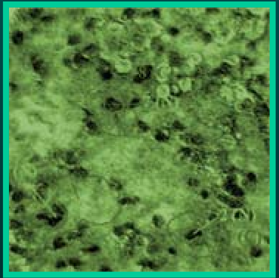
Experiments with Synchrotron Radiation: Basic Facts and Challenges for Accelerator Science



Giorgio Margaritondo

Dean, School of Basic Sciences, EPF-Lausanne

Summary:

- How to build a good x-ray source using relativity
- Recent progress -- ultrahigh brightness and how to use it:
  - Ultrahigh resolution spectroscopy
  - From spectroscopy to spectromicroscopy
- Coherent x-rays: The new radiology
- From synchrotrons to FEL's and their use:
  - Microscopy beyond $\lambda/2$
- The future: SASE X-ray FEL's

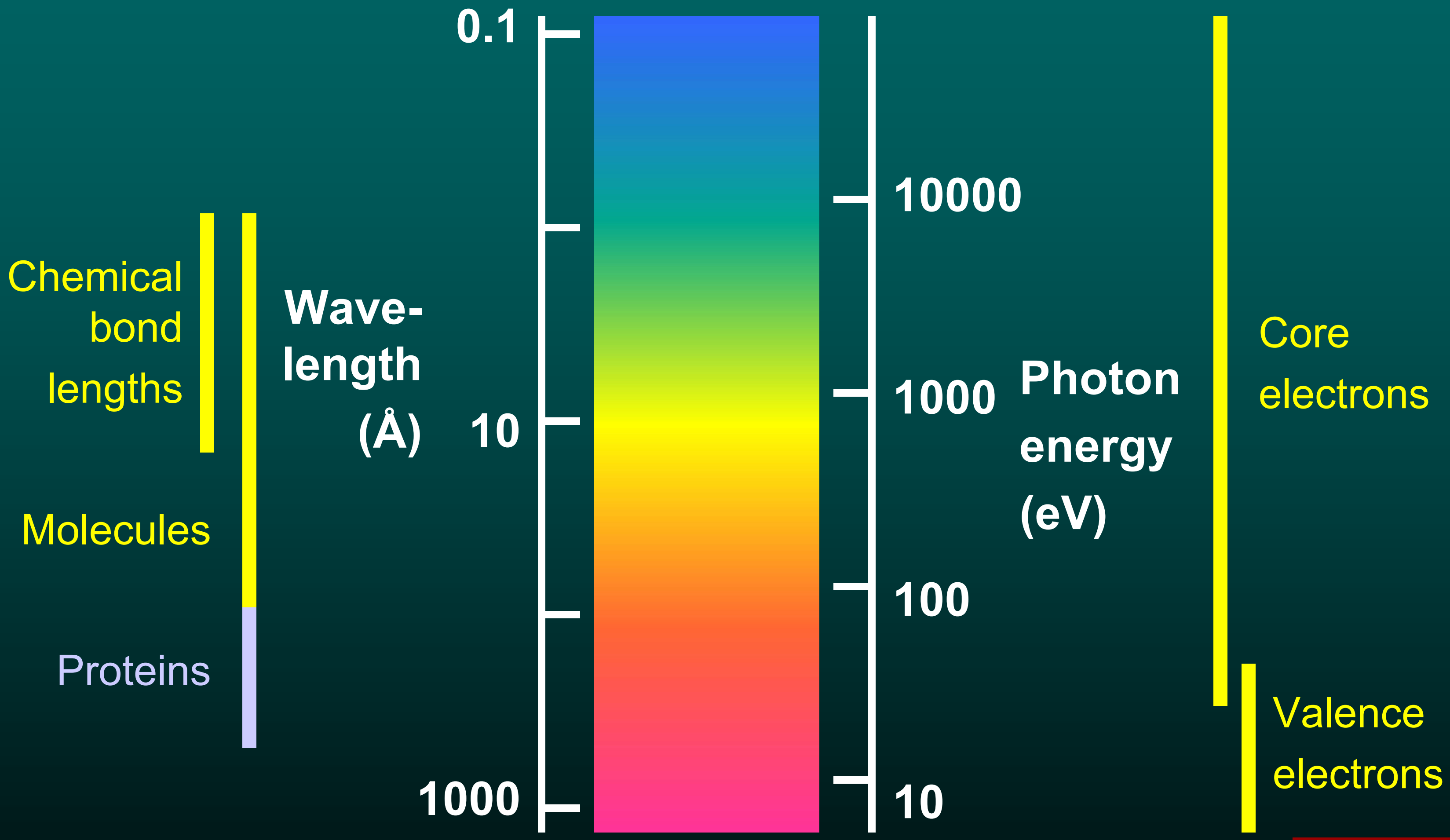
From ancient fires to synchrotrons and FEL's, the same problems:

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

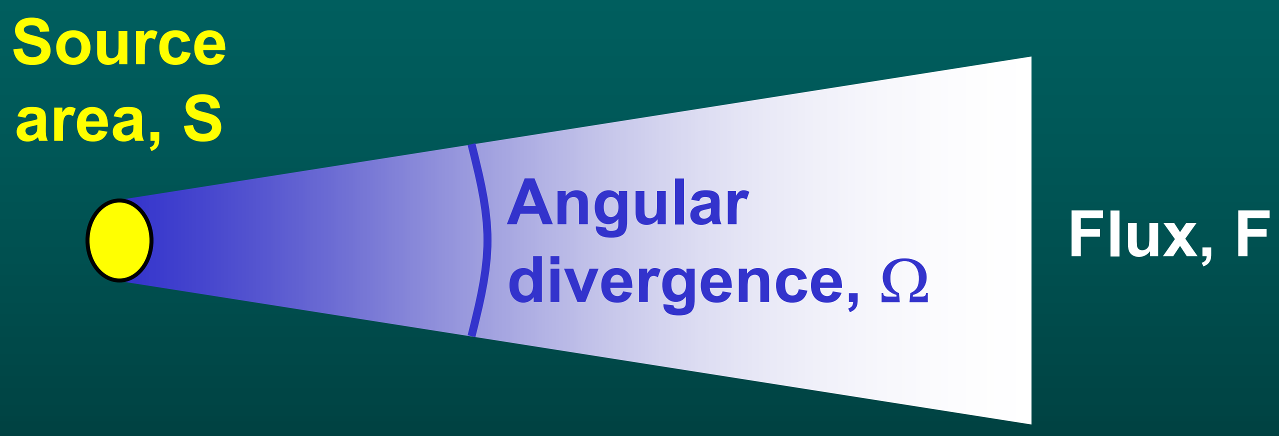
A torchlight is much more effective: it is a small-size source with emission concentrated within a narrow angular spread -- it is a "bright" source

Likewise, we would like to use "bright" sources for x-rays (and ultraviolet light)

Why x-rays and ultraviolet?



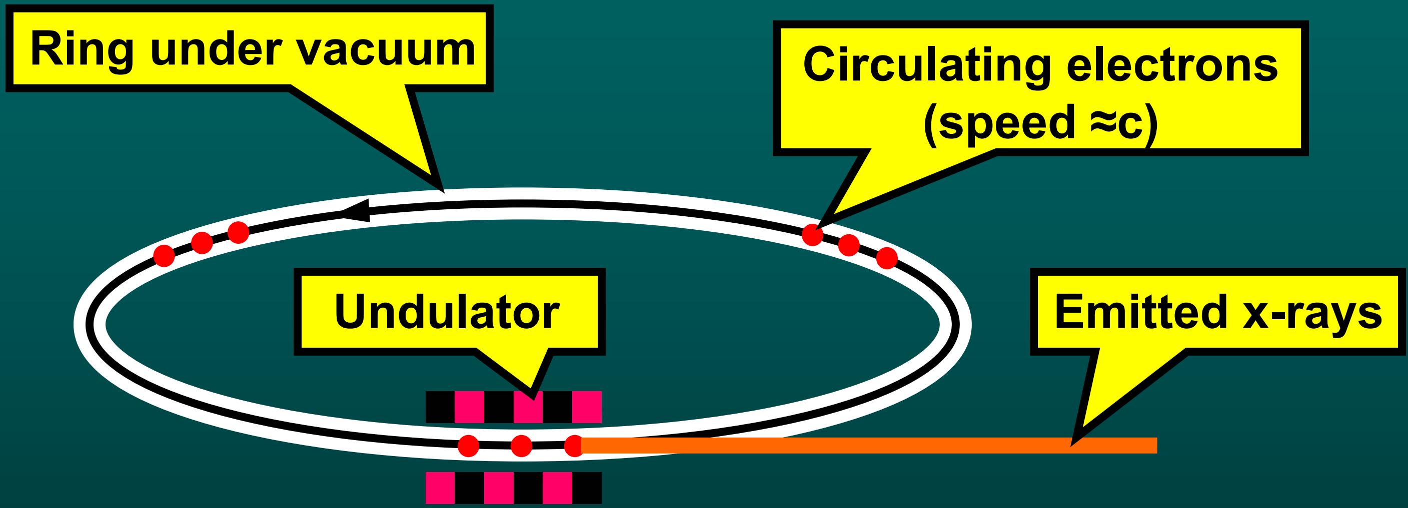
The “brightness” of a light source:



$$\text{Brightness} = \text{constant} \times \frac{F}{S \times \Omega}$$

Objective: building a very bright x-ray source.

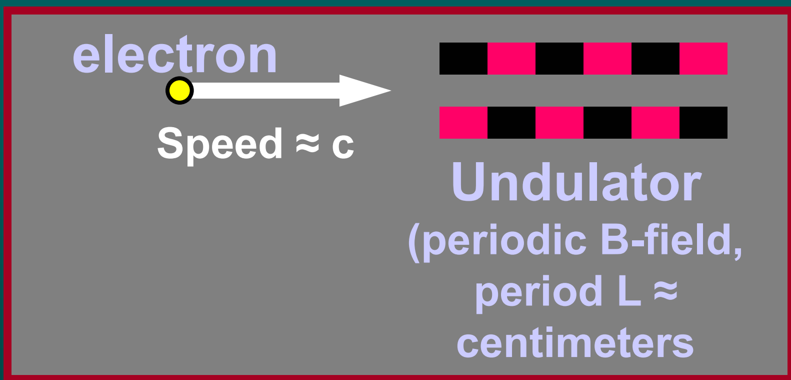
Solution: relativity!!



- The undulator (periodic magnet array) period determines the emitted wavelength. This period is **shortened by the relativistic "Lorentz contraction"** giving x-ray wavelengths
- The emitted x-rays are "**projected ahead**" by the motion of their sources (the electrons), and therefore collimated. Relativity enhances the effect

Objective: building a very bright x-ray source

Details of the solution:



In the electron reference frame:

- Periodic B-field → periodic B & E-fields moving at speed ≈c, similar to electromagnetic wave
- Lorentz contraction: L → L/γ
- Undulation of electron trajectory → emission of waves with wavelength L/γ

In the laboratory frame:

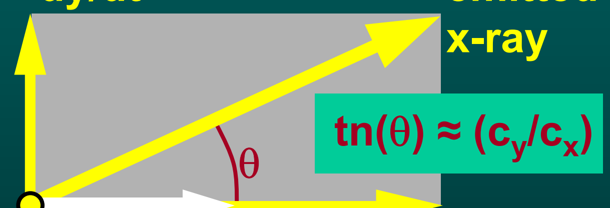
- Doppler effect → wavelength further reduced by a factor of ≈2γ, changing from L/γ to L/2γ^2

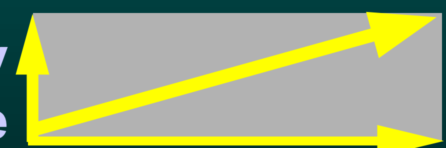
Overall: L → L/2γ^2

Centimeters → 0.1-1,000 Å (x-rays, UV)

What causes the high brightness?

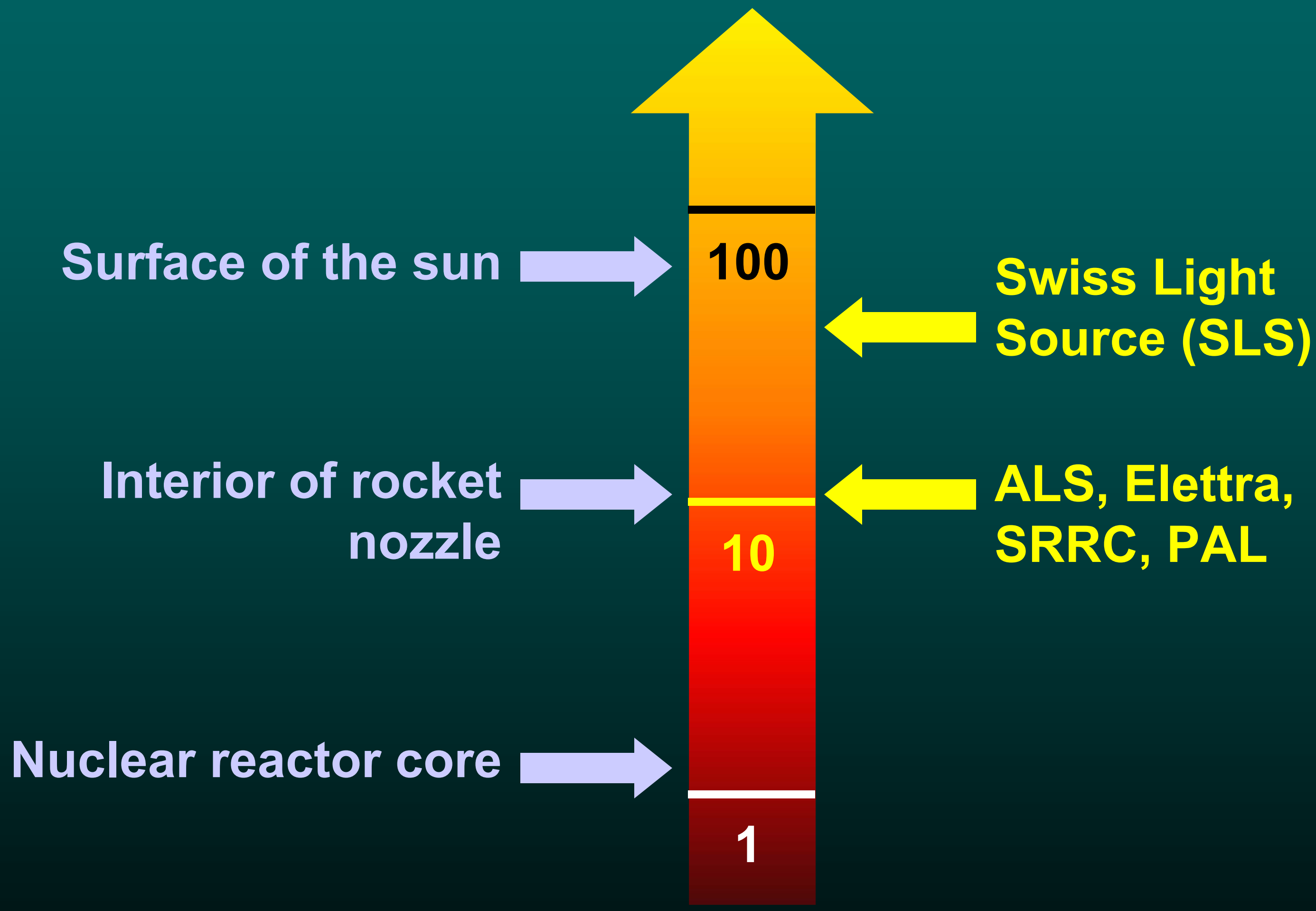
- Free electrons can emit more light than bound electrons ⇒ **high flux**
- The electron beam control is very sophisticated: small transverse beam cross section ⇒ **small synchrotron source size**
- Relativity **collimates** the emitted synchrotron radiation:





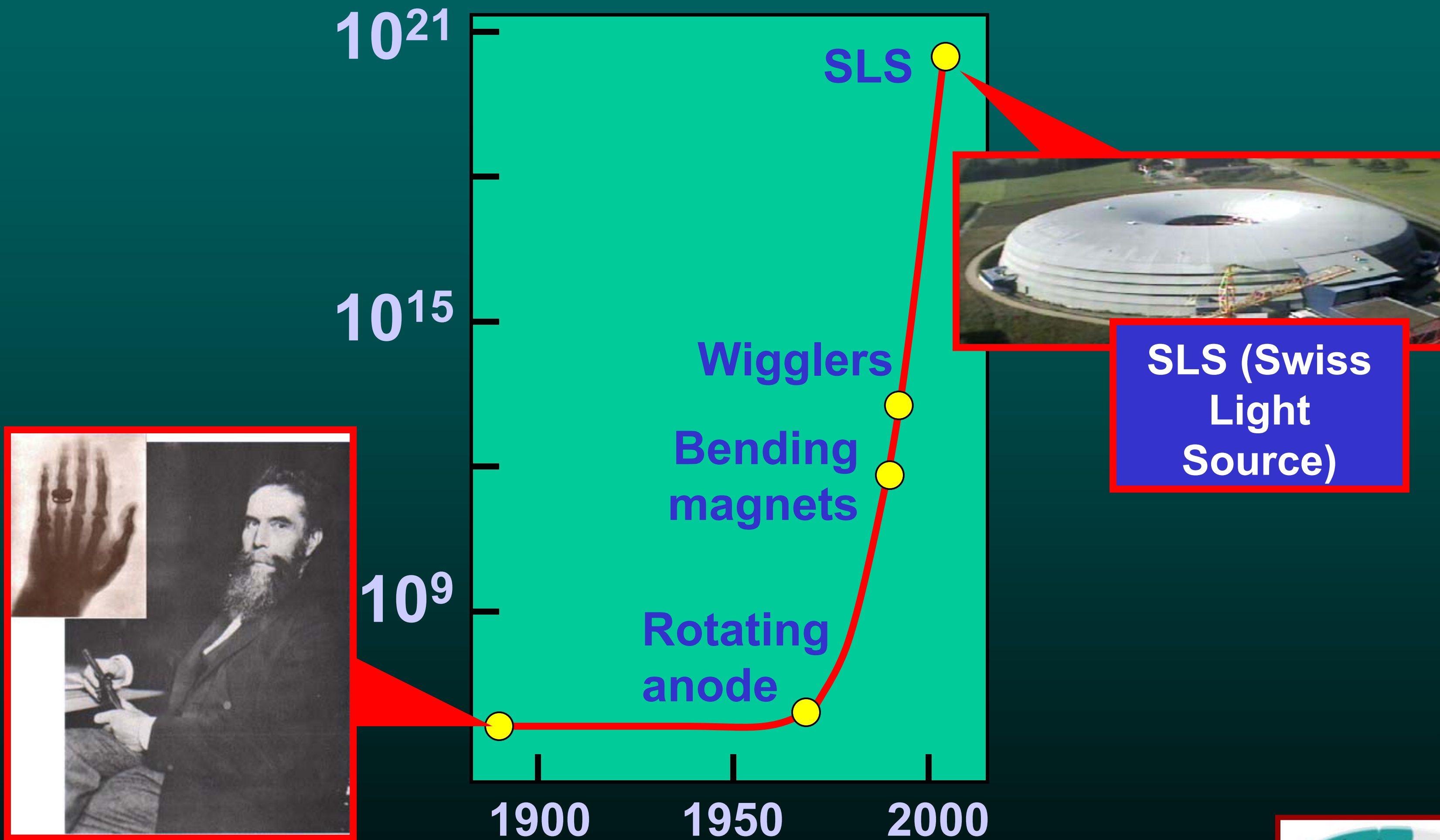
Lorentz transformation: γ-factor for x' and t' but not for y' ⇒ tn(θ) reduced by a factor ≈1/γ

Heat flux (watt/mm^2)



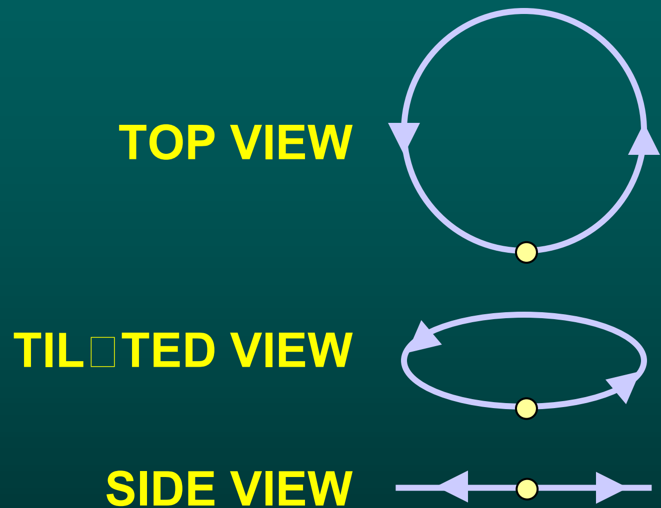
The historical growth in brightness/brilliance

(units: photons/mm²/s/mrad², 0.1% bandwidth)



