

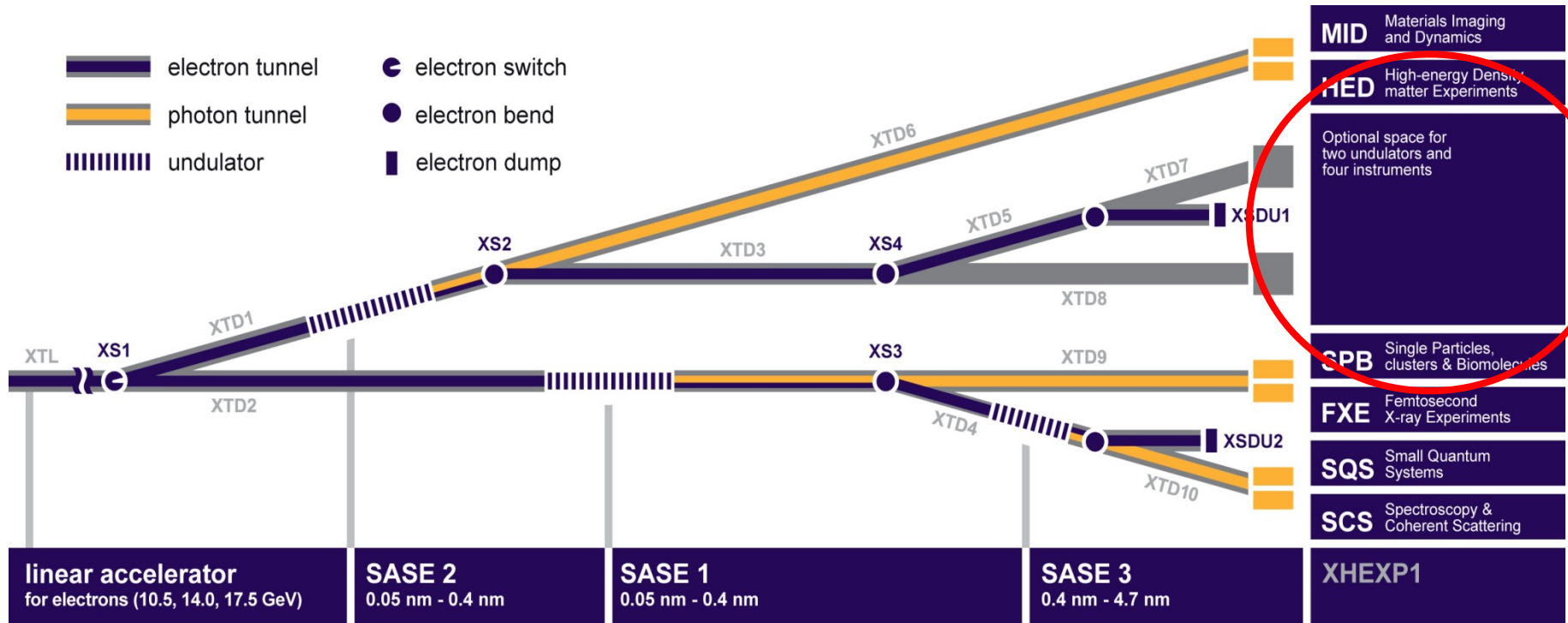
From Coherent to Ghost Imaging at XFEL sources

Ivan Vartanants

*Photon Science, DESY, Hamburg, Germany
NRNU, 'MEPhI', Moscow, Russia*

How we can fill two empty tunnels?

Undulator Segment	FEL radiation energy [keV]	Wavelength [nm]
SASE 1	3 - over 24 (Hard XR)	0.4 - 0.05
SASE 2	3 - over 24	0.4 - 0.05
SASE 3	0.27 - 3 (Soft XR)	4.6 - 0.4



What are coherence properties of X-ray free-electron lasers?

I. A. Vartanyants & A. Singer, "Coherence Properties of Third-Generation Synchrotron Sources and Free-Electron Lasers", Synchrotron Light Sources and Free-Electron Lasers. Editors: E. Jaeschke, S. Khan, J.R. Schneider and J.B. Hastings Springer International Publishing Switzerland (2016), pp.821-863.

Coherence - correlations



Nobel prize: 2005

1-st order correlation function:

$$g^{(1)}(x_1, x_2) = \frac{\langle E(x_1)E(x_2) \rangle}{\langle \sqrt{I(x_1)} \rangle \langle \sqrt{I(x_2)} \rangle}$$

N-th order correlation function:

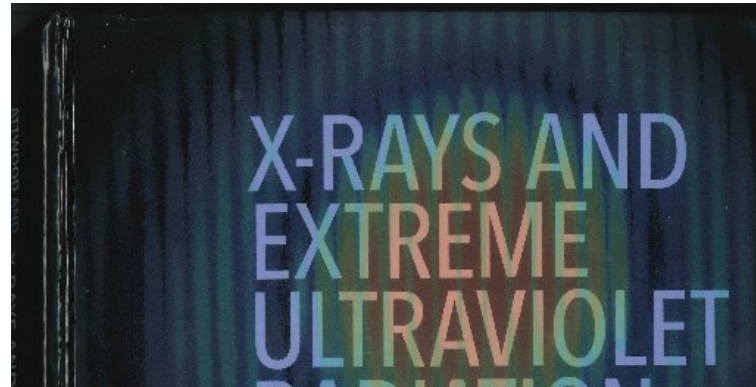
$$g^{(n)}(x_1, \dots, x_n) = \frac{\langle I(x_1) \dots I(x_n) \rangle}{\langle I(x_1) \rangle \dots \langle I(x_n) \rangle}$$

Glauber has formulated:

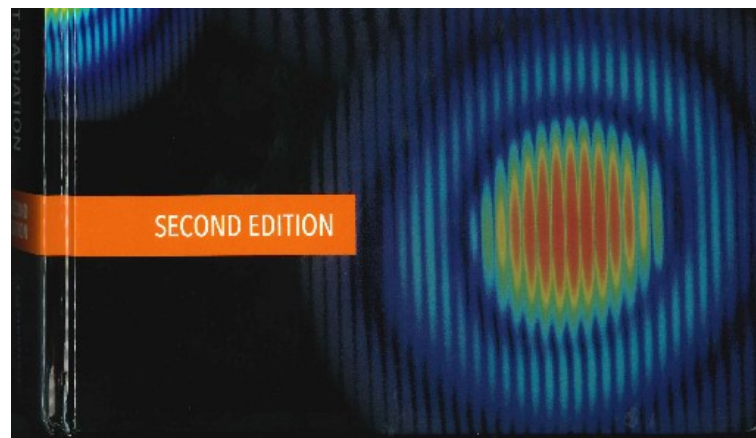
**The source to be coherent, has to be coherent in
all orders of correlation function**



1-st order transverse coherence@XFELs

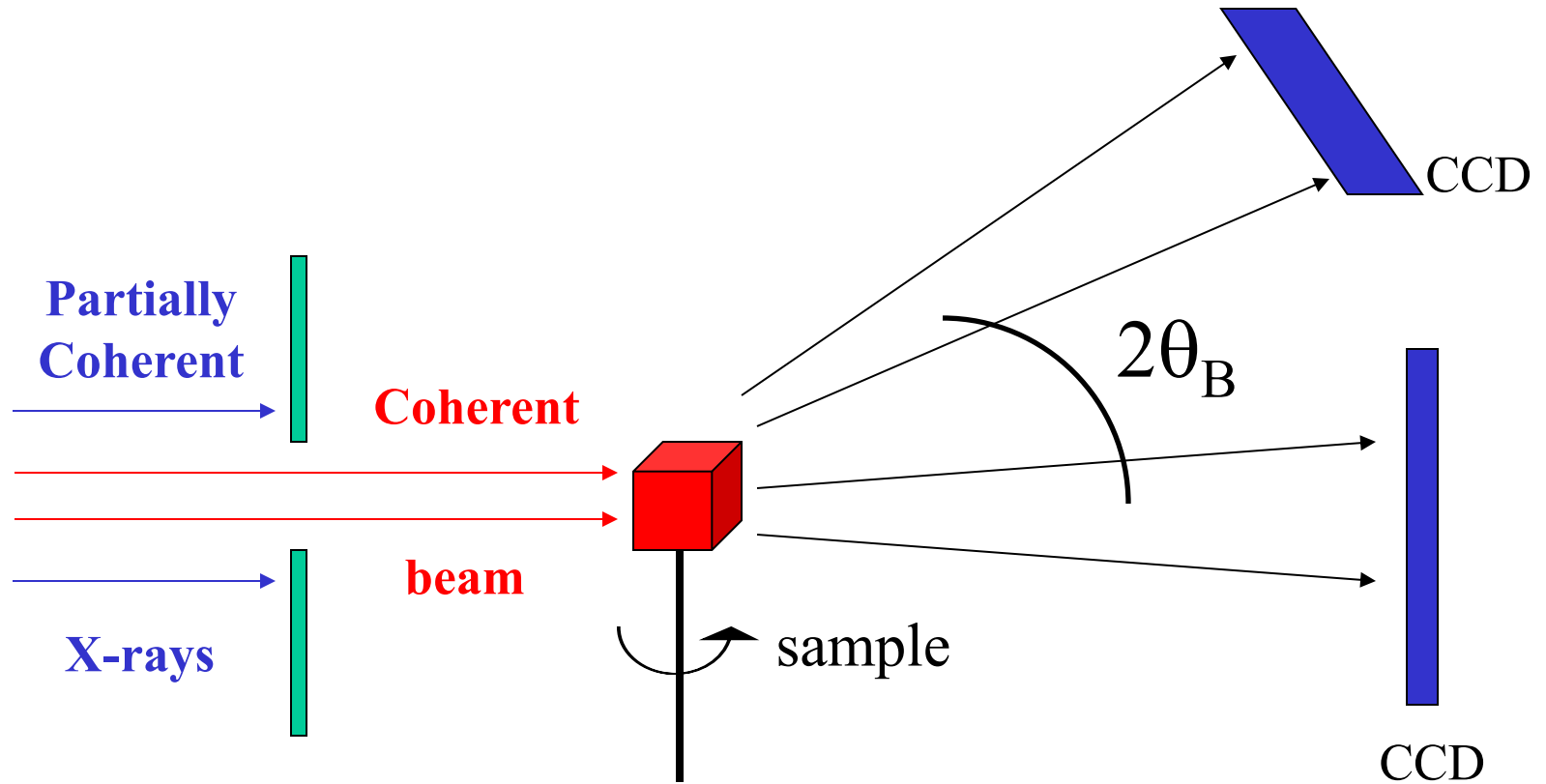


**1st order transverse coherence
at FELs ~ 80-90%**



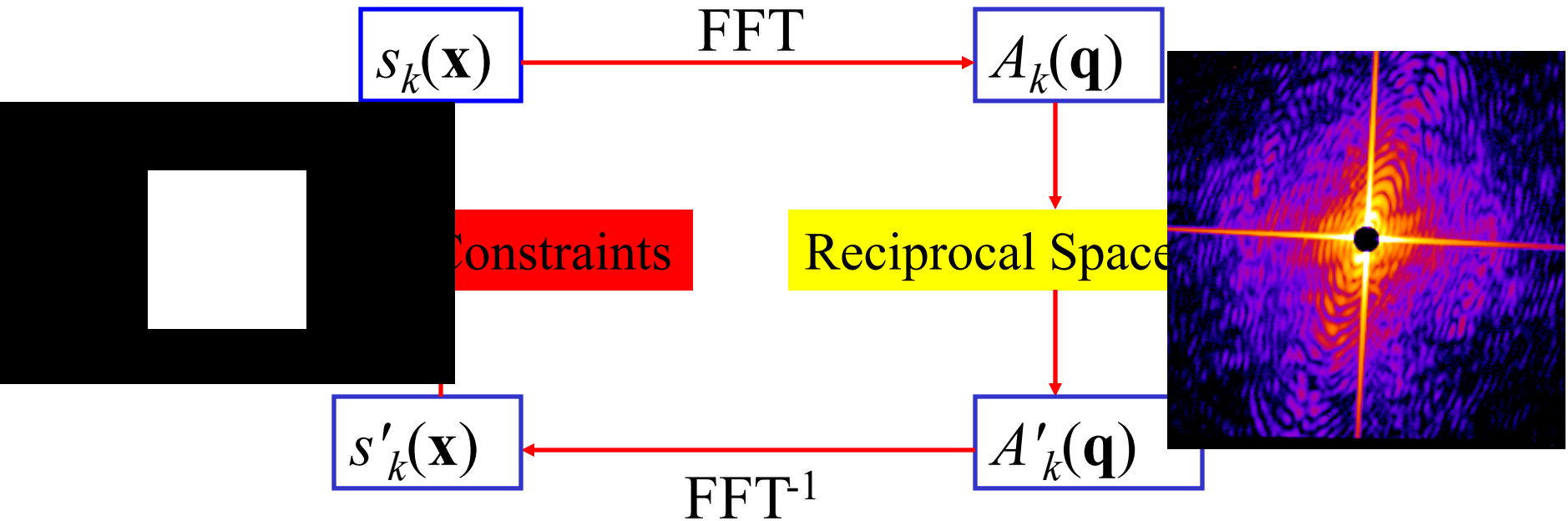
Coherent X-ray Diffraction Imaging

Lensless X-ray Microscopy



Optics: → **Phase Retrieval**

Iterative phase retrieval algorithm



Real space constraints:

use all *a priori* knowledge:

- finite support
- positivity
- ...

