Applications of Synchrotrons Radiation

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A word about the Ecole Polytechnique Fédérale de Lausanne (EPFL):

Site of the Hydroptère research: world record for sailing speed

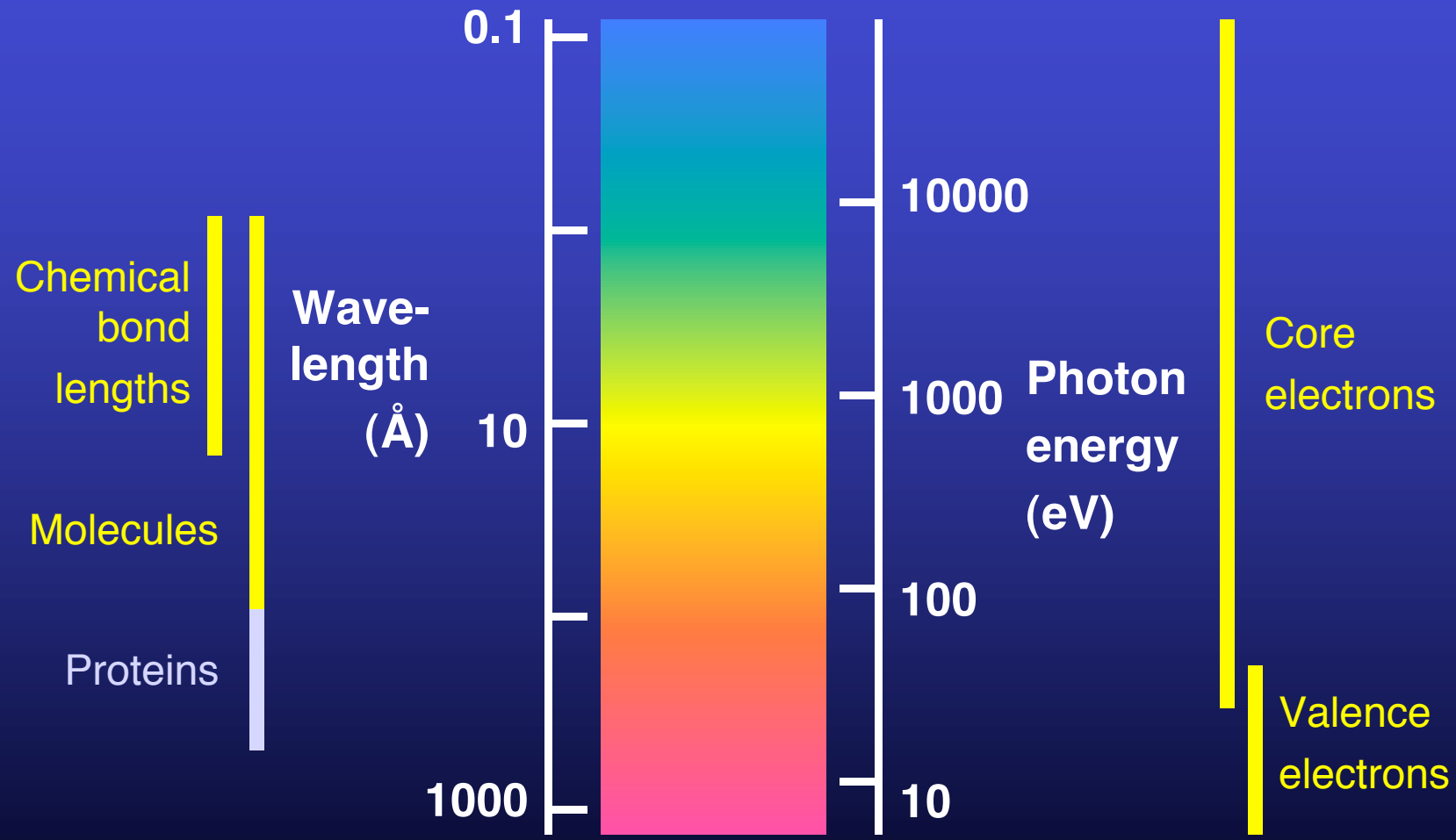
Birthplace of the mouse, >1 billion Logitech mice ago

- The most international engineering-technology university in the world (Times Higher Education)
- The top engineering-technology university in Europe, with Cambridge (Shanghai ranking)
- English is our official language for graduate education

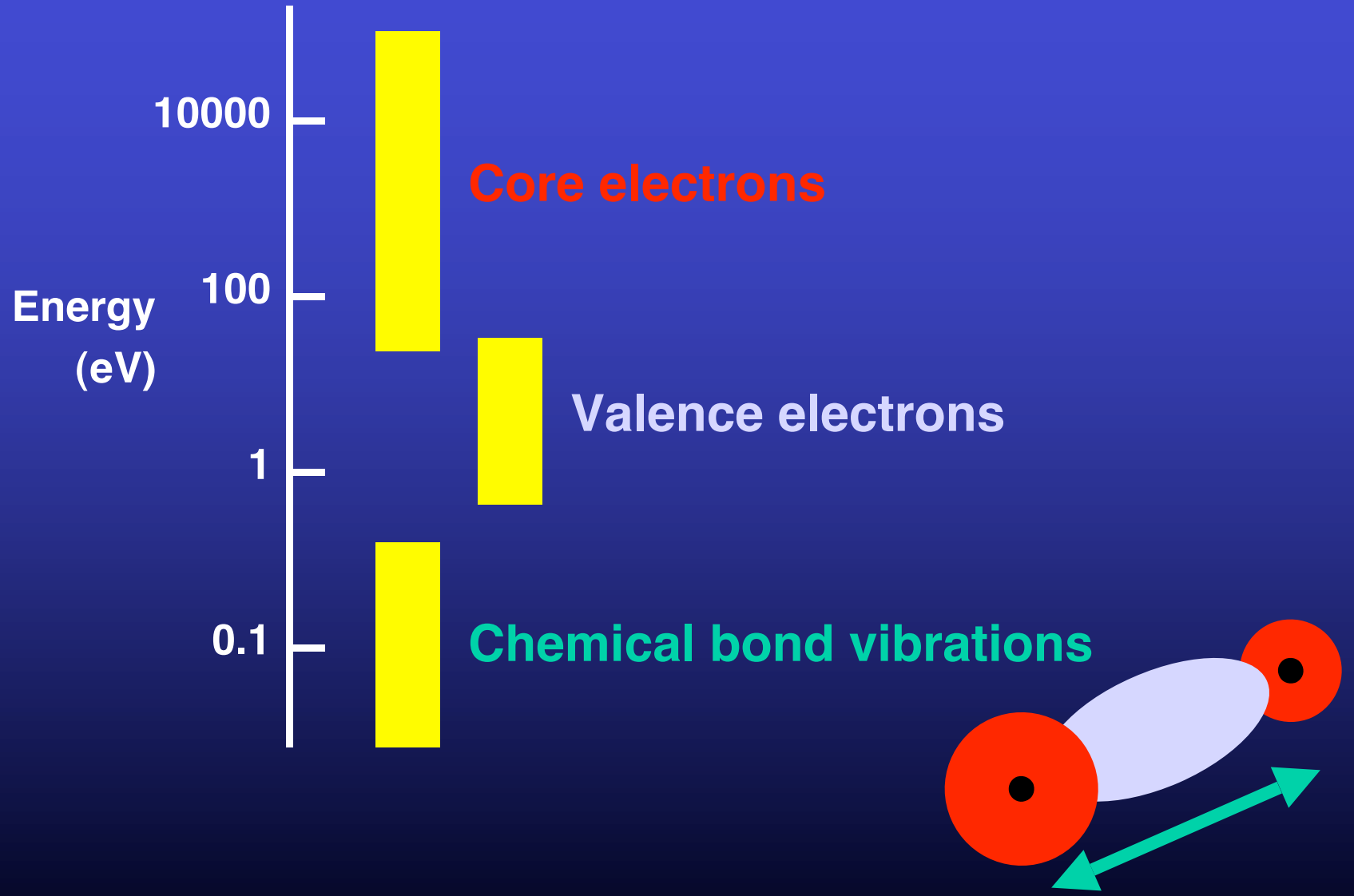
Outline:

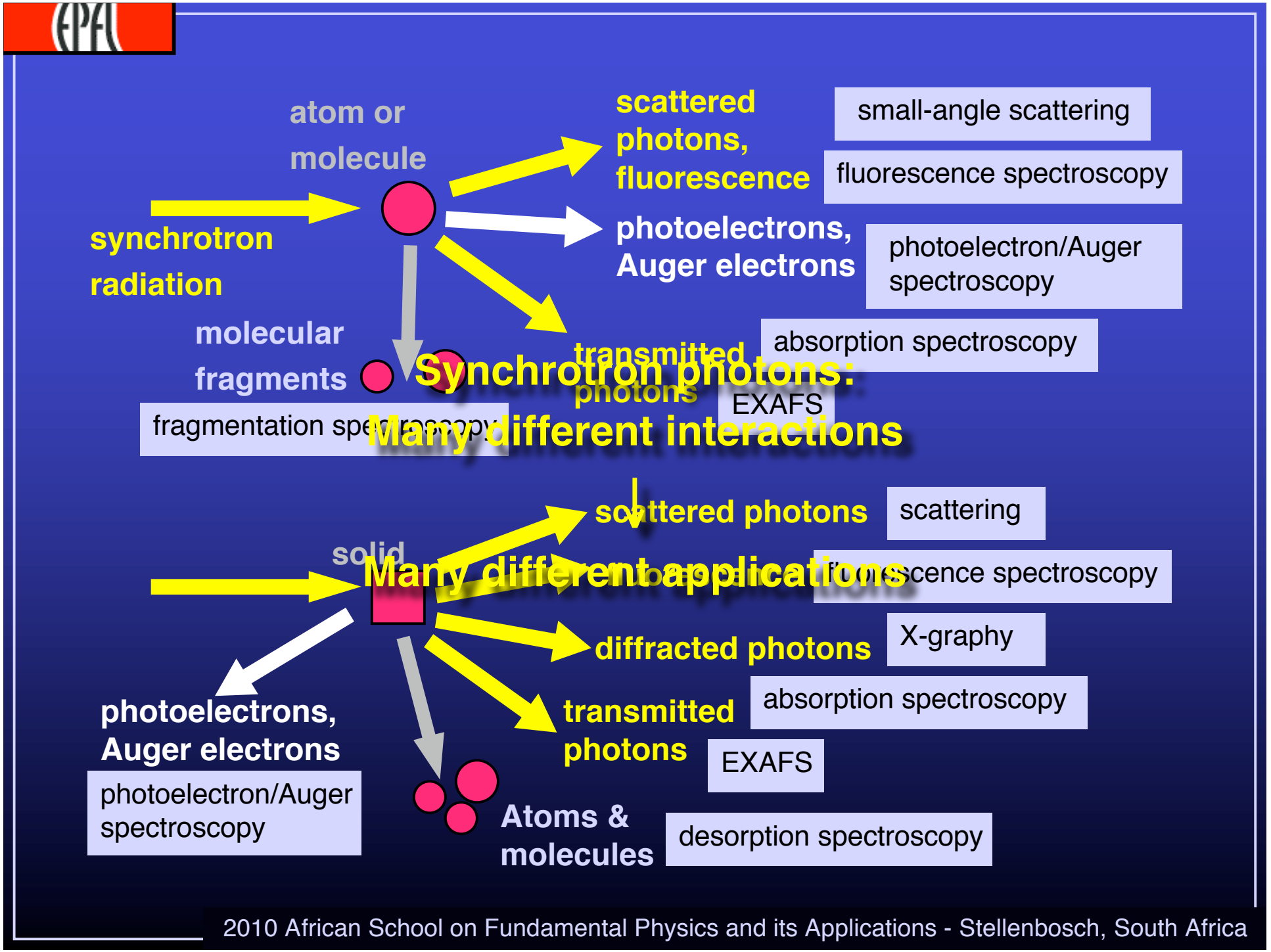
- Why do we need x-rays?
- Spectroscopy and spectromicroscopy
- Scattering and diffraction -- protein crystallography
- Coherence: a revolution in radiology and x-ray tomography
- Future: from storage rings to free electron lasers

Why x-rays and ultraviolet?

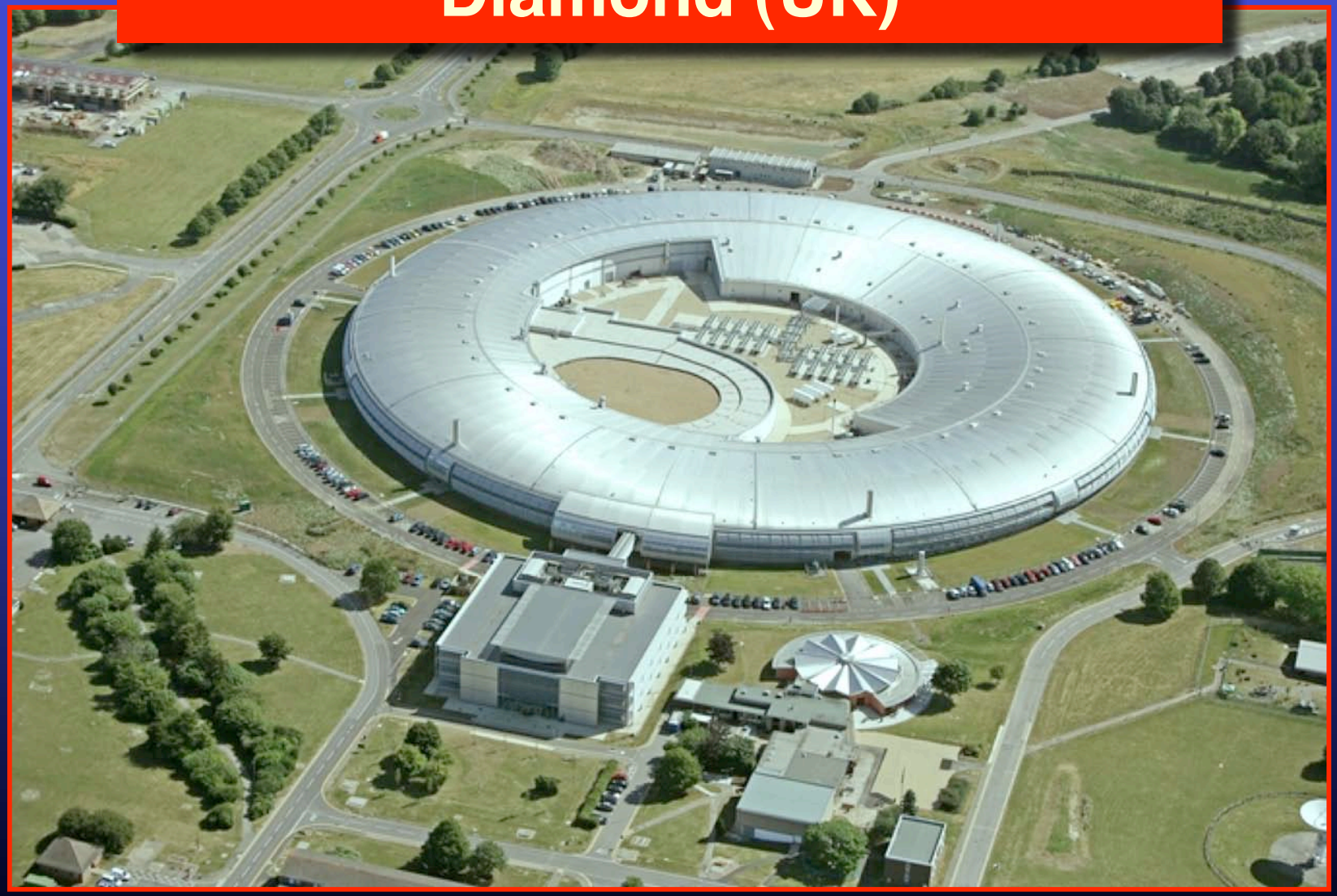


Over a broader spectrum:

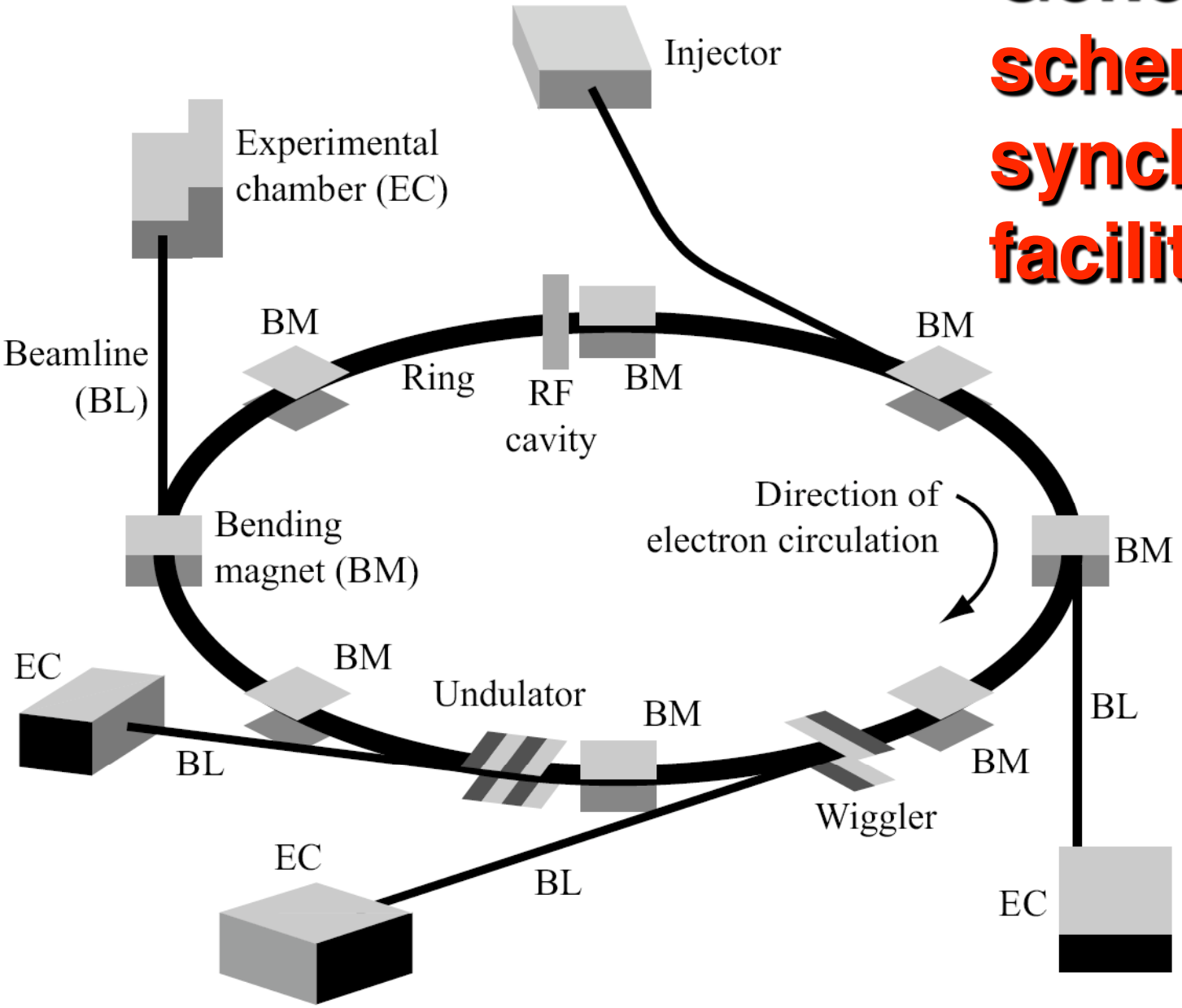



atom or molecule
synchrotron radiation
scattered photons, fluorescence
small-angle scattering
fluorescence spectroscopy
photoelectrons, Auger electrons
photoelectron/Auger spectroscopy
absorption spectroscopy
molecular fragments
fragmentation spectroscopy
transmitted photons
EXAFS
Synchrotron photons:
Many different interactions
Many different applications
scattered photons
scattering
fluorescence spectroscopy
diffracted photons
X-graphy
absorption spectroscopy
transmitted photons
EXAFS
photoelectrons, Auger electrons
photoelectron/Auger spectroscopy
Atoms & molecules
desorption spectroscopy

A real synchrotron facility: Diamond (UK)



General scheme of a synchrotron facility

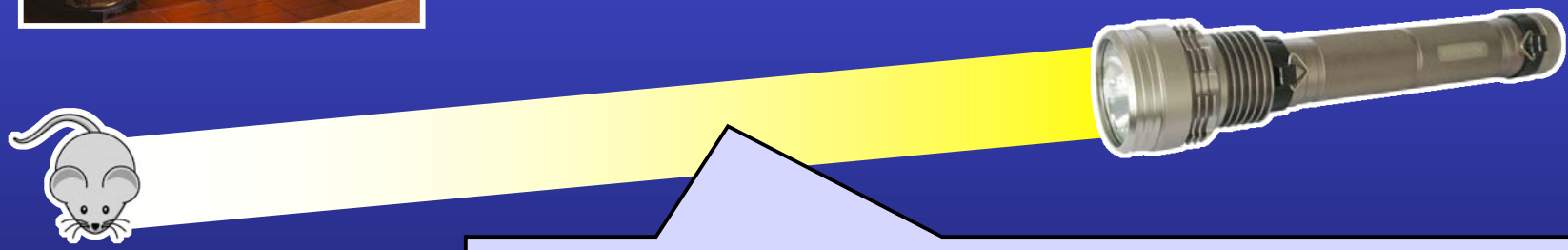


Synchrotron sources offer many advantages for practical applications:

- High photon flux
- Tunable wavelength/photon energy over a broad band
- High brightness/brilliance
- Polarization
- Time structure
- Coherence

Fireplaces and torchlights :

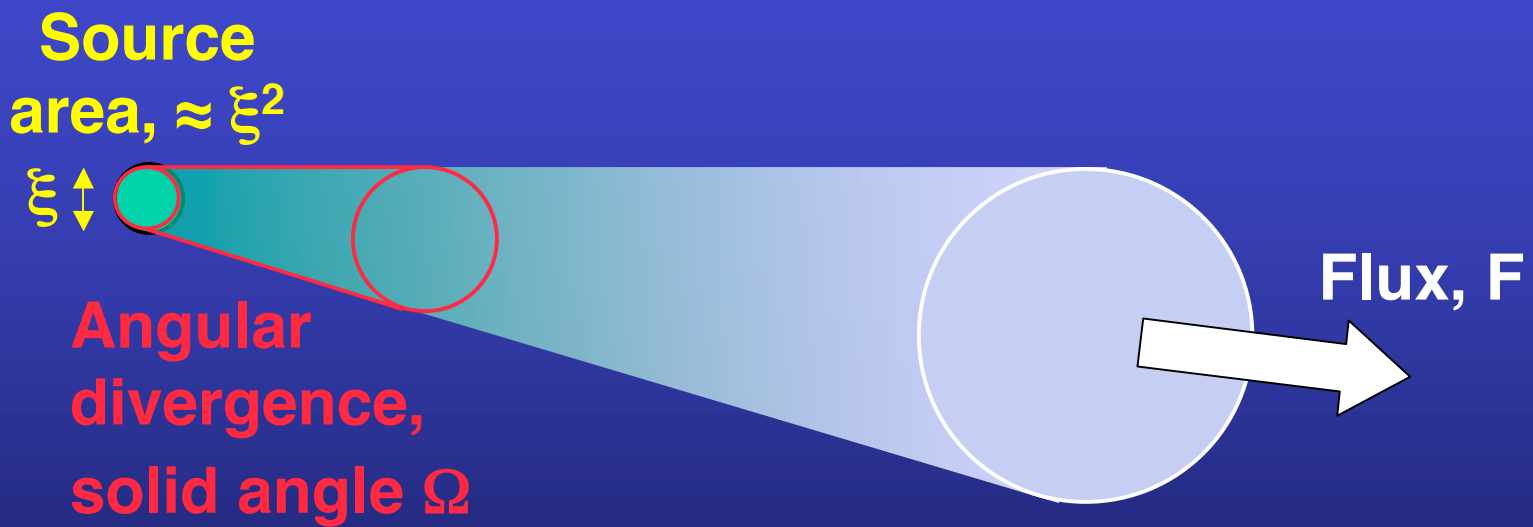
A fireplace is not very effective in "illuminating" a specific target: its emitted power is spread in all directions



A torchlight is much more effective: it is a small-size source with emission concentrated within a narrow angular spread

This can be expressed using the “brightness”