Synchrotron light polarization:

<u>Electron in a storage ring:</u>



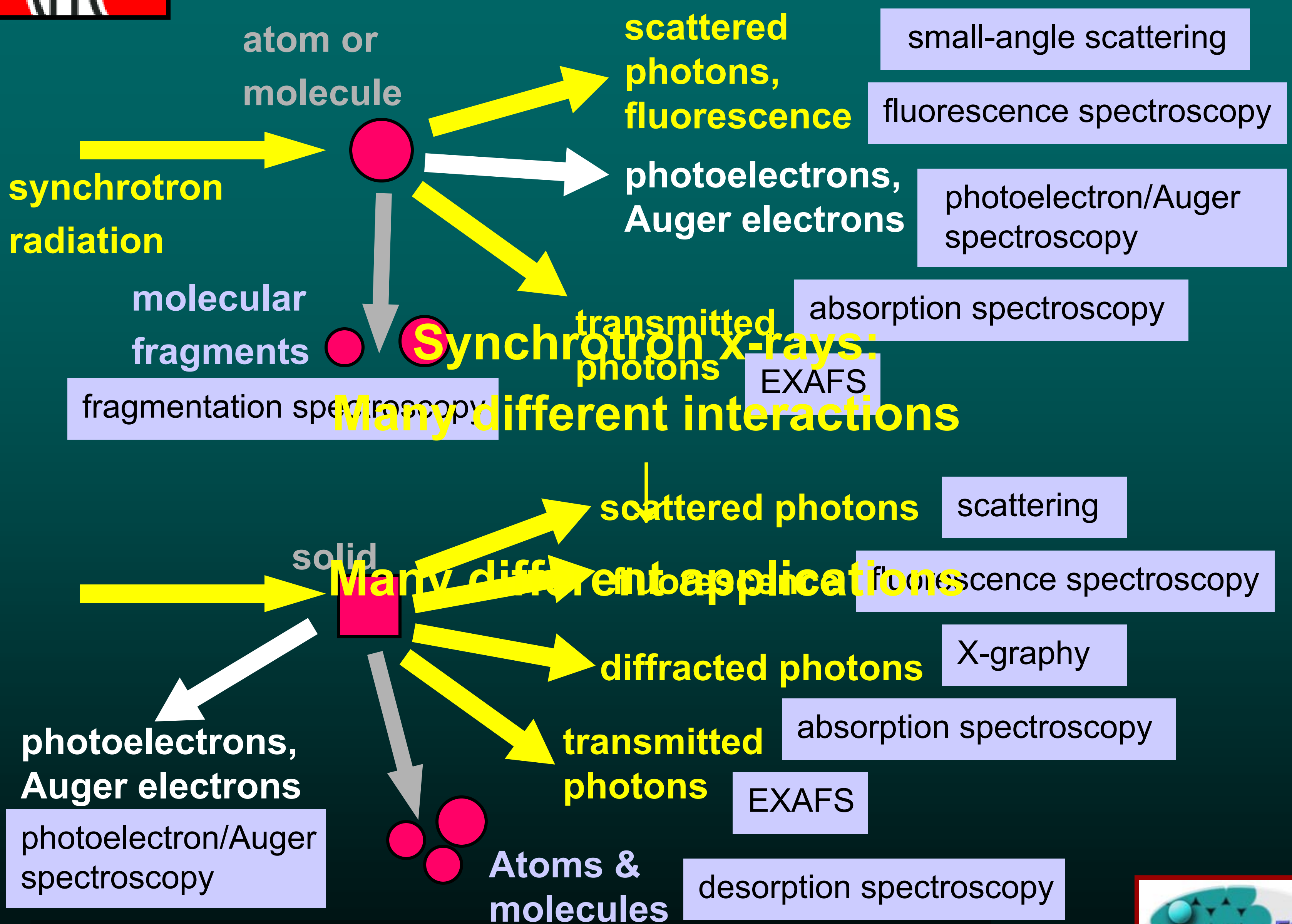
<u>**Polarization:**</u>

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

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

Synchrotron X-rays: Many different interactions

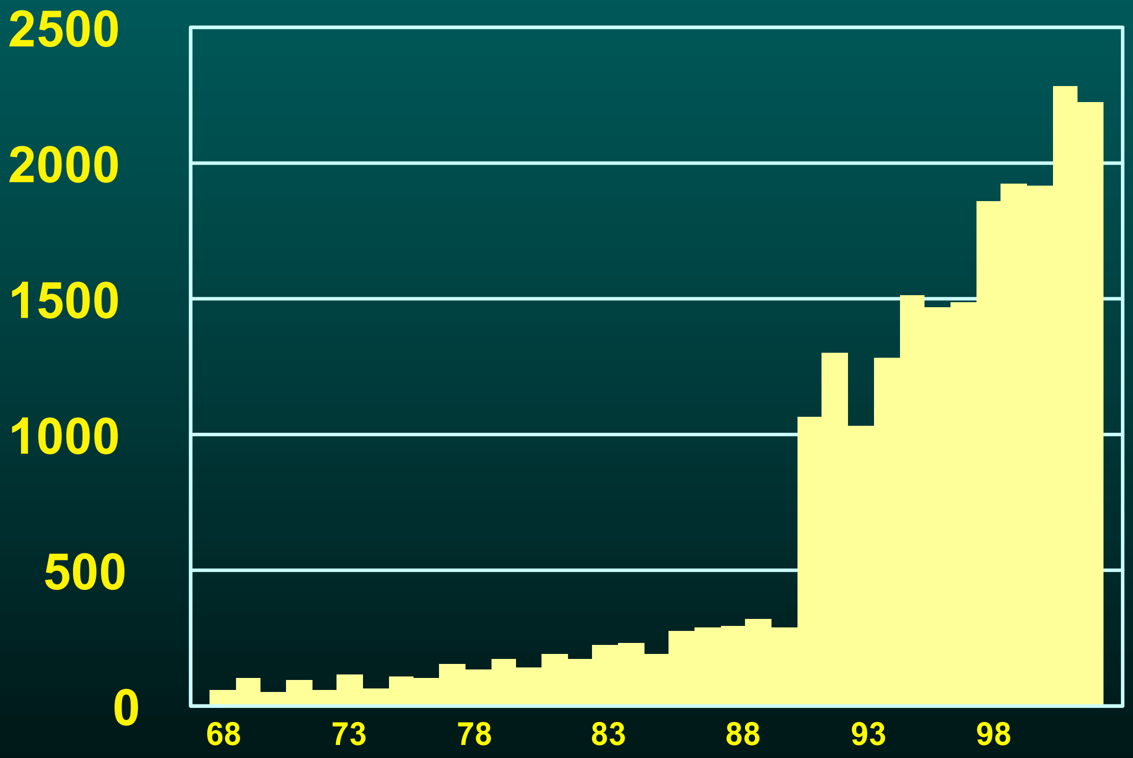
Many different applications



Historical Growth of Synchrotron Publications

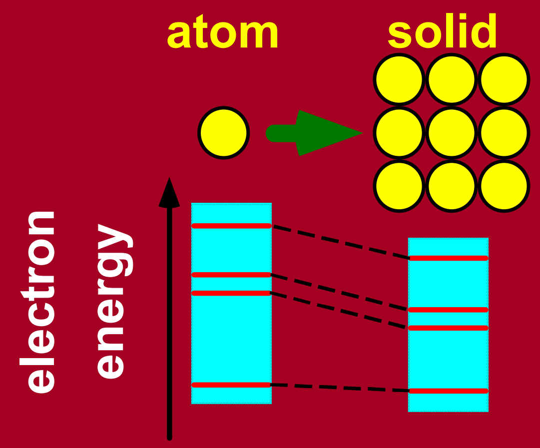
Worldwide ISI data 1968 to 2002

Keyword: “synchrotron”

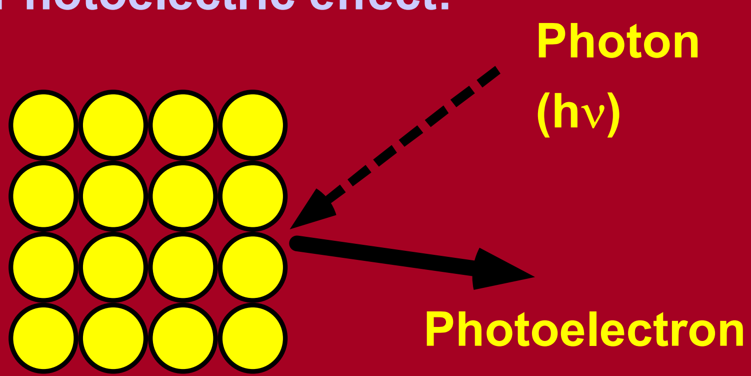


Photoelectron spectroscopy: basic ideas

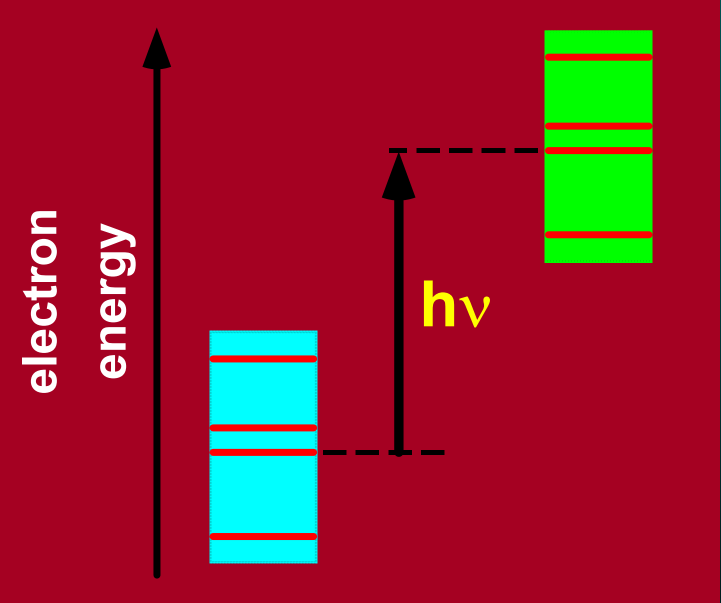
Formation of chemical bonds:



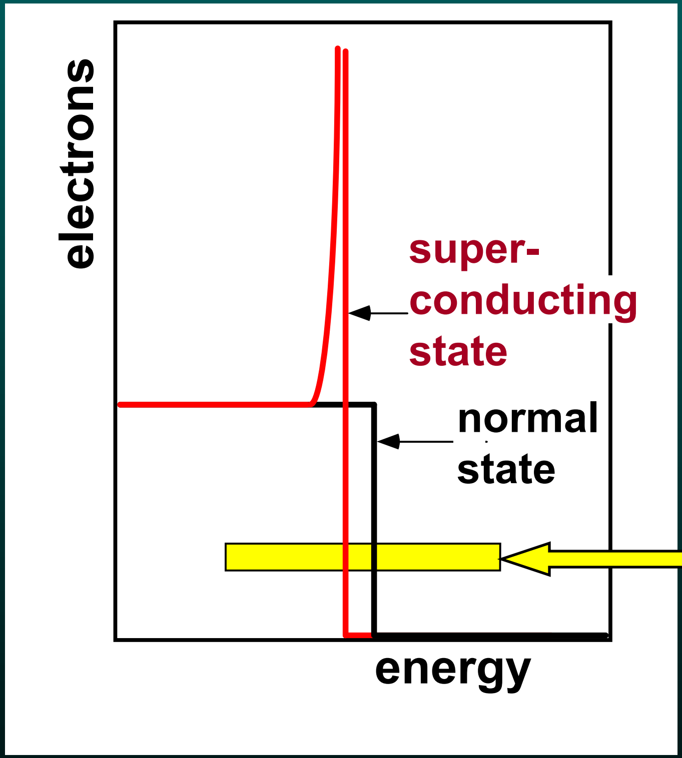
Photoelectric effect:



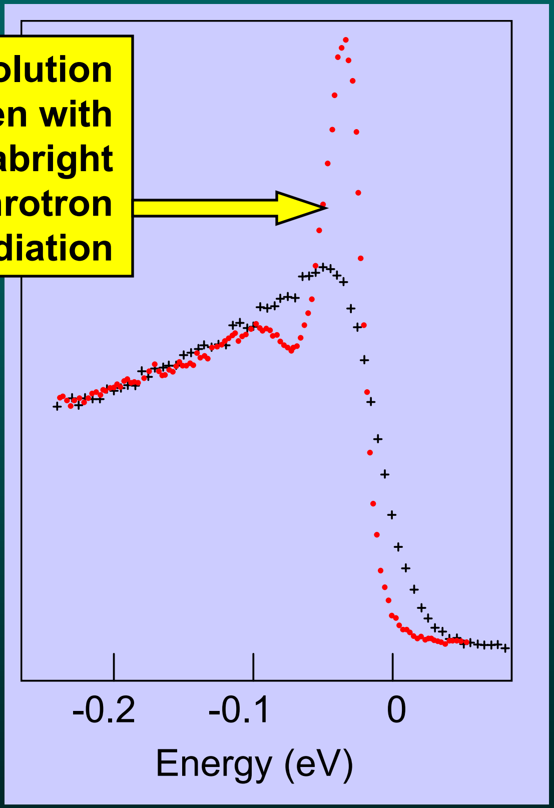
The photon absorption increases the electron energy by hν before ejection of the electron from the solid



Photoelectron spectroscopy of high-temperature superconductivity:

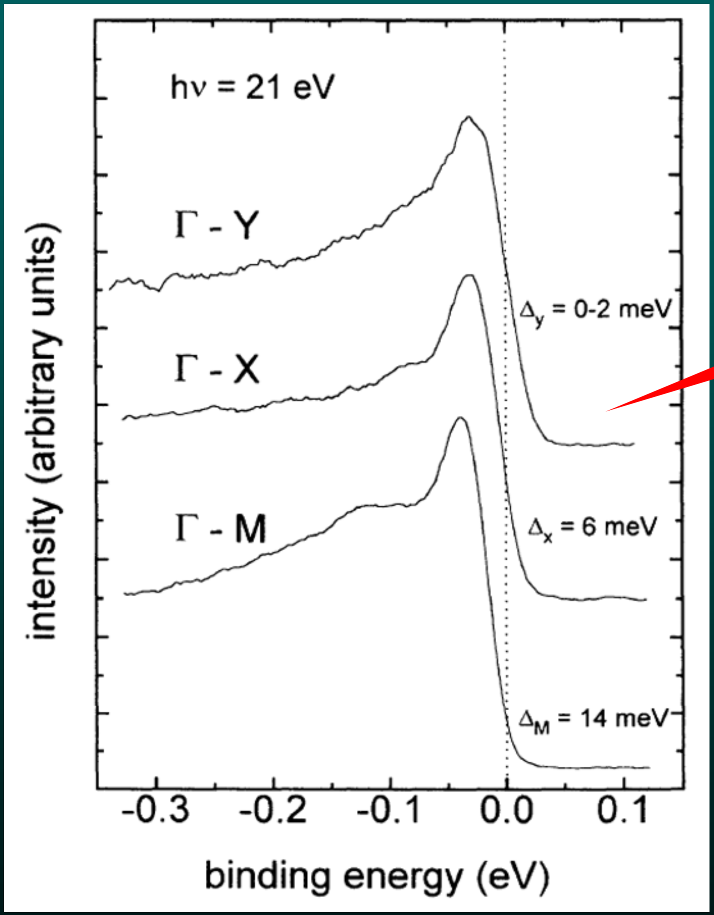


High-resolution spectra taken with ultrabright synchrotron radiation



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

Superconducting gap spectroscopy: Gap anisotropy in BCSCO



Different gaps in different directions (Kelly, Onellion et al.

NO conventional s-waves:

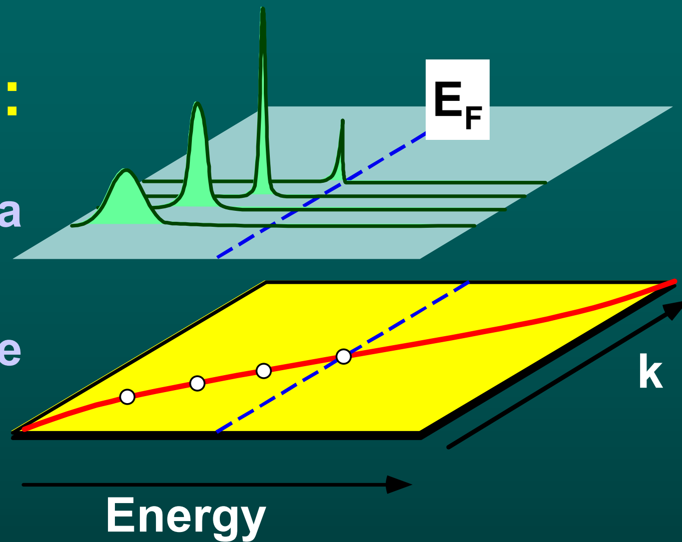
- **d-waves?**
- **Mixed d-symmetries or s+d?**
- **Other symmetries?**

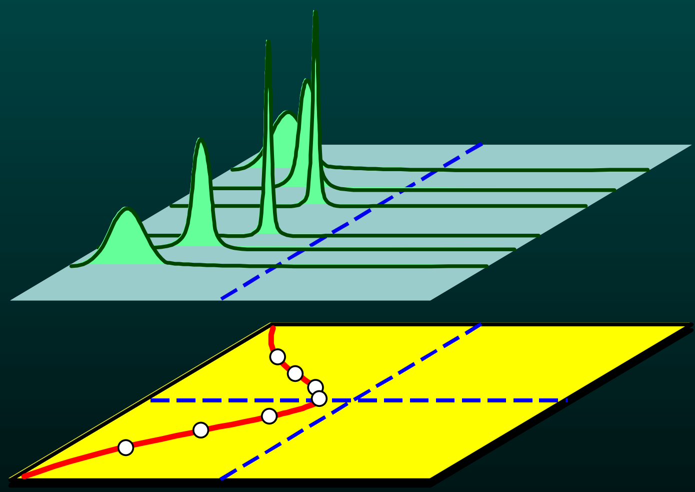
Angle-resolved photoemission: simple picture

In a Fermi-liquid metal:

Photoemission spectra

(quasi-particle) energy vs k curve

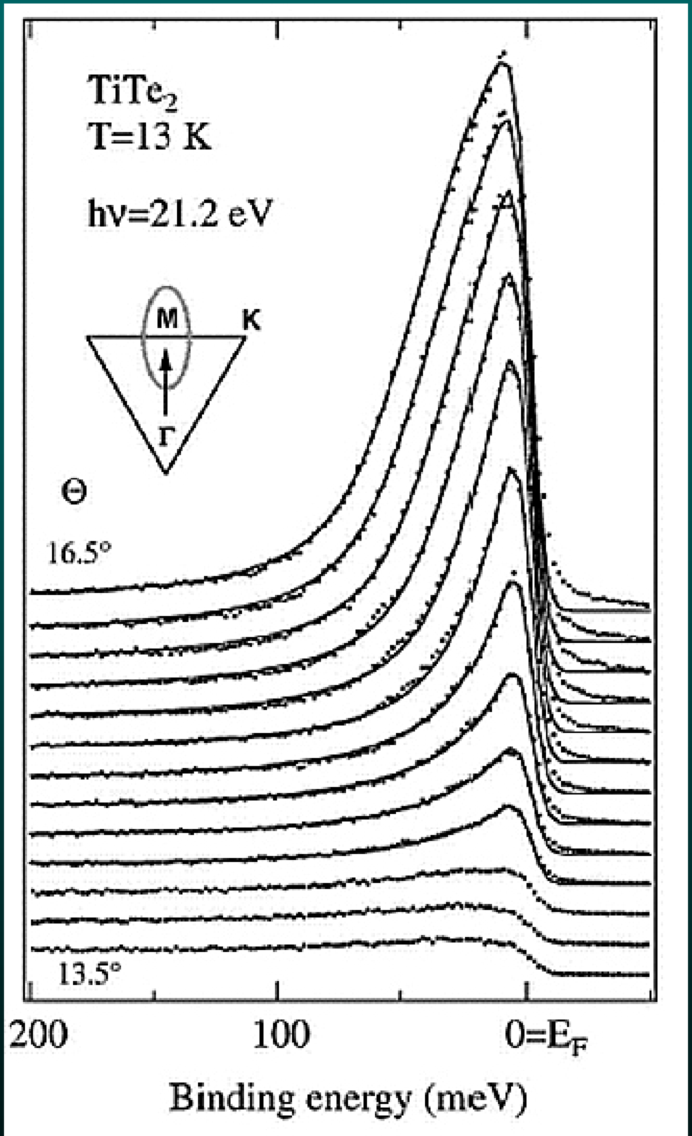


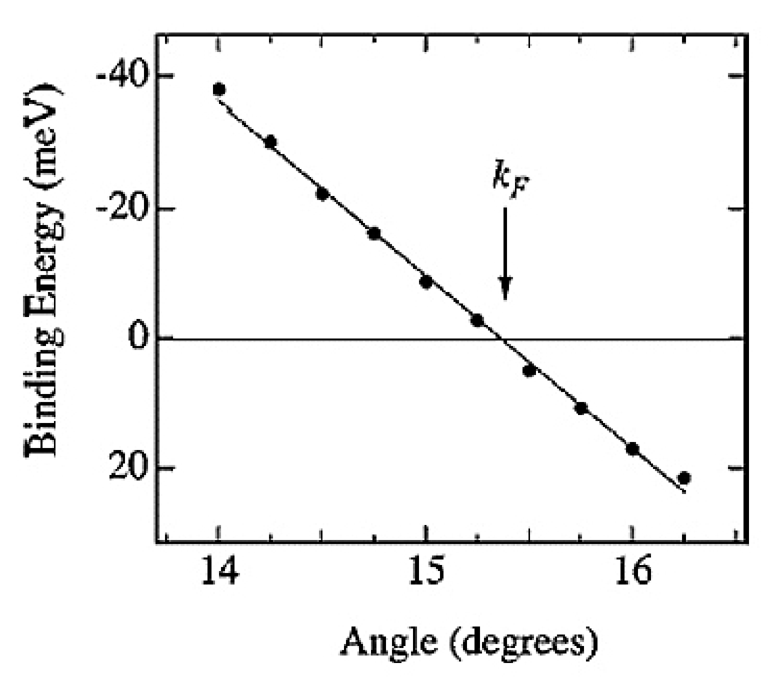


In a “Peierls” insulator (or in an electron-instability insulator in general)

