Imaging via photon-photon correlation of X-ray fluorescence

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SPB/SFX

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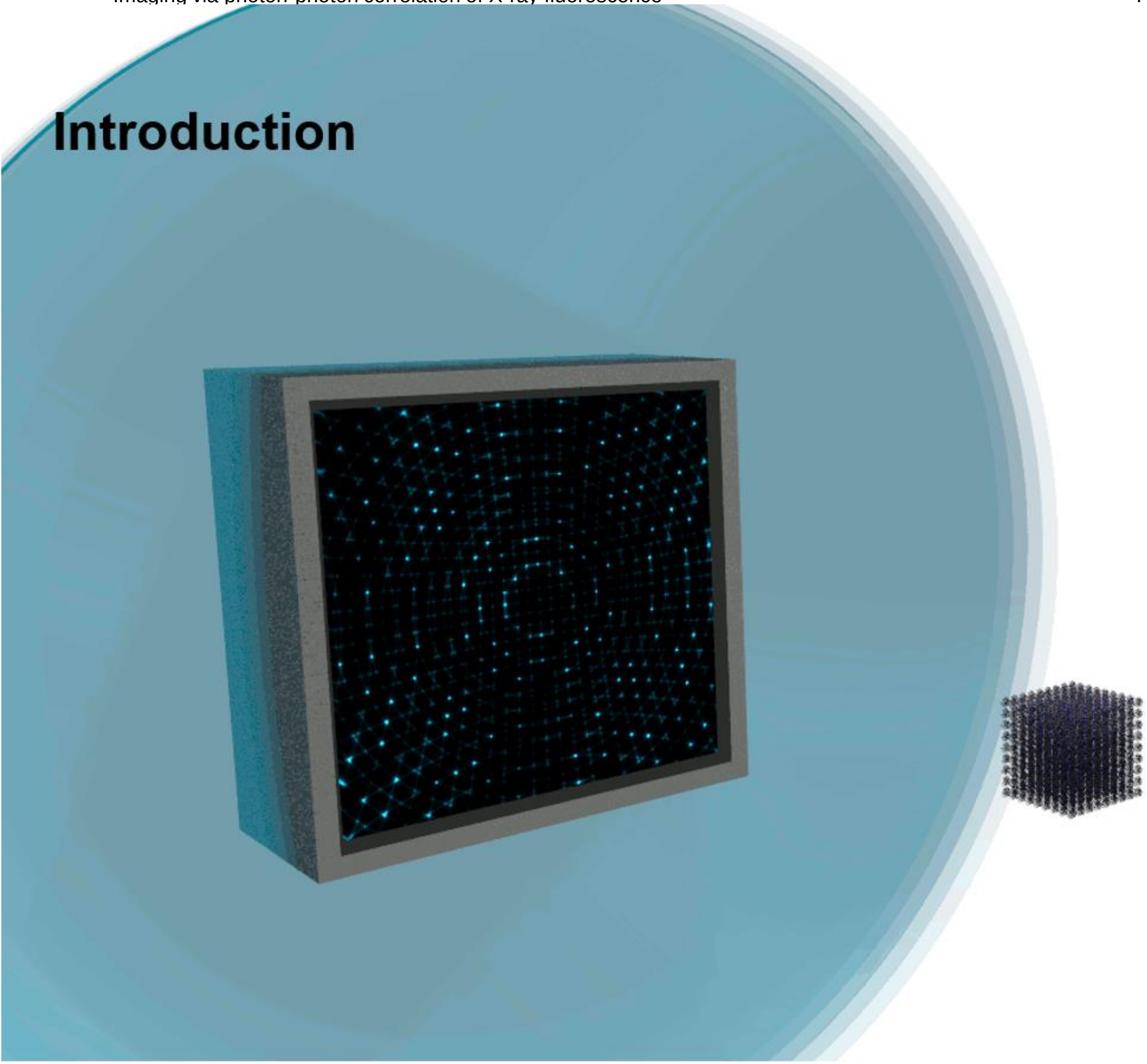
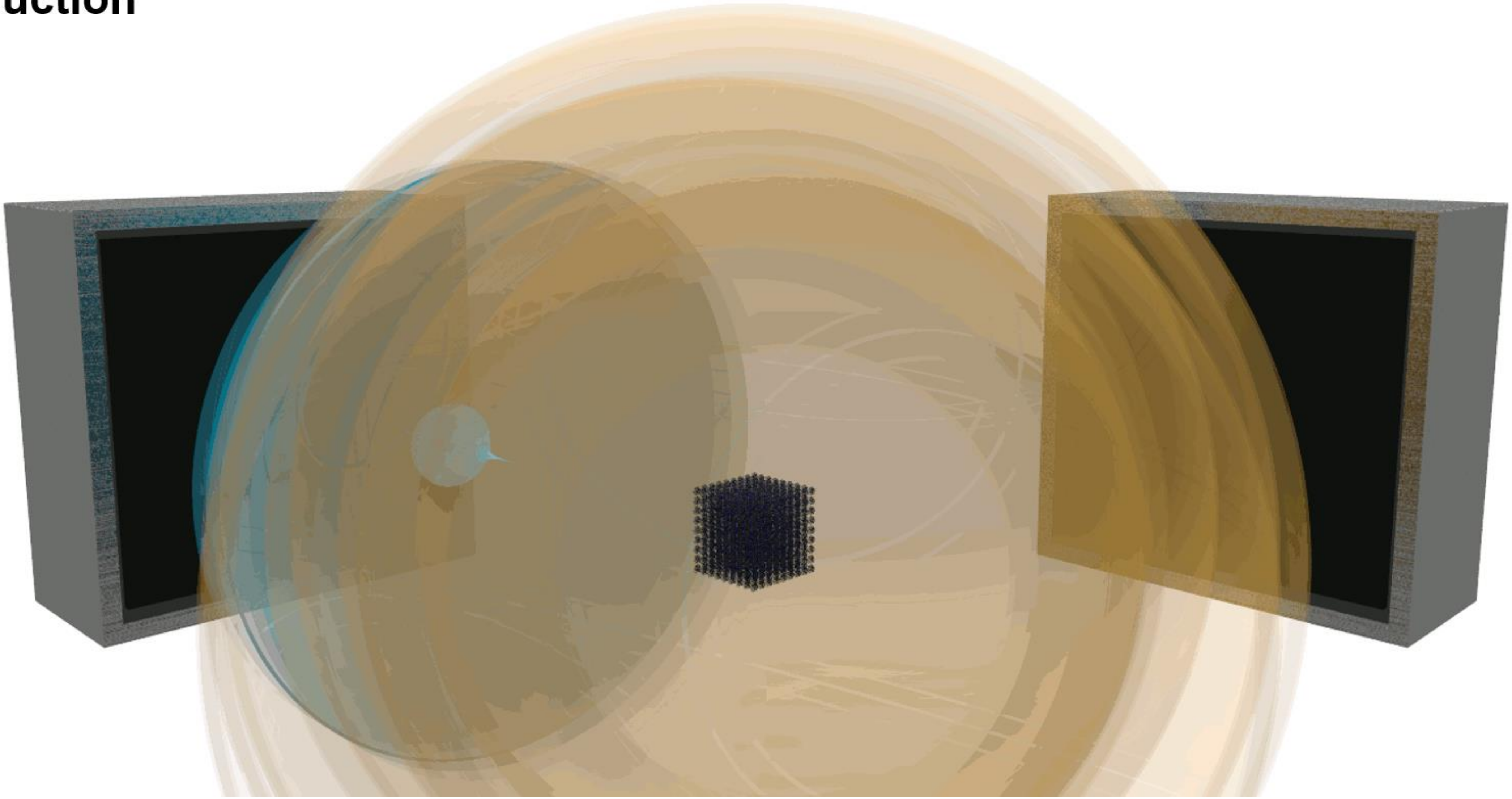
Introduction



