Foto: Anne Günther

Correlated X-ray photons for incoherent diffraction imaging

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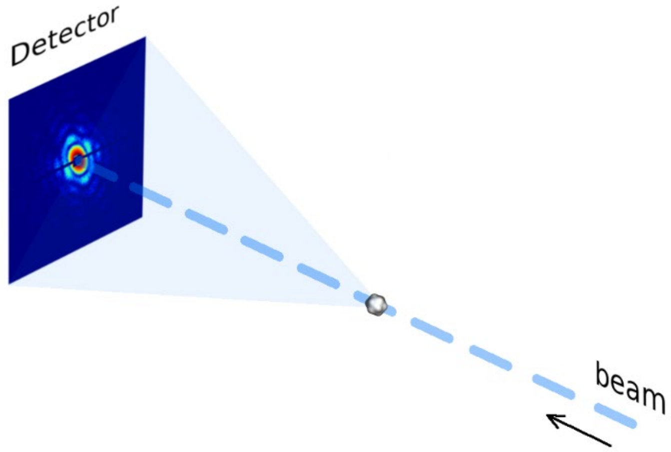
DESY.

Coherent diffractive imaging (CDI)

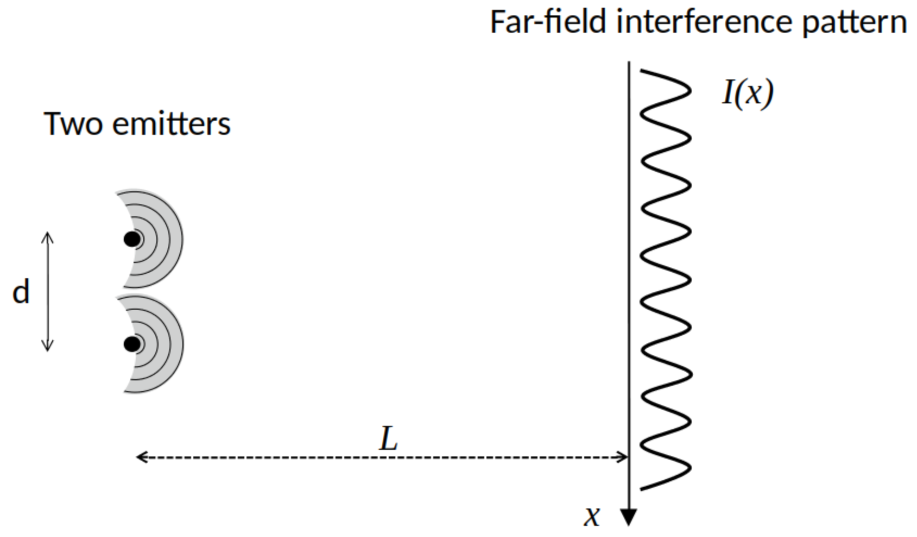
High-resolution structure determination via CDI:

- Fixed phase relation between the incoming and scattered photons is assumed
- First-order coherence of the radiation field is maintained
- Stationary interference pattern of a large number of photons



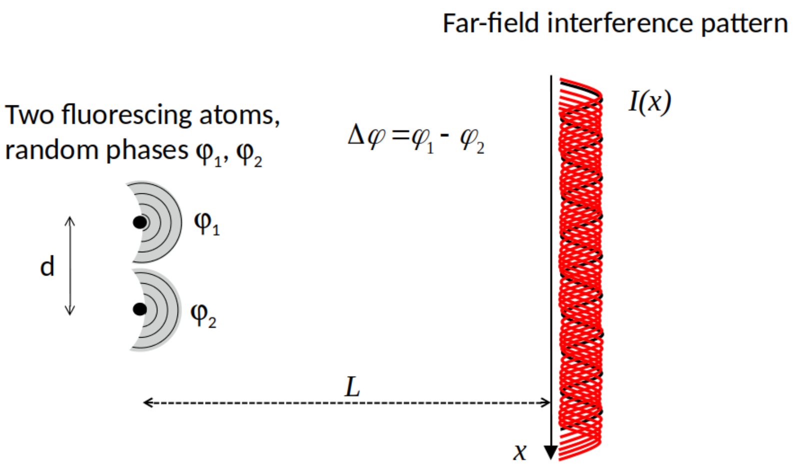
Incoherent processes, sometimes predominant, generate a constant intensity and produce background