High-resolution angle-resolved photoemission investigation of the quasiparticle scattering processes in a model Fermi liquid: 1T-$TiTe_2$

L. Perfetti, C. Rojas, A. Reginelli, L. Gavioli, H. Berger, G. Margaritondo, M. Grioni, R. Gaál, L. Forró, and F. Rullier Albenque





SCIENCE VOL 290 20 OCTOBER 2000

Electronic Structure of Solids with Competing Periodic Potentials

J. Voit,[1*] L. Perfetti,[2] F. Zwick,[2] H. Berger,[2] G. Margaritondo,[2] G. Grüner,[3] H. Höchst,[4] M. Grioni[2*]

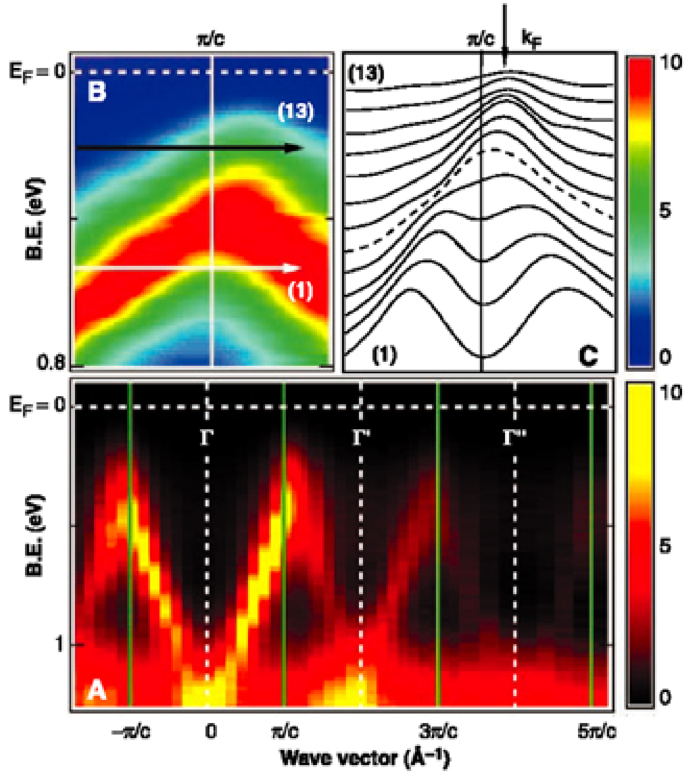
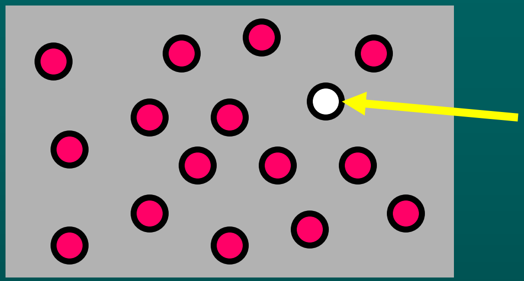


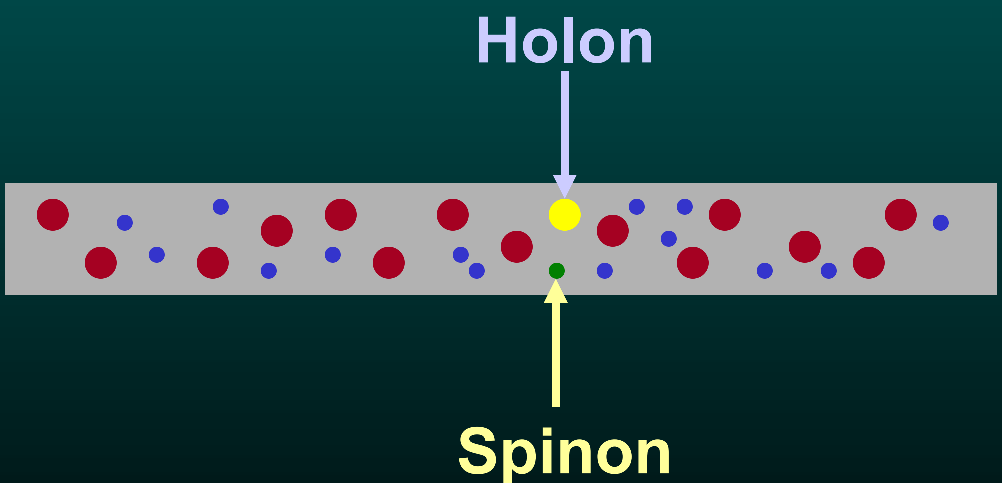
Fig. 3. (**A**) ARPES intensity map [light $h\nu = 21.2$ eV (h is Planck's constant and ν is photon frequency); $T = 300$ K] of $(TaSe_4)_2I$ along the 1D chain direction. B.E., binding energy. (**B**) Detailed view of the region near the zone boundary. For clarity, the raw spectra have been normalized to the same peak intensity and interpolated with respect to energy and wave vector. (**C**) Constant-energy cuts through the intensity map shown in (B), taken at equal energy intervals between the lines marked 1 and 13. The dashed line is a cut at the minimum peak binding energy $E = E^*$.

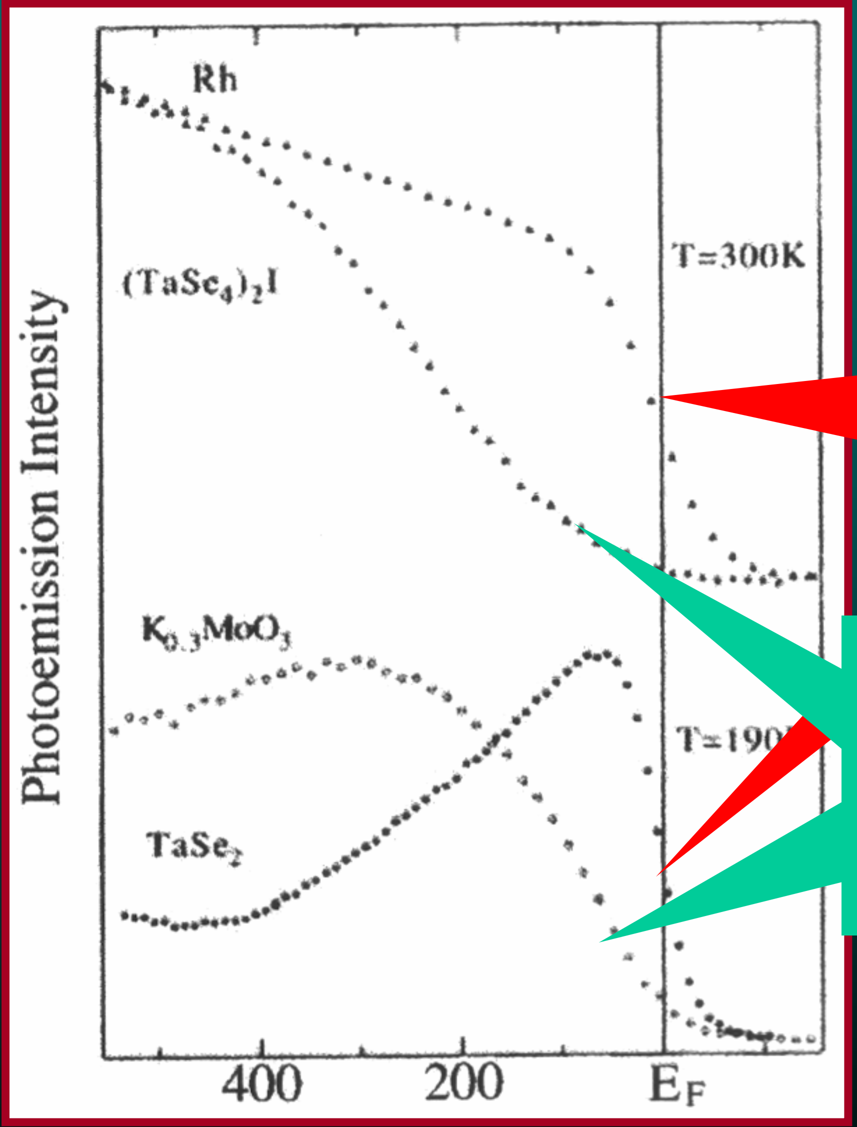
Breakdown of the "Fermi liquid":



The elementary excitation in a Fermi liquid is a "hole" (empty electron state), a quasiparticle with charge +e and spin 1/2

But this is not always true: when observed with high-resolution photoemission, excitations in one-dimensional solids sometimes behave as separated electron-"charge" quasiparticles (holons) and "spin" quasiparticles (spinons)





Fermi Edge in Fermi-liquid Metals

Non-Fermi-liquid Metals: No Fermi Edge

From spectroscopy to spectromicroscopy:

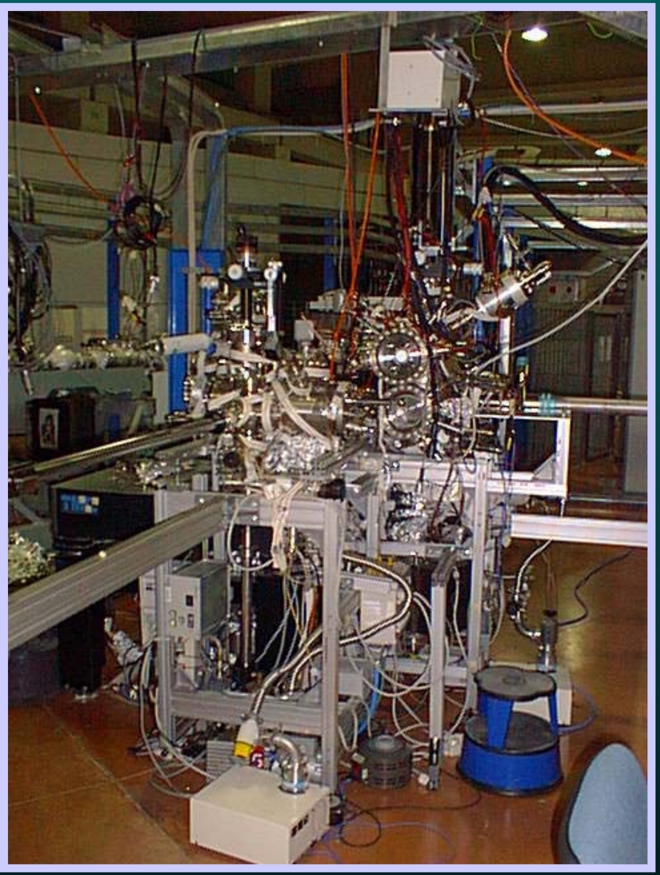
Spectroscopy (energy and momentum resolution)

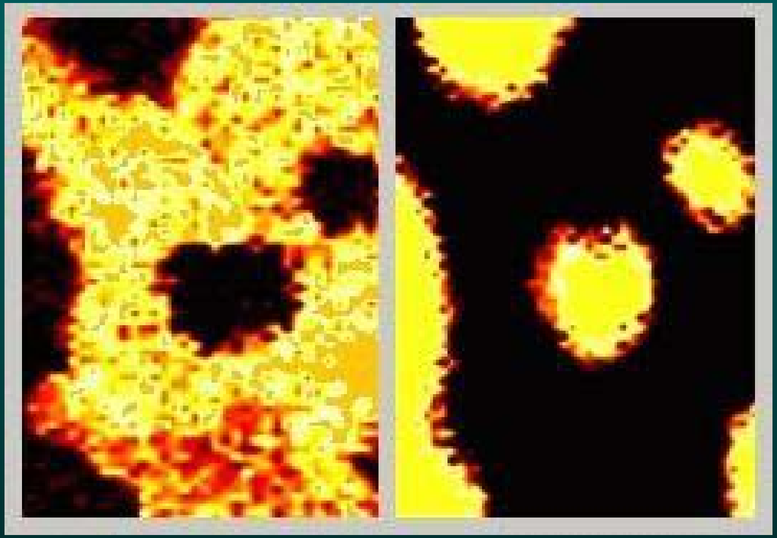
Chemical information

Microscopy (spatial resolution)

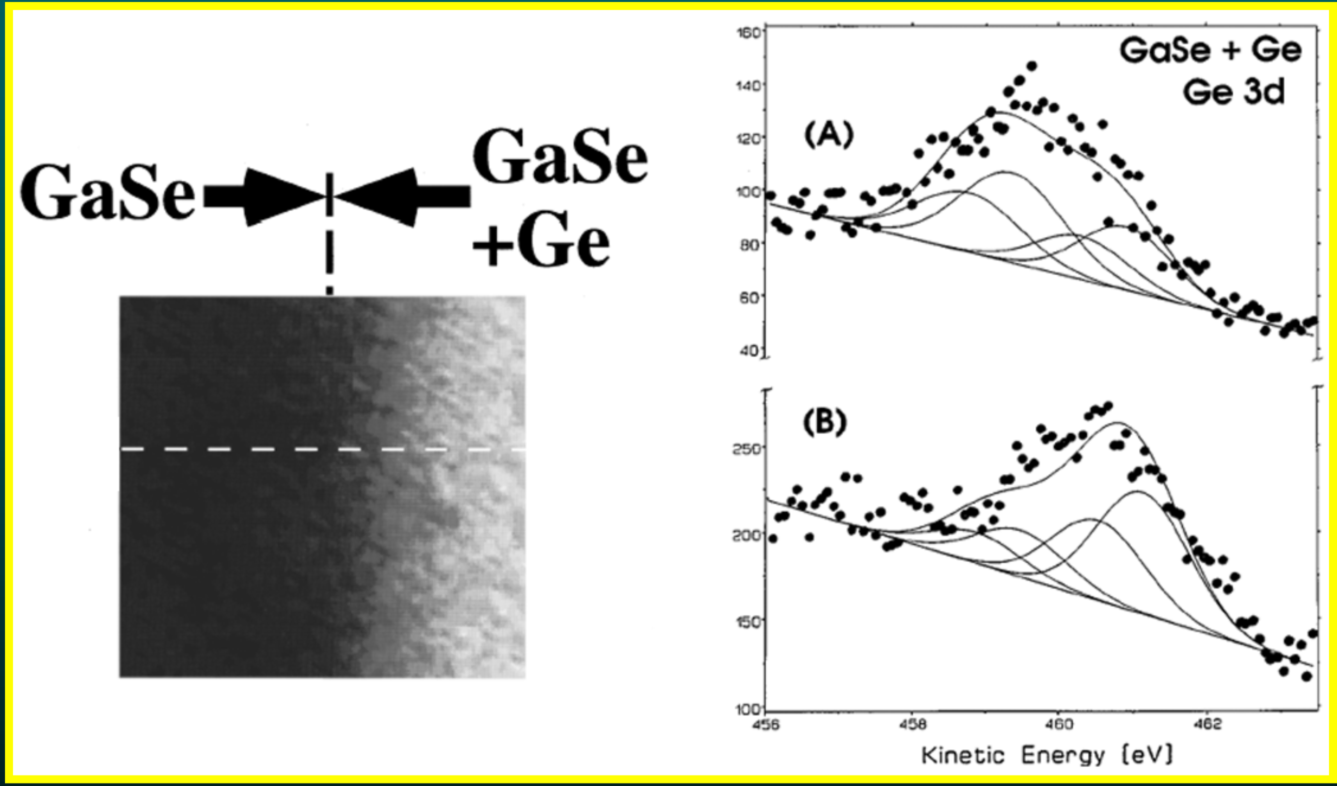
Spectromicroscopy

The ESCA Microscopy Beamline at ELETTRA



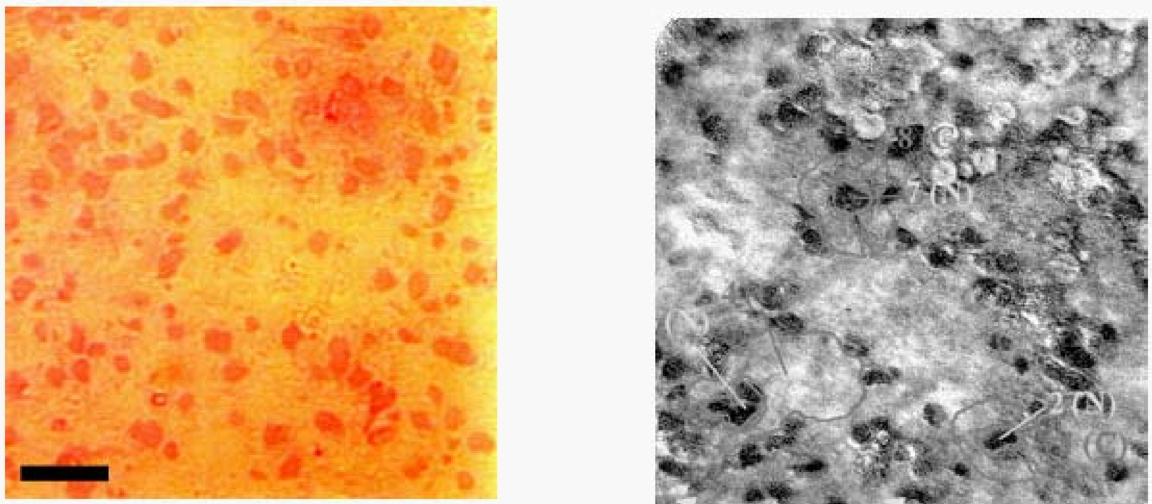


Inhomogeneous chemical reactions at the “unreactive” (!) Gase-Ge interface (J. Almeida et al.)