Reciprocal space constraint:

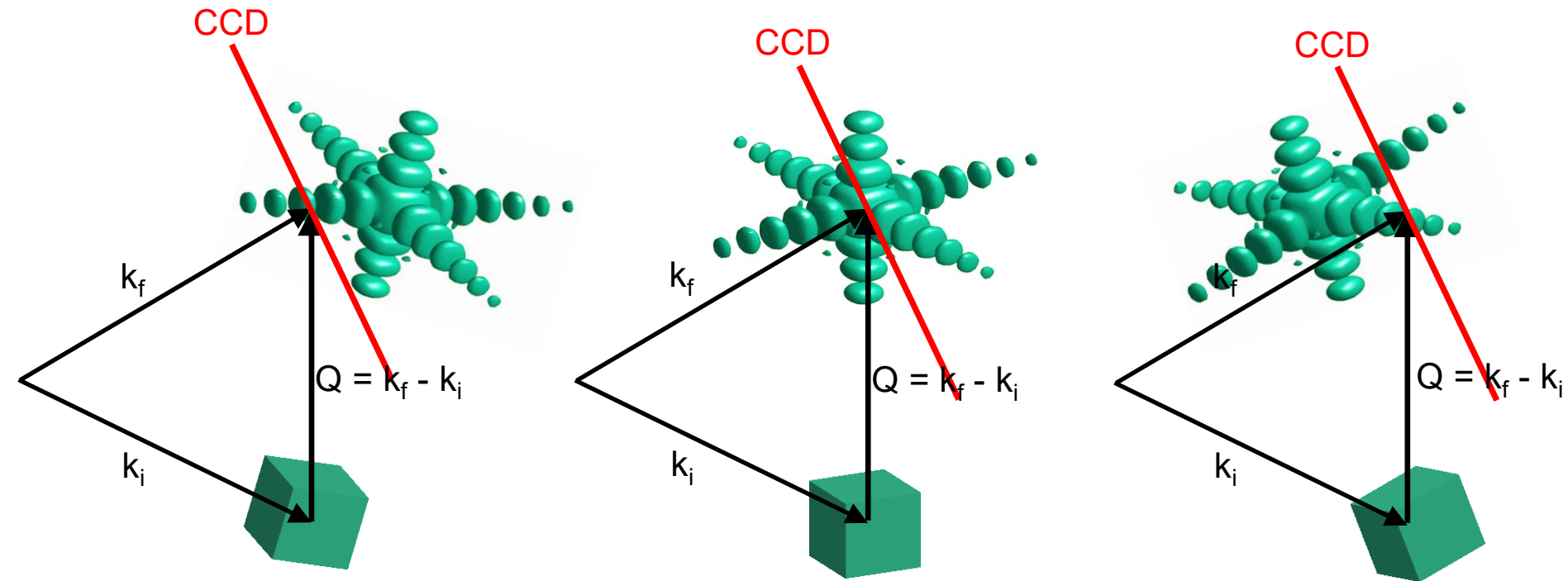
$$|A_k(\mathbf{q})| \rightarrow \sqrt{I_{exp}(\mathbf{q})}$$

R.W.Gerchberg & W.O. Saxton, *Optic* (1972) **35**, 237

J.R. Fienup, *Appl Opt.* (1982) **21**, 2758

V. Elser, *J. Opt. Soc. Am. A* (2003) **20**, 40

Bragg CXDI – 3D Measurements



Change the incidence angle

Catalysis

Collaboration with DESY Nano Lab and A. Stierle group

Catalysis

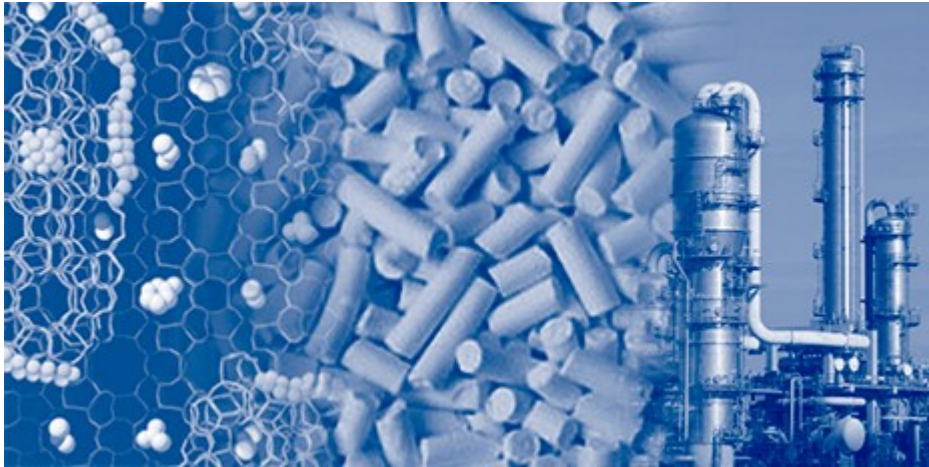


Photo: iStockphoto / collage in ETH Zurich news

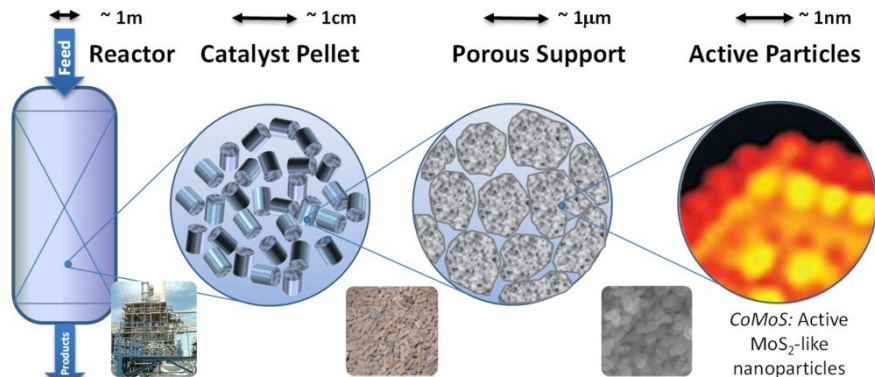
Present Use of Catalysts

Aluminosilicates (Zeolite)
: small hydrocarbons
(e.g. ethylene, propene)

Iron
: Making ammonia

Nickel
: Making synthesis gas
(e.g. carbon monoxide and hydrogen)

Platinum
: Converter
(e.g. carbon monoxide to carbon dioxide)



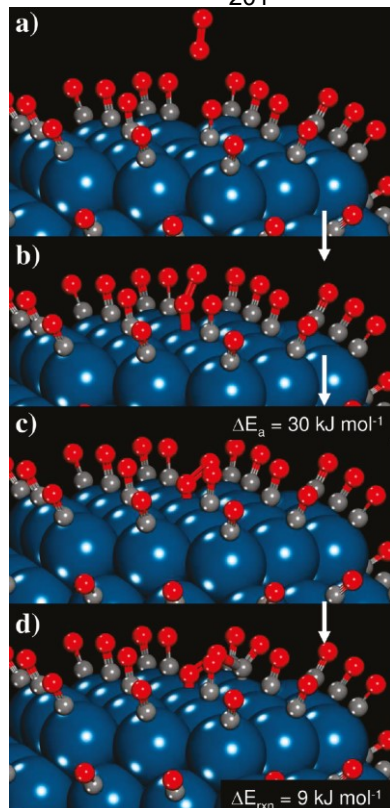
<http://www.catalysiscluster.eu/>

Catalysis as a key technology for Europe

Catalyst : Platinum

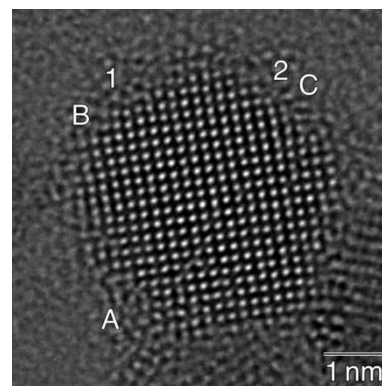
To understand strain on atomic length scales

DFT-calculated reaction on the (111) terrace of Pt₂₀₁ cluster

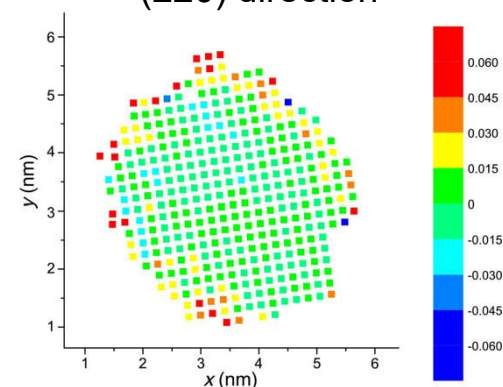


Ayman D. Allian et al. *JACS*. 133 (2011) 4498.

HRTEM

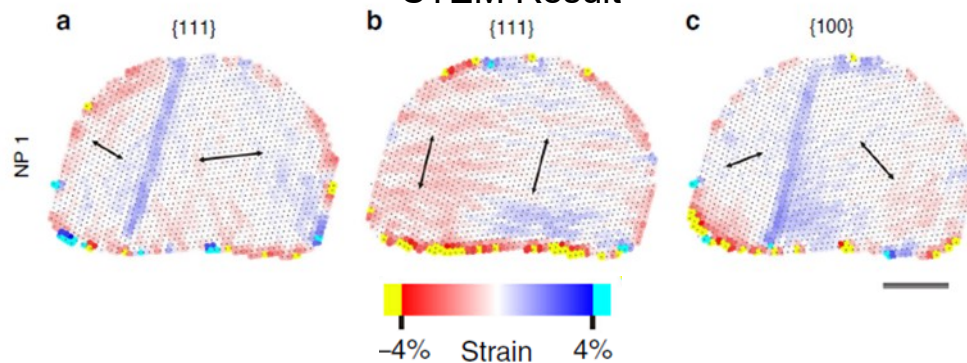


Map of elastic strain along the (220) direction



K. Du et al. *Acta Materialia* 58 (2010) 836–845.

STEM Result



T. N. Pingel et al. *Nature Communications*, 9, 2722 (2018)

Experiment (ESRF ID01)

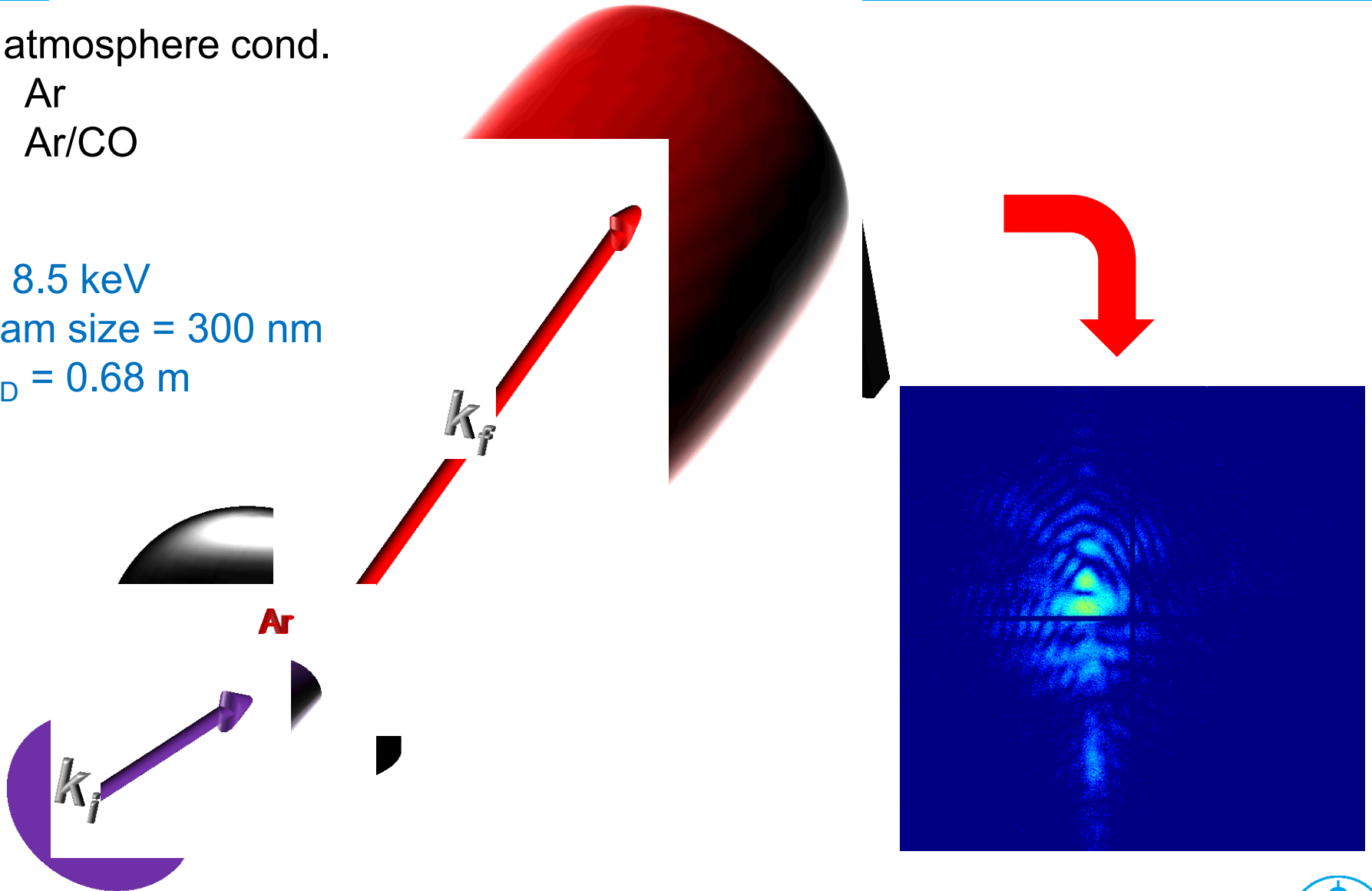
2 atmosphere cond.