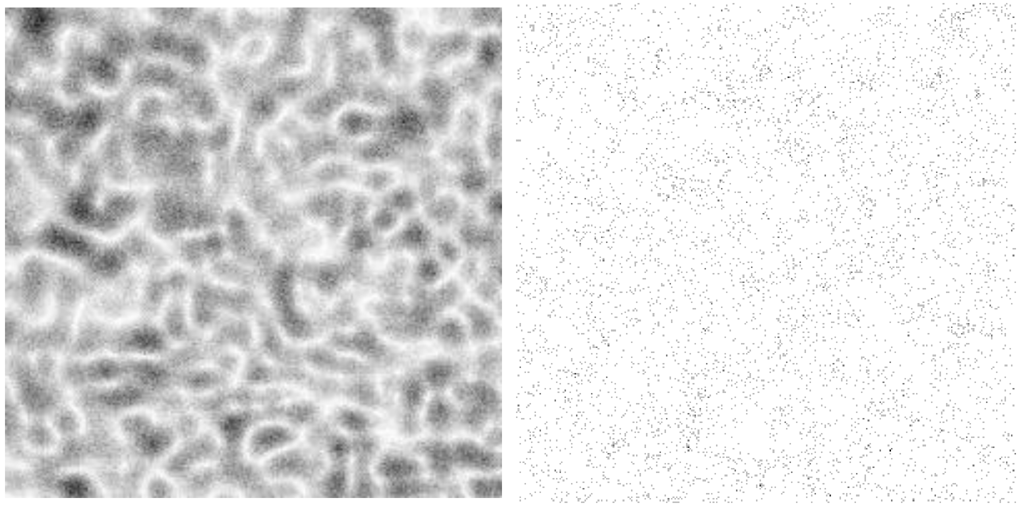
Introduction



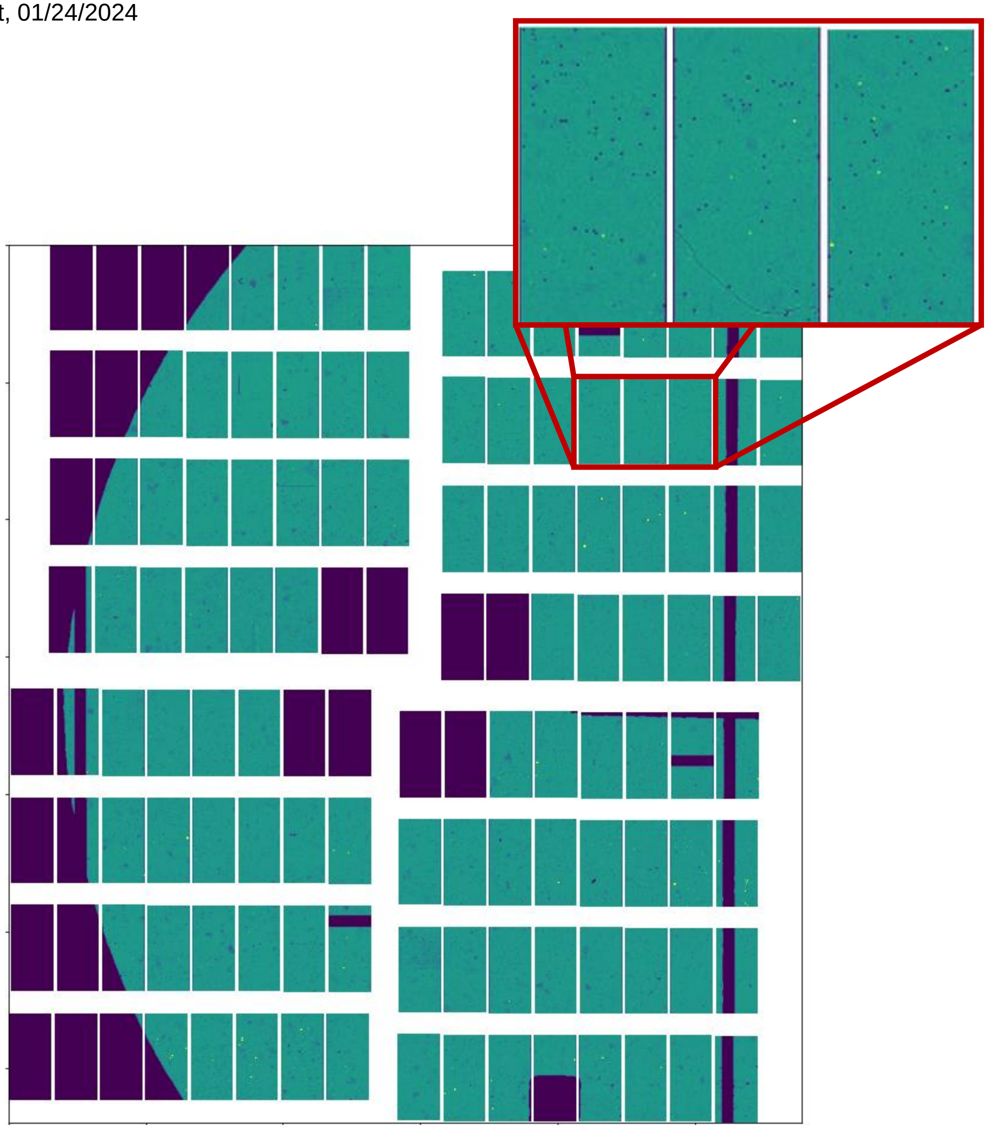
X-ray Flouresence

- Fluorescence is incoherent.
  - It averages to a flat intensity distribution,
  - Where all structural information is lost.

