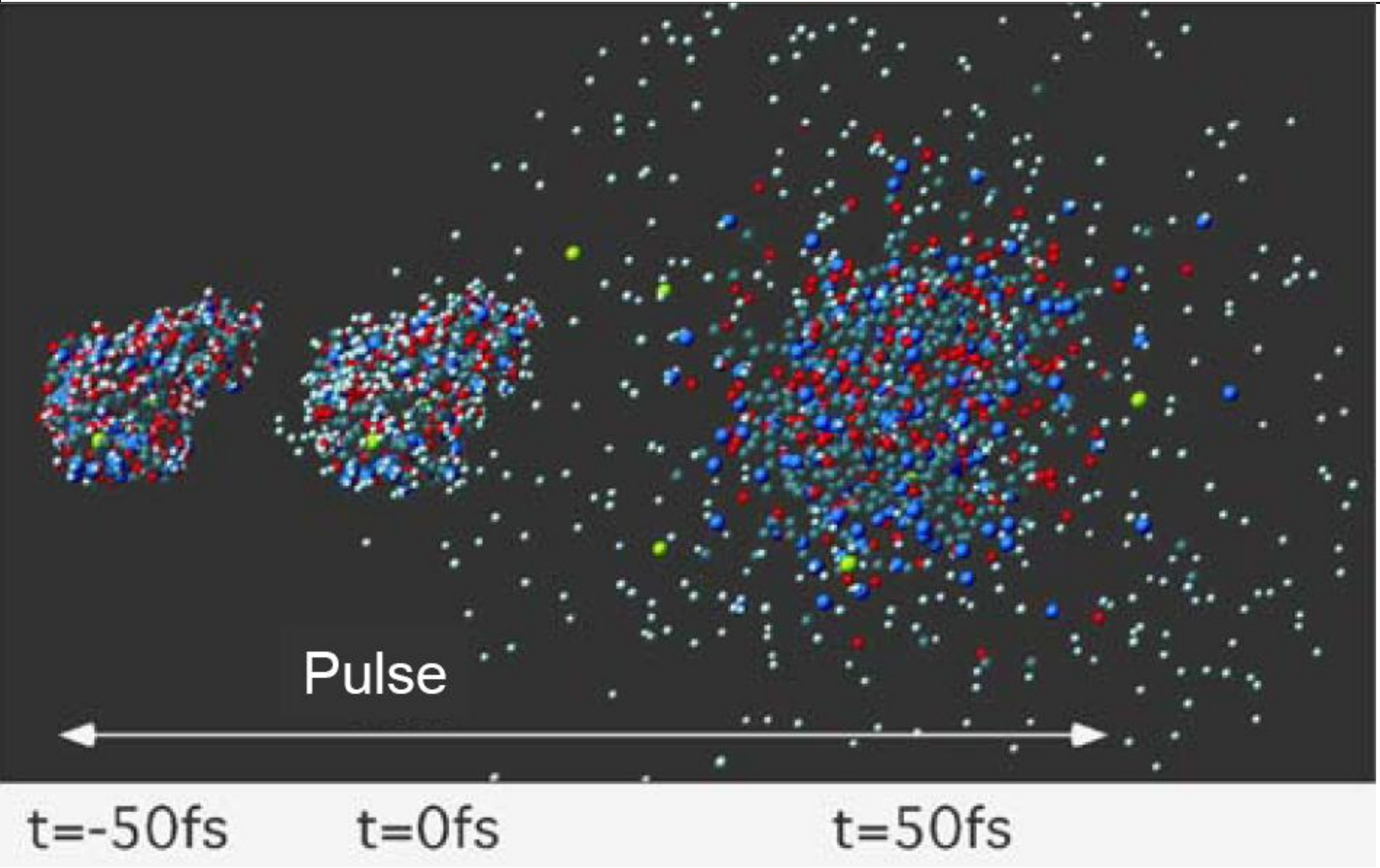


Pump-probe experiment



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

Obstacle: Coulomb-Explosion

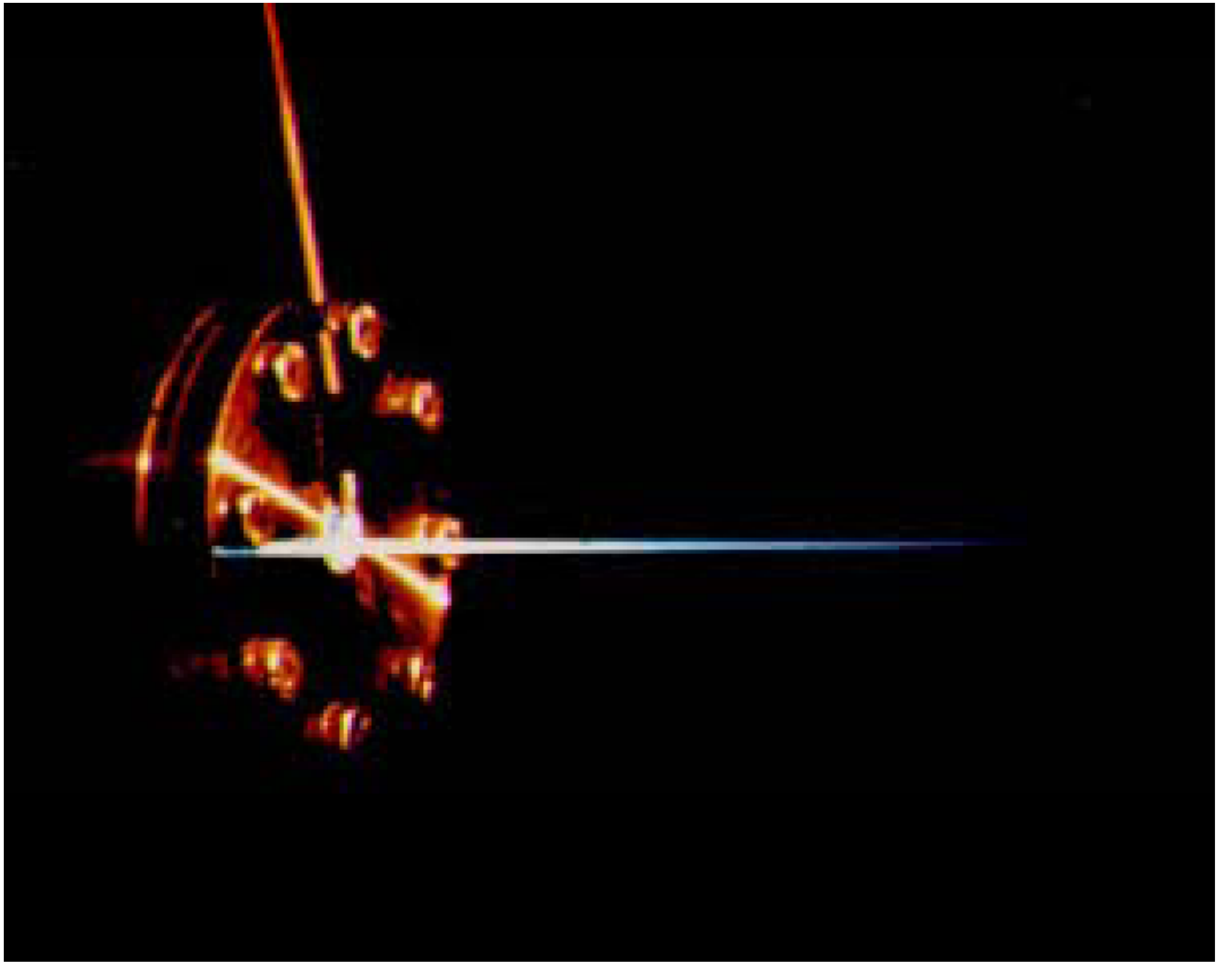


<u>Example:</u>
Lysozyme
white: Hydrogen,
grey: Carbon,
blue: Nitrogen,
red: Oxygen,
yellow: Sulfur

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.

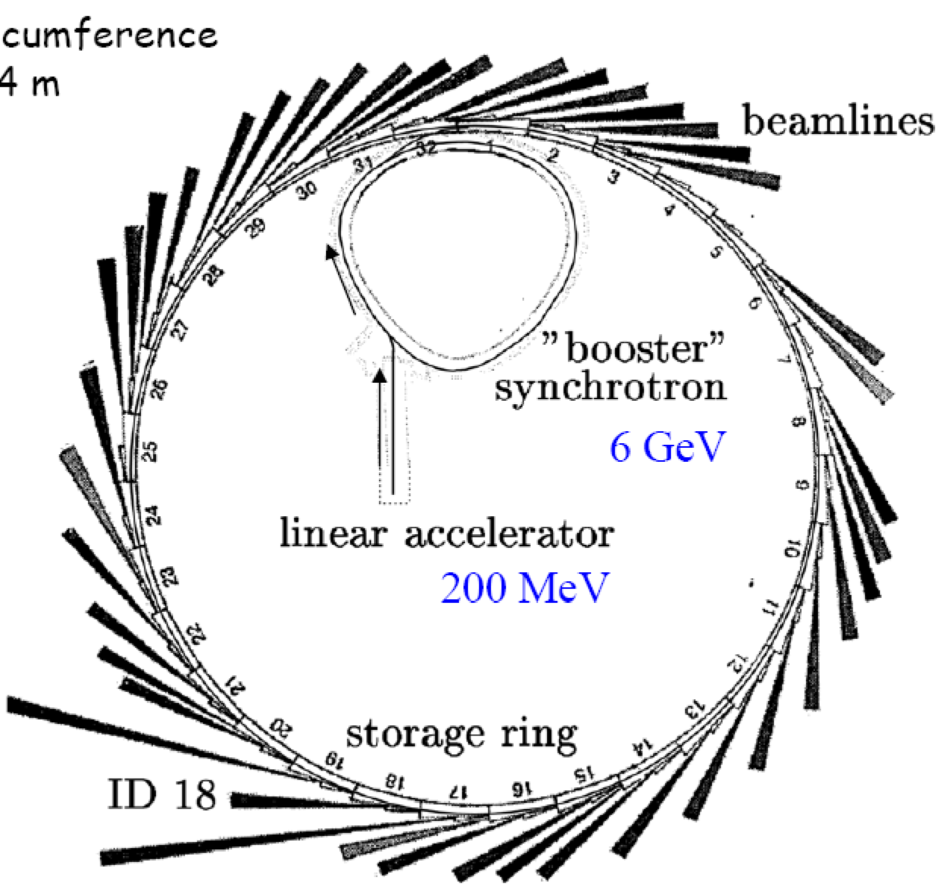
Crab Nebula – an astronomical synchrotron source

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

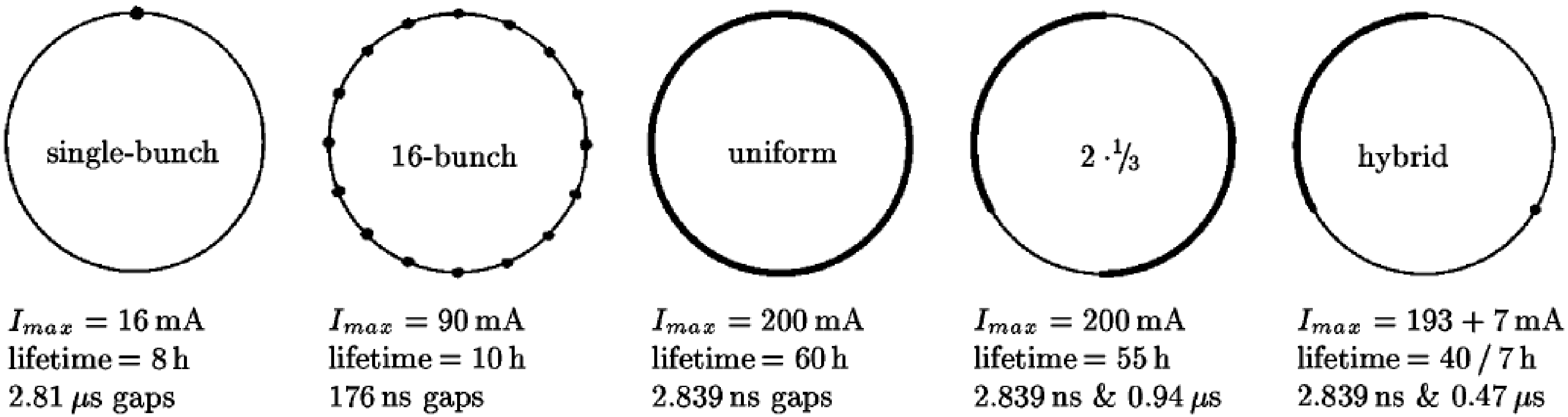
$$N = \nu_{RF} \cdot \frac{L}{c}$$

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

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

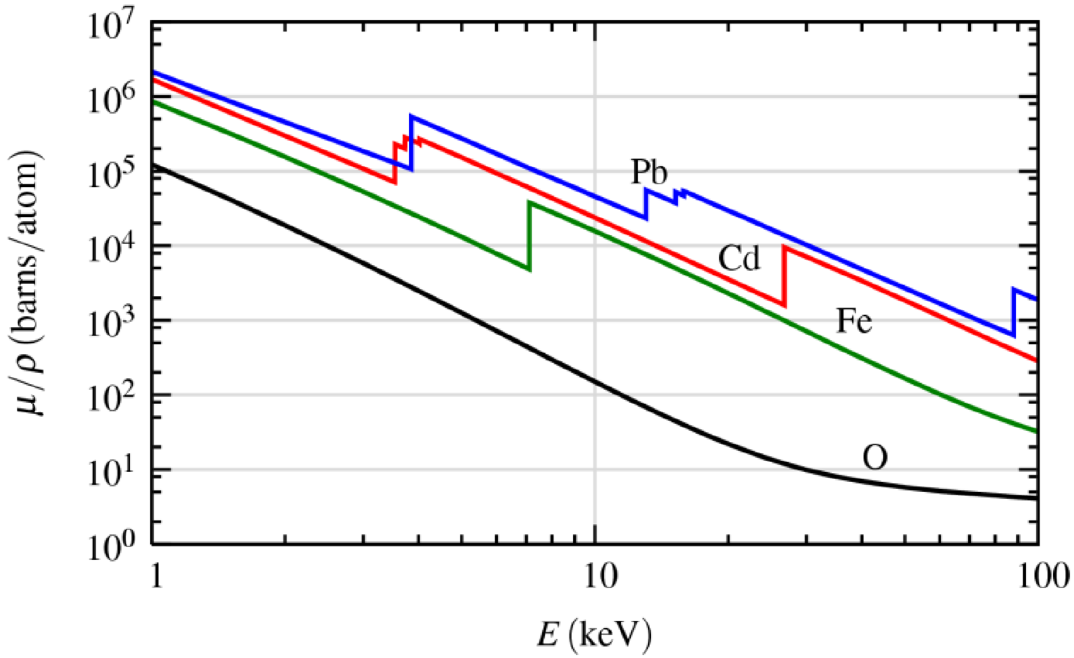
$$\Delta t = \frac{L}{c} \cdot \frac{1}{N} = 2.84\,ns \quad (\,flashes\,)$$

Time structure of Synchrotron Radiation



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**

X-ray adsorption spectroscopy



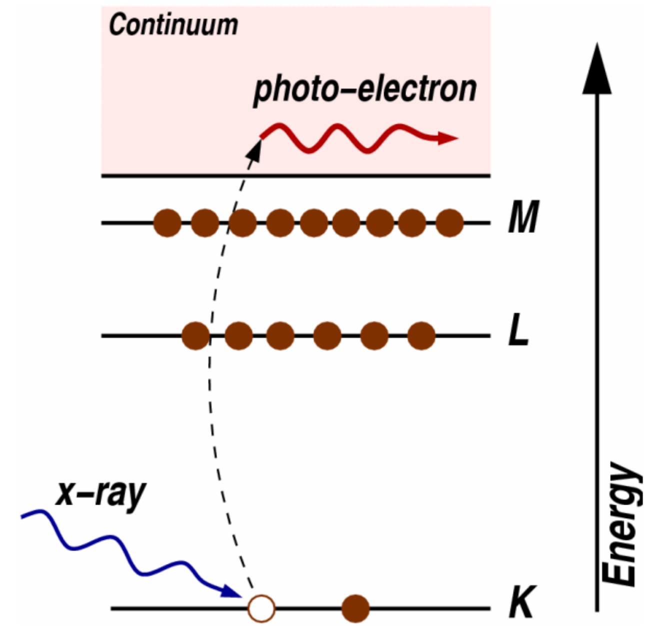


Figure 2.3: The absorption cross-section μ/ρ for several elements over the x-ray energy range of 1 to 100 keV. Notice that there are at least 5 orders of magnitude in variation in μ/ρ, and that in addition to the strong energy dependence, there are also sharp rises corresponding to the core-level binding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

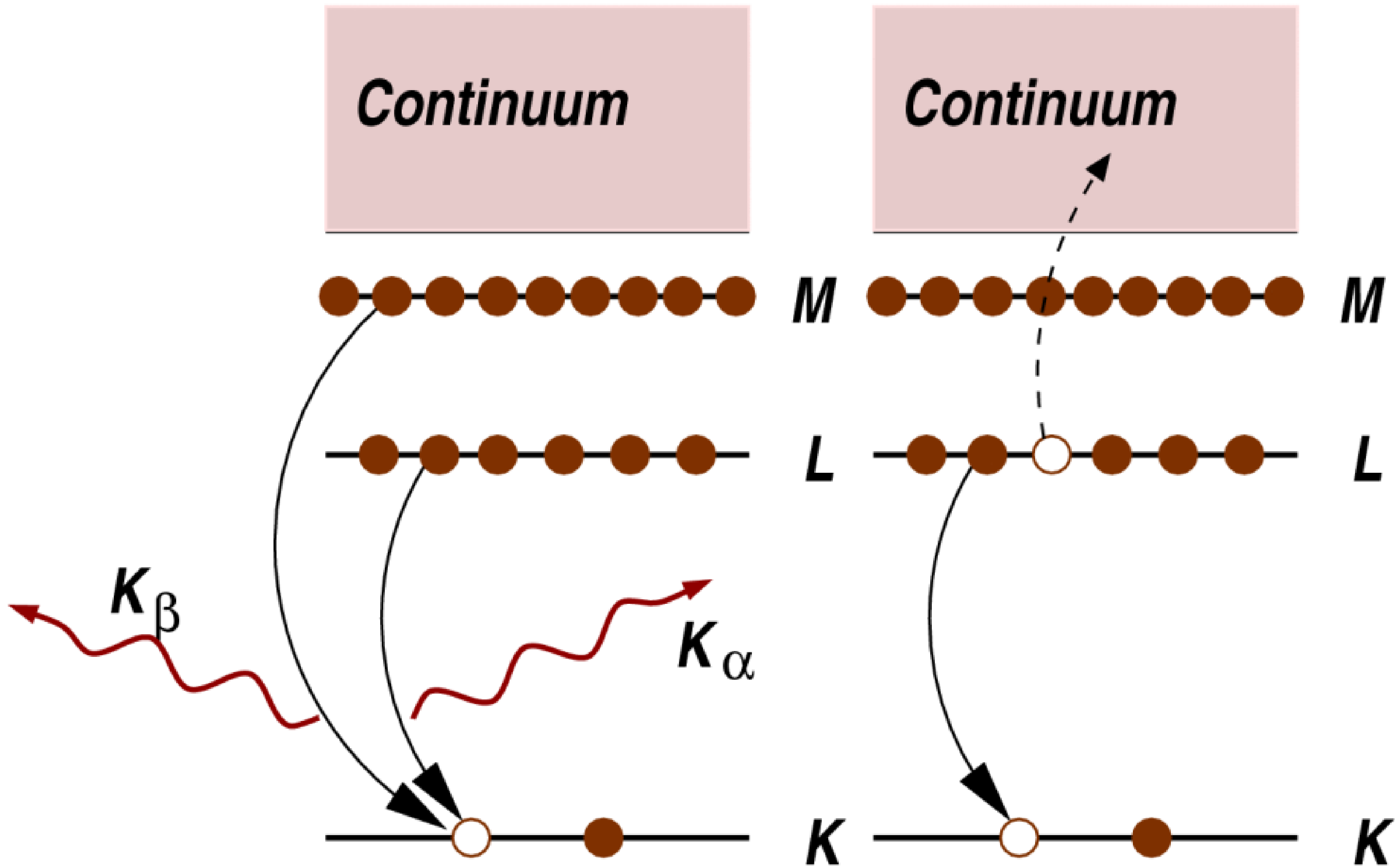
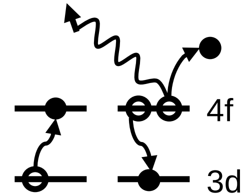


Figure 2.5: Decay of the excited state: x-ray fluorescence (left) and the Auger effect (right). In both cases, the probability of emission (x-ray or electron) is directly proportional to the absorption probability.

