# Quantum imaging with incoherently scattered X-rays from a Free-Electron Laser

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I. Imaging with coherently diffracted vs. incoherently scattered light:

Young interference, Hanbury Brown Twiss measurement & higher order photon correlations

II. Imaging of arbitrary 1D and 2D source distributions with FLASH at XUV wavelengths using higher order photon correlations

**III. Outlook:** 

Incoherent Diffraction Imaging (IDI) of 3D atomic source distributions with hard X-rays



#### Young's double slit : wave description



Intensity distribution:

$$I(r_1) = \langle E^{\star}(r_1) E(r_1) \rangle = G^{(1)}(r_1)$$

 $I(r_1) \propto 1 + \cos(kd\sin\theta)$ 



Th. Young (1773 – 1829)

Young's double slit : quantum path description





#### Dirac: "Each photon interferes only with itself. Interference between two different photons never occurs."



Young's double slit experiment with single atoms



U. Eichmann et al., PRL 70, 2359 (1993)



FIG. 2. Interference pattern for three different ion separations: (a) 5.4  $\mu$ m, (b) 4.3  $\mu$ m, and (c) 3.7  $\mu$ m. The two white spots are caused by stray reflections of the laser beam.





steady state



Itano et al., PRA 57, 4176 (1998) Skornia, JvZ, Agarwal, Werner, Walther, PRA 64, 063801 (2001)

igstarrow Young's double slit experiment with two atoms in excited state |e,e
angle

spontaneous decay

incoherent emission of radiation



$$|\phi\rangle = \frac{1}{\sqrt{2}} \left( e^{i\Phi_1} |1\rangle |0\rangle + e^{i\Phi_2} |0\rangle |1\rangle \right)$$

 $\longrightarrow$  <u>no interference fringes in G<sup>(1)</sup></u>

two atoms in state |e,e
angle

with 
$$\hat{E}^{(+)}(\vec{r}_j) \sim \sum_{l=1}^2 e^{-i\Phi_{lj}} \hat{s}_l^-$$
 and  $\hat{s}_l^- = |g\rangle_l \langle e|$   
 $G^{(1)}(\mathbf{r}_1) = \langle e, e | \mathbf{E}^{(-)}(\mathbf{r}_1) \mathbf{E}^{(+)}(\mathbf{r}_1) | e, e \rangle \sim const.$ 

- Hanbury-Brown Twiss measurement for two atoms in state  $\ket{e,e}$ 

double star



Hanbury BrownTwiss, Nature 177, 27 (1956) Hanbury BrownTwiss, Nature 178, 1046 (1956) Hanbury Brown, Nature 218, 637 (1968)



#### Photon-photon-correlation of fluorescence light of two atoms



$$G^{(2)}(r_1, r_2) \sim \langle E^-(r_1) E^-(r_2) E^+(r_2) E^+(r_1) \rangle$$

- Hanbury-Brown Twiss measurement for two atoms in state  $\ket{e,e}$ 

$$G^{(2)}(\mathbf{r}_1;\mathbf{r}_2) = \langle e, e | \mathbf{E}^{(-)}(\mathbf{r}_1) \, \mathbf{E}^{(-)}(\mathbf{r}_2) \, \mathbf{E}^{(+)}(\mathbf{r}_2) \, \mathbf{E}^{(+)}(\mathbf{r}_1) | e, e \rangle$$

$$= 1 + \cos[kd(\sin\theta_2 - \sin\theta_1)]$$

two 2-level atoms

Two different quantum paths contribute to same final state:



$$|e,e
angle |0,0
angle igsim |g,e
angle |1,0
angle igsim |g,g
angle |1,1
angle \ igsim |e,g
angle |1,0
angle igsim |1,0
angle$$

interference fringes in G<sup>(2)</sup> expected

Skornia, JvZ, Agarwal, Werner, Walther, PRA 64, 063801 (2001) Agarwal, JvZ, Skornia, Walther, PRA 65, 053826 (2002)