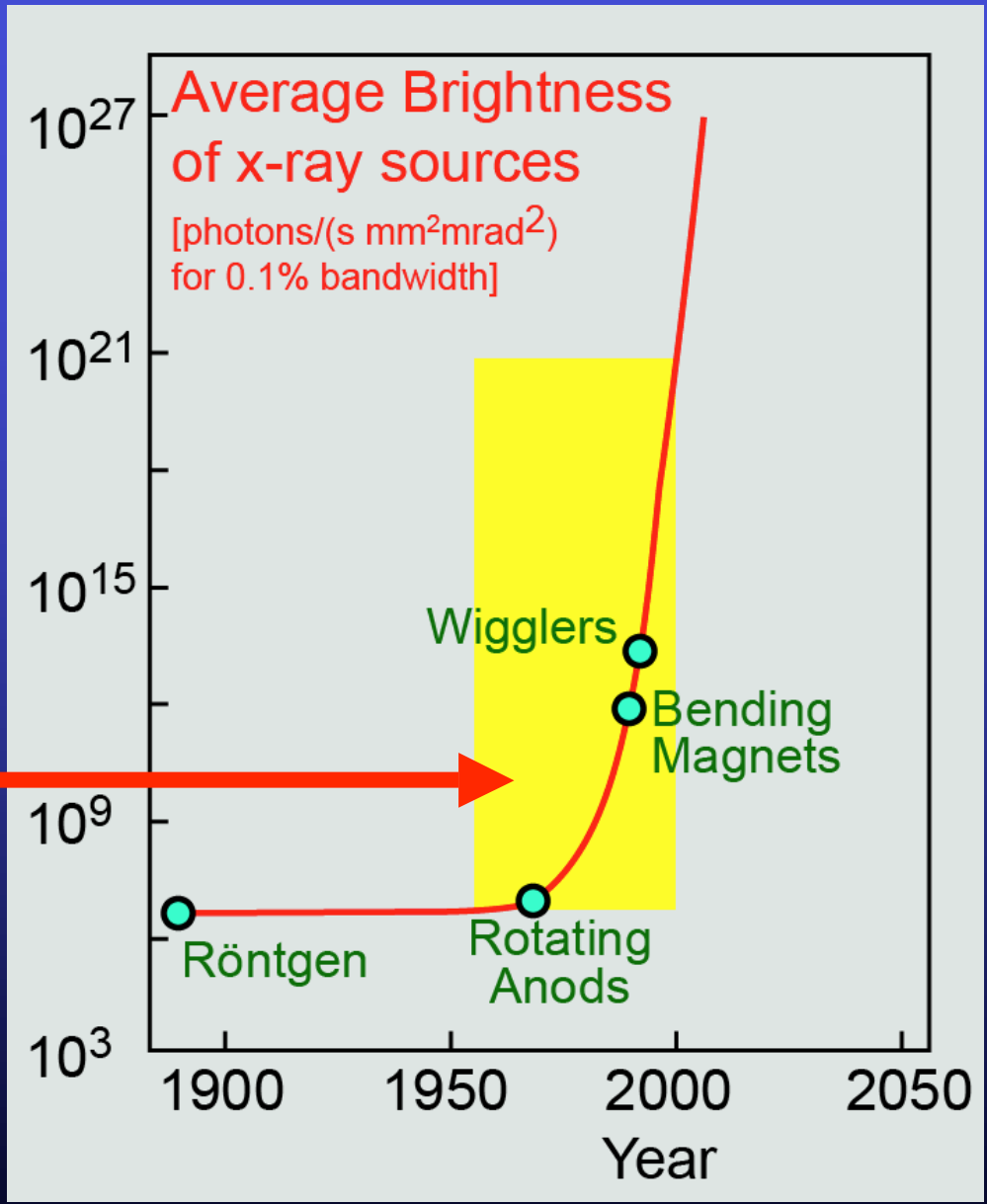
The "brightness" (or brilliance) of a source of light :

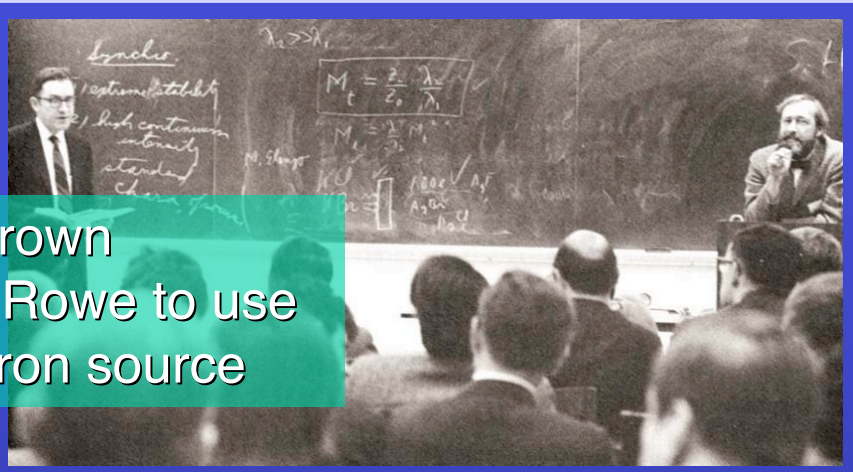


$$\text{Brightness} = \text{constant} \frac{F}{\xi^2 \Omega}$$

The historical growth in x-ray brightness

Between 1955 and 2000, the brightness increased by more than 15 orders of magnitude… whereas the top power of computing increased “only” by 6-7 orders of magnitude





The early steps -- 1966: Fred Brown proposes to Tantalus' father Ed Rowe to use it as the first dedicated synchrotron source

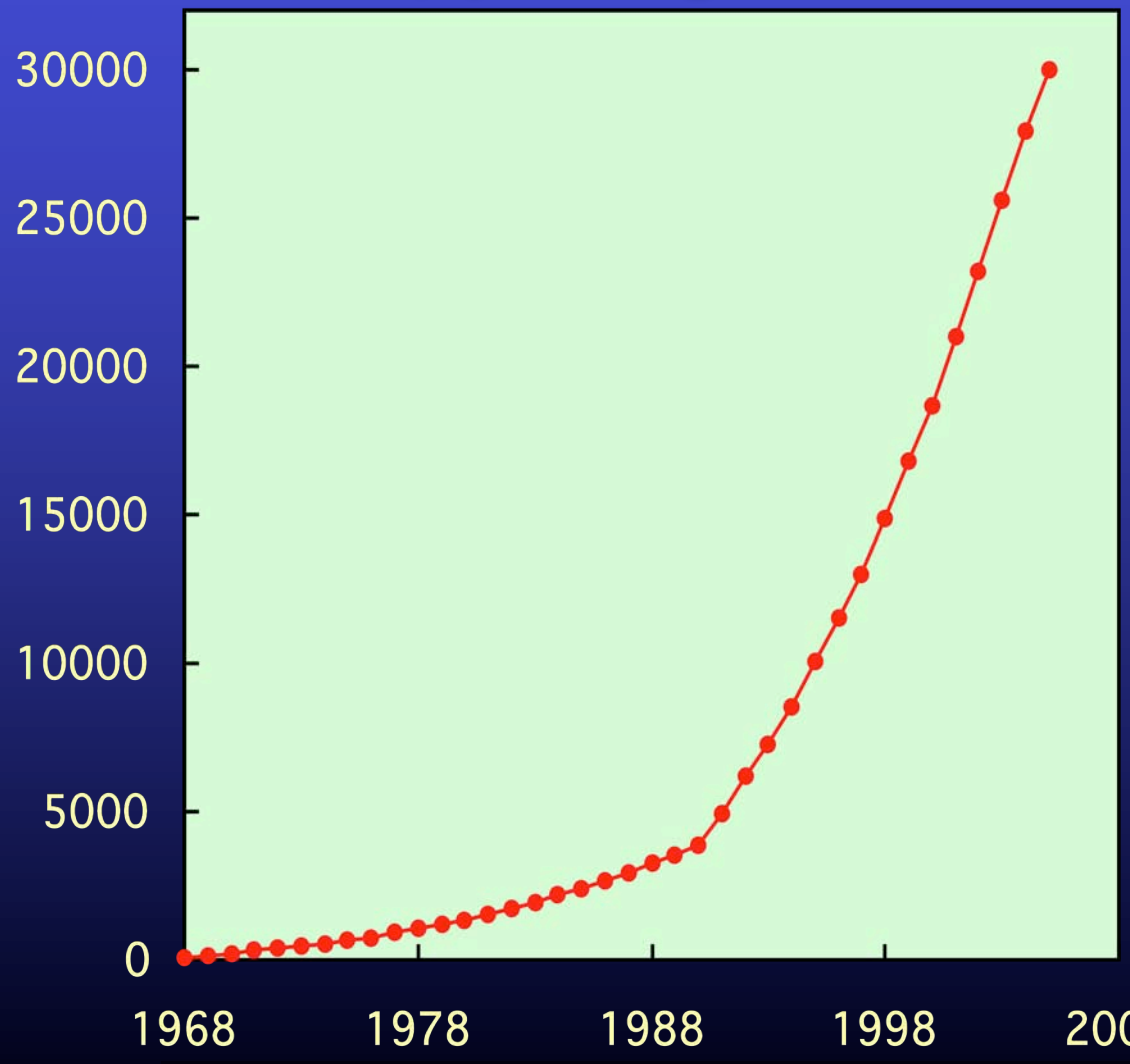
7 August 1968, 10:40 a.m.: Ulrich Gehrardt's team performs on Tantalus the first experiment with a dedicated synchrotron source:

ONLY 42 YEARS AGO!!!

Synchrotron Facilities in the World (2010):

69 in 25 Countries

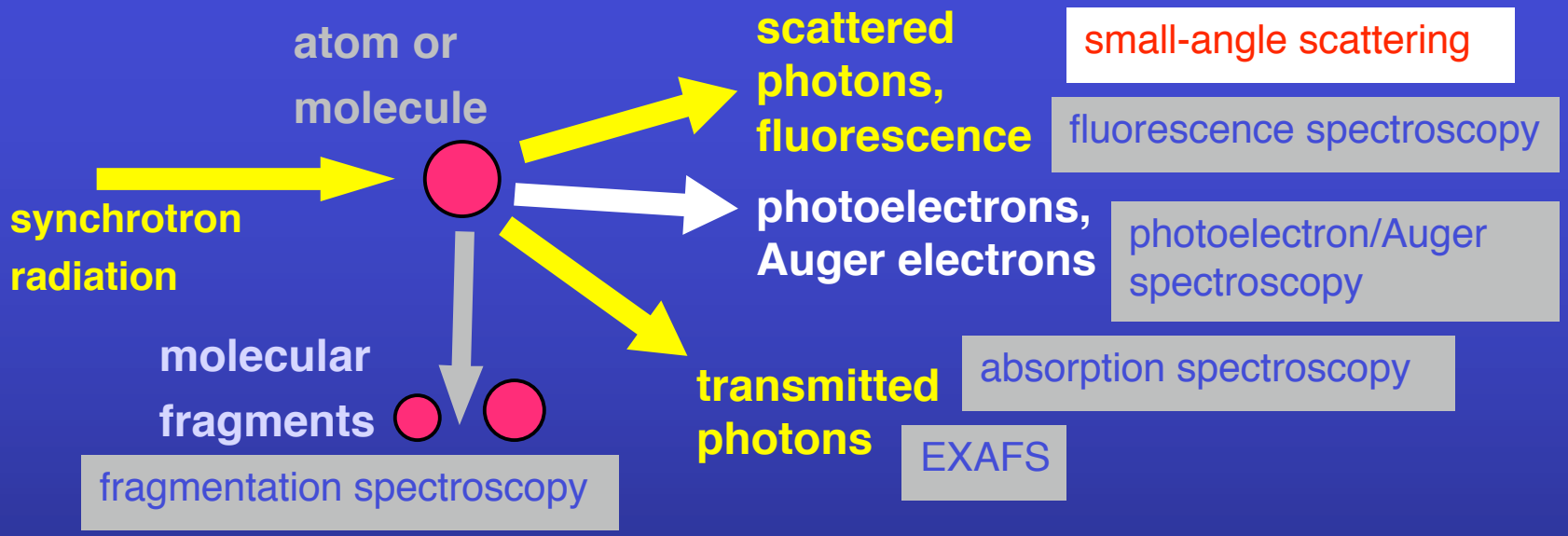
(operating or under construction)

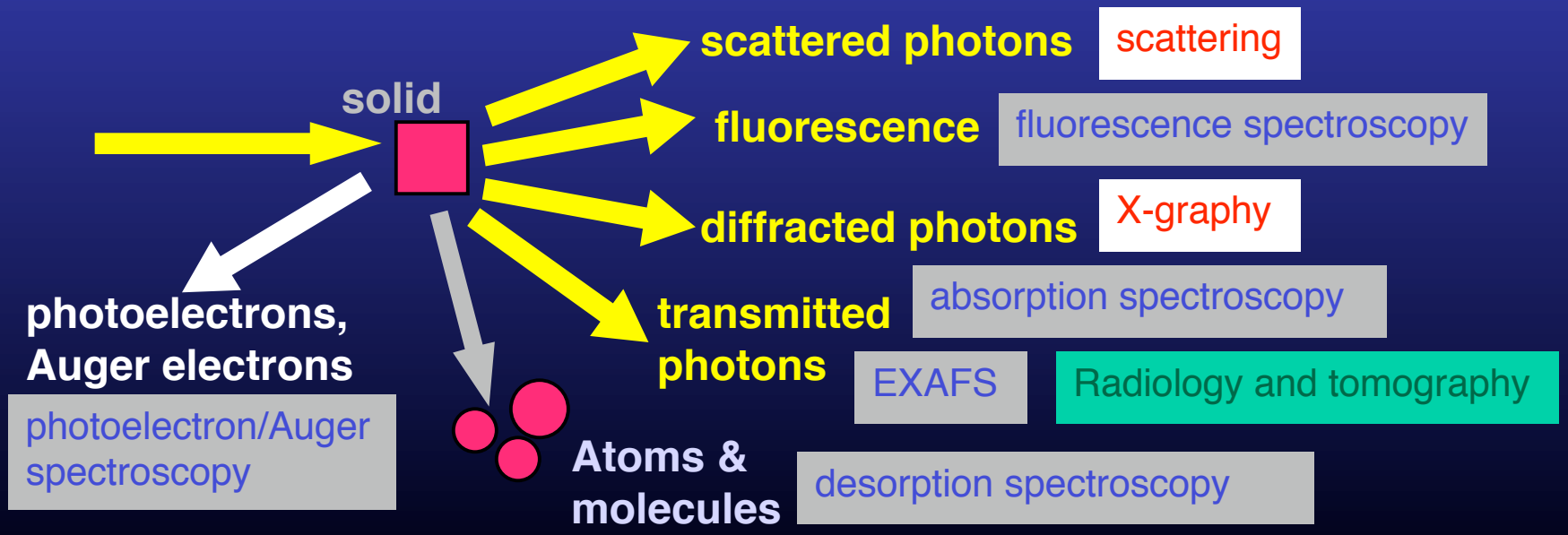


Historical Growth:

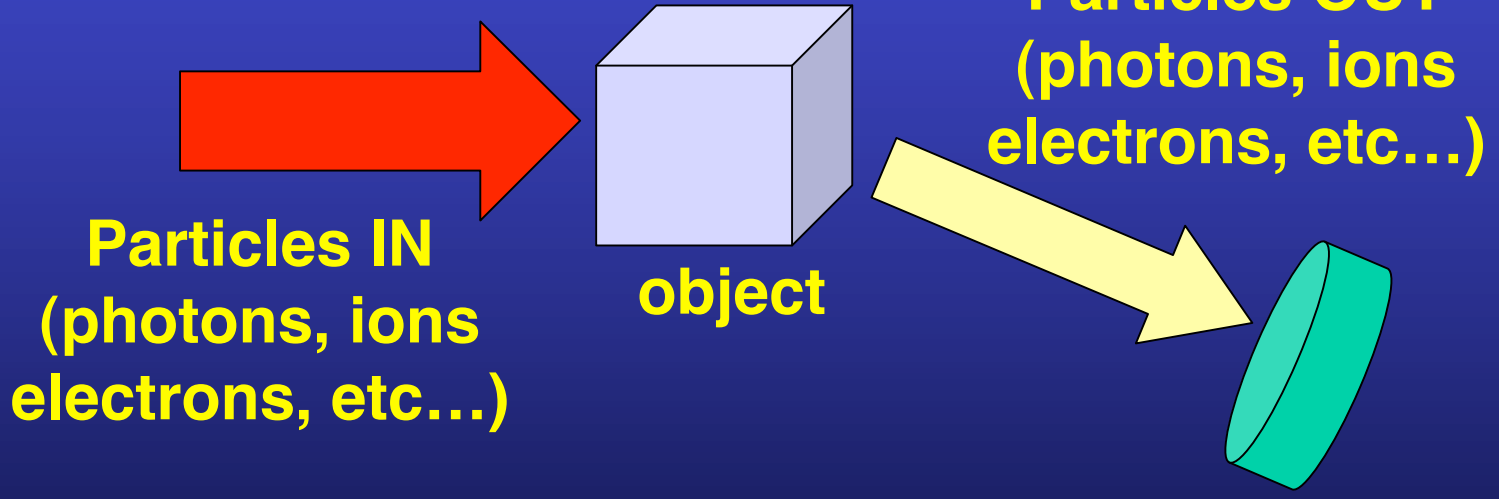
Worldwide ISI data 1968-2006, Keyword: "synchrotron"

Three classes of experiments:



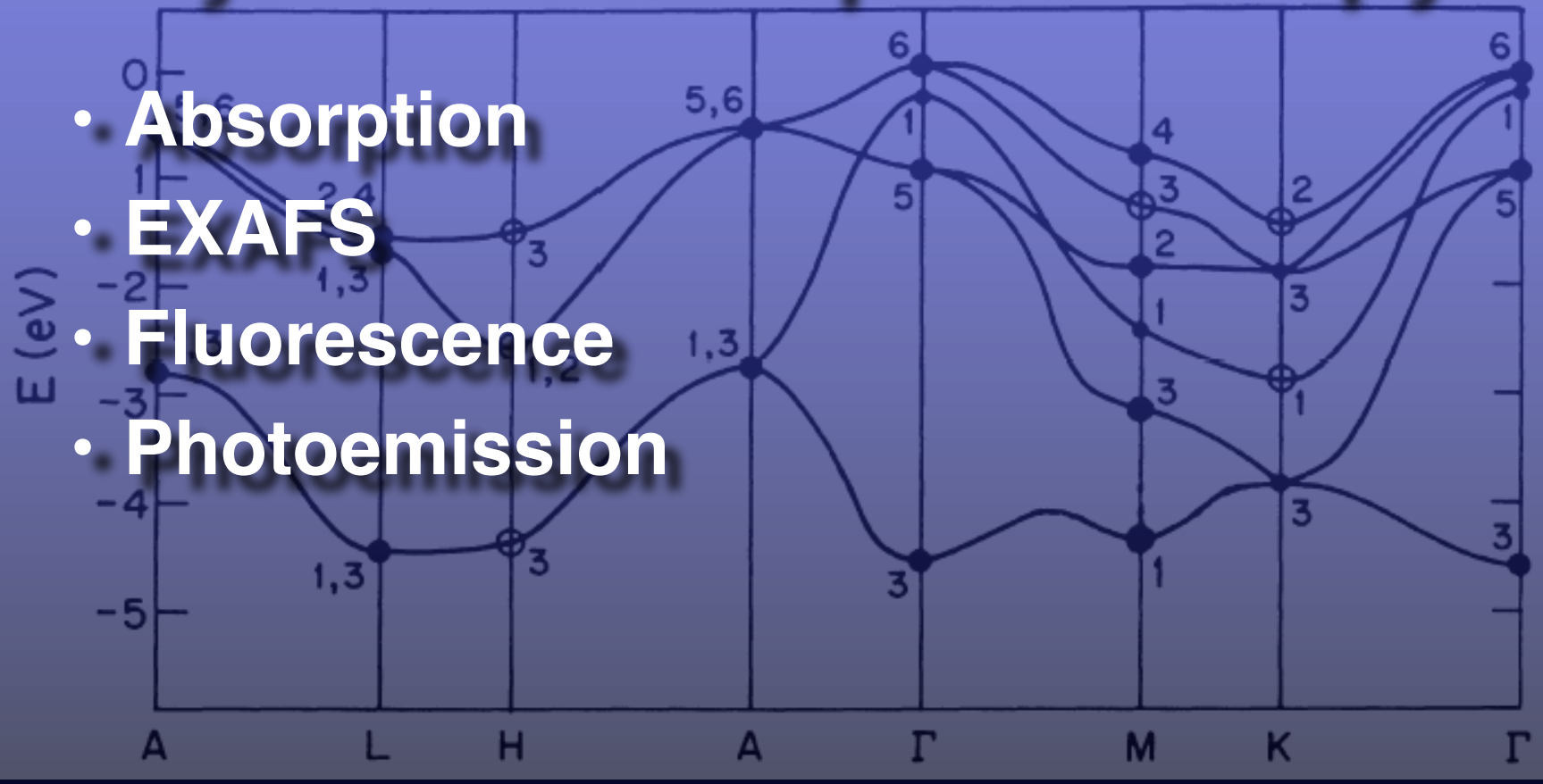


“Spectroscopy”

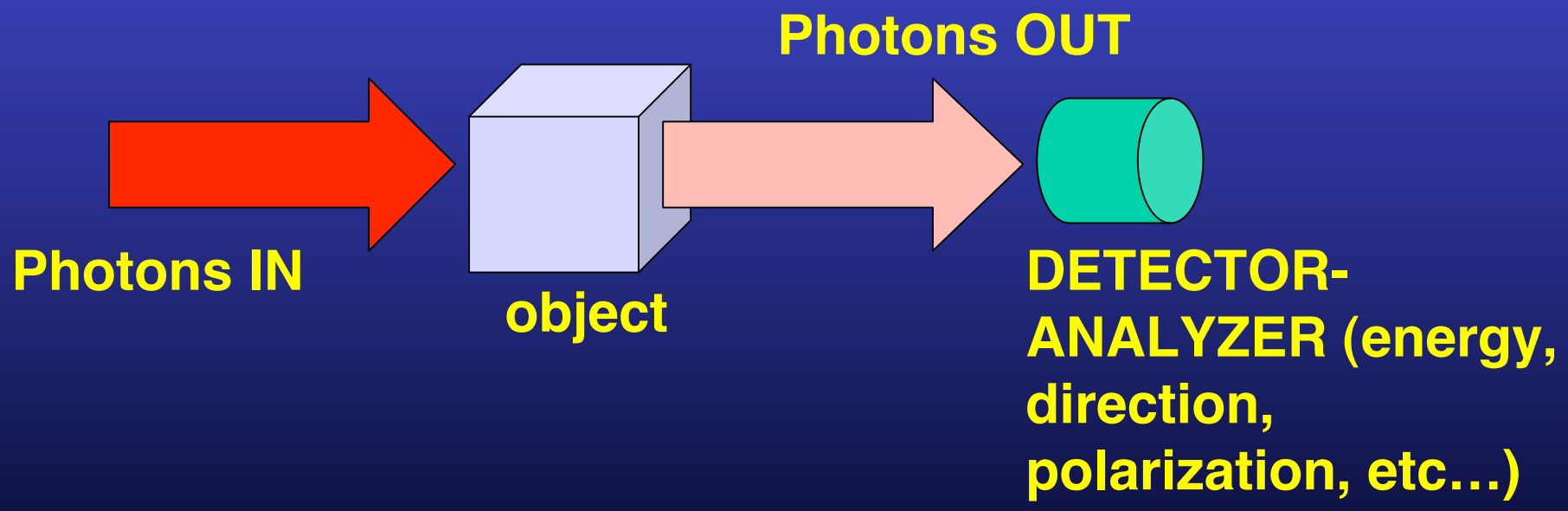


Important examples of synchrotron spectroscopy:

- Absorption
- EXAFS
- Fluorescence
- Photoemission



Absorption Spectroscopy and EXAFS:

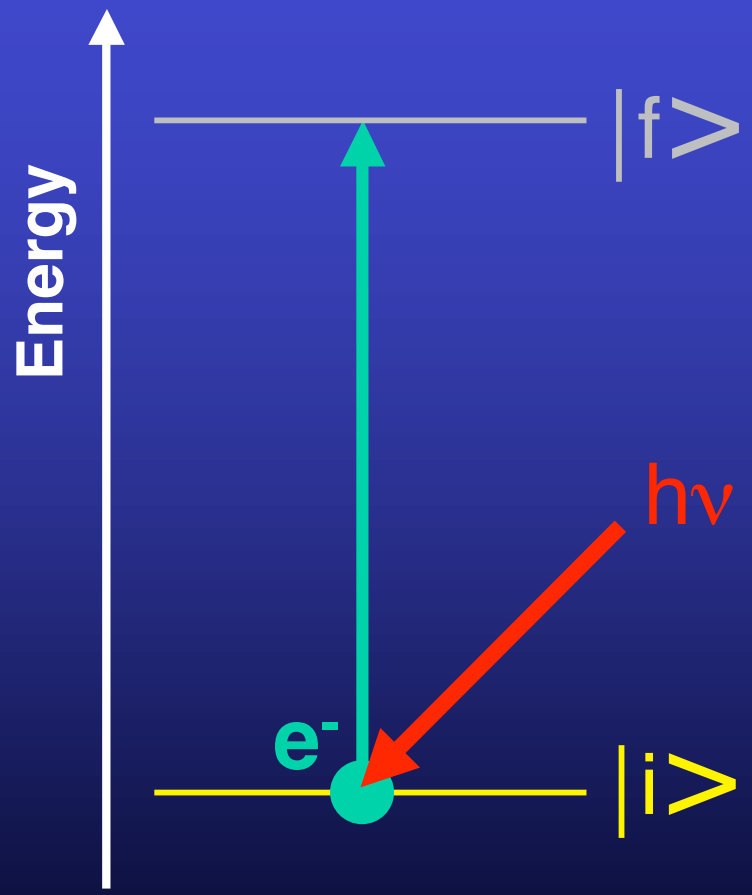


Theoretical background:

Transition Probability:

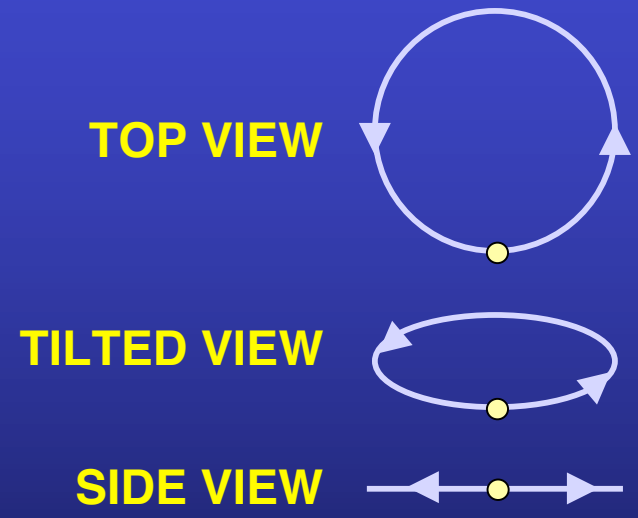
$$\propto |\langle i|\mathbf{A}\cdot\mathbf{p}|f\rangle|^2 \delta(E_f - E_i - h\nu)$$

Selection rules
Polarization effects



Synchrotron light polarization:

Electron in a storage ring:



Polarization:

Linear in the plane of the ring, elliptical out of the plane

Special (elliptical) wigglers and undulators can provide elliptically polarized light with high intensity

A formal approach:

- Wave in vacuum: $\sin(kx - \omega t) \rightarrow \exp[i(kx - \omega t)$
- Wave in a material:

  $k \rightarrow nk$

  $n = n_R + i n_I$

  $\exp[i(kx - \omega t)] \rightarrow \exp(-n_I kx) \exp[i(n_R kx - \omega t)]$
- $\exp(-n_I kx)$ → attenuation factor → absorption:
- Intensity: I ∝ wave square = $A^2 \exp(-2n_I kx)$
- Absorption coefficient α: by definition, $I(x) = I_o \exp(-\alpha x)$ -- hence, $\alpha = 2n_I k$
- $n_R k$ → phase shifts (refraction, diffraction, interference)

Kramers-Kroenig (KK) relations:

- They link the real and imaginary parts of n:

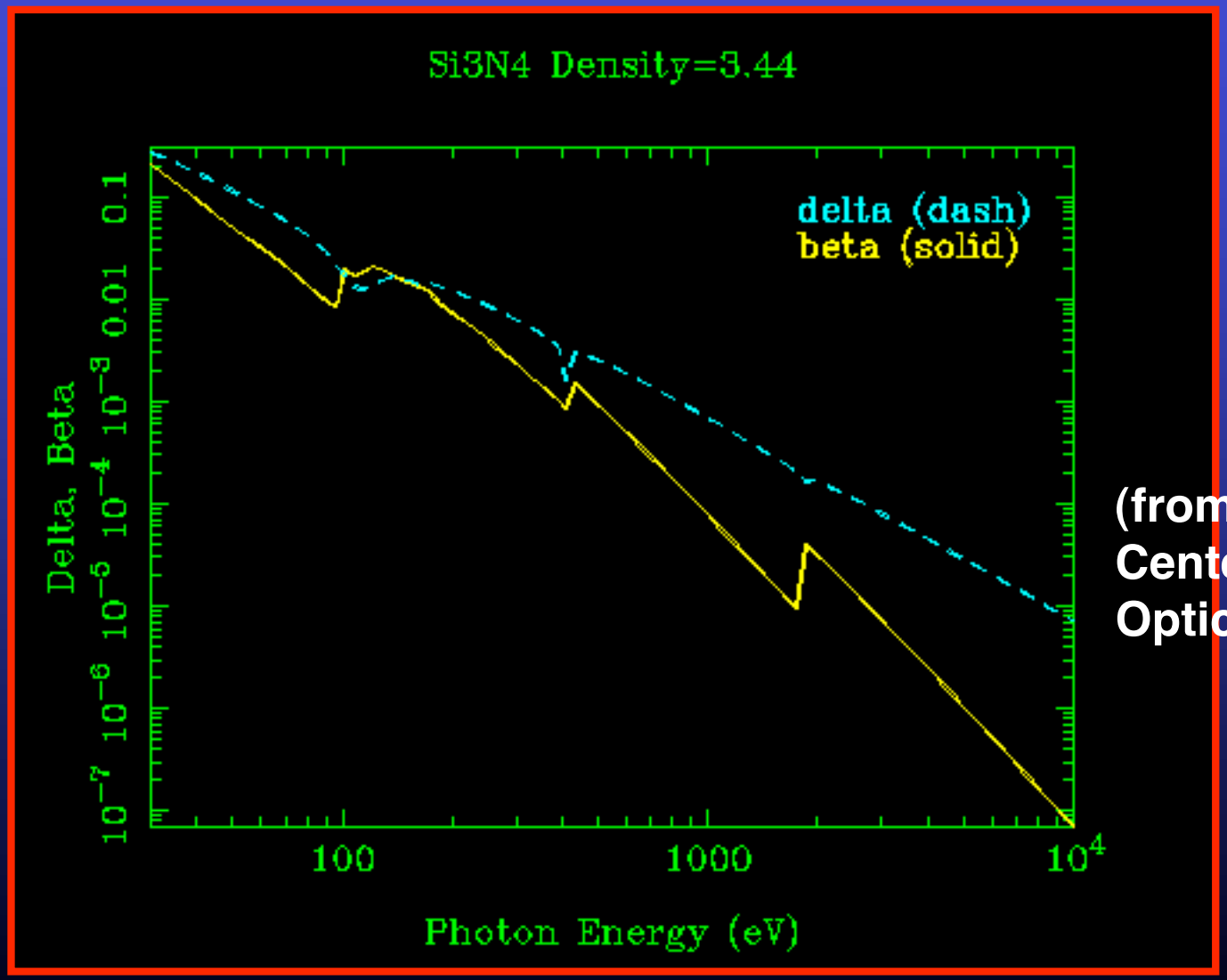
$$n_R - 1 = (2/\pi)\int d\omega' \, n_I(\omega')[\omega'/(\omega^2 - \omega'^2)]$$
$$n_I = (2/\pi)\int d\omega' n_R(\omega')[\omega'/(\omega^2 - \omega'^2)]$$

  with integrals performed over the entire ω spectral range from 0 to ∞

- From measurements of the real part from 0 to ∞, we can also derive the imaginary part -- and voce-versa
- In practice, the measurements **cannot be from 0 to ∞** -- but must be performed over a spectral range **as wide as possible: synchrotrons are very helpful!**

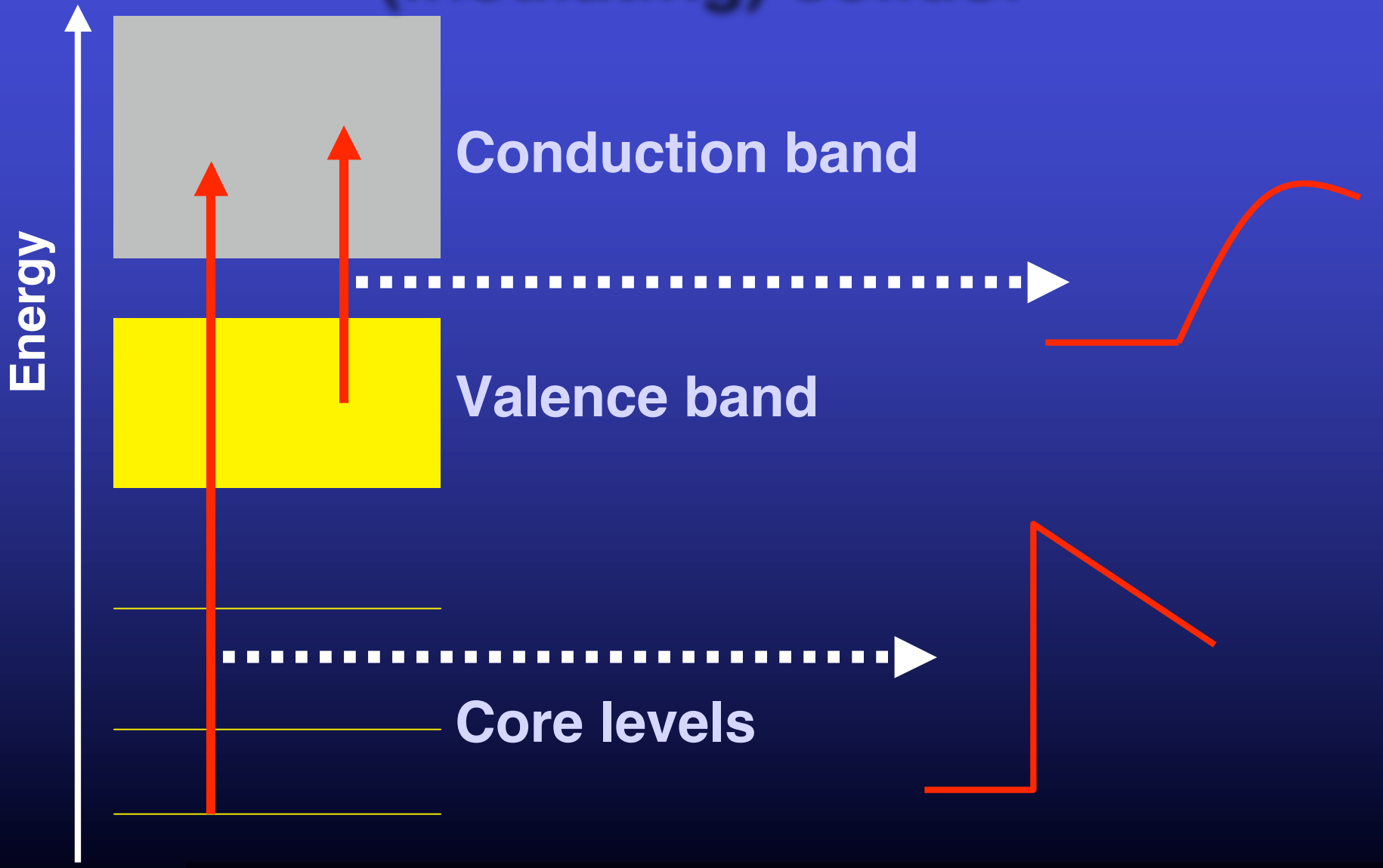
Kramers-Kroenig relations -- example:

Complex refractive index = n_R + in_I = 1-delta - i beta



(from the Berkeley Center for X-ray Optics databank)

Photon absorption in (insulating) solids:



A real example: transitions from core levels (I. Koprinarov and A. P. Hitchcock, tutorial)

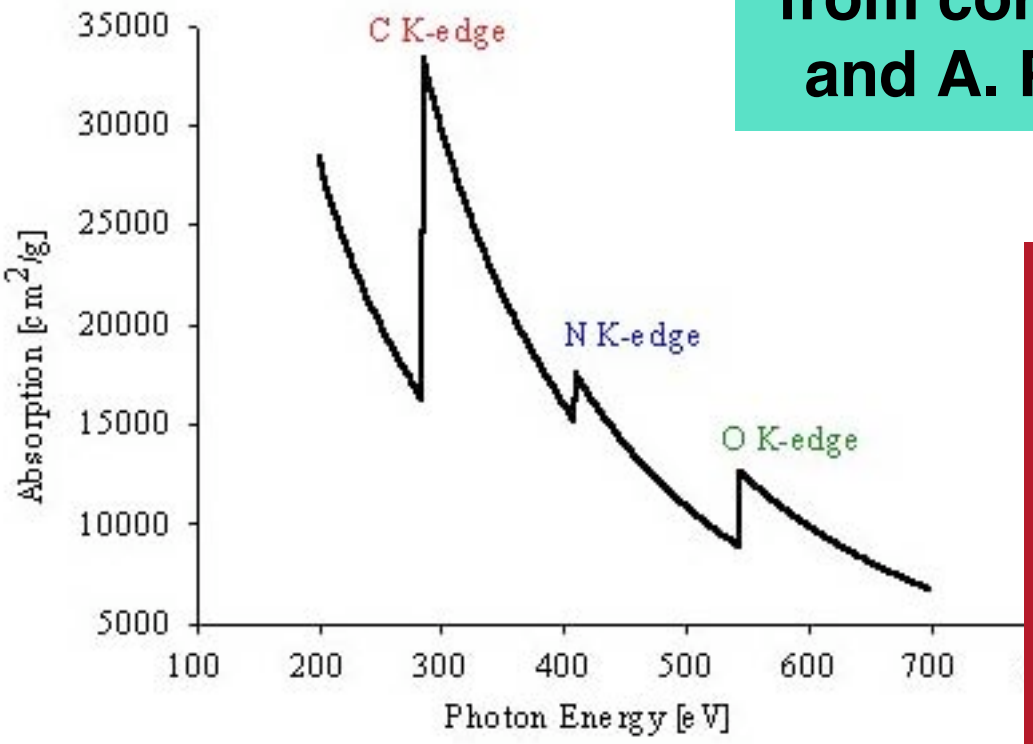
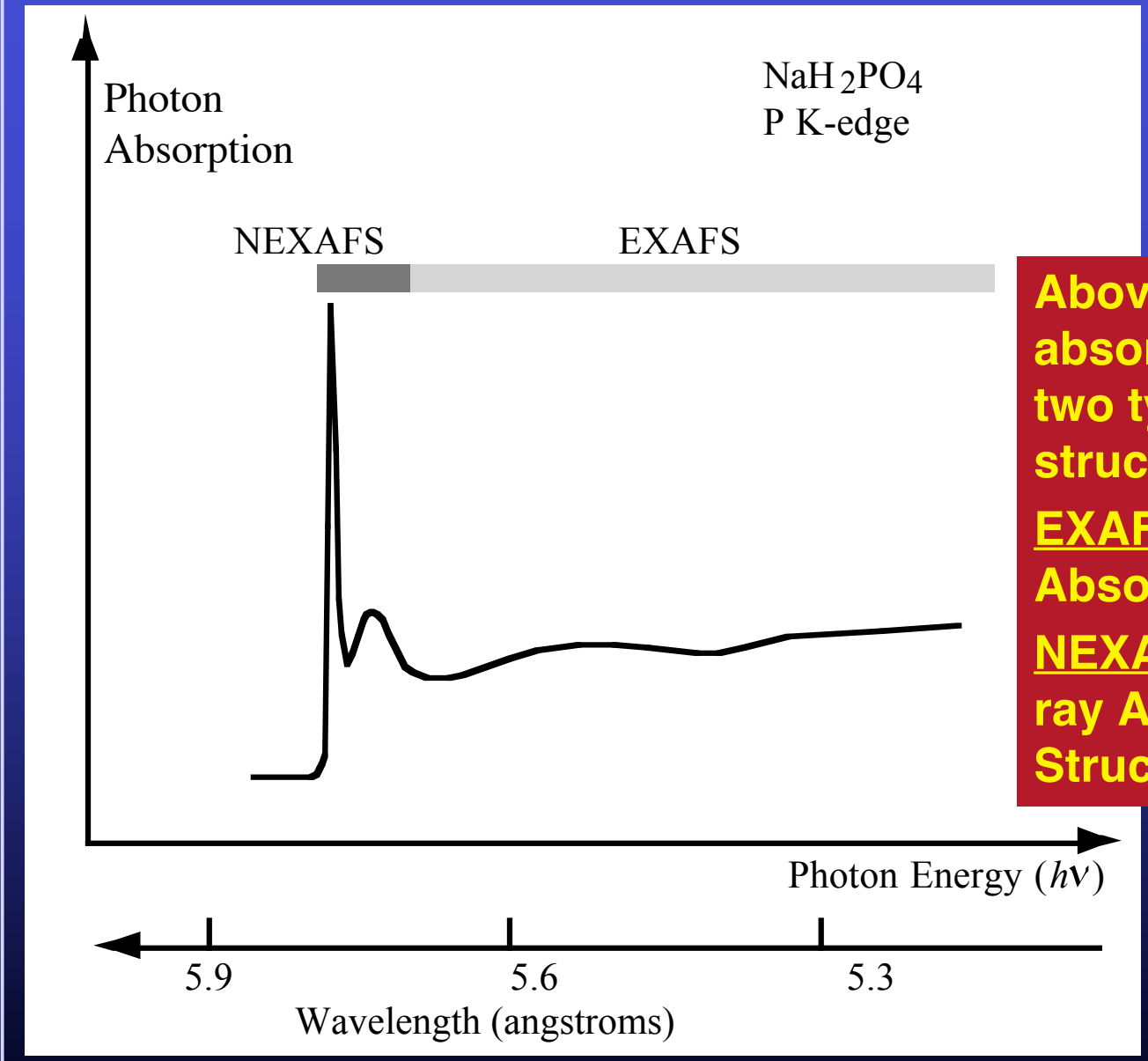


Fig. 1 X-ray absorption edges of carbon, nitrogen and oxygen of a composition typical of a polymer. A rapid increase of the absorption occurs at the threshold of energy required to excite electrons from these inner shells.

Information in general:

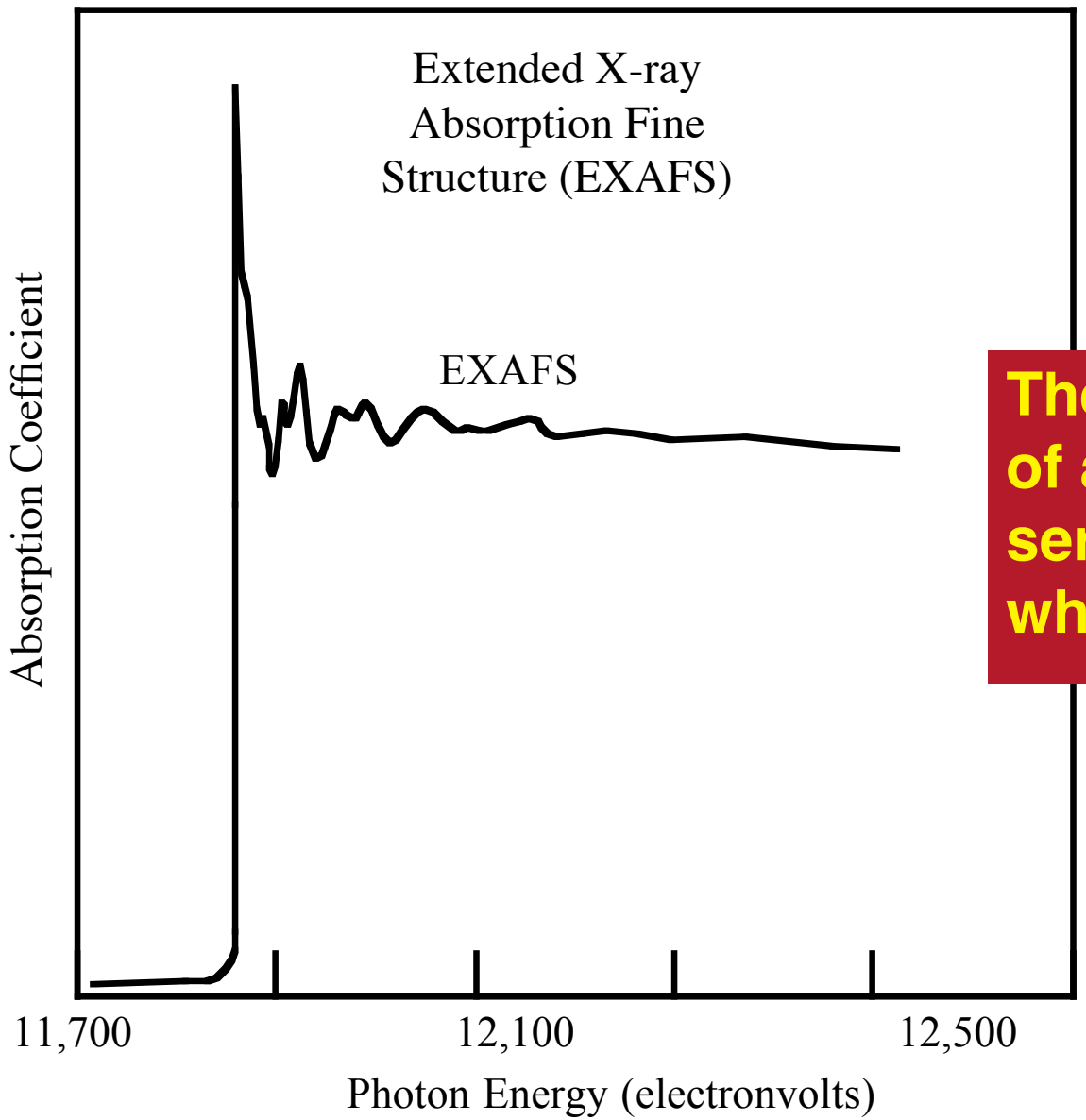
- **From core-level transitions: core-level binding energies (influenced by the charge distribution of the chemical bonds)**
- **From band-band transitions: band structure (determined by the bond formation)**



Above each core-level absorption threshold -- two types of fine structure:

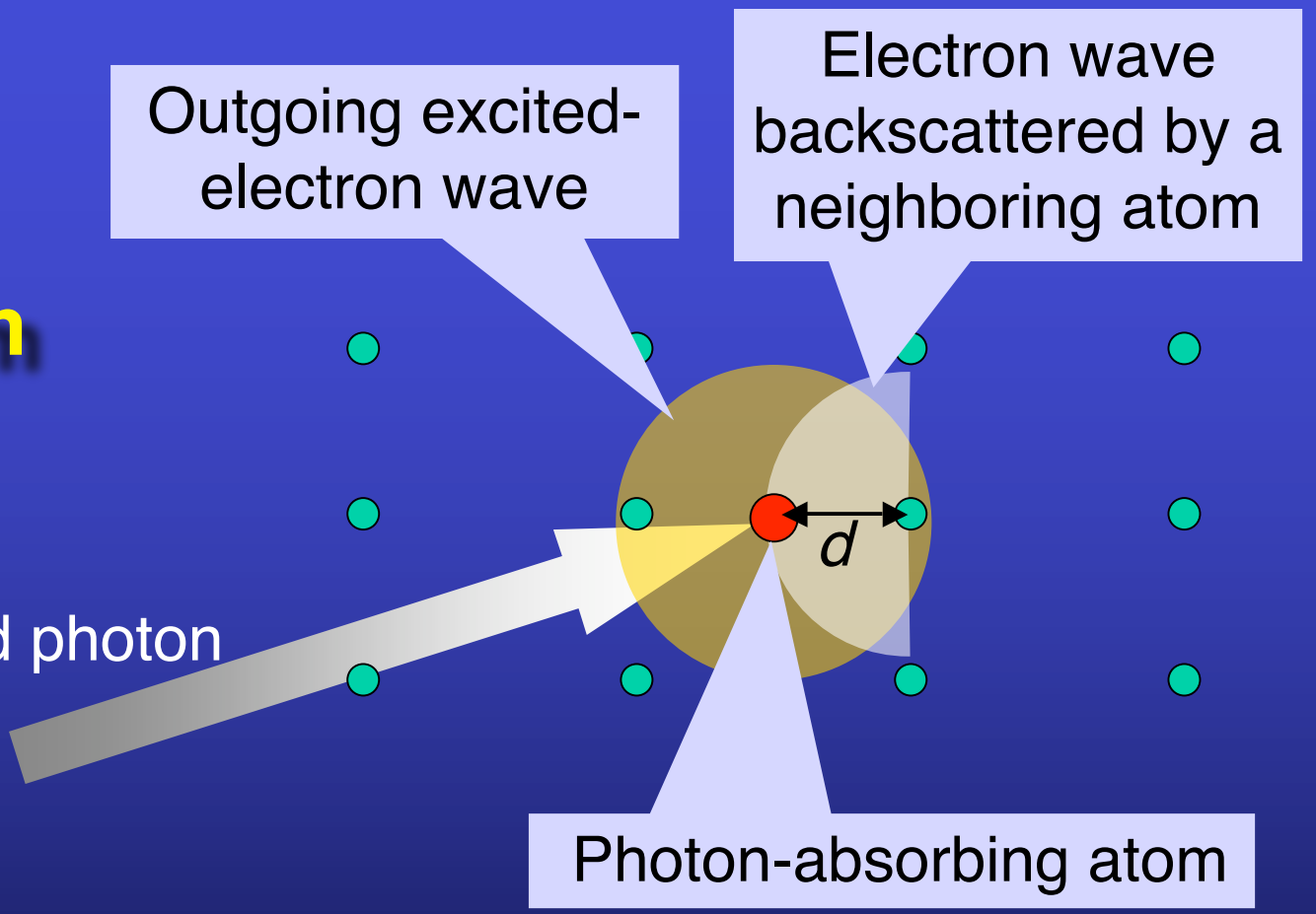
EXAFS = Extended X-ray Absorption Fine Structure

NEXAFS = Near-edge X-ray Absorption Fine Structure



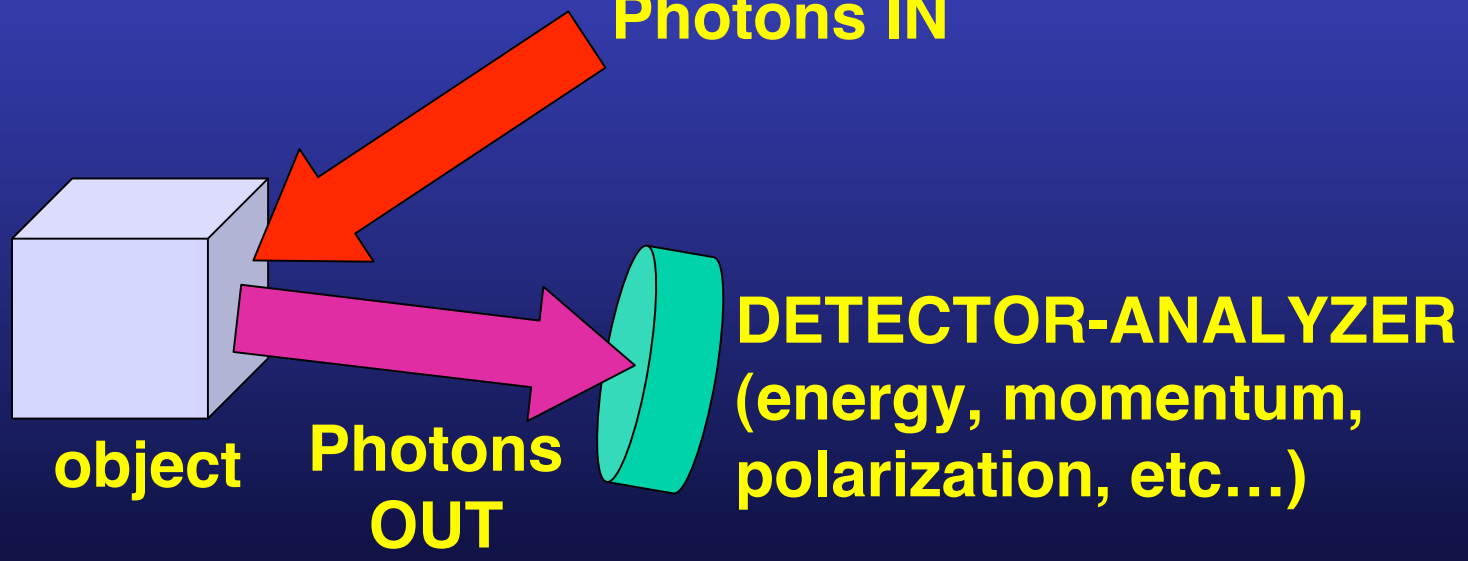
The EXAFS consists of a characteristic series of oscillations: what is their origin?

