#### 2023 School on Synchrotron Light Sources and their Applications

### Fundamentals of Synchrotron Radiation from Storage Rings Fundamentals of X-ray Interactions with Matter



**ICTP** 

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#### Two ways to understand synchrotron radiation:

1865-1973:

Maxwell's

equations



$$\vec{r}(t) = \left(\rho \sin \frac{r}{\rho} t, \rho \left(1 - \cos \frac{r}{\rho} t\right), 0\right).$$
the limit of small angles we compute
$$\hat{n} \times \left(\hat{n} \times \vec{\beta}\right) = \beta \left[-\vec{\varepsilon}_{\parallel} \sin \left(\frac{\beta c t}{\rho}\right) + \vec{\varepsilon}_{\perp} \cos \left(\frac{\beta}{\rho}\right)\right]$$

$$\omega \left(t - \frac{\hat{n} \cdot \vec{r}}{c}\right) = 1945-49: \text{ Schwinger's full}$$



ctron lasers

 $\vec{\epsilon} \perp \cos ($ 

radiation emission theory

(1) The hard way, with a complete theory: 8 decades of development, complex formalism



Synchrotron radiation and X-ray free-electron lasers (X-FELs) explained to all users, active and potential

Substituting into the

 $\xi = rac{
ho \omega}{3 c \gamma^3} ig( 1 + \gamma^2 heta^2 ig)$ 

Yeukuang Hwu<sup>a,b,c</sup>\* and Giorgio Margaritondo<sup>d</sup>\*



(2) Considering only the pure relativistic case

Simple analysis, elementary math

rimoz kebernik kibic

on Radiation 18, 101 (2011)



Why are x-rays from electron accelerators so important? In other words, what <u>can we probe</u> with their wavelengths and photon energies?



## So, we need excellent x-ray sources: how can we build them with relativity?





#### The five relativistic ingredients:



-9 **~**3-

(4) **<u>Time</u> <u>Dilation</u>**: a

time interval  $\Delta t_{\rm e}$  in

 $\mathcal{R}_e$  increases in  $\mathcal{R}_c$ 

 $\rightarrow V$ 

(5) <u>Relativistic</u> <u>mass</u>: the electron mass  $m_{\rm e}$  in  $\mathcal{R}_{e}$ (rest mass) changes in  $\mathcal{R}_{\mathcal{L}}$  to  $m_{\rm L} = \gamma m_{\rm e}$ 

(2) <u>**Doppler shift</u>**: due to the source motion (speed *v*), the wavelength  $\lambda_e$  emitted by an electron in  $\mathcal{R}_e$  decreases to  $\lambda_L \approx \lambda_e/(2\gamma)$  when seen in  $\mathcal{R}_{\mathcal{L}}$ </u>





#### To understand how a synchrotron source works, we start from electrons oscillating in an antenna

...they are <u>accelerated</u> electric charges, thus they <u>emit electromagnetic waves</u>

But the typical emission is <u>long-wavelength</u> radio waves, not <u>short-wavelength</u> x-rays!

The electron mass is small: this enhances the acceleration and the emission

waves

To get x-rays, we need relativity: consider an electron oscillating transversally, but also moving longitudinally with speed  $\underline{v} \approx c$ 

#### waves

 $V \approx \gamma$ 

#### How can we obtain this complex motion?

### A solution:

The undulator forces the electron to oscillate in a transverse direction: being accelerated, it emits electromagnetic waves – "synchrotron radiation"

the emitted

wavelength 1 in

 $\mathcal{R}_{\mathcal{L}}$  is <u>related</u> to the

undulator period L

Add an "undulator": a periodic series of magnets, period *L* 

> relativistic electron,  $v \approx c$

...consider an electron forced to circulate in vacuum in a storage ring by a special system of magnets (not shown)



velocity  $-v \approx -c$ :

 $L/\gamma <$ 

Furthermore, it "sees" the undulator period shrunk to  $\approx L/\gamma$  by the relativistic Lorentz contraction. This is also the emitted wavelength  $\lambda_e$  as detected in  $\mathcal{R}_e$ :  $\lambda_e \approx L/\gamma$ 



...however, in the laborate  $\gamma$  <u>me</u>  $\mathcal{R}_{\mathcal{L}}$  the motion of the source (the elec n) causes the (relativistic) <u>Doppler shift</u> -- furt or decreasing the detected wavelength by a f tor  $\approx 1/(2\gamma)$ 

