# **Microscopy with Coherent X-Rays**

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Broad field of applications:

>Physics, chemistry, biology, earth- and environmental science, nano-technology, ...

>Main advantage: large penetration depth

in-situ and in-operando studies

>X-ray analytical contrasts: XRD, XAS, XRF, ...

atomic scale resolution

Today: "mesoscopic gap"

real-space resolution: down to about 10 nm XRD and XAS: atomic scale

What happens on (mesoscopic) 1 - 10 nm scale?



#### quantum dots



M. Hanke, et al., APL **92**, 193109 (2008)





Today: "mesoscopic gap"

real-space resolution: down to about 10 nm

XRD and XAS: atomic scale

What happens on (mesoscopic) 1 - 10 nm scale?

Many interesting physics and chemistry questions:

investigate local states:

- > individual defects (0D): changes in electron density, charge ordering
- > (structural) domain boundaries (2D), e.g., in multiferroics
- > mesoscopic dynamics at (solid-state) phase transitions
- > catalytic nanoparticles (activity in operando)

>...

Mesoscale also very important for nanotechnology (e.g., defects in devices)!



## **Current State of X-Ray Microscopy**

#### Conventional x-ray microscopy

optics limit spatial resolution: diffraction limit



(typically: a few tens of nanometers)

optics are technology limited! Theoretical extrapolation of x-ray optical performance to the atomic level.

[PRB 74, 033405 (2006); H. Yan, et al., PRB 76, 115438 (2007)]

Coherent x-ray imaging techniques (CXDI, ptychography)

→ no imaging optic!

— limited by statistics of far-field diffraction patterns ...

highest resolution: a few nanometers, focusing coherent beam [PRL 101, 090801 (2008); Y. Takahashi, et al., PRB 80, 054103 (2009); A. Schropp, et al., APL 100, 253112 (2012)]





X-rays are electromagnetic radiation

propagation is described by homogeneous Maxwell equation.

Simplification: scalar wave field (small angles, paraxial)

$$\nabla^2 \varphi - \frac{1}{c^2} \frac{\partial^2 \varphi}{\partial t^2} = 0, \quad \varphi: \text{ scalar wavefield}$$

Monochromatic radiation: Helmholtz equation

$$\nabla^2 \psi_{\omega}(\vec{r}) - k^2 \psi_{\omega}(\vec{r}) = 0, \quad \varphi(\vec{r}, t) = \psi_{\omega}(\vec{r}) \cdot e^{i\omega t}, \quad k = \frac{\omega}{c}$$

Polychromatic radiation is superposition of different frequencies  $\boldsymbol{\omega}$ 



Helmholtz equation

$$\nabla^2 \psi_{\omega}(\vec{r}) - k^2 \psi_{\omega}(\vec{r}) = 0, \quad \varphi(\vec{r}, t) = \psi_{\omega}(\vec{r}) \cdot e^{i\omega t}, \quad k = \frac{\omega}{c}$$

can be solved with appropriate boundary conditions using Green's theorem (Born & Wolf, pp. 417-425).

Boundary conditions:





#### **Free Propagation of X-Rays**

Particular solution is the Fresnel-Kirchhoff integral: (Propagation mainly along the *z* direction)



X-rays interact with electrons in sample. Sample can be described by complex index of refraction:

$$n(x, y, z) = 1 - \delta(x, y, z) + i\beta(x, y, z)$$

Thin sample:

No multiple scattering (1. Born approximation)

$$\psi_{\Delta z}(\vec{x},\omega) = O_{\Delta z}(\vec{x}) \cdot \psi_0(\vec{x},\omega)$$

With the transmission function

$$O_{\Delta z}(\vec{x}) = e^{ik \int n \, dz} = e^{ik\Delta z} \cdot e^{-ik \int \delta \, dz} \cdot e^{-k \int \beta \, dz}$$







#### object: Cu



thickness: >frame: 2 μm >thin stripes: 0.5 μm

energy: 20 keV

#### 15 µm

characteristic distance:  $d^2/\lambda = (30 \ \mu m)^2/0.62 \ \text{\AA} = 14.5 \ m$ 





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### **Fraunhofer Diffraction: Far Field**

#### Extreme far field:

$$r = r_0 - \frac{x}{r_0}x' - \frac{y}{r_0}y'$$

with

$$r_0 = \sqrt{z^2 + x^2 + y^2}$$



$$\begin{split} \psi_z(\vec{x},\omega) &= -\frac{ie^{ikr_0}}{2\lambda r_0} \int \psi_0(\vec{x}',\omega) \cdot \exp\left\{-ik\frac{xx'+yy'}{r_0}\right\} dx'dy' \\ &= -\frac{ie^{ikr_0}}{2\lambda r_0} \int \psi_0(\vec{x}',\omega) \cdot \exp\left\{-i\vec{\xi}\vec{x}'\right\} dx'dy', \quad \vec{\xi} = \frac{k}{r_0} \begin{pmatrix} x\\ y \end{pmatrix} \end{split}$$

cos in inclination vanishes due to projection on sphere!