EXAFS Mechanism in a solid:

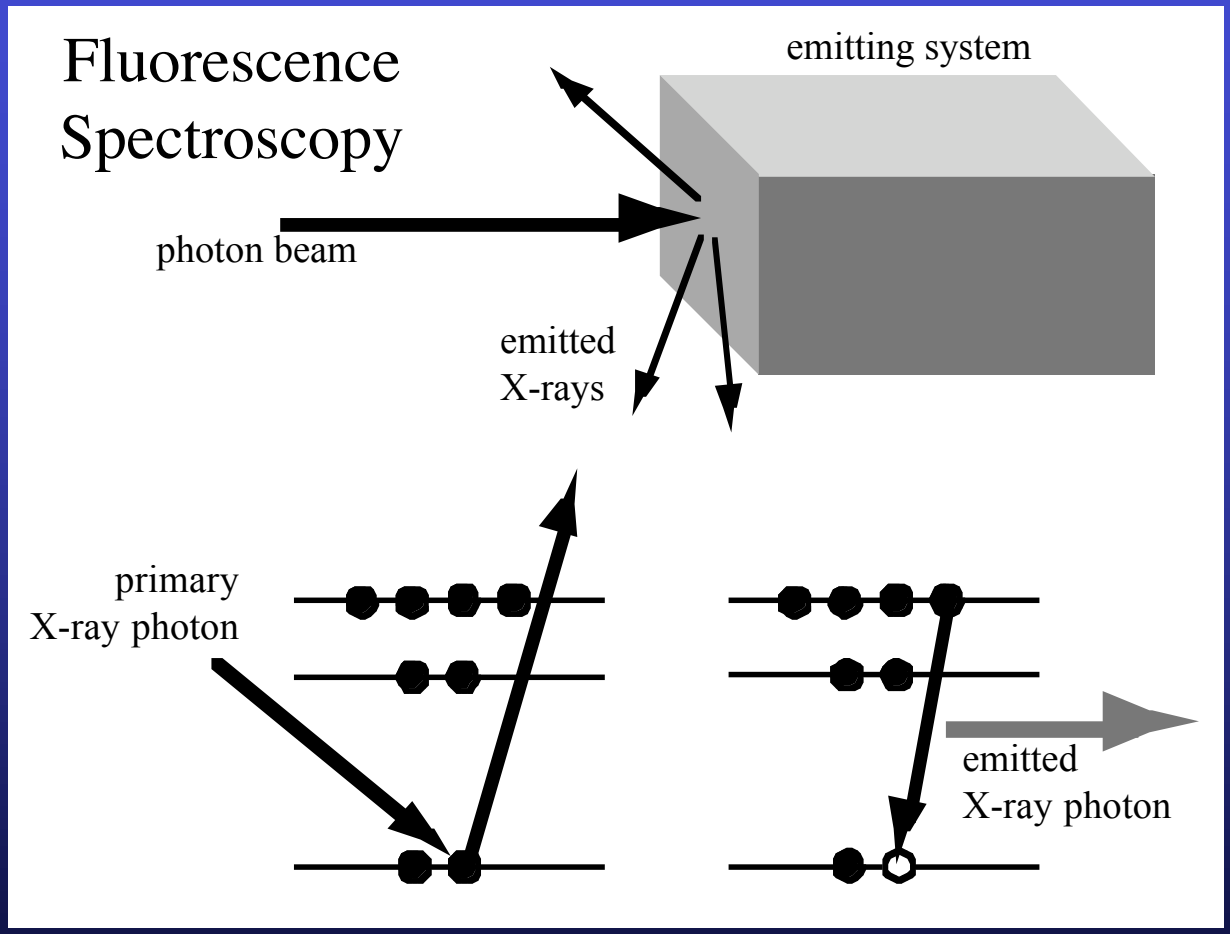


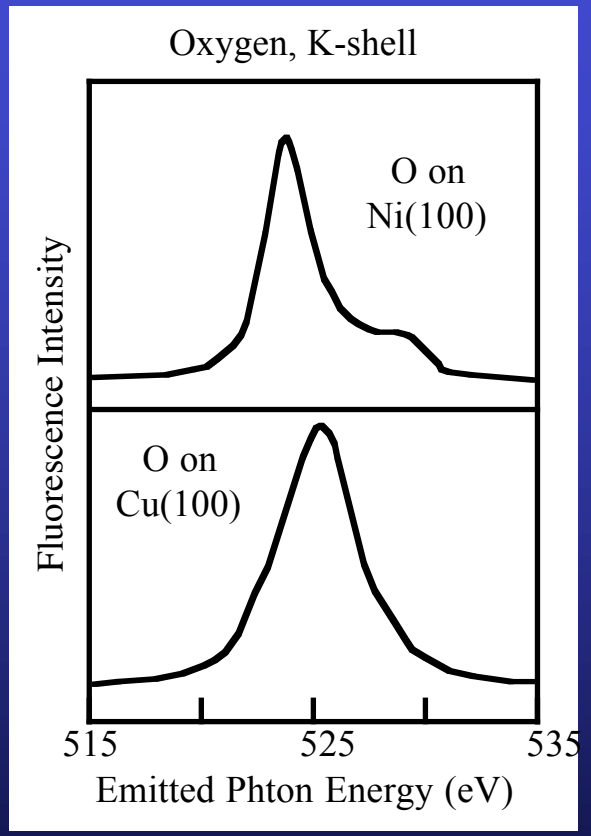
For a given distance *d*, the combination of the outgoing and backscattered electron waves at the photon-absorbing atom goes from constructive to destructive etc. as the photon energy increases, changing the electron energy and wavelength: hence the EXAFS oscillations. From these, one can derive local chemical bond lengths *d* around specific atomic species.

“Fluorescence Spectroscopy”



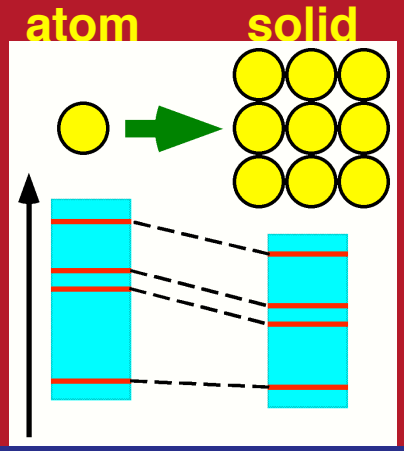
Fluorescence Spectroscopy



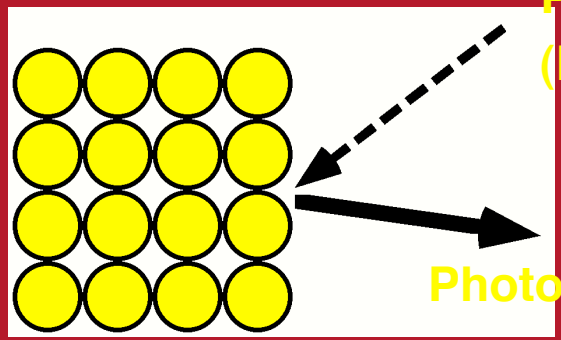


Photoelectron spectroscopy: basic ideas

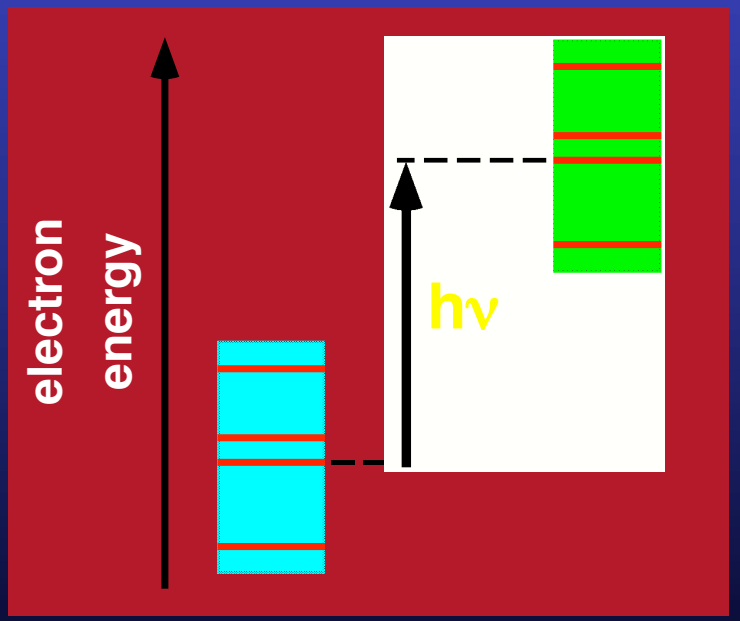
Formation of chemical bonds:



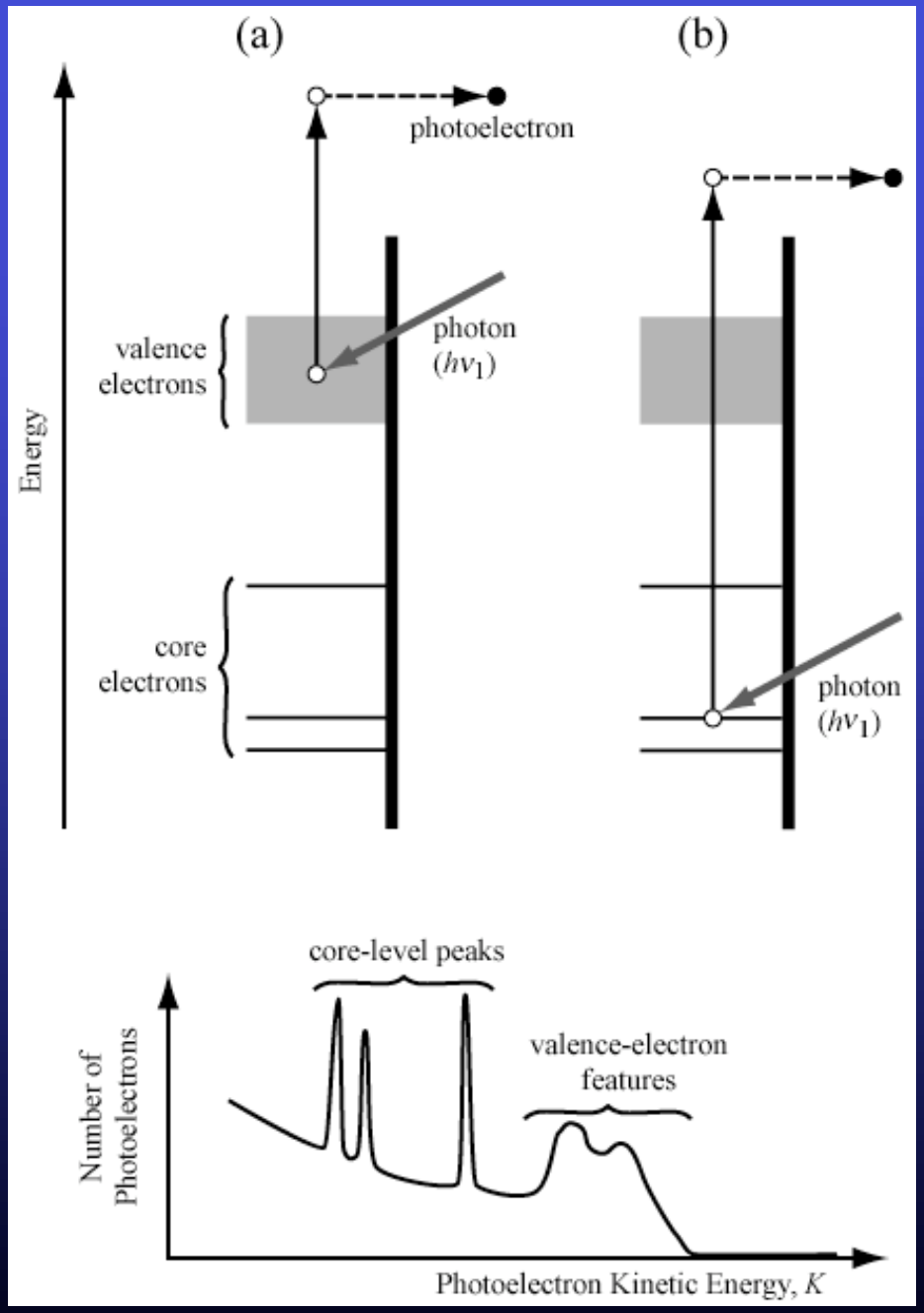
Photoelectric effect:

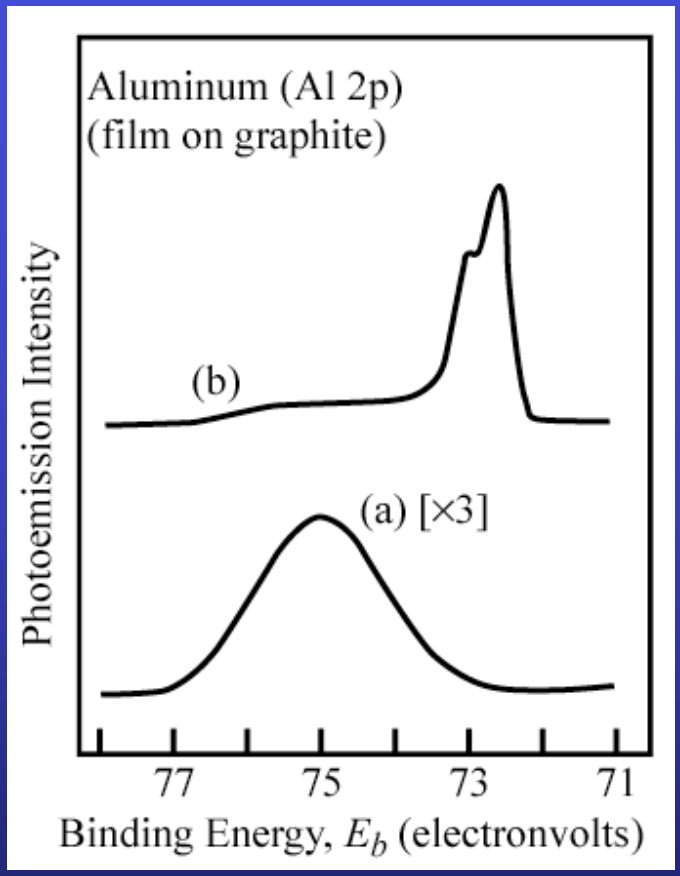


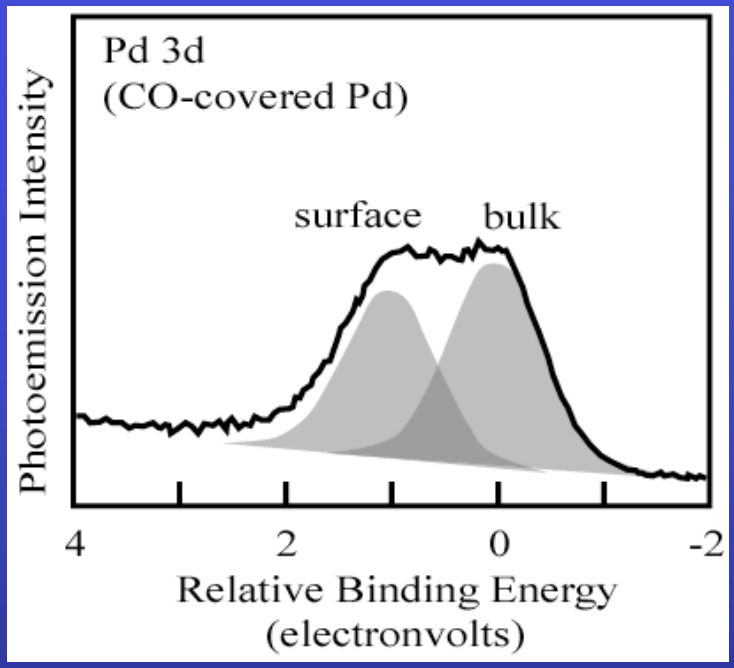
The photon absorption increases the electron energy by hν before ejection of the electron from the solid

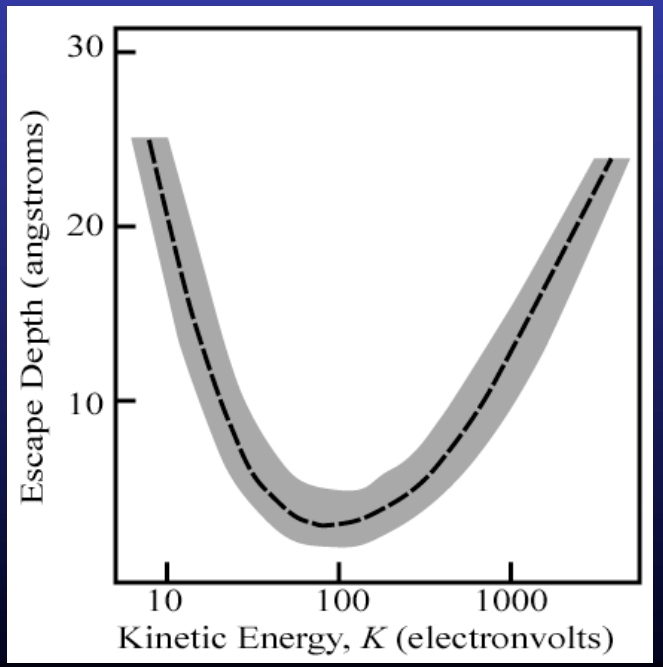


Photoemission in solids: valence electrons and core electrons





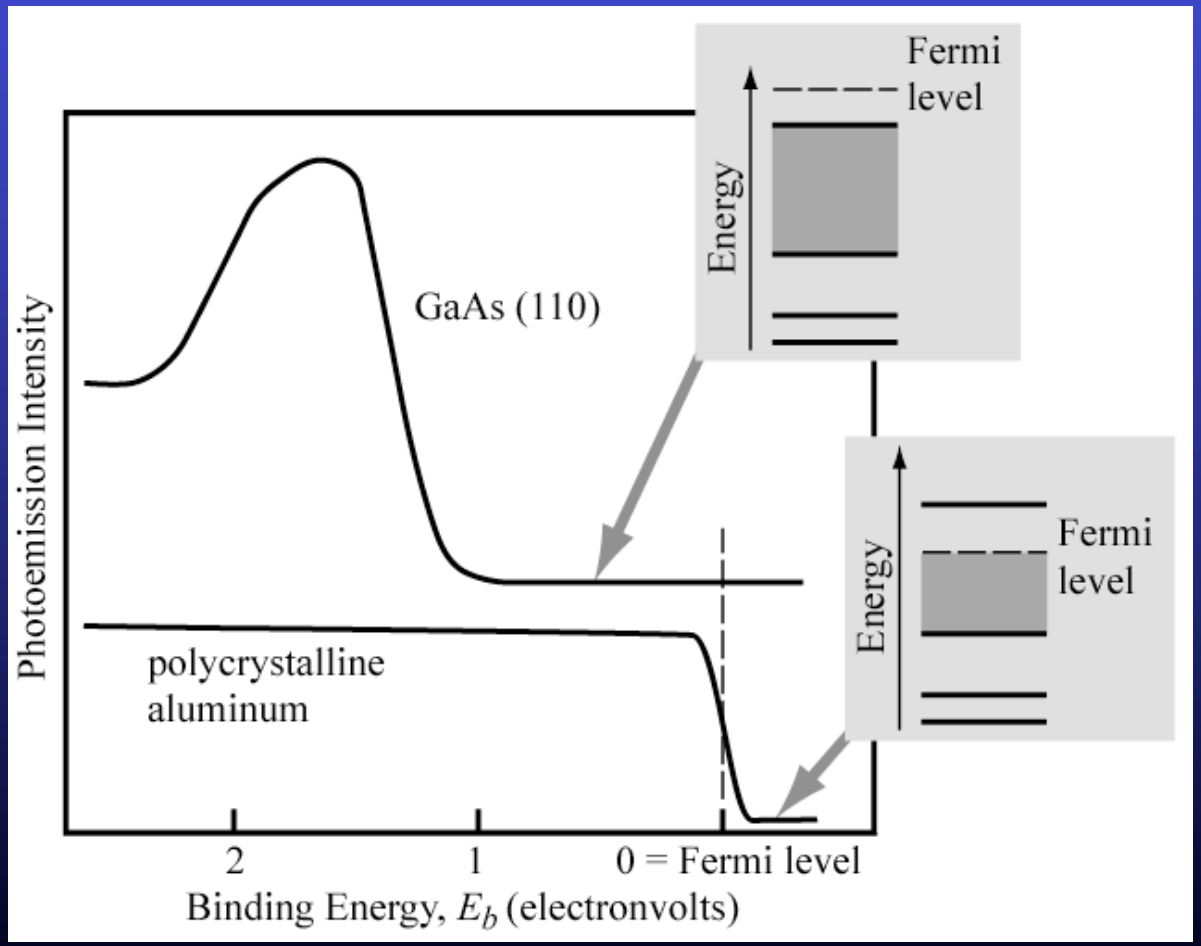


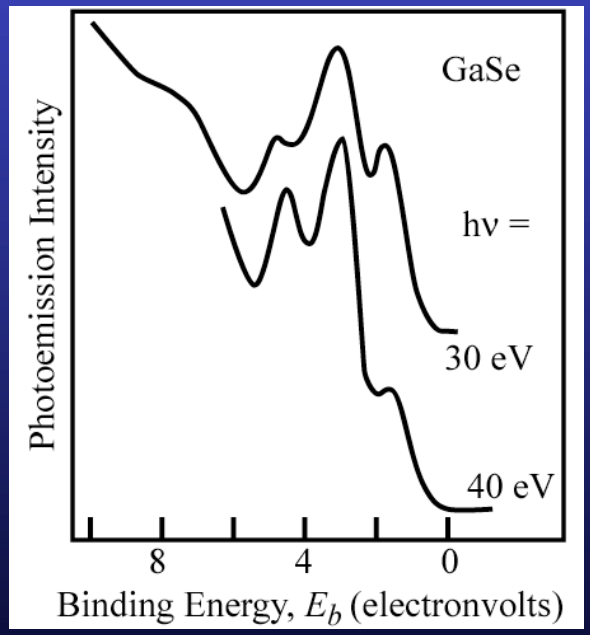


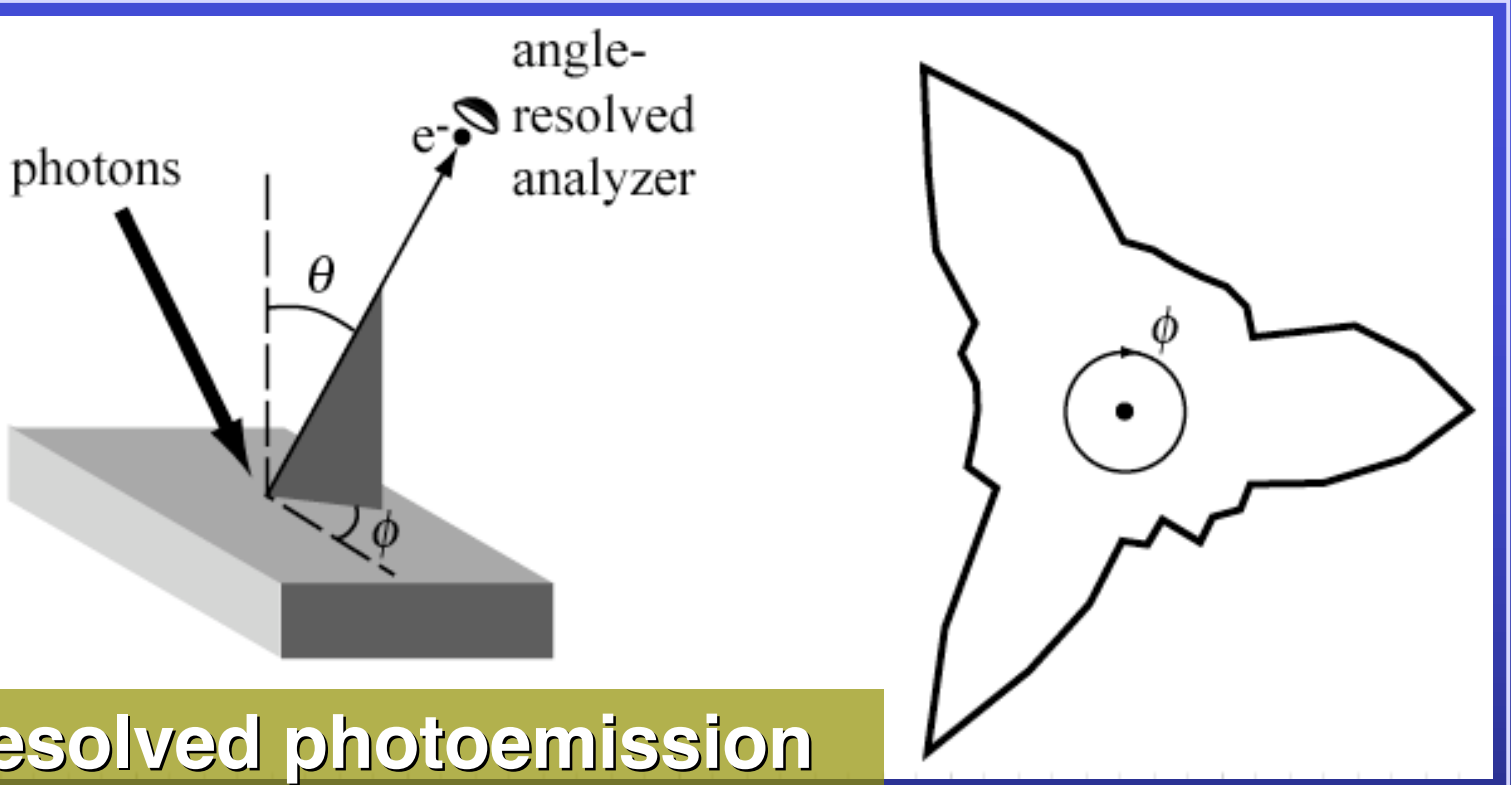
Core-level peaks: they depend on the chemical environment …