- When measured within timescales of the coherence time a speckle pattern is obtained.



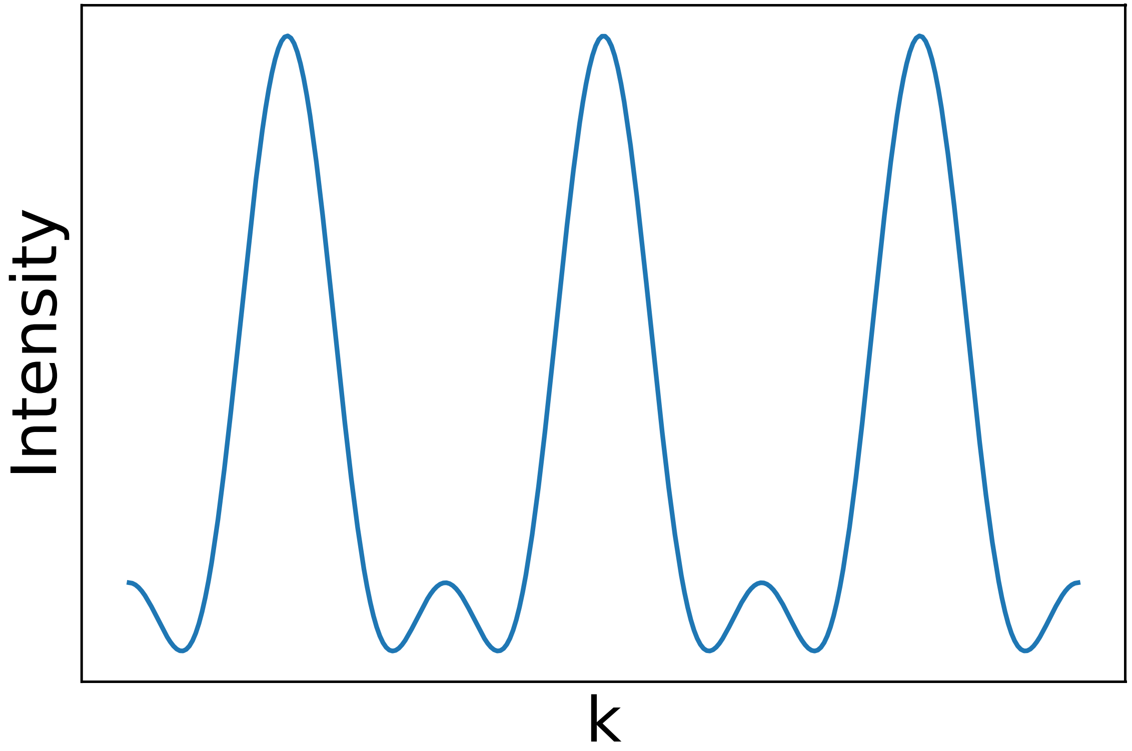
(speckles origin from interference)



average over 1 000 000 exposures of Cu K_{α}-fluorescence
(no structure, just flat intensity distribution)