Coherent diffractive imaging (CDI) - 2 emitters



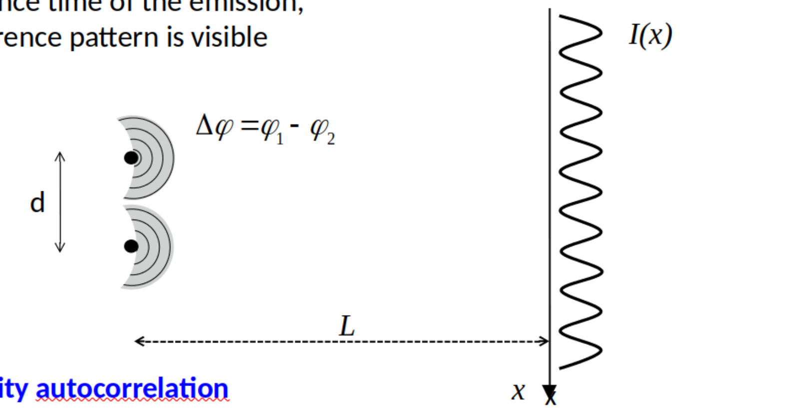
Fringe spacing $l = \frac{L\lambda}{d}$

Incoherent diffractive imaging (IDI) - 2 emitters



Fringes shifted by $l\frac{\Delta\varphi}{\pi}$

However, when recorded within the coherence time of the emission, interference pattern is visible



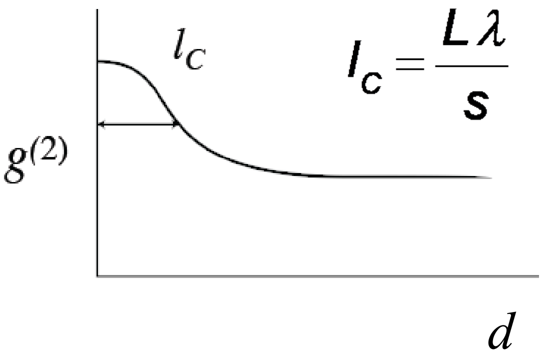
Intensity autocorrelation
is independent of the fringe shift

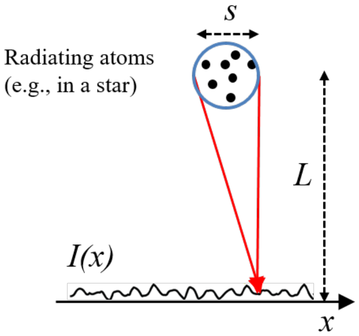
Structural information can be retrieved !

Incoherent diffractive imaging (IDI)

- After pulsed excitation of the sample, incoherently scattered photons are recorded coincidentally at different locations
- Photon correlations of higher order are considered rather than the photon distribution itself

$$g^{(2)}(d) = \frac{\langle I(x)I(x+d)\rangle}{\langle I(x)\rangle^2}$$





Detection time has to be shorter than the fluctuation time (coherence time) – the relative phases of the scattered photons considered as stable – stationary fringe pattern

Pulse duration has to be shorter than the emission time

Imaging with incoherent radiation: milestones

Starlight intensity interferometry

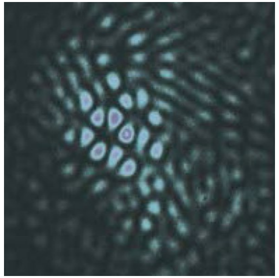
Hanbury Brown and Twiss, Nature (1956)

Quantum theory of optical coherence

Treatment of photon correlations extended to arbitrary orders

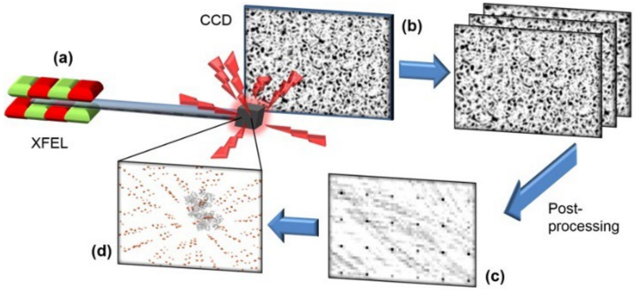
R.J. Glauber, Phys. Rev. (1963)