Far field is Fourier transform of wave field in initial plane!



Example: (E = 8 keV) Au (150 nm) on Si<sub>3</sub>N<sub>4</sub>:

t μm

$$\frac{(3 \ \mu \text{m})^2}{1.55 \ \text{\AA}} = 58 \ \text{mm} = z$$

far-field

image



 $L_2 = 3.1 \text{ m}$ 



#### Coherence

So far, we considered the propagation of pure states only! The ideal experiment:

>prepare probe (x-ray beam) in well defined initial state:

 $\psi_{\omega}(\vec{x},0)$  unimodal source

>propagate through **fixed** experimental setup

$$\psi_{\omega}(\vec{x}, \det) = \int P(\vec{x}, \vec{x}'') \psi_{\omega}(\vec{x}'', 0) \ d^2 x''$$

Also sample static and in well-defined state!

>detection:

probability to detect photon ~  $|\psi_{\omega}(\vec{x}, \det)|^2$ 

Repeat experiment many times under identical conditions!!



#### Coherence

Main problem:

We have no full control over the experiment:

>initial state not fully determined: ensemble of initial states that are realized statistically:

 $\psi_{\omega_i}(\vec{x}, 0)$  from statistical ensemble with density  $\rho_i$ 

>experimental setup varies in time

$$\psi_{\omega_{(i,j)}}(\vec{x}, \det) = \int P(\vec{x}, \vec{x}'', t_j) \psi_{\omega_i}(\vec{x}'', 0) \ d^2 x''$$

dynamics of sample and setup described by *j*.

time dependence of sample (dynamics)!

>detection:

$$I_{\det}(\vec{x}) = \sum_{i,j} \rho_{(i,j)} \cdot |\psi_{\omega_{(i,j)}}(\vec{x}, \det)|^2$$

ensemble average under assumption of ergodicity!



#### Coherence

Main problem:

What information can be extract from

$$I_{\det}(\vec{x}) = \sum_{i,j} \rho_{(i,j)} \cdot |\psi_{\omega_{(i,j)}}(\vec{x}, \det)|^2$$

about the experiment! Can we deduce

$$\psi_{\omega_{(i,j)}}(ec{x},\det)$$
 and  $ho_{(i,j)}$  ?

If the states  $\psi_{\omega_{(i,j)}}(\vec{x}, \det)$  are very different, e. g., when the initial state is badly prepared, this becomes more and more difficult!

coherence:

visibility of interference: how much does  $I_{det}(\vec{x})$  resemble the probability density  $|\psi_{\omega_{(i,j)}}(\vec{x}, \det)|^2$ ?



Visibility of interference:

how much does  $I_{\det}(\vec{x})$  resemble the probability density  $|\psi_{\omega_{(i,j)}}(\vec{x},\det)|^2$  ?

States  $\psi_{\omega_{(i,j)}}(\vec{x}, \det)$  must be sufficiently similar that the interference fringes are not washed out!

Influence of the source

>transverse coherence

spatial differences of  $\psi_{\omega_{(i,j)}}(\vec{x}, det)$  as function of *i*.

>longitudinal coherence

energetic differences of  $\psi_{\omega_{(i,j)}}(\vec{x}, \det)$  as function of *i*.



### Example: speckle size varies inversely with object size





#### **Transverse Coherence**



Interference observable:

$$\frac{\lambda}{d} > \frac{D}{L_1} \qquad \qquad \frac{\lambda}{d} : \text{angle between two speckles as seen from sample} \\ \frac{D}{L_1} : \text{angle of source as seen from sample}$$

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#### **Transverse Coherence**



Transverse coherence length:

$$l_t = \frac{\lambda L_1}{D} \longrightarrow d < l_t$$
: interference visible



## Example: Transverse Coherence Length





### **Example: Transverse Coherence Length**





#### **Longitudinal Coherence: Monochromaticity**

Wave packet generated from superposition of monochromatic waves:





#### **Longitudinal Coherence: Monochromaticity**

Difference in length of different paths through sample must not exceed  $I_{\rm l}$ :



reduction of longitudinal coherence destroys visibility of interference at large momentum transfer!



