# Lecture : Coherence School of Crystallography 2024



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# Coherence Outline of the Lecture

Reflections on Coherence... From Visible to X-rays

Generating and Using X-ray Coherence – From the Source Point to the Detector

- Definition of Beam Coherence: Volume and Degree of Coherence
- > Natural Emergence of Coherence: Generating Coherent X-rays from Incoherent Sources
- Measuring X-ray Coherence: Diffracting Object and Near/Far-field Detection
- Current Use of Coherent X-rays in Condensed Matter
  - What Can We Expect to See in X-ray Diffraction on Disordered Systems?
  - Structural Studies through Direct Analysis of Speckle Patterns
  - Coherent Diffraction Imaging:
    - Forward Techniques: CDI, Holography, Ptychography, Phase Contrast Tomography
    - In Bragg Condition: CDI, Holography, Ptychography
  - Slow Dynamics Experiments: XPCS (Forward or Bragg)
- New Opportunities Offered by Next-Generation Sources

#### A Standard Coherent Source: The Visible Laser

 $\lambda$  = 530nm What is the typical object size that can diffract light?

Diffraction of a Grating by a Laser Beam?

√ОК

Grating through binoculars



#### Fourier Transform

# Diffraction of a hair by a laser beam?



a~100µm...



Following these observations, we can ask a few questions...

- $\blacktriangleright$  What are the conditions on  $\lambda$  and *a* to observe a diffraction pattern?
- What are the properties of the laser necessary to get diffraction by these objects?
- Would the same experiments work with X-rays?
- > What is coherence?
- How can coherent X-rays be generated in practice?
- How could a coherent X-ray beam be useful in condensed matter?

#### What can be learned from Ewald's construction ?









To observe the diffraction of a beam having a wavelength  $\lambda$  by a crystal with lattice parameter a, it is necessary to have:



# Generating and Using X-ray Coherence – From the Source Point to the Detector

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#### Basic Experiment to Study an Object: Illuminate It

Necessary Equipment: A Source, an Object, a Detector



How to Introduce Coherence into This Experiment?

- On the source side ?







- On the object side ?





- Set Up Under Specific Observation Conditions / Use a Special Detector ?

# Where does the coherence of a beam come from?

What limits the validity of the summation of light amplitudes?



#### Longitudinal coherence length (temporal)



$$2\xi_L = \frac{\lambda^2}{\Delta\lambda}$$

Proportional to  $\lambda$  , inversely proportional to  $\Delta\lambda/\lambda$ 

 $\rightarrow$  The smaller  $\lambda$  , the smaller  $\xi_1$  ...

#### Transverse coherence length (spatial)



 $\xi_{\text{T}}: \text{length perpendicular to } \textbf{k} \text{ for} \\ \text{which two beams having an} \\ \text{incidence difference } \alpha \text{ will be} \\ \text{out-of-phase} \\ \end{cases}$ 

$$tg(\alpha) = \frac{S}{R} = \frac{\lambda}{2\xi_{\alpha}}$$

$$2\xi_T = \frac{\lambda R}{S}$$

 $\frac{S}{R}$  Numerical aperture of the source

 $\begin{array}{l} \mbox{Proportional to } \lambda \\ \mbox{Proportional to the source-sample distance} \\ \mbox{Inversely proportional to the source size} \end{array}$ 

Link Between the Coherence Volume and the Sample Volumes



#### How to Gain Spatial Coherence (easily)?

Propagation of a partially coherent beam



We can get a coherent beam from an intrinsically incoherent source (like the sun)

2. Coherence Degree:

$$\beta = \frac{\xi(z)}{\sigma(z)} = \frac{\xi}{\sigma}$$

The coherence degree is constant through propagation

#### A Bit of Pragmatism: Coherence in Numbers...

With wavelengths more than 1000 times smaller than visible wavelengths, what values do we obtain?



#### Increase Coherent Flux Using Large Scale Instruments



#### With an undulator beamline at SOLEIL





# Influence of Optical Elements on Coherence Surface rugosity



With X-rays, the surface roughness must be compared to  $\lambda$ ...