Higher order photon correlation functions:

$$G^{(2)}(\mathbf{r}_1, \mathbf{r}_2) = \langle \hat{E}^{(-)}(\mathbf{r}_1) \, \hat{E}^{(-)}(\mathbf{r}_2) \, \hat{E}^{(+)}(\mathbf{r}_2) \, \hat{E}^{(+)}(\mathbf{r}_1) \rangle$$



R. J. Glauber

Phys. Rev. 130, 2529 (1963); ibid. 131, 2766 (1963) Nobel prize 2005



$$G_N^{(m)}(\mathbf{r}_1,\ldots,\mathbf{r}_m) = \langle \hat{E}^{(-)}(\mathbf{r}_1)\ldots\hat{E}^{(-)}(\mathbf{r}_m)\hat{E}^{(+)}(\mathbf{r}_m)\ldots\hat{E}^{(+)}(\mathbf{r}_1)\rangle$$

#### can be used for improved imaging?

# Quantum imaging with incoherently scattered light from a Free-Electron Laser

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N incoherent light sources (TLS) on a 1D grid with lattice constant d



#### **Detection scheme to determine F in 1D:**

Magic positions for *m*-1 fixed det.

$$\delta_j = 2\pi \frac{j-2}{m-1} \quad \text{for} \quad j = 2, \dots, m$$

$$R_{1}$$

$$R_{2}$$

$$\vdots$$

$$R_{N}$$

$$D_{1}(\delta_{1})$$

$$g_{N \text{ TLS}}^{(4)}(\delta_{1})$$

One moving detector  $\Rightarrow g_N^{(m)}(\delta_1)$ 

#### **Spatial frequency filtering**:

$$g_{N}^{(m)}(\delta_{1}) = A_{0}^{(m)} + \sum_{\substack{a,l=1\\a \neq l}}^{N} A_{la}^{(m)} \cdot \begin{cases} \cos((x_{l} - x_{a}) \cdot \delta_{1}) & \text{if } (x_{l} - x_{a}) \in \mathbb{N}_{0} \cdot (m-1) \\ 0 & \text{else} \end{cases}$$

only particular spatial frequencies appear within a given correlation order m

#### m > 2: resolution below the Abbe limit

Classen, Waldmann, Giebel, Schneider, Bhatti, Mehringer, JvZ, PRL 117, 253601 (2016)



#### Set of spatial frequencies

source geometry can be described on a grid with basis  $\begin{pmatrix} d_x \\ d_y \end{pmatrix} = \begin{pmatrix} \sqrt{3} \\ 1 \end{pmatrix} a$ 

Set of spatial frequencies F consists of 9 different spatial frequency vectors

Knowing F allows to determine the source distribution

## **Detection scheme to determine F:**

m-1 fixed detectors located at magic positions (MP)

along x-axis

$$\delta_{j,x} = 2\pi \frac{j-2}{m-1} \quad \text{for} \quad j = 2, ..., m$$

one moving detector D<sub>1</sub> in the x-y-plane

$$\Rightarrow g_N^{(m)}(\delta_{1,x},\delta_{1,y},MP_x)$$



detector plane

#### **Spatial frequency filtering**:

$$g_{N}^{(m)}(\delta_{1,x},\delta_{1,y},MP_{x}) = A_{0} + \sum_{\substack{a,l=1\\a\neq l}}^{N} A_{la} \cdot \begin{cases} \cos\left[(x_{l} - x_{a})\delta_{1,x} + (y_{l} - y_{a})\delta_{1,y}\right] & \text{if } (x_{l} - x_{a}) \in \mathbb{Z} \cdot (m-1) \\ 0 & \text{else} \end{cases}$$

- For m-1 fixed detectors at MP along x-axis: Filtering of 2D spatial frequencies, depending on correlation order m and x-component of spatial frequency
- Analogue filtering conditions for m-1 fixed detectors at MP along y-axis