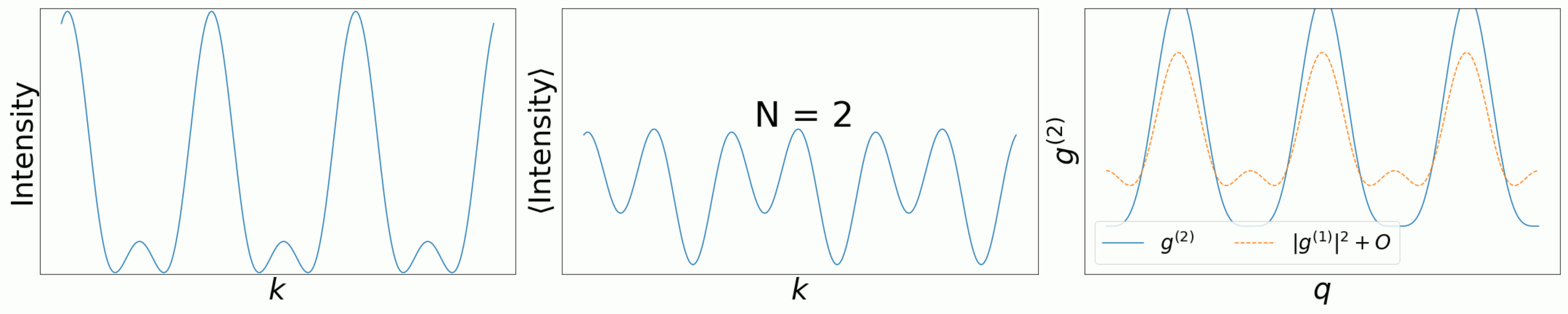
Photon-photon correlation

Think of a triple slit:



Photon-photon correlation

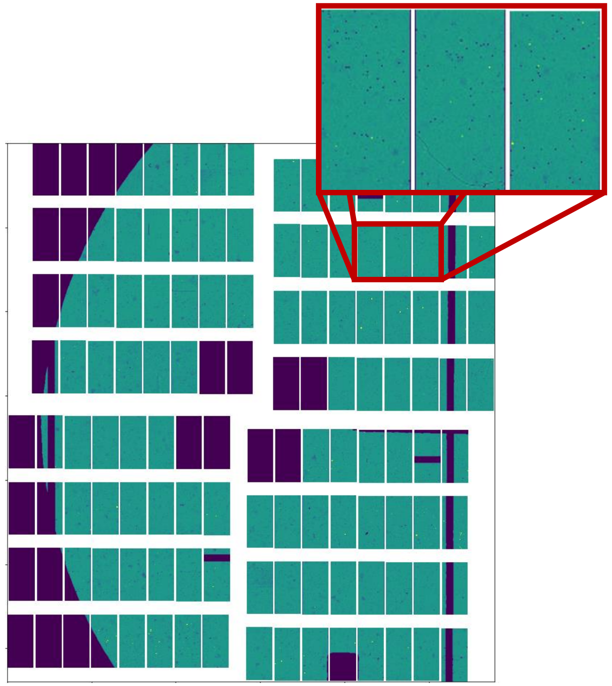
Think of a triple slit, with a random phase shift after each exposure:



$$g^{(2)} = \frac{\langle I(k)I(k+q)\rangle_k}{\langle I(k)\rangle_k^2}$$

Photon-photon correlation

- Fluorescence is incoherent.
  - Averages to flat intensity distribution.
- When measured within timescales of the coherence time τ_c, a speckle pattern is obtained.
- The speckle pattern encodes the "coherent pattern", which can be obtained via autocorrelation of the measured photons: $g^{(2)}(\vec{q}) = \frac{\langle I(\vec{k})\, I(\vec{k}+\vec{q})\rangle_{\vec{k}}}{\langle I(\vec{k})\rangle_{\vec{k}}^2}$.
- $g^{(2)}(\vec{q}) = 1 + \beta\left|g^{(1)}(\vec{q})\right|^2$ encodes the emitter distribution ρ: $\mathrm{g}^{(1)}(\vec{q}) = \frac{\mathfrak{F}[\rho(\vec{r})](\vec{q})}{\mathfrak{F}[\rho(\vec{r})](0)}$.