1. Ar
2. Ar/CO

$E = 8.5 \text{ keV}$

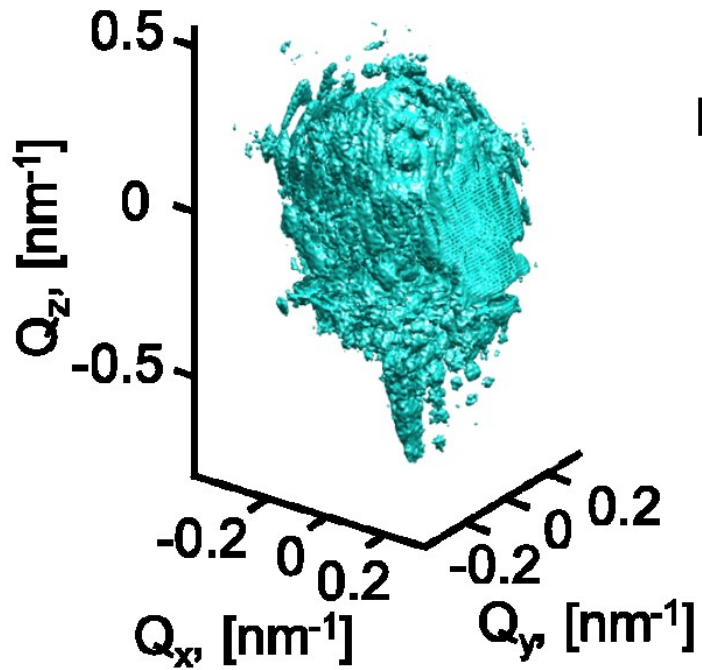
Beam size = 300 nm

$L_{S-D} = 0.68 \text{ m}$



3D Intensity Distribution & Reconstruction

Ar atmosphere



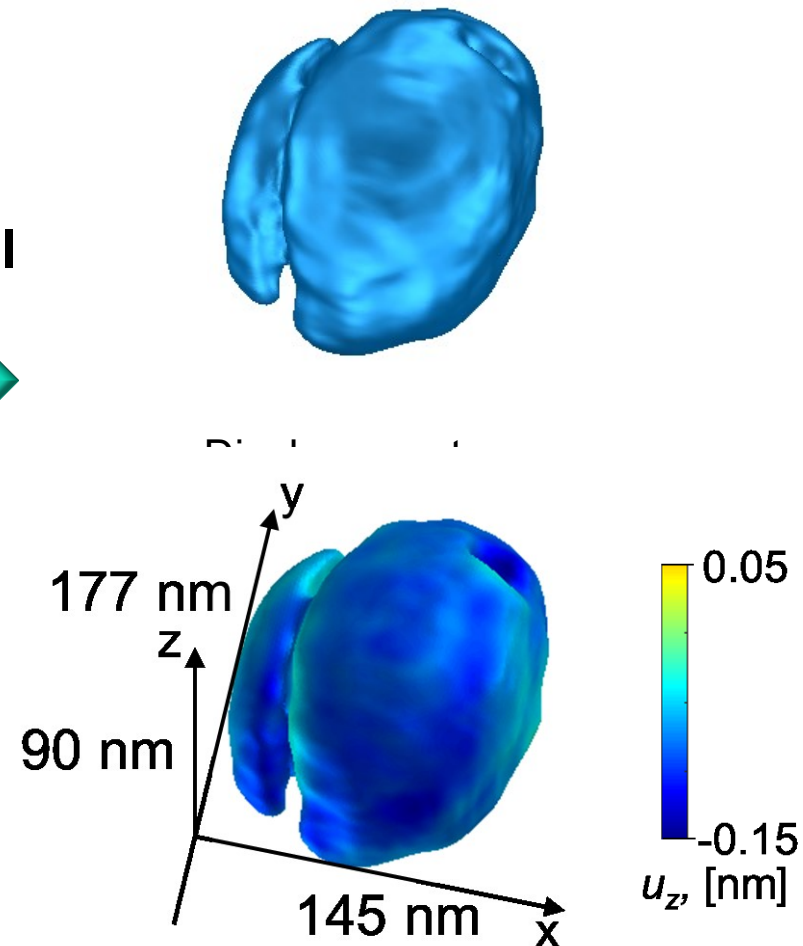
Pt 111 Bragg peak

Phase Retrieval



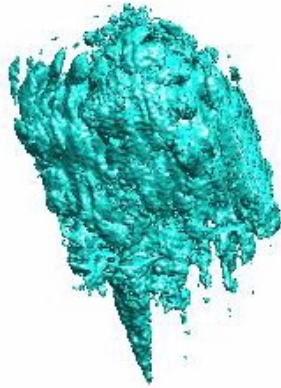
HIO+ER

Amplitude isosurface map

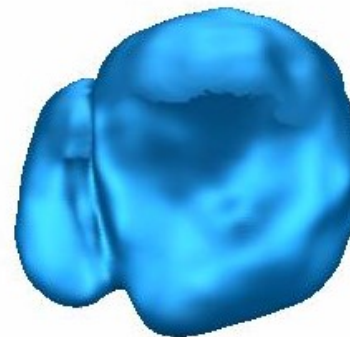
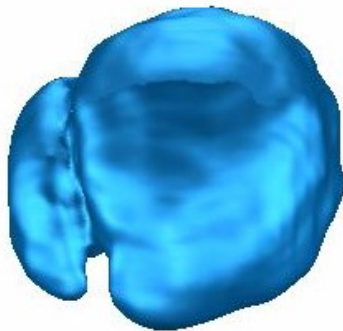
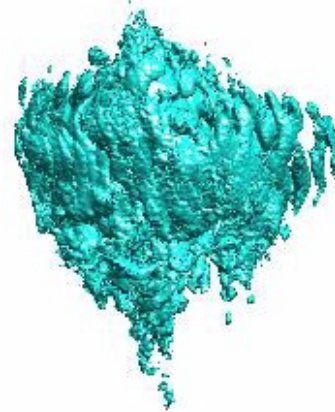


Results of reconstruction

Ar atmosphere



Ar/CO atmosphere

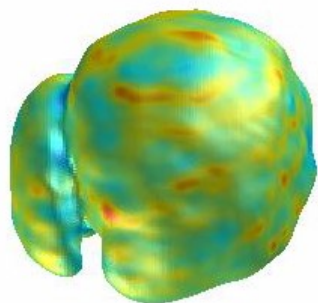


3D coherent scattered intensities and Pt particle shape reconstruction

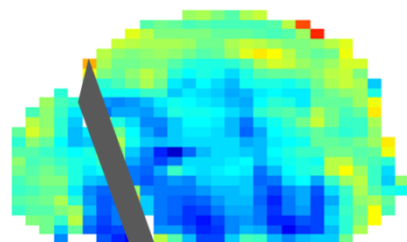


Strain

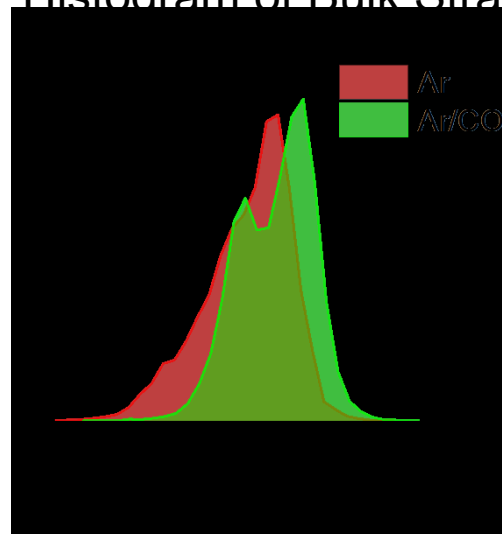
Ar



Slice through the center of the particle



Histogram of Bulk Strain

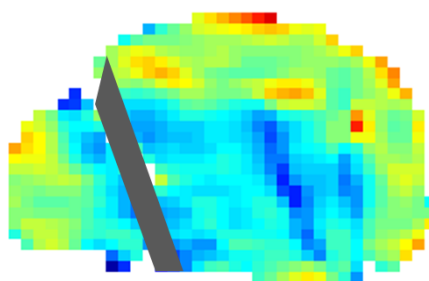
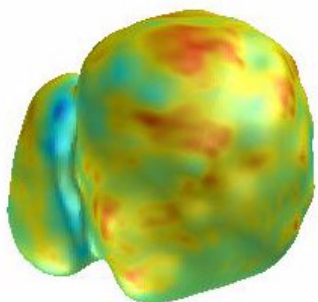


Mean values [%]

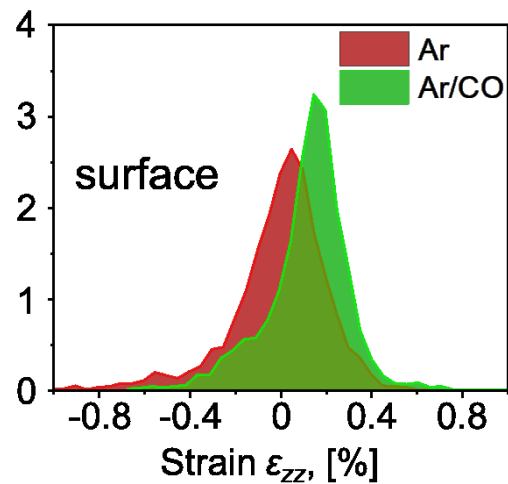
Ar : -0.15

Ar/CO : -0.02

Ar/CO



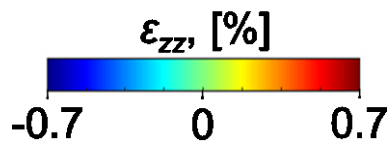
Histogram of Surface Strain



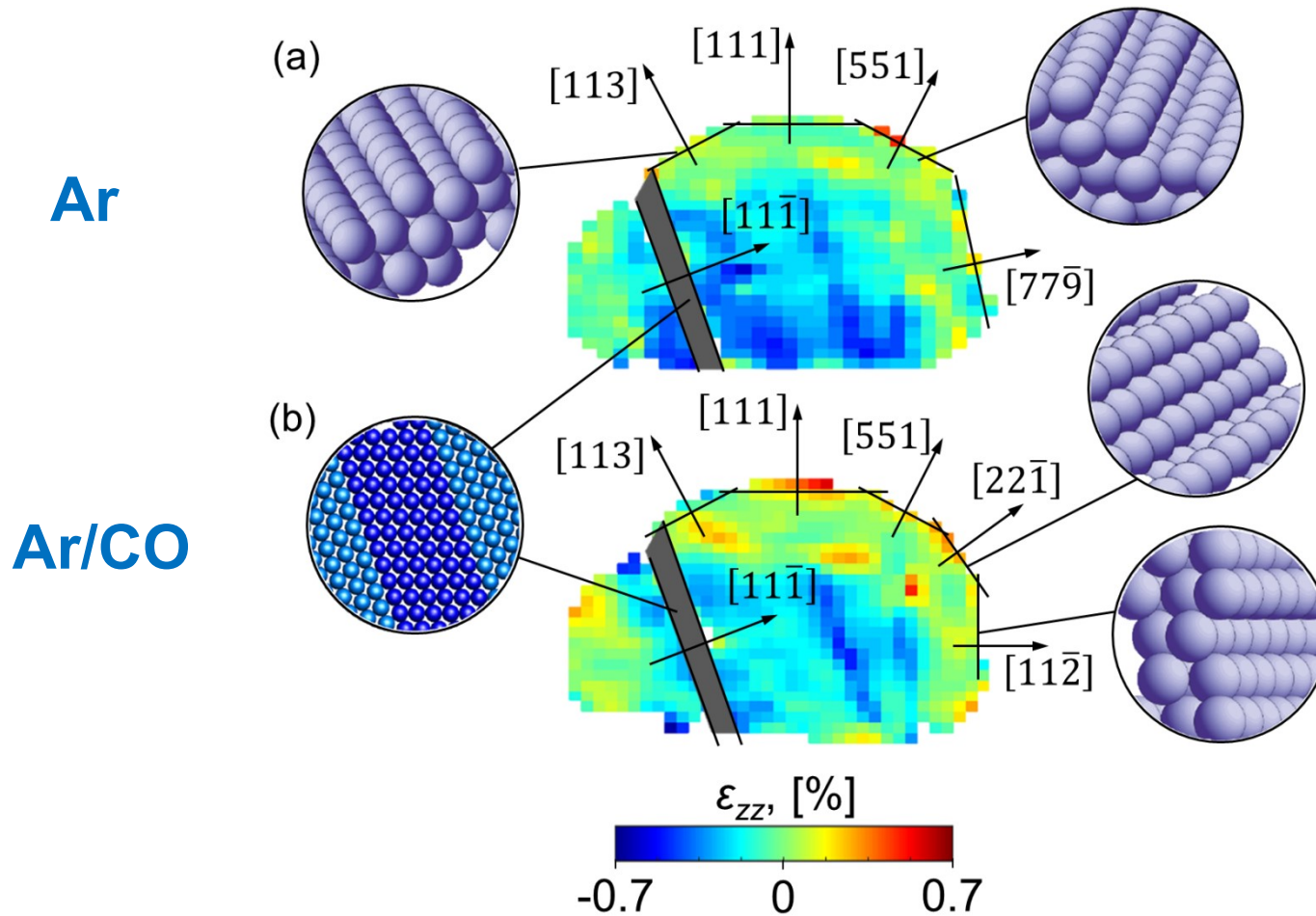
Mean values [%]

Ar : -0.01

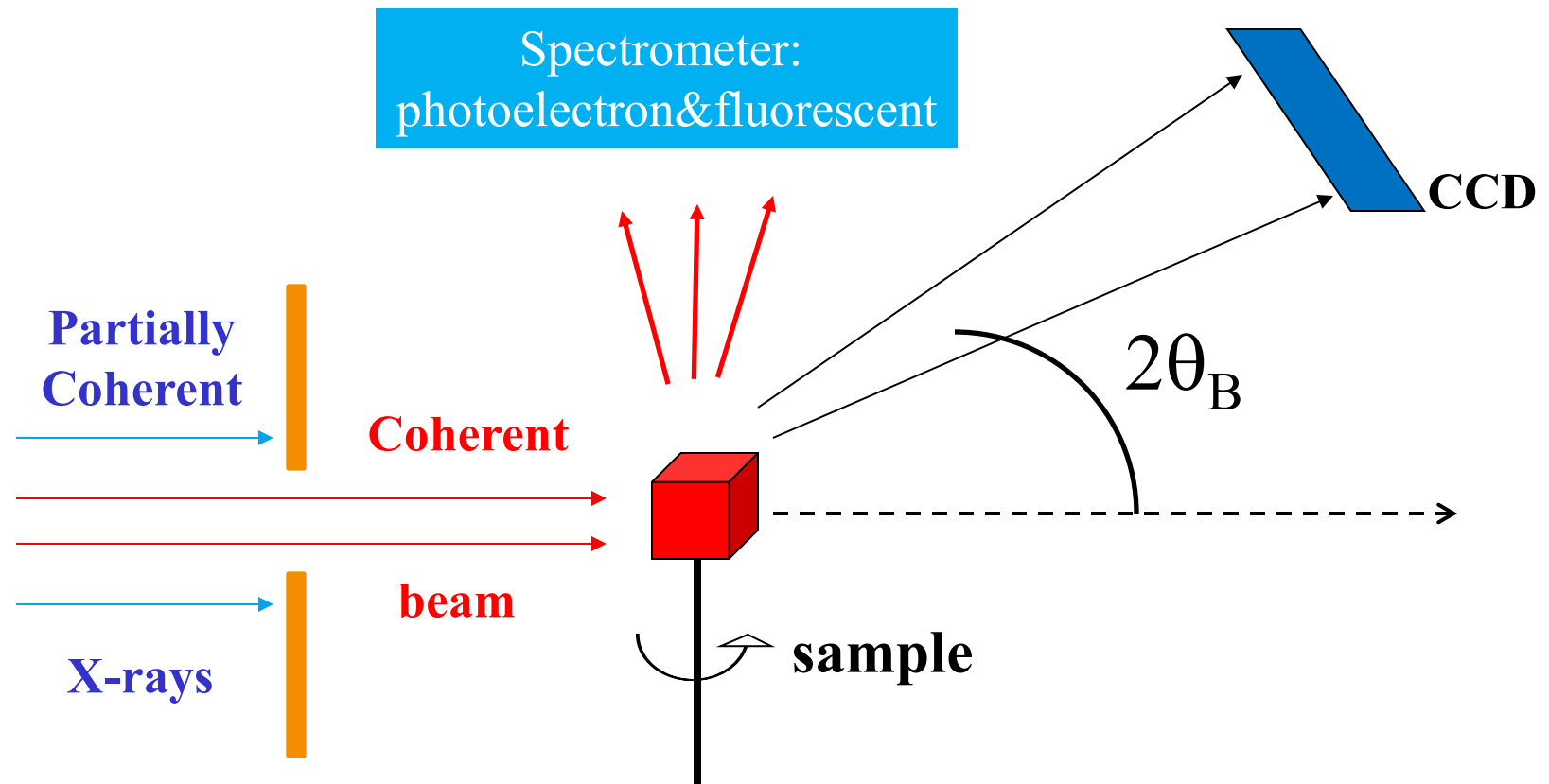
Ar/CO : 0.12



Strain with the surface facets



Strain field with assignment of the vicinal surface facets

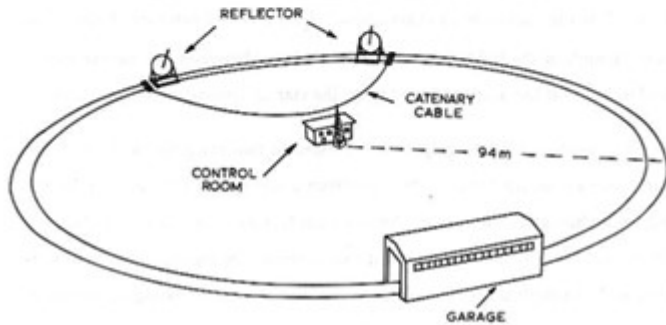
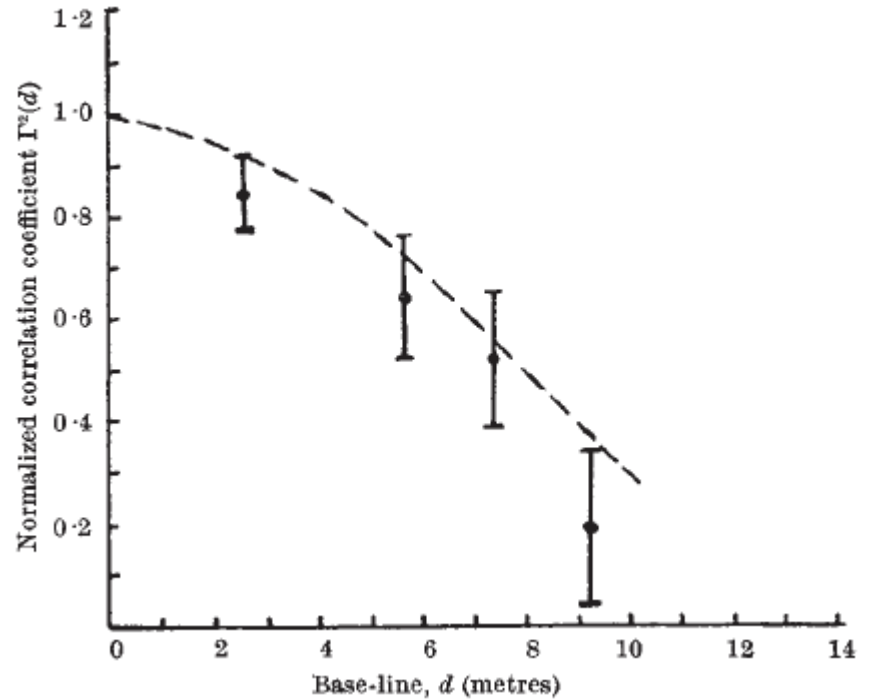
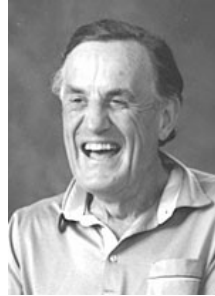


Combination of Ultrafast Spectroscopy and CXDI

What are the 2-nd order coherence properties of X-ray free-electron lasers?