#### **Detection scheme to determine F:**





**Detector plane** 

- Measure  $g^{(m)}(\delta_{1,x}, \delta_{1,y}, MP_x)$  for m = 3, 4, ..., with m-1 fixed detectors at MP<sub>x</sub>
- Measure  $g^{(m)}(\delta_{1,x}, \delta_{1,y}, MP_y)$  for m = 3, 4, ..., with m-1 fixed detectors at MP<sub>y</sub>

• Measuring sequentially  $g^{(m)}(\delta_{1x}, \delta_{1y}, MP_x)$  and  $g^{(m)}(\delta_{1x}, \delta_{1y}, MP_y)$  for m > 2 enables determination of complete set of different spatial frequencies F



#### FLASH at DESY, Hamburg



*N* artificial incoherent sources on a 2D-grid (with lattice constant  $d_x$  and  $d_y$ ) using 6 holes arranged in form of a benzene molecule



**REM-image** 



#### **Experimental results:**

#### 10,000 images:





#### Experimental results: Example $g^{(4)}(\delta_{1,x}, \delta_{1,y}, MP_x)$



### **Detection scheme to determine F:**

#### **REM Image**



#### Result for F:

F	$\binom{1}{1}$	$\begin{pmatrix} 1 \\ -1 \end{pmatrix}$	$\binom{2}{0}$	$\binom{2}{2}$	$\binom{2}{-2}$	$\binom{3}{1}$	$\binom{3}{-1}$	$\binom{4}{0}$	$\binom{0}{2}$
<b>F</b> <sub>exp</sub>			$\binom{1.94}{0.02}$	$\binom{1.96}{2.00}$	$\binom{1.97}{-1.95}$	$\binom{2.97}{0.97}$	$\binom{2.97}{-1.03}$	$\binom{3.90}{0.04}$	$\binom{0.01}{1.98}$

Schneider, Mehringer, Mercurio, Wenthaus, Classen, Brenner, Gorobtsov, Benz, Bhatti, Bocklage, Fischer, Lazarev, Obukhov, Schlage, Skopintsev, Wagner, Waldmann, Willing, Zaluzhnyy, Wurth, Vartanyants, Röhlsberger, von Zanthier, Nature Phys. 14, 126 (2018)

#### **Reconstructed Image of Source Distribution:**



#### Result for F:

F	$\binom{1}{1}$	$\begin{pmatrix} 1\\ -1 \end{pmatrix}$	$\binom{2}{0}$	$\binom{2}{2}$	$\binom{2}{-2}$	$\binom{3}{1}$	$\binom{3}{-1}$	$\binom{4}{0}$	$\binom{0}{2}$
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Schneider, Mehringer, Mercurio, Wenthaus, Classen, Brenner, Gorobtsov, Benz, Bhatti, Bocklage, Fischer, Lazarev, Obukhov, Schlage, Skopintsev, Wagner, Waldmann, Willing, Zaluzhnyy, Wurth, Vartanyants, Röhlsberger, JvZ, Nature Phys. 14, 126 (2018)

#### **Reconstructed Image of Source Distribution:**

#### news & views

#### X-RAY IMAGING

#### Incoherent success

The diffraction of coherent X-rays is routinely used to determine the structure of crystals and molecules, and underpinned the discovery of the double-helix structure of DNA in 1953. As the method relies on diffraction and interference it requires the X-rays to be scattered coherently. When this is not the case, the incident and diffracted waves are not in phase, and X-ray imaging methods cannot generate the diffractive patterns needed to reconstruct the arrangement of the atoms in a crystal. This poses a big limit to coherent X-ray diffractive imaging since incoherent scattering predominates in the X-ray domain and much effort is needed to ensure coherence.