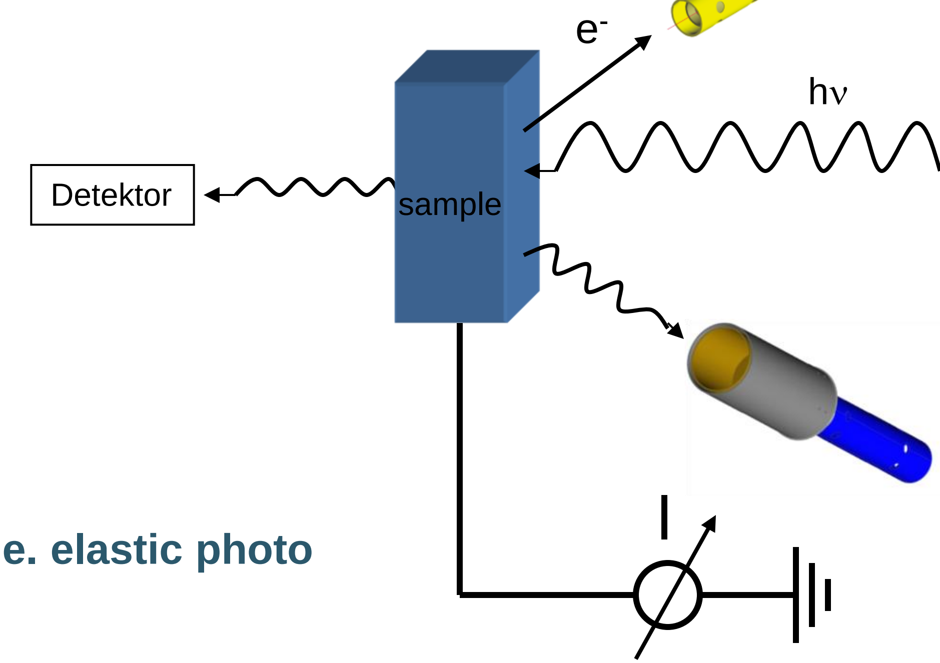
XAS experimentally



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

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

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



<u>All methods can be measured simultaniously to get more information</u>

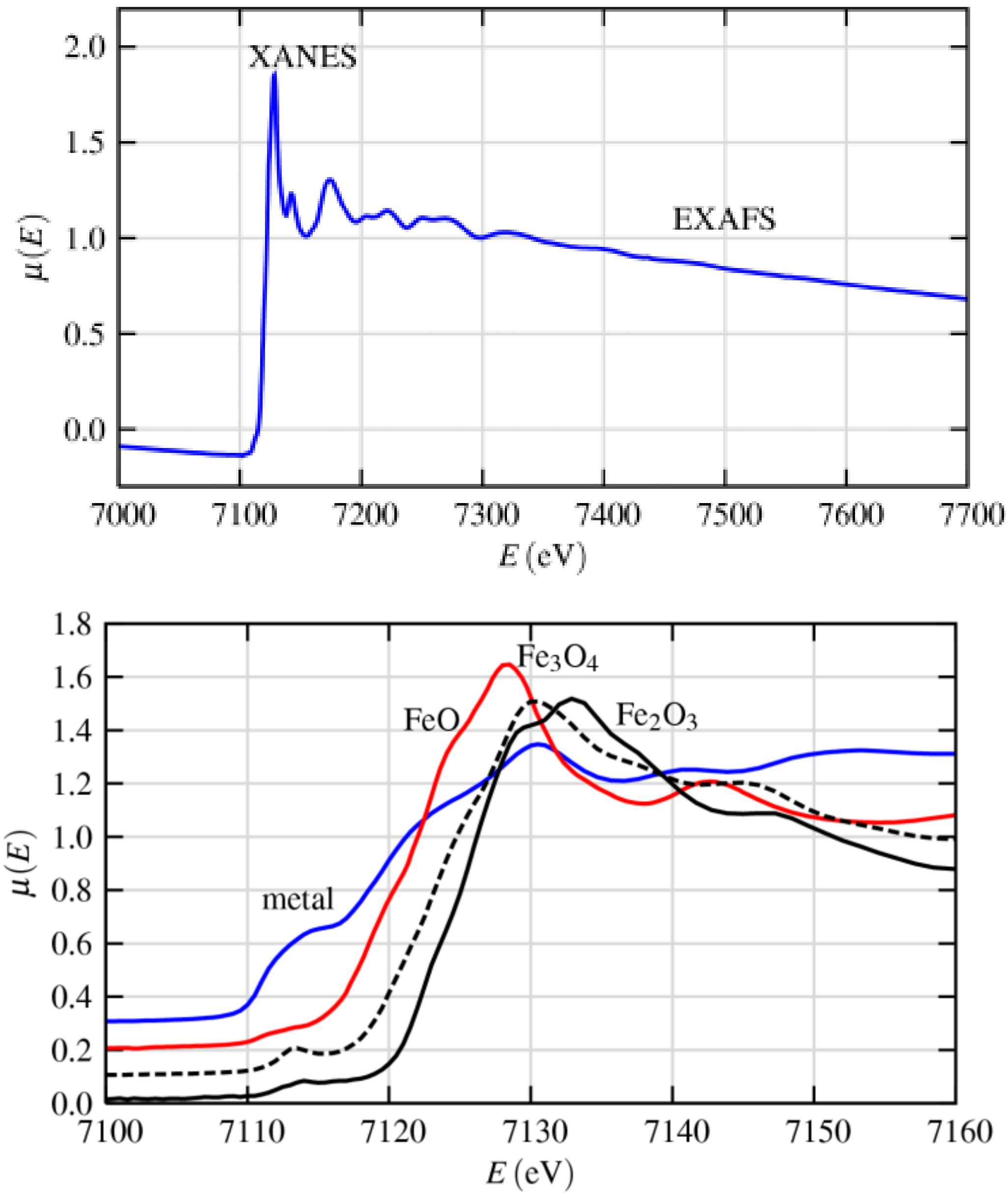
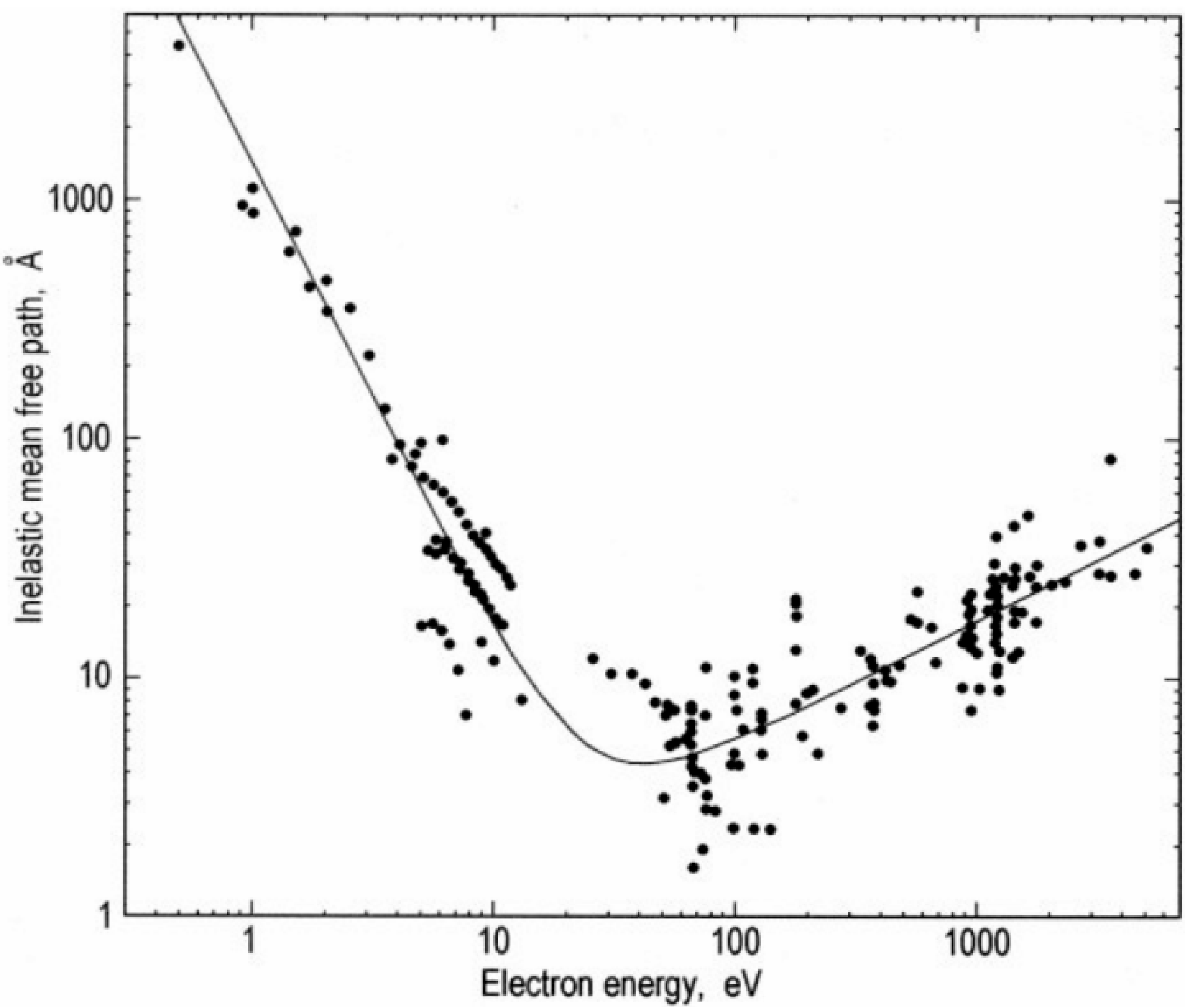
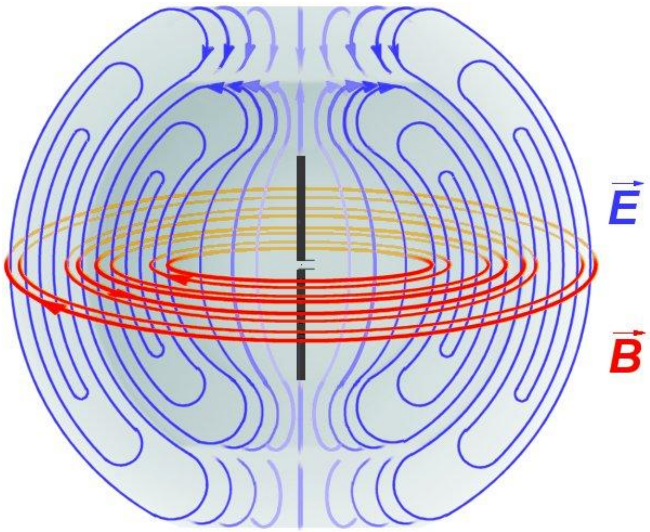


Figure 2.6: XAFS $\mu(E)$ for FeO. On top, the measured XAFS spectrum is shown with the XANES and EXAFS regions identified. On the bottom, $\mu(E)$

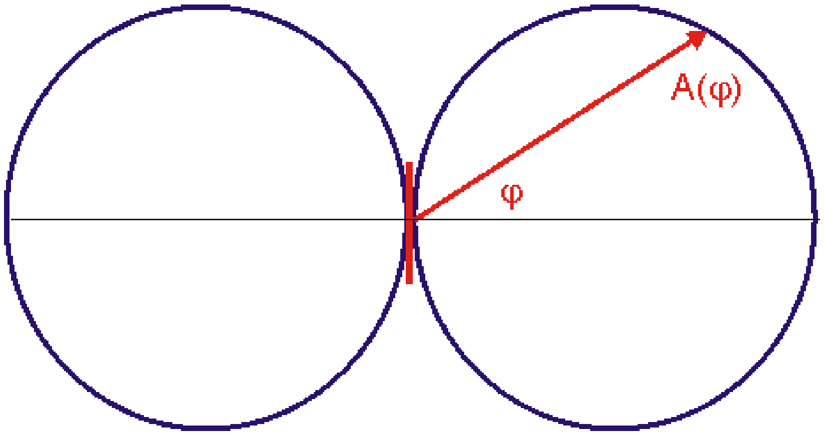
The Universal Curve for the Electron Mean Free Path



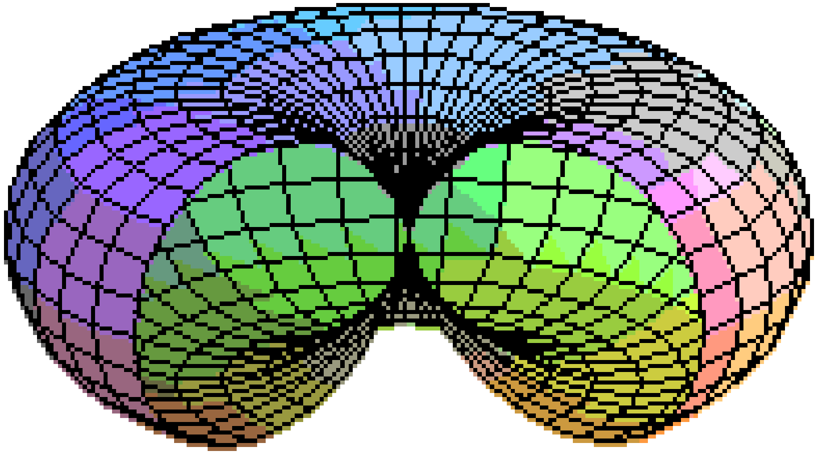
Dipole radiation



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



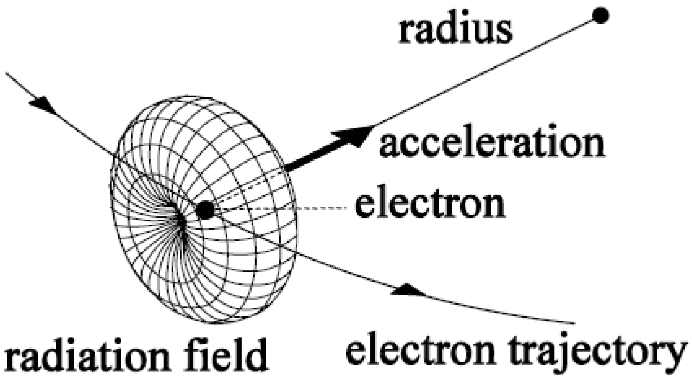
Emission characteristics (A=intensity)



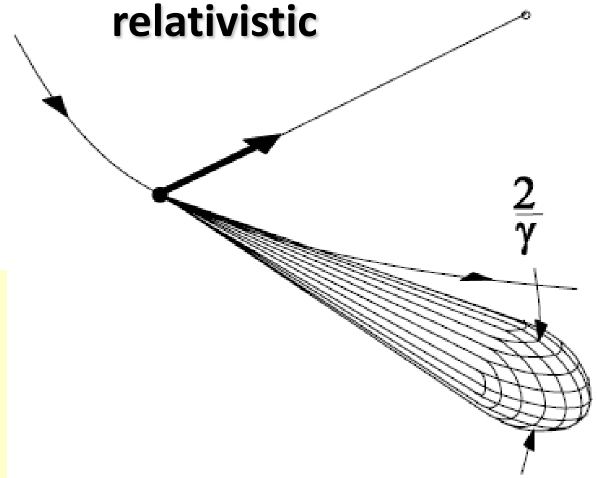
3D-view

Electrons on circular orbit

nonrelativistic



relativistic



opening angle θ

Radiation Power *P*

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

nonrelativistic:

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

$\Rightarrow$ 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:

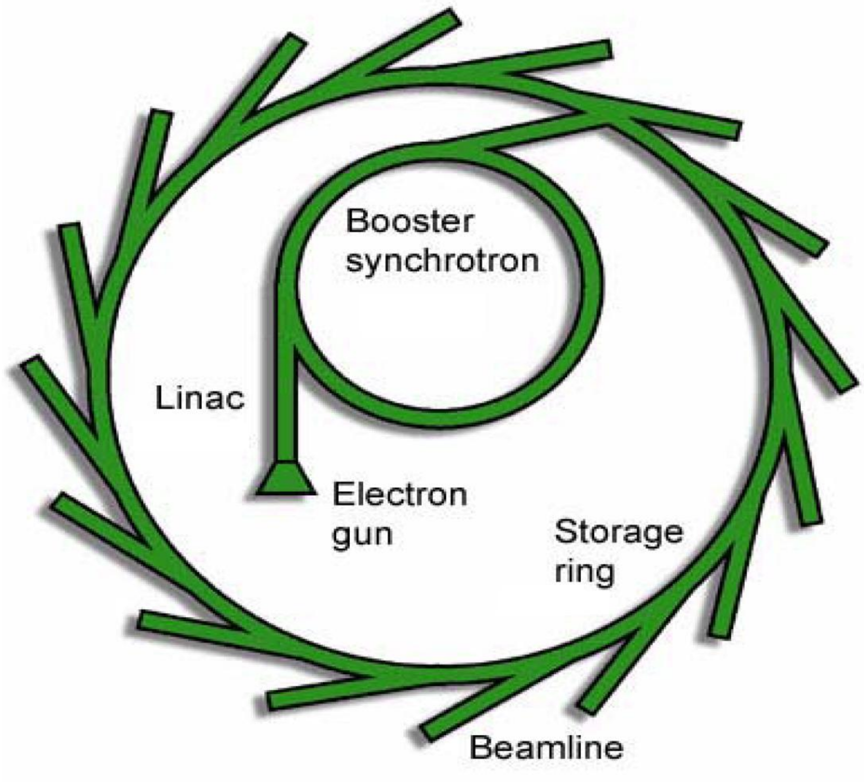
$\rightarrow v \approx c \rightarrow \beta \approx 1$

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

$\Rightarrow$ extremely high radiation power, emitted in a sharp forward cone!

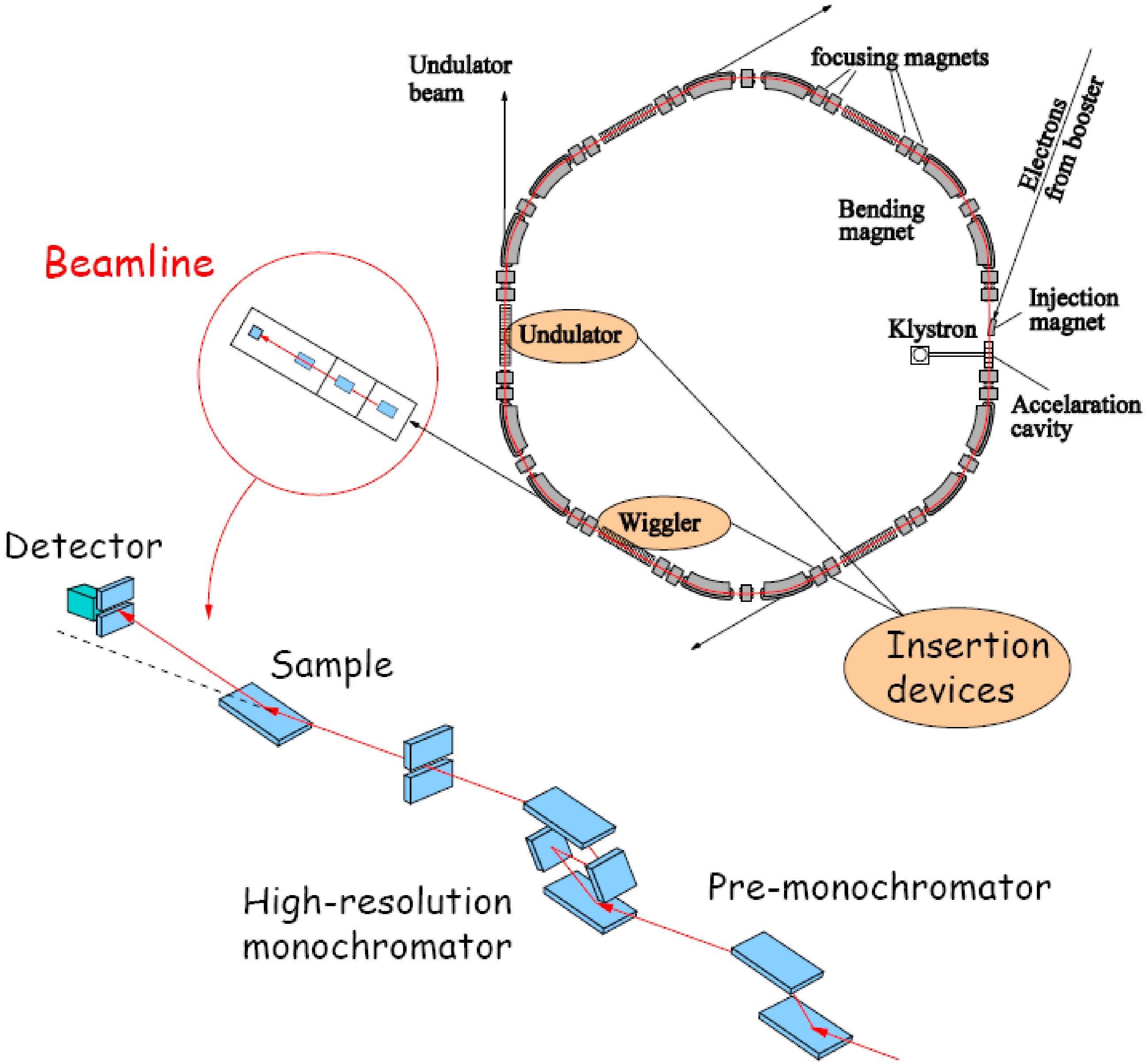
Generation of Synchrotron Radiation

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)
   $\rightarrow$ 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



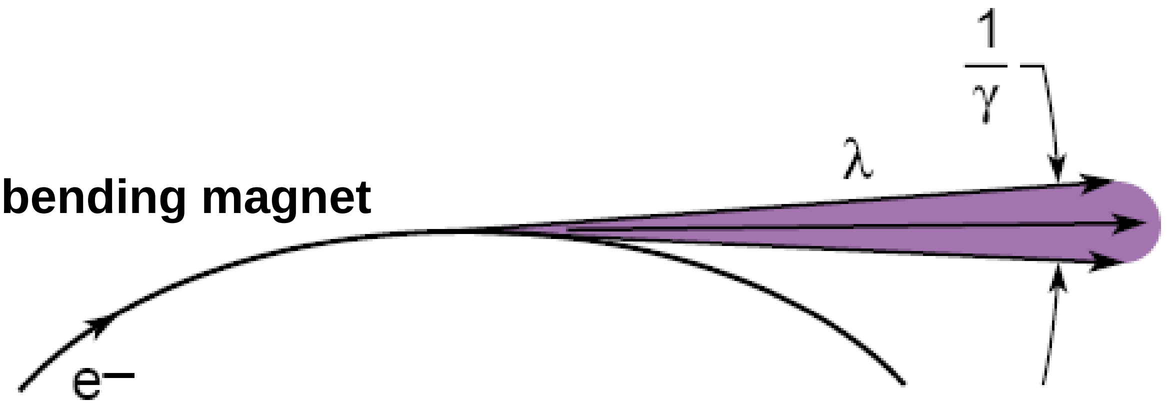
Velocity of relativistic electrons (6 GeV) *v* is only 107 cm/s slower than the velocity of light

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)

$\gamma = E/mc^2 \cong 1820$

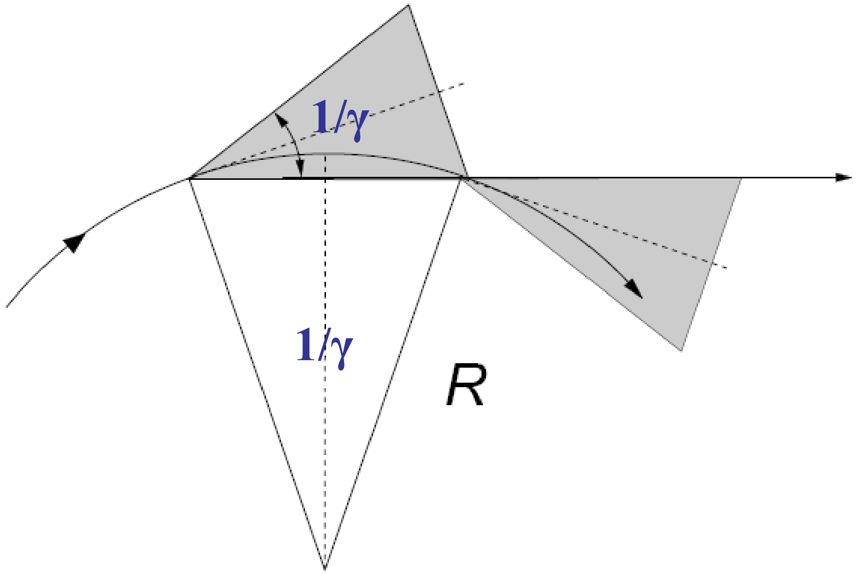
$\theta \approx 8 \times 10^{-5}$ rad (0.08 mrad)

The emitted radiation is a sharp cone with an opening angle $\theta \approx 0.08$ mrad

$\Rightarrow$ Excellent collimation!