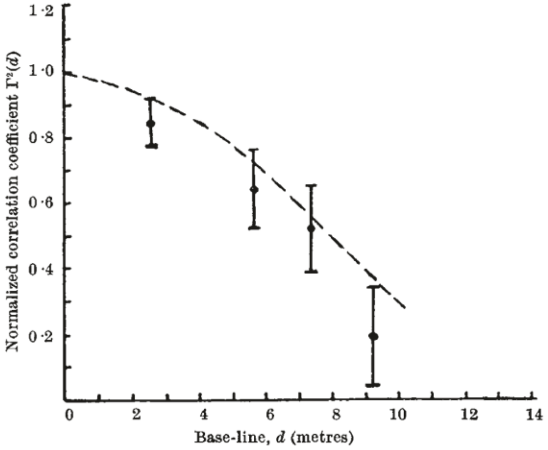


averaged fluorescence

Photon-photon correlation (short history)

- Robert Hanbury Brown and Richard Twiss used intensity interferometry to measure the diameter of stars.
- Intensity correlation signal must origin from multi-photon interference.

[Sky & Telescope, vol. 28, pp. 2-7, 1964]

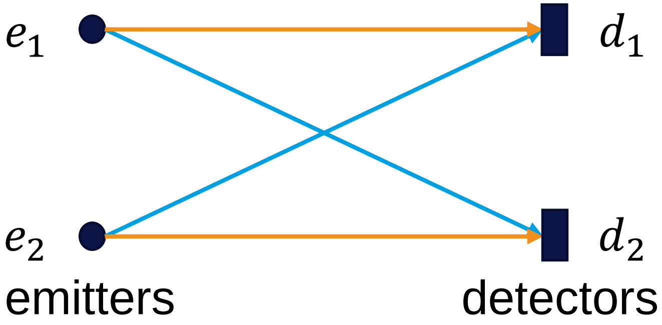


[R. Hanbury Brown, R. Twiss, Nature, Vol 178 Springer p. 1046-1048 (1956)]
Comparison between the values of the normalized correlation coefficient $\Gamma^2(d)$observed from Sirius and the theoretical values for a star of angular diameter 0.0063".

- Contradicted Paul Diracs view that "each photon [...] only interferes with itself "

[Paul Dirac, The principles of quantum mechanics, 1930]

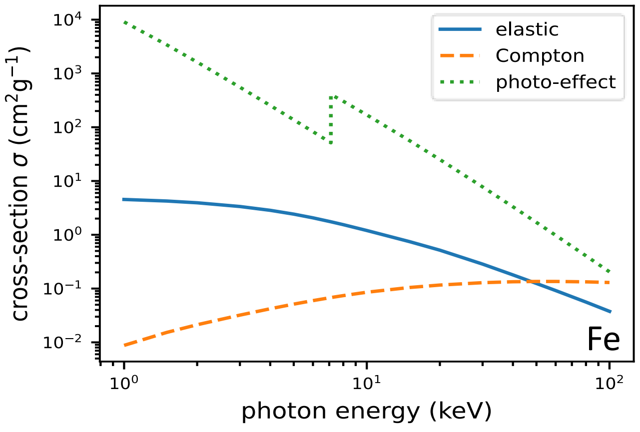
- Explanation by Ugo Fano: photon paths are not distinguishable and therefore interfere.