I. A. Vartanyants & A. Singer, "Coherence Properties of Third-Generation Synchrotron Sources and Free-Electron Lasers", Synchrotron Light Sources and Free-Electron Lasers. Editors: E. Jaeschke, S. Khan, J.R. Schneider and J.B. Hastings Springer International Publishing Switzerland (2016), pp.821-863.

Hanbury Brown and Twiss interferometry



Hanbury Brown, R. , and Twiss, R. Q. , *Nature*, 178, 1046 (1956)

HBT telescopes measuring light intensity from Sirius

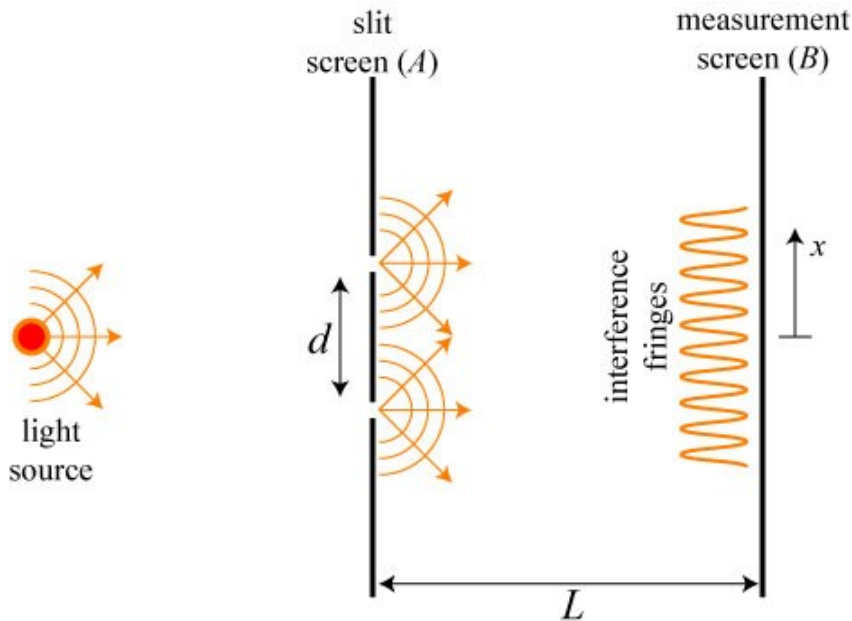
G. Goldhaber, Proc. Int. Workshop on Correlations and Multiparticle production (CAMP - LESIP IV), World Scientific (1991).

$$g^{(2)}(\mathbf{r}_1, \mathbf{r}_2) = \frac{\langle I(\mathbf{r}_1)I(\mathbf{r}_2) \rangle}{\langle I(\mathbf{r}_1) \rangle \langle I(\mathbf{r}_2) \rangle}$$



Young's and HBT interferometry

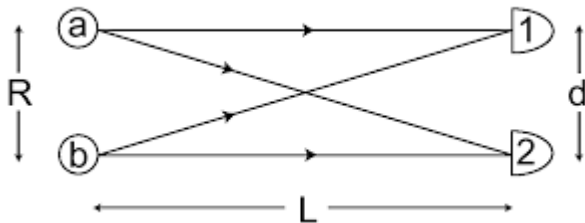
Young's double pinhole interferometry



1-st order correlation function:

$$g^{(1)}(x_1, x_2) = \frac{\langle E(x_1)E(x_2) \rangle}{\langle \sqrt{I(x_1)} \rangle \langle \sqrt{I(x_2)} \rangle}$$

HBT interferometry

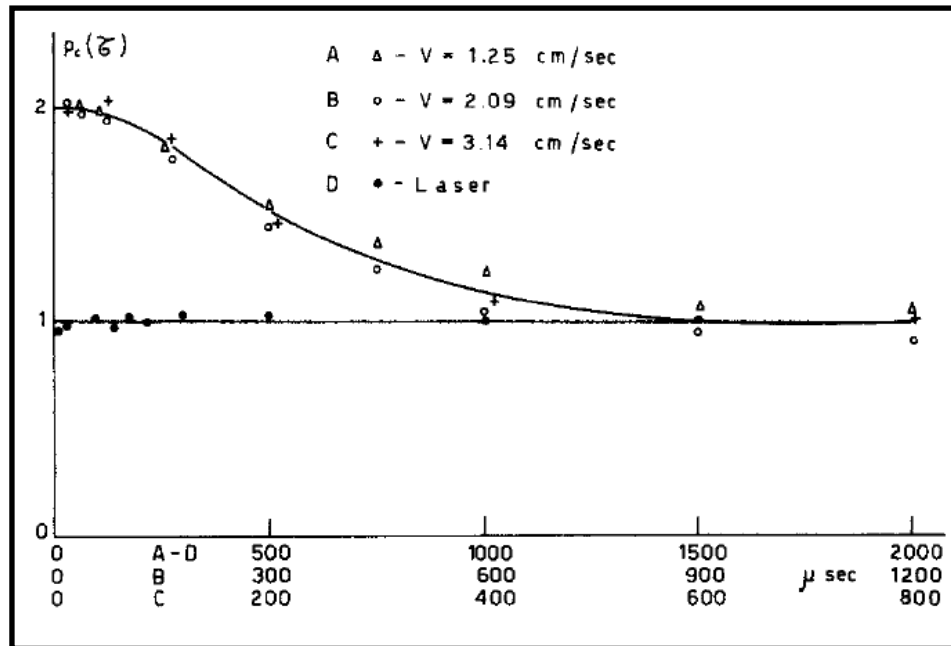


2-nd order correlation function:

$$g^{(2)}(x_1, x_2) = \frac{\langle I(x_1) \cdot I(x_2) \rangle}{\langle I(x_1) \rangle \cdot \langle I(x_2) \rangle}$$



Laser sources and chaotic sources

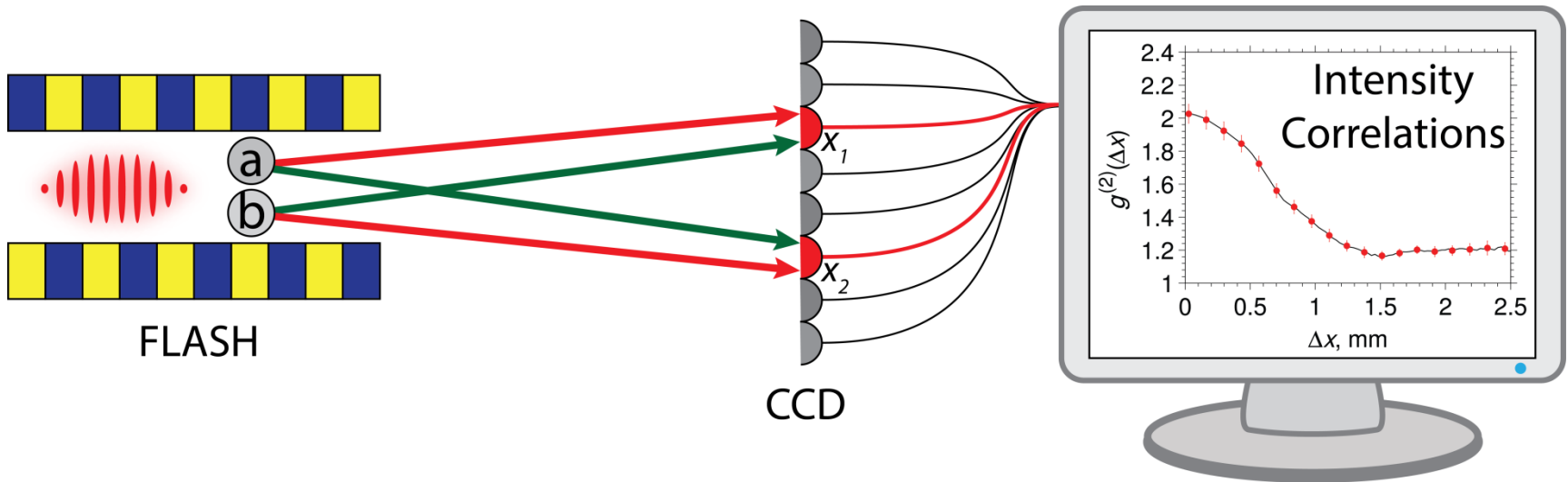


Chaotic sources (Gaussian Statistics)

$$g^{(2)}(\mathbf{r}_1, \mathbf{r}_2) = 1 + \frac{\tau_c}{T} |\gamma_{12}|^2$$

τ_c : coherence time
 T: pulse duration
 τ_c/T high at FELs

Hanbury Brown - Twiss Interferometry at FELs



A. Singer *et al.*, *Phys. Rev. Lett.* **111**, 034802 (2013)

O. Gorobtsov *et al.*, *Phys. Rev. A* **95**, 023843 (2017)

O. Gorobtsov *et al.*, *Sci. Rep.* **8**, 2219 (2018)

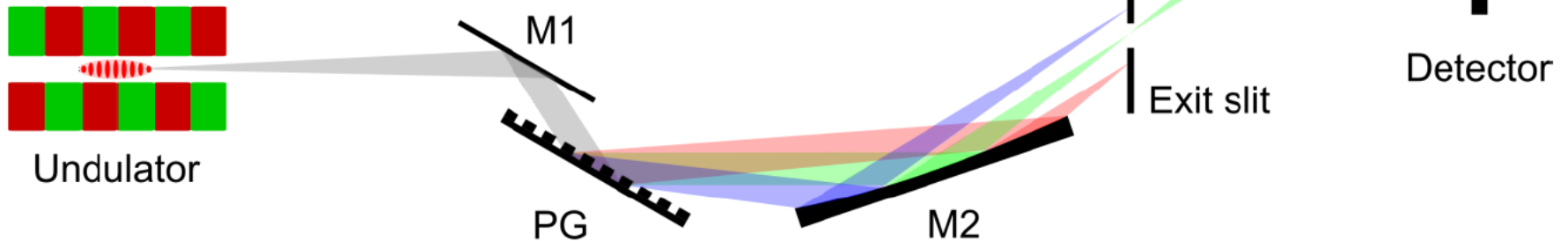
O. Gorobtsov *et al.*, *Nat. Commun.* **9**, 4498 (2018)

Intensity-Intensity Correlation Measurements at FLASH

$\lambda=5.5$ nm

Bunch charge 0.6 nC

Pulse Energy 100 μ J



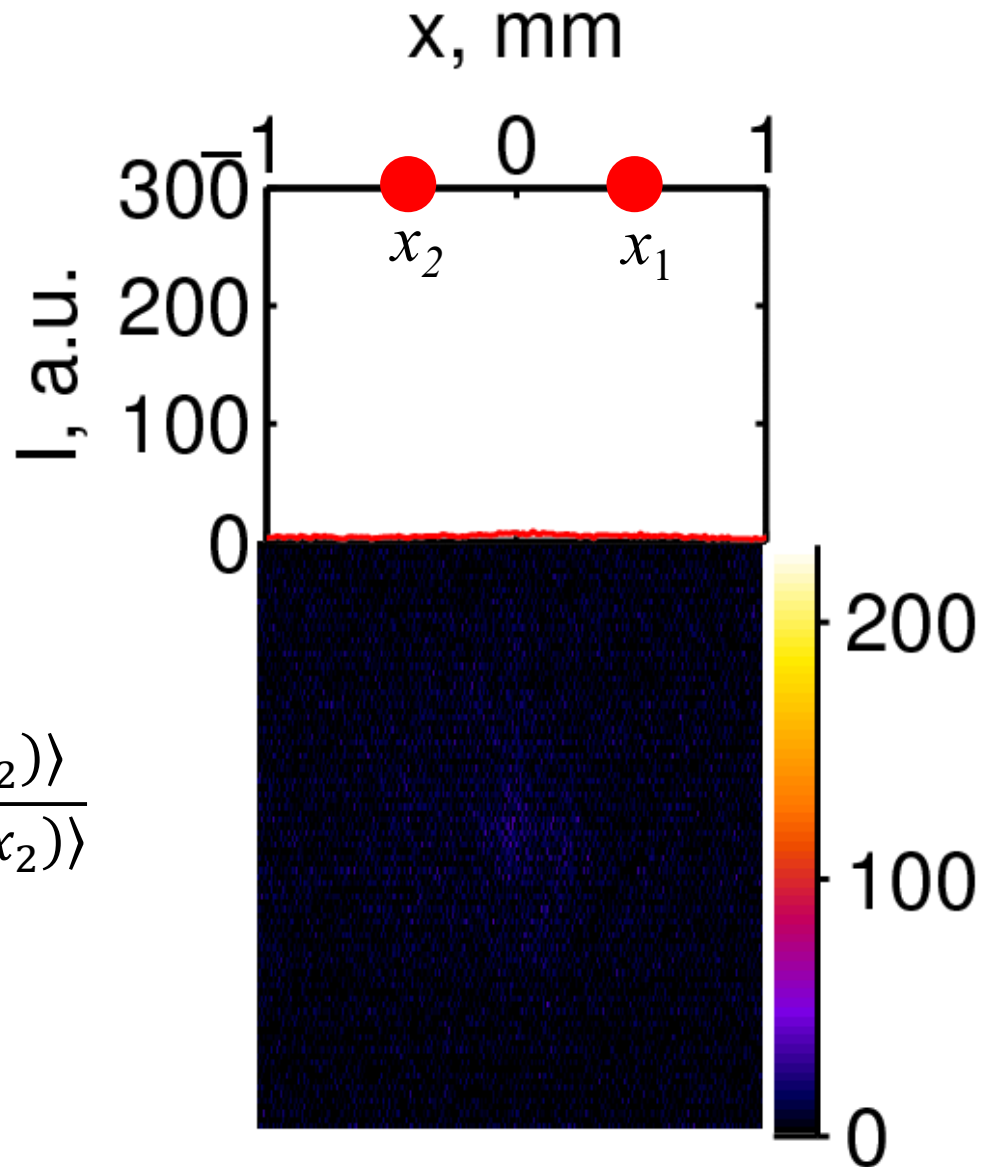
$$g^{(2)}(\mathbf{r}_1, \mathbf{r}_2) = 1 + \frac{\tau_c}{T} |g^{(1)}(\mathbf{r}_1, \mathbf{r}_2)|^2$$