#### **Example: Longitudinal Coherence Length**



$$\frac{\Delta\lambda}{\lambda} = 0.12$$





## **Lensless Imaging: Coherent Diffraction Imaging**





#### **Reconstruction by Phase Retrieval**



Difficulty: no direct image

Diffraction pattern: intensity (phase of wave field lost)



#### What Information is Coded in Phase and Modulus?





### What Information is Coded in Phase and Modulus?

Amplitudes: square root of diffraction pattern





## What Informa

Phase: modulus of (shown: real part

phases are most important!



Is diffraction pattern unique?

>NO! Half of the information is missing! (complex amplitudes vs. real diffraction pattern)

What additional information can be used to help solve the problem?

>Knowledge of part of the object:



Example:

- Object confined to certain area! (rest of image empty) Support constraint!
- >Object must be positive: positive electron density [small (real) object]
- Number of unknowns reduced below the number of measured data!





2k x 2k detector Large pixel and large field of view: number of unknowns too large!



#### How to Obtain a Large Support?



2k x 2k detector

Small pixel and small field of view:

very few unknowns (a lot of redundant information)!



#### How to Obtain a Large Support?



2k x 2k detector pixel half the size as in first example: sufficiently few unknowns & high information content!
## **Right Information Content: Oversampling**



important result:

in order for the diffraction pattern to contain sufficient information to recover the real and imaginary part of the field amplitude, the speckles have to be nicely resolved:

appropriate sampling:

oversampling



No!! Object can be arbitrarily shifted!

Both objects yield same diffraction pattern!





### **Is Solution Now Unique?**



Diffraction pattern nearly point symmetric! transmission function: >real: pure absorption FT symmetric >imaginary: pure phase object FT anti-symmetric

In both cases:

Diffraction pattern has point symmetry!



No!! Object and its point inversion are both solutions

Both objects yield same diffraction pattern!





No!! Object and its point inversion are both solutionsBoth objects yield same diffraction pattern!(except for central beam usually covered by beam stop.)



Babinet's theorem!!

### **Iterative Phase Reconstruction**



real-space constraint:

Fourier-space constraint:

> support

> positivity

```
\psi_i'(\vec{q}, \text{obj}) := \frac{\psi_i'(\vec{x}, \text{obj})}{|\psi_i'(\vec{q}, \text{obj})|} \cdot \sqrt{I_{\text{det}}(\vec{q})}
```

>...

R.W.Gerchberg & W.O. Saxton, *Optic* (1972) **35**, 237 J.R. Fienup, *Appl Opt.* (1982). **21**, 2758 R.P. Millane & W.J. Stroud, *J. Opt. Soc. Am.* (1997) **A14**, 568 Christian G. Schroer | Cheiron School 2014 | 29. September 2014 | page 42







### **Reconstruction**







## **Reconstruction: Stagnation**







## **Repeat Reconstruction**

Deviation of model from diffraction pattern:





# **Coherent Diffraction Imaging with Soft X-Rays**

#### Object: Gold on Si<sub>3</sub>N<sub>4</sub>

*****	2322222222	**************************************
antisztett.	****** ******	**************************************
	1µ	m

#### diffraction pattern:



#### light micrograph:



#### reconstruction:



J. Miao, et. al., *Nature* **400**, 342(1999)



### **XFEL: Image and Destroy**





## **XFEL: Image and Destroy**



#### first pulse



second pulse: sample destroyed





## **CXDI of Biological Samples: Mimivirus**





# **Spatial Resolution**

Diffraction pattern of a yeast cell:





# **Nanofocusing Optics**

reflection:

>mirrors (25 nm)

>capillaries

>wave guides (~10 nm)

diffraction:

Fresnel zone plates (~20 nm)multilayer mirrors (7 nm)

>multilayer Laue lenses (16 nm)

>bent crystals

refraction:

>lenses (43 nm, 18 nm)



## **Refractive X-Ray Lenses**

- > first realized in 1996 (Snigirev et al.)
- > a variety of refractive lenses have been developed since
- > applied in full-field imaging and scanning microscopy
- > most important to achieve optimal performance:

parabolic lens shape



## Nanofocus

Large focal length *f*: aperture limited by absorption

$$D_{\rm eff} = 4\sqrt{\frac{f\delta}{\mu}} \propto \sqrt{f}$$

→ minimize 
$$\mu/\delta$$
 (⇒ small atomic number *Z*)  
→  $NA = \frac{D_{\text{eff}}}{2f} \propto \frac{1}{\sqrt{f}}$  (⇒ minimize focal length *f*)





transition to nanofocusing lenses (NFLs)





## Nanofocusing Lenses (NFLs) Made of Silicon



Made at IHM at TU Dresden together with SAW Components.