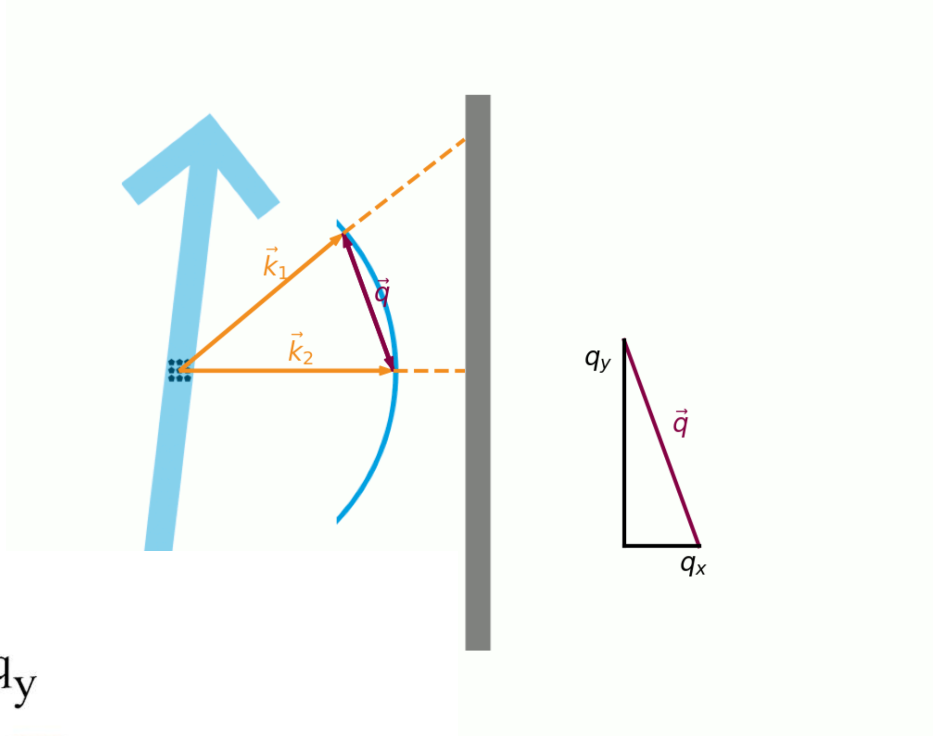


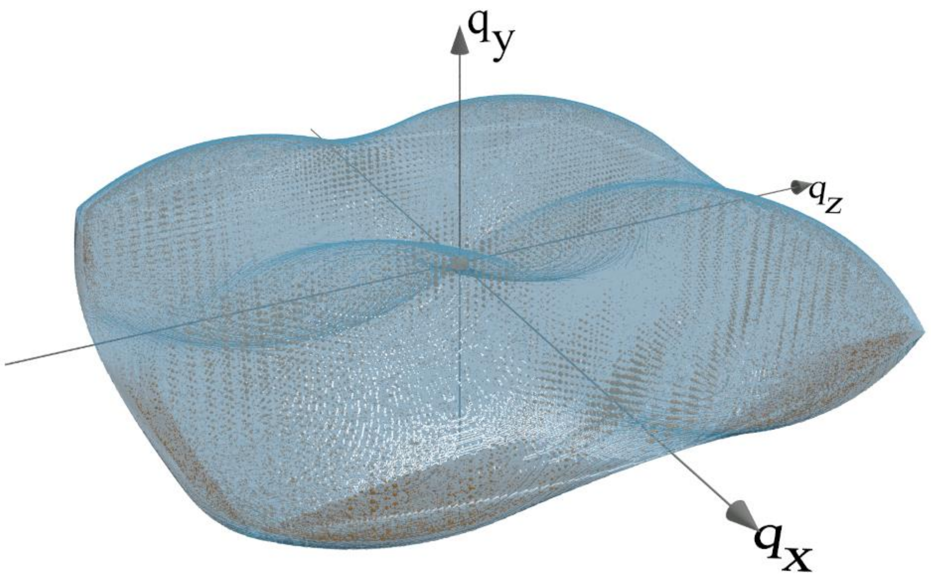
[U. Fano, "Quantum theory of interference effects in the mixing of light from phase-independent sources." *American Journal of Physics* 29.8 (1961): 539-545.]