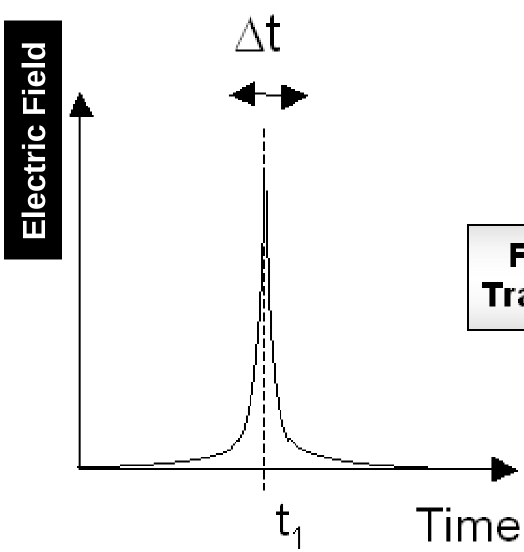
$\Rightarrow$ in a distance of 50 m from the source, one obtains a spot of only ~ 4 mm!

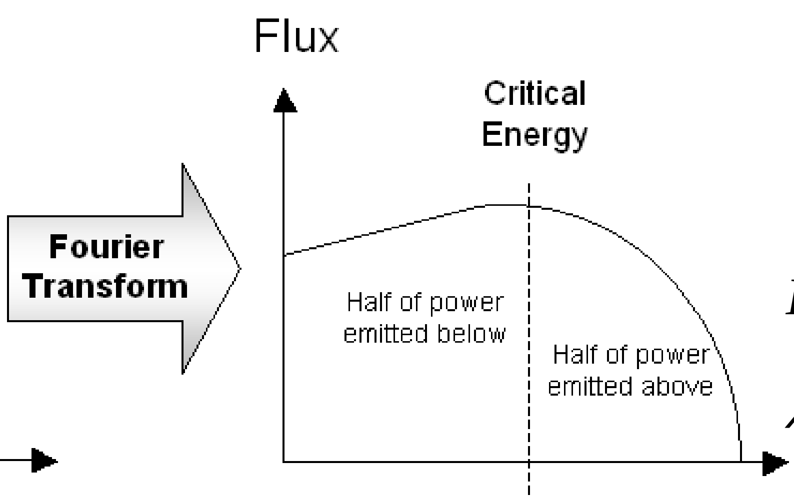
Pulse duration and energy spectrum



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

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

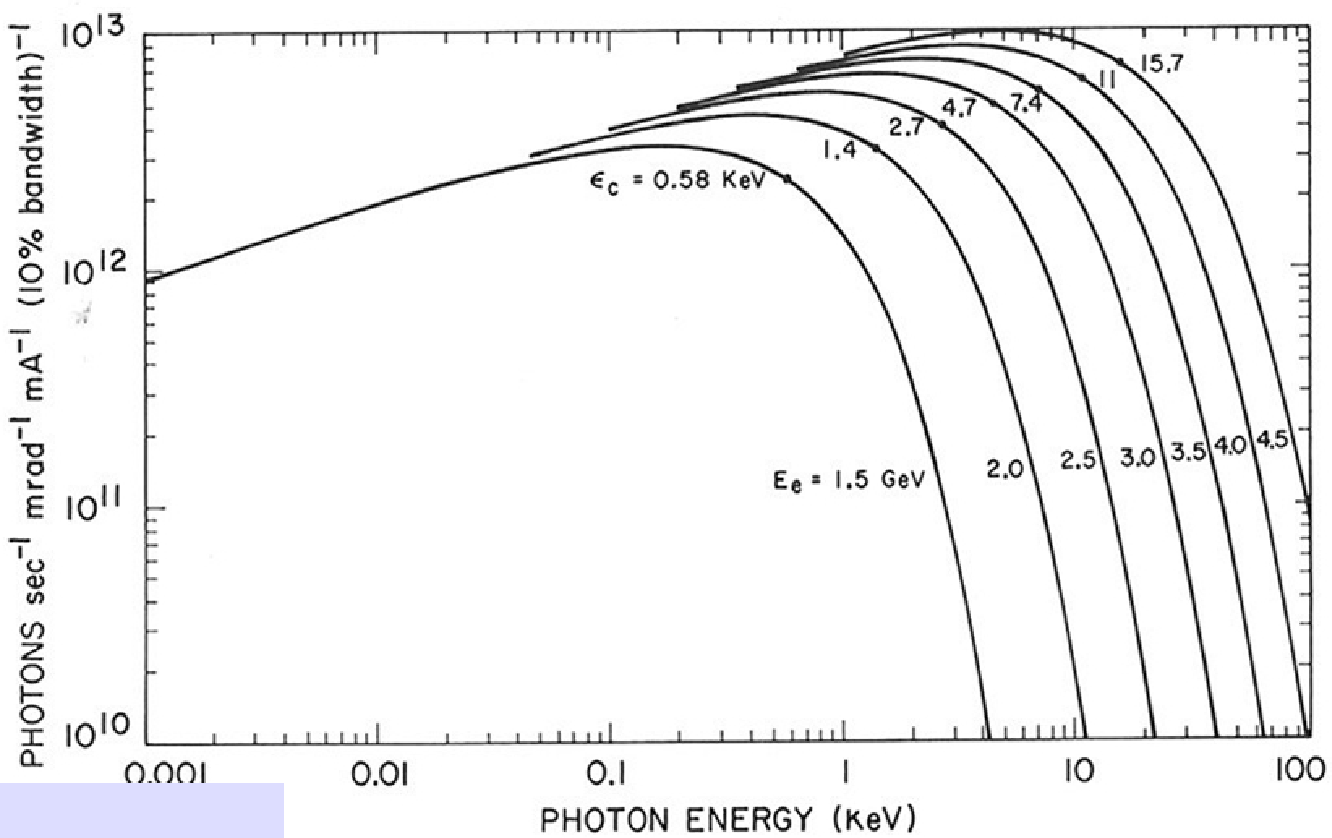
$$\text{Spectral flux} = \frac{\text{Total flux}}{0.1\%\text{bandwidth}} \quad \left[\frac{\text{Photons/s}}{0.1\%\text{bandwidth}}\right]$$

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

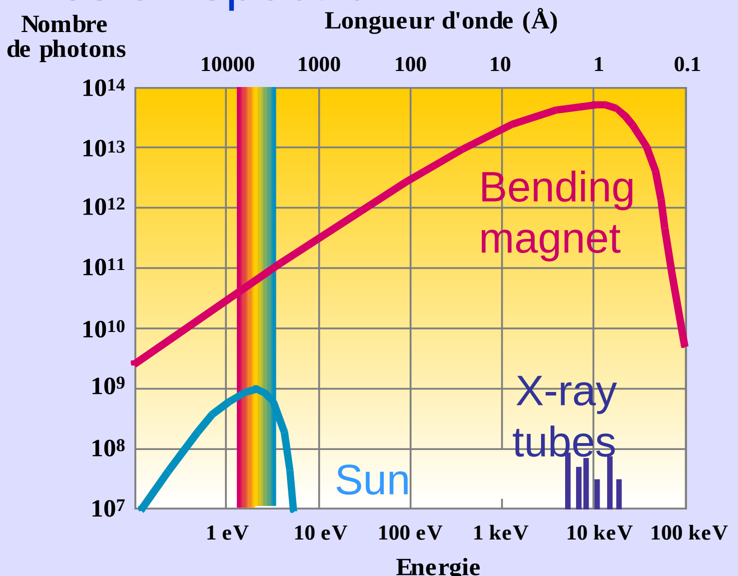
$$\text{Brilliance} = \frac{\text{Total flux}}{\text{solid angle} \cdot \text{source area} \cdot 0.1\%\text{bandwidth}}$$

$$\left[\frac{\text{Photons/s}}{\text{mrad}^2 \cdot \text{mm}^2 \cdot 0.1\%\text{bandwidth}}\right]$$

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



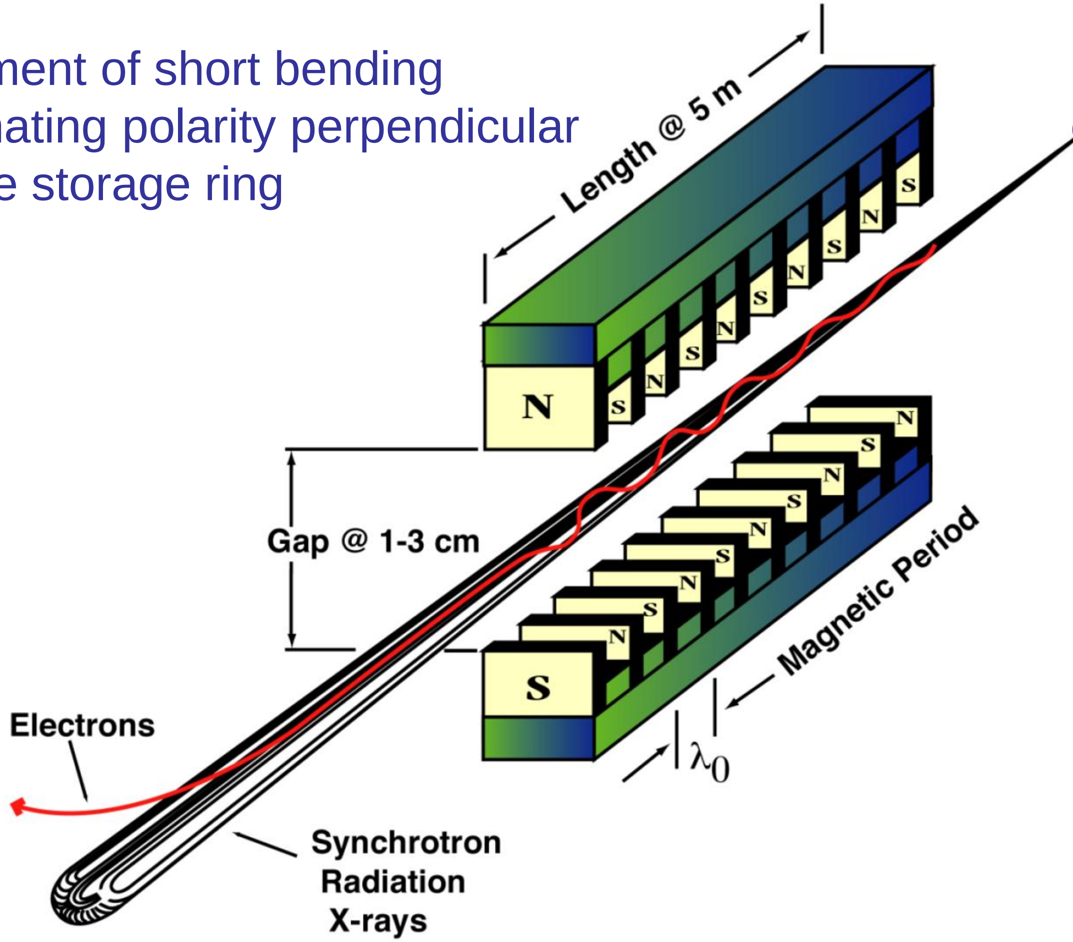
Emission spectrum



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

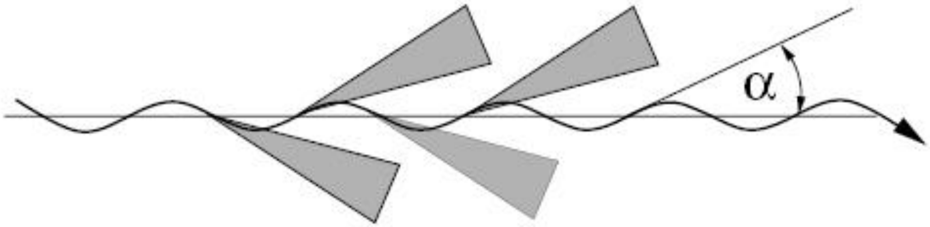
⇒ Radiation is emitted during each individual wiggle

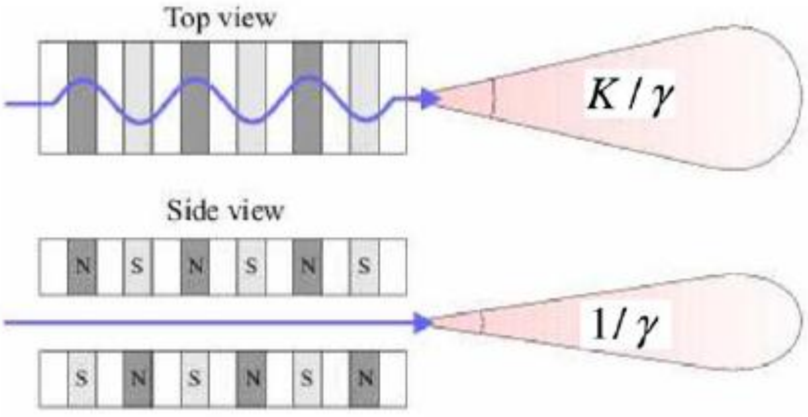
⇒ increase of the intensity

wiggler and undulator

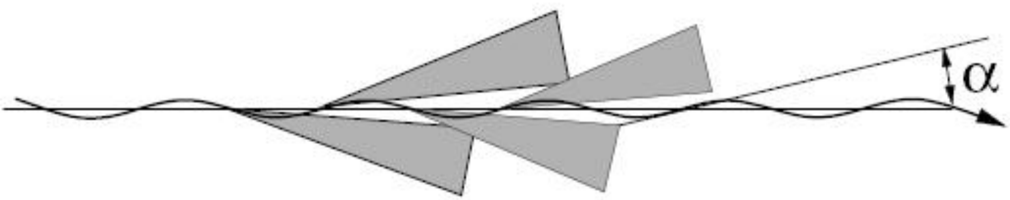
$$K := \alpha \cdot \gamma = \frac{e\, B_0\, \lambda_0}{2\pi\, m_e\, c}$$

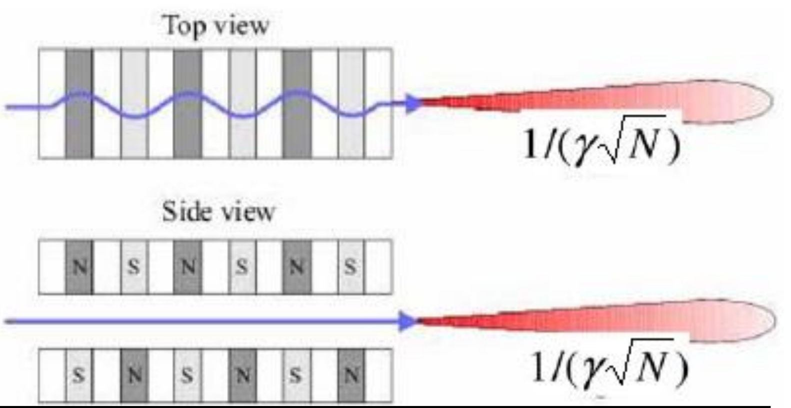
Wiggler regime: α > 1/γ





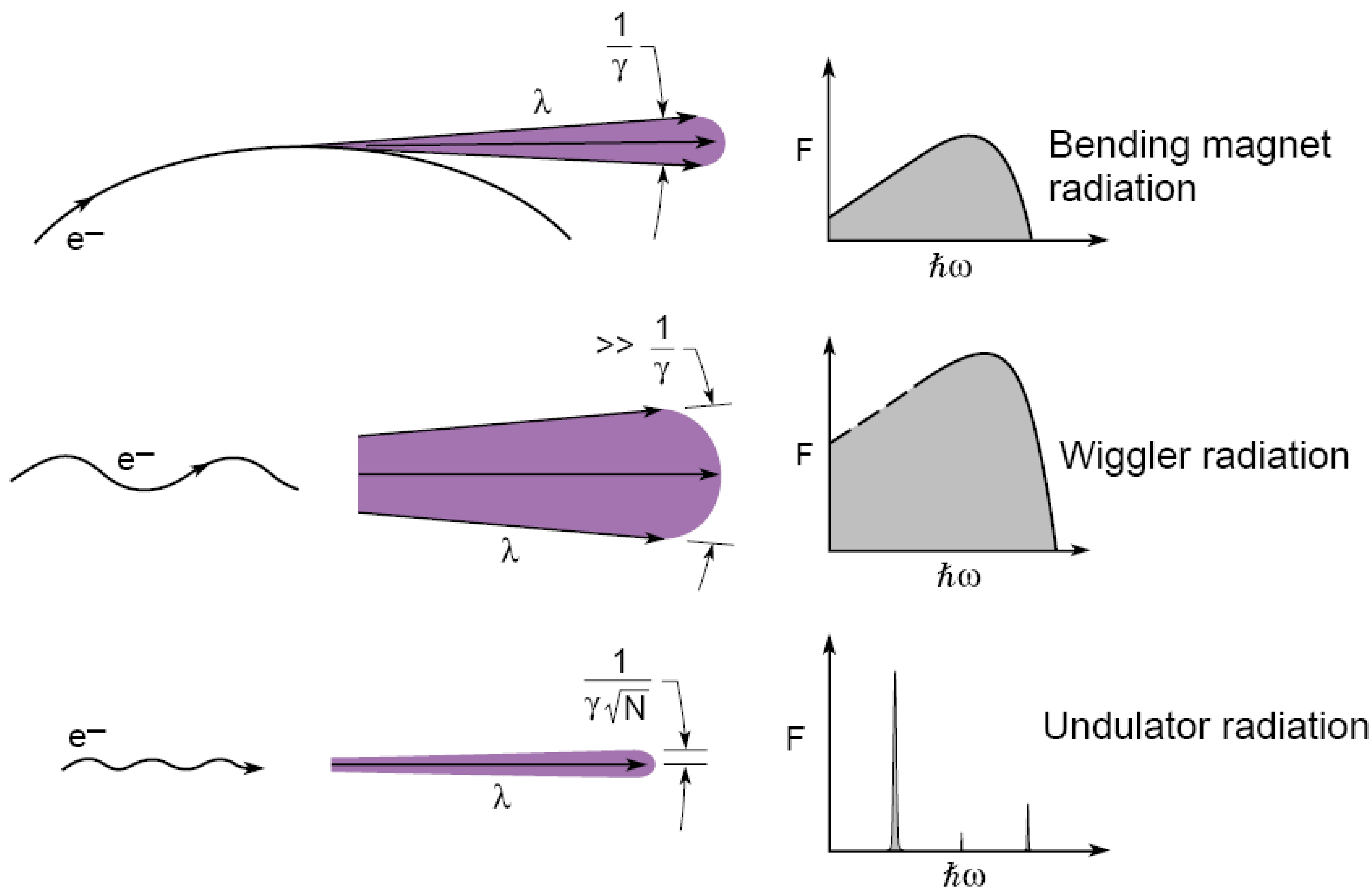
Undulator regime: α ~ 1/γ



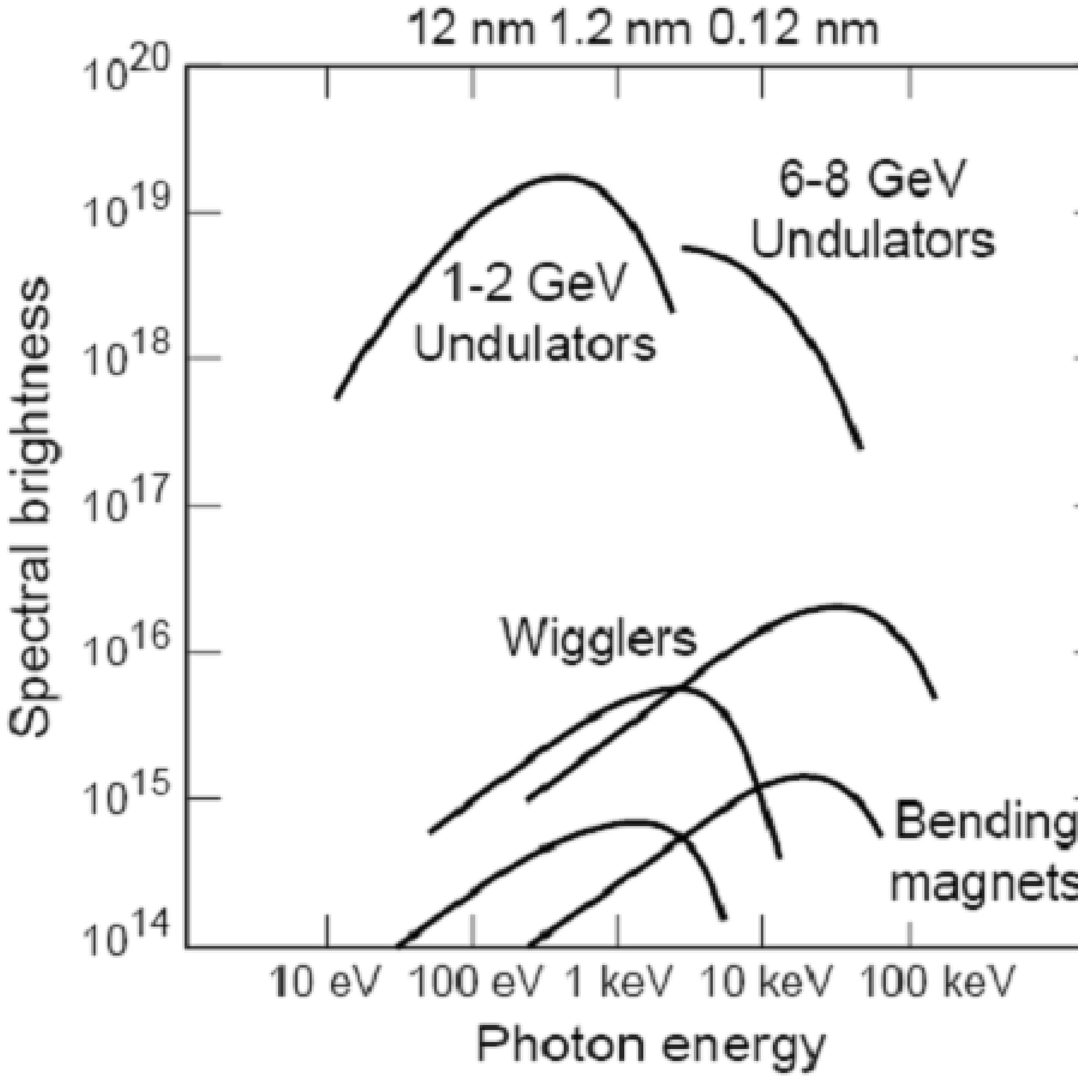


In the undulator regime the radiation cones overlap and the wave trains can interfere