Various source distributions in 1D and 2D



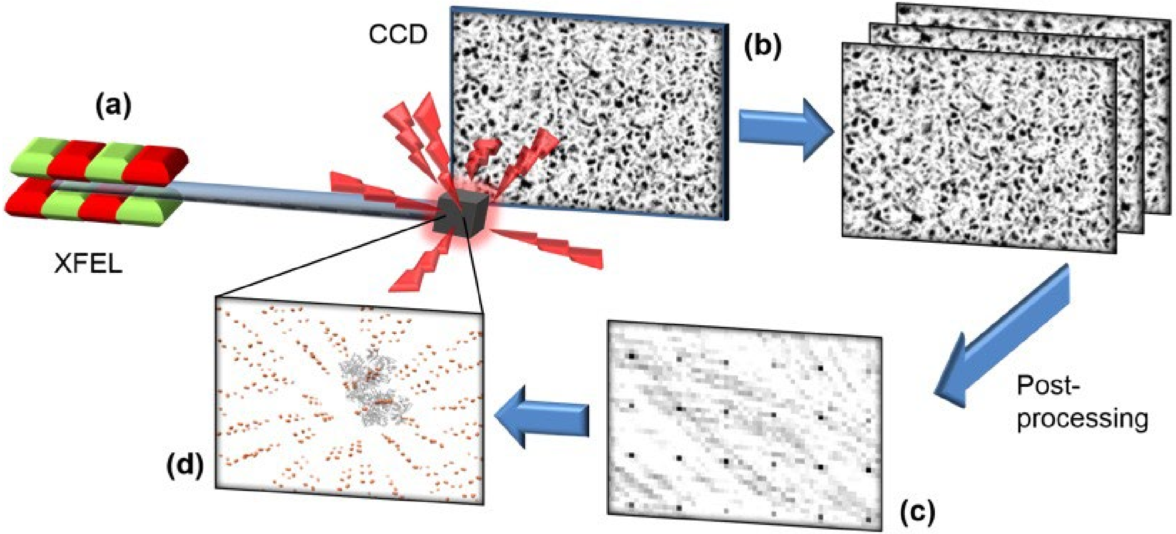
R. Schneider et al., Nat. Phys. (2017)

Potential to reveal full 3D information



A. Classen et al., PRL (2017)

IDI – 3 Dimensions



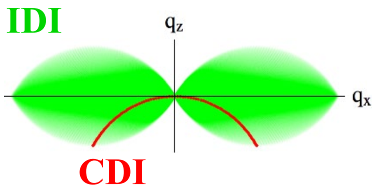
A. Classen et al., PRL (2017)

(a) FEL radiation impinges crystalline sample
(b) CCD records incoherently scattered fluorescence light, a large number of snapshots are accumulated
(c) Calculate photon correlations of each snapshot and average over snapshots – yields source distribution in reciprocal space
(d) Distribution of sources obtained via Fourier transform

IDI – 3 Dimensions

Advantages compared to coherent diffractive imaging (CDI)

- Atomic absorption cross section leading to incoherent fluorescence emission is typically significantly larger than the cross section of coherent scattering – larger signals
- No Bragg peaks, but homogeneous intensity distribution – no large dynamic range of detector needed
- Access to large range of momentum transfers – high spatial resolution already for moderate X-ray photon energies
- Volumetric information in a single image – a few orientations are sufficient
- Element-specific imaging – different species in the crystal can be selectively resolved



Reciprocal space coverage

Correlated X-ray photons

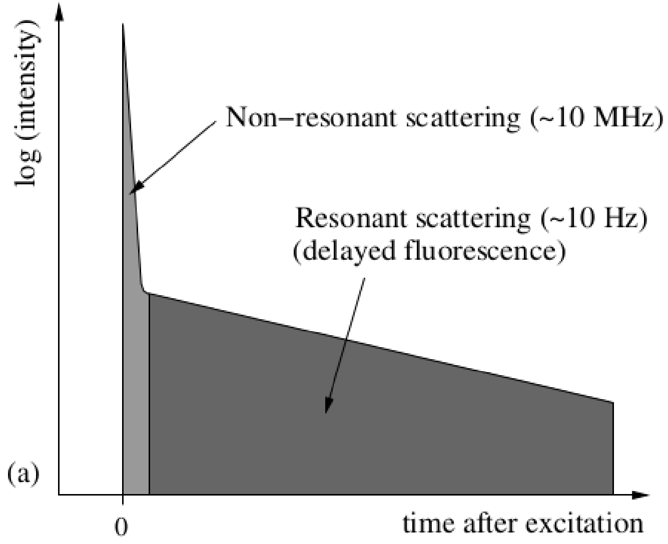
X-ray structure determination:

- Atomic inner-shell fluorescence
  - Structured foils of different materials (e.g. transition metals, Ti to Ge)
  - Nanoparticles or nanocrystals

- Nuclear resonant fluorescence from Mössbauer isotopes

Emission/coherence time		**Pulse duration**	
		XFEL: 5 fs	**Synchrotron:** 50 ps
Atomic fluorescence (Fe K-shell):	1 – 2 fs	●	●
Nuclear fluorescence (^{57}Fe):	100 ns	●	●

IDI – Nuclear resonant fluorescence



R. Röhlsberger, Springer Tracts in Modern Physics 208 (2004)

Mössbauer isotopes: ^{57}Fe or ^{119}Sn

Lifetime (^{57}Fe) = 141 ns

Emission time (^{57}Fe) = 100 ns

Detector for nuclear resonant fluorescence

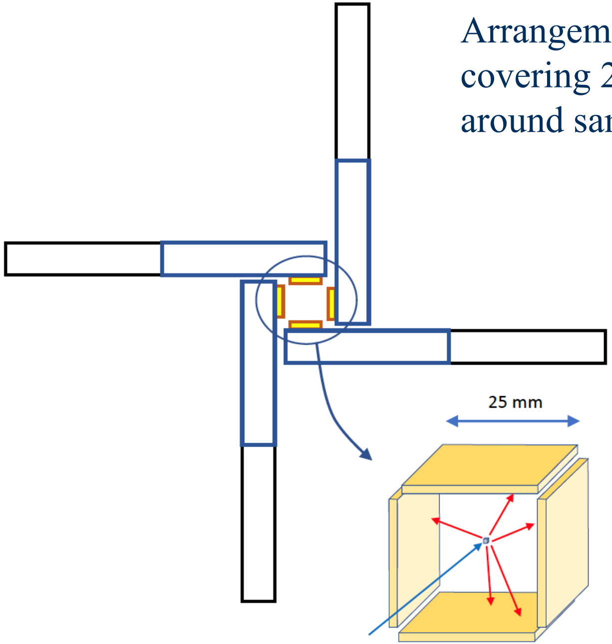
Challenge to discriminate multi-photon events :

- Energy selection
- Timing

Requirements for detector:

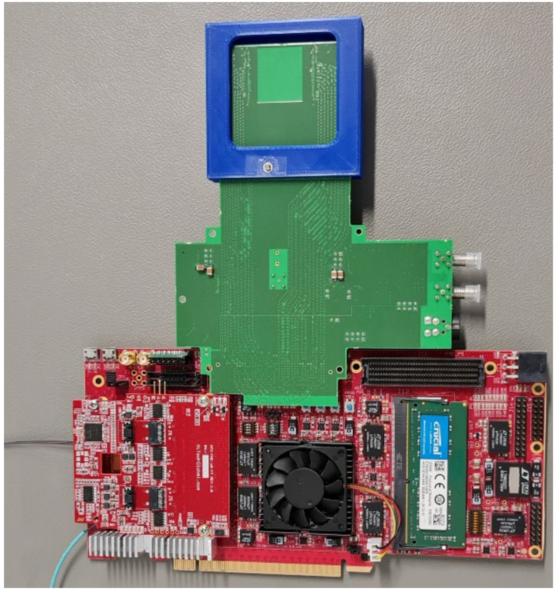
- Energy resolution < 1 keV – no absorber foil needed to block atomic fluorescence photons
- Spatial resolution, pixel size ~50 µm
- Compact shape – coverage of large solid angle
- Event-based detection with ns time resolution – efficient data handling and post-processing

IDI – Nuclear resonant fluorescence radiation from Mössbauer isotopes



Arrangement of 4 modules covering 2/3 of solid angle around sample position

Sample illuminated with sub-µm focused beam



1 module: carrier board with TimePix4 chip and Xilinx Zynq Ultrascale+ board with FPGA fabric

Summary

- **Transfer of the technique from the domain of quantum optics to the field of X-ray structure analysis**
- **New paradigm to obtain structural information on the atomic scale**
- **Crystals or crystallized proteins: the full 3D structure of the sample can be determined with higher resolution compared to conventional CDI**
- **Resonant fluorescence radiation: signal can be efficiently discriminated against non-resonant background – perspective for imaging of metal clusters in active centers of biological macromolecules**

Thank you for your attention!