… and can be different for the surface and for the bulk

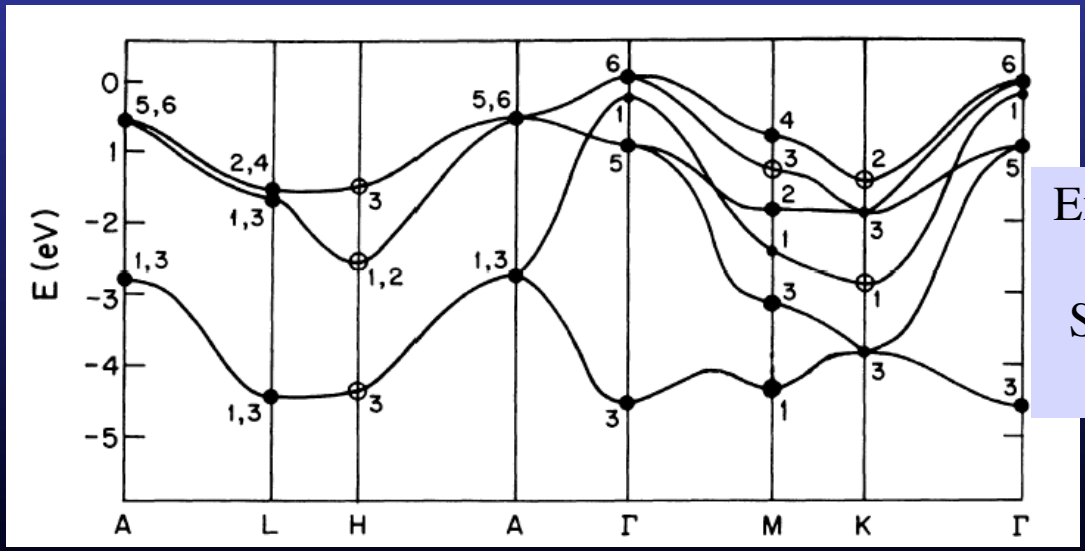
Valence-electron ultraviolet photoemission: semiconductors vs simple metals





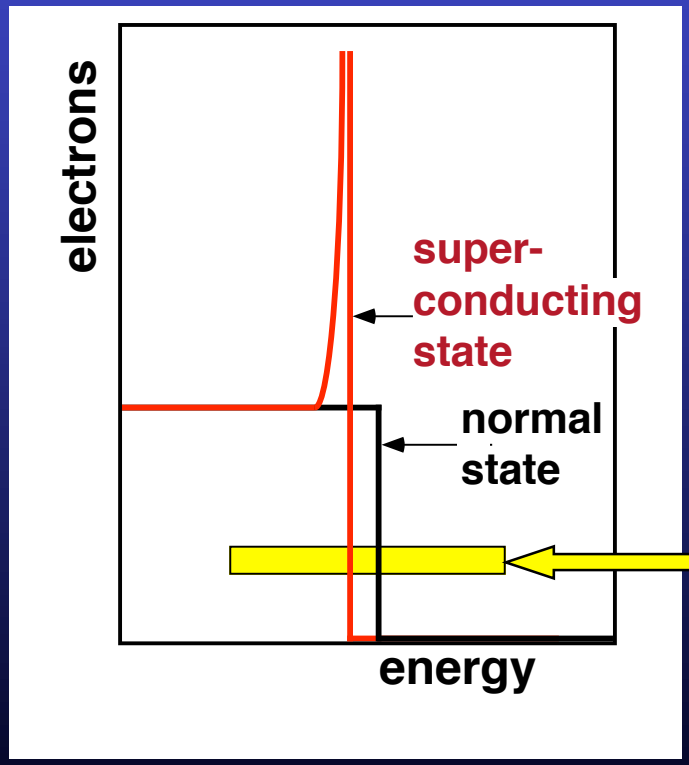


Angle-resolved photoemission

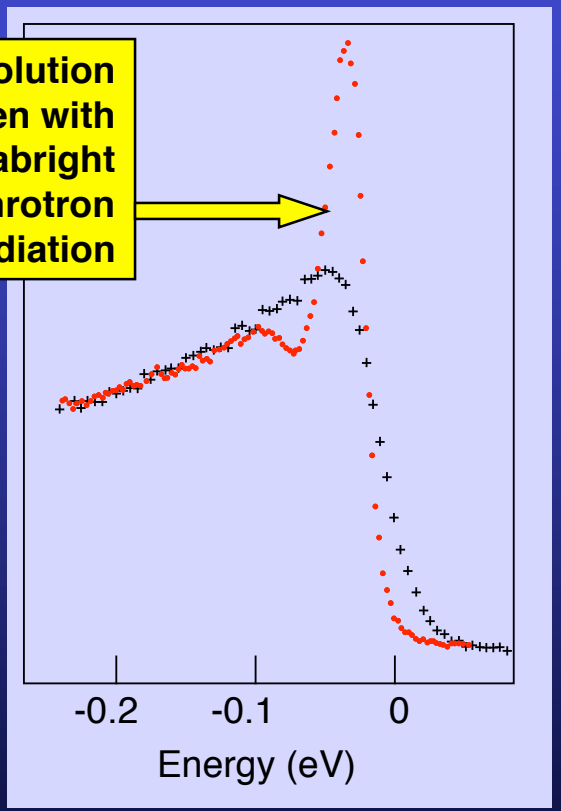


Experimental Band Structure of CdS

Photoelectron spectroscopy of high-temperature superconductors:

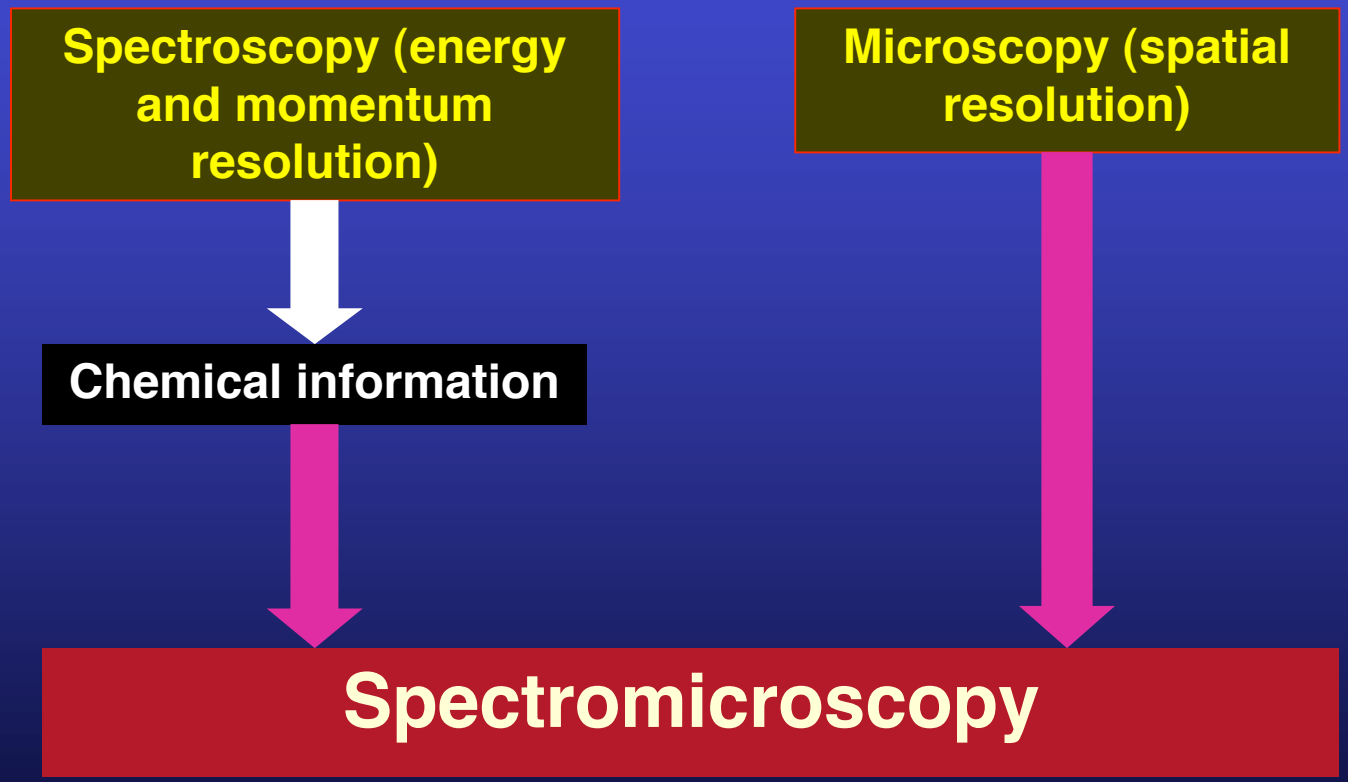


High-resolution spectra taken with ultrabright synchrotron radiation



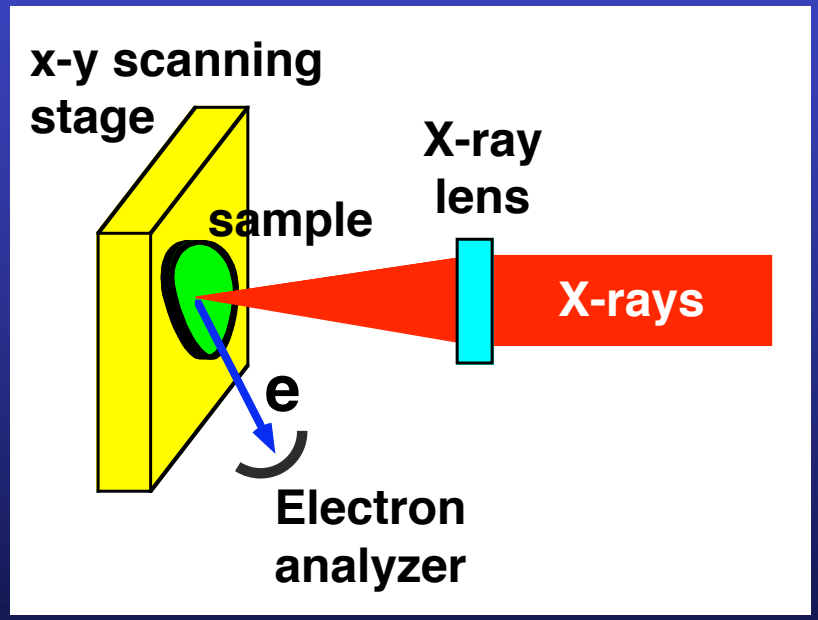
The limited energy resolution of conventional photoemission makes it impossible to observe the phenomenon

From spectroscopy to spectromicroscopy:

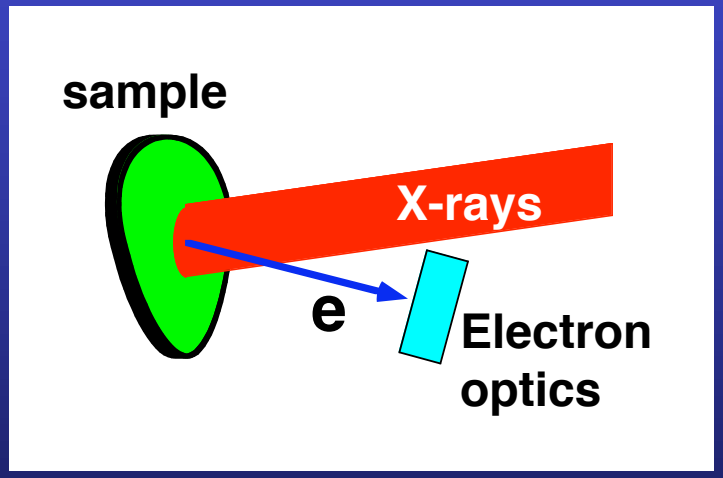


The two modes of photoemission spectromicroscopy:

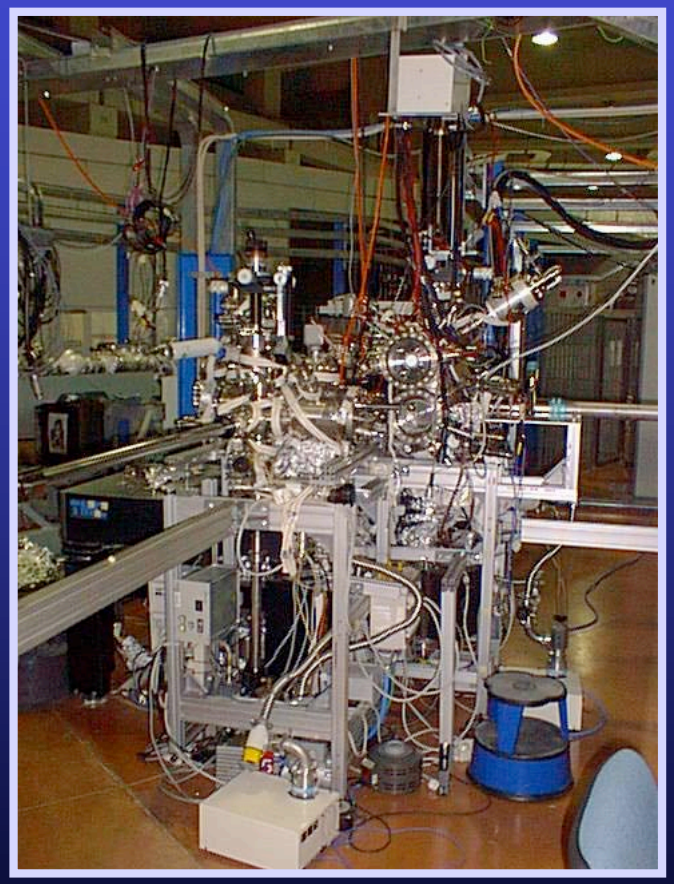
SCANNING

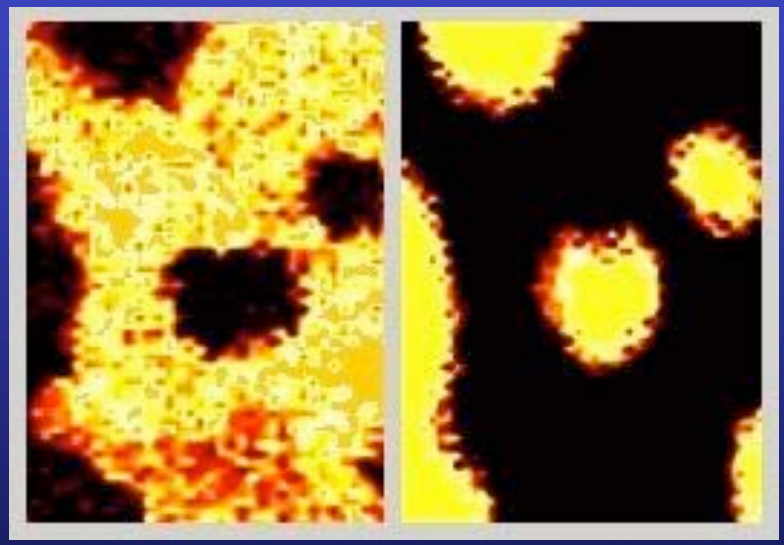


ELECTRON IMAGING



The ESCA Microscopy Beamline at ELETTRA, Trieste

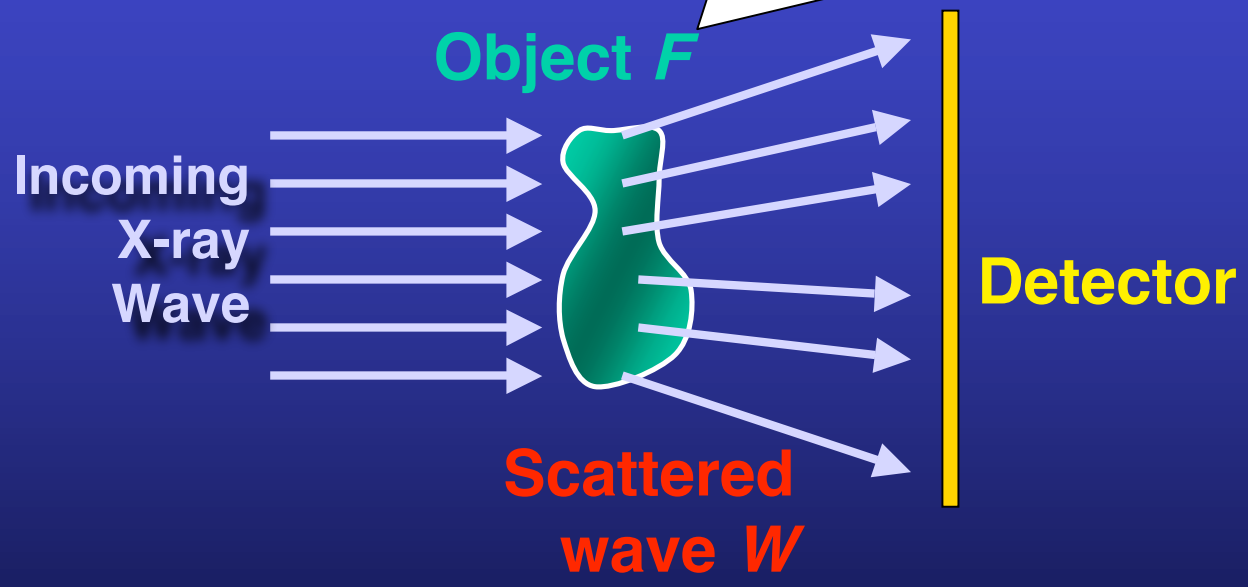




Synchrotron scattering and diffraction: exploring the microstructure of objects

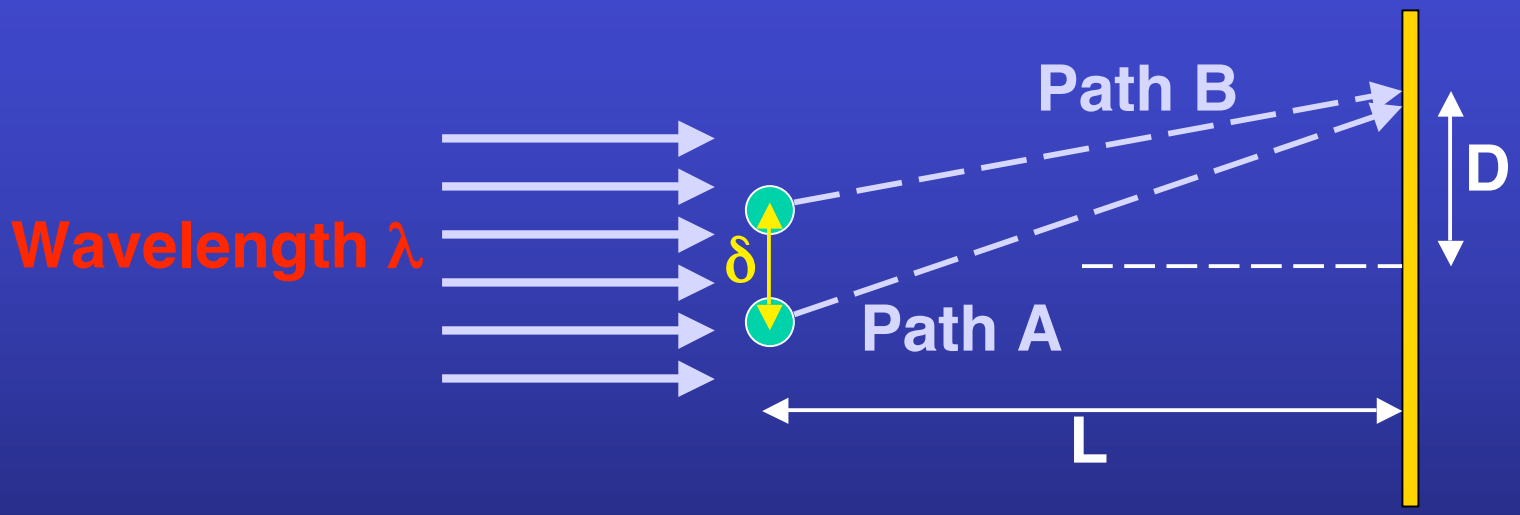
X-rays are scattered by electrons: the “object function” *F* is the space distribution of the electronic charge

Conceptual foundation: the Fourier transforms



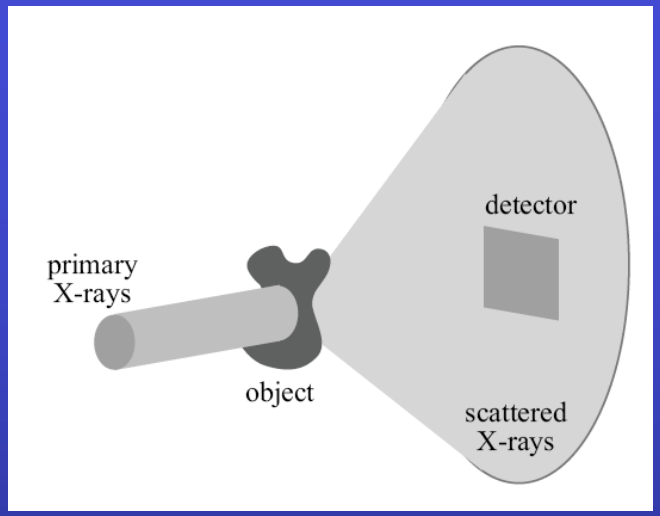
- **Fourier transform of *F* → *W***
- **(inverse) Fourier transform of *W* → *F***