Optical elements quickly degrade the coherence of an X-ray beam...It is necessary to use secondary sources after the optics and let the beam propagate freely as much as possible.

#### Coherence setup of the CRISTAL beamline at SOLEIL



#### Measuring the Coherence Degree of an X-ray Beam



The visibility of the fringes is related to the degree of coherence. At synchrotron facilities  $\rightarrow$  measure the degree of coherence with slits?







X-rays  $(\lambda \sim 1 \text{ Å})$  with a=2µm slit: d<sub>f</sub>= 2 cm









#### Measurement at the CRISTAL beamline of SOLEIL





2m

Ligne Cristal Soleil

#### Visibility measurement in the far-field at CRISTAL



#### Measuring visibility in the near-field at CRISTAL

![](_page_32_Figure_1.jpeg)

Slit gap (μm)

# Current Use of Coherent X-rays in Condensed Matter

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- Generating and Using X-ray Coherence From the Source Point to the Detector
- Current Use of Coherent X-rays in Condensed Matter
  - What Can We Expect to See in X-ray Diffraction on Disordered Systems?
  - Structural Studies through Direct Analysis of Speckle Patterns
  - Coherent Diffraction Imaging:
    - Forward Techniques: CDI, Holography, Ptychography, Phase Contrast Tomography
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#### What does coherence bring in diffraction ?

Classical diffraction allows for a perfect determination of the average structure of crystals! This is done by measuring *a portion of reciprocal space*, applying *symmetry constraints*, and carefully analyzing the *intensities of the Bragg peaks*.

Coherent diffraction provides information about deviations from the perfect arrangement.

For a crystal with disorder:

=

$$A(\mathbf{q}) = \sum_{uvw} F_{uvw}(\mathbf{q}) e^{-i\mathbf{q}\cdot\mathbf{R}_{uvw}} = \sum_{n} F_{n}(\mathbf{q}) e^{-i\mathbf{q}\cdot\mathbf{r}_{n}}$$

$$A^*(\mathbf{q})A(\mathbf{q}) = \sum_m \left(\sum_n F_n^* F_{n+m}\right) e^{-i\mathbf{q}\cdot\mathbf{r}_m}$$

 $\phi_n = F_n - \langle F_n 
angle$  Deviation from the Average Structure Factors

#### Intensity obtained in the case of a disordered crystal

$$I_{tot}(q) = I_B(q) + I_{DD}(q) + I_S(q)$$

$$I_{B}(\mathbf{q}) = |\langle F \rangle|^{2} \sum_{hkl} \frac{|\Sigma(\mathbf{q} - \mathbf{Q}_{hkl})|^{2}}{v^{2}} \longrightarrow \text{Diffraction term}$$

$$I_{DD}(\mathbf{q}) = \sum_{m} N(m) \langle \phi_{0}^{*} \phi_{m} \rangle e^{-i\mathbf{q} \cdot \mathbf{r}_{m}} \longrightarrow \text{Diffuse scattering term}$$

$$I_{S}(\mathbf{q}) = \sum_{m} N(m) \Delta_{m} e^{-i\mathbf{q} \cdot \mathbf{r}_{m}} \longrightarrow \text{Speckle term}$$

#### Fourier transform of fluctuations

Produces interference on the diffracted signal, called speckles.

Visible with a coherent beam, if the temporal fluctuations are slow compared to the acquisition time.

![](_page_35_Picture_6.jpeg)

#### Several Ways to Exploit X-ray Coherence

![](_page_36_Figure_1.jpeg)

A.S. Poulos, J. Chem. Phys. 132, 091101 (2010)

#### Effect of a Single Dislocation on Coherent Bragg Diffraction

![](_page_37_Figure_1.jpeg)

#### Dislocation Loops in a Specific Si Sample

![](_page_38_Picture_1.jpeg)

![](_page_38_Picture_2.jpeg)

![](_page_38_Picture_3.jpeg)

Prismatic loop

#### Sample preparation:

Growth under  $O_2$ Annealing 35h under  $O_2$ Defect Concentration :  $10^{18}$  cm<sup>-3</sup>

![](_page_38_Figure_7.jpeg)

# Direct Imaging of the Dislocation Line Using a Coherent Beam

![](_page_39_Picture_1.jpeg)

![](_page_39_Picture_2.jpeg)

Resolution: 5µm

#### Revealing a Large-Scale Dissociation

![](_page_40_Figure_1.jpeg)

V. Jacques et al., PRL 107, 199602 (2011)

# Lensless Imaging using coherent x-rays

Principle: retrieve the phase lost during the measurement ... Several techniques...