3136 NFLs on wafer! about 600000 single lenses!

high accuracy, reproducibility







# **Nanofocusing Lenses (NFLs)**



## Nanoprobe: Coherent X-Ray Diffraction Imaging





## **Imaging of Small Objects**





### **Reconstruction**





#### record resolution: 5 nm (1.5·10<sup>6</sup> ph/nm<sup>2</sup>)





diffraction at single gold particle PRL **101**, 090801 (2008). 4 gold particles:

- >fluence 10 x smaller
- >resolution reduced by

 $\sqrt[4]{10} \approx 1.8$ 



## **CXDI of Silver Nanocube**



# Hard X-ray Scanning Microscopy at PETRA III



Microscope:

~98 m from source

different contrasts:

> fluorescence

- > diffraction (SAXS, WAXS)
- > absorption (XAS)
- > ptychography & CXDI

spatial resolution: down to < 50 nm down to < 5 nm (CXDI)

> X-ray energy: 10 - 50 keV











## **Scanning Coherent Diffraction Imaging: Ptychography**

Sample is raster scanned through confined beam
At each position of scan: diffraction pattern is recorded
Overlap in illumination between adjacent points



## **Ptychographic Microscopy**



Experiment at P06:

detector:

Pilatus 300k (172µm pixel size)

sample-detector distance: 2080 mm

exposure time: 1.5 s per point

#### Sample: NTT AT test pattern





## **Ptychography: Reconstruction**



Maiden & Rodenburg, Ultramicroscopy 109, 1256 (2009).



## **Scanning Microscopy: Fluorescence Imaging**



Ta Lα fluorescence

E = 15.25 keV50 x 50 steps of 40 x 40 nm<sup>2</sup> 2 x 2  $\mu$ m<sup>2</sup> FOV exposure: 1.5 s per point





## **Scanning Microscopy: Ptychography**



0.0  
-0.1  
$$-0.2 \boxed{p}$$
  
-0.3  
-0.4

A. Schropp, et al., APL 96, 091102 (2010), S. Hönig, et al., Opt. Exp. 19, 16325 (2011).

*E* = 15.25 keV 50 x 50 steps of 40 x 40 nm<sup>2</sup> 2 x 2 µm<sup>2</sup> FOV exposure: 1.5 s per point detected fluence: 2.75·10<sup>4</sup> ph/nm<sup>2</sup>

A. Schropp, et al., APL 100, 253112 (2012).

#### 50 nm lines and spaces



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-0

-0

## **Scanning Microscopy: Ptychography**



A. Schropp, et al., APL 100, 253112 (2012).

## Scanning Microscopy: Ptychography



A. Schropp, et al., APL 100, 253112 (2012).



## **Contrast in Coherent Diffraction Imaging**

#### Mimivirus (TEM)





M. Seibert, et al., Nature **469**, 78 (2011).

#### Microchip



Only tungsten! Where is the -AI, Si, ... ?



A. Schropp, et al., J. Microscopy, **241**(1), 9 (2011).



## **Resolution and Contrast in Ptychography**



Schropp & Schroer, NJP 12, 035016 (2010).



## Imaging at the X-Ray Free-Electron Laser

#### Linac Coherent Light Source (LCLS at SLAC)



Investigating the dynamics of matter on nanoscale.

- > short, dense electron bunches
- > long undulator
  - ultra short, very intense
     x-ray pulses (10 100 fs)


## **Focusing Hard X-Ray FEL Pulses**



Diffraction limited imaging of source



X-ray optics:

beryllium parabolic refractive x-ray lenses

B. Lengeler, RXOptics

Focusing needed:

- >increase fluence for imaging
  - resolution determined by fluence on sample
- >increase intensity on sample
  - induce non-linear optical effects
- >increase energy deposited in sample
  - create matter in extreme conditions



>...

# Experiment at MEC at LCLS unattenuated beam



### **Experiment at MEC at LCLS**



single shot exposure: 2 exposures per point 384 out of 400 (800) images for reconstruction

A. Schropp, et al., Sci. Rep. 3, 1633 (2013).

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#### **Reconstruction**



### **Nanofocused LCLS Beam Profile**



A. Schropp, et al., Sci. Rep. 3, 1633 (2013). Christian G. Schroer | Cheiron School 2014 | 29. September 2014 | page 79



# **High Resolution X-Ray Imaging at XFEL Sources**

#### Method: magnifying phase contrast imaging



imaging with secondary point source: efficient use of fluence
phase contrast: for quantitative results, phase retrieval required
Alternatively:

Time-resolved diffraction in focus of nanobeam



## **An X-Ray Microscopists Dream**

Quantitive in-situ measurement of physical properties of matter

- >on all relevant length scales
- >on all relevant time scales

Key technology: brilliant, coherent x-rays with time structure

Fusion of real and reciprocal space:

access to all length scales (in principle) from Å to millimeters

Requirement: a lot of coherent light

X-ray free-electron lasers
diffraction limited storage rings