Forms of Synchrotron Radiation

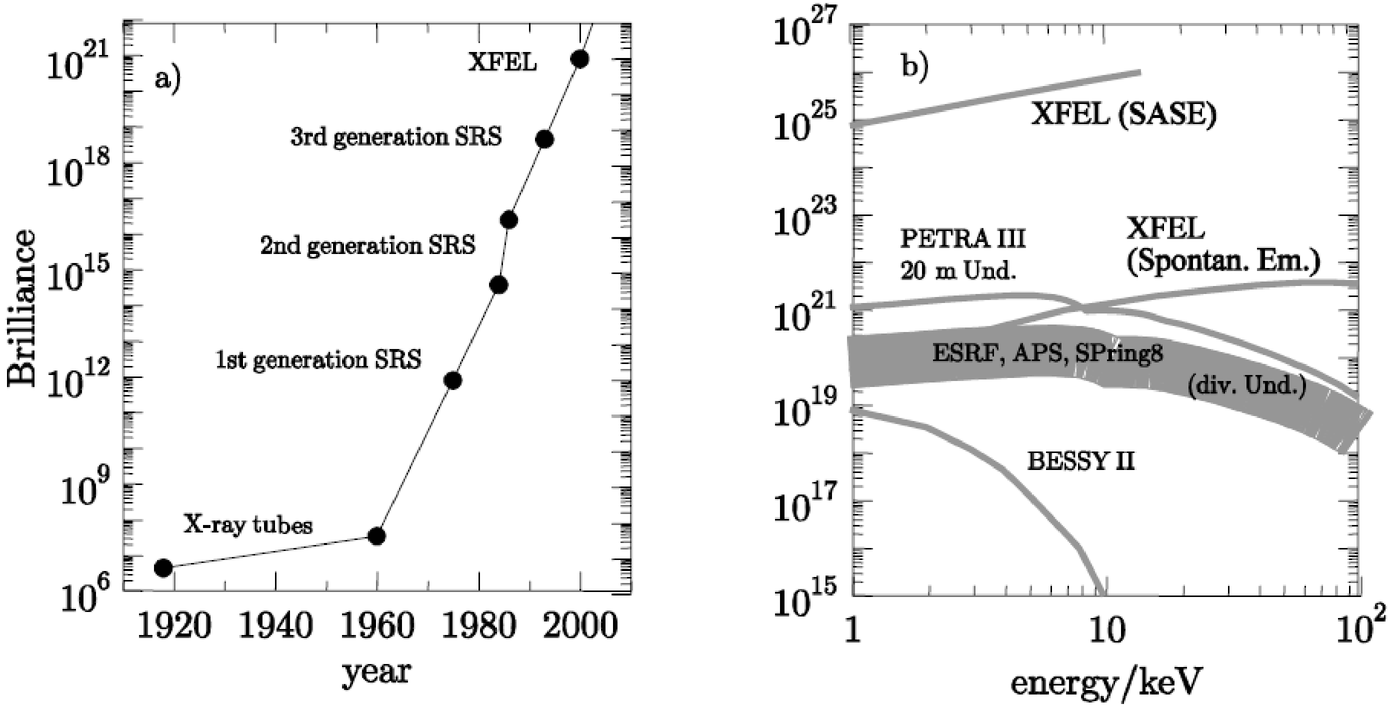


Spectral Brightness



Examples of Wigglers and Undulators

Evolution of Brilliance



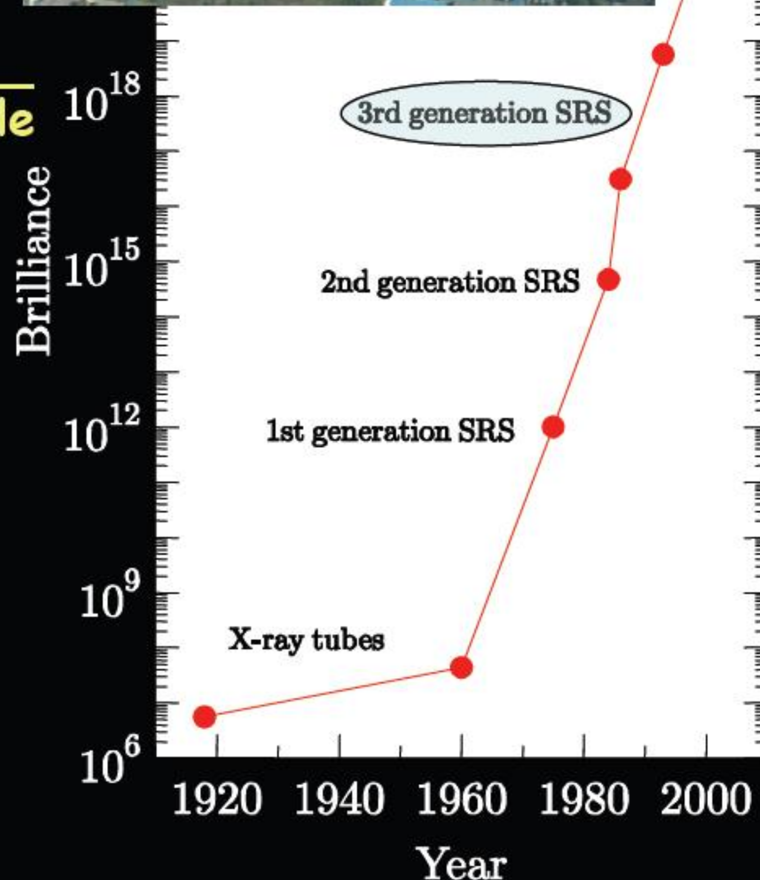
1st generation: Exploitation of the light from the bending magnets of e+/e- colliders originally built for elementary particle physics

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

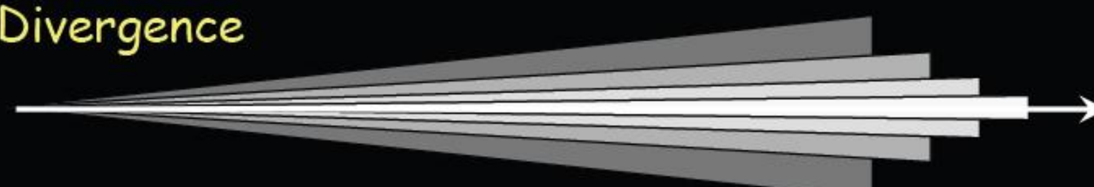
Evolution of Source Brilliance

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



Source size

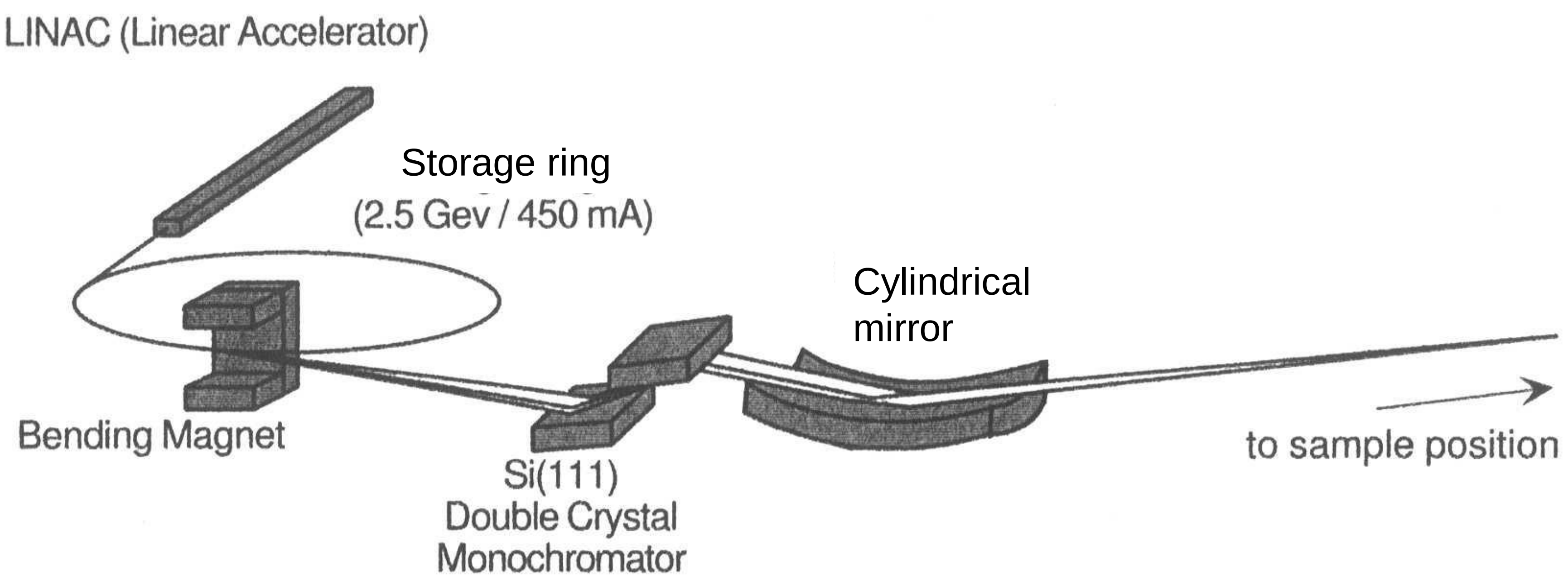
Divergence



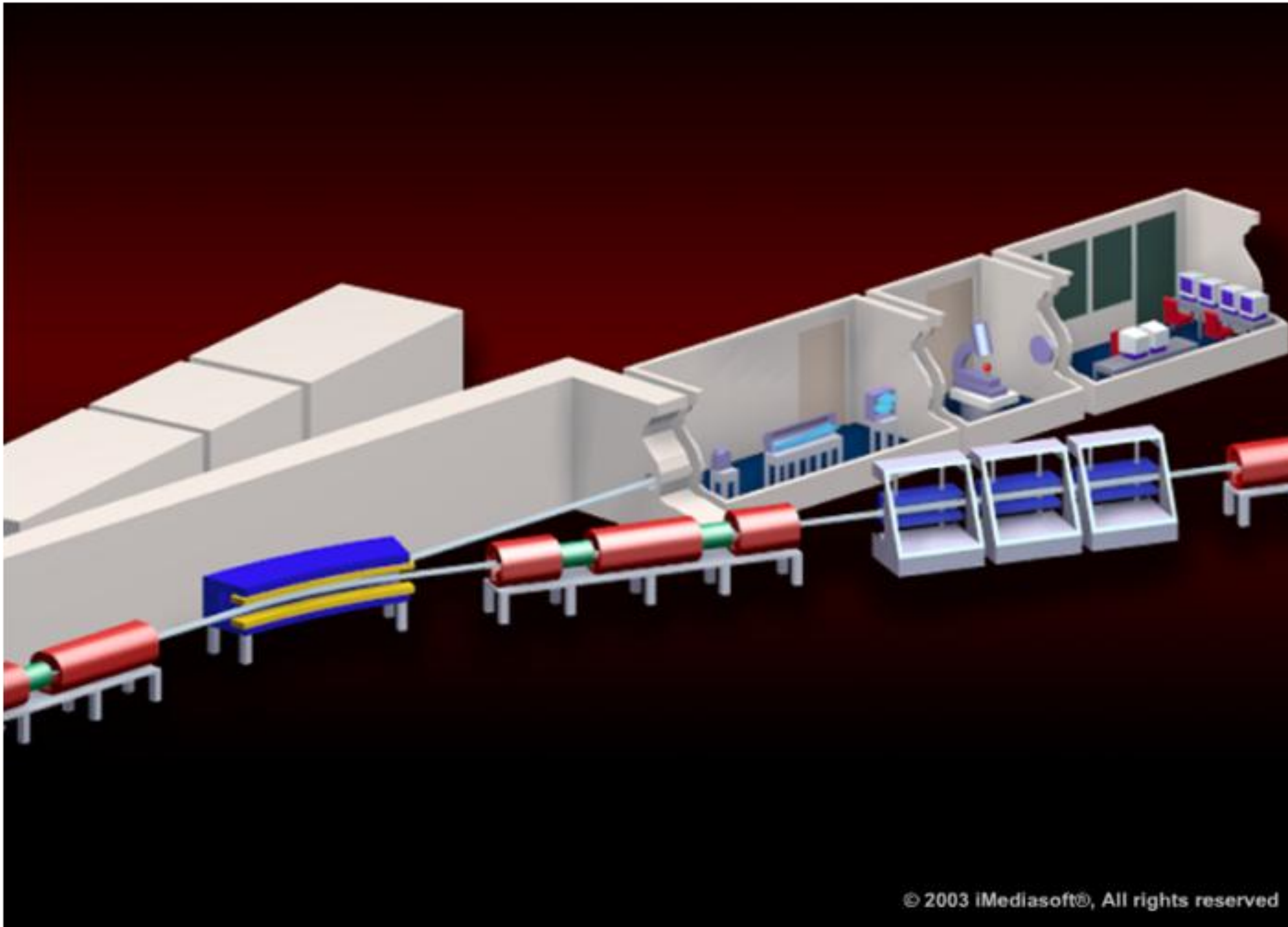
European Synchrotron Radiation Facility (ESRF)

Beamline organization

This is a typical x-ray beamline.
Optics hutch contains elements for conditioning the x-ray beam



How does a beamline work?

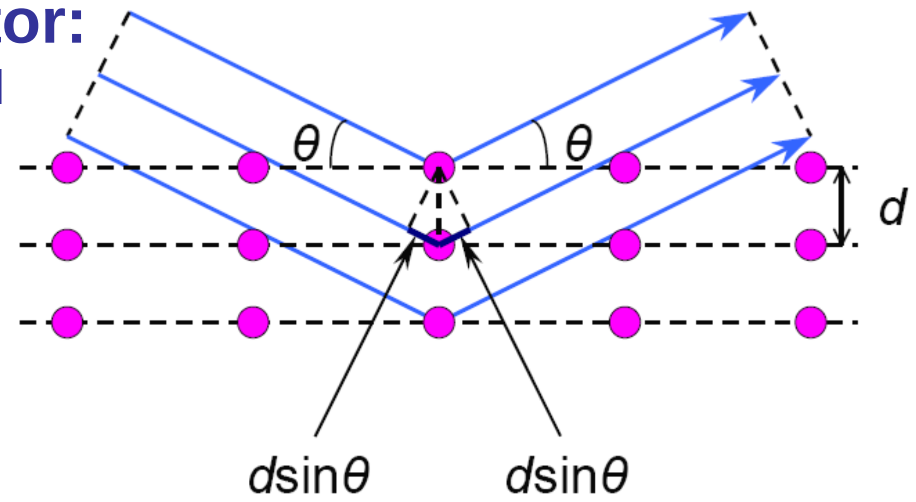


X-ray monochromator using perfect crystal

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

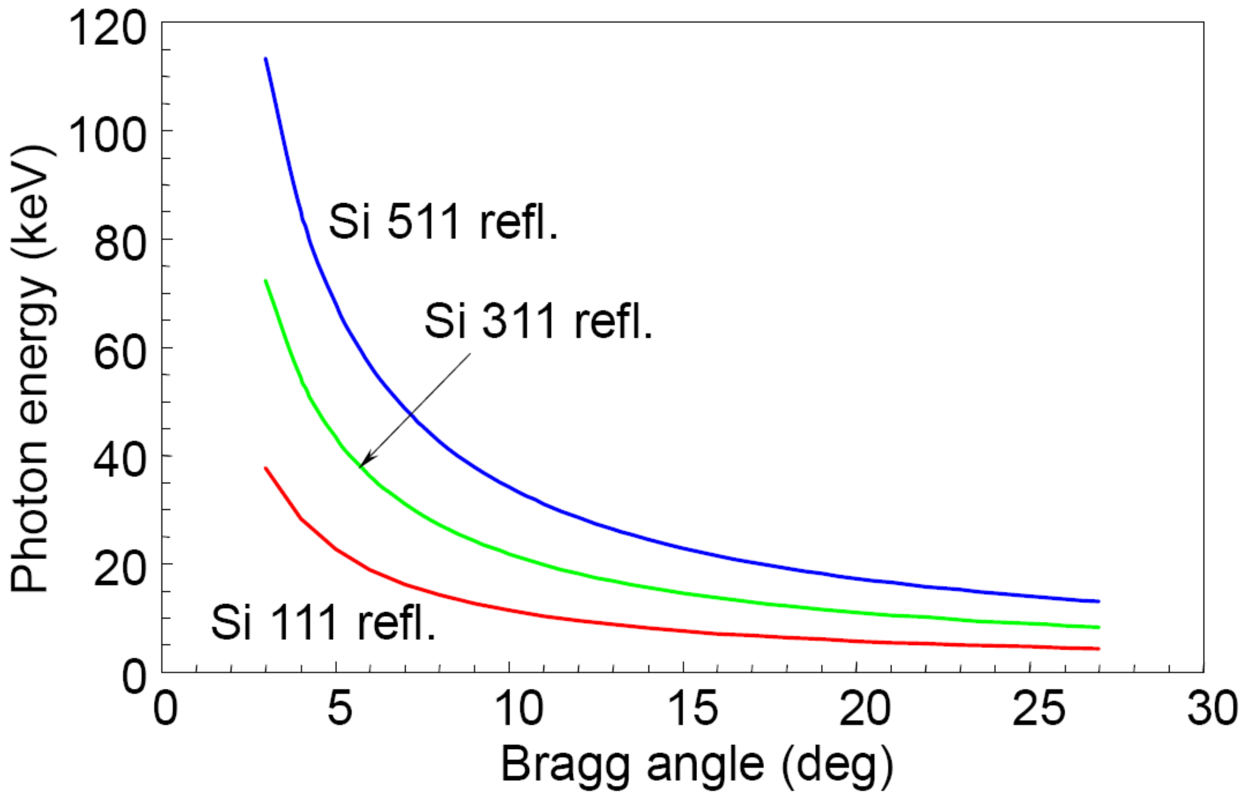
.....

Bragg angles

3~27°

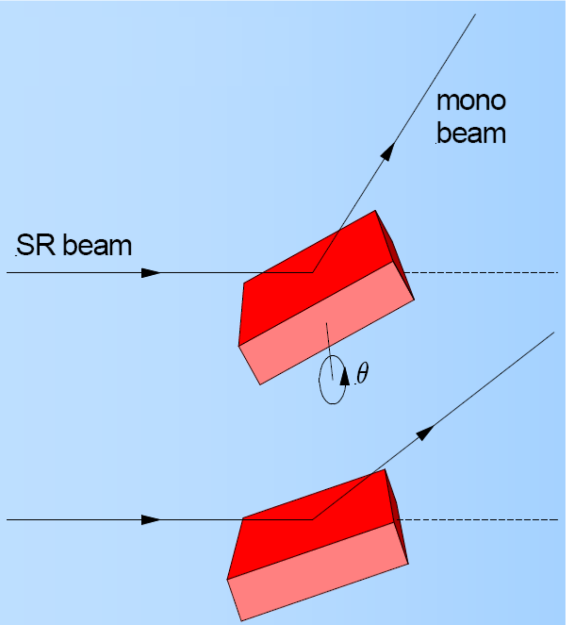
Energy range

4.4~110 keV



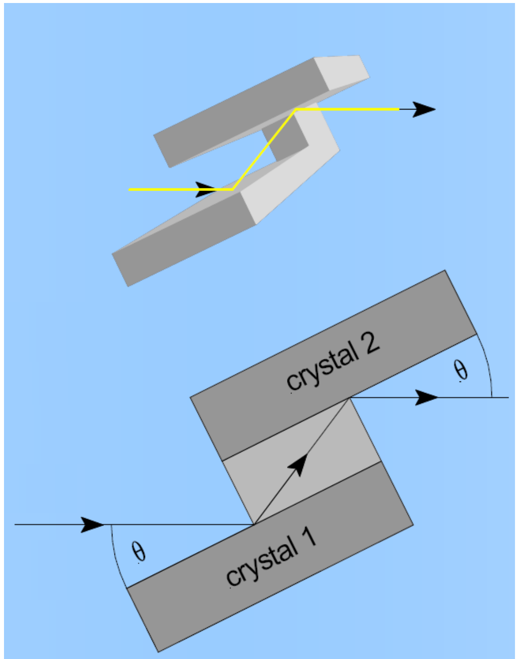
Photon energy (wavelength) can be selected by crystal, net planes, and Bragg angle.