Photoelectron spectromicroscopy (on untreated specimens) beats optical microscopy + staining in revealing cell nuclei

(B. Gilbert , M. Neumann , S. Steen , D. Gabel , R. Andres, P. Perfetti, G. Margaritondo and Gelsomina De Stasio)

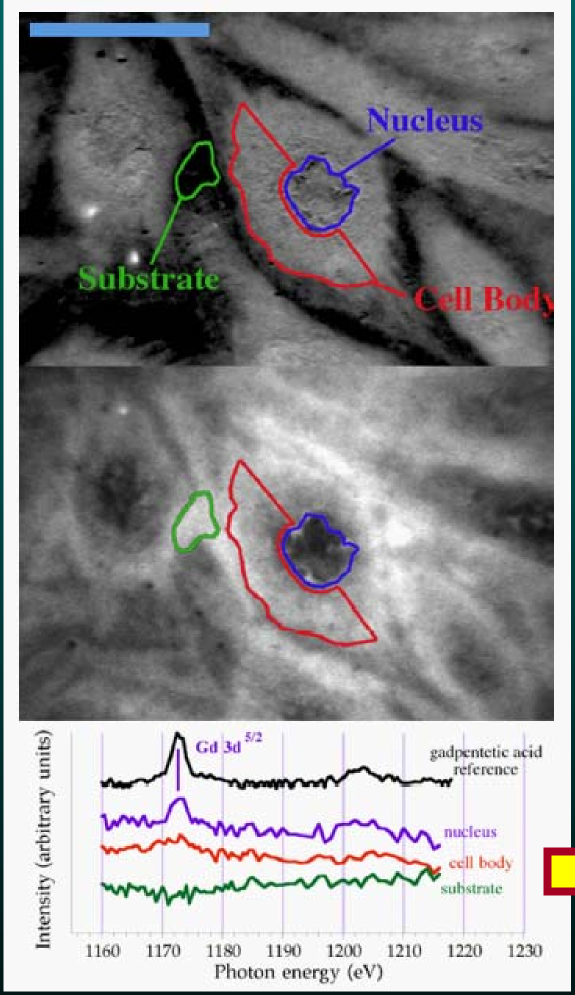


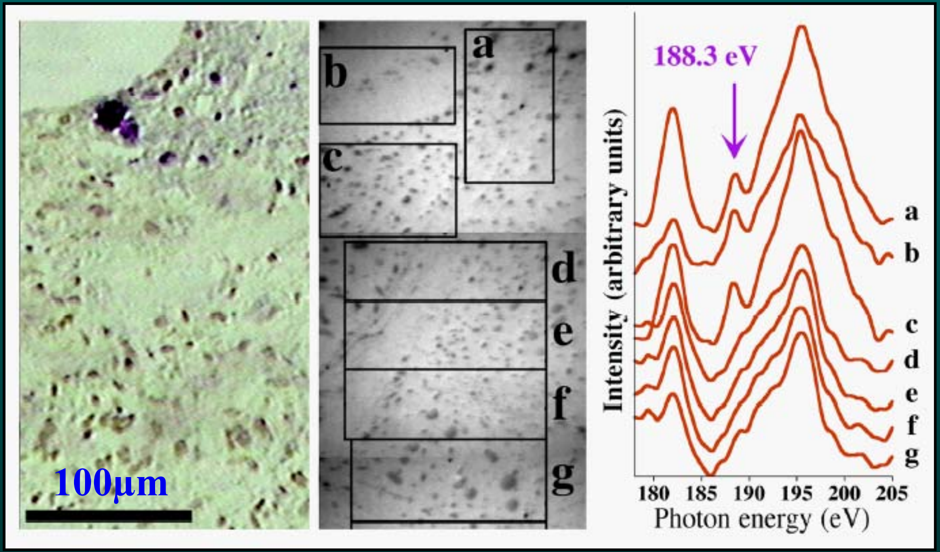
20μm

The distribution of nuclei in human glioblastoma tissue, revealed (left) by staining for optical microscopy and (right) by a MEPHISTO phosphorus map on ashed tissue (phosphorus is shown dark). The MEPHISTO section on gold had no treatment other than ashing. The imaged areas are in adjacent tissue sections, so the exact pattern of nuclei distributions is not identical.

Photoelectron spectromicroscopy explores fine chemical details in boron uptake in cells, in preparation for neutron cancer therapy

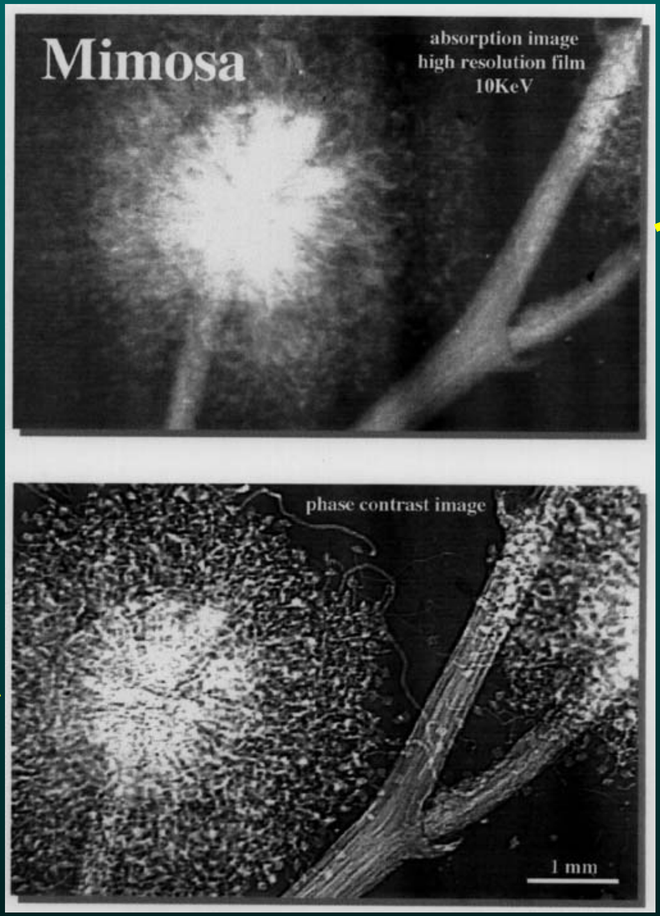
(B. Gilbert , Gelsomina De Stasio et al.)





Gadolinium may work better!

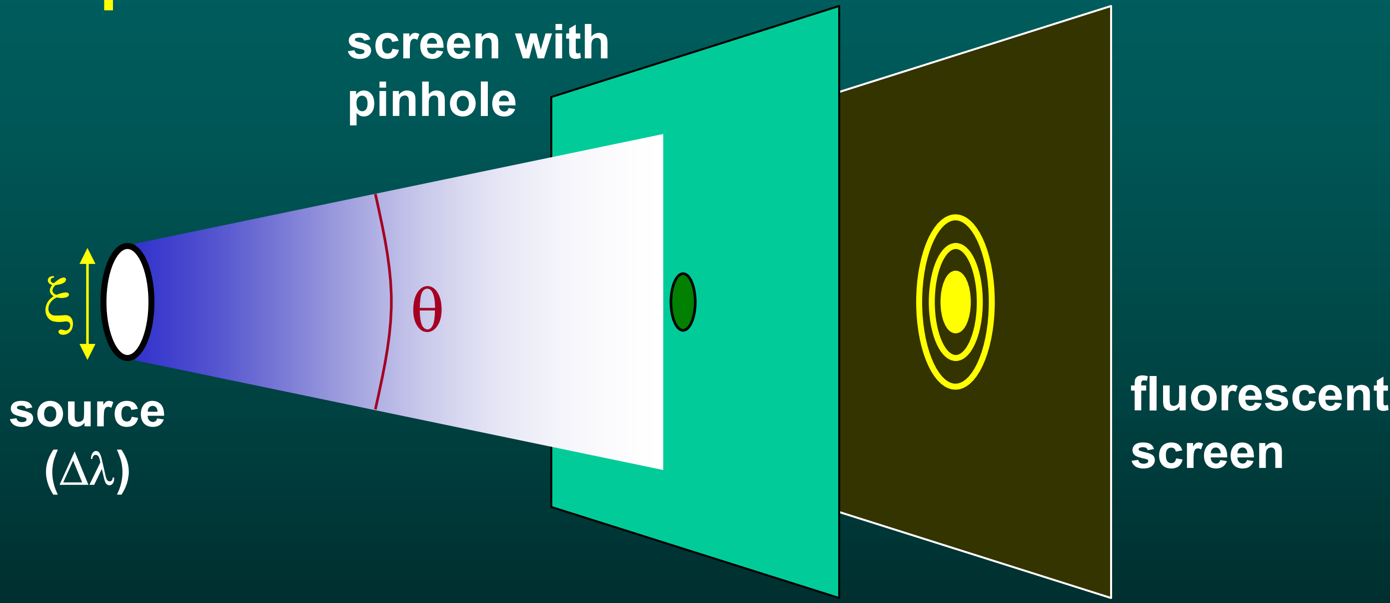
Conventional radiology



Refractive-index radiology

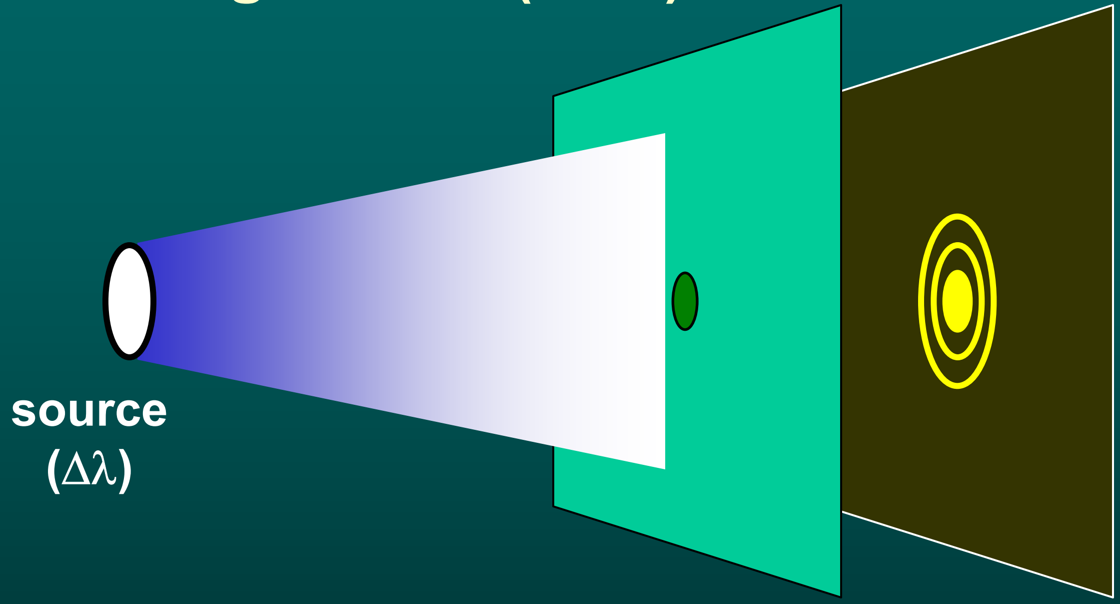
Coherence: "the property that enables a wave to produce visible diffraction and interference effects"

Example:



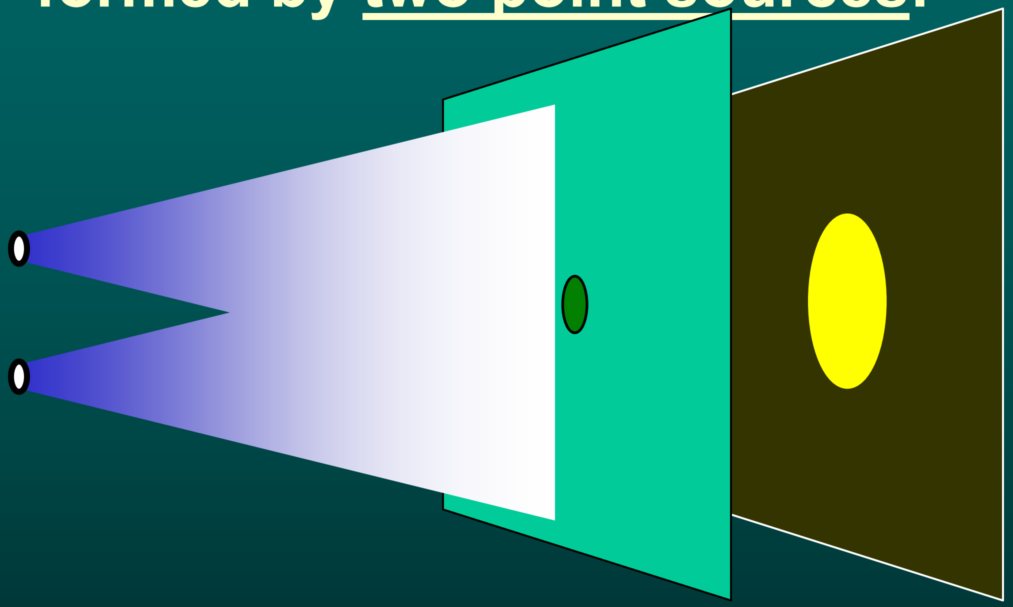
The diffraction pattern may or may not be visible on the fluorescent screen depending on the source size ξ, on its angular divergence θ and on its wavelength bandwidth $\Delta\lambda$

Longitudinal (time) coherence:



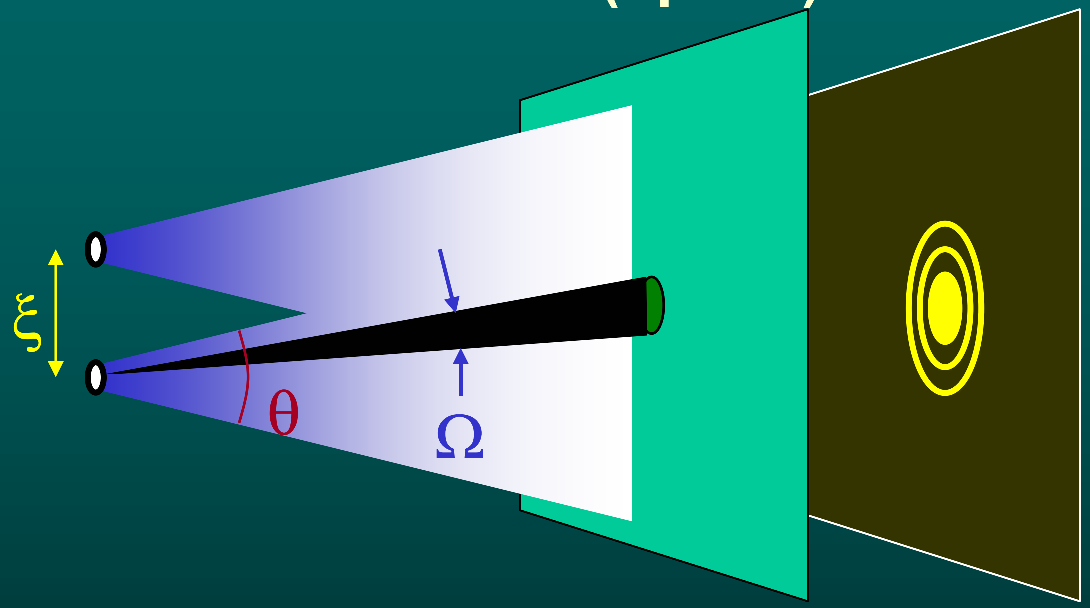
- Condition to see the pattern: $\Delta\lambda/\lambda < 1$
- Parameter characterizing the longitudinal coherence: "coherence length": $L_c = \lambda^2/\Delta\lambda$
- Condition of longitudinal coherence: $L_c > \lambda$

Lateral (space) coherence — analyzed with a source formed by <u>two point sources</u>:



- Two point sources produce overlapping patterns: diffraction effects are no longer visible.
- However, if the two source are close to each other an overall diffraction pattern may still be visible: the condition is to have a **large "coherent power" $(2\lambda/\xi\theta)^2$**

Conditions for lateral (space) coherence:

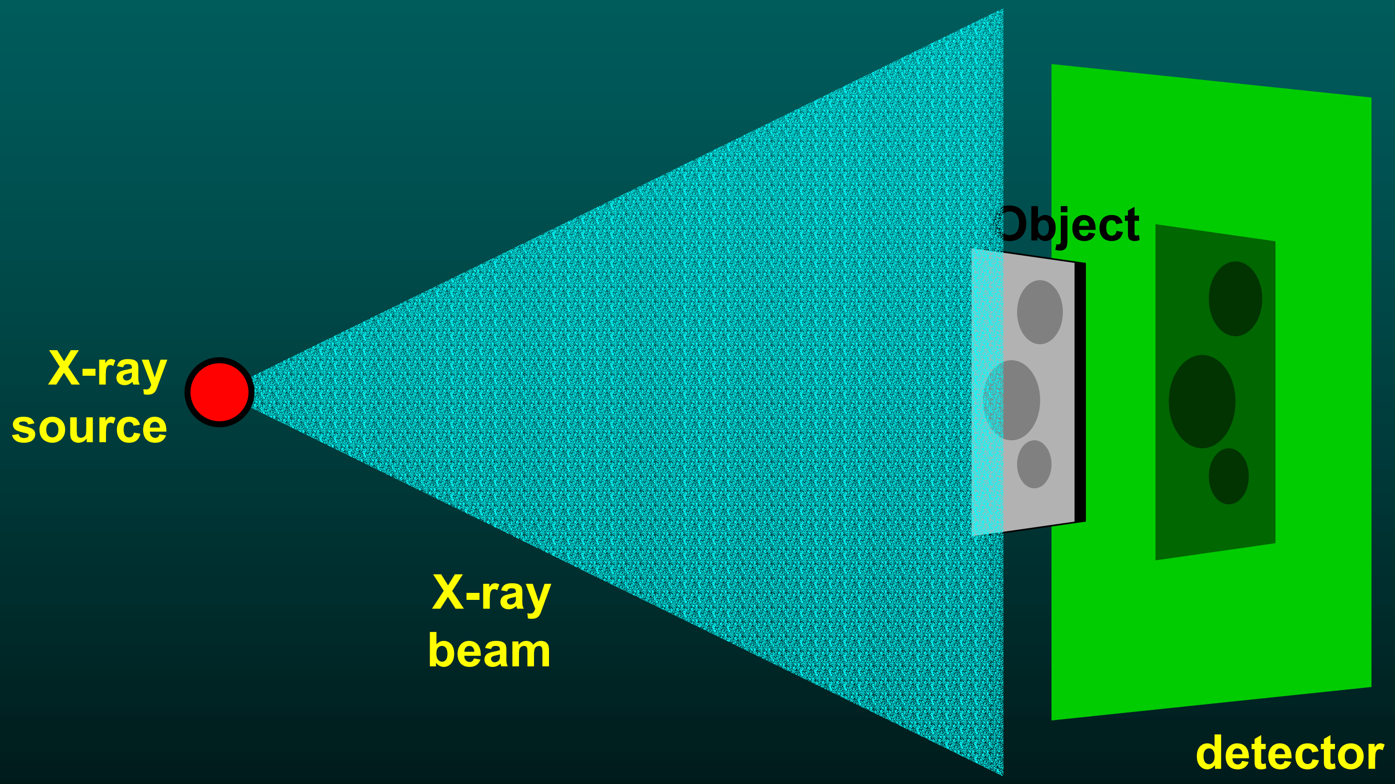


- Condition to see the pattern: $\xi\Omega < 2\lambda$
- If the emission occur over θ, only a fraction $(\Omega/\theta)^2 < (2\lambda/\xi\theta)^2$ produces diffraction. This defines the (lateral) coherent power: $(2\lambda/\xi\Omega)^2$
- Full (lateral) coherence — diffraction limit: $\xi\Omega = 2\lambda$

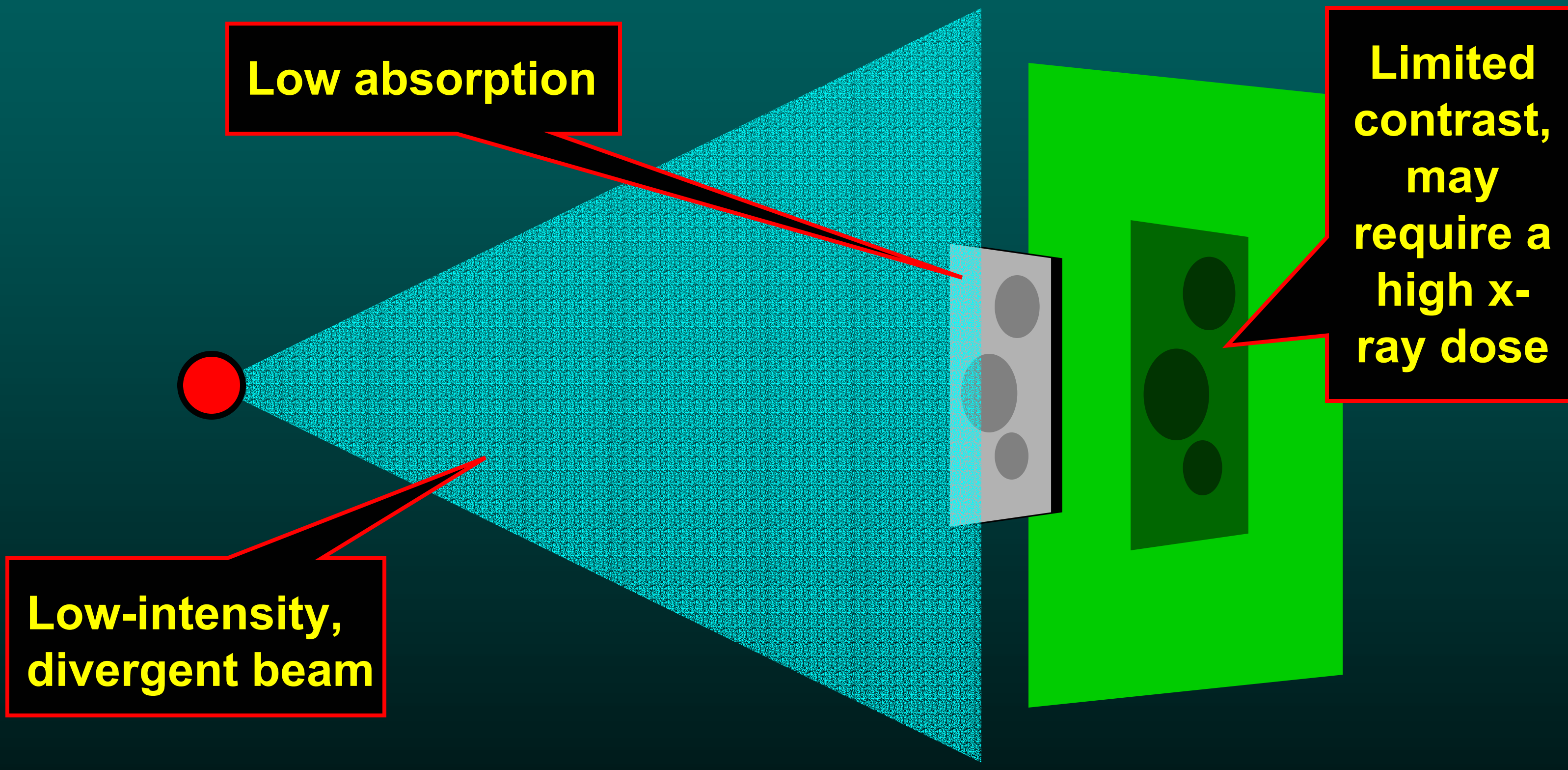
Coherence — summary:

- Large coherence length $L_c = \lambda^2/\Delta\lambda$
- Large coherent power $(2\lambda/\xi\theta)^2$
- **Both difficult to achieve for small wavelengths (x-rays)**
- **The conditions for large coherent power are equivalent to the geometric conditions for high brightness**

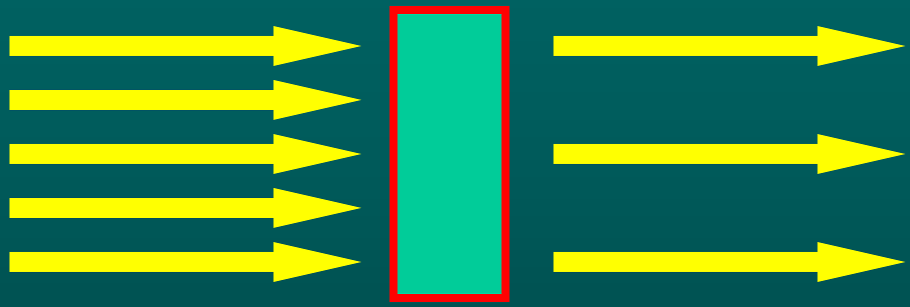
Conventional (Absorption) Radiology:



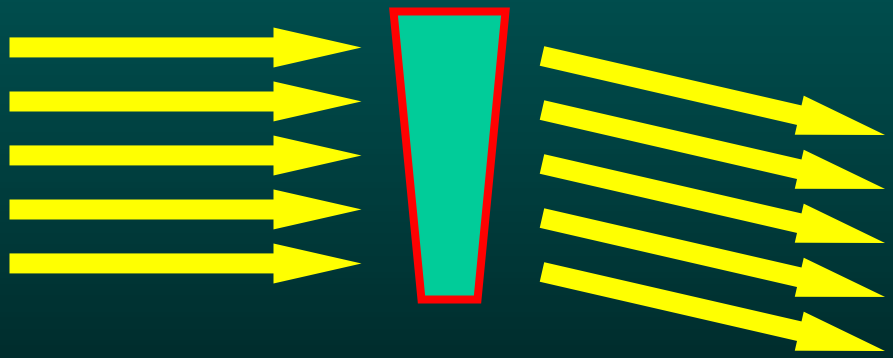
Some Problems in Conventional Radiology:



Light-matter Interactions:



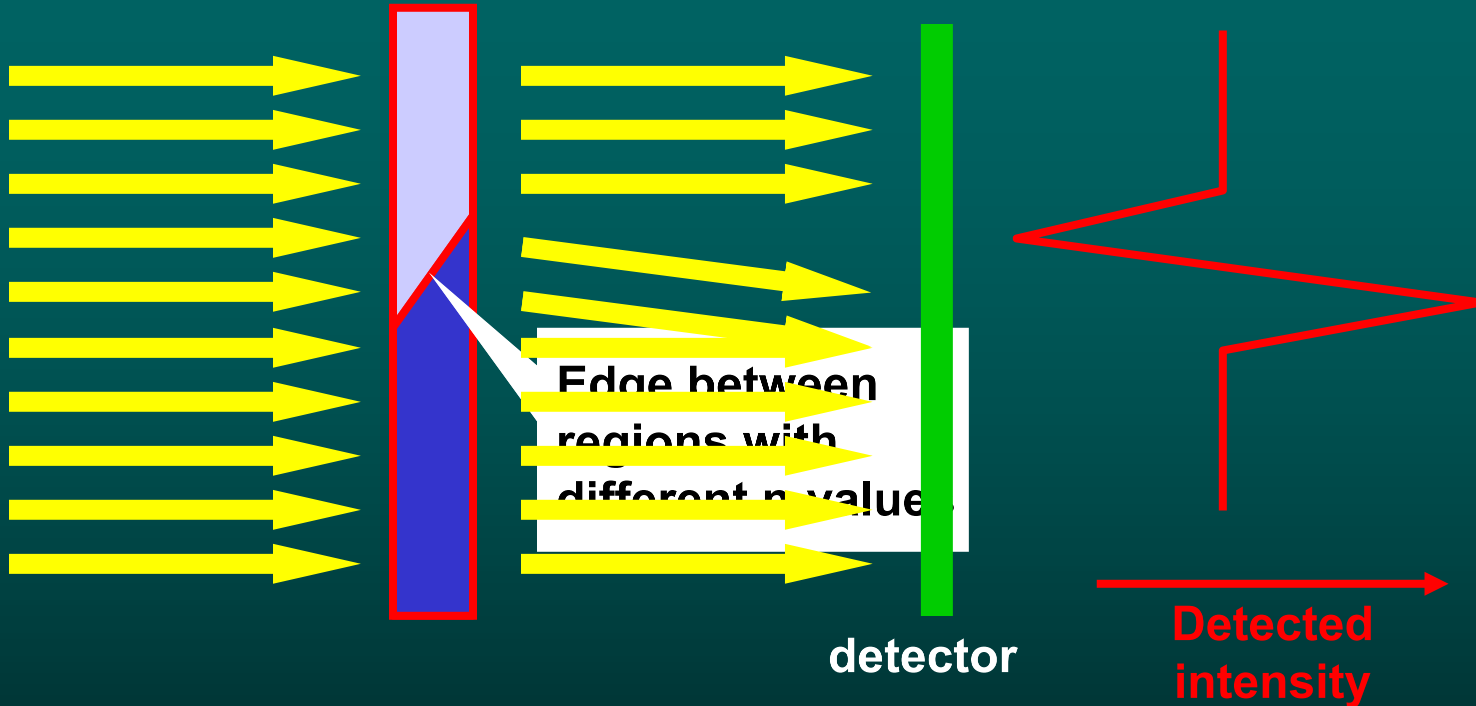
Absorption -- described by the absorption coefficient α

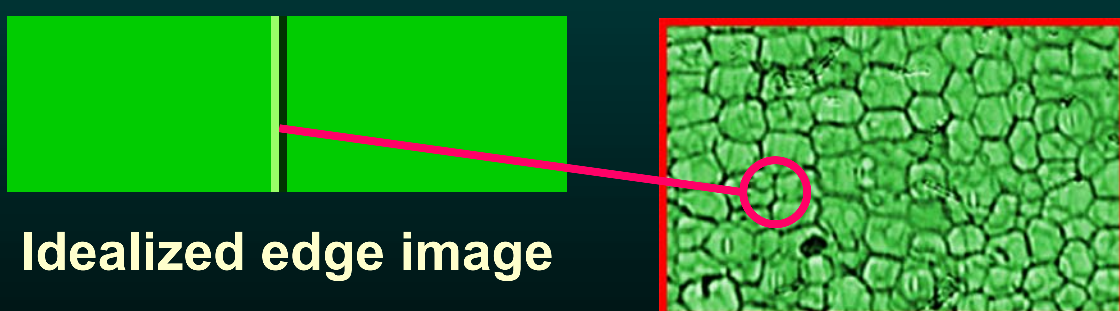


Refraction (and diffraction/interference) -- described by the refractive index n

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

“Refraction” x-ray imaging:





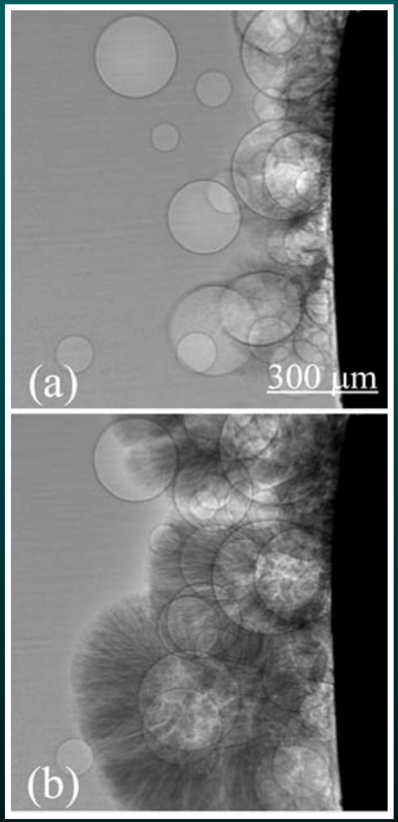
Idealized edge image

Real example (leaf)

"Refraction" x-ray imaging -- potential advantages over absorption:

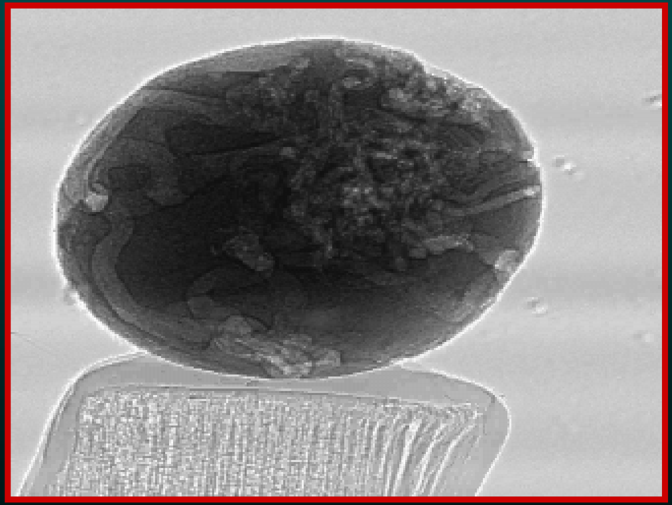
- Differences between object and vacuum: small in both cases, but larger for n than for α
- This advantages increases as the wavelength decreases
- Better edge visibility, better contrast, smaller dose

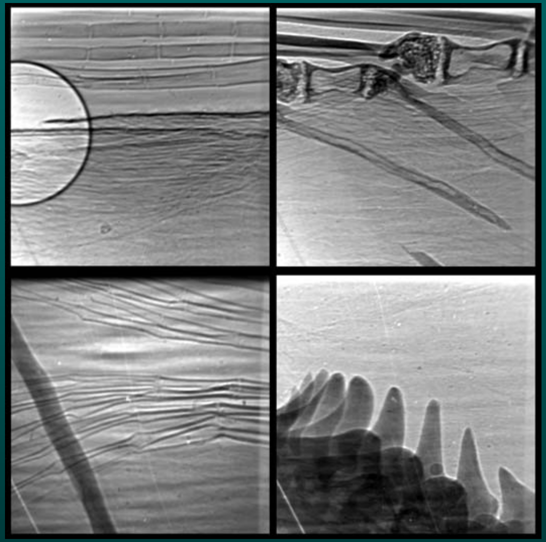
Examples of "refraction" radiology:



We can show that one can build on bubbles …

… study a fossil embryo...



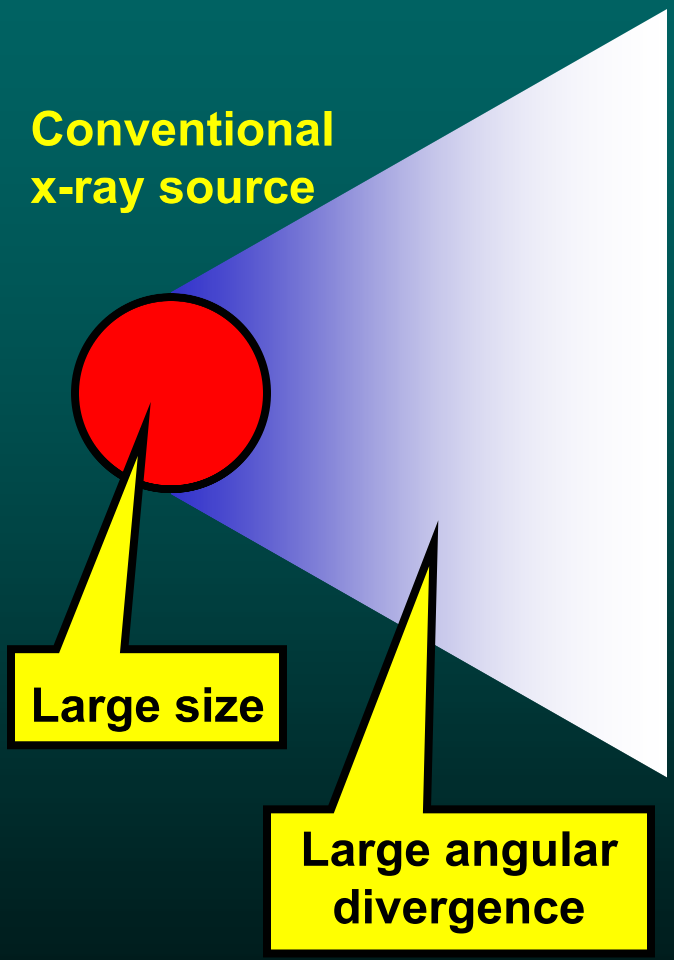


… or take x-rays images of a 0.5 mm <u>live</u> microfish

Building on bubbles:

QuickTime™ et un décompresseur
GIF sont requis pour visualiser
cette image.

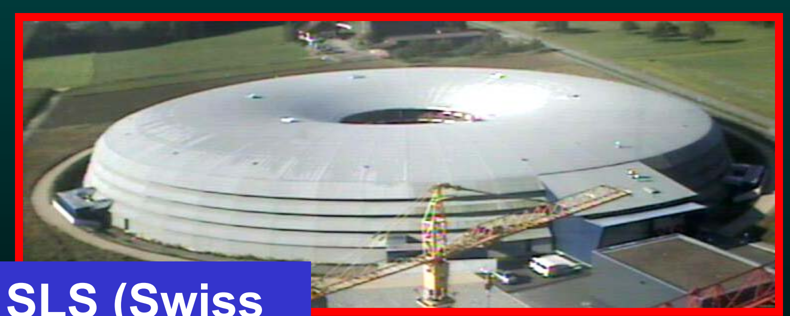
“Refraction” radiology -- the problems



Required, instead:

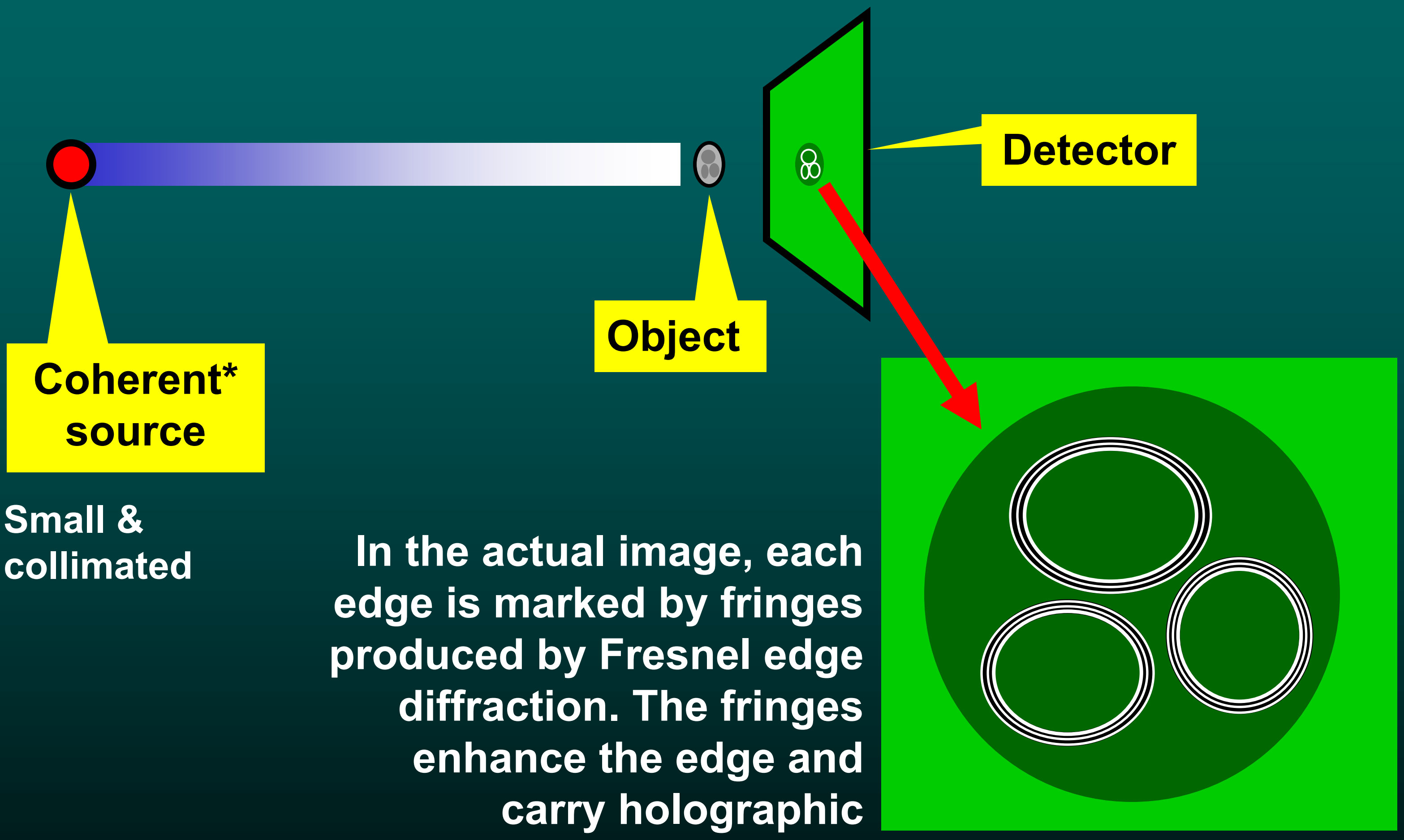
Solution:

synchrotron sources



SLS (Swiss Light Source)

A bit more sophisticated description



* Small & collimated

In the actual image, each edge is marked by fringes produced by Fresnel edge diffraction. The fringes enhance the edge and carry holographic information

Our own results:

- Analytical modeling and validation tests. Main results: interplay between refraction and diffraction regimes; limited longitudinal coherence needed
- Numerical modeling
- First tests on live animals (PAL-Korea and SRRC-Taiwan)
- Materials science experiments (Argonne, PAL, SRRC, Elettra): electrodeposition, fracture)
- First tests on anomalous-scattering

PhysicsWorld

Double take makes the most of X-rays to enhance synchrotron images

Physics in Action: March 1998

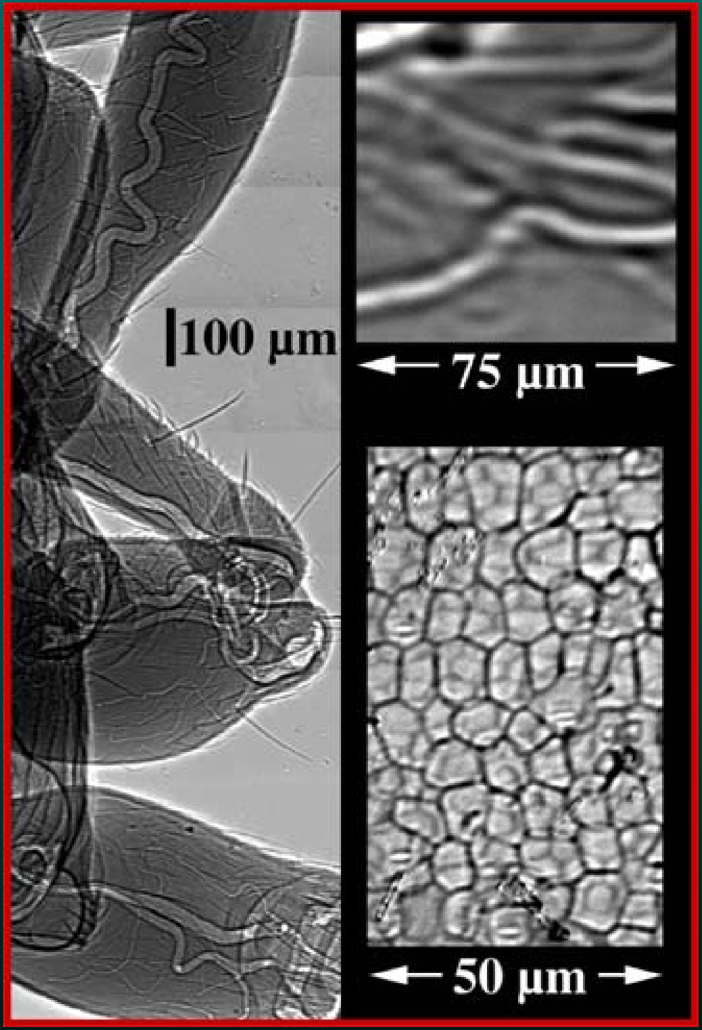
Is there something basically wrong with present-day radiology techniques? The answer would seem to be yes, on the basis of recent results from the National Synchrotron Light Source in the US.