Together, the Lorentz contraction and the Doppler effect decrease the wavelength in  $\mathcal{R}_{\mathcal{L}}$  to:

 $\lambda_{\rm L} \approx \frac{L/\gamma}{2\gamma}$ 

Example: *L* = 1 cm, γ= 5000, →λ<sub>L</sub> ≈ 2 Å: <u>x-rays!!!</u>





large flux, small area, small divergence

## This is how the brightness of x-ray sources evolved in history: synchrotrons boosted it!

(units: photons/mm<sup>2</sup>/s/mrad<sup>2</sup>, 0.1% bandwidth)

1033

1027

(peak values)

-ra

electron

tree

an increase by 26 orders of magnitude since 1970, while computer chips "only" improved by 7 orders of magnitude!!!



Synchrotron sources reach <u>very high</u> <u>brightness levels</u> (some 10<sup>15</sup> times more than conventional x-ray sources) -- how? Thanks to four factors:

- 1. <u>Relativity drastically boosts the emitted power</u>
- 2. <u>Relativity</u> sharply reduces the <u>angular divergence</u>
- Electrons in vacuum can handle more emitted power than those in a solid since the power does not damage their environment
- 2. Different electrons circulate in the accelerator along slightly different paths. The <u>source size</u> is the transverse cross section of all paths. Effective electron beam controls make it very small

#### Relativity at work: <u>emitted power</u>



<u>"Larmor law"</u>: the emitted power is proportional to the square of the transverse acceleration,  $a_e^2$  emission If v = zero,  $a_e = a_1$ : the power is proportional to  $a_1^2$ 

If  $v \neq \text{zero:}$  from  $\mathcal{R}_e$  to  $\mathcal{R}_{\mathcal{L}}$  the time is dilated by  $\gamma$  and the <u>transverse</u> coordinate remains invariant; the acceleration = coordinate/time<sup>2</sup> is divided by  $\gamma^2$ :  $a_{\text{L}} = a_{\text{e}}/\gamma^2$ , and  $a_{\text{e}} = \gamma^2 a_{\text{L}}$ 

...the power is proportional to  $a_e^2 = \gamma^4 a_L^2$ , thus to  $\gamma^4 = (\text{energy})^4 / (m_o c^2)^4$ 

The emission increases as the 4<sup>th</sup> power of the electron energy, to <u>very high levels</u>

...and it decreases as  $1/m_0^4$ : electrons emit a lot, protons much less

oscillating

electron

#### Relativity at work again: angular collimation of synchrotron radiation

...but seen in  $\mathcal{R}_{\mathcal{L}}$  the range shrinks to a <u>very</u> narrow forward cone

in the electron  $\mathcal{R}_{e}$  frame, x-rays are emitted in a we ve wide angular range, like se the waves from an antenna  $\mathcal{R}_{e}$ 

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V≋Ć

wave velocity in  $\mathcal{R}_e$  (magnitude *c*, relativistic invariant) wave velocity in  $\mathcal{R}_{\mathcal{L}}$ (same magnitude *c*)

 $\mathcal{R}_e \rightarrow \mathcal{R}_{\mathcal{L}}$ : the time is dilated by  $\gamma$ ; the transverse coordinate is invariant; the transverse velocity, proportional to 1/time, changes from  $c_{ye}$  to  $c_{yL} = c_{ye}/\gamma \approx c/\gamma$ . The vector velocity rotates to  $\theta \approx c_{yL}/c \approx 1/\gamma$ 

#### Small angular spread $\approx 2\theta \approx 2/\gamma$ : milliradians!!!





Around a storage ring, there are other types of x-ray sources besides undulators







brightness than from bending magnets

### Synchrotron emission is also polarized!

Why? Imagine an electron in an undulator or a wiggler:





Linear polarization in the horizontal

[TOP VIEW]

0

### [FRONT VIEW]

plane, where the electric field perturbations by the wave occur

...same conclusion for bending magnets



# Summary of the amazing properties of synchrotron sources discovered so far:



<u>COHERENCE</u> is "what enables radiation to produce <u>visible</u> wave effects (e.g., diffraction or interference)"



With a more realistic source, we discover TWO kinds of coherence: "time" and "spatial"

source  $(\Delta \lambda)$ 

Example: pinhole diffraction - a point source emitting only <u>one wavelength</u> always produces a visible diffraction pattern: it has full coherence



...if the source emits a <u>band</u> of wavelengths, each one of them produces a pattern; in the superposition of patterns the fringes may be too blurred to be visible: this leads to the notion of "time (or longitudinal) coherence"

Likewise, if the source is not a point but has a finite size the pattern may become impossible to see: this leads to "spatial (or lateral) coherence"

Effects of multiple wavelengths: longitudinal (time) coherence

different wavelengths produce different patterns...