Why x-rays? Look at the diffraction by a 2-atom molecule

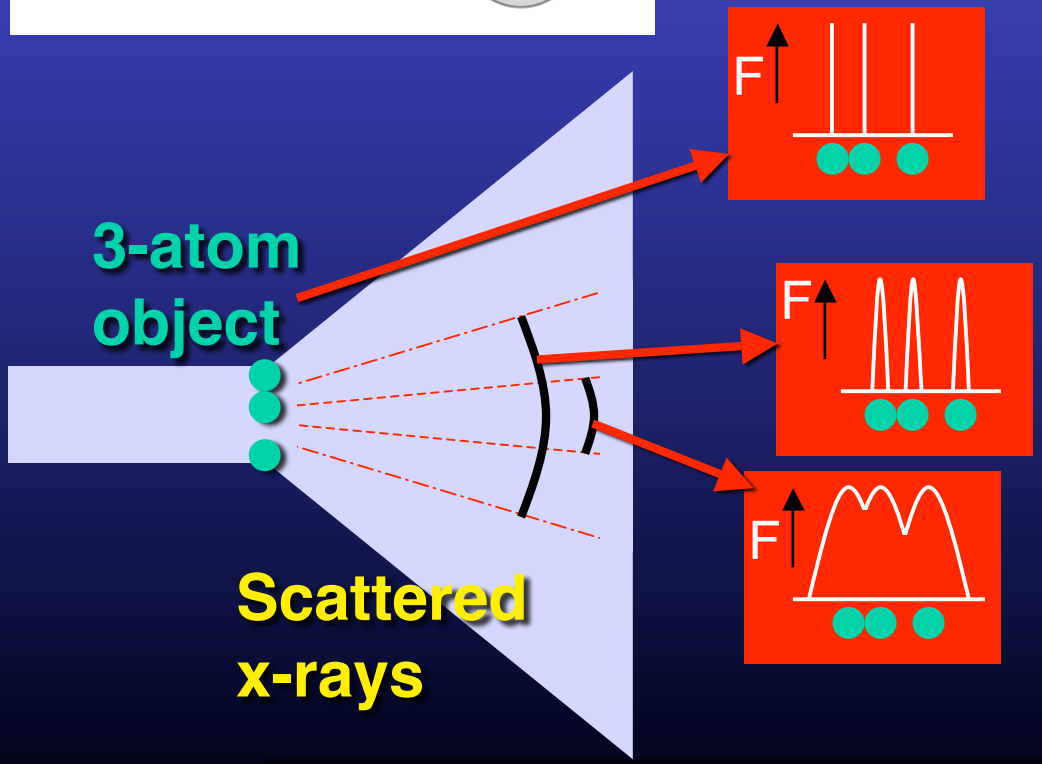


- A-B path difference ≈ (D/L)δ
- First diffracted intensity maximum: (D/L)δ = λ
- Thus, D = (λ/δ)L: if λ is not comparable to δ, L becomes too big (unrealistically large detector)

Small scattering angles and large scattering angles



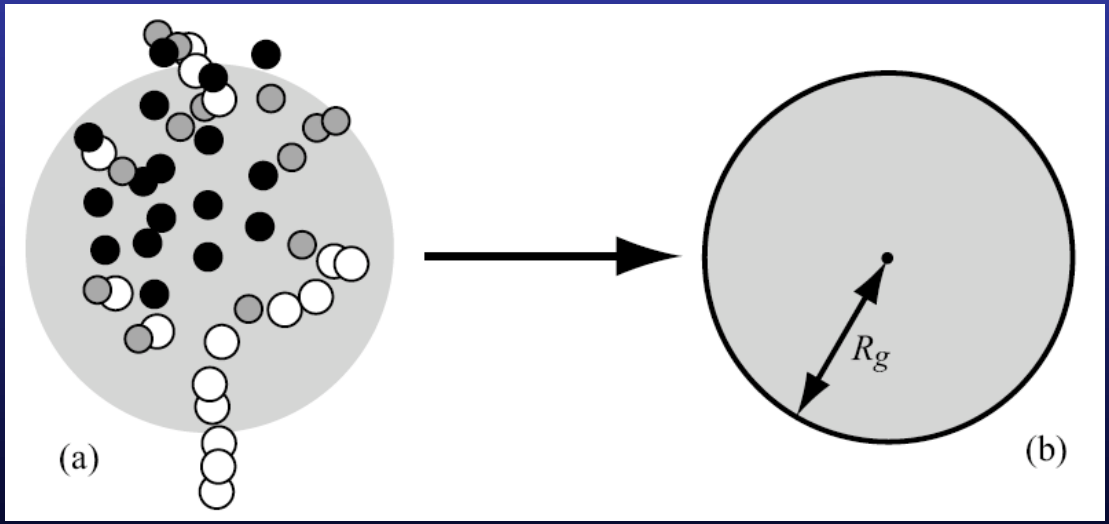
In a real experiment, the detector is small and captures only a portion of the scattering directions



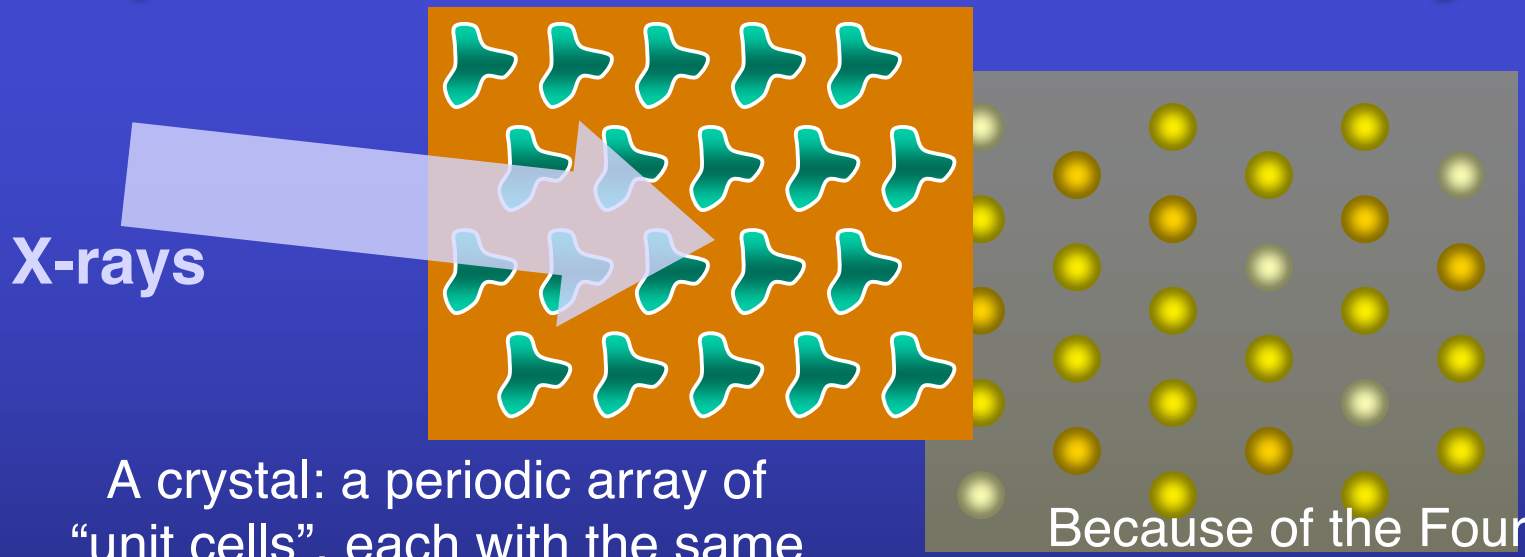
Because of the Fourier transform properties, when only small scattering angles are detected, then the inverse transform only gives the general shape of the object and not its fine details

Small-angle x-ray scattering (SAXS)

Consider the scattering angular parameter λ/δ: for $\lambda \approx 1$ Å and $\delta \approx 10$ Å, the angle magnitude is 0.1 radians or ≈ 6 degrees: SAXF reveals features 10 Å big or larger. In first approximation, it simulates the object as a sphere, measuring its radius



A special case: x-ray scattering from periodic structures -- such as crystals:



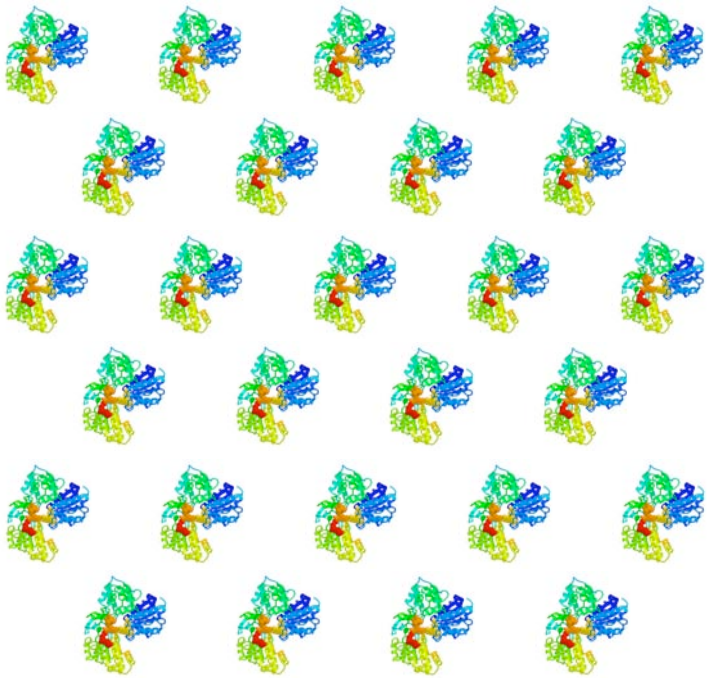
A crystal: a periodic array of "unit cells", each with the same electronic charge distribution

Because of the Fourier transform properties, the x-ray diffraction pattern consists of periodic "Bragg spots"

From the intensities of the individual Bragg spots, we can derive the electronic distribution inside each unit cell

Protein crystallography

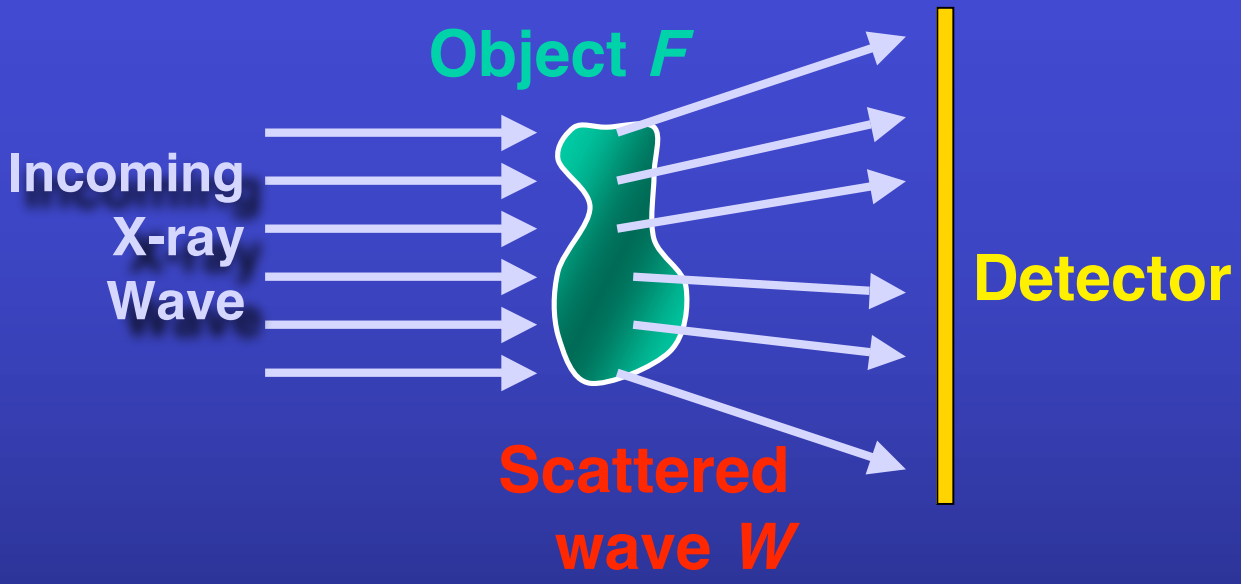
The accurate identification of protein structures with tens of thousands of atoms is one of the most important and challenging tasks for science today



However, damage induced by x-rays is a major problem. The standard solution is to measure simultaneously many molecules organized in a “crystal” and spread the damage

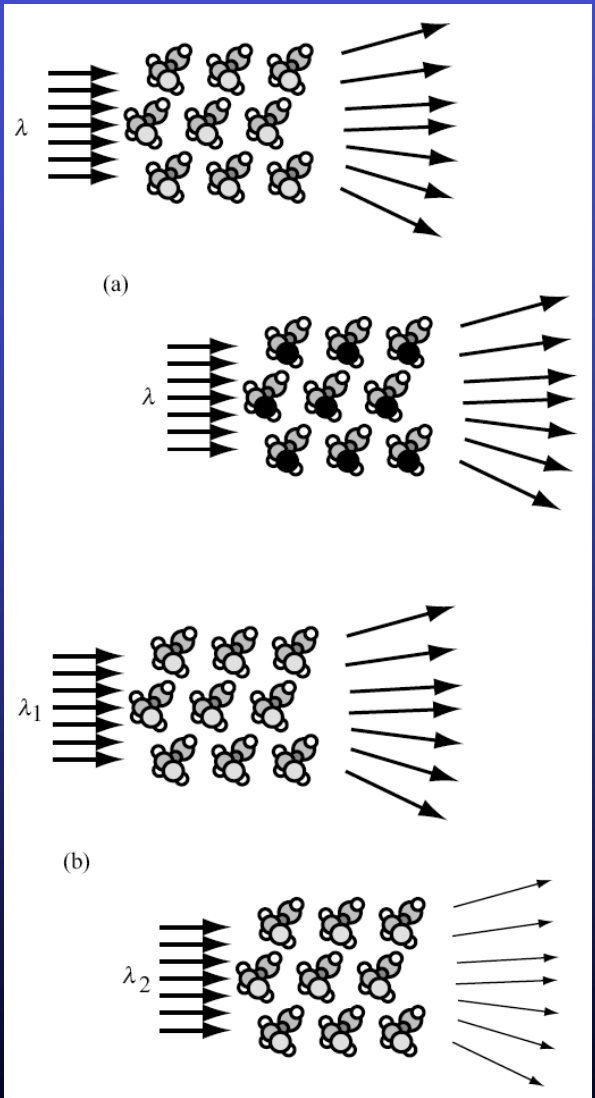
Problems : difficulties in obtaining large stable crystals, in particular for hydrophobic molecules -- e.g., many membrane proteins

The "phase problem":



- The detector does not measure the scattered wave W but its intensity $|W|^2$
- If the wave is $W_o \exp[i(kx - \omega t + \phi)]$, then $|W|^2 = W_o^2$: the information on the phase factor exp(…) is lost. This prevents the direct derivation of the molecular structure

Solving the phase problem in protein crystallography ("MAD" techniques)

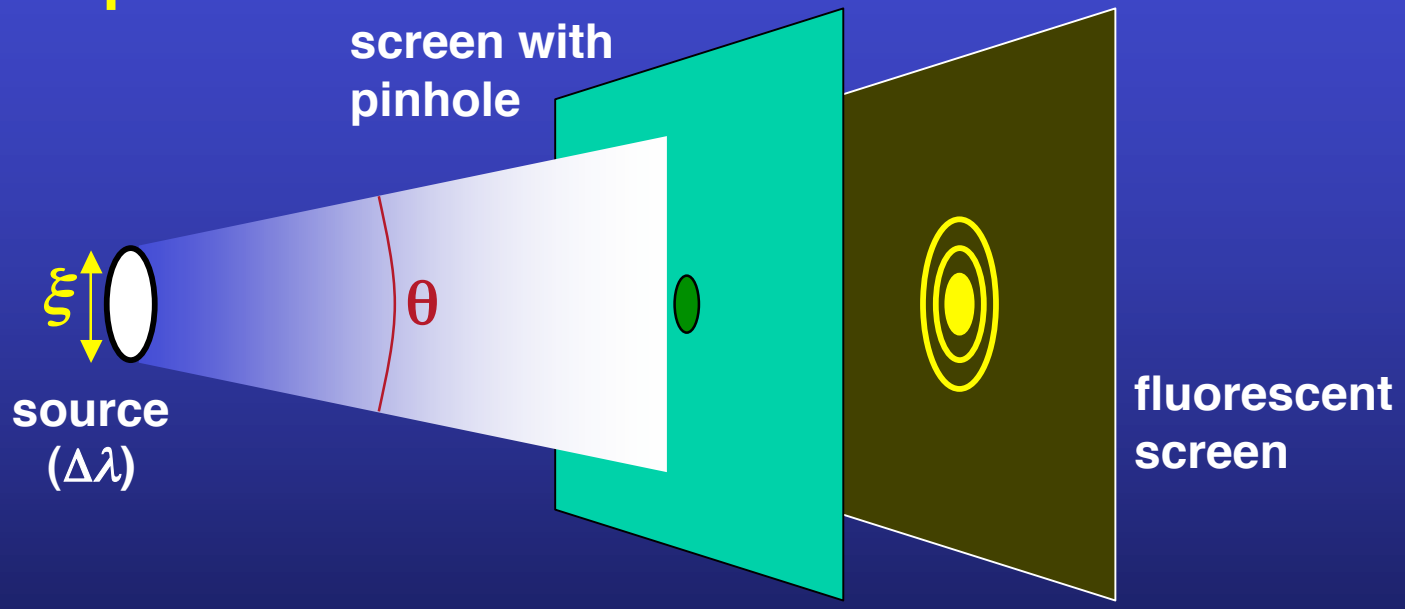


The classical solution is to measure diffraction patterns for the target molecule and for two (or more) modified versions ("isomorphous derivative") obtained by changing one or more atomic species. This complicated the task of getting good crystals

With wavelength-tunable synchrotron sources, patterns are now taken at different wavelengths. The phase problem is solved by exploiting the "anomalous scattering": rapid changes in the refractive index at wavelengths close to x-ray absorption thresholds

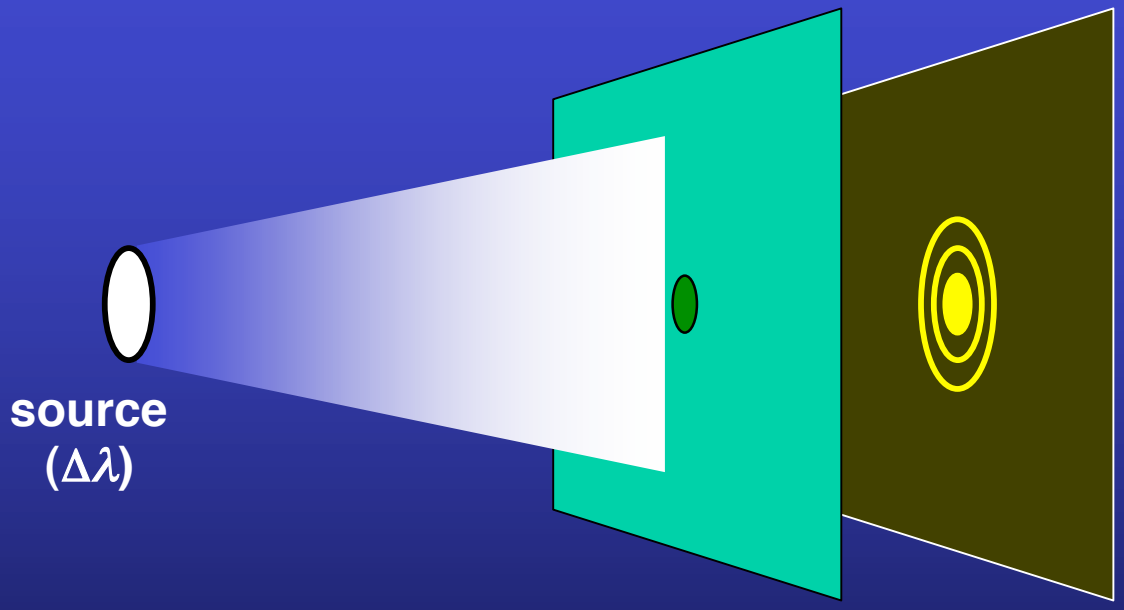
Synchrotrons make possible a **new radiology** based on **coherence:** "the property that enables a wave to produce **visible** diffraction and interference effects"

Example:



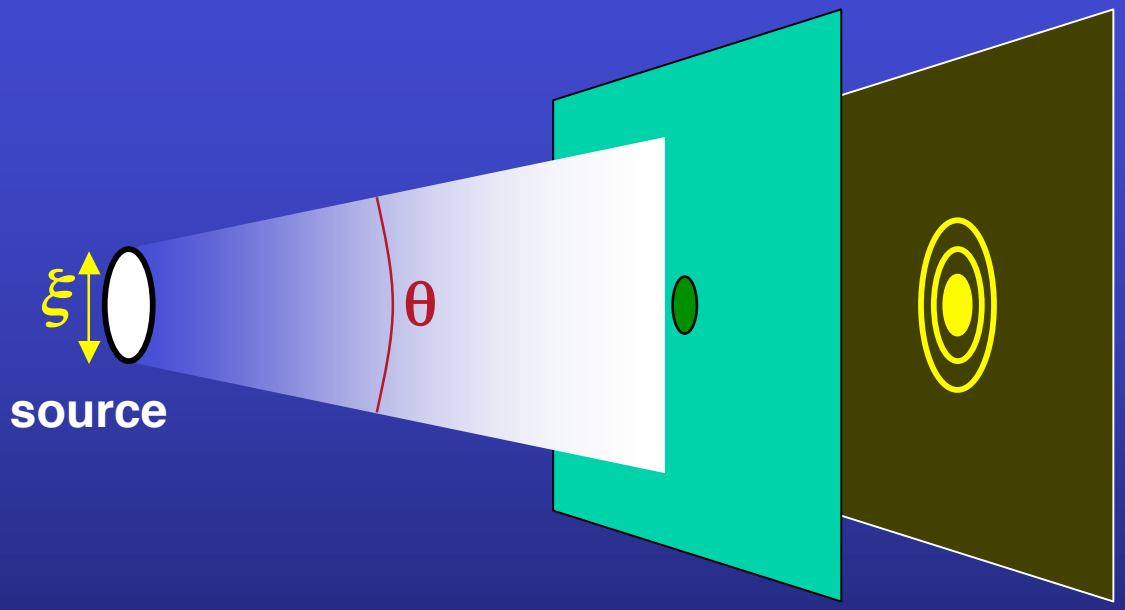
- **A point-source emitting only one wavelength always produces a visible diffraction pattern on the fluorescent screen**
- **For a real source, the pattern may or may not be visible depending on the source size ξ, on its angular divergence θ and on its wavelength bandwidth $\Delta\lambda$**

Longitudinal (time) coherence:



- **One wavelength ⇒ one diffraction pattern**
- **Several wavelength in a band $\Delta\lambda$ ⇒ several overlapping diffraction patterns ⇒ the fringes may or may not still be visible, depending on how large is $\Delta\lambda$**

Lateral (space) coherence:

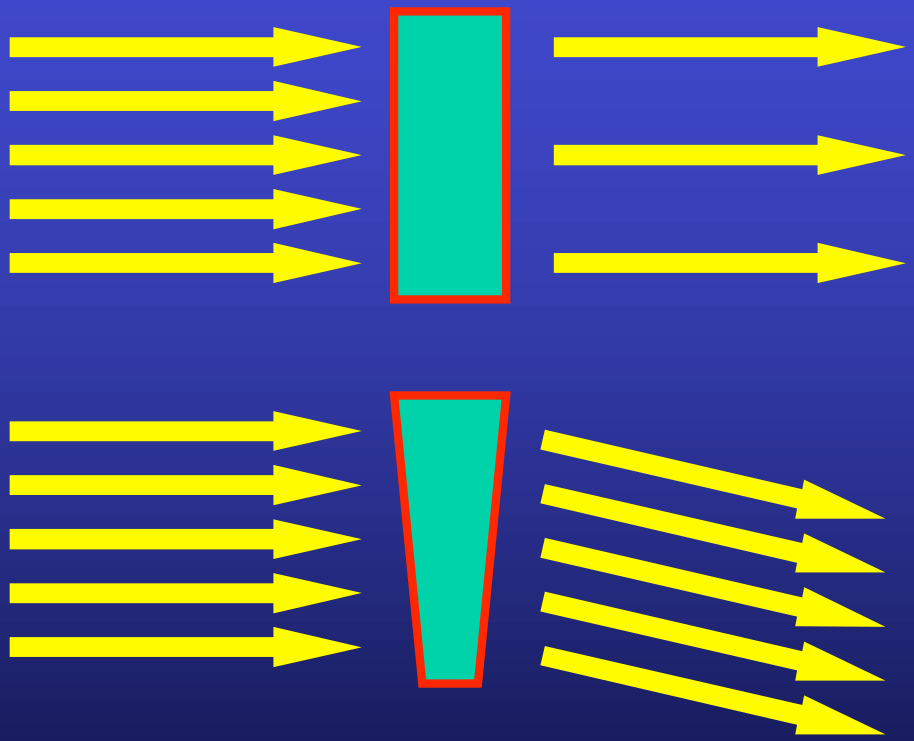


- One point-source (large divergence) ⇒ one diffraction pattern
- Extended source = a collection of point sources ⇒ several overlapping diffraction patterns ⇒ fringes may or may not still be visible, depending on how large are ξ and θ

Coherence — summary:

- Longitudinal coherence: requires a large coherence length $L_c = \lambda^2/\Delta\lambda$
- Lateral coherence: requires a large coherent power $\approx (\lambda/\xi\theta)^2$
- **Both difficult to achieve for small wavelengths (x-rays)**
- The geometric conditions for large $(\lambda/\xi\theta)^2$ are **the same as for high brightness**

Light-matter interactions in radiology:



Absorption -- described by the absorption coefficient α

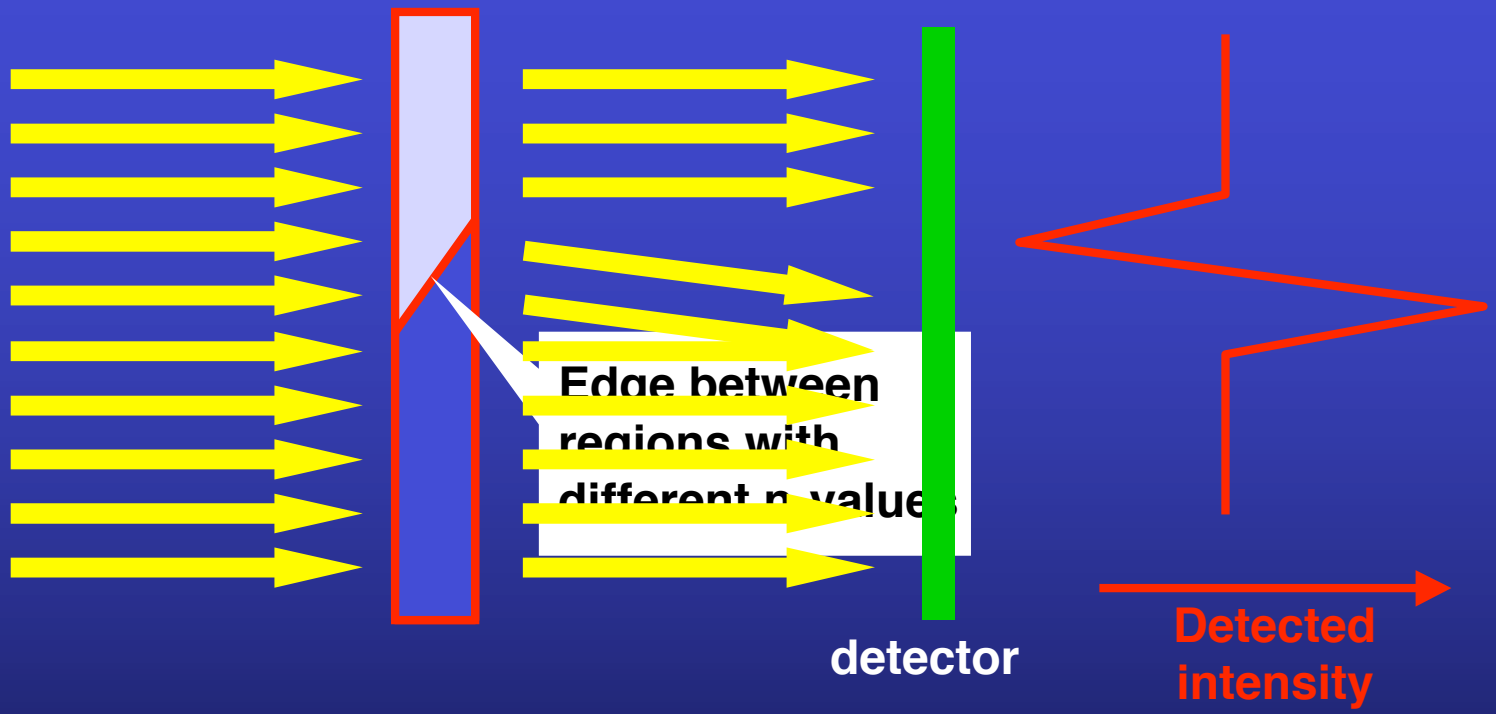
Refraction (and diffraction/interference) -- described by the refractive index *n*

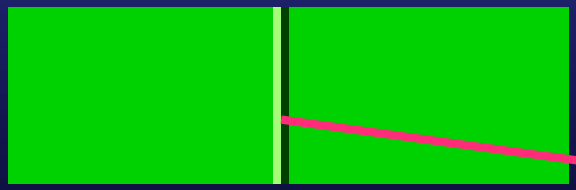
For over one century, radiology was based on absorption: why not on refraction /diffraction?