Double crystal monochromator



Problems with single crystal monochromators

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



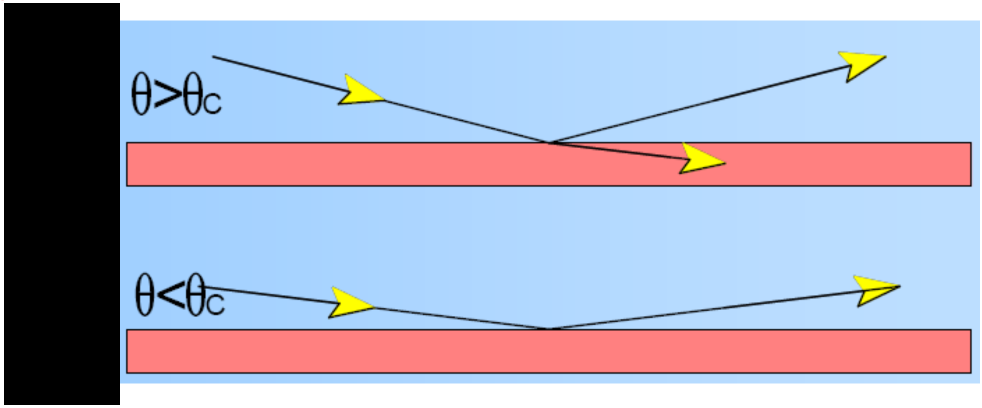
Solution: double crystal design!
Simplest design: cutting a channel for the beam in a silicon block (**channel cut monochromator**)

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

X-ray Mirrors

reflectivity at grazing angles:

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

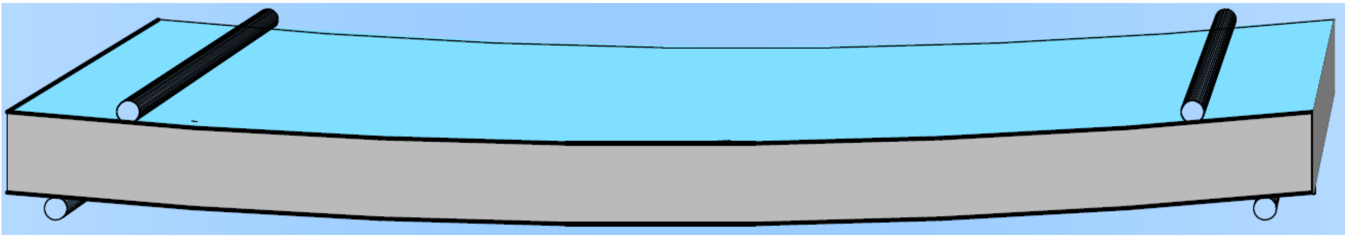


By Snell's law ($n_1 \cos(\theta_1) = n_2 \cos(\theta_2)$ with θ the grazing angle) in the absence of absorption (total reflection), we find total external reflection for angles less than $\theta_c \approx \lambda(r_0\, \rho / \pi)^{1/2}$

θ_c typically a few mrad for x-ray mirrors

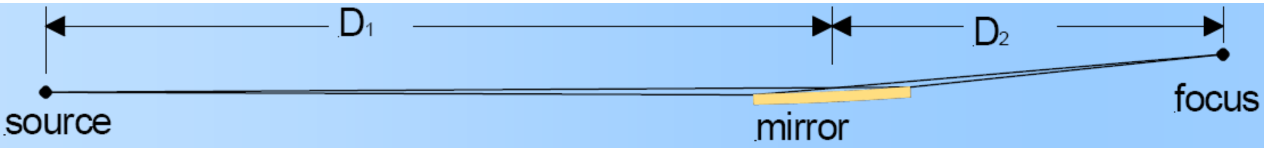
Surface roughness must be considered around critical energy (angle).

Bent mirrors (focusing and collimating)



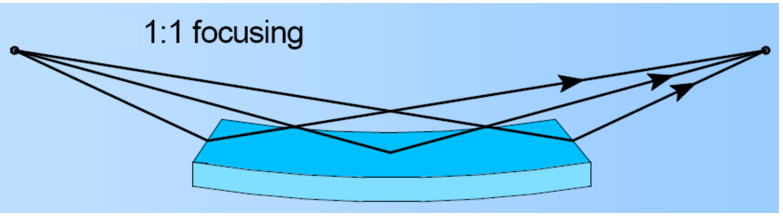
Focusing of the x-ray beam → reflecting surface must have some curvature (achieved e.g. by bending mirror, **mirror focuses in one plane only**!)

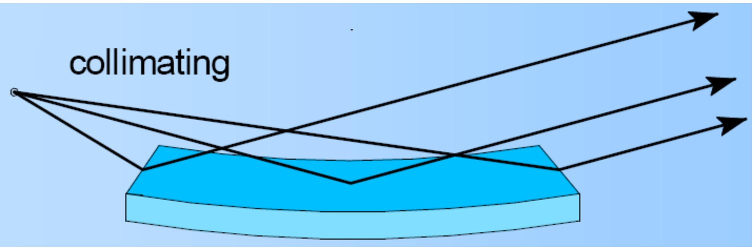
Bending radius R (can be ~ 10 km)



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

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)

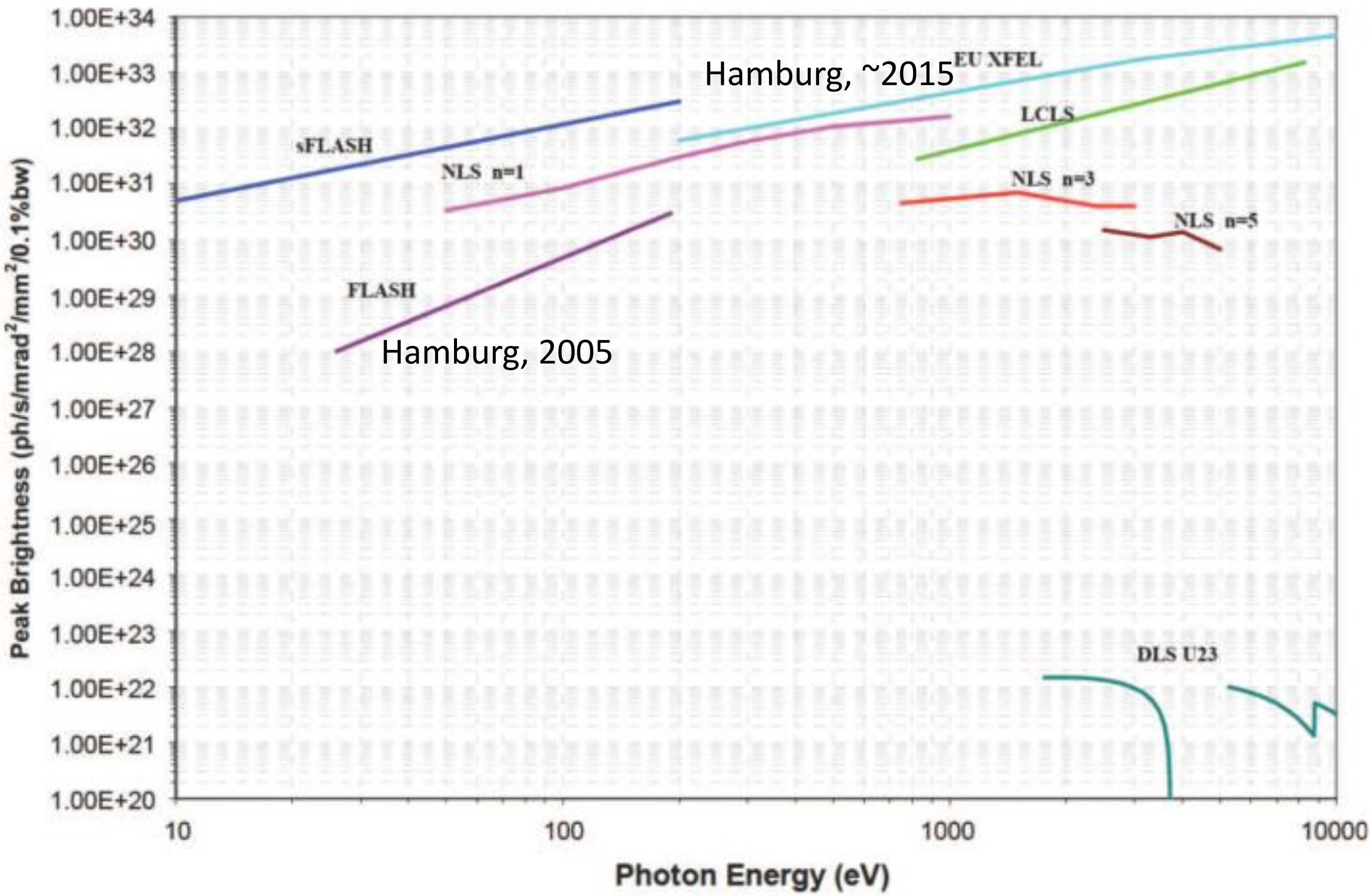


Figure 1. Comparison of several recently commissioned FELs (FLASH and LCLS) and several planned FELs (sFLASH, Euro XFEL, NLS) with a state-of-the-art undulator beamline on the Diamond Light source. The standard definition of brightness is given in photons/unit time/unit solid angle/unit area/normalised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

SASE – spontaneous amplified self-emission

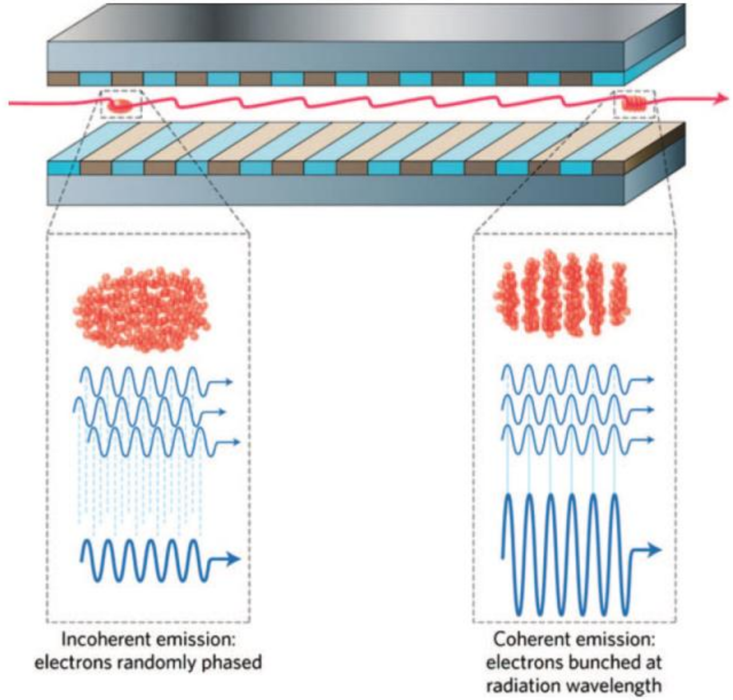
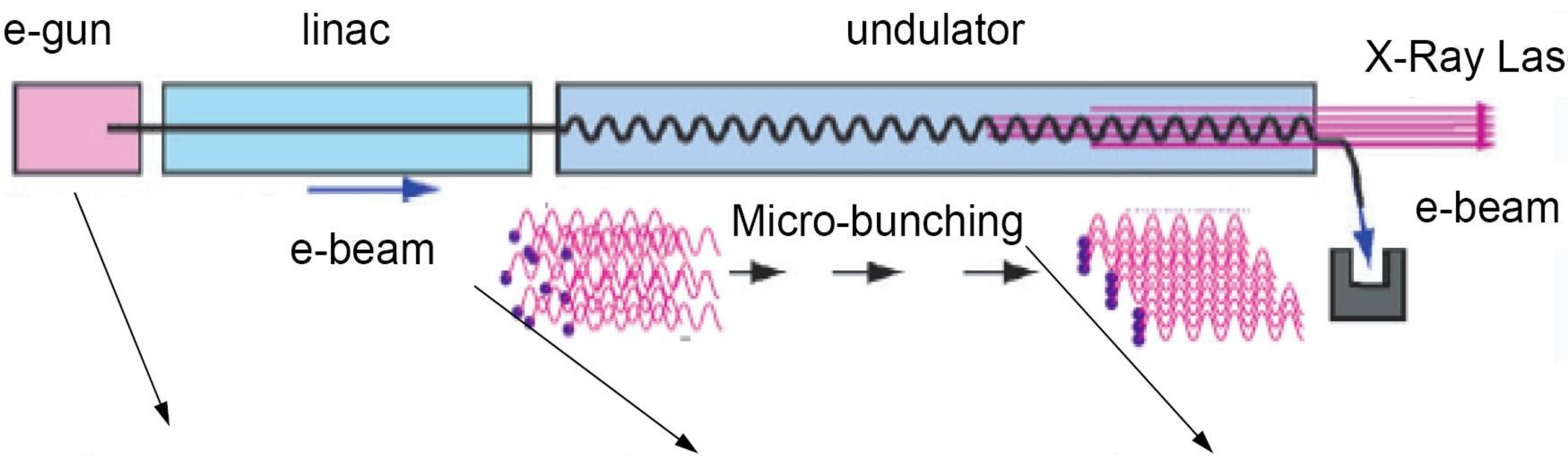


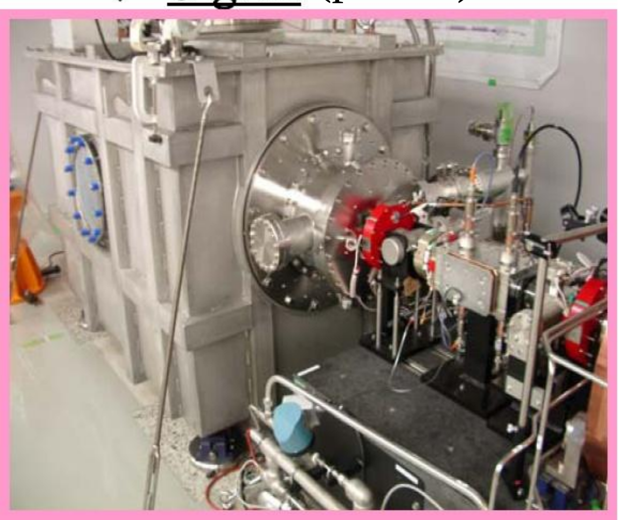
Figure 3. Schematic of SASE operation where the oscillation induced in the electron beam by the periodic magnetic field leads to radiation emission and, at the end of a sufficiently long undulator, self-organisation of the electrons gives rise to coherent X-ray emission. Reprinted by permission from 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).

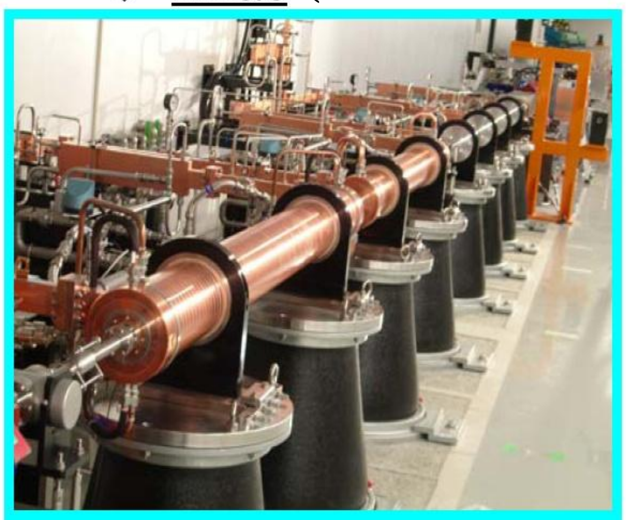
Linac-based Free Electron Laser
Self-Amplified Spontaneous Emission (SASE)

Principle design (SPring-8, Japan):