> bandwidth  $\Delta \lambda$ bandwidth

 $\bigcirc$ 

...and their superposition blurs the pattern features

Using the "<u>coherence length</u>"  $L_c = \lambda^2 / \Delta \lambda$ , the condition for time coherence is:  $L_c > \lambda$ 

Source geometry: spatial (lateral) coherence Each point in the source produces a

diffraction pattern – and the superposition blurs the pattern fringes

solid angle  $\Omega$ 

D

When are such fringes visible?  $\xi H/D \approx$  maximum distance between centers of patterns given by different source points fringe spacing  $\approx (H/\delta)\lambda$ ; To see the pattern features:  $\xi H/D \leq (H/\delta)\lambda \rightarrow \delta \leq \lambda D/\xi$ 

**for later** 

coherence

Another way to look at lateral coherence: Illuminated screen area:  $\Omega D^2$ ; pinhole area  $\approx \delta^2$ ; portion of waves contributing to diffraction  $\approx \delta^2/(\Omega D^2) \leq (\lambda D/\xi)^2/(\Omega D^2) =$ 

"coherent power factor": if it is large, there is lateral coherence

#### Spatial coherence — summary:

- It requires a large coherent power factor  $\lambda^2/\xi^2\Omega$ : we need a synchrotron source with small  $\xi^2$  and  $\Omega$
- Due to the  $\lambda^2$  term, it is difficult to achieve for small x-ray  $\lambda$ 's (by the way: a  $\lambda^2$  term is also present for longitudinal coherence)
- NOTE: the brightness is proportional to  $1/[\xi^2 \Omega]$  the efforts to enhance the brightness by decreasing  $\xi$  and  $\Omega$  also increased the spatial coherence

Coherent x-rays allow phase-contrast imaging, a powerful radiology that produces pictures with sharp contrast and very small details: we shall see later how





### From storage rings to X-ray Free Electron Lasers (X-FELS)

Synchrotron sources have several laser-like properties: collimation, high intensity, coherence... are they lasers? <u>NO</u>: the emission mechanism is different!

But there is now a new class of x-ray sources more similar to lasers: the FELs (Free Electron Lasers) – with optical amplification caused by the interaction of electrons with their emitted waves



...this is the FEL optical amplification mechanism

### Why are not all undulators behaving like FELs?

The relativistic "longitudinal mass" of high-speed electrons,  $\gamma^3 m_{\rm e}$ , is very heavy. It takes a (relatively) long time to move the electrons to the "slices": micro-bunching requires <u>very long</u> undulators

...and the microbunching period is very short (x-ray wavelengths): the structure is delicate and easily destroyed

#### ...but x-ray free electron lasers are now a fantastic reality, notably at ELETTRA

The X-FEL "FERMI" (Free Electron Radiation for Multidisciplinary Investigations)

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### **Fundamentals of X-ray Interactions with Matter**



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Interactions between x-rays and solids: general formal background Wave function in vacuum:  $W_{o} exp[i(kx - \omega t)]$ "PHASE In the solid: k changes to nk, where  $n = n_{\rm B} + in_{\rm I}$  (complex refractive index) Wave function:  $W_0 \exp[i(nkx - \omega t)]$  $= W_{o} \exp(-n_{l}kx) \exp[i(n_{R}kx - \omega t)]$ Factor decreasing with the distance: absorption  $n_{\rm B}k$  determines the phase, causing phase effects: refraction, diffraction, interference...

## The real and imaginary parts of the refractive index, $n_{\rm R}$ and $n_{\rm I}$ , are linked to each other

- $n = n_{\rm R} + in_{\rm I}$  is the <u>"response function"</u> describing how the system interacts with photons
- General rule for complex response functions: the real and imaginary parts are not independent but <u>linked by relations</u> <u>like the "Kramers-Kroenig" (KK) equations</u>
- This explains the relations between different types of phenomena:
  - strong reflection corresponds to strong absorption (think about a mirror)
  - refraction and absorption phenomena occur for the same wavelengths
  - Faraday rotation is linked to circular dichroism
  - Hi-fi systems: the amplification is linked to the phase changes

#### Kramers-Kroenig relations for x-rays: example

#### $Si_3N_4$



...if something happens for absorption, something also happens for refraction The mathematical forms of the Kramers-Kroenig relations include integrals over the entire v frequency range, from zero to infinity