The results show that the limited quality of conventional X-ray images can be dramatically improved by exploiting synchrotron light (D Chapman et al. 1997 *Phys. Med. Biol*. 42 2015). Moreover, there is no need for a high radiation dose.

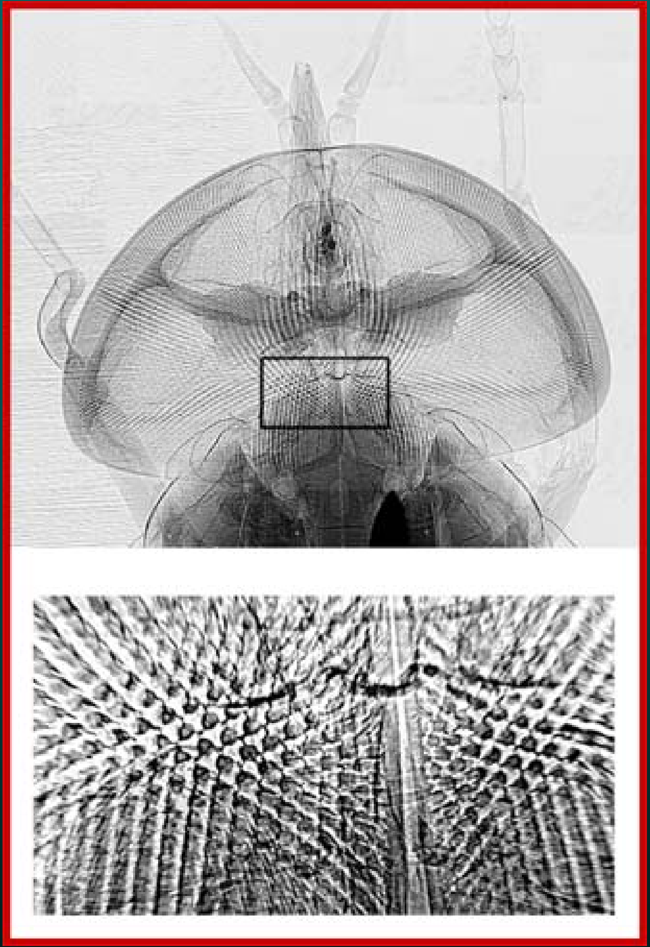
Ineffective radiology has a big impact on society. Relatively high X-ray doses can overcome the limitations of conventional techniques, but they act as a deterrent for the mass screening of killer diseases like breast cancer. This is regrettable, since screening allows early detection and, in most cases, very successful therapy.

Author
Giorgio Margaritondo is from the Ecole Polytechnique Fédérale de Lausanne, Switzerland, and at Sincrotrone, Trieste, Italy.

Our own results:

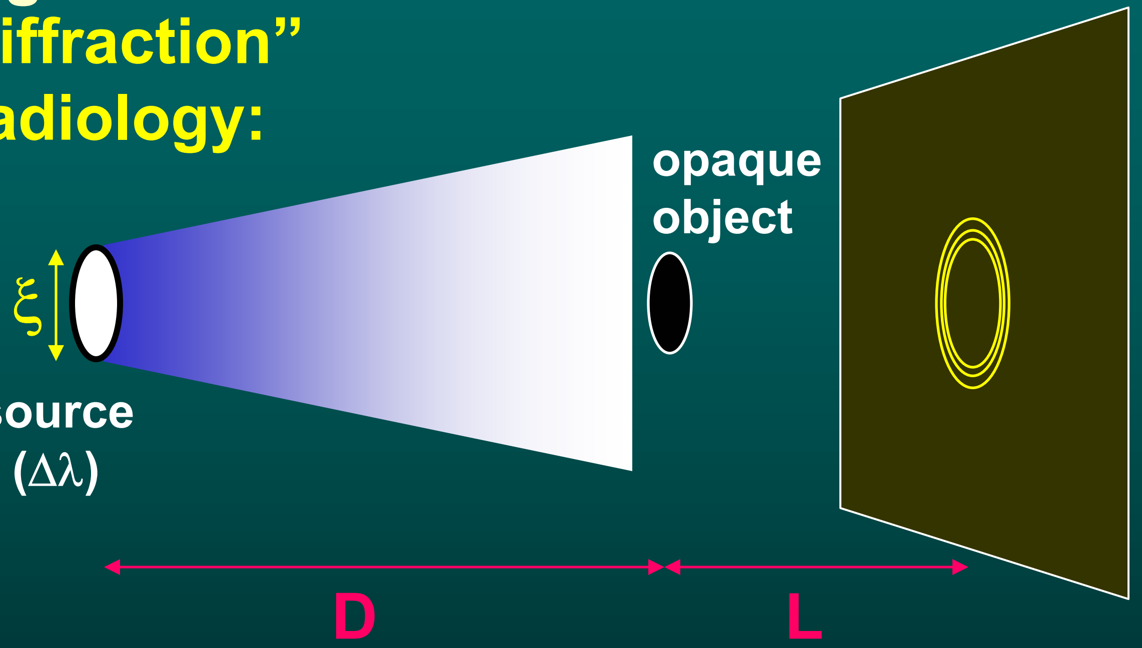


Our own results:



Modeling: conditions for coherence-based radiology

“Diffraction” radiology:



Conditions to see the edge diffraction fringes:

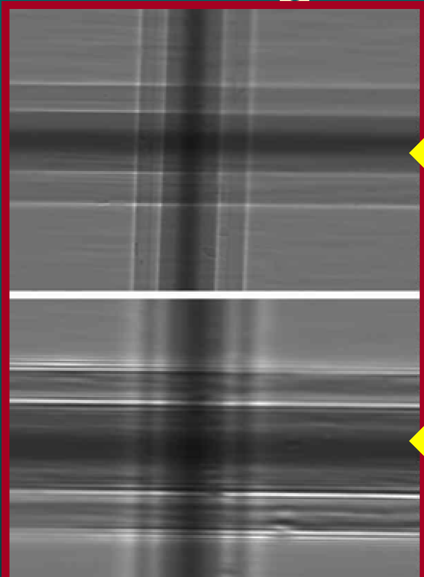
$\xi < 0.8\, D \sqrt{(\lambda/2L)} \approx 100$ micron

$\boxed{\Delta\lambda/\lambda < \sqrt{2}}$ ← **no monochromator!!**

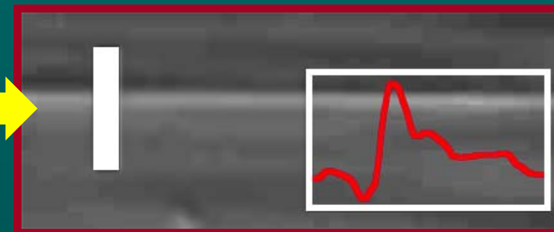
Equivalent condition for “refraction” radiology:

$\xi/D < \theta$

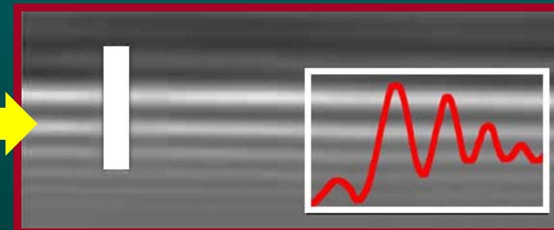
Modeling: interplay of "refraction" and "diffraction"

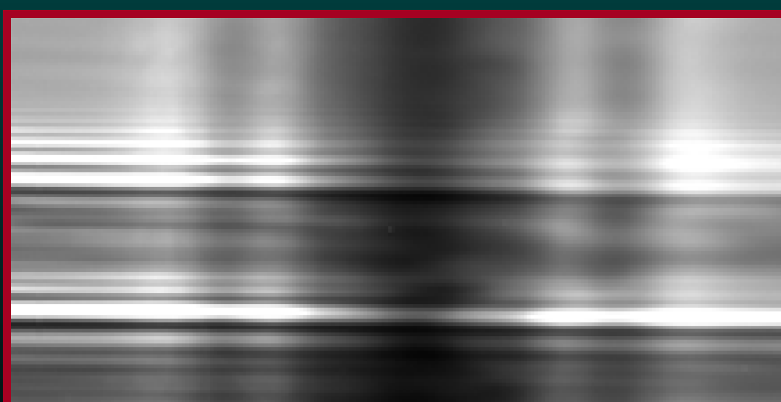


Refraction radiographs



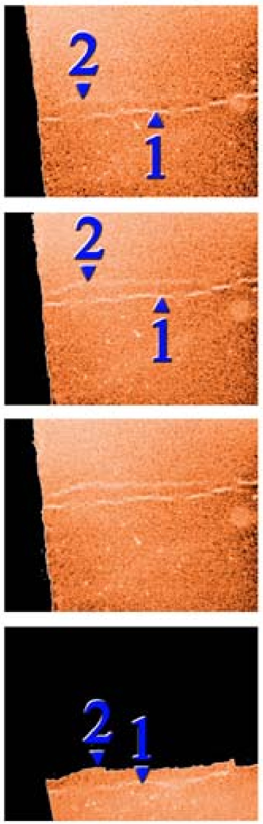
Diffraction radiographs





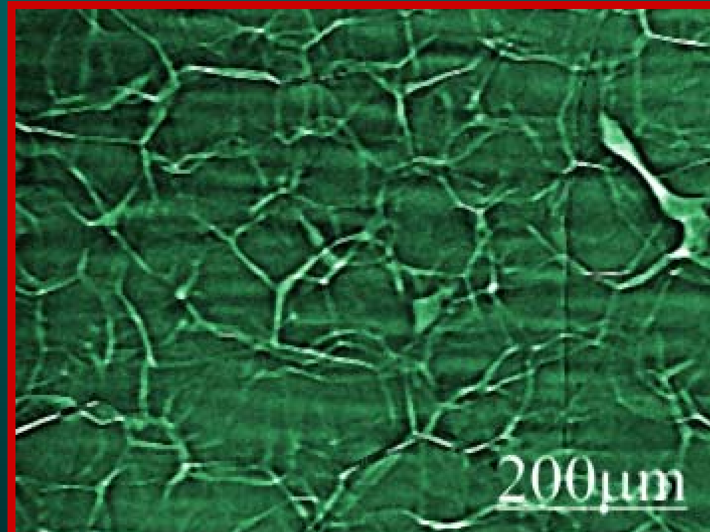
Note: with bending-magnet emission, the effects are only in the vertical direction (no space coherence in the horizontal direction)

Our own results -- materials science:



Time-resolved collimation-enhanced micro-radiology study of the breaking of a Cu foil (Y. Hwu et al.)

Note the development of two defect lines (1) and (2)

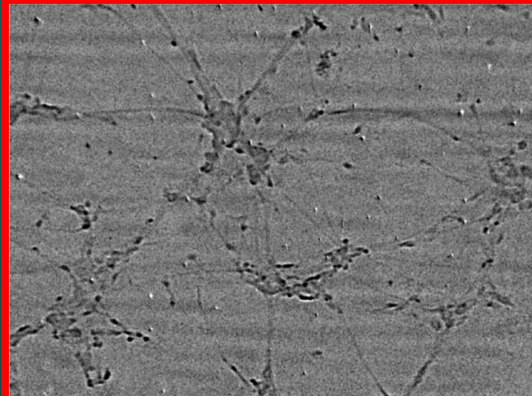


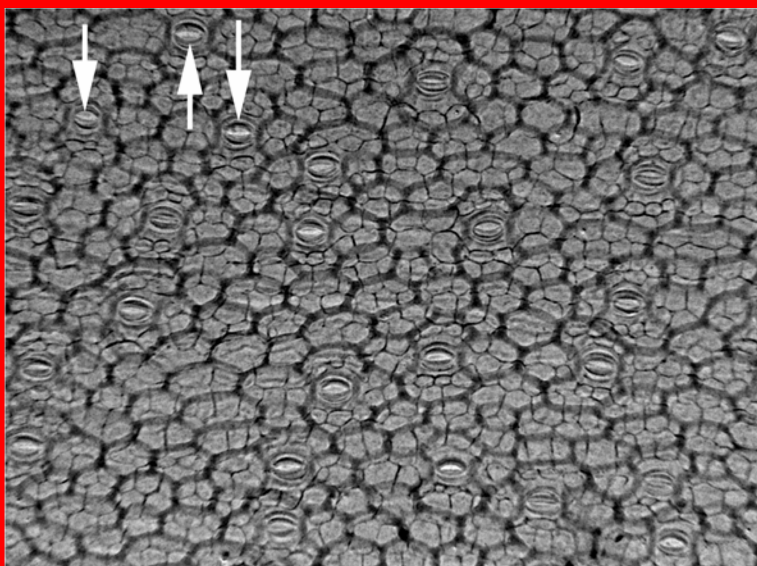
Imaging grain boundaries without any decoration

Radiography of individual cells

Opening of a “stoma”

Neurons

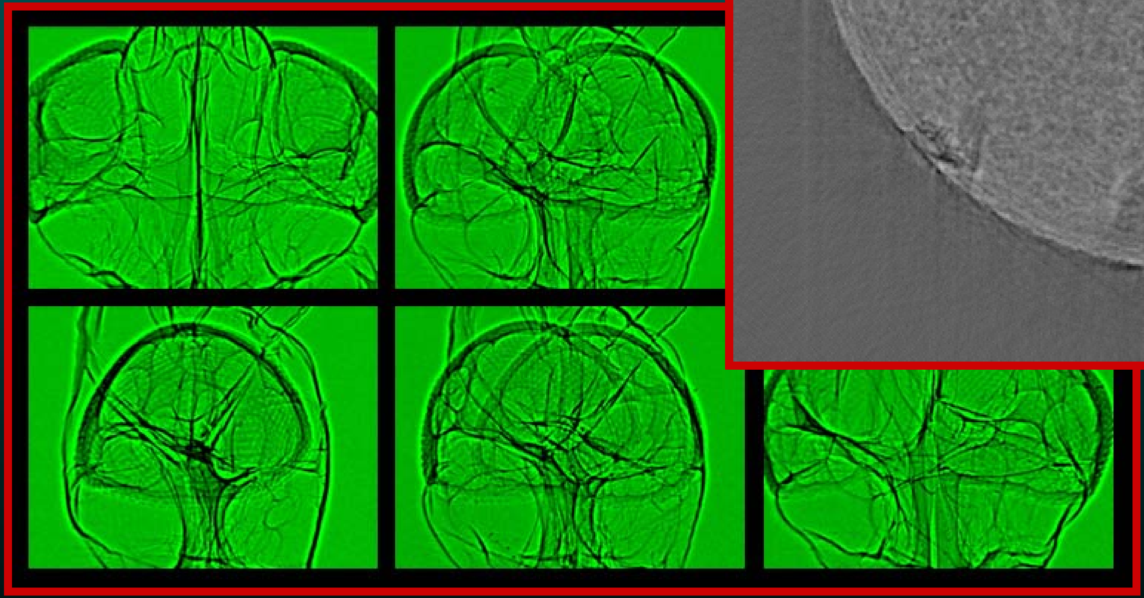


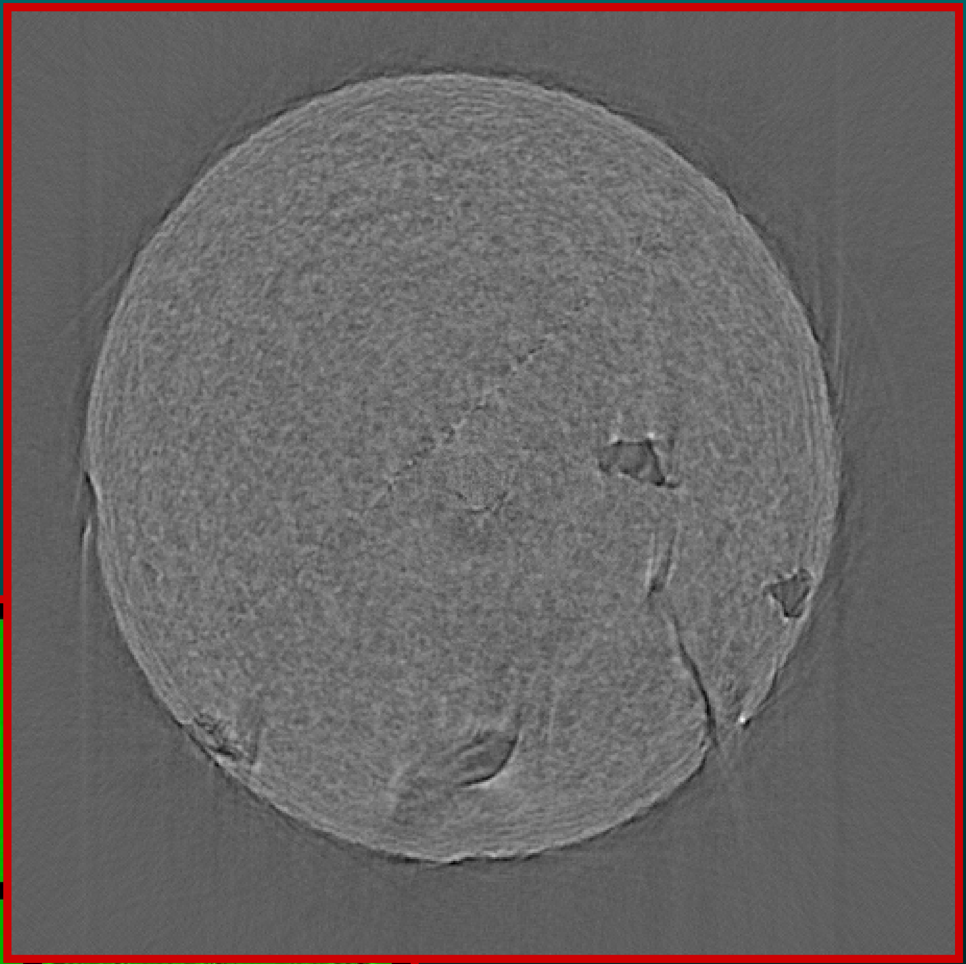


Leaf

QuickTime™ et un décompresseur
GIF sont requis pour visualiser
cette image.