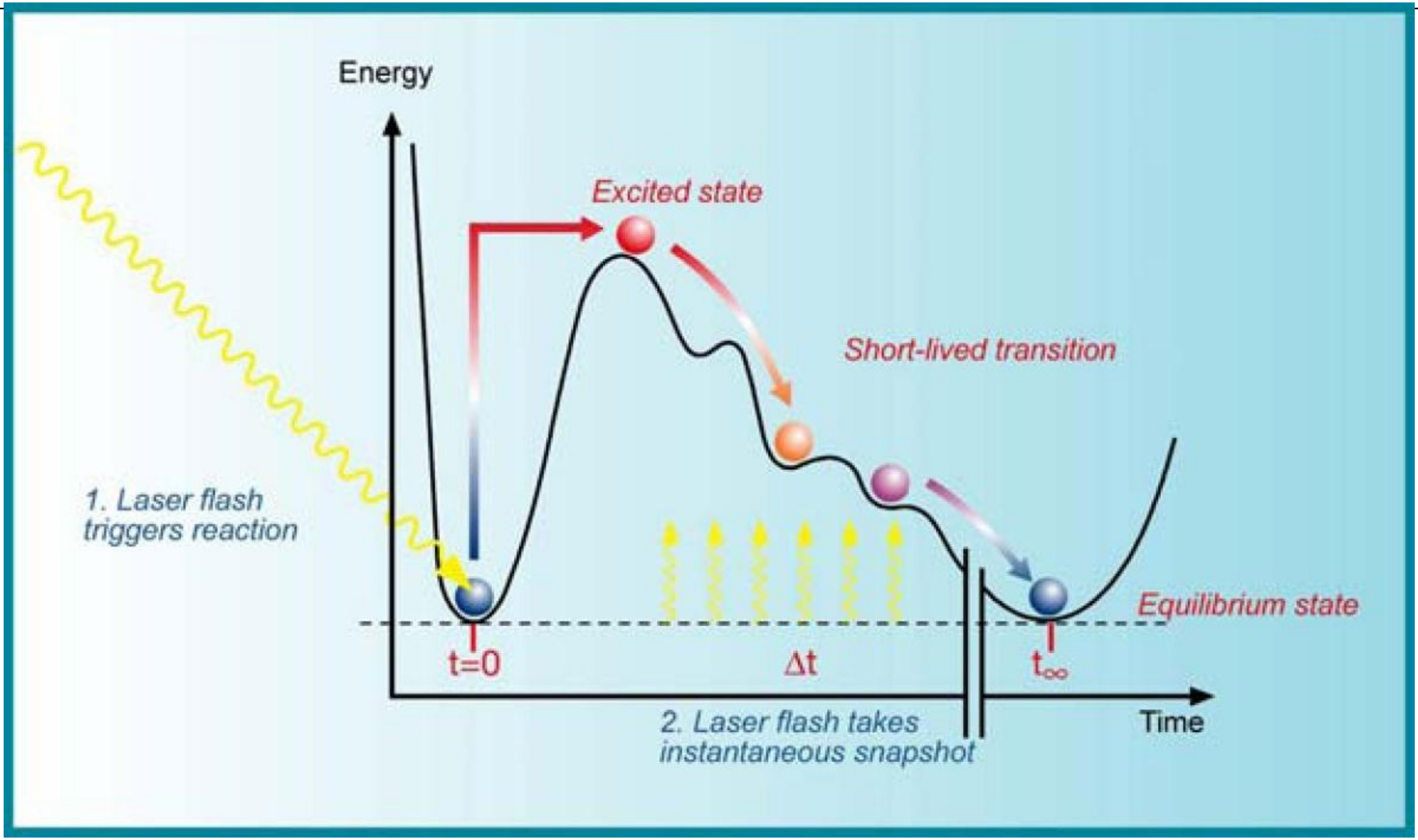


◆ <u>e-gun</u> (pulsed) ◆ <u>linac</u> (linear accelerator) ◆ <u>undulator</u>



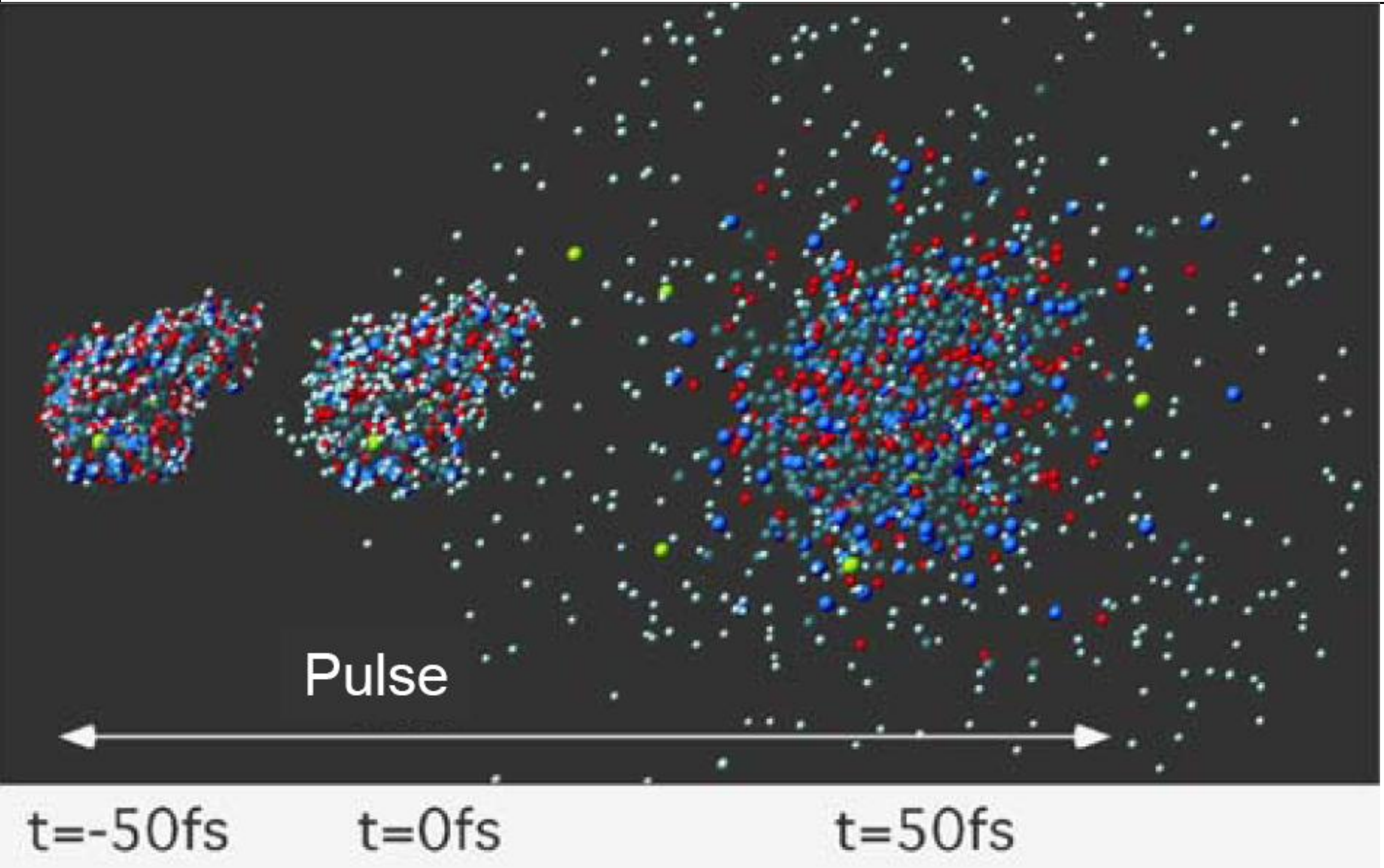


Pump-probe experiment



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

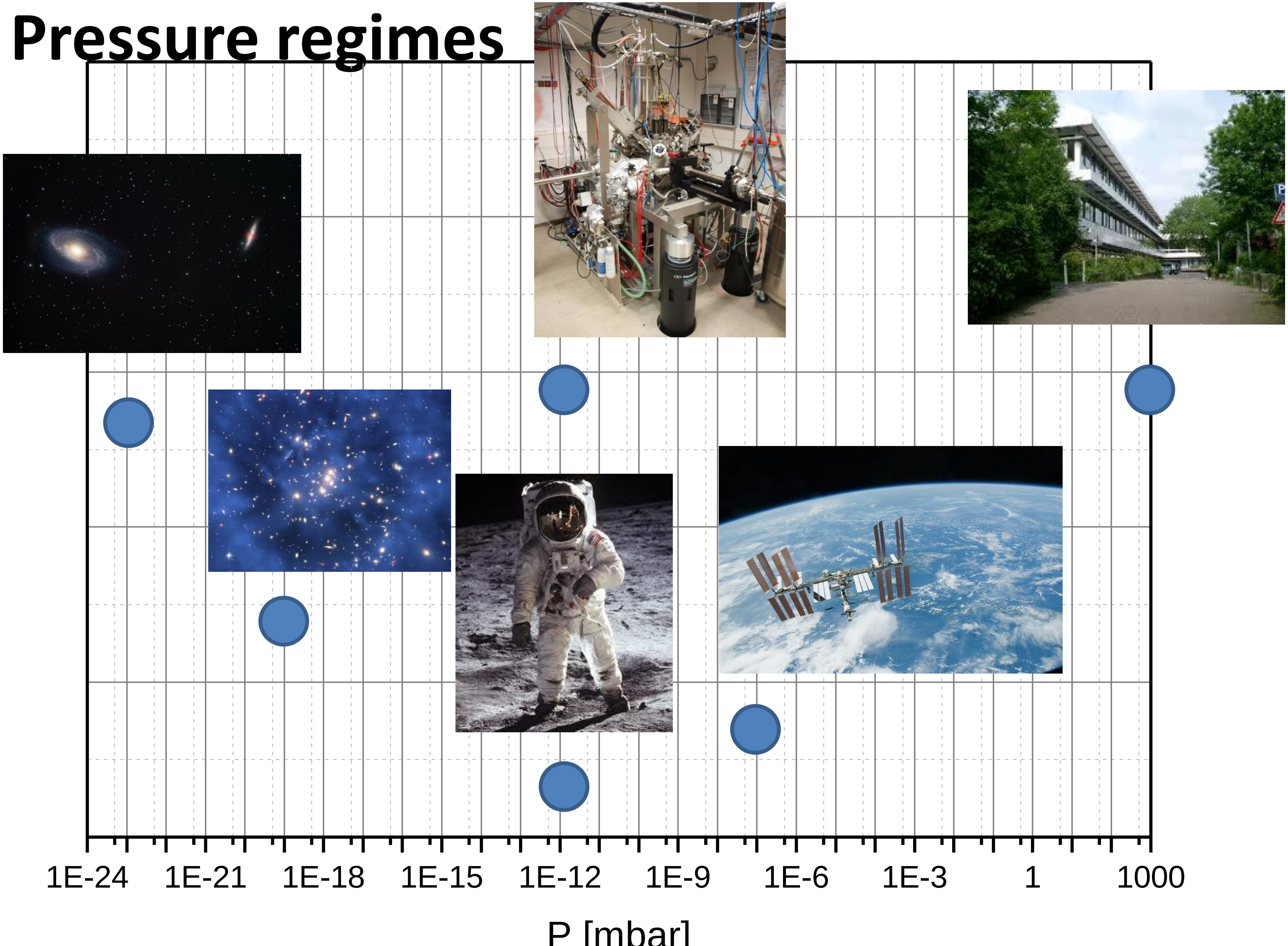
Requirement: Pulse must be short enough and not to intense,
to take picture before molecule disintegrates !

R. Neutze et al., Nature, August 2000

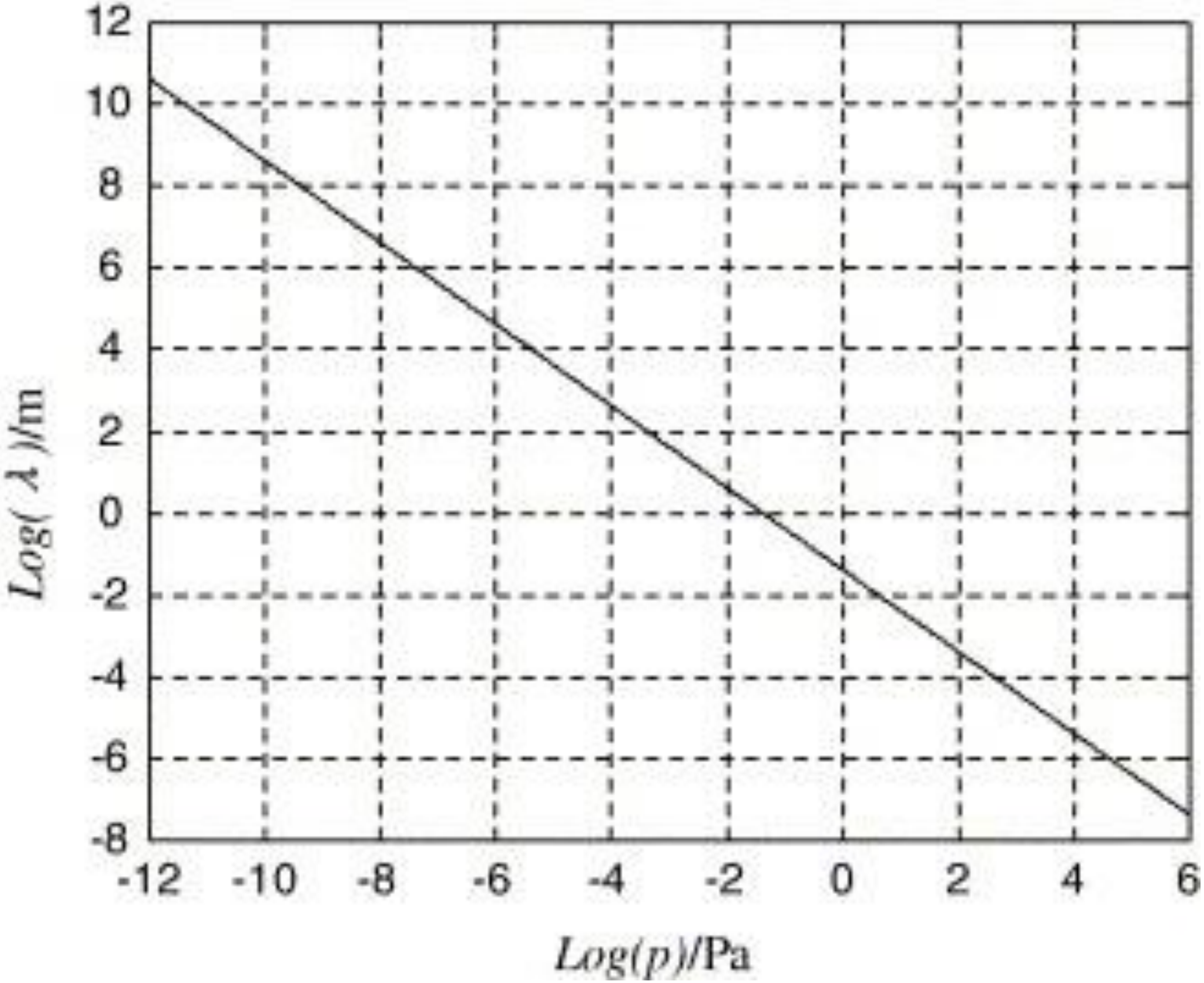
Properties of vacuum

pressure	monolayer time constant	molecular density	mean free path
p [mbar]	t (s)	n [m-3]	l [m]
1,00E+03	3E-09	2E+25	8E-09
1,00E+00	3E-06	2E+22	8E-06
1,00E-03	3E-03	2E+19	8E-03
1,00E-06	3E+00	2E+16	8E+00
1,00E-09	3E+03	2E+13	8E+03
1,00E-12	3E+06	2E+10	8E+06
1,00E-15	3E+09	2E+07	8E+09

Pressure regimes



Mean free path of electrons



Turbomolecular pump

Ion pump

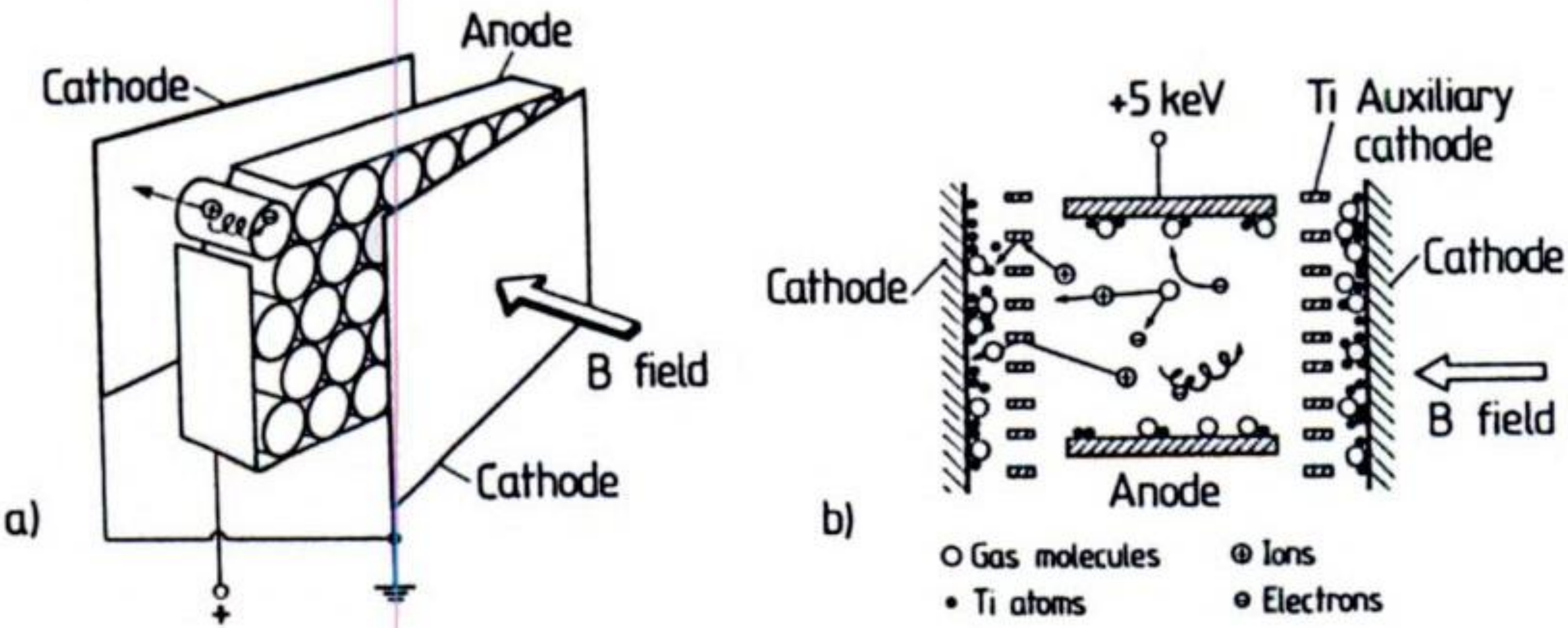


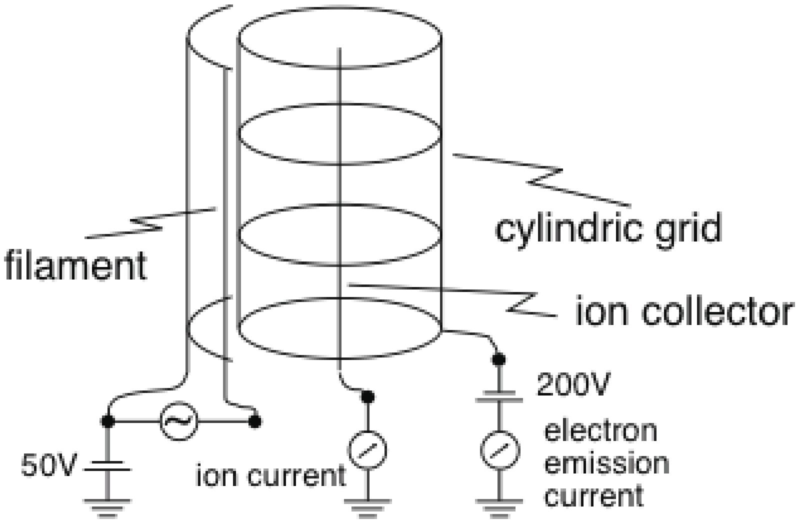
Fig. I.7a,b. Schematic view of an ion-getter pump: (**a**) The basic multicell arrangement. Each cell consists essentially of a tube-like anode. The cells are sandwiched between two common cathode plates of Ti, possibly together with auxiliary cathodes of Ti. (**b**) Detailed representation of the processes occurring within a single cell. Residual gas molecules are hit by electrons spiralling around the magnetic field B and are ionized. The ions are accelerated to the cathode and/or auxiliary cathode; they are trapped on the active cathode surface or they sputter Ti atoms from the auxiliary cathode, which in turn help to trap further residual gas ions

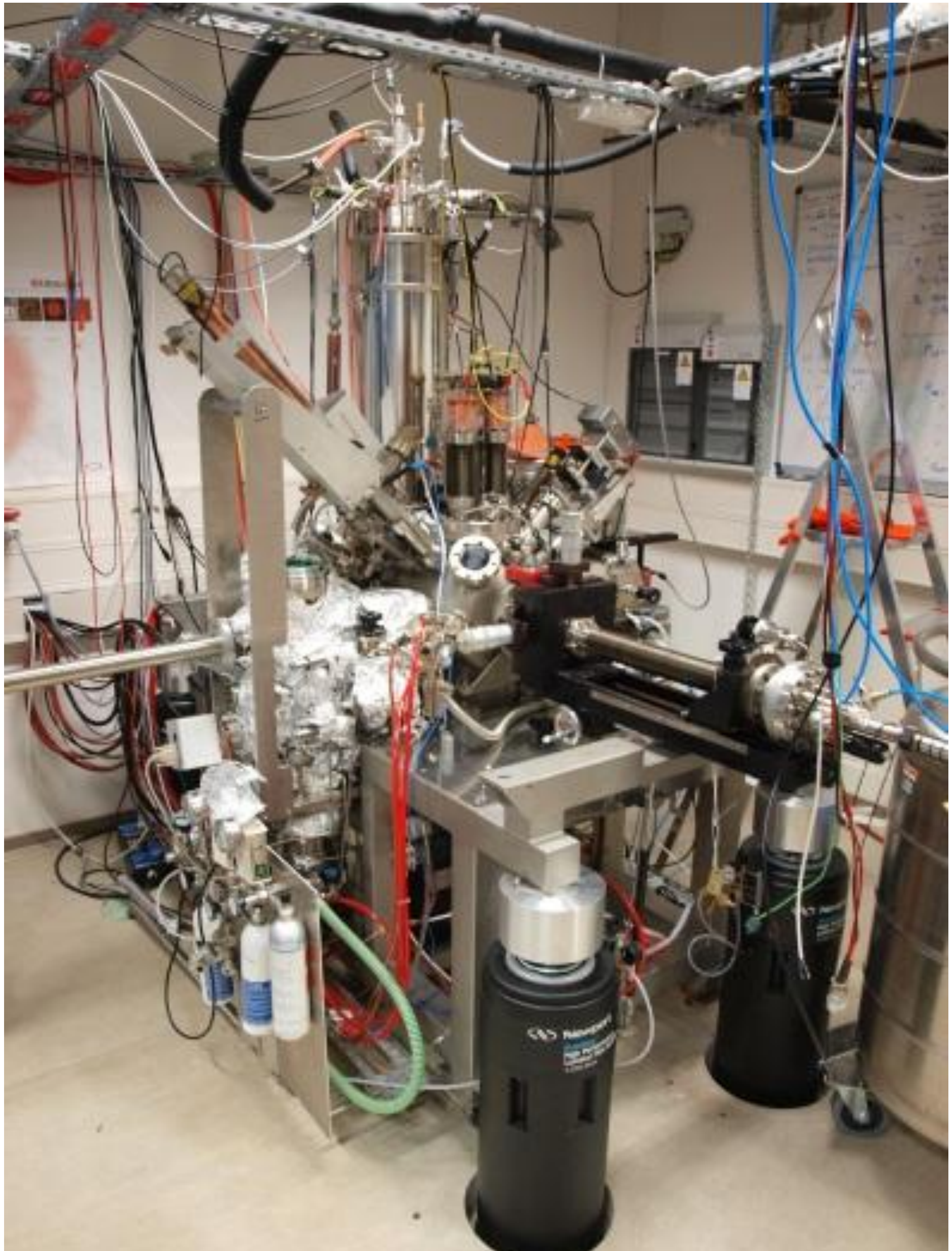
Vapor pressure

	10^{-10}mbar	10^{-6}mbar
Na	310K	400K
Zn	355K	450K
Cd	310K	390K
Hg	150K	230K
Mg	405K	505K
Al	860K	1100K
Fe	1000K	1300K
W	2160K	2680K

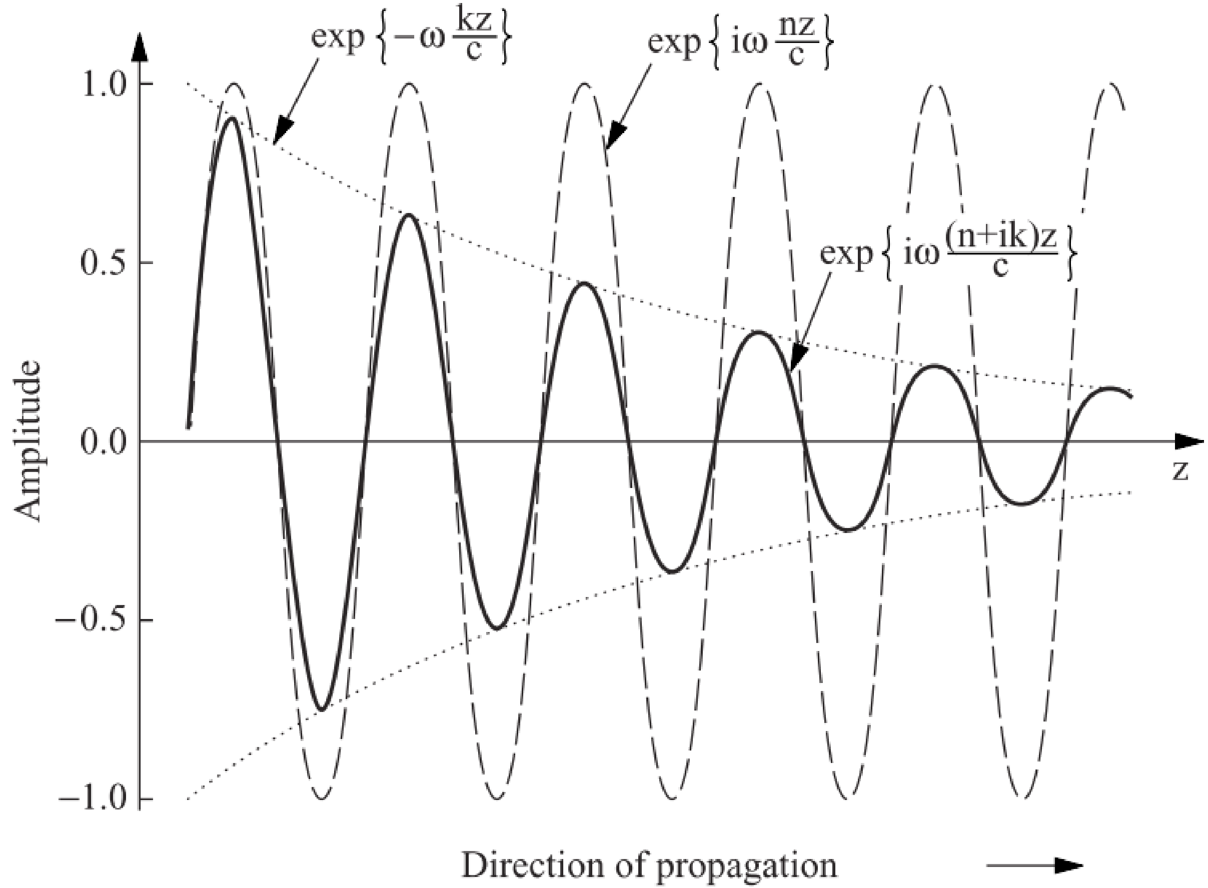
CF-flange

Ionization gauge





$$\vec{E} = \vec{E}_0 e^{-\frac{k\omega}{c}\vec{n}_q\cdot\vec{r}}\, e^{i(\frac{n\omega}{c}\vec{n}_q\cdot\vec{r}-\omega t)}$$