![](_page_41_Picture_2.jpeg)

These techniques can be used:

- in transmission near the reciprocal space origin : imaging local density
- In diffraction at wide angles: probing local displacement field

$$A(\mathbf{q}) = \sum_{n} F_{n}(\mathbf{q})e^{-i\mathbf{q}\cdot\mathbf{r}_{n}} \qquad r_{n} = r_{0} + u_{n} \qquad A(q) = \sum_{n} F_{n}(q)e^{-iq.r_{0}}e^{iq.u_{n}}$$
Phase

# X-ray Holography on a system having magnetic domains (CoPt)

![](_page_42_Picture_1.jpeg)

### Lensless imaging of magnetic nanostructures by X-ray spectro-holography

S. Eisebitt $^1$ , J. Lüning $^2$ , W. F. Schlotter $^{2,3}$ , M. Lörgen $^1$ , O. Hellwig $^{1,4}$ , W. Eberhardt $^1$  & J. Stöhr $^2$ 

![](_page_42_Picture_4.jpeg)

Resolution : 50nm

Éisebitt et al., Nature432, 885 (2004)

#### X-ray Holography in Bragg condition

![](_page_43_Figure_1.jpeg)

#### Phase Retrieval Algorithms for CDI

Recover the phase lost during the measurement using an algorithm that performs Fourier Transforms (FT) and Inverse Fourier Transforms (FT<sup>-1</sup>) in a loop, until it converges to a solution, applying constraints in both real space and Fourier space.

Coherent Diffraction Imaging (CDI): phase retrieval algorithm

![](_page_44_Figure_3.jpeg)

#### First Implementation of Forward CDI

Fourier space constraints real space constraints  $g_i(\mathbf{x}) \xrightarrow{\mathcal{F}} G_i(\mathbf{k}) = |G| e^{i\phi} \longrightarrow G'_i(\mathbf{k}) = |F(\mathbf{k})| e^{i\phi} \xrightarrow{\mathcal{F}^{-1}} g'_i(\mathbf{x}) \longrightarrow g_{i+1}(\mathbf{x})$ 

- The error can only decrease... or stay the same.
- J. R. Fienup Optics Letters 3 27-29 (1978)
- J. Miao, D. Sayre and H. N. Chapman JOSA A 15 1662-1669 (1988)

![](_page_45_Figure_5.jpeg)

![](_page_45_Figure_6.jpeg)

### **CDI in Bragg Condition: Access to the Strain Field**

# Three-dimensional mapping of a deformation field inside a nanocrystal

Mark A. Pfeifer<sup>1</sup><sup>†</sup>, Garth J. Williams<sup>1</sup><sup>†</sup>, Ivan A. Vartanyants<sup>1</sup><sup>†</sup>, Ross Harder<sup>1</sup> & Ian K. Robinson<sup>1</sup><sup>†</sup>

![](_page_46_Figure_3.jpeg)

Sensitivity to strain  $\Delta \phi = \mathbf{k}_{f} \cdot \mathbf{u} - \mathbf{k}_{i} \cdot \mathbf{u} = \mathbf{Q} \cdot \mathbf{u}$ 

![](_page_46_Picture_5.jpeg)

Nature 442, 63 (2006)

#### A Dislocation Loop Appearing Under Strain

![](_page_47_Figure_1.jpeg)

Applied Force : 560 nN

![](_page_47_Figure_3.jpeg)

M. Dupraz et al., Nano Letters 17, 6696 (2017)

![](_page_48_Picture_0.jpeg)

![](_page_48_Picture_2.jpeg)

ISSN 1600-5767

Signature of dislocations and stacking faults of facecentred cubic nanocrystals in coherent X-ray diffraction patterns: a numerical study

Maxime Dupraz,<sup>a,b</sup>\* Guillaume Beutier,<sup>a,b</sup> David Rodney,<sup>a,b,c</sup> Dan Mordehai<sup>d</sup> and Marc Verdier<sup>a,b</sup>

![](_page_48_Figure_6.jpeg)

# Ptychography: lensless imaging on 'big' samples

'big' = larger than the coherent beam used...