Now, this prerequisite no longer stands. Joachim von Zanthier and colleagues have demonstrated that incoherently scattered photons can be used to image tiny, complex structures (*Nat. Phys.* https://dx.doi. org/10.1038/nphys4301; 2017). Specifically, they have shown that incoherently scattered X-rays from a free-electron laser (FEL) can image 2D objects with a spatial resolution close to or even below the Abbe limit. The imaging capability in two dimensions was surprising to the researchers considering the much enlarged parameter space for the possible phase combinations used to determine the



Credit: Macmillan Publishers Ltd

higher-order correlation functions. It allows, for example, imaging of arbitrary 2D objects on a substrate, and potentially transfers the ideas of quantum imaging from visible wavelengths to shorter wavelengths.

The team performed the experiment at the PG2 beamline of the Free-Electron Laser Hamburg (FLASH) at Deutsches Elektronen-Synchrotron (DESY), Hamburg. The FEL beam runs in a 10 Hz pulsed mode at 13.2 nm. It passes a monochromator and impinges on a moving diffusor. The pseudothermal light scattered by the diffusor is used to illuminate an object and the light passing through the object is measured by a chargecoupled device (CCD) image sensor. In the experiment, a 2D object mask, consisting of six square-cut holes in a hexagonal arrangement to mimic the carbon atoms in a benzene molecule, on the micrometre scale, was used to generate six quasimonochromatic independently radiating incoherent sources. The researchers showed that they were able to determine the entire benzene structure based on the 10,800 single-shot speckle patterns (see image) obtained by the CCD detector.

"The requirements for the implementation — high brilliance, ultrashort excitations and high repetition rates — are well met by the FEL facility at DESY. Our next step is to apply the scheme in the hard X-ray regime to reveal structures of crystals, nanoparticles, or even single molecules at the atomic scale," said von Zanthier, who also added that the approach will likely improve structural analyses in biology and medicine.

#### **Rachel Won**

Published online: 22 December 2017 https://doi.org/10.1038/s41566-017-0080-5

#### **Reconstructed Image**



Schneider, Mehringer, Mercurio, Wenthaus, Classen, Brenner, Gorobtsov, Benz, Bhatti, Bocklage, Fischer, Lazarev, Obukhov, Schlage, Skopintsev, Wagner, Waldmann, Willing, Zaluzhnyy, Wurth, Vartanyants, Röhlsberger, JvZ, Nature Phys. 14, 126 (2018)

Workshop "Frontiers of X-rays in the Physical Sciences", Imperial College, London, 13.11.2019

#### News & Views, Nature Photon. 12, 6 (2018)

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**Coherent Diffraction Imaging (CDI)** 

**Coherently diffracted light field** 

Probe



Ferredoxin (PDB: 1FDN)



$$f(\mathbf{q}) = -r_e \int \rho(\mathbf{r}) e^{i\mathbf{q}\cdot\mathbf{r}} \,\mathrm{d}\mathbf{r}$$

 $\mathbf{q} = \mathbf{k}_{\mathrm{out}} - \mathbf{k}_{\mathrm{in}}$  momentum transfer

 $ho({f r})$  electron density



**Coherent Diffraction Imaging (CDI)** 



#### **Coherently diffracted light field**

$$\psi(\mathbf{k}_{\text{out}}) = e^{i\mathbf{k}_{\text{in}}\cdot(\mathbf{r})} + \frac{e^{ikr}}{r}f(\mathbf{q})$$

$$f(\mathbf{q}) = -r_e \int \rho(\mathbf{r}) e^{i\mathbf{q}\cdot\mathbf{r}} \,\mathrm{d}\mathbf{r}$$

 $\mathbf{q} = \mathbf{k}_{\mathrm{out}} - \mathbf{k}_{\mathrm{in}}$  momentum transfer