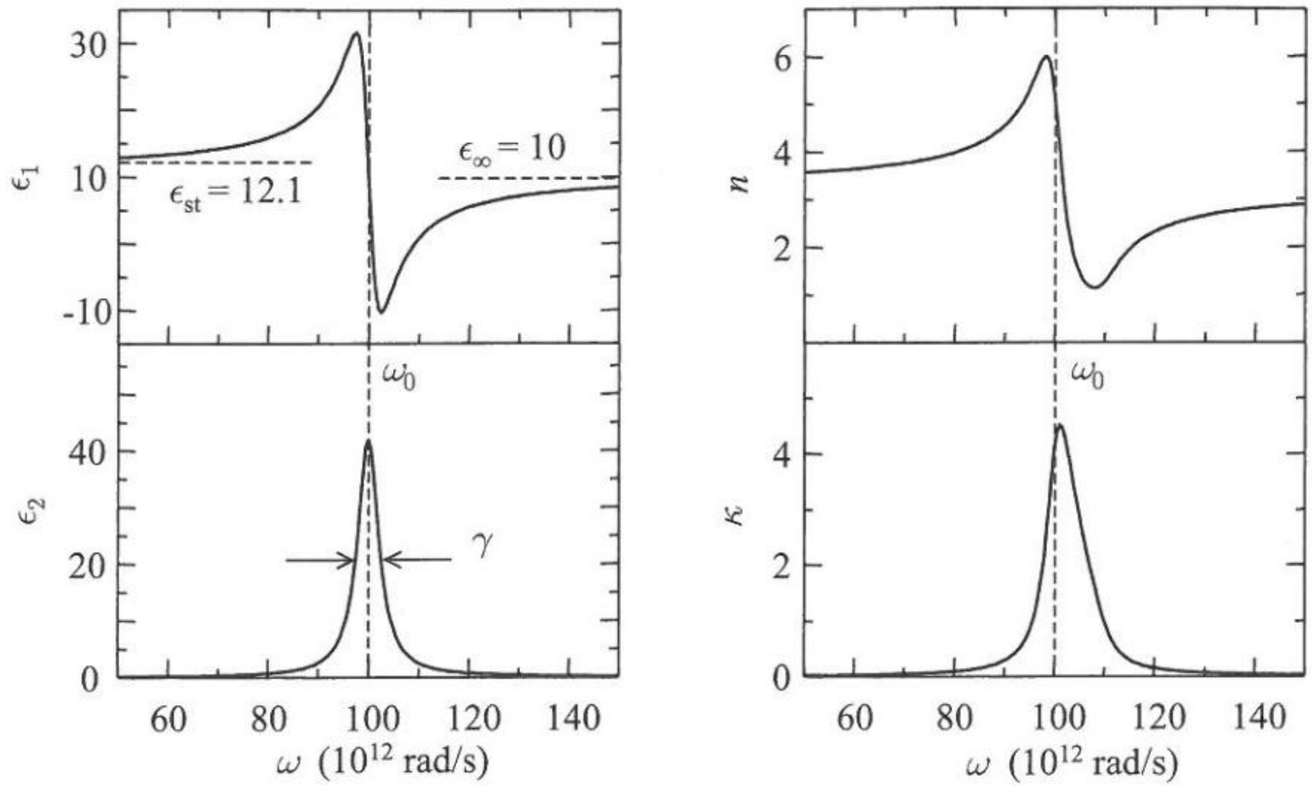
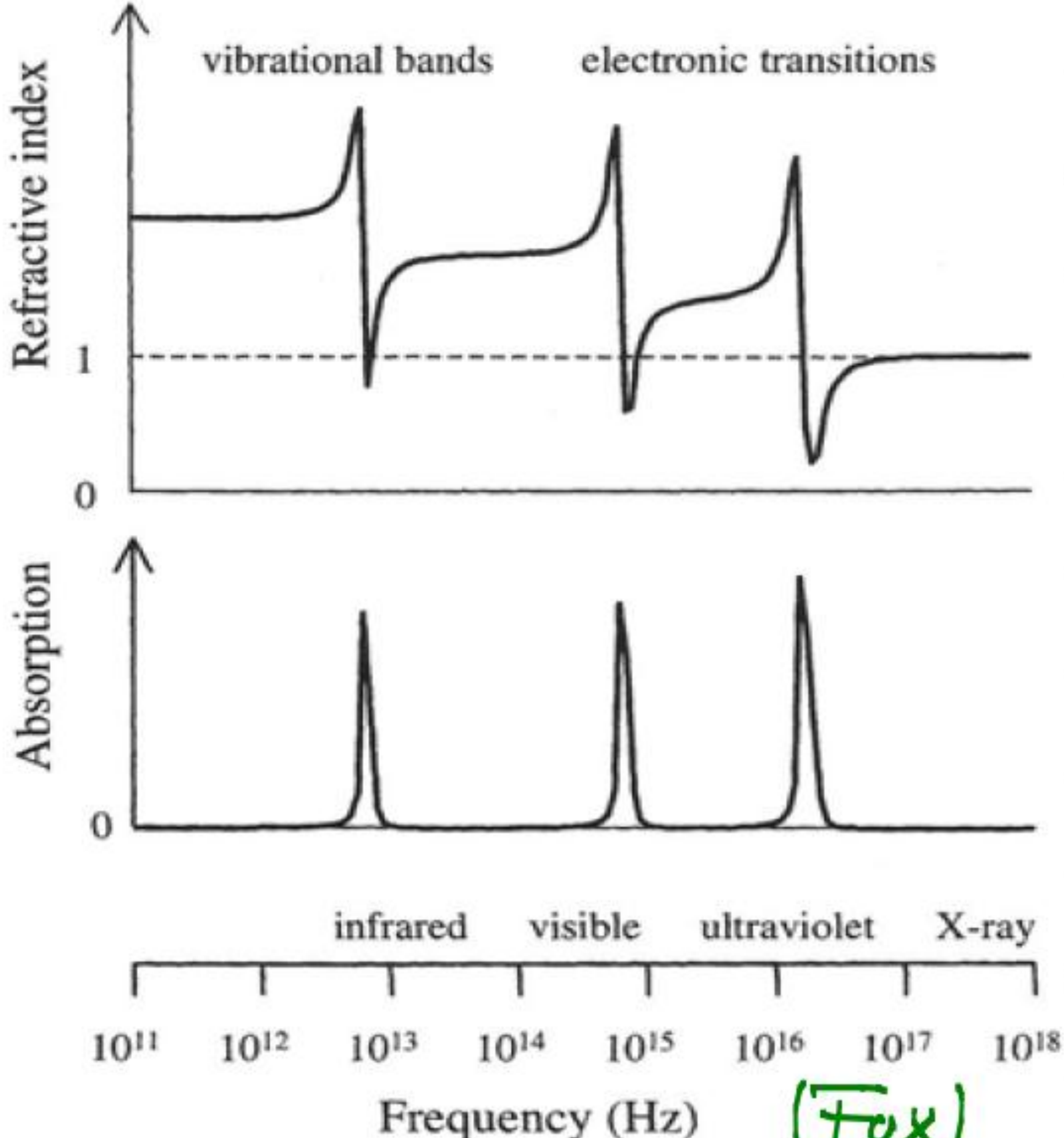
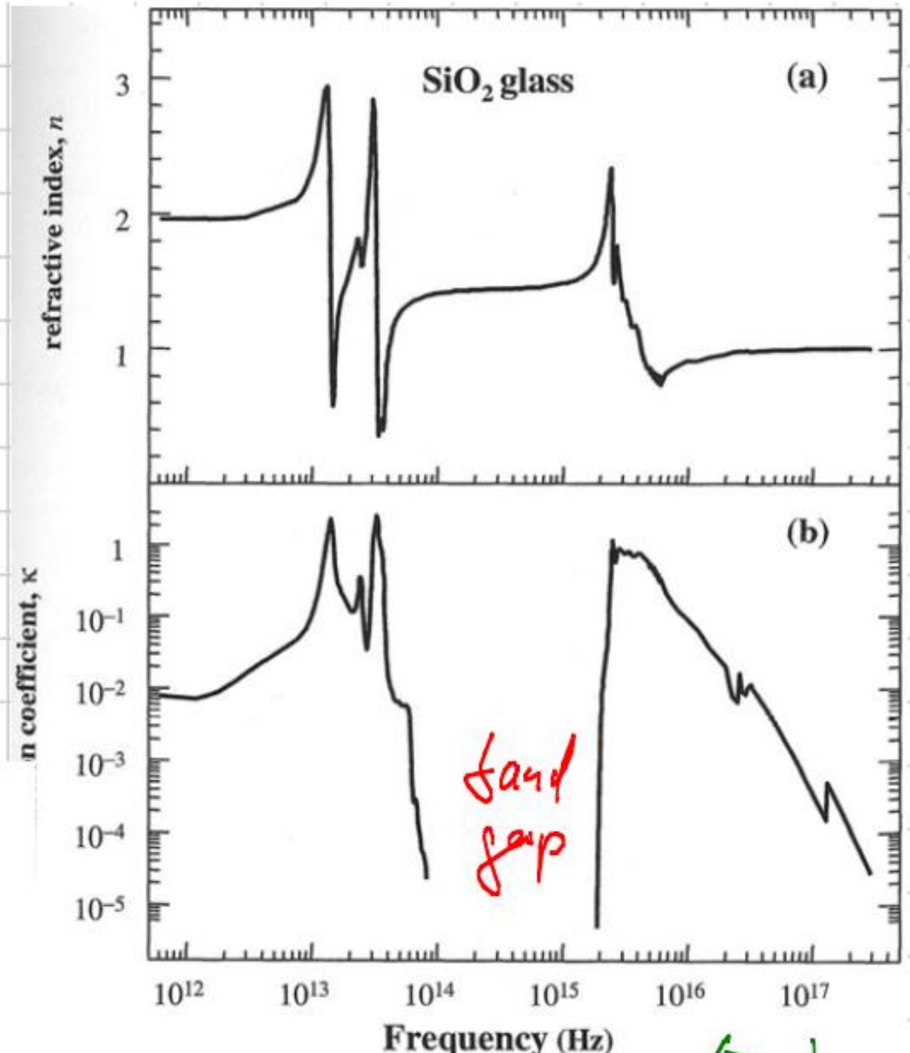


Fig. 2.4 Frequency dependence of the real and imaginary parts of the complex dielectric constant of a dipole oscillator at frequencies close to resonance. The graphs are calculated for an oscillator with $\omega_0 = 10^{14}$ rad/s, $\gamma = 5 \times 10^{12}\,\mathrm{s}^{-1}$, $\epsilon_{st} = 12.1$,(and $\epsilon_\infty = 10$. Also shown is the real and imaginary part of the refractive index calculated from the dielectric constant.


vibrational bands
electronic transitions
Refractive index
1
0
Absorption
0
infrared
visible
ultraviolet
X-ray
10^{11}
10^{12}
10^{13}
10^{14}
10^{15}
10^{16}
10^{17}
10^{18}
Frequency (Hz)

(Fox)


SiO$_2$ glass
(a)
(b)
refractive index, n
n coefficient, κ
3
2
1
1
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{12}
10^{13}
10^{14}
10^{15}
10^{16}
10^{17}
Frequency (Hz)
band gap

(Fox)

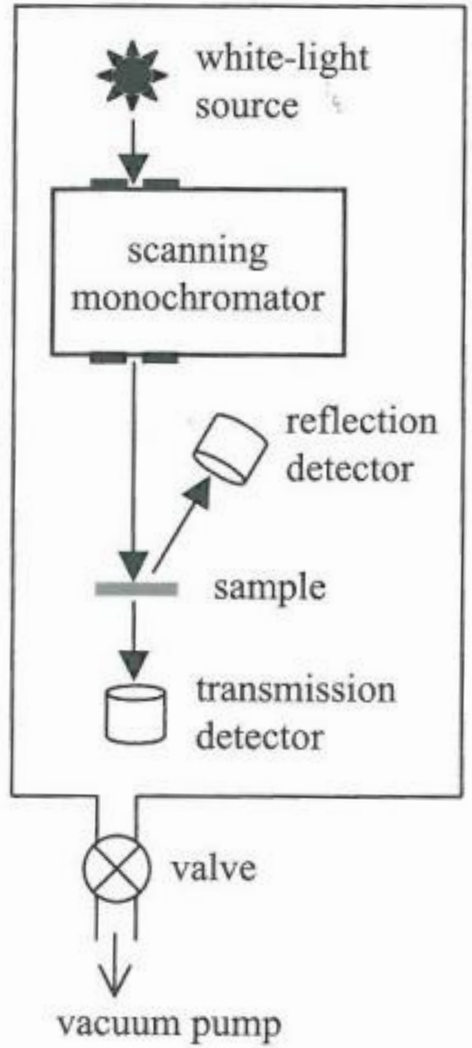
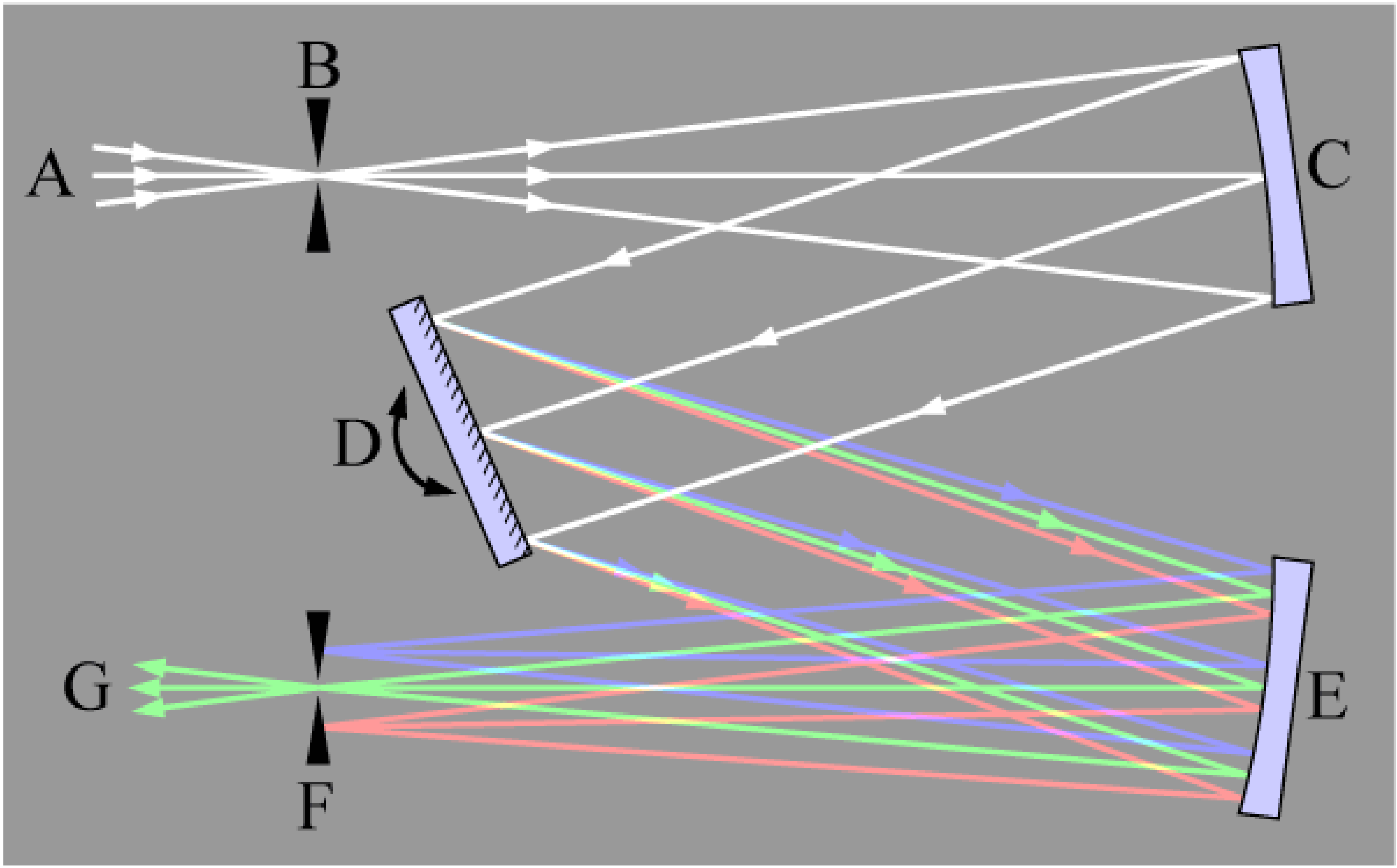


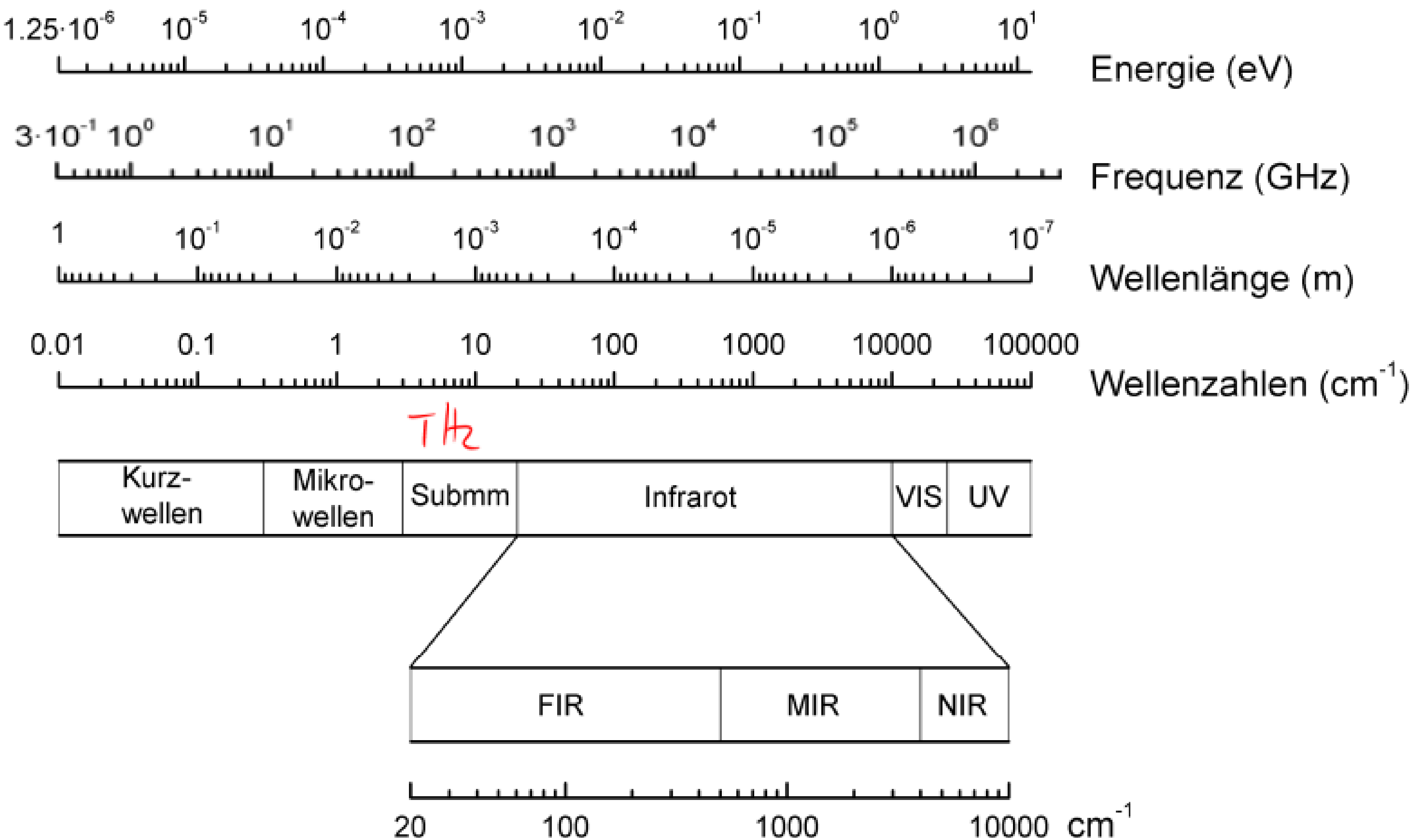
Fig. 3.14 Schematic diagram of the experimental arrangement required to determine the absorption coefficient over a wide spectral range by making reflectivity and transmissivity measurements.