![](_page_49_Figure_2.jpeg)

The CDI algorithm is applied to each image taken at each position. Real space constraint: overlap between positions.

#### Tomographic Ptychography on a Mouse Bone

![](_page_50_Picture_1.jpeg)

Dierolf et al., Nature 436, 467 (2010)

#### Bragg Ptychography and Strain Field

![](_page_51_Figure_1.jpeg)

#### Strain field around dislocations !

Takahashi et al., PRB 87, 121201 (2013)

# Study of the Temporal Dynamics of Speckles: XPCS (X-ray Photon Correlation Spectroscopy)

![](_page_52_Figure_1.jpeg)

#### New opportunities offered by next-generation sources

- Introduction: What are the key quantities to consider for coherence?
- Coherence: From the source point to the detector
- Current use of coherent X-rays
- New opportunities offered by next-generation sources
  - Coherence and nanofocusing
  - Next-generation synchrotron sources: ultimate storage rings
  - Coherence and time-resolved measurements at XFELs

![](_page_54_Figure_0.jpeg)

![](_page_55_Picture_0.jpeg)

Circumference	528 m
No. of long straight sections / no. of available ID straights	20/19
njection	Full-energy top-up from MAX IV Linac
Stored current	500 mA
lorizontal emittance	~200 - 330 pm rad (depending on ID gap settings)
/ertical emittance	2 - 8 pm rad (depending on user demand)
ypical horizontal beam size at ID center	42 - 54 micron (depending on horizontal emittance)
ypical horizontal beam divergence at ID center	4.7 - 6.1 urad (depending on ID gap settings)
ypical vertical beam size at ID center	2 - 4 micron (depending on choice of vertical emittance)
vpical vertical beam divergence at ID center	1 - 2 urad (depending on choice of vertical emittance)

# **Machine characteristics**

- Very low emittance (divided by ~10)
- Very small source sizes (divided by 10)
- Brighter sources

# Techniques

# COHERENCE

# NANOFOCUSING

# **TIME-RESOLVED**

## Propriétés combinables pour explorer la matière de manière inédite!

![](_page_56_Figure_1.jpeg)

#### Coherent X-ray Nanodiffraction Setup

![](_page_57_Figure_1.jpeg)

Enders, Thibault, Proc. R. Soc. A 472: 20160640 (2016)

# Reconstruction of the Phase and Amplitude Profile of a Focused Beam by Ptychography

Astigmatism Correction, Measurement of the Size, and Phase Profile of the Focused Beam

![](_page_58_Figure_2.jpeg)

#### Single InAs Nanowires Probed by Coherent Nanodiffraction

![](_page_59_Figure_1.jpeg)

![](_page_59_Figure_2.jpeg)

Diaz A. et al., PRB 79, 125324 (2009)

# Free-Electron X-ray Lasers: Naturally Coherent Sources with Ultrafast Pulses

![](_page_60_Picture_1.jpeg)

**Complementary** to Synchrotrons for Specific Studies

Advantages: Brightness, fs Temporal StructureDisadvantages: Stability, Sample Damage, Limited Access

#### Combining coherence, nanodiffraction, and time-resolution at XFELs...

![](_page_61_Picture_1.jpeg)

Ultrafast Three-Dimensional Imaging of Lattice Dynamics in Individual Gold Nanocrystals J. N. Clark *et al. Science* **341**, 56 (2013); DOI: 10.1126/science.1236034

![](_page_61_Figure_3.jpeg)

... Imaging an acoustic phonon in a gold nanocrystal

![](_page_62_Figure_1.jpeg)