Thus, if one measures  $n_{\rm R}$  over the entire <u>v</u> spectral range, one can derive  $n_{\rm I}$  mathematically avoid measuring it -- and vice-versa

Realistically, however, one cannot measure  $n_R$ or  $n_I$  for all frequencies, but over a frequency range as wide as possible: broadband synchrotron sources are helpful! We shall now explore effects linked to the imaginary part  $n_{\rm I}$  of the complex refractive index, "inelastic" phenomena in which x-ray waves lose energy. The three most important ones are (plots for water):



#### Photon absorption in an (insulating) solid:



### Photon absorption: theory



The direction of  $\vec{A}$  is that of the photon polarization: polarized synchrotron radiation can explore the electronic state symmetry (parity) A core-level absorption edge reveals the presence of the corresponding element and its chemical status -- plus more information

EXAFS =Extended X-ray **Absorption Fine** Structure NEXAFS =Near-Edge Xray Absorption **Fine Structure** 



#### EXAFS mechanism:



From the oscillations, one can derive the local interatomic distance *d*, a very valuable piece of information

The outgoing and backscattered electron waves interfere constructively or destructively depending on the distance *d* and on the electron wavelength --which in turn depends on the electron energy and therefore on the photon energy *hv*. This produces oscillations in the absorption vs. *hv* plots



What happens to the energy of a photon absorbed by a solid? It can cause the emission of another photon (fluorescence)





<u>Photoemission</u>, a leading synchrotron technique based on x-ray absorption





The photoelectric effect increases the electron energy by hv: one can derive from measured photoelectron energies the electron energies in the solid

### Photoemission detects valence electrons and core electrons



...synchrotron photoemission actually transformed my bookish quantum notions into very tangible realities!

## Photoemission: a metal compared to a semiconductor



#### Angle-resolved photoemission

photon

angle-resolved
electron detector

photoelectron





by detecting the energy and direction of a photoelectron, one can obtain its k-vector and derive the experimental band structure E(k) – here the results for CdS

## Photoemission of a (high-temperature) superconductor



## From photoemission spectroscopy to spectromicroscopy:



But, as the probed area decreases, the signal is lower: one needs a bright synchrotron source!

# Two ways to implement photoemission spectromicroscopy:



### Photon beam focusing

Photoelectron detection with magnifying electron lenses

## Photoemission spectromicroscopy results: "chemical maps":



Photoelectron images at three different photoelectron energies: note the chemical contrast inversion [M. Marsi et al., J. Electron Spectroscopy **84**, 73 (1997)] We shall now describe elastic phenomena corresponding to the real part  $n_R$  of the refractive index, such as refraction or reflection (both very weak for x-rays), and:

> Elastic x-ray scattering: a tool to explore the microscopic structure of objects



Since x-rays are mainly scattered by electrons, the object is described by the space distribution of the <u>electronic charge</u>, *F* 

The key property:

- W corresponds to the Fourier transform of F
- The inverse Fourier transform of W corresponds to F

Thus, we can hope to retrieve the electronic charge distribution of an object by performing an inverse Fourier transform of the pattern created by the scattered detector x-rays on the detector inverse **Fourier transform** 

BUT: we do not measure the scattered wave  $W = W_0 \exp(i\vec{r} \cdot \vec{k'})$ , only its intensity,  $W^*W = W_0^*\exp(-i\vec{r} \cdot \vec{k'}) W_0\exp(i\vec{r} \cdot \vec{k'}) = W_0^*W_0$ . Thus, we do not know its PHASE. This severely affects the inverse Fourier transforms, creating the PHASE PROBLEM. Special synchrotron-based methods are used to solve it Grasping the role of Fourier transforms: x-ray scattering by a point-charge q



Using as a reference a wave scattered by a charge  $q_0$ at the origin, the path difference is:  $L_a - L_b = \vec{r} \cdot (\lambda / 2\pi) \vec{k} - \vec{r} \cdot (\lambda / 2\pi) \vec{k'} = \vec{r} \cdot \vec{s} \cdot (\lambda / 2\pi)$ , where  $\vec{s} = \vec{k} - \vec{k'} =$  "scattering vector"

Phase difference =  $(2\pi / \lambda)$ (path difference) =  $\vec{r} \cdot \vec{s}$ : the wave scattered by q is proportional to  $q \exp(i\vec{r} \cdot \vec{s})$ 

Now look at an entire charge distribution  $F(\vec{r})$ :  $F(\vec{r})$ 

The charge in the infinitesimal volume  $d^3\vec{r}$  is  $F(\vec{r})d^3\vec{r}$ The wave scattered by this charge is proportional to  $F(\vec{r})d^3\vec{r} \exp(i\vec{r}\cdot\vec{s})$ 

 $\vec{s} = \vec{k} - \vec{k'}$ 

Thus, the total scattered wave  $W(\vec{s})$  is proportional to  $\int F(\vec{r}) \exp(i\vec{r}\cdot\vec{s}) d^3\vec{r}$ 

...indeed, the Fourier transform of the distribution F!