Our own results - tomography:





New types of sources:

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

The New European Commission Round Table

For Synchrotron Radiation And Free Electron Lasers (2000-2004)

Proposal Submission Forms

EUROPEAN COMMISSION

6th Framework Programme for Research, Technological Development and Demonstration

Support for research Infrastructures
Integrating Activities/Communication Network Development

Integrated Infrastructure Initiative

A1

Proposal Number[1]		Proposal Acronym[2]	IA-SFS

General Information on the Proposal

Proposal Title[3] (max. 200 char.)	Integrating Activity on Synchrotron and Free Electron Laser Science		
Duration in months[4]	60	*Call (part) identifier[5]*	FP6-2002-Infrastructures-1
Activity code(s) most relevant to your topic[6]	INFRASTR-2.1		
Keyword 1[7]	04.00.00.00.00.00.00		
Keyword 2[7]	03.02.03.23.00.00.00		
Keyword 3[7]	04.02.10.00.00.00.00		
Class of infrastructure[8]	Synchrotrons, Free Electron Lasers		

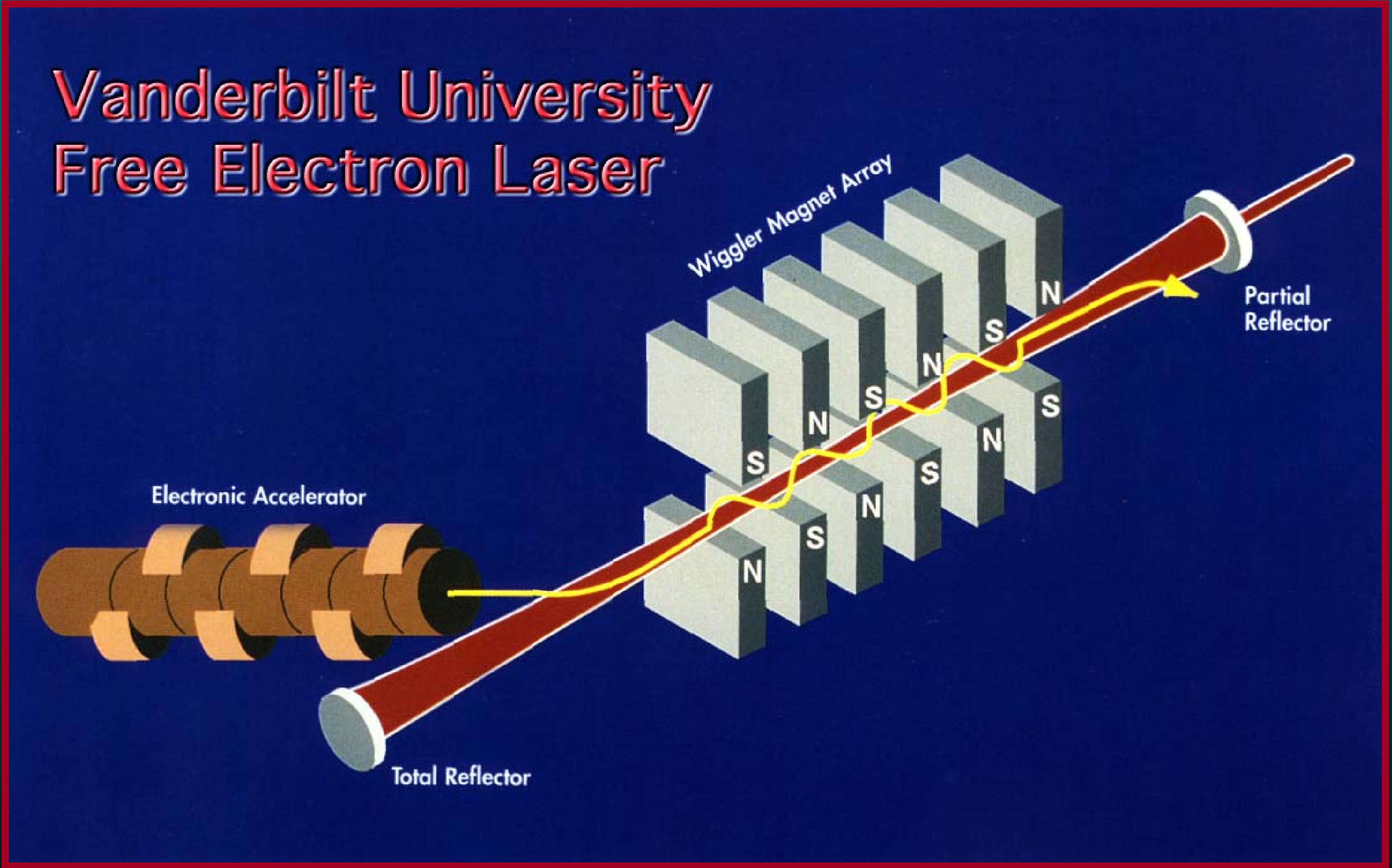
Abstract[9] (max. 2000 char.)

This I3 has two strategic objectives: (1) to support transnational users of national facilities in the domain of synchrotron and FEL science; (2) to support joint research activities (JRA's) with the purpose of (i) enhancing the effectiveness of the facilities in serving users and in particular transnational users, and (ii) contributing to the development of novel sources in this domain. The initiative is based on the very solid record of success of transnational access contracts at individual facilities in the past decade, of the coordinating role of the European Round Table for Synchrotron Radiation and Free Electron Lasers, and of several instrumentation collaboration projects. The key element of the success is that the EC support has practically opened the most advanced network of synchrotron and FEL sources in the world to all qualified European users based on merit. The present I3 will continue and boost this very successful approach. The impact is positive on scientists from all over Europe and in particular on those from countries without this type of facilities. Many new experiments will be made possible fully exploiting the intellectual resources of thousands of scientists. The JRA's target strategic areas of instrumentation with specific emphasis on electron guns and experimental tools for future x-ray lasers.

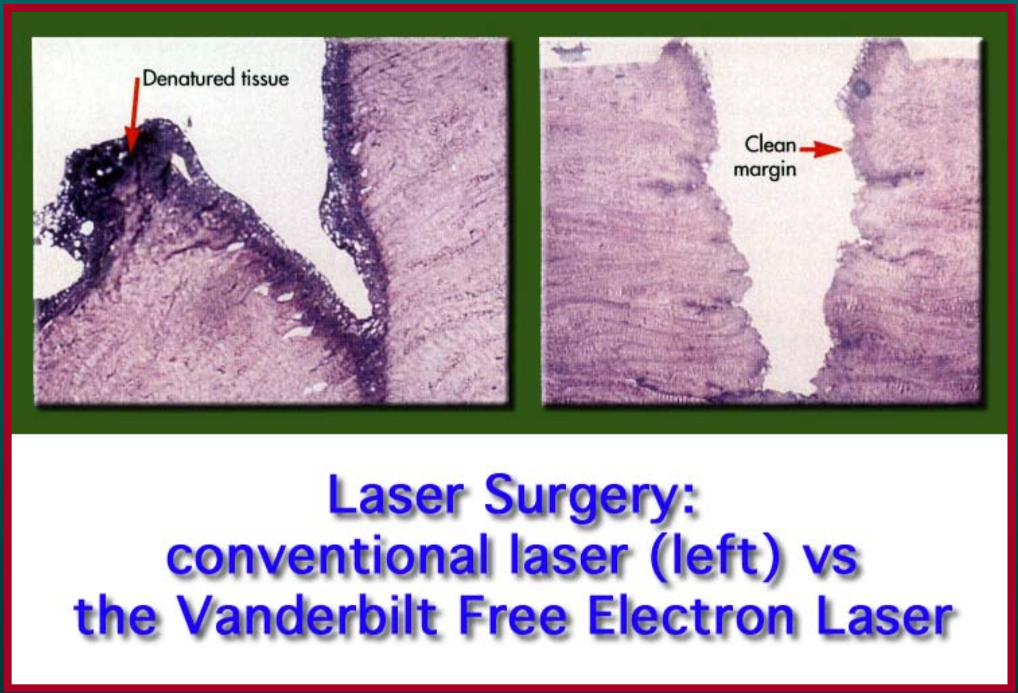
The access and research initiatives are complemented and enhanced by targeted networking activities. The I3 will continue the Round Table support for specialized workshops, meetings and schools with the objectives to (i) stimulate new ideas for transnational collaboration, and (ii) prepare new generations of users. The I3 will also support sabbatical exchanges between facilites and user institutions.

To appreciate the dimension of this initiative, note that it involves ESRF and all major national facilities in Europe. The national facilities alone provide over 200 experimental stations and will expand this patrimony. The I3 facility network puts Europe at the forefront of this strategic area of science and technology and the I3 will enable Europe to fully exploit it beyond national barriers.

Free-electron lasers:

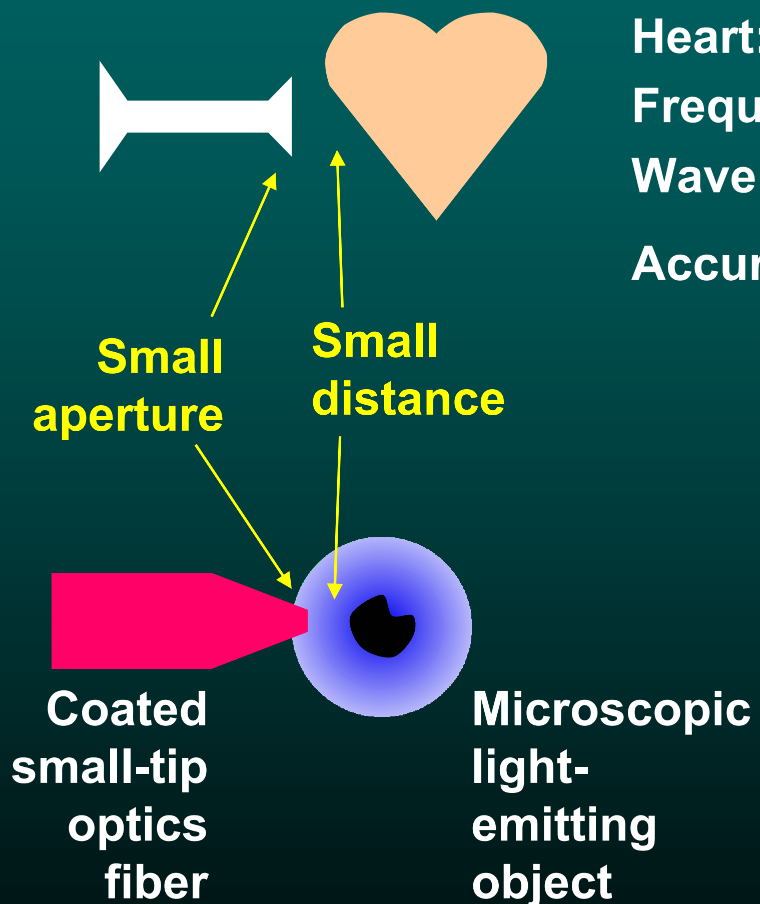


Free-electron laser surgery:



Wavelength selection → much less collateral damage:

The scanning near-field optical microscope (SNOM): like the stethoscope



Heart:

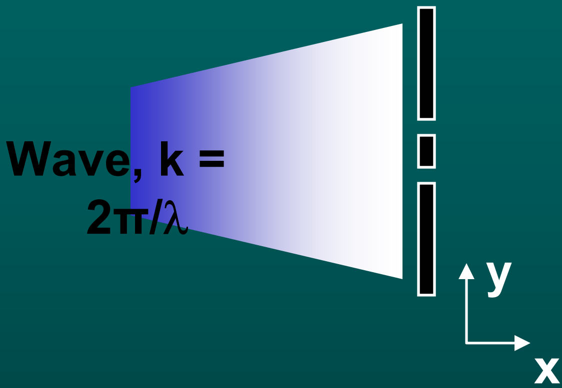
Frequency ≈ 30-100 Hz

Wavelength λ ≈ 102 m

Accuracy in localization ≈ 10 cm ≈ λ /1000

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

SNOM: why does it work? Consider two slits:



$\Delta y \, \Delta k_y > 2\pi \rightarrow \Delta y > 2\pi/\Delta k_y$

$\Delta k_y < k_y = \sqrt{(k^2 - k_x^2)}$

k_x real $\rightarrow \Delta k_y < k = 2\pi/\lambda$

$\Delta y > \lambda$

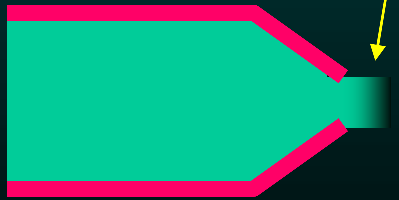
(diffraction limit)

However, for k_x imaginary the condition does not apply and

$\Delta y < \lambda$

becomes possible

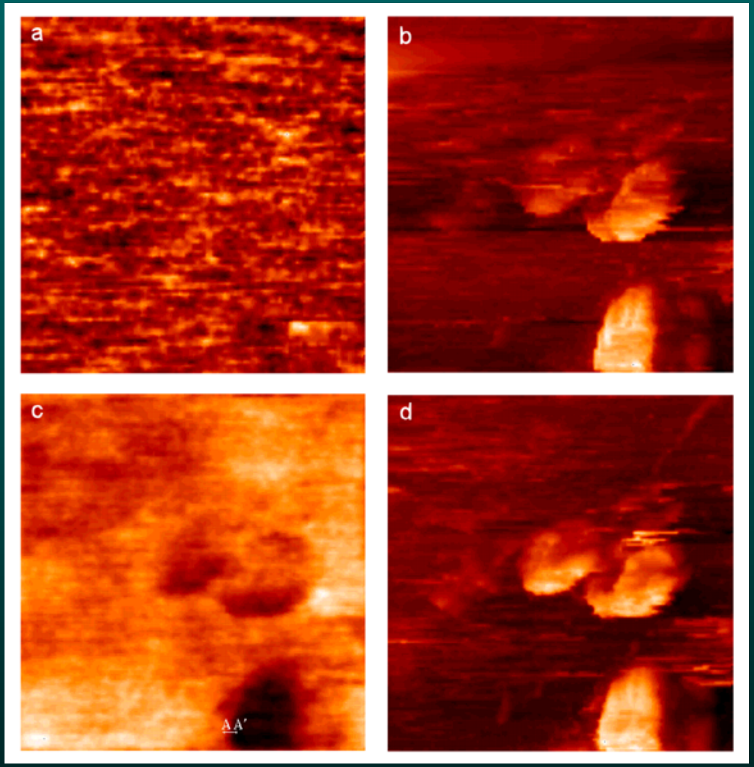
After a narrow optics fiber tip, there is an "evanescent wave" with imaginary in the x-direction k_x



20x20 μm² SNOM image of growth medium (A. Cricenti et al.):

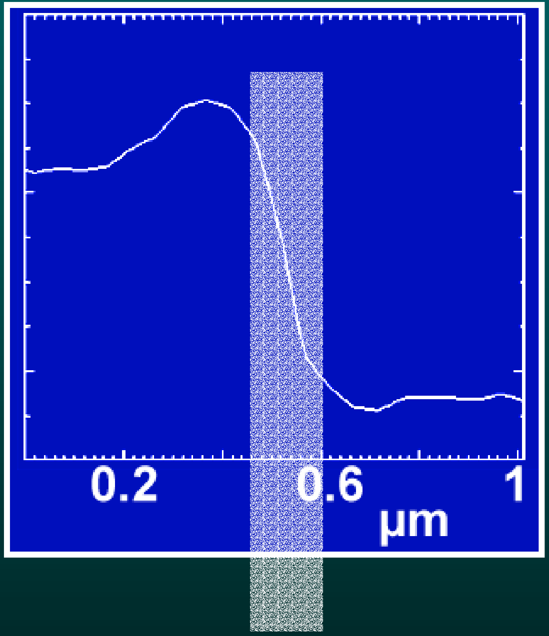
λ = 6.6 μm

S-O & N-O vibrations (λ = 6.95 μm)



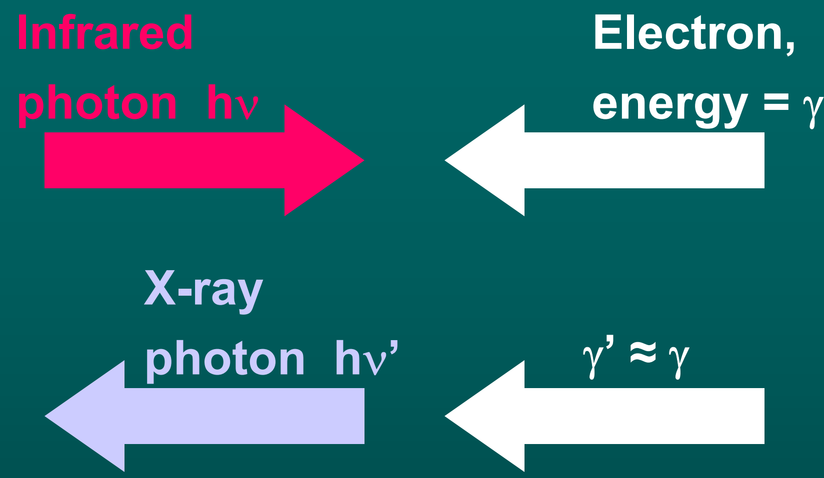
SNOM   topography

Intensity line scan



Resolution ≈ 0.15 μm << λ

The Magic of Compton Backscattering



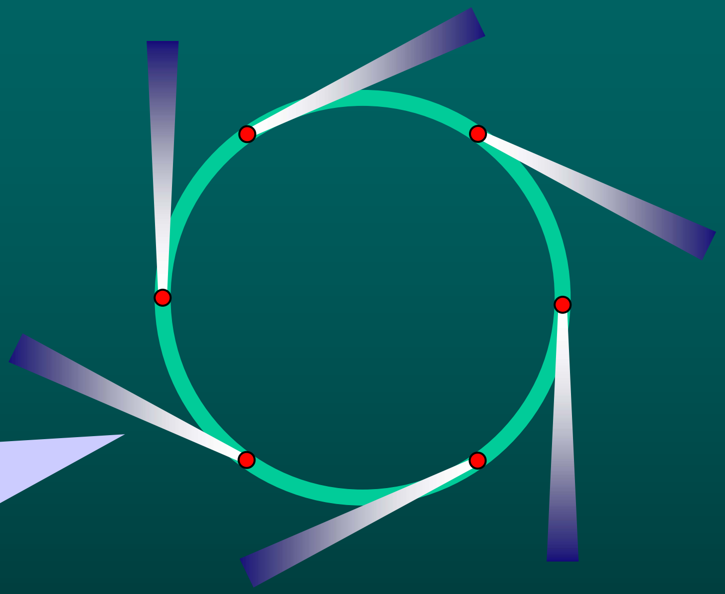
<u>Doppler effect:</u> in the electron beam frame, the photon energy ≈ $2\gamma\, h\nu$. This is also the energy of the backscattered photon in the electron-beam frame.

In the laboratory frame, there is <u>again a Doppler shift</u> with a 2γ factor, thus:

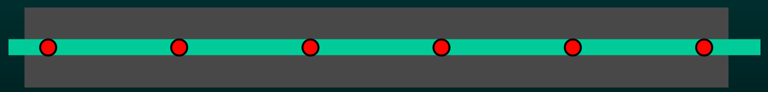
$$h\nu' \approx 4\gamma^2\, h\nu$$

Energy-recovery LINAC sources

The brightness depends on the geometry of the source, i.e., of the electron beam

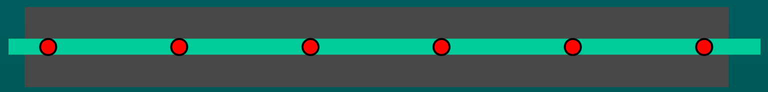


In a storage ring, the electrons continuously emit photons. This “warms up” the electron beam and negatively affects its geometry



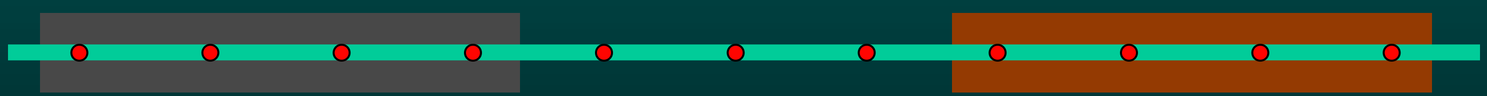
Controlling the electron beam geometry is much easier in a linear accelerator (LINAC). Thus, LINAC sources can reach higher brightness levels

Energy-recovery LINAC sources



However, contrary to the electrons in a storage ring, the electrons in a LINAC produce photons only once: the power cost is too high