Czerny-Turner monochromator



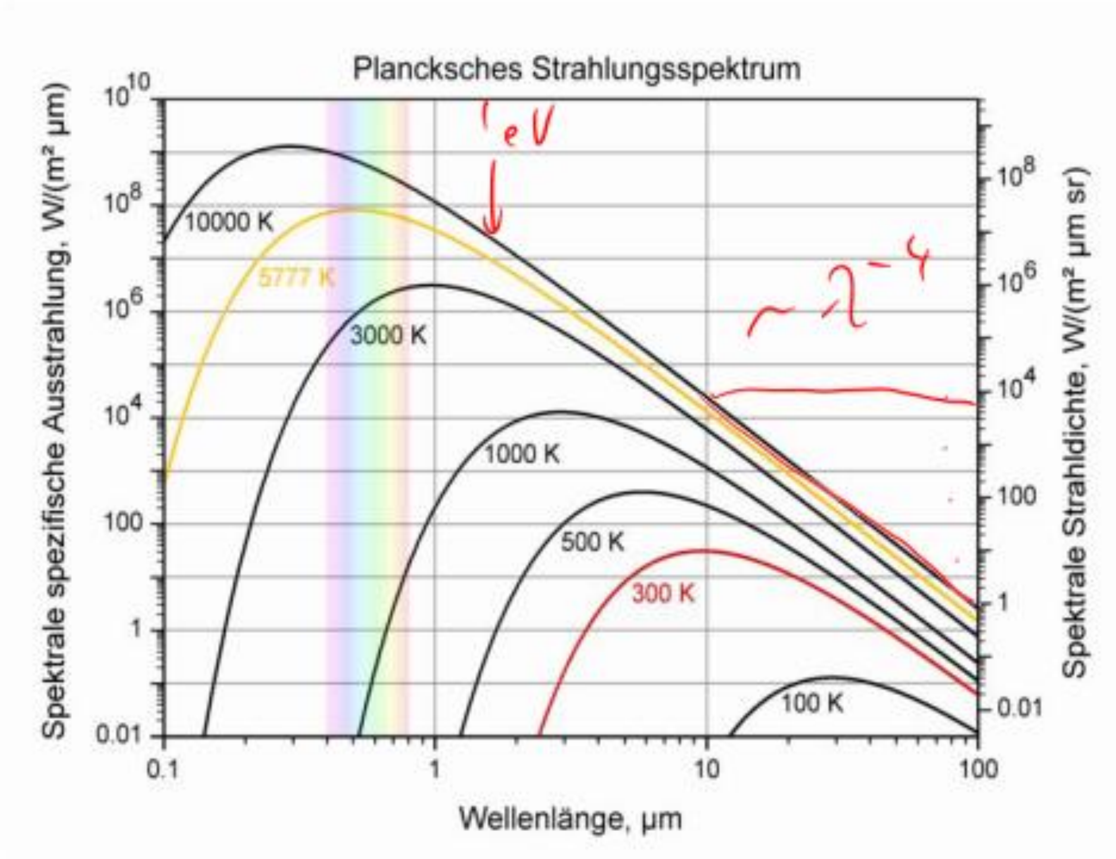
Spectal region	Wavelength (nm)	Source	detector
Infrared	> 1600	Black body	Cooled semiconductor
Near infrared	700-1600	Black body	Semiconductor
Visible	400-700	Black Body	Photomultiplier
Ultraviolet	200-400	Xenon lamp	Photomultiplier

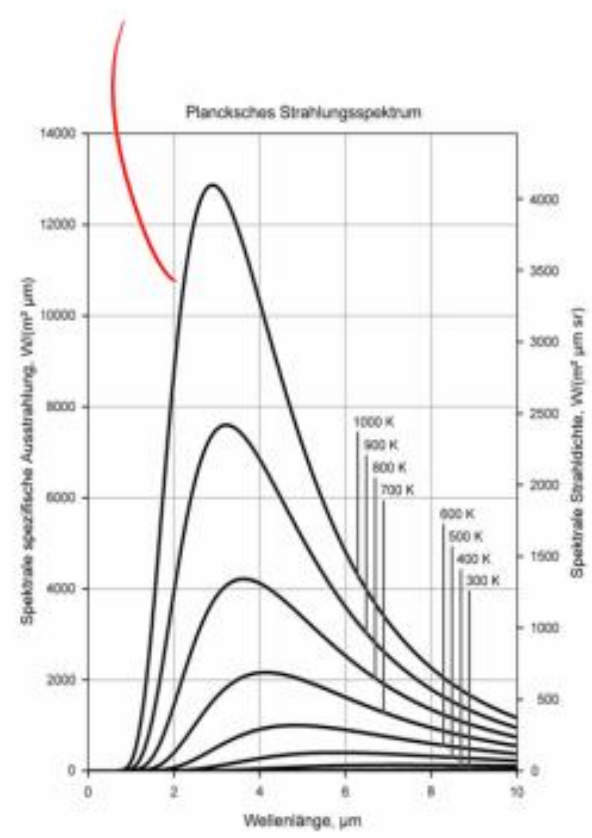
Spectral units



A black body's radiation

$e^{-1/\lambda}$





Photomultiplier

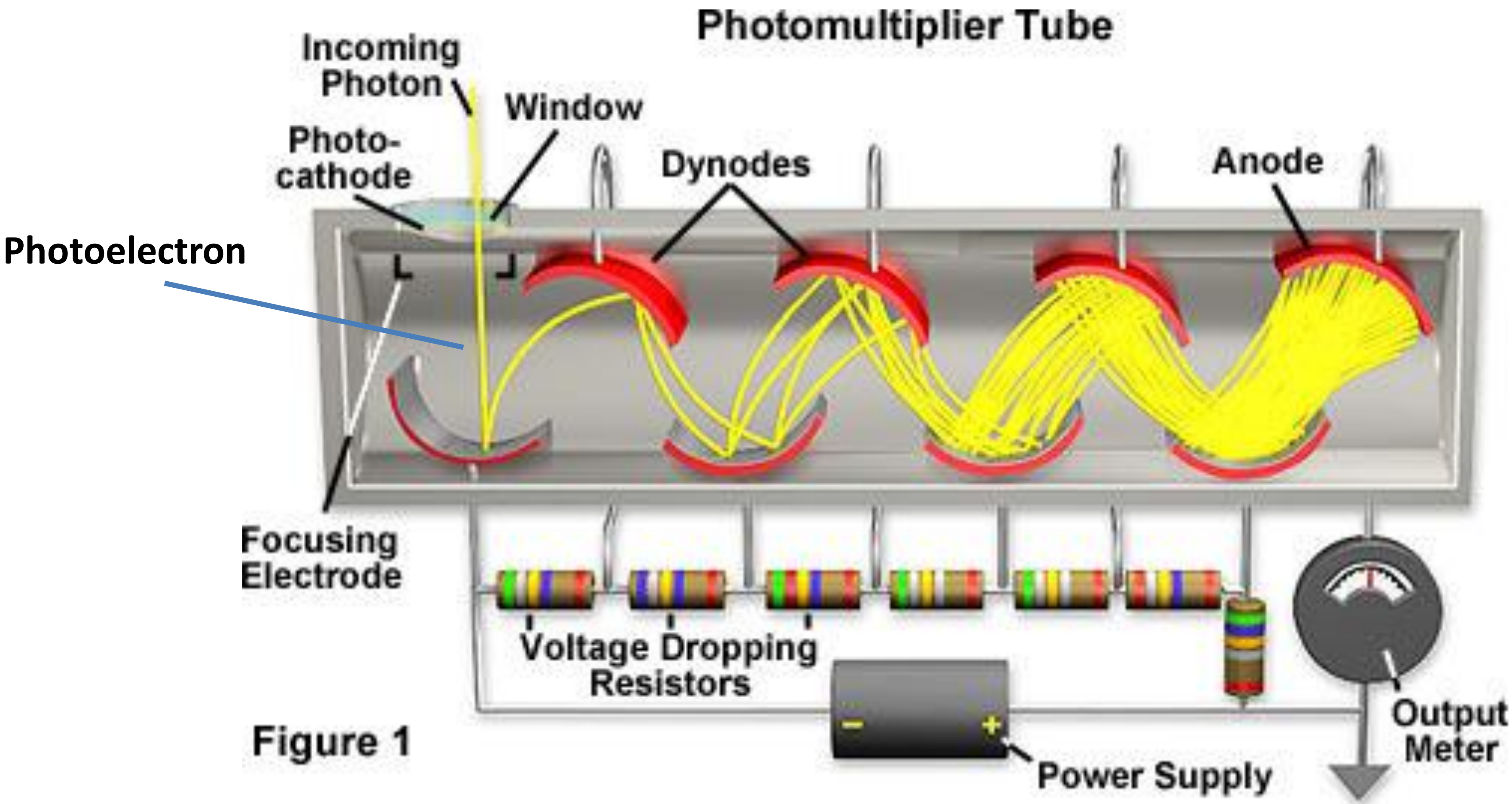
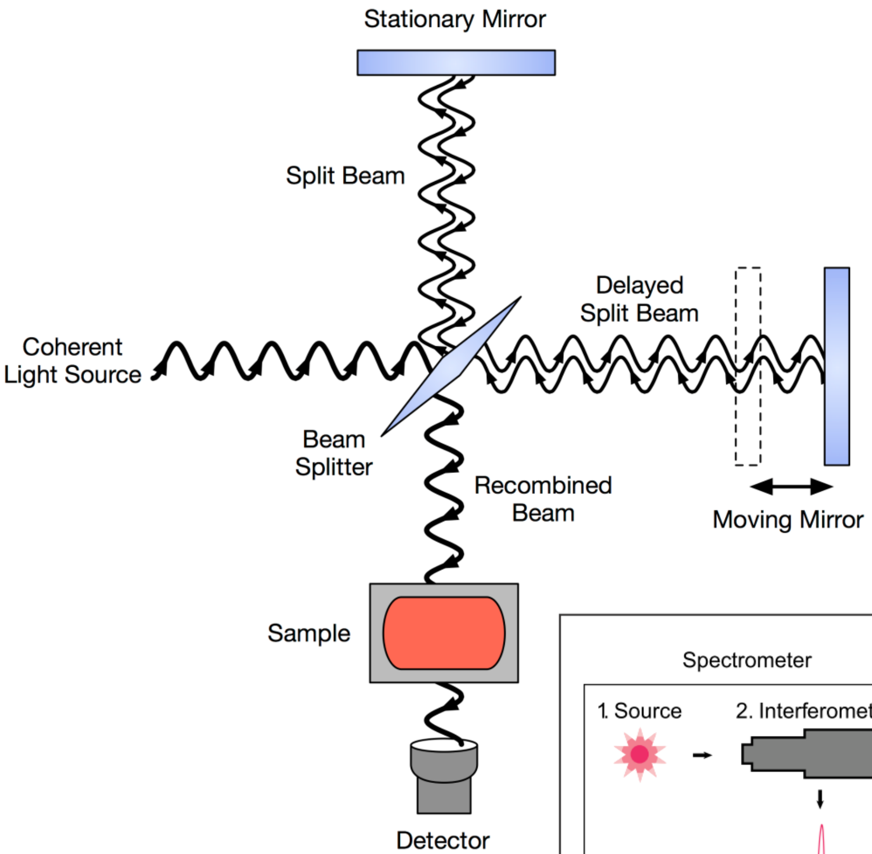
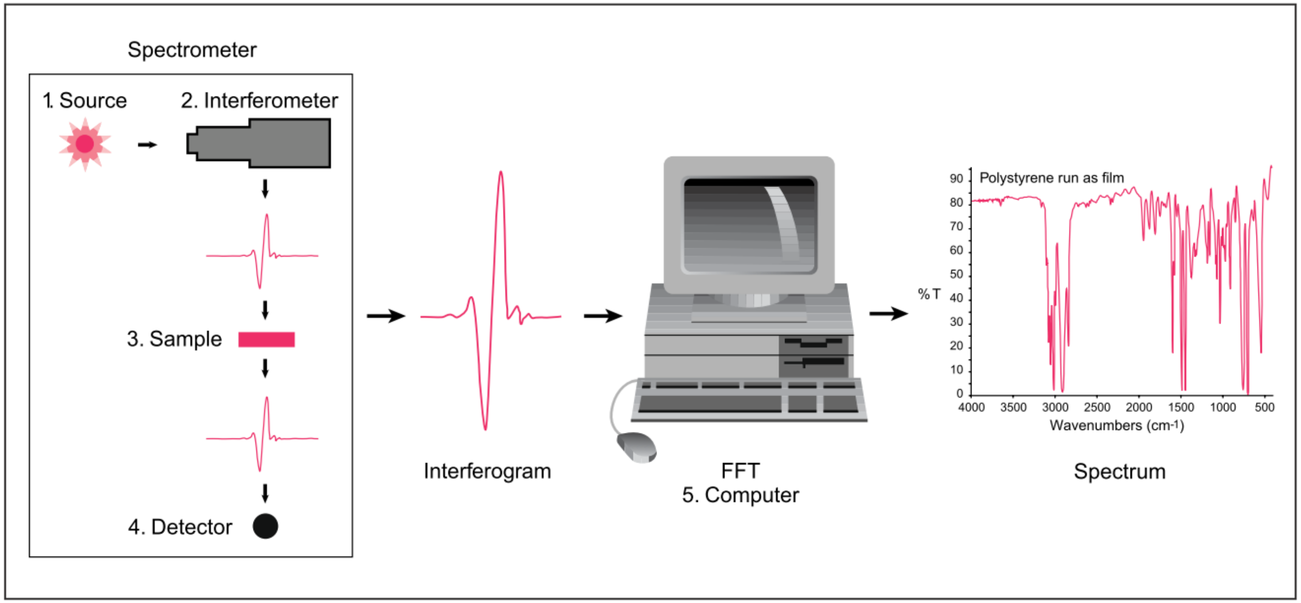


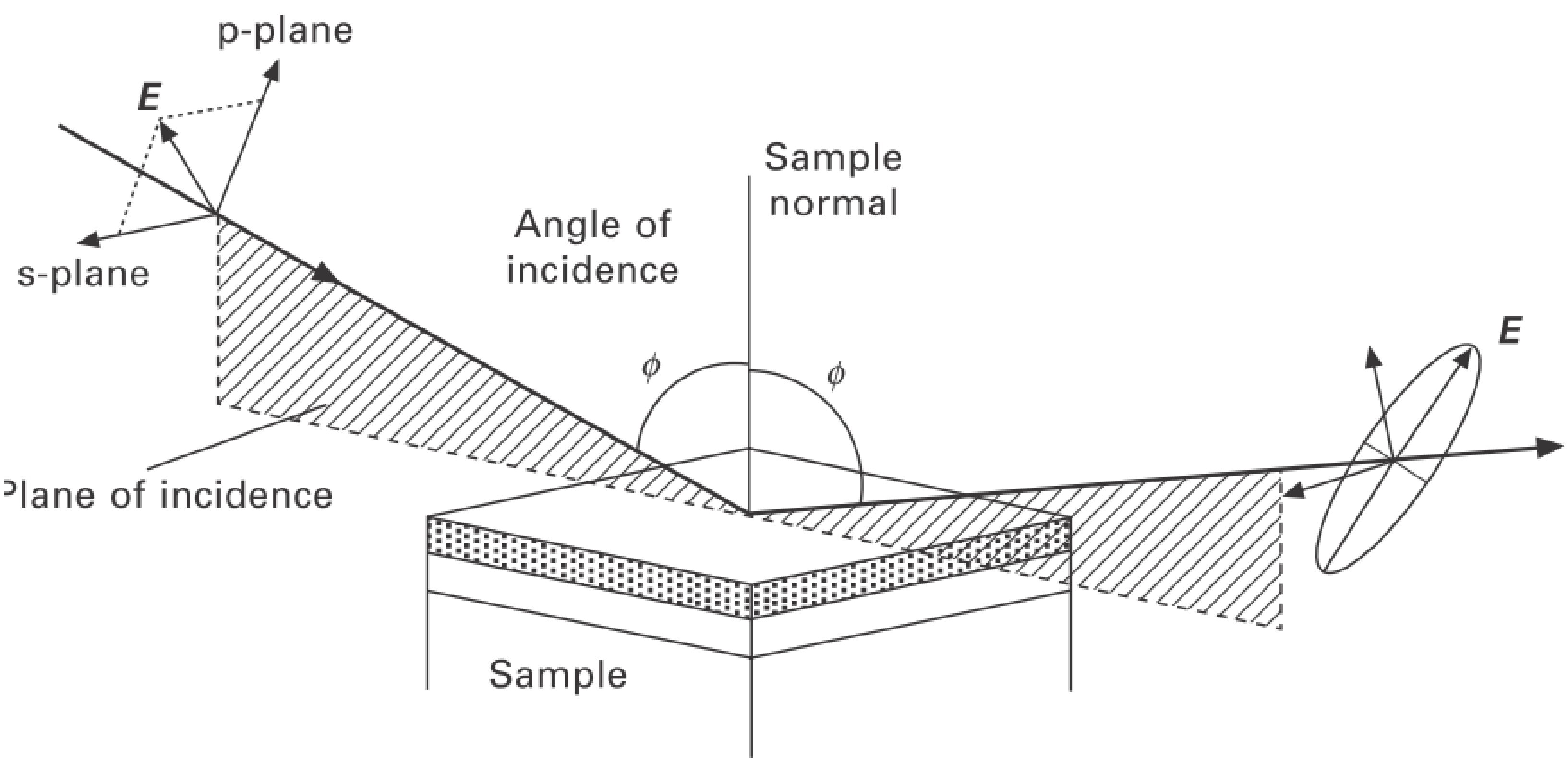
Figure 1

Fourier-transform infrared spectrometer



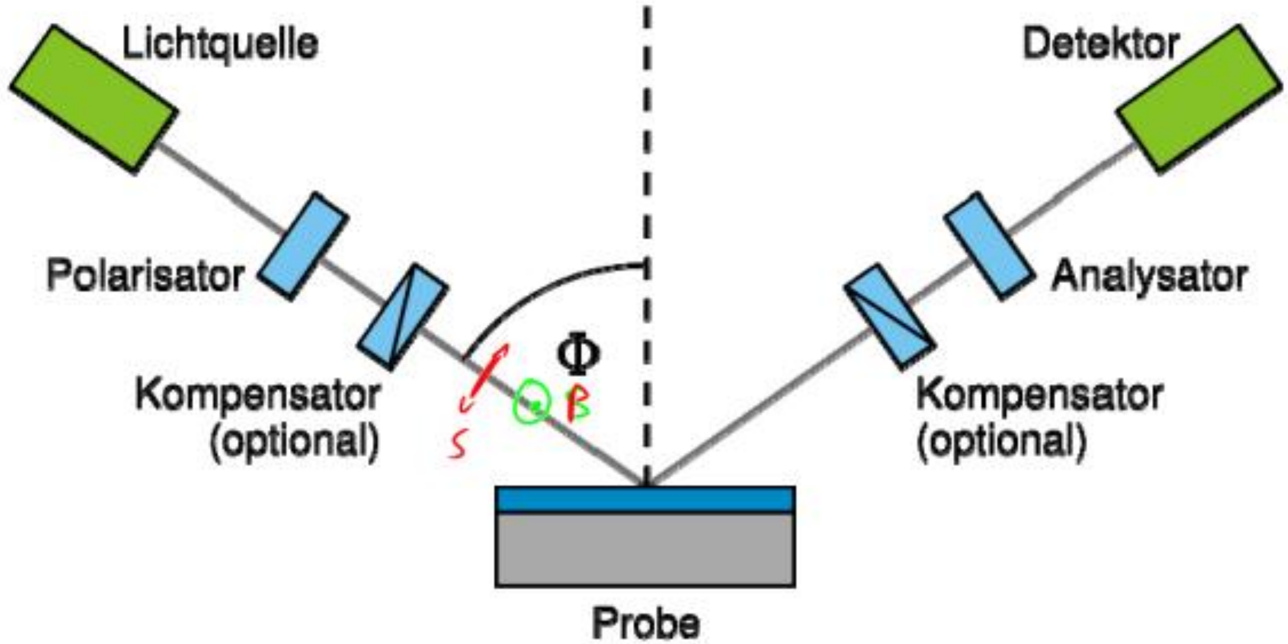


Ellipsometry



5.1 When linearly polarized incident light, consisting of p- and s-orthogonal polarization components, is reflected from a surface at oblique angle of incidence (ϕ) the result is often elliptical polarization. Ellipsometry measurements determine the change in polarization that occurs when light interacts with the sample.

Ellipsometry



$$\rho = \frac{R_p}{R_s} = \tan\Psi \exp(i\Delta)$$ $\rightarrow$ n, k $\quad \varepsilon', \varepsilon''$

Optical conductivity

- complex dielectric constant: $\epsilon = \epsilon_1 + i\epsilon_2$
- by definition: $\sqrt{\epsilon} = n + ik = \hat{n}$

$$\boxed{\epsilon_1 = n^2 - k^2 \qquad \epsilon_2 = 2nk}$$

- with complex conductivity: $\sigma = \sigma_1 + i\sigma_2$

  and $\epsilon = 1 + \dfrac{i\sigma}{\epsilon_0\omega}$

$$\boxed{\sigma_1 = \epsilon_0\epsilon_2\omega \qquad \sigma_2 = \epsilon_0(1 - \epsilon_1)\omega}$$

	Dielectric constant $\hat{\epsilon}$	Conductivity $\hat{\sigma}$	Refractive index $\hat{N}$
$\hat{\epsilon}$	$\hat{\epsilon} = \epsilon_1 + i\epsilon_2$	$\epsilon_1 = 1 - \frac{4\pi\sigma_2}{\omega}$ $\epsilon_2 = \frac{4\pi\sigma_1}{\omega}$	$\epsilon_1 = \frac{n^2-k^2}{\mu_1}$ $\epsilon_2 = \frac{2nk}{\mu_1}$
$\hat{\sigma}$	$\sigma_1 = \frac{\omega\epsilon_2}{4\pi}$ $\sigma_2 = (1-\epsilon_1)\frac{\omega}{4\pi}$	$\hat{\sigma} = \sigma_1 + i\sigma_2$	$\sigma_1 = \frac{nk\omega}{2\pi\mu_1}$ $\sigma_2 = \left(1 - \frac{n^2-k^2}{\mu_1}\right)\frac{\omega}{4\pi}$
$\hat{N}$	$n = \left\{\frac{\mu_1}{2}\left[\epsilon_1^2 + \epsilon_2^2\right]^{1/2} + \frac{\epsilon_1\mu_1}{2}\right\}^{1/2}$ $k = \left\{\frac{\mu_1}{2}\left[\epsilon_1^2 + \epsilon_2^2\right]^{1/2} - \frac{\epsilon_1\mu_1}{2}\right\}^{1/2}$	$n = \left\{\frac{\mu_1}{2}\left[\left(1 - \frac{4\pi\sigma_2}{\omega}\right)^2 + \left(\frac{4\pi\sigma_1}{\omega}\right)^2\right]^{1/2} + \frac{\mu_1}{2} - \frac{2\pi\mu_1\sigma_2}{\omega}\right\}^{1/2}$ $k = \left\{\frac{\mu_1}{2}\left[\left(1 - \frac{4\pi\sigma_2}{\omega}\right)^2 + \left(\frac{4\pi\sigma_1}{\omega}\right)^2\right]^{1/2} - \frac{\mu_1}{2} + \frac{2\pi\mu_1\sigma_2}{\omega}\right\}^{1/2}$	$\hat{N} = n + ik$

Table 7.1 Free electron density and plasma properties of some metals. The figures are for room temperature unless stated otherwise. The electron densities are based on data taken from Wyckoff (1963). The plasma frequency ω_p is calculated from eqn 7.6, and λ_p is the wavelength corresponding to this frequency.

Metal	Valency	N ($10^{28}\,\mathrm{m}^{-3}$)	$\omega_p/2\pi$ (10^{15} Hz)	λ_p (nm)
Li (77 K)	1	4.70	1.95	154
Na (5 K)	1	2.65	1.46	205
K (5 K)	1	1.40	1.06	282
Rb (5 K)	1	1.15	0.96	312
Cs (5 K)	1	0.91	0.86	350
Cu	1	8.47	2.61	115
Ag	1	5.86	2.17	138
Au	1	5.90	2.18	138
Be	2	24.7	4.46	67
Mg	2	8.61	2.63	114
Ca	2	4.61	1.93	156
Al	3	18.1	3.82	79

Experimentelles Beispiel: Aluminium