$$\tau_c \sim 1/\Delta\nu$$

Second Order Correlation Measurements

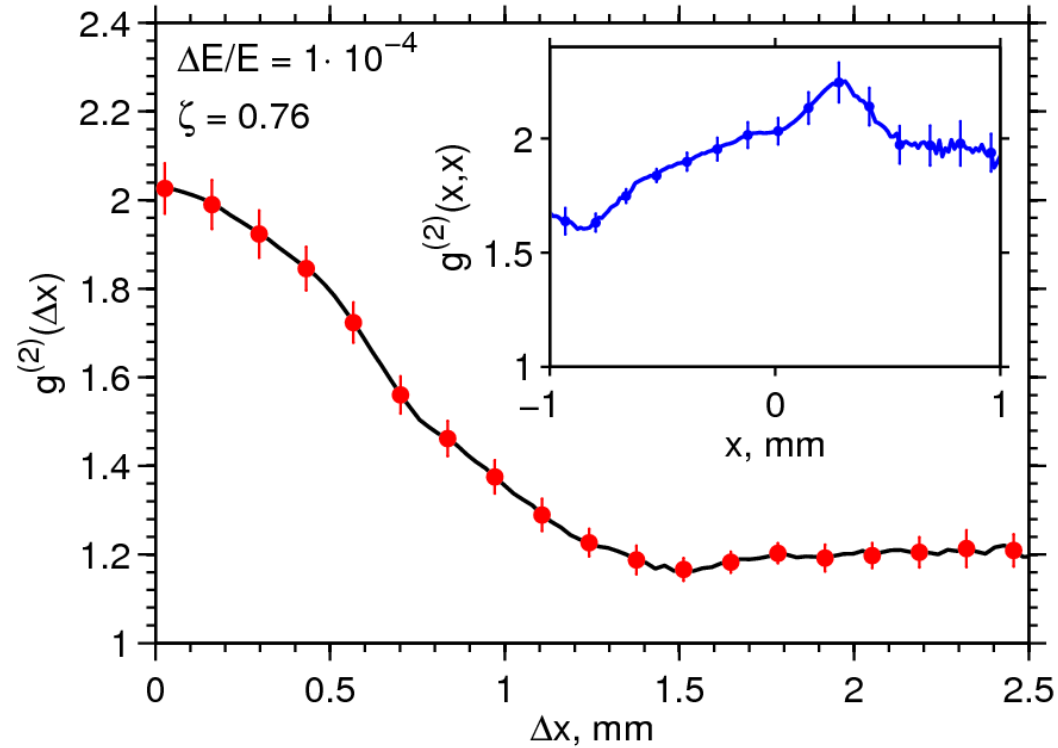
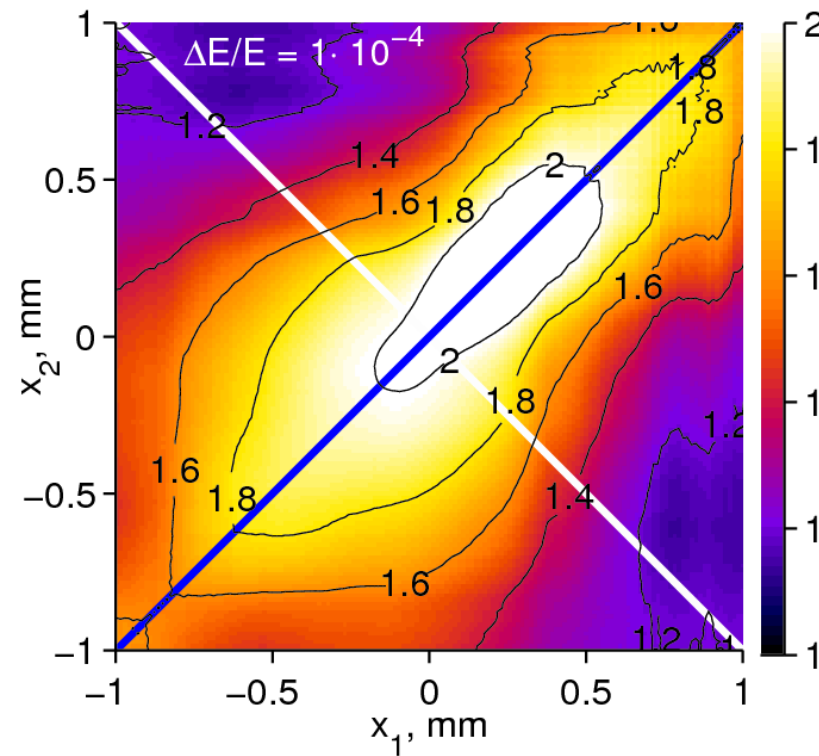
$30 \mu\text{m}$
 $\Delta E/E = 0.8 \cdot 10^{-4}$

$$g^{(2)}(x_1, x_2) = \frac{\langle I(x_1)I(x_2) \rangle}{\langle I(x_1) \rangle \langle I(x_2) \rangle}$$



Second Order Correlation Measurements

Exit slit: $30\ \mu\text{m}$

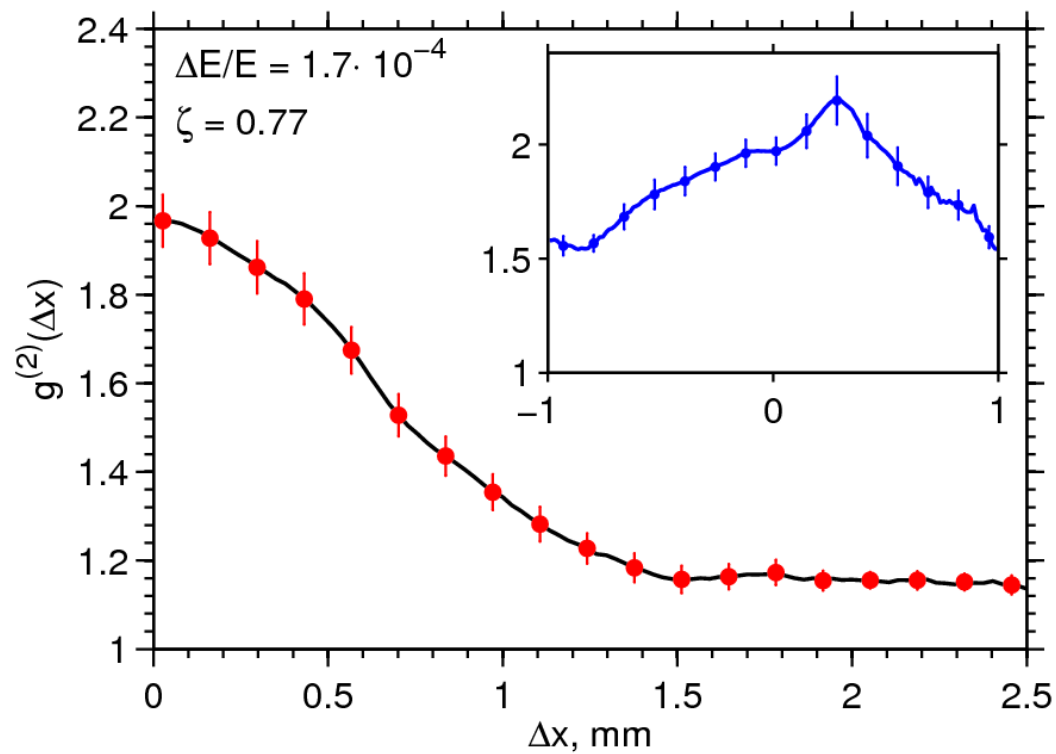
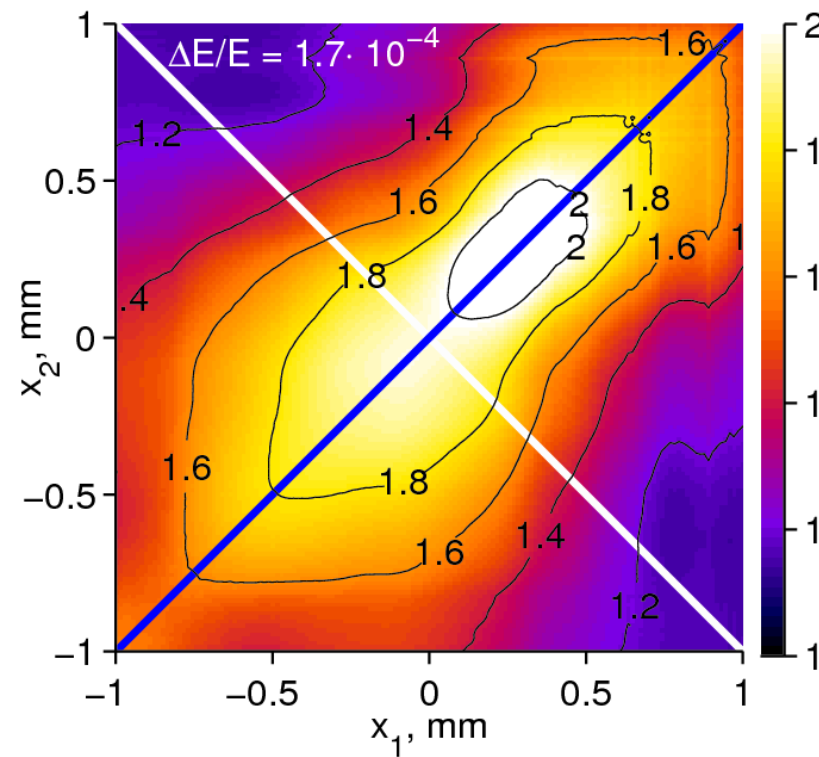


Single longitudinal mode!

Coherence length: $l_c = 0.93$ mm
Beam size: 0.45 mm
Degree of coherence: 76%

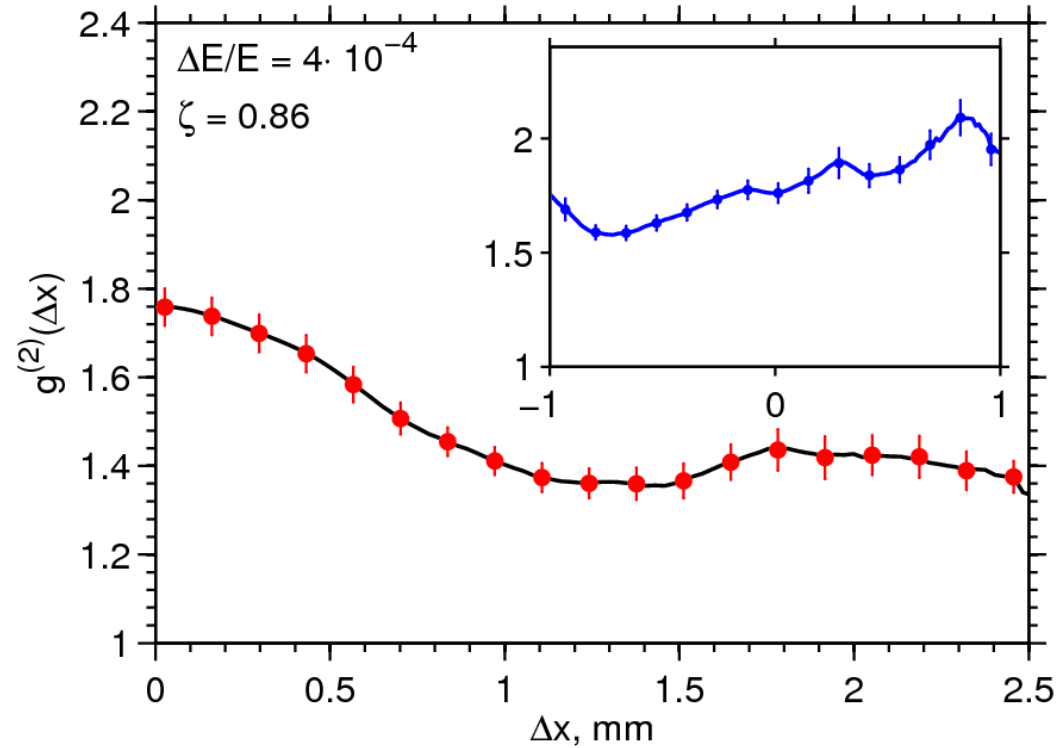
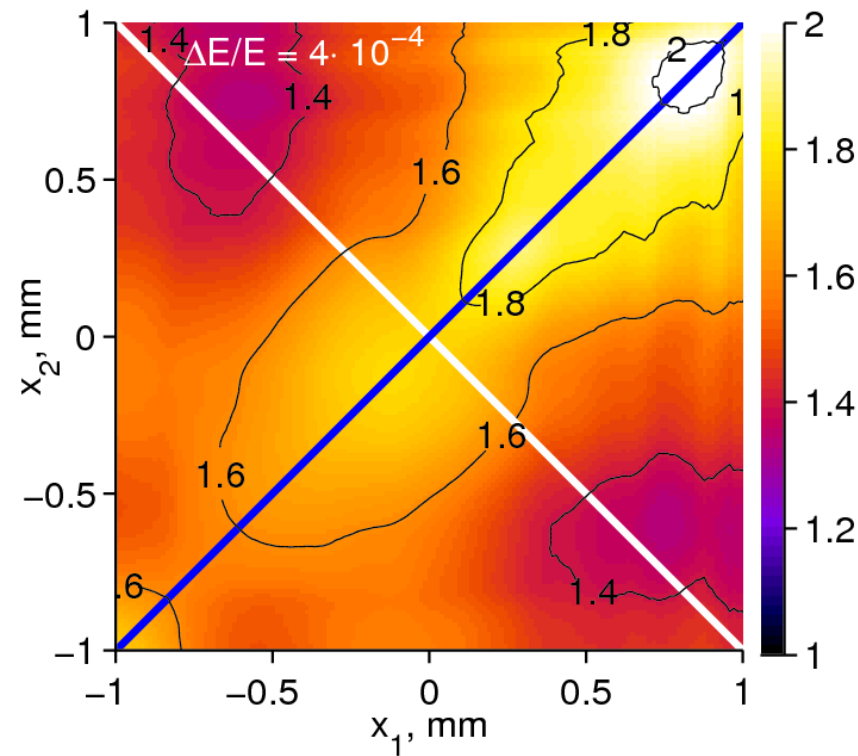
Second Order Correlation Measurements