 $ho({f r})\sim FT^{-1}\{f({f q})$ electron (density  $\cdot^{{f r}}\,{
m d}{f q}$ 





$$G^{(1)}(\mathbf{k}_1) = I(\mathbf{k}_1) \sim const.$$

$$G^{(2)}(\mathbf{k}_1, \mathbf{k}_2) \sim 1 - \frac{2}{N} + |f(\mathbf{q})|^2$$
  
 $\approx 1 + |f(\mathbf{q})|^2$ 

$$\rho(\mathbf{r}) \sim FT^{-1}\{f(\mathbf{q})\} = \int f(\mathbf{q}) e^{-i\mathbf{q}\cdot\mathbf{r}} \,\mathrm{d}\mathbf{q}$$

$$\mathbf{>}$$







#### dose advantage at X-ray energies



Incoherently diffracted light

$$G^{(1)}(\mathbf{k}_1) = I(\mathbf{k}_1) \sim const.$$

$$G^{(2)}(\mathbf{k}_1, \mathbf{k}_2) \sim 1 - \frac{2}{N} + |f(\mathbf{q})|^2$$
  
 $\approx 1 + |f(\mathbf{q})|^2$ 

$$\rho(\mathbf{r}) \sim FT^{-1}{f(\mathbf{q})} = \int f(\mathbf{q})e^{-i\mathbf{q}\cdot\mathbf{r}} \,\mathrm{d}\mathbf{q}$$



dose advantage

Incoherently diffracted light

- volumetric information for a single orientation
  - increased resolution
  - increased statistics

$$G^{(1)}(\mathbf{k}_1) = I(\mathbf{k}_1) \sim const.$$

$$G^{(2)}(\mathbf{k}_1, \mathbf{k}_2) \sim 1 - \frac{2}{N} + |f(\mathbf{q})|^2$$
  
 $\approx 1 + |f(\mathbf{q})|^2$ 

$$\rho(\mathbf{r}) \sim FT^{-1}{f(\mathbf{q})} = \int f(\mathbf{q})e^{-i\mathbf{q}\cdot\mathbf{r}} \,\mathrm{d}\mathbf{q}$$

Classen, Ayyer, Chapman, Röhlsberger, JvZ, PRL 119, 053401 (2017)

**Improvements for Incoherent Diffractive Imaging (IDI)** 

- shorter pulse length of FEL ( < 1 fs)
- high reprate (with detectors keeping up)

- larger detectors

- automatized evaluation (e.g., dropletization)

# **Conclusion and Outlook**

- Photons emitted by incoherent sources can produce diffraction patterns if measured coincidentally in the far-field ( = higher order correlation functions)
- Arbitrary source distribution in 1D and 2D: filtering of spatial frequencies

 $g_N^{(m)}(\delta_1) = A_0^{(m)} + \sum A_{la}^{(m)} \cdot \cos((x_l - x_a) \cdot \delta_1) \quad \text{if} \quad (x_l - x_a) \in \mathbb{N}_0 \cdot (m - 1)$ 

$$g_{N}^{(m)}(\delta_{1,x},\delta_{1,y},MP_{x}) = A_{0} + \sum_{\substack{a,l=1\\a\neq l}}^{N} A_{la} \cdot \begin{cases} \cos\left[(x_{l} - x_{a})\delta_{1,x} + (y_{l} - y_{a})\delta_{1,y}\right] & \text{if } (x_{l} - x_{a}) \in \mathbb{Z} \cdot (m-1) \\ 0 & \text{else} \end{cases}$$

using XUV light from FEL:



PRA 64, 063801 (2001); PRL 99, 133603 (2007); PRL 109, 233603 (2012); PRL 117, 253601 (2016) Nature Physics 14, 126 (2018); News & Views, Nature Photonics 12, 6 (2018)

Incoherent Diffraction Imaging (IDI): 3D structure determination with hard x-rays

Theory: PRL 119, 053401 (2017) Experiment at LCLS in 2018, evaluation ongoing

### The Team:

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# **Collaborations:**

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