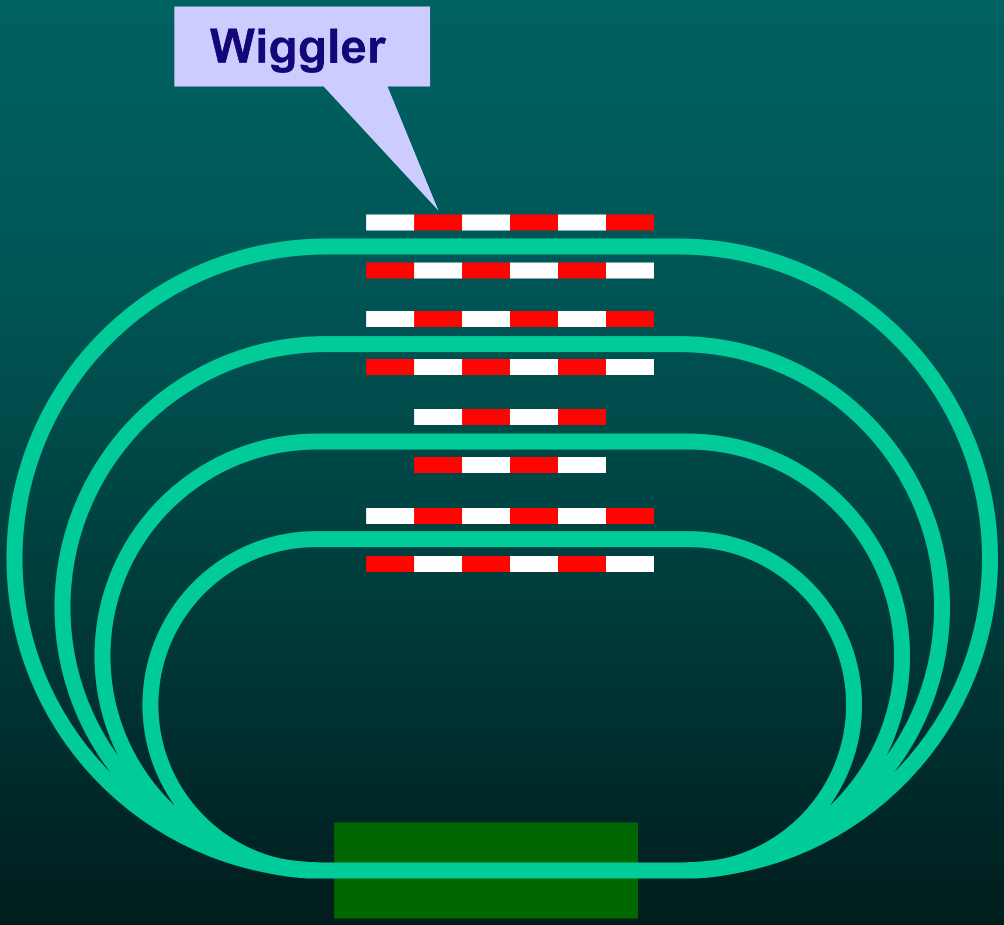
Solution: recovering energy



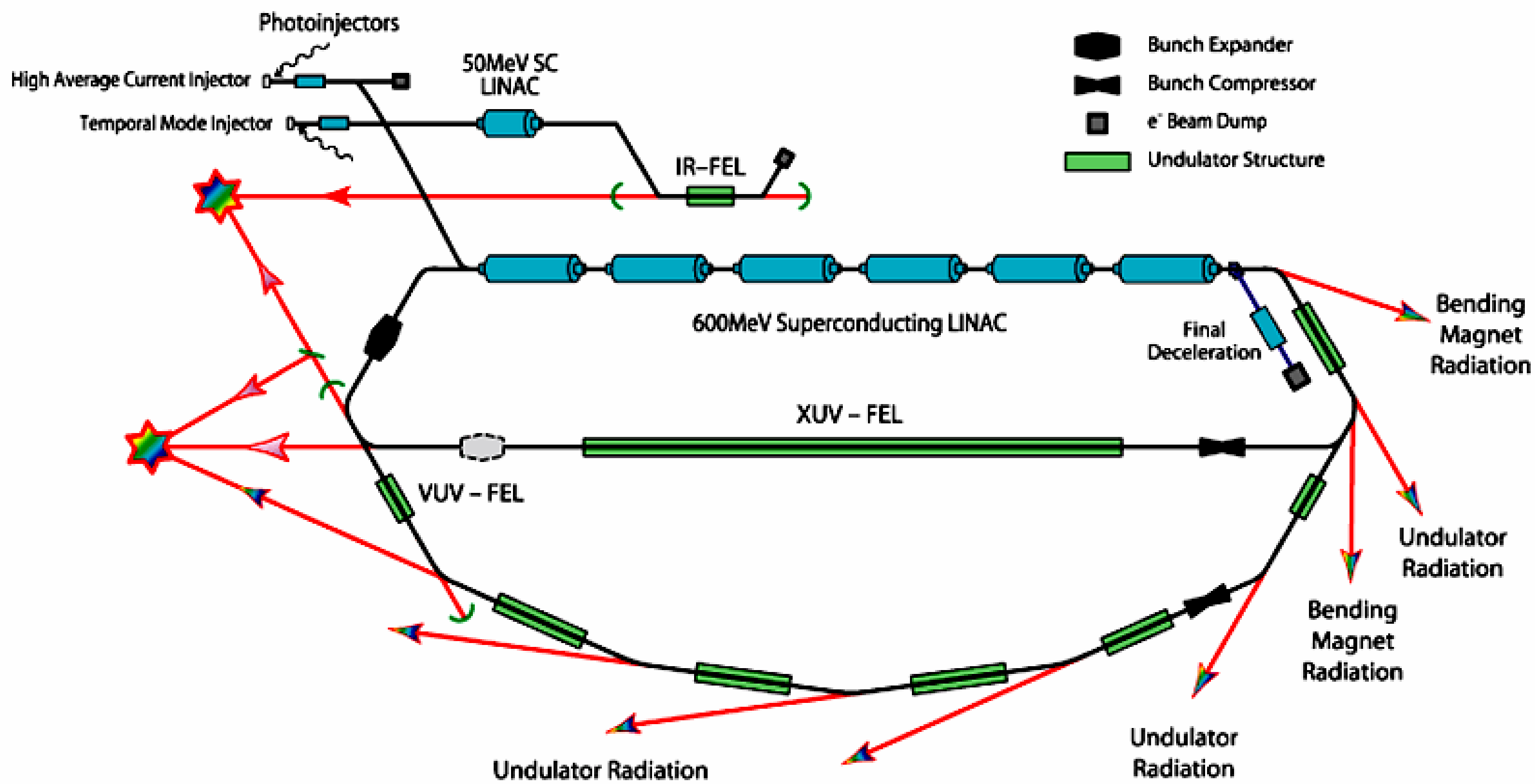
Accelerating section

Energy-recovery section

Example: Kulipanov's "super-microtron" ER LINAC



THE “4 GLS” CONCEPT AT DARESBURY



The 4GLS concept involves the use of an energy recovery linac (ERL) in a ring configuration. This is extremely flexible and will allow the easy incorporation of, for example, a single pass XUV-FEL that would allow the generation of coherent pulses of electron laser radiation up to several hundreds of eV in energy.

Self-amplified spontaneous emission x-ray free-electron lasers (SASE X-FEL's)

Normal (visible, IR, UV) lasers:

optical amplification in amplifying medium

plus optical cavity (two mirrors)

X-ray lasers: no mirrors → no optical cavity → need for one-pass high optical amplification

SASE strategy:

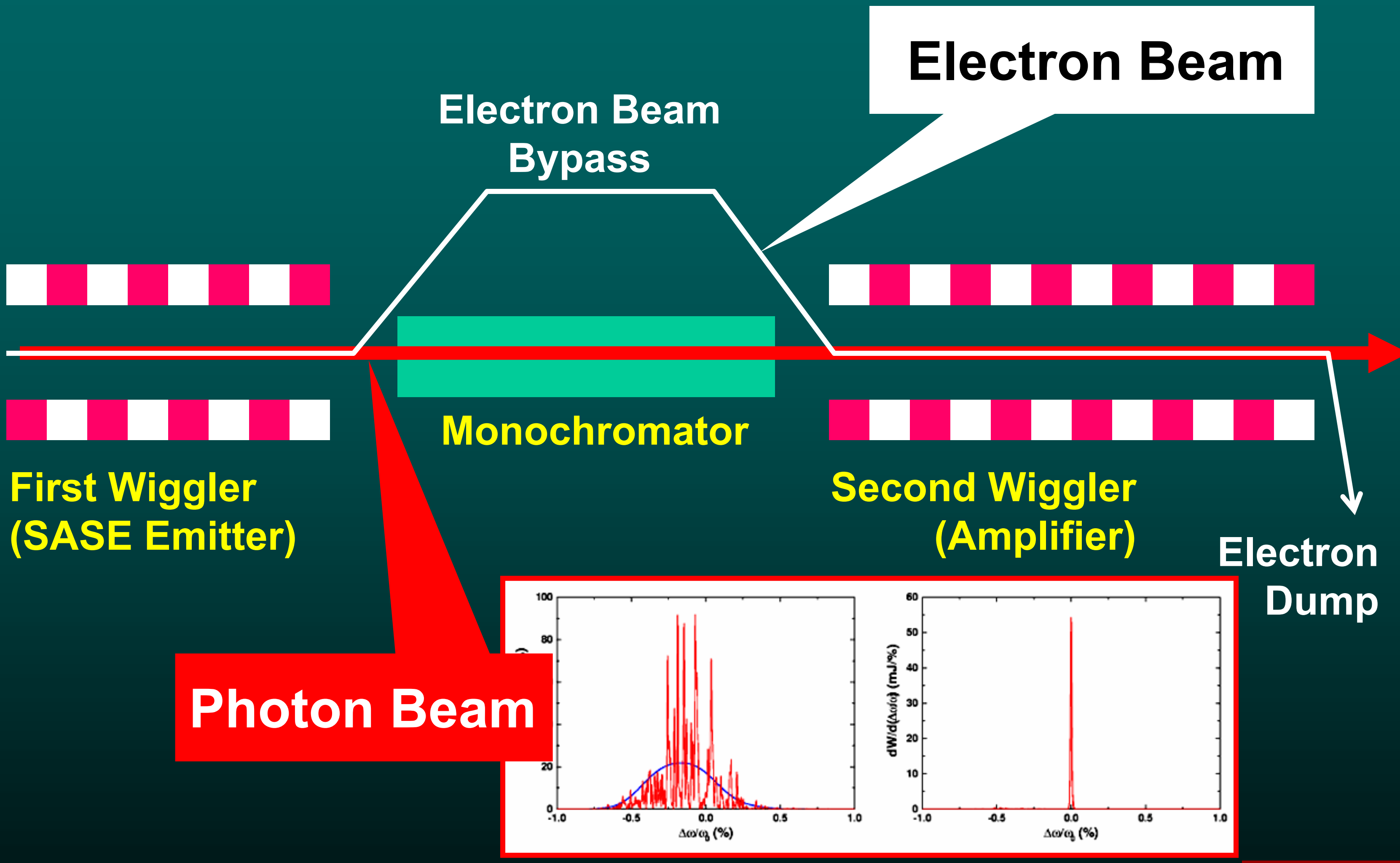
Wiggler

electron bunch

LINAC (linear accelerator)

The microbunching increases the electron density and the amplification and creates very short pulses

Seeding-Amplifier X-FELs




N
TESLA - HAMBURG

SASE x-ray FEL's

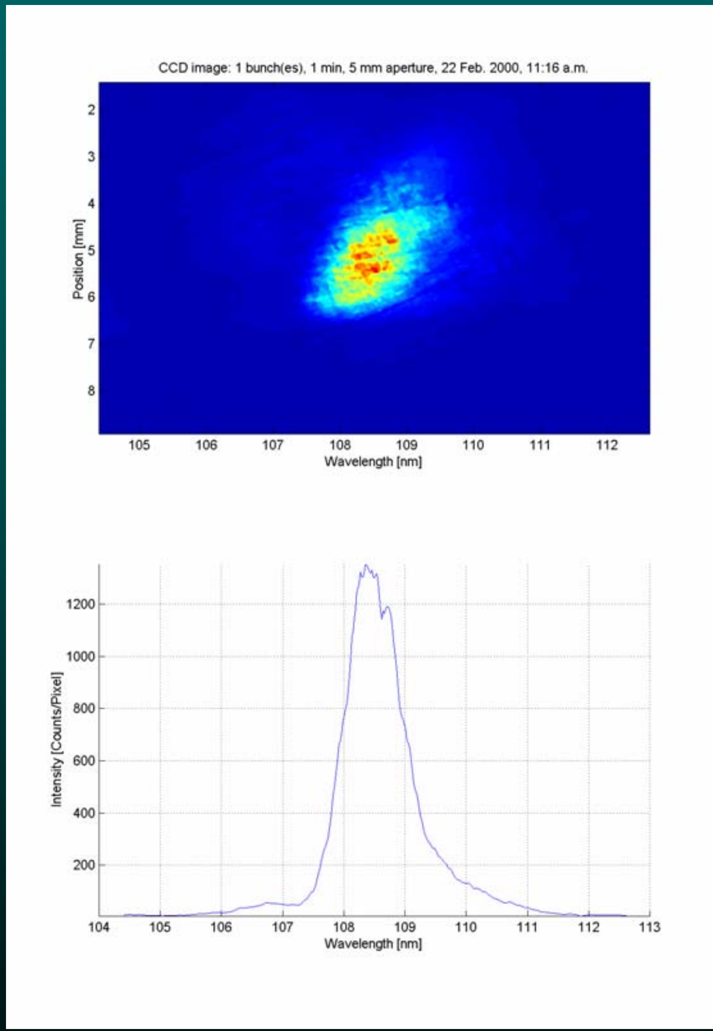


Image of CCD at focal plane of a 1 m monochromator

Dispersion

Vertical Position

The FEL at the Tesla Test Facility

Extracted spectrum

SASE at λ = 108.5 nm

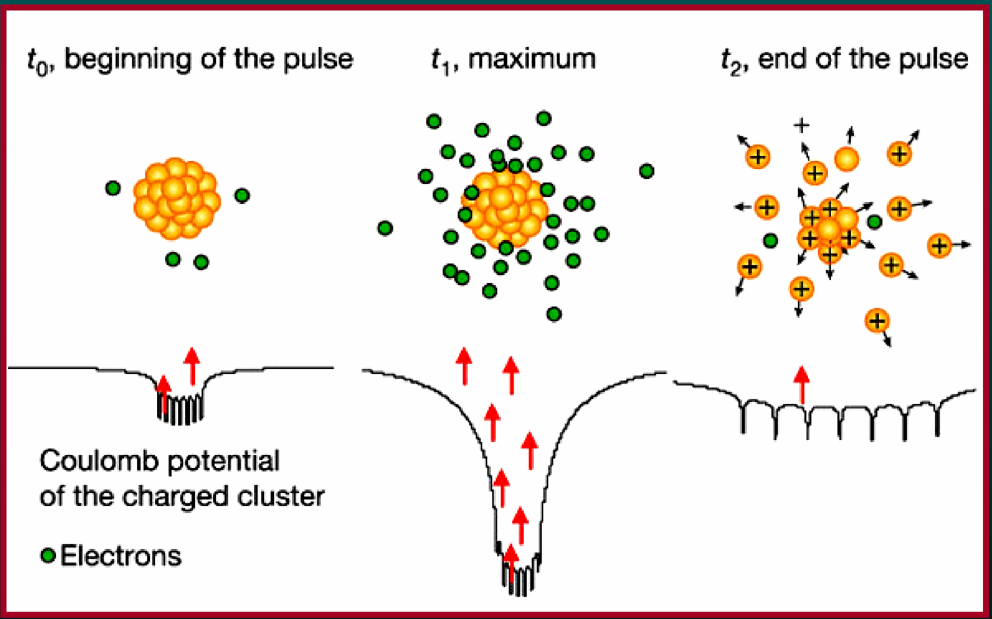
First Real Experiments at the TESLA X-FEL's

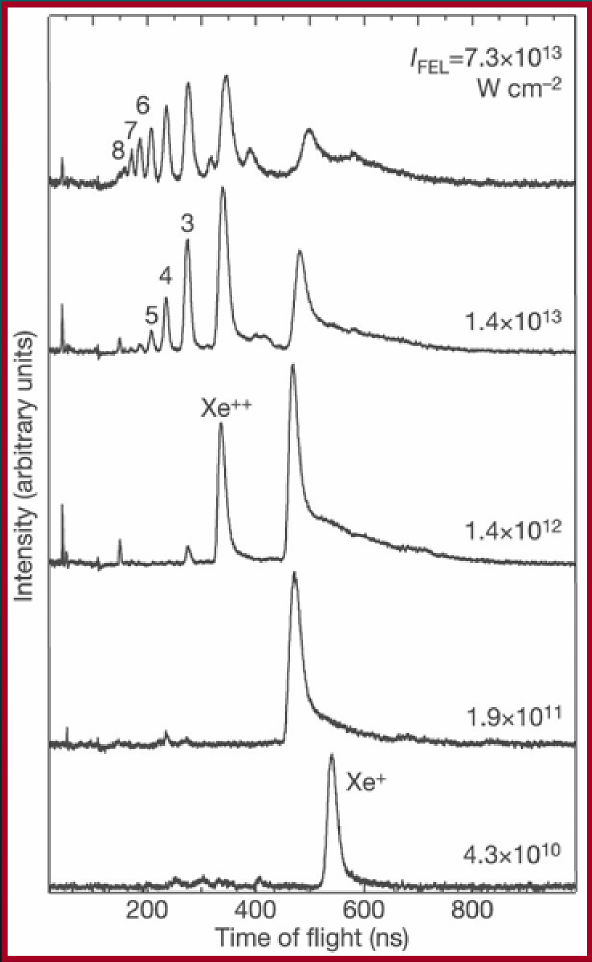
letters to nature

Nature **420**, 482 - 485 (2002); doi:10.1038/nature01197

Multiple ionization of atom clusters by intense soft X-rays from a free-electron laser

H. WABNITZ*, L. BITTNER*, A. R. B. DE CASTRO†, R. DÖHRMANN*, P. GÜRTLER*, T. LAARMANN*, W. LAASCH*, J. SCHULZ*, A. SWIDERSKI*, K. VON HAEFTEN*‡§, T. MÖLLER*, B. FAATZ*, A. FATEEV‡§, J. FELDHAUS*, C. GERTH*, U. HAHN*, E. SALDIN‡, E. SCHNEIDMILLER‡, K. SYTCHEV‡, K. TIEDTKE*, R. TREUSCH* & M. YURKOV‖

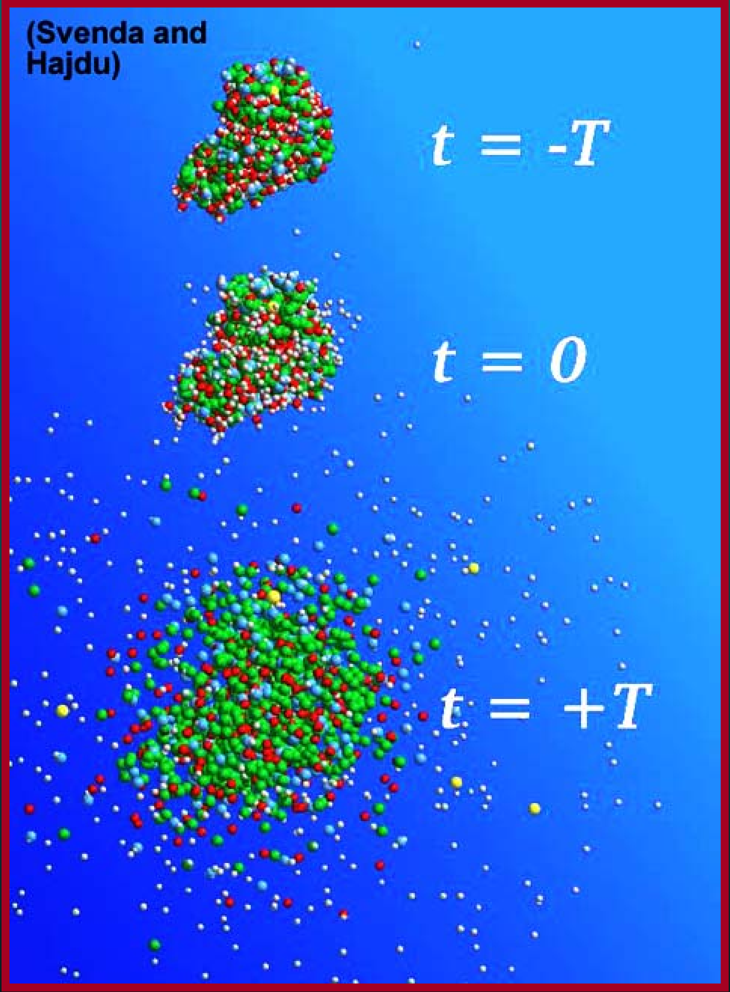




SASE X-FEL's:

superbright (orders of magnitude more than present sources) femtosecond pulses:

- New chemistry?
- One-shot crystallography (no crystals)?
- Total coherence
- Unprecedented electromagnetic energy density
- Is this "vacuum"?



New physics?

Consider the parameters of the Swiss Light Source:

Circumference: 288 m

Bunch Length: 4 □ 10^{-3} m

Single Bunch Current: 10^{-4} A

Electron speed ≈ c ≈ 3 □ 10^{8} m/s

The charge per bunch is 10^{-4} □ 288/(3 □ 10^{8}) ≈ 10^{-8} coulomb, corresponding to 6 □ 10^{10} electrons. The horizontal bunch size is < 2 □ 10^{-5}. Assuming 0.1% coupling, the bunch volume is < 1.6 □ 10^{-15} m^3. Thus, **the electron density exceeds 4 x 10^{19} cm^{-3}.**

What is this: a gas of independent electrons? Or a **correlated multi-particle system?**

What **kind of thermodynamics should we use**? The covalent form of thermodynamics is still an open issue!

For example: T can be defined using the entropy law or the equipartition principle. The two definitions are equivalent in classical physics, but in relativity they lead to different Lorentz transformations of T!

Conclusions:

1. The technology of storage rings and FEL's solved the ancient problem of brightness.
2. The brightness increase was so rapid that applications are still trailing behind.
3. Nevertheless, many exciting results were obtained, for example in spectromicroscopy and high resolution spectroscopy.
4. The most important new achievements will be linked to the interdisciplinary use of photon sources — exporting physics and chemistry techniques to medical research and the life sciences in general.
5. Coherence-based applications will play a special role.
6. New FEL's like the SASE machines are beyond imagination: towards one-shot crystallography?

Thanks:

1. The EPFL colleagues (Marco Grioni, Davor Pavuna, Laszlo Forro Mike Abrecht, Amela Groso, Luca Perfetti, Helmuth Berger, Daniel Ariosa et al.).
2. The Wisconsin colleagues (groups of Marshall Onellion and Pupa De Stasio).
3. The Academia Sinica Taiwan colleagues (group of Yeukuang Hwu).
4. The Vanderbilt colleagues (group of Norman Tolk).
5. The POSTECH colleagues (group of Jung Ho Je).
6. The ISM-Frascati colleagues (groups of Antonio Cricenti and Paolo Perfetti)
7. The facilities: PAL-Korea, Elettra-Trieste. Vancerbilt FEL, SRRC-Taiwan, APS-Argonne, SLS-Villigen, LURE-Orsay