...and  $F(\vec{r})$  is proportional to

k

 $\int W(\vec{s}) \exp(-i\vec{r}\cdot\vec{s}) d^3\vec{s}$  (inverse Fourier transform)

Fourier transform relation between some simple objects and their scattered waves:



Note: periodic object  $\rightarrow$  periodic *F*  $\rightarrow$  periodic pattern ("Bragg spots")!



#### Small-angle and large-angle scattering



In a real experiment, the detector captures only a portion of the solid angle, i.e., only part of the scattered x-rays

Because of the Fourier transform properties, when scattering is only detected at small angles the inverse transform gives the general shape of the object but not its fine details



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#### 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 a large number of molecules organized in a "crystal"



Problem: obtaining large stable crystals, in particular for hydrophobic molecules – for example, many membrane proteins

We must now look at phenomena in which the effects of  $n_R$  and  $n_I$  are both present, such as <u>imaging with</u> <u>coherent x-rays</u>. We can use a simple analogy: seeing a bottle of red wine

we see the wine because it absorbs certain wavelengths and looks red but we also see the <u>edges</u> of the (transparent) glass bottle because they deviate the light by refraction/scattering likewise, "phase contrast" (refraction/scattering) causes sharp <u>edges</u> in synchrotron radiographs



...to see the edges we need x-rays with a well defined direction: fortunately, the high spatial coherence of synchrotron sources implies <u>angular collimation</u> A simple model of how edges are caused by refraction in phase-contrast radiology [G. Margaritondo and G. Tromba, J. Appl. Phys. **85**, 3406 (1999); Y. Hwu et al., J. Appl. Phys. **86**, 4613 (1999)]



... high lateral coherence is required (x-rays with a well-defined direction); on the contrary, high longitudinal (time) coherence is not needed

## Example of study with phase contrast radiology: the magic light of fireflies



microtomography of a firefly "lantern" [Y. L. Tsai, Y. Hwu et al.]

...being able to detect all vessels, including the smallest ones, we could identify the incredibly effective emission mechanism

#### Synchrotron tomography reads ancient manuscripts:

#### ports: fortions books and genarics of free fortions books and genarics of the offer fortions books and the offer fortions

#### ...even under seal:







so, Lady Cataruçia Savonario of Venice could speak to us after seven centuries [results of Fauzia Albertin]

#### Imaging with coherent x-rays: exploring the brain, neuron by neuron





SYnchrotrons for Neuroscience – an Asia-Pacific Strategic Enterprise

(SARI/SSRF-China, PAL-Korea, AS-Taiwan, RIKEN/Spring8-Japan, NUS/SSLS-Singapore, ANSTO-Australia, SLRI-Thailand, SESAME-Jordan)

At the end of our journey, I would like to thank: Maya Kiskinova and Yeukuang Hwu for disclosing their unpublished results Primoz Rebernik for contributing to our synchrotron radiation theory The school organizers for their kind invitation

...and thank you, young folks, for attending my lectures: your future looks brighter than ever!

### For further reading:

#### Y. Synchrotron radiation and X-ray free-electron lasers Show and K-Gs Margaritonco ve and potential ynchrotron Radiation and X-ray Electron ty, Academia Sinica, Taipei 11529 Science, National Cheng ity, aina ersity, Hsinchu 30013, 5 Lausanne, Switzerland. **Received 8 September** \*Correspondence e-mail: phhwu@sinica.edu.tw, giorgio.margaritondo@epfl.ch Accep **S**. enterprise involving tens of thousands of researchers. Initially, almost all users Edited by M. Yamamoto, RIKEN SPrin ph Center, Keywords: synchrotron; X-FEL; re and others. This poses a challenge: explaining synchrotron sources without pond , physicated background in theoretical physics. Here this challenge n novative approach that only involves elementary notions, commonly possessed by scientists of all domains.