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

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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?

- Photon energy (eV): 10, 100, 1000, 10000
- Wavelength (Å): 0.1, 10, 100

Chemical bond lengths
Molecules
Proteins
Core electrons
Valence electrons

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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/2γ²

Overall: L → L/2γ²

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: \( \gamma \)-factor for \( x' \) and \( t' \) but not for \( y' \) ⇒ \( t_n(\theta) \) reduced by a factor \( \approx 1/\gamma \)
Heat flux (watt/mm²)

- Surface of the sun: 100
- Interior of rocket nozzle: 10
- Nuclear reactor core: 1
- Swiss Light Source (SLS)
- ALS, Elettra, SRRC, PAL
The historical growth in brightness/brilliance
(units: photons/mm²/s/mrad², 0.1% bandwidth)

10²¹
10¹⁵
10⁹

1900       1950        2000

Wigglers
Bending magnets
Rotating anode

SLS
SLS (Swiss Light Source)
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.
Synchrotron x-rays:

Many different interactions

- Scattered photons, fluorescence
- Photoelectrons, Auger electrons
- Transmitted photons
- Diffraeted photons
- Scattered photons
- Photoelectrons, Auger electrons

Many different applications

- Scattering
- Fluorescence spectroscopy
- X-graphy
- Absorption spectroscopy
- EXAFS
- Desorption spectroscopy

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Historical Growth of Synchrotron Publications

Worldwide ISI data 1968 to 2002
Keyword: “synchrotron”
Photoelectron spectroscopy: basic ideas

Formation of chemical bonds:
- Atom
- Solid
- Electron energy
- Photon ($h\nu$)

The photon absorption increases the electron energy by $h\nu$ before ejection of the electron from the solid.

Photoelectric effect:
- Photon ($h\nu$)
- Photoelectron

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Photoelectron spectroscopy of high-temperature superconductivity:

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

High-resolution spectra taken with ultrabright synchrotron radiation.
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$_2$

Fig. 3. (A) ARPES intensity map [light $h \nu = 21.2$ eV ($h$ is Planck's constant and $\nu$ is photon frequency); $T = 300$ K] of (TaSe$_4$)$_2$ 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”:

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)

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

Non-Fermi-liquid Metals: No Fermi Edge
From spectroscopy to spectromicroscopy:

Spectroscopy (energy and momentum resolution)

Microscopy (spatial resolution)

Chemical information

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


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 thanashing. 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 $\xi$, on its angular divergence $\theta$ 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 two point sources:

- 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 occurs over \( \theta \), 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 $L_c = \frac{\lambda^2}{\Delta \lambda}$
- Large coherent power $(2\frac{\lambda}{\xi \theta})^2$
- 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:

- X-ray source
- X-ray beam
- Object
- Detector
Some Problems in Conventional Radiology:

- Low absorption
- Low-intensity, divergent beam
- Limited contrast, may require a high x-ray dose
Light-matter Interactions:

Absorption -- described by the absorption coefficient $\alpha$

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:

Edge between regions with different n-value

detector

Idealized edge image

Real example (leaf)

Detected intensity
“Refraction” x-ray imaging -- potential advantages over absorption:

- Differences between object and vacuum: small in both cases, but larger for $n$ than for $\alpha$.
- 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 live microfish
Building on bubbles:

QuickTime™ et un décompresseur GIF sont requis pour visualiser cette image.
“Refraction” radiology -- the problems

Conventional x-ray source

- Large size
- Large angular divergence

Required, instead:

Solution:

synchrotron sources

SLS (Swiss Light Source)
A bit more sophisticated description

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

* Small & collimated
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
Our own results:
Our own results:
Modeling: conditions for coherence-based radiology

"Diffraction" radiology:

Conditions to see the edge diffraction fringes:
\[ \xi < 0.8 D \sqrt{\frac{\Delta\lambda}{2L}} \approx 100 \text{ micron} \]

\[ \Delta\lambda/\lambda < \sqrt{2} \]