Exit slit: $60\ \mu\text{m}$



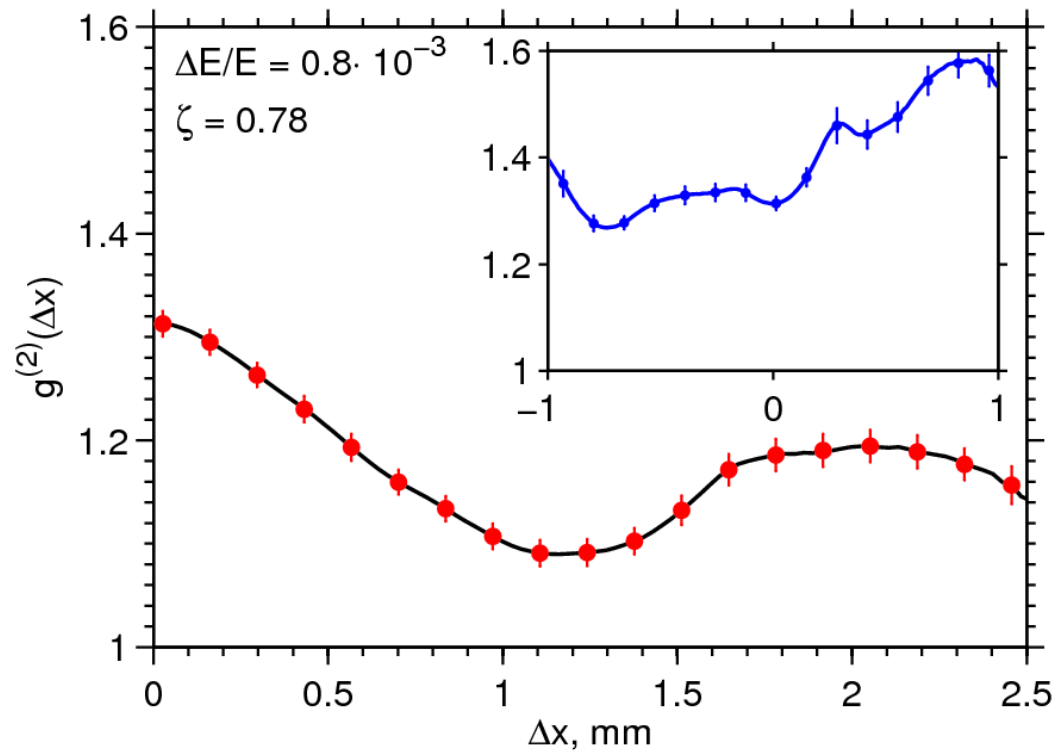
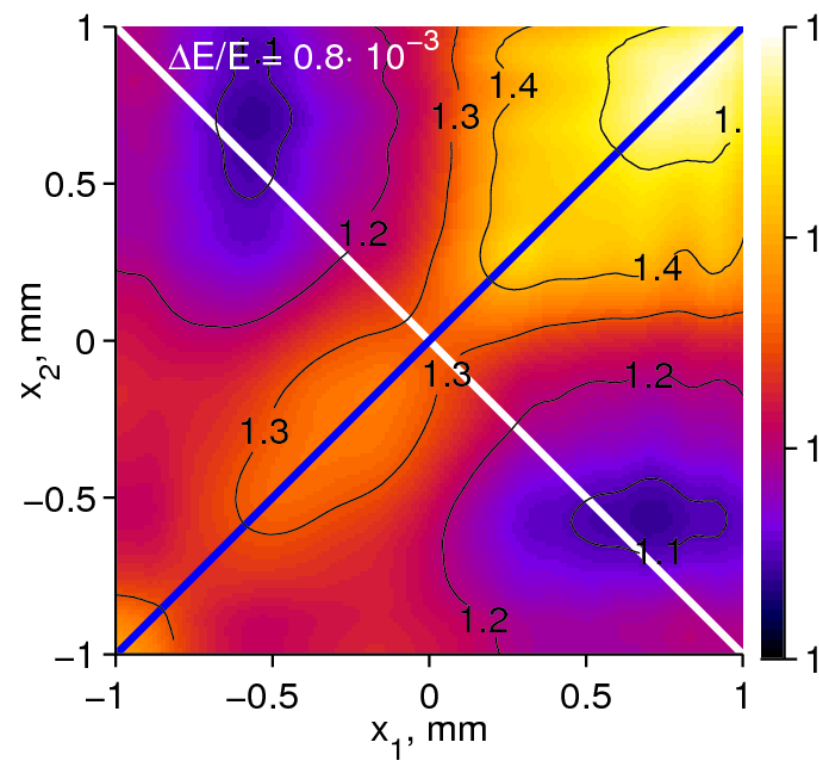
Second Order Correlation Measurements

Exit slit: 150 μm



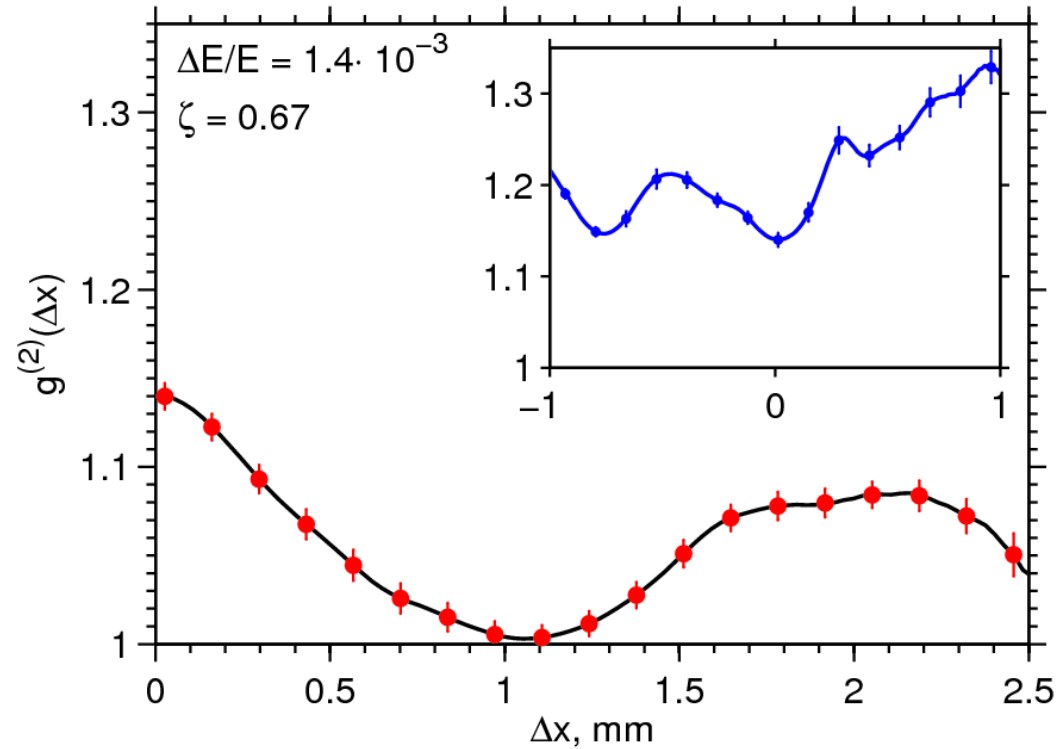
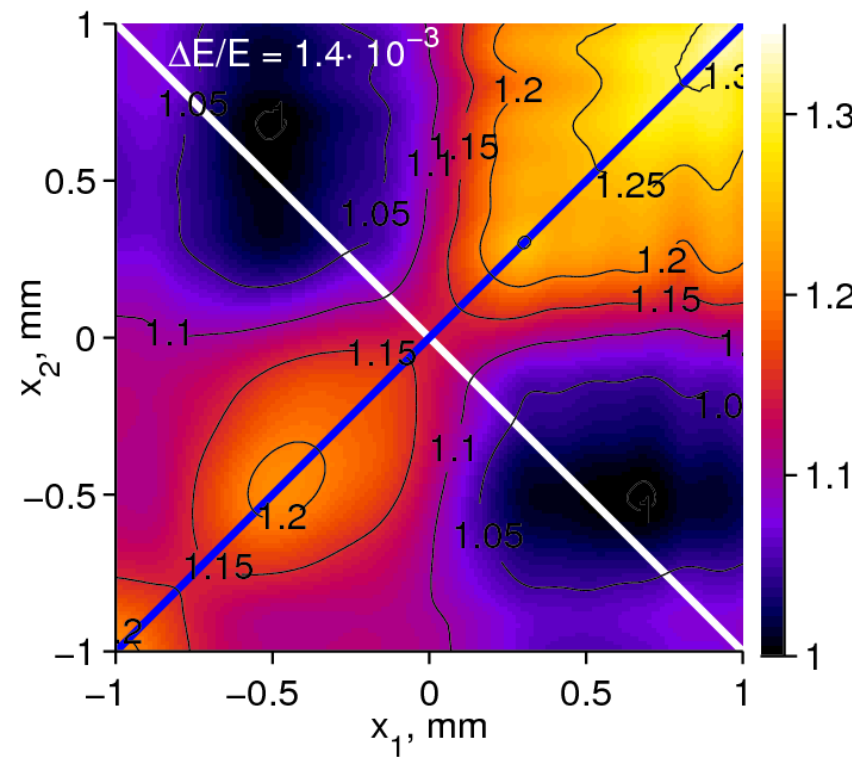
Second Order Correlation Measurements

Exit slit: 300 μm



Second Order Correlation Measurements

Exit slit: 500 μm

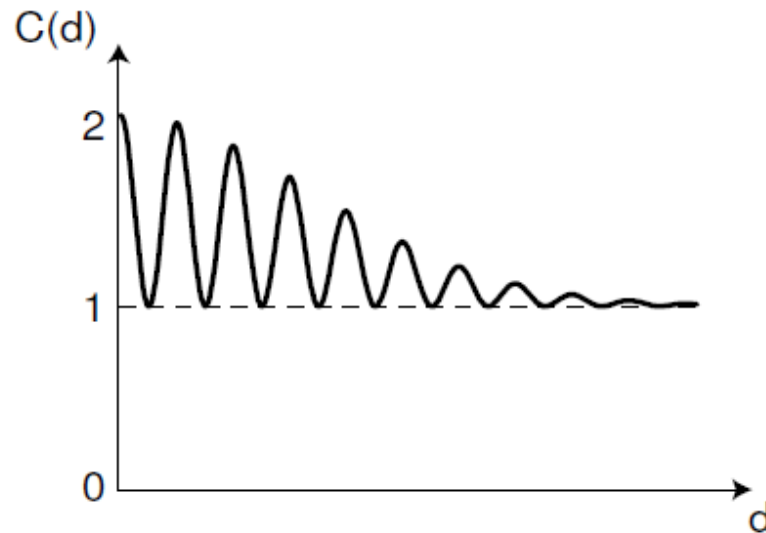


What is the origin of the 2-nd maximum?

Second order correlation measurements



(a)



(b)

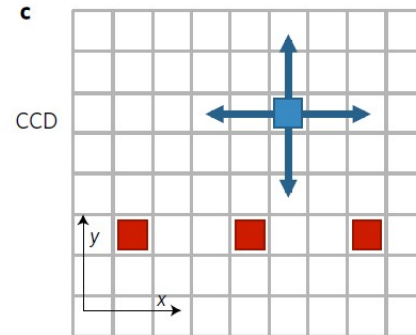
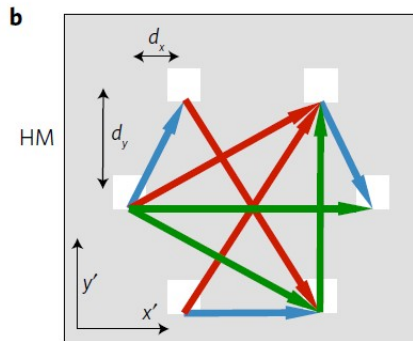
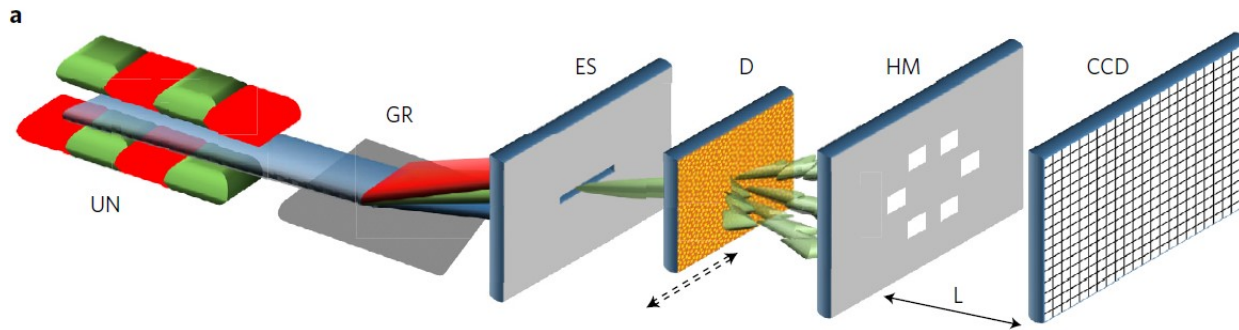


Quantum imaging with incoherently scattered light from a free-electron laser

Collaboration with J. von Zanthier group (Uni. Erlangen)
R. Röhlsberger and W. Wurth groups (DESY)

Quantum imaging using high order correlations

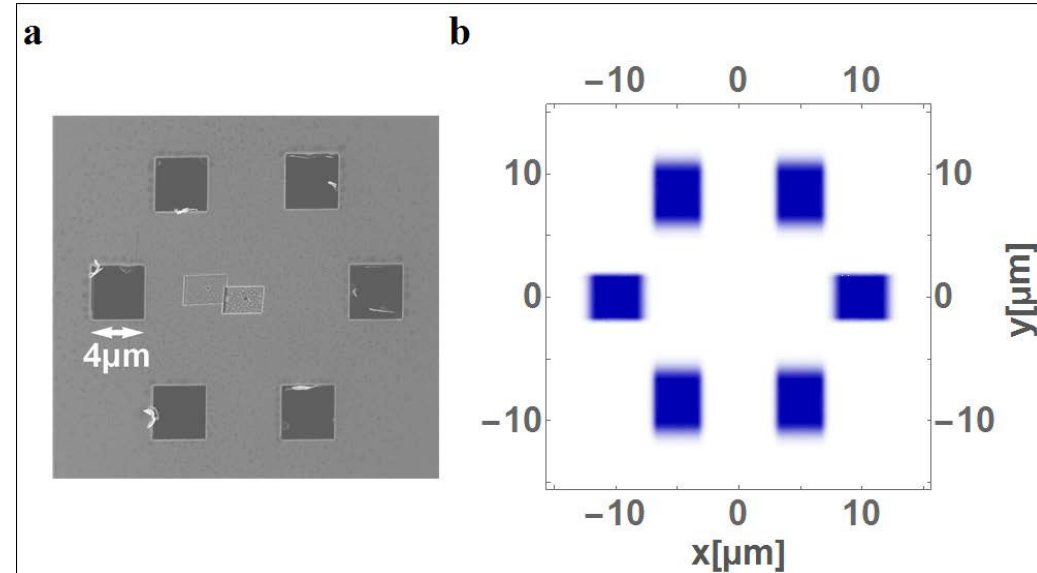
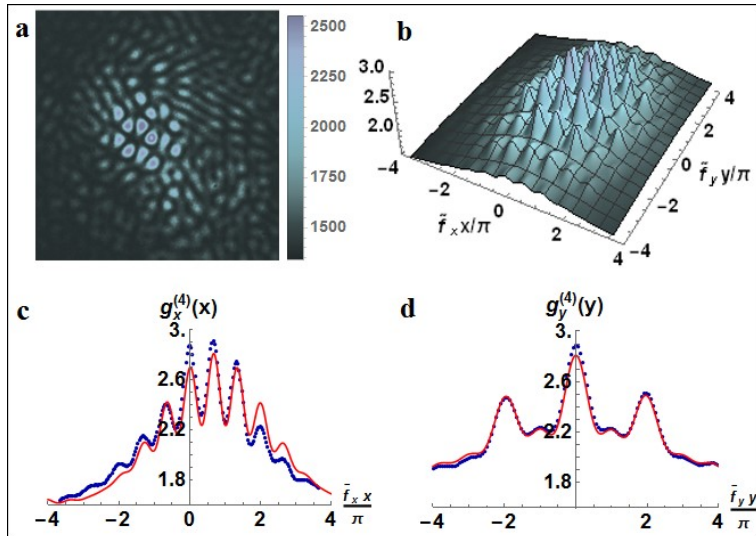
Scheme of experiment at FLASH