Conventional radiology

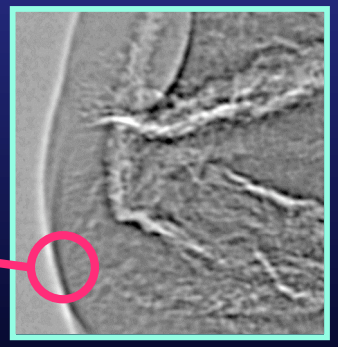
Refractive-index radiology (Giuliana Tromba)

“Refraction” x-ray imaging:





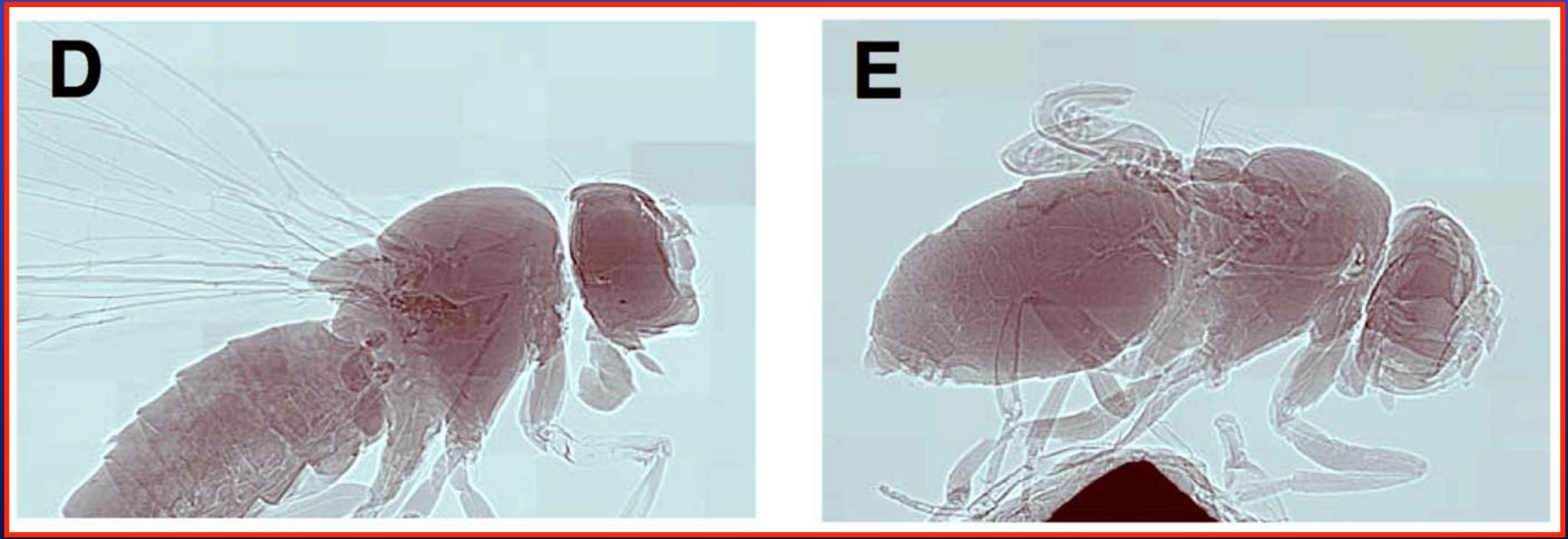
Idealized edge image



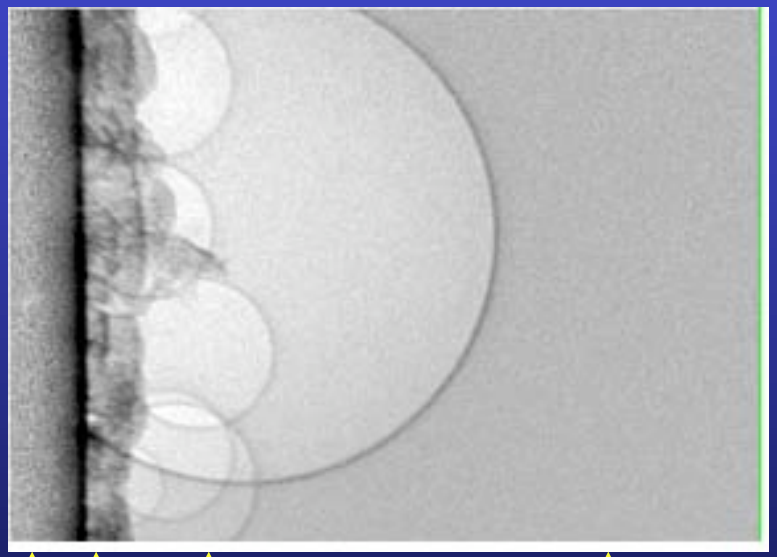
Real
example
(frog egg)

Microradiology study of mutant drosophila fly evolution

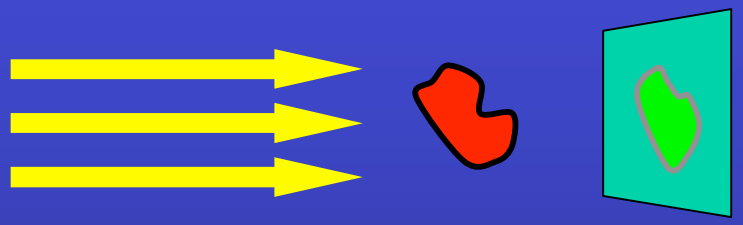
[Charron, Vassalli et al.]



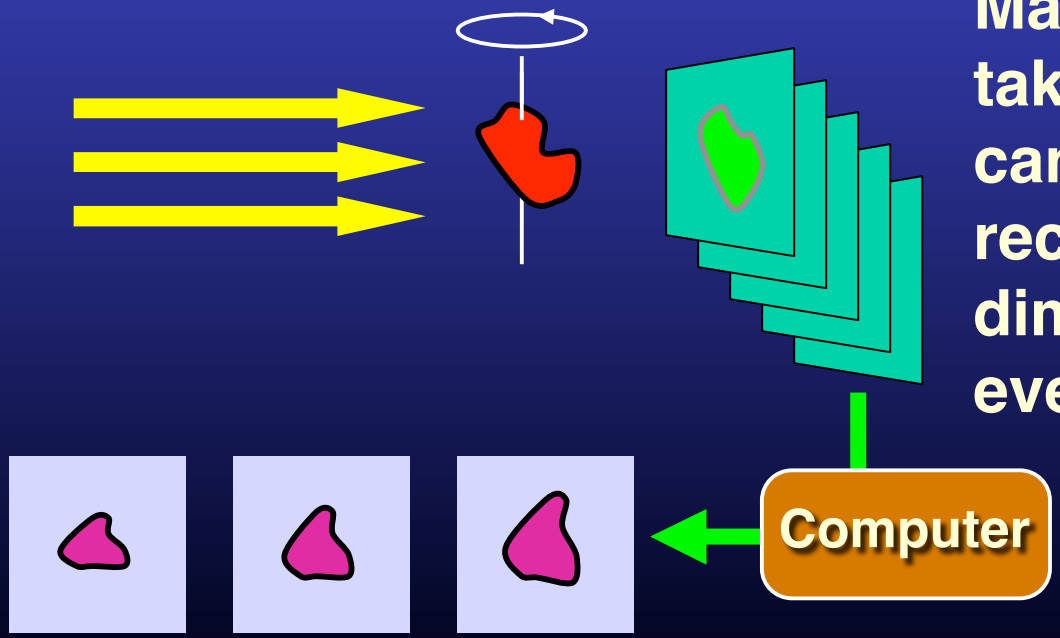
Building on bubbles (zinc electrodeposition):



X-ray (micro)tomography:

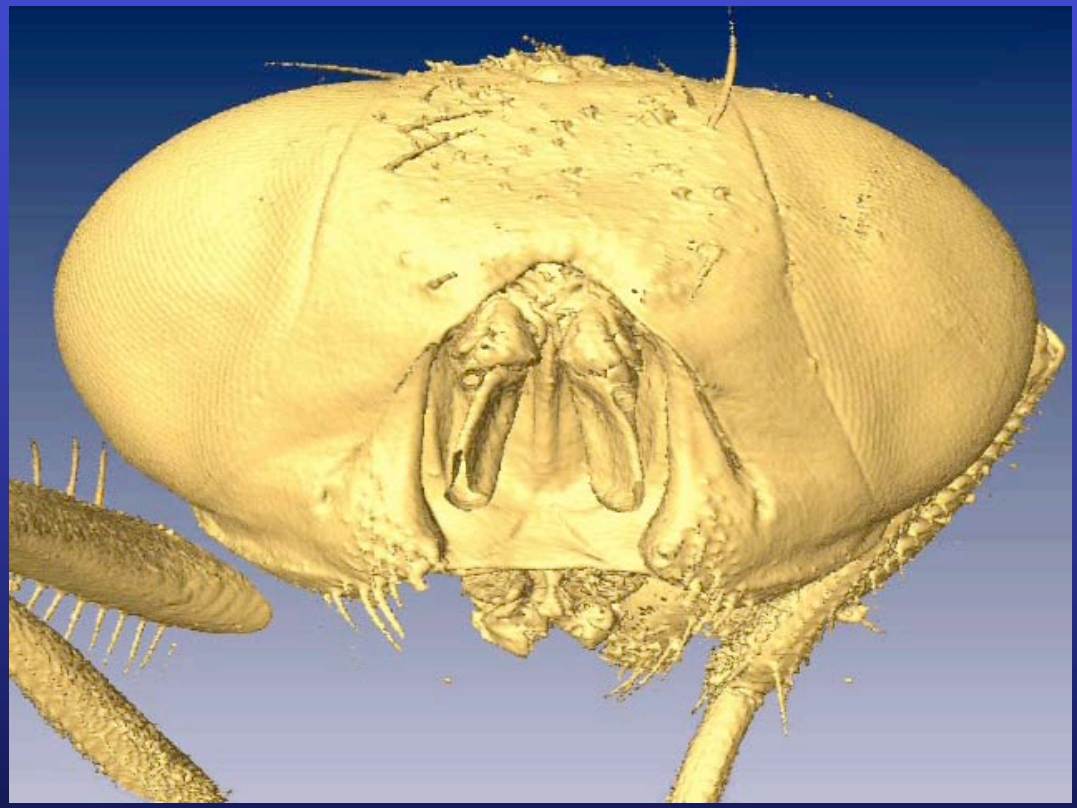


A single (projection) x-ray image does not deliver three-dimensional information



Many x-ray images taken at different angles can be computer-reconstructed in three dimensions -- and can even give movies

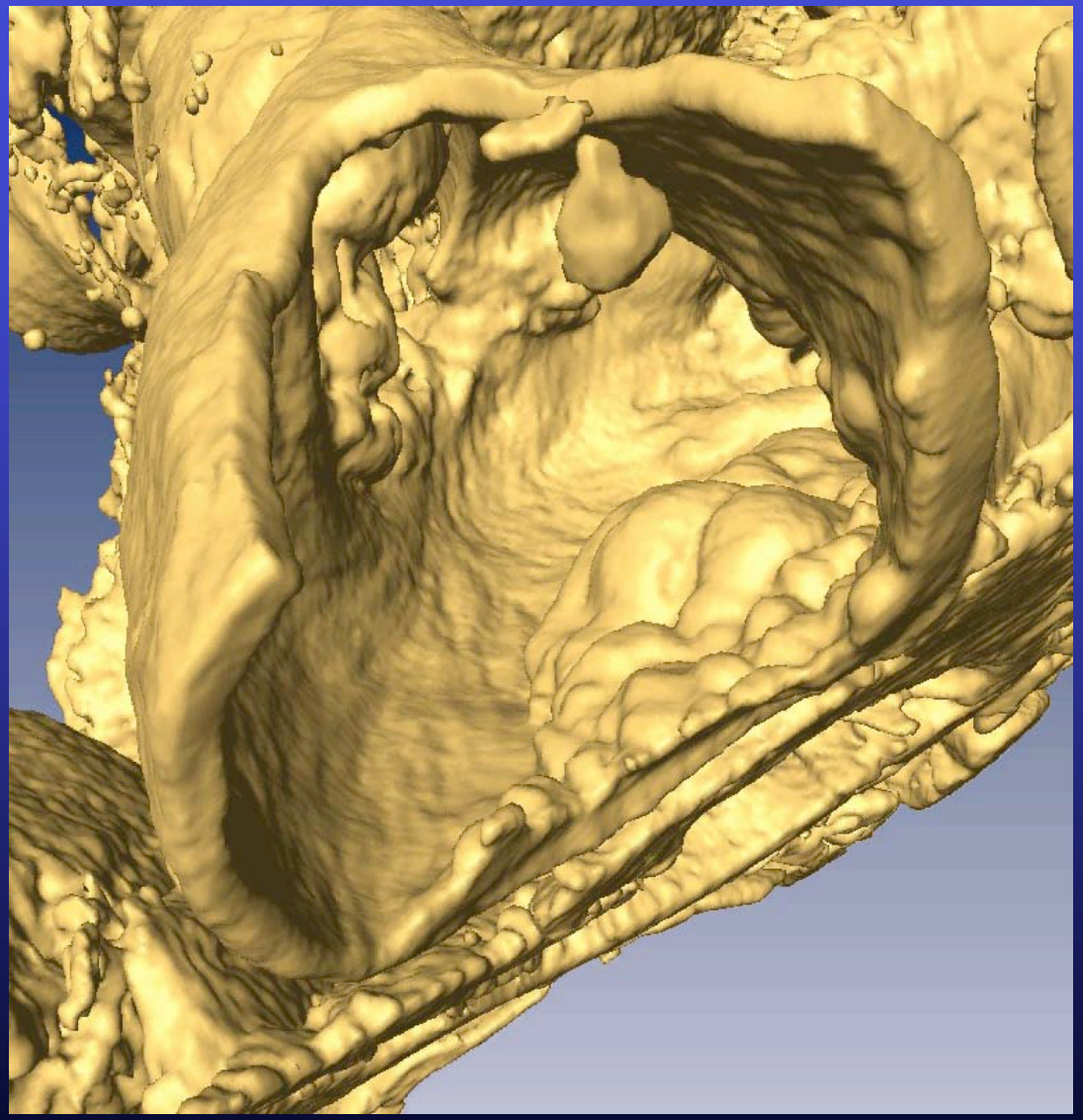
Phase contrast micro-tomography: housefly



Yeukuang Hwu, Jung Ho Je et al.

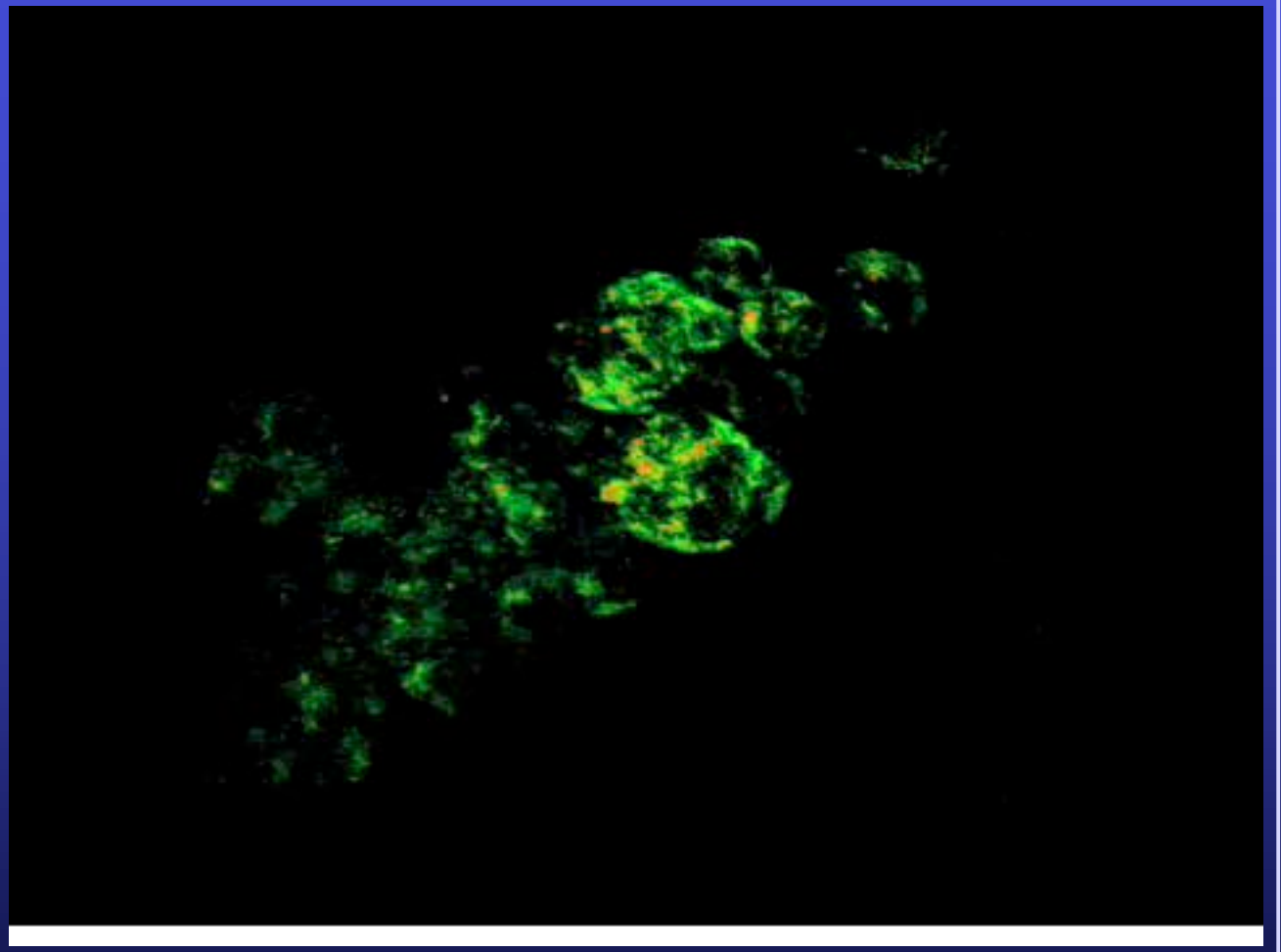
Phase contrast micro-tomography: navigating inside micro-vessels

Yeukuang Hwu, Jung Ho Je et al.