Equivalent condition for "refraction" radiology:
\[ \xi/D < \theta \]

no monochromator!!
Modeling: interplay of “refraction” and “diffraction”

Refraction 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

200μm
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

### Proposal Submission Forms

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<table>
<thead>
<tr>
<th>Proposal Title (max. 200 char.)</th>
<th>Integrating Activity on Synchrotron and Free Electron Laser Science</th>
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<td>Class of Infrastructure</td>
<td>Synchrotrons, Free Electron Lasers</td>
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This B3 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 infrastructure 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 B3 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 by exploiting the intellectual resources of thousands of scientists. The JA'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 B3 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 B3 will also support bilateral exchanges between facilities 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 portfolio. The B3 facility network puts Europe at the forefront of this strategic area of science and technology and the B3 will enable Europe to fully exploit it beyond national barriers.
Free-electron lasers:

Vanderbilt University
Free Electron Laser
Free-electron laser surgery:

Wavelength selection → much less collateral damage:

Laser Surgery: conventional laser (left) vs the Vanderbilt Free Electron Laser

Vanderbilt's new laser gets raves from patient
The scanning near-field optical microscope (SNOM): like the stethoscope

Heart:
Frequency $\approx 30-100$ Hz
Wavelength $\lambda \approx 102$ m
Accuracy in localization $\approx 10$ cm $\approx \frac{\lambda}{1000}$

SNOM resolution: well below the "diffraction limit" of standard microscopy ($\approx \lambda$)
SNOM: why does it work? Consider two slits:

\[ \Delta y \Delta k_y > 2\pi \rightarrow \Delta y > 2\pi/\Delta k_y \]
\[ \Delta k_y < k_y = \sqrt{(k^2 - k_x^2)} \]
\[ k_x \text{ real } \rightarrow \Delta k_y < k = 2\pi/\lambda \]
\[ \Delta y > \lambda \]
(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.):

\[ \lambda = 6.6 \text{ µm} \]

S-O & N-O vibrations
\[ (\lambda = 6.95 \text{ µm}) \]

Intensity line scan

Resolution
\[ \approx 0.15 \text{ µm} \ll \lambda \]
The Magic of Compton Backscattering

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

In the laboratory frame, there is again a Doppler shift with a $2\gamma$ factor, thus:

$$h\nu' \approx 4\gamma^2 h\nu$$
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.
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 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:

LINAC (linear accelerator)→ Wiggler

The microbunching increases the electron density and the amplification and creates very short pulses
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 $\lambda = 108.5$ nm
First Real Experiments at the TESLA X-FEL’s

letters to nature


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


$t_0$, beginning of the pulse  $t_1$, maximum  $t_2$, end of the pulse

Coulomb potential of the charged cluster

- Electrons

Intensity (arbitrary units)

Time of flight (ns)

$\lambda_{FEL}=7.3 \times 10^{-13}$

$1.9 \times 10^{11}$

$4.3 \times 10^{10}$

$1.4 \times 10^{12}$

$1.4 \times 10^{13}$
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”? 

(Svenda and Hajdu)

$t = -T$

$t = 0$

$t = +T$
Consider the parameters of the Swiss Light Source:

- Circumference: 288 m
- Single Bunch Current: $10^{-4}$ A
- Bunch Length: $4 \times 10^{-3}$ m
- Electron speed $\approx c \approx 3 \times 10^8$ m/s

The charge per bunch is $10^{-4} \times \frac{288}{3 \times 10^8} \approx 10^{-8}$ coulomb, corresponding to $6 \times 10^{10}$ electrons. The horizontal bunch size is < $2 \times 10^{-5}$. Assuming 0.1% coupling, the bunch volume is < $1.6 \times 10^{-15}$ m$^3$. Thus, the electron density exceeds $4 \times 10^{19}$ cm$^{-3}$.

What is this: a gas of independent electrons? Or a correlated multiparticle 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 $T$!
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.).
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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.