Nine source separation vectors of a benzene-like mask generating six independently radiating incoherent light sources

Fourth-order correlation scheme

Quantum imaging using high order correlations



Single-shot speckle pattern and experimentally derived fourth-order intensity correlation function

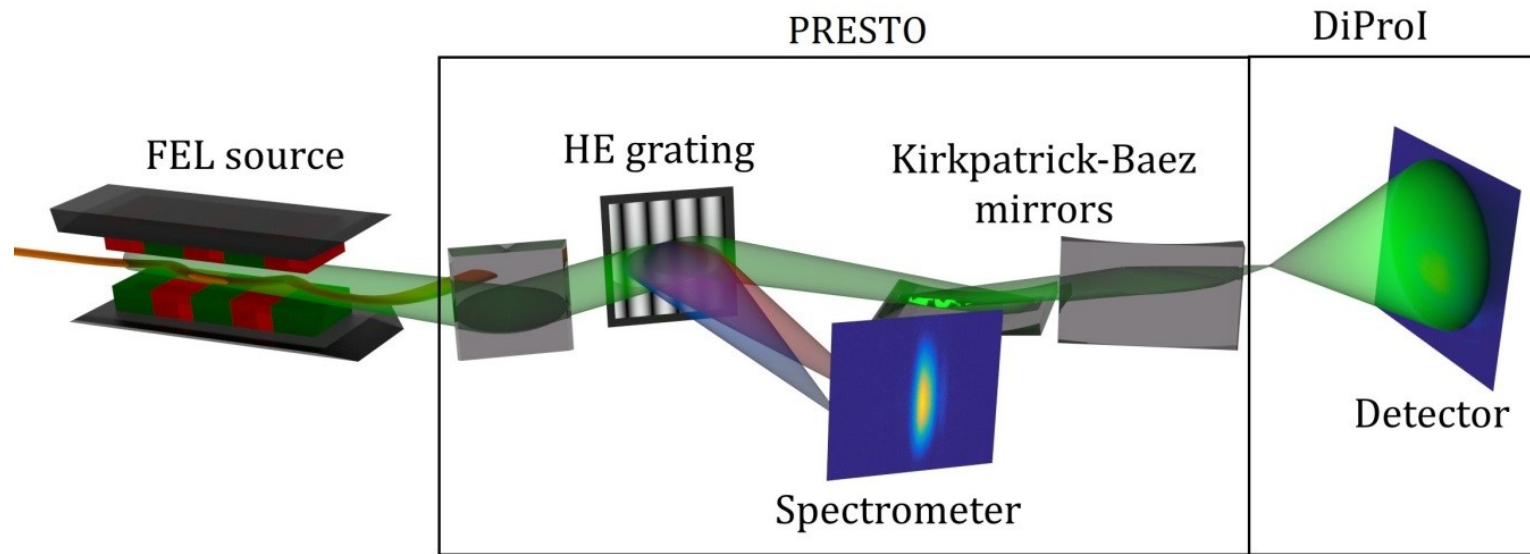
SEM image and quantum protocol reconstruction of the artificial benzene molecule

**Our results demonstrate definitely that SASE
XFELs statistically behave as chaotic sources
and not as a fully coherent sources!**

What about seeded FELs?



HBT experiment at FERMI

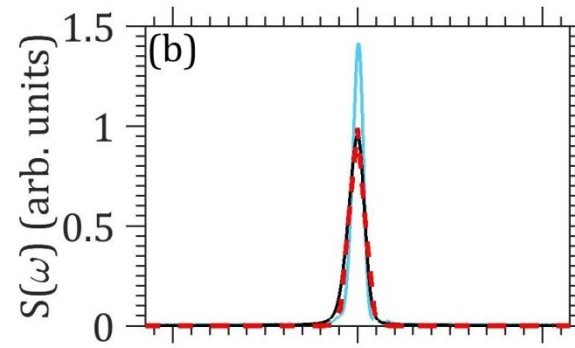
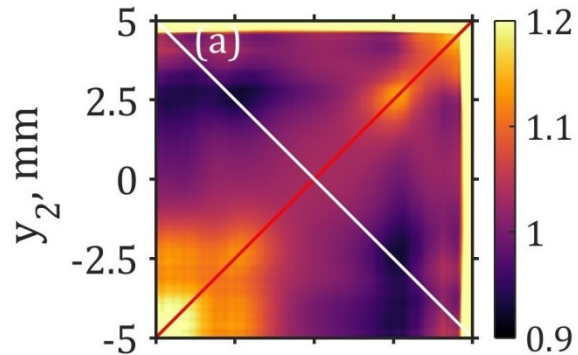


**Layout of experiment at the seeded FEL
FERMI**



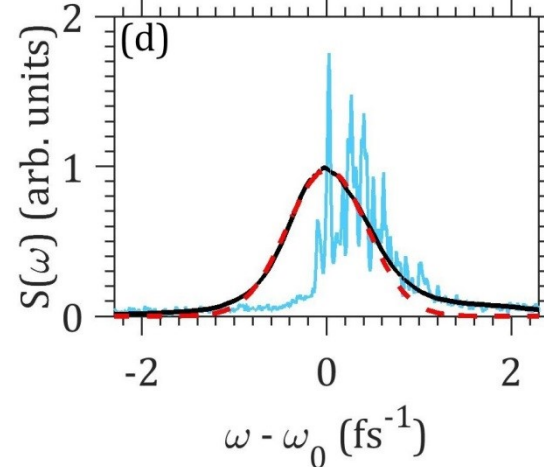
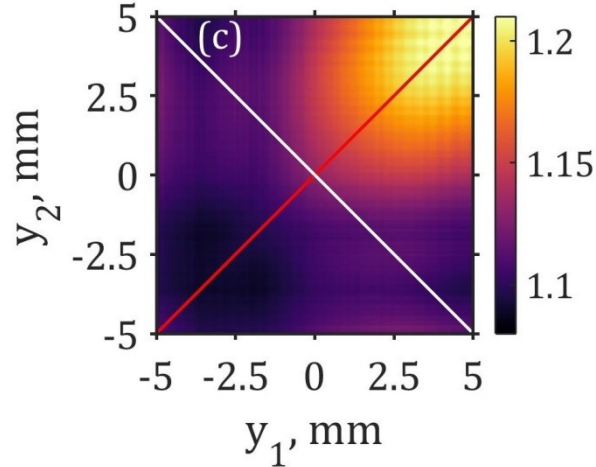
HBT experiment at FERMI

Seeded



$$g^{(2)} - 1 = 0.03$$

SASE



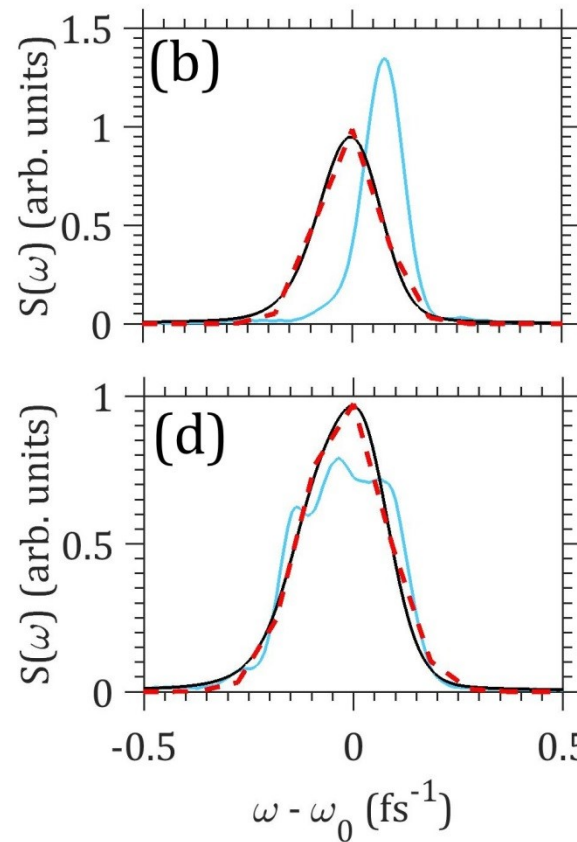
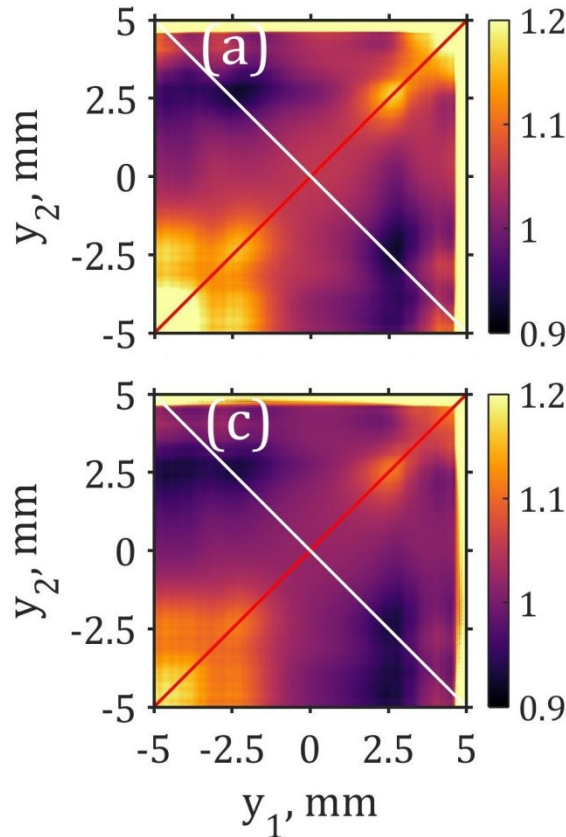
$$g^{(2)} - 1 = 0.1$$

Seeded and SASE mode correlation functions and spectra



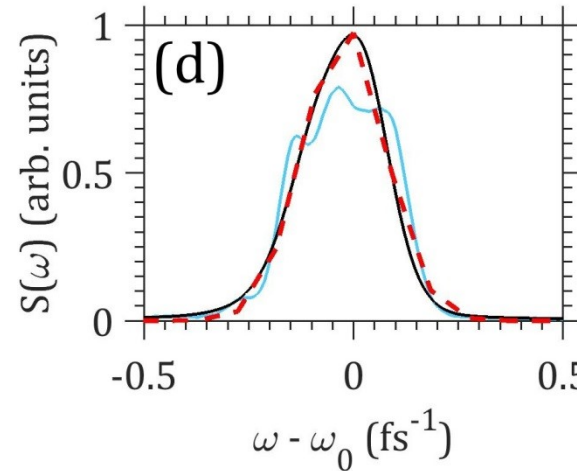
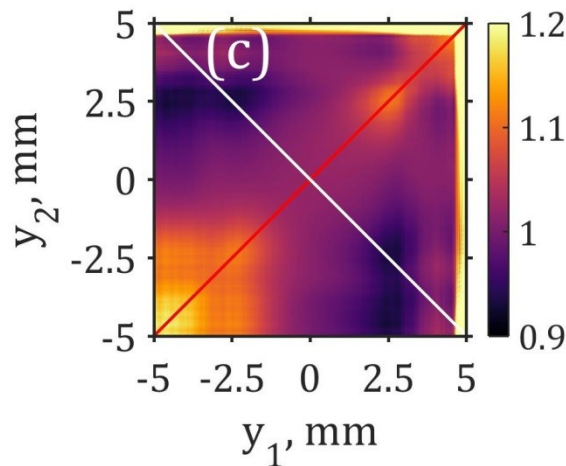
HBT experiment at FERMI

Single pulse



$$g^{(2)} - 1 = 0.02$$

Multiple pulses



$$g^{(2)} - 1 = 0.07$$

Seeded mode single and multiple pulses analysis



**Seeded FEL source FERMI statistically
behaves as a real laser-like source
according to Glauber's definition**

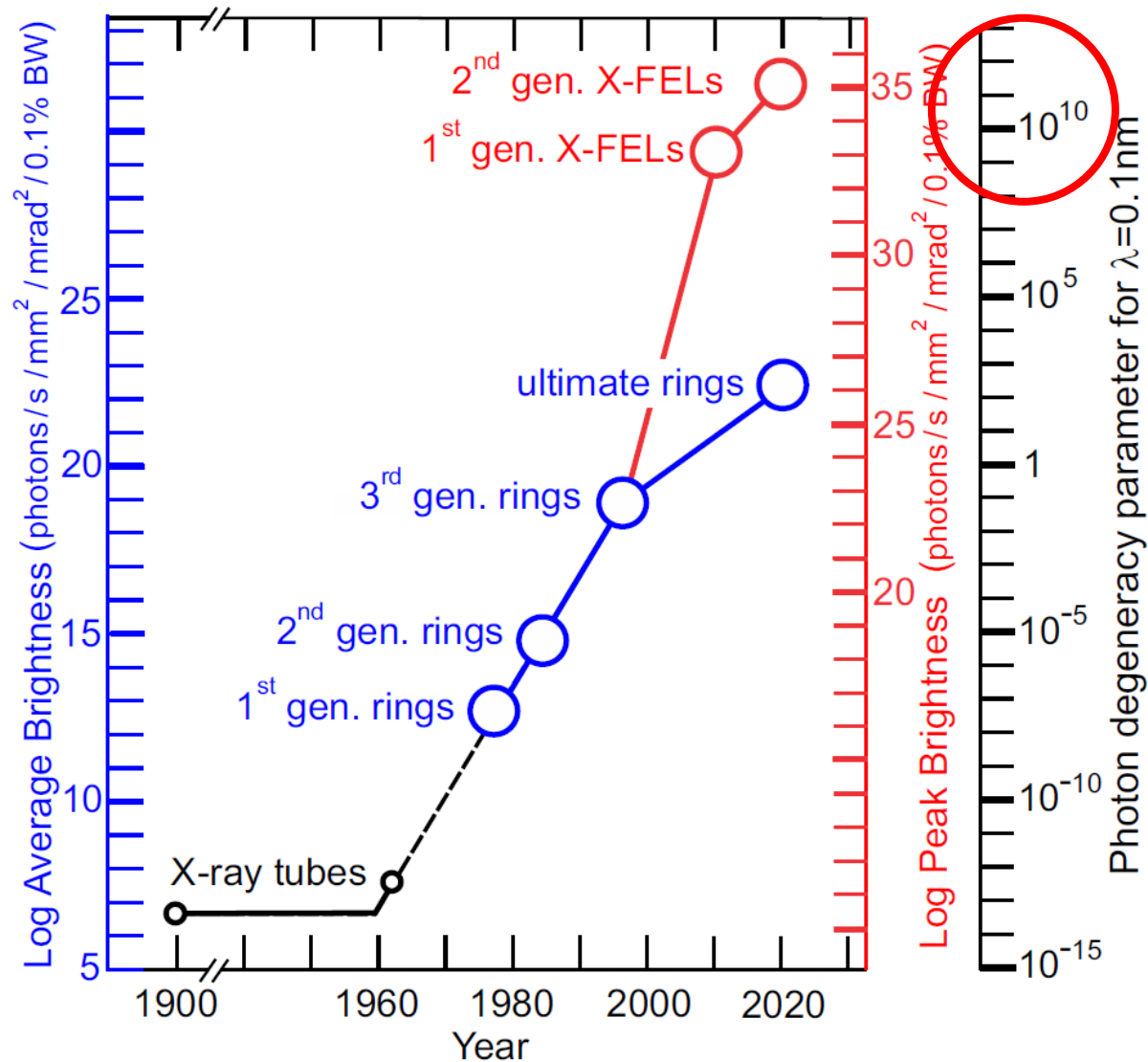
Still open question!