From nanofabrication to nanoanalysis

Y. S. Chu, J. M. Yi, F. De Carlo, Q. Shen, W.-K-Lee, H. J. Wu, C. L. Wang, J. Y. Wang, C. J. Liu, C. H. Wang, S. R. Wu, C. C. Chien, Y. Hwu, A. Tkachuk, W. Yun, M. Feser, K. S. Liang, C. S. Yang, J. H. Je, G. Margaritondo



Agglomerated Au nanoparticles attached to cancer cells

New types of sources:

- Ultrabright storage rings (SLS, new Grenoble project) approaching the diffraction limit
- Self-amplified spontaneous emission (SASE) X-ray free electron lasers
- VUV FEL's (such as CLIO)
- Energy-recovery machines
- Inverse-Compton-scattering table-top sources

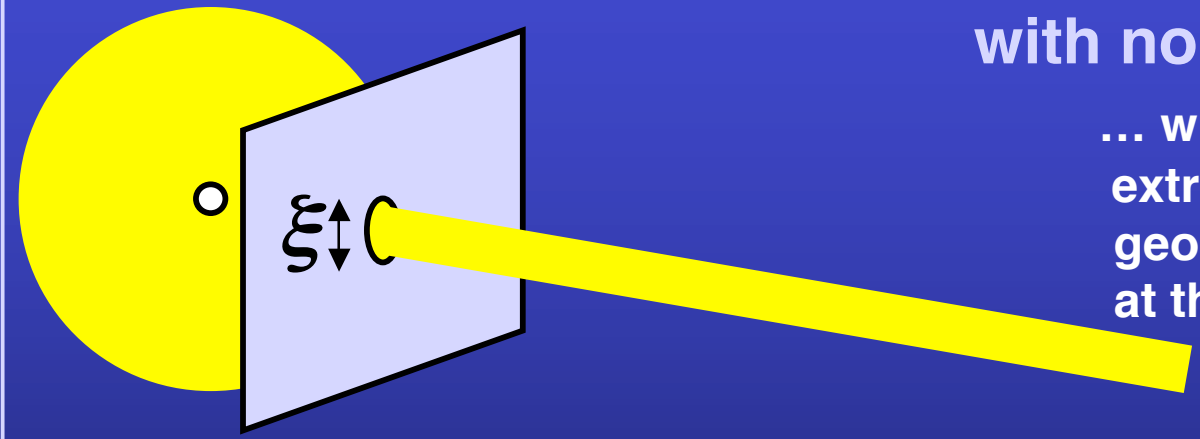
So far, we dealt with synchrotron sources with laser-like characteristics -- but not with real lasers:

- Can we go to true lasing?
- ANSWER: not only we can, but we must -- the present synchrotron sources are reaching their natural limits

The absolute geometric limit for coherence and brightness:

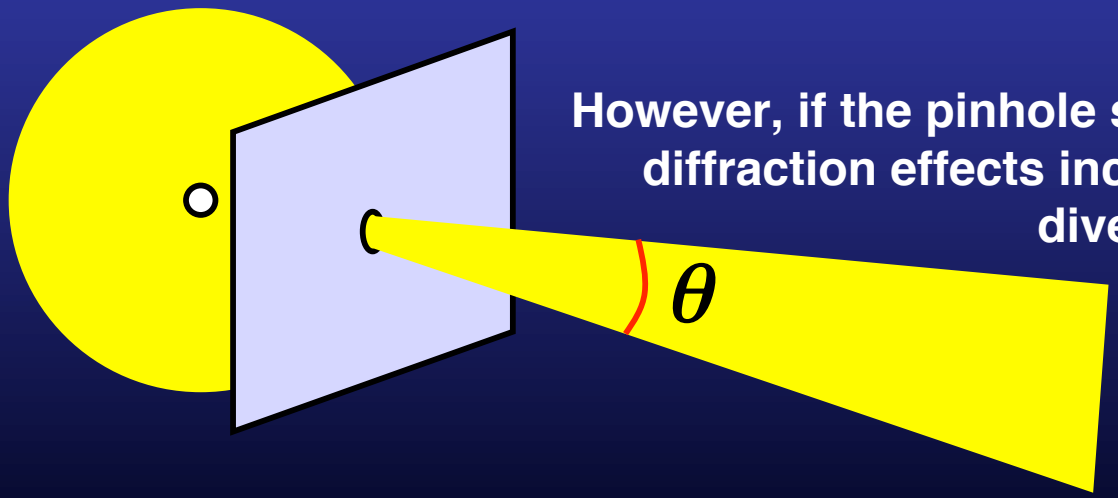
Take a standard photon source with no lateral coherence …

… with a pinhole (size ξ), we can extract coherent light with good geometrical characteristics (but at the cost of losing most of the emission)



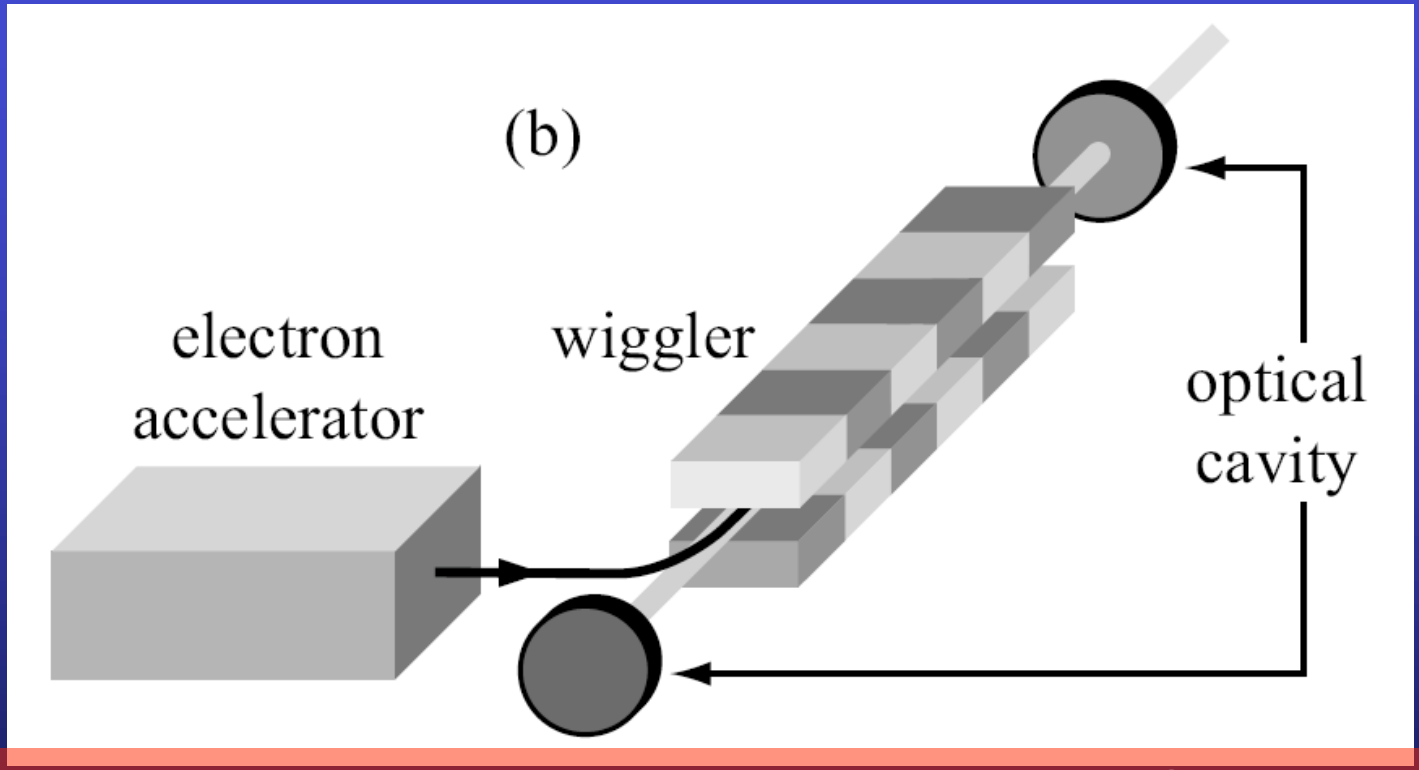
However, if the pinhole size is too small diffraction effects increase the beam divergence so that:

$$\xi\theta > \lambda$$



No source geometry beats this diffraction limit

Free-electron lasers (FEL's):



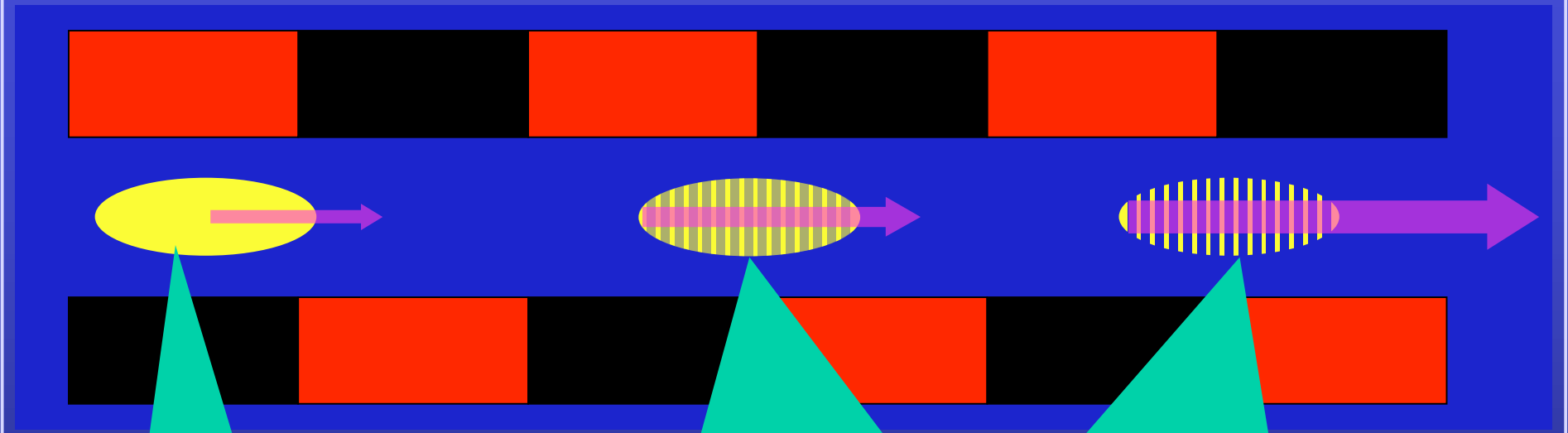
A lasing mechanism requires optical amplification: for an FEL, the amplification is given by the interaction of the electrons passing through a wiggler (or undulator) with previously emitted photons waves

Why do we need a wiggler?

The transverse electron oscillations induced by the wiggler are required to activate the interaction between electrons and emitted photon waves that produces optical amplification:

1. **They enable energy transfer between the electrons and the photon waves**
2. **Furthermore, they assist the photon waves in "microbunching" the electrons**

Details of FEL mechanism:



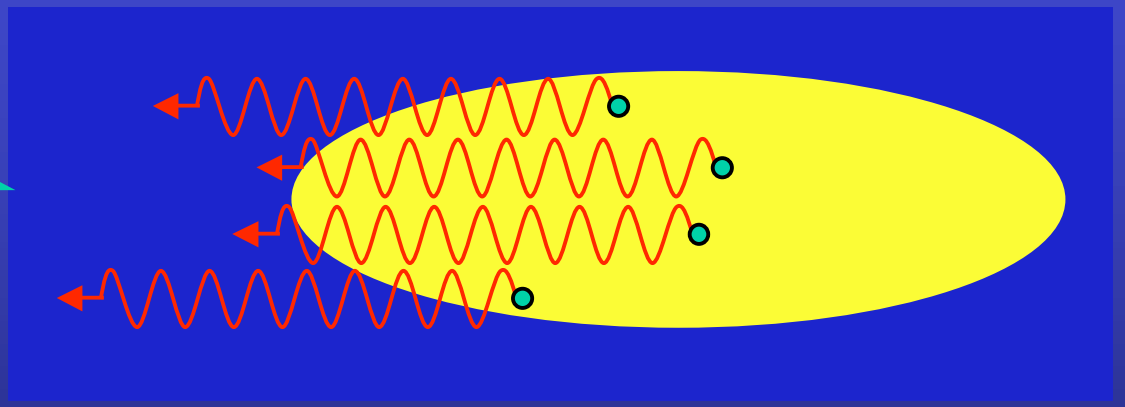
Electrons enter the wiggler: some of them stochastically start emitting waves

The combined wiggler+wave action progressively microbunches the electrons. The emission of microbunched electrons enhances the previously emitted waves

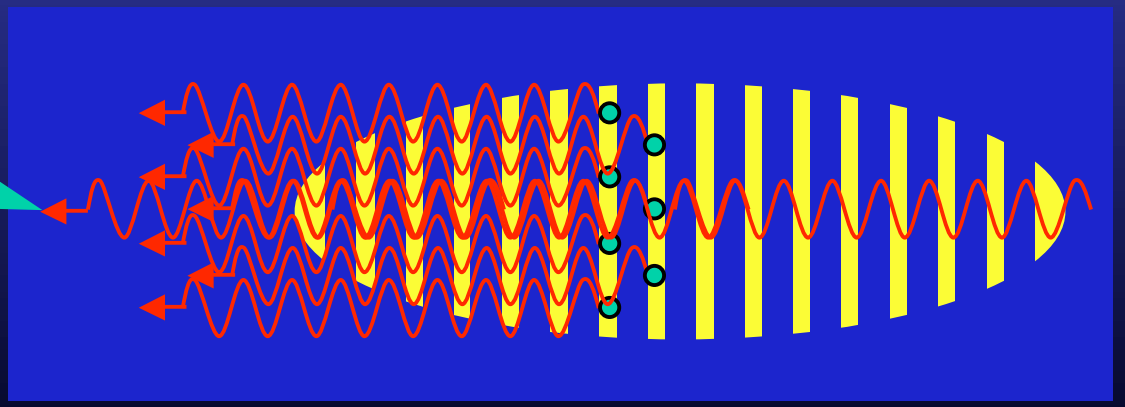
- The initial emission intensity is proportional to the number of electrons, n
- The subsequent microbunching increases with the wave intensity, thus is also proportional to n
- The final amplified intensity is proportional to n^2

Microbunching makes the difference!

With no microbunching, electrons emit in an uncorrelated way: no optical amplification



Instead, the electrons in wiggler-induced microbunches emit coherently, amplifying previously emitted waves



Key factors for strong optical amplification, leading to FEL action:

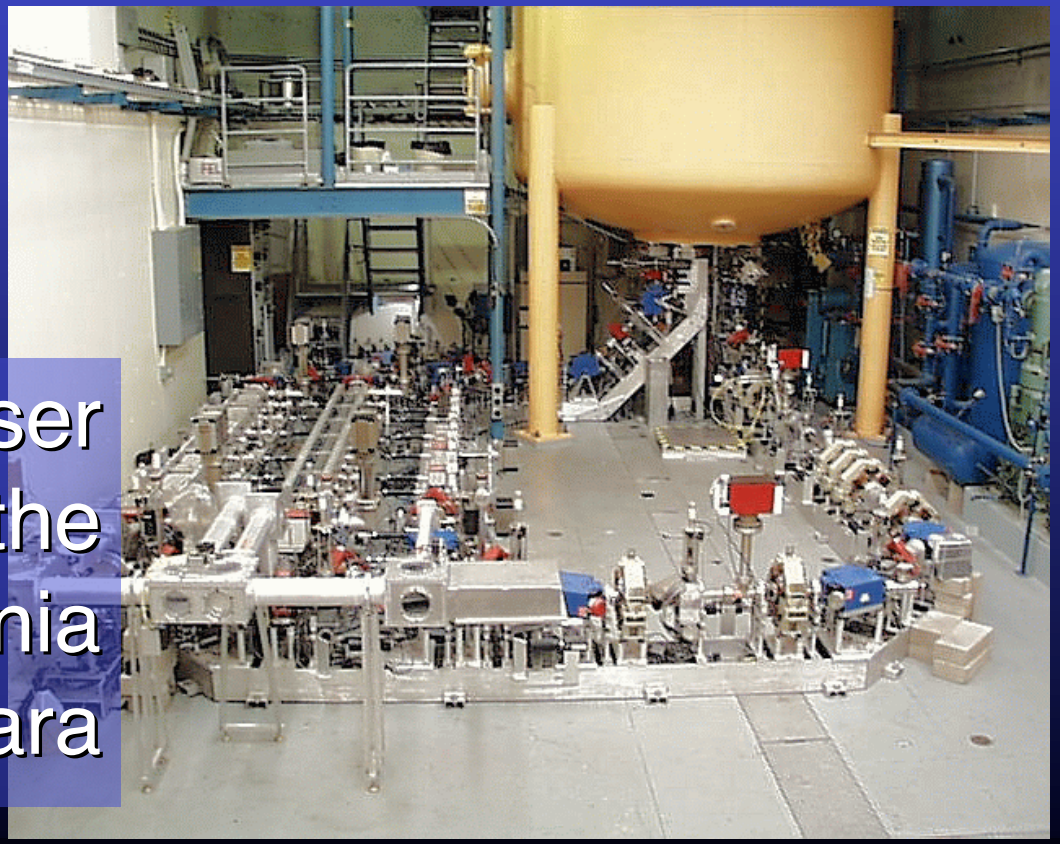
1. A well-defined electron energy: limited “energy spread”
2. A small transverse cross section of the electron beam.
3. Small angular deviations of the real electron trajectories from the “reference path”
4. The optical amplification increases with:
   - The wiggler length
   - The number of wiggler periods, squared
   - The electron beam current density
   - *The wavelength*

FELs are much easier to build and operate for infrared photons than for x-rays

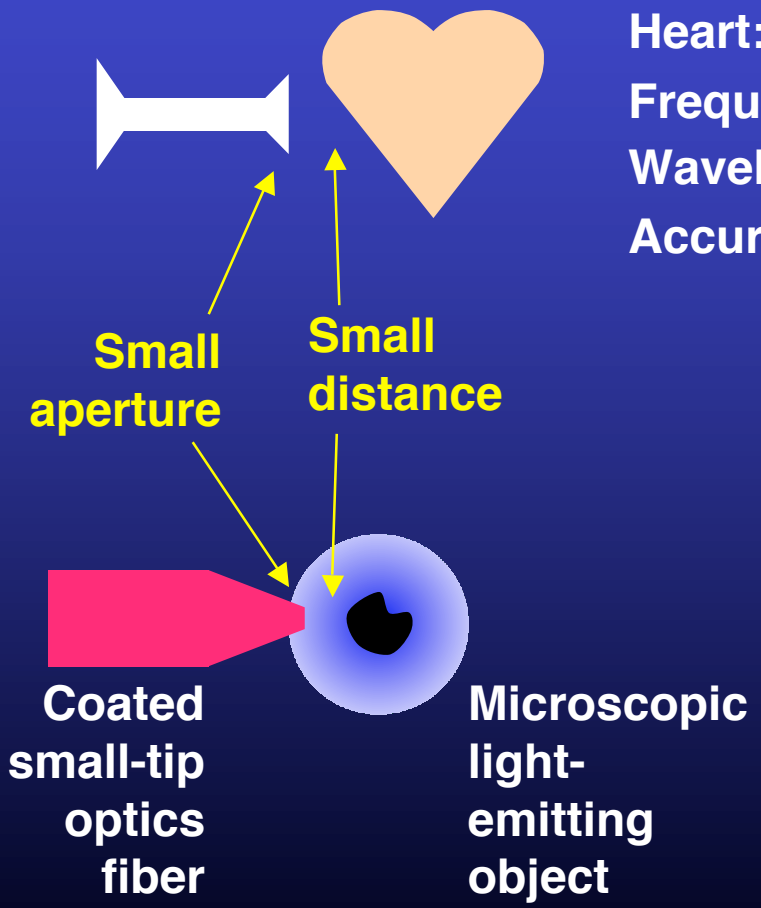
Furthermore, for infrared FELs the optical gain can be assisted by the use of an optical cavity (two mirrors).
This is **not possible** for x-ray FELs **(no x-ray mirrors!)**

Example of working infrared FELs:

The free electron laser facility of the University of California at Santa Barbara



Use of infrared FEL's: The scanning near-field optical microscope (SNOM) -- like a "stethoscope"



Heart:

Frequency ≈ 30-100 Hz

Wavelength $\lambda \approx 102$ m

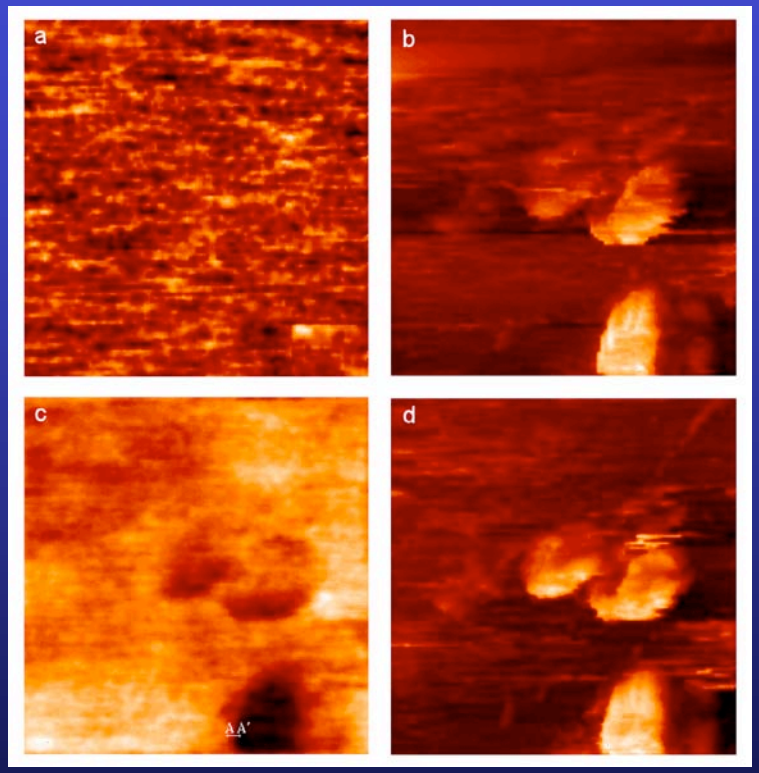
Accuracy in localization ≈ 10 cm ≈ λ /1000

SNOM resolution: well below the "diffraction limit" of standard microscopy (≈ λ)

20x20 μm^2 SNOM image of growth medium (A. Cricenti et al.):

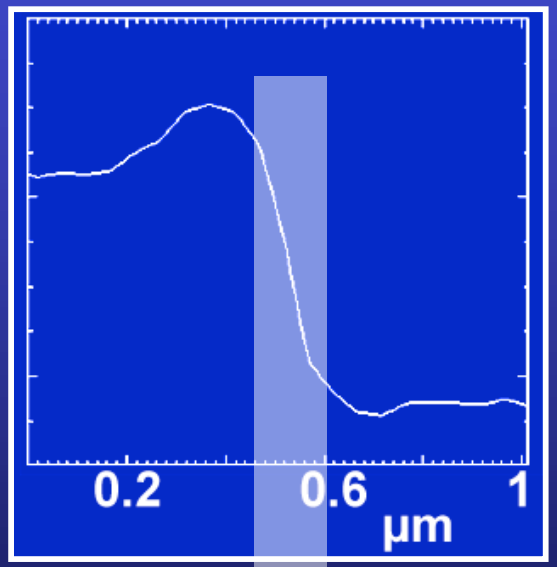
λ = 6.6 μm

S-O & N-O vibrations (λ = 6.95 μm)



SNOM topography

Intensity line scan



Resolution

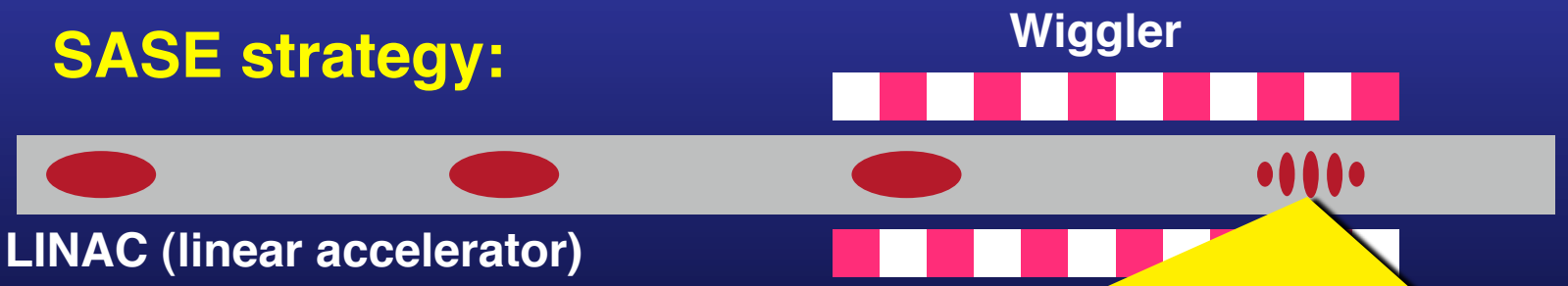
≈ 0.15 μm << λ

Self-Amplified Spontaneous Emission X-ray Free-Electron Lasers (SASE X-FEL's)

For most lasers: the optical amplification is enhanced by an optical cavity (two mirrors)

X-ray lasers: no mirrors → no optical cavity → lasing requires enough optical amplification <u>in one pass</u>

SASE strategy:



Microbunching must produce enough optical amplification for one-pass lasing

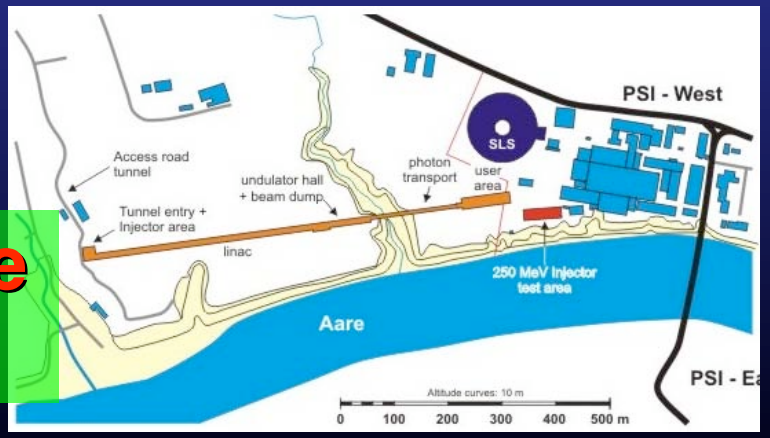
April 21, 2009 - New Era of Research Begins as World's First Hard X-ray Laser Achieves "First Light"

X-ray laser pulses of unprecedented energy and brilliance produced at SLAC

The FERMI X-FEL at Elettra, Trieste

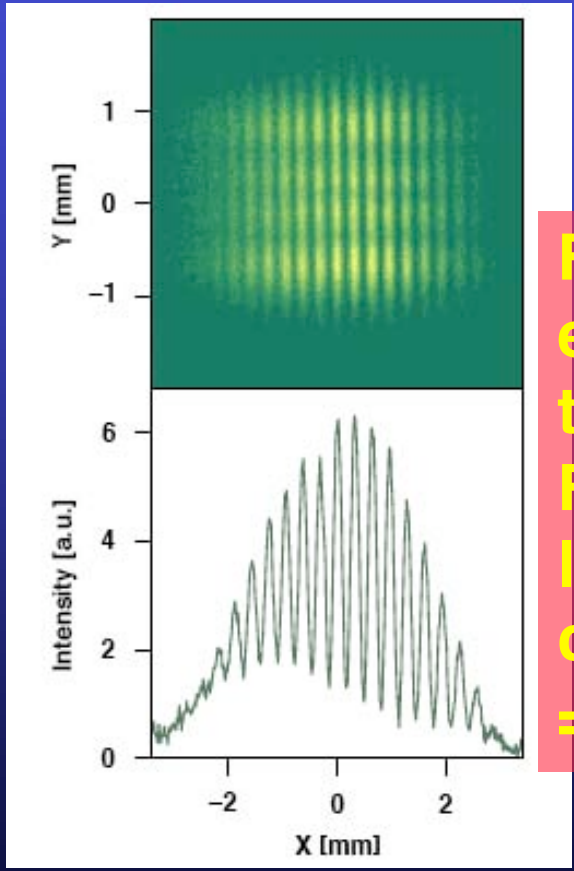
The European X-FEL project underway at DESY, Hamburg

The Swiss X-FEL at the Paul-Scherrer Institut



SASE-FEL coherence:

Full lateral (space) coherence all the way to the hard x-rays



First coherence experiments on the Tesla Test Facility: full lateral coherence at λ = 95 nm

SASE-FEL's: an exciting future

- Full coherence
- Unprecedented concentration of electromagnetic energy in space and time: new physics?
- Ultrashort (femtosecond) pulses: real time chemistry
- One-shot diffraction -- taking the data "before" the x-rays cause the object explosion: protein crystallography without crystals?

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