**What is the statistical behavior of self-seeded
XFEL pulses?**

**We hope to clarify this question at
European XFEL!**



Degeneracy parameter

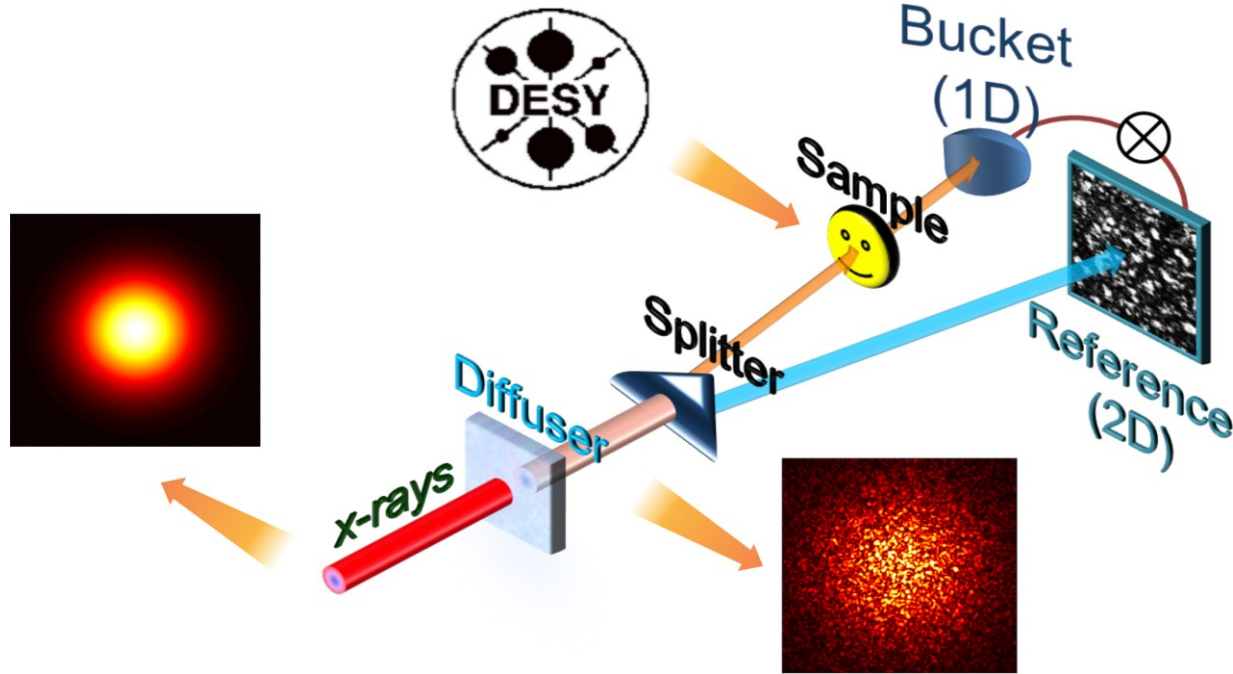


**How to exploit this high degeneracy
parameter at XFEL sources?**



Classical Ghost Imaging at X-ray Free-Electron Lasers

What is Classical Ghost Imaging



Indirect imaging by correlation of

- a) Photons that pass through the sample and are measured by bucket detector
- b) Photons which never pass through the sample and are measured by the pixel detector

$$G(x, y) = \frac{\langle \Delta I_B \cdot \Delta I_R(x, y) \rangle_M}{\sqrt{\langle \Delta I_B^2 \rangle_M \langle \Delta I_R^2(x, y) \rangle_M}}$$

$G(x, y)$: Ghost Image

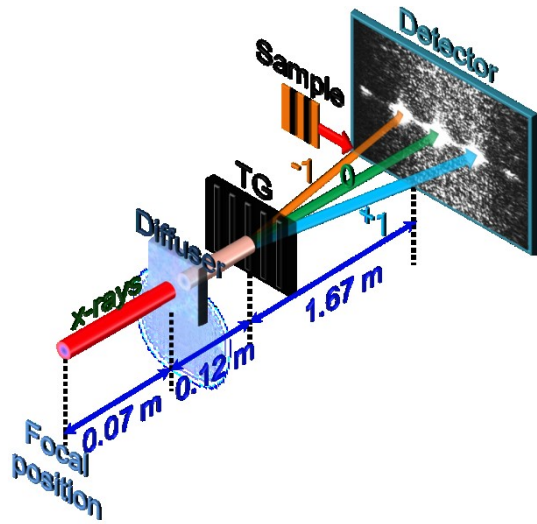
M : # of frames

I_B : Bucket signal

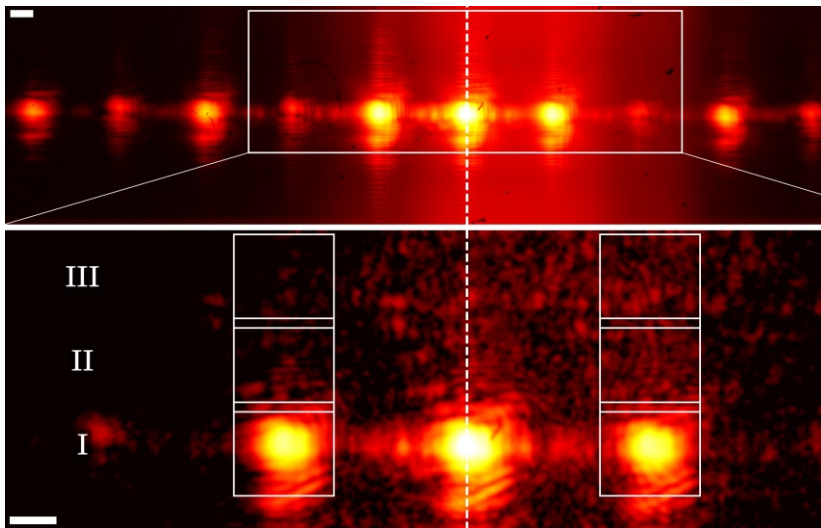
$I_R(x, y)$: Reference Image

$\Delta I : I - \langle I \rangle_M$

Experiment at FLASH



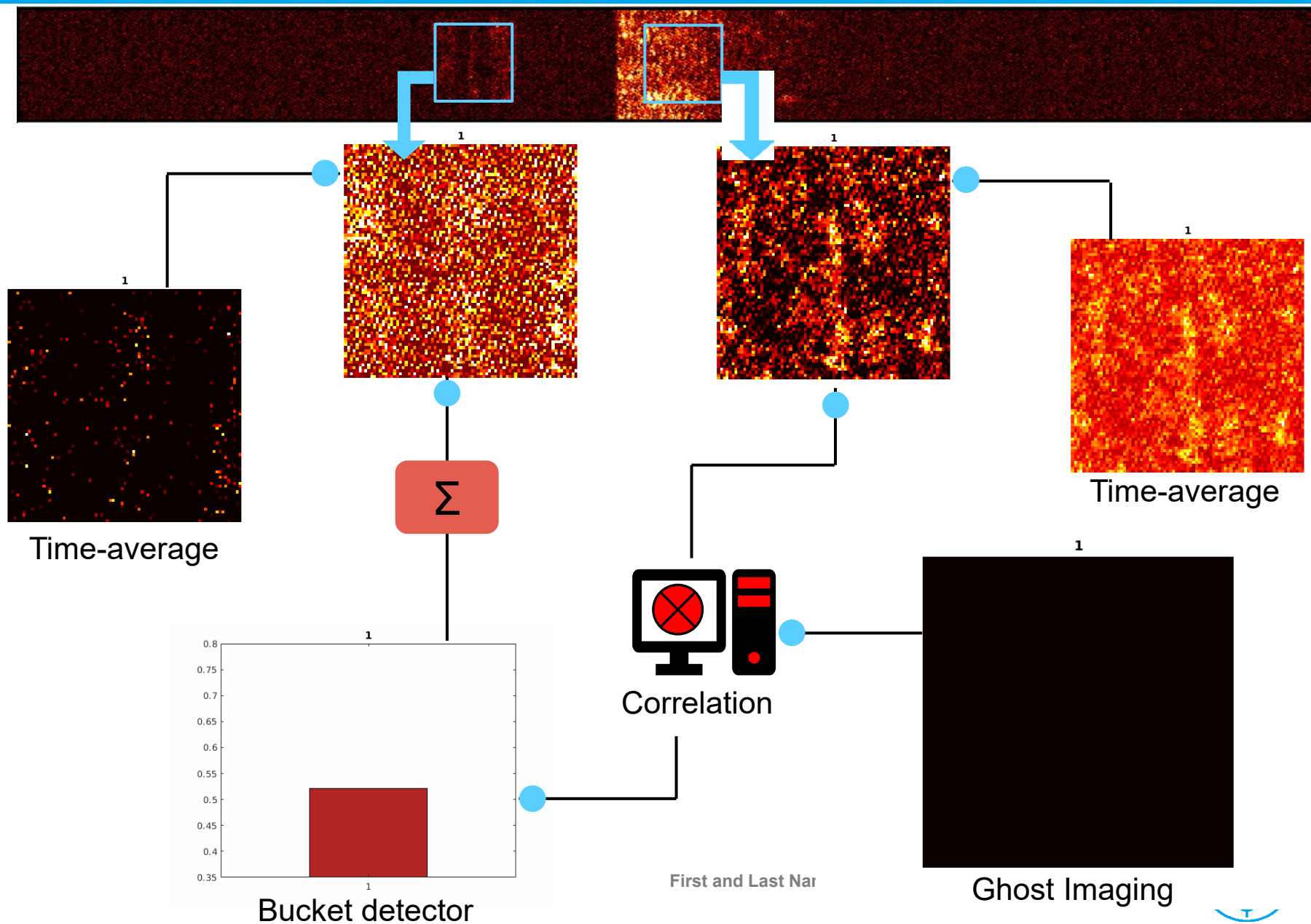
Setup II
(Diffuser + Transmission Grating)



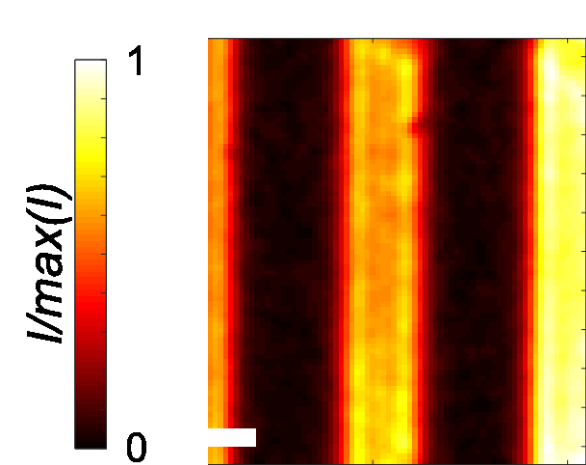
Example of detected intensities (averaged) with several orders from the transmission grating clearly visible.

The speckle field generated in a single realization.

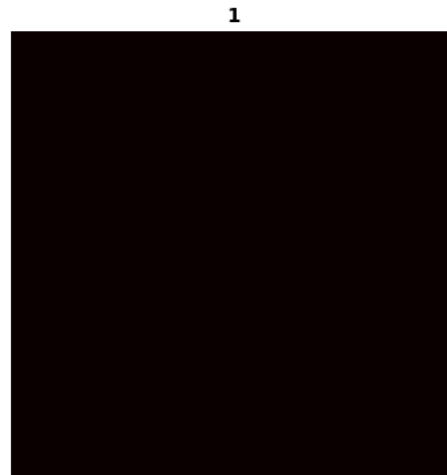
Ghost Imaging Protocol



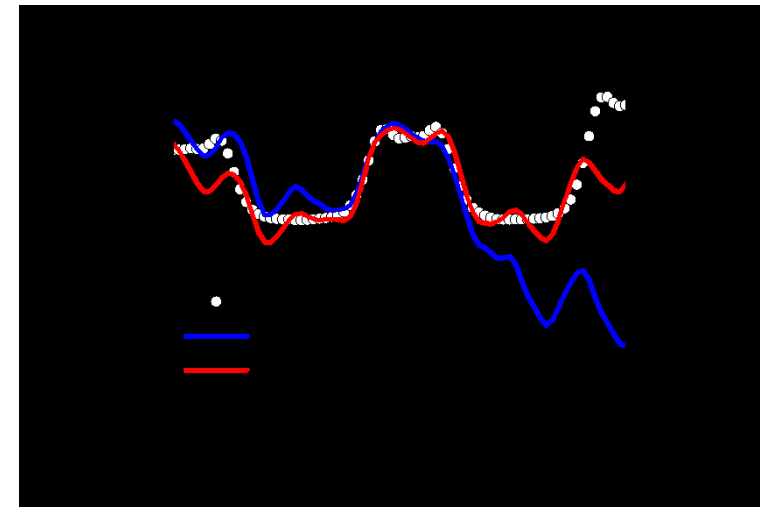
Experiment at FLASH – result of GI



An average image at the bucket detector



Ghost imaging reconstruction
(average of $\sim 40\ 000$)

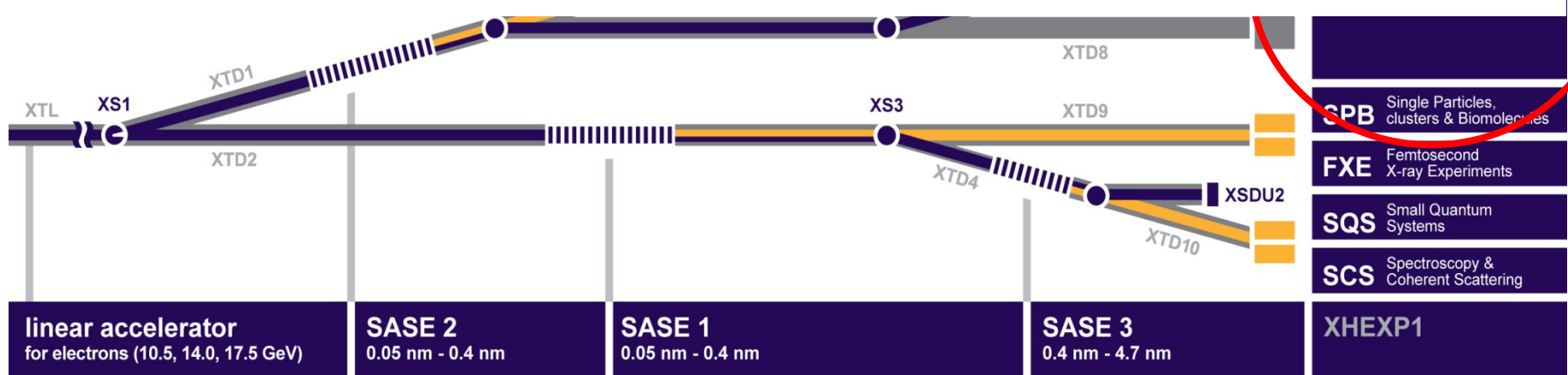


Line profile of the structure

How we can fill two empty tunnels?

Undulator Segment	FEL radiation energy [keV]	Wavelength [nm]
SASE 1	3 - over 24 (Hard XR)	0.4 - 0.05

**We would like to exploit more
of coherence and quantum properties
of XFELs at new beamlines**



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FLASH, LCLS, FERMI teams



We have open PostDoc positions!

Thank you for your attention!