Photon-photon correlation imaging (motivation)

- Element sensitivity.
- High cross-section.
- Fluorescence is isotropic.
- Translation invariant.
- 3D information and higher resolution.
- Can be used for FEL pulse length determination.







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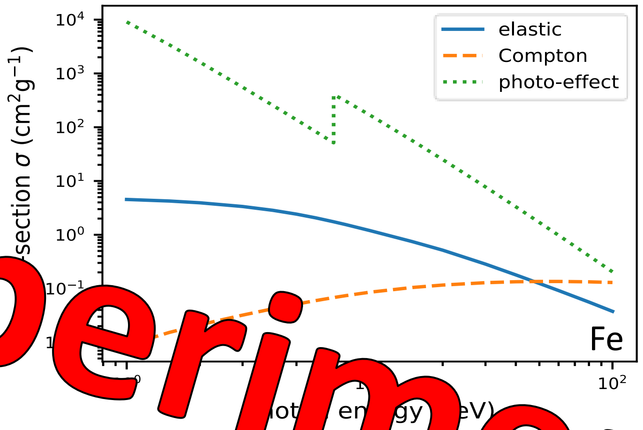
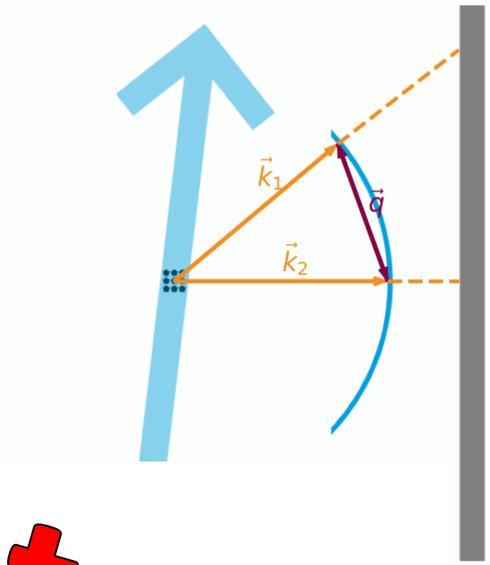
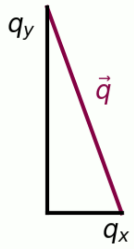
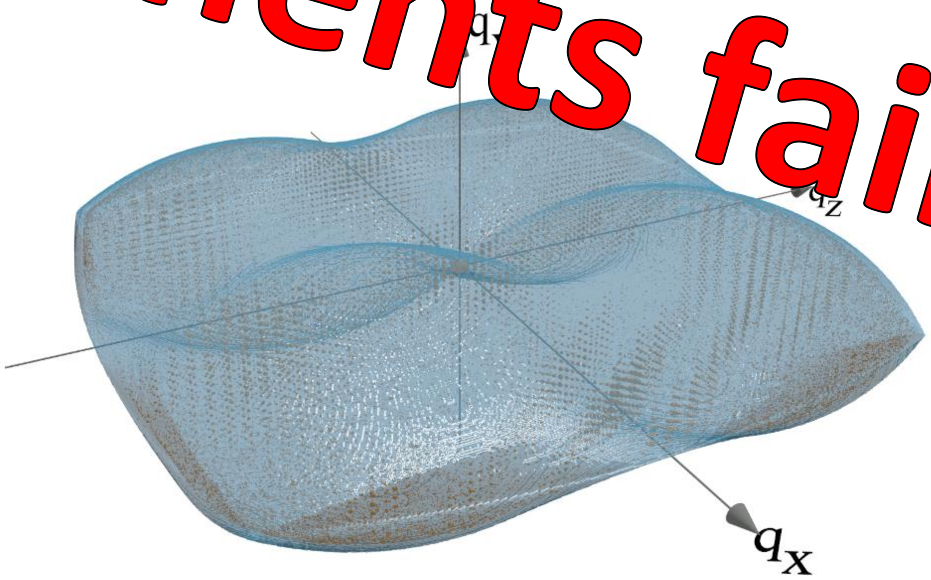








First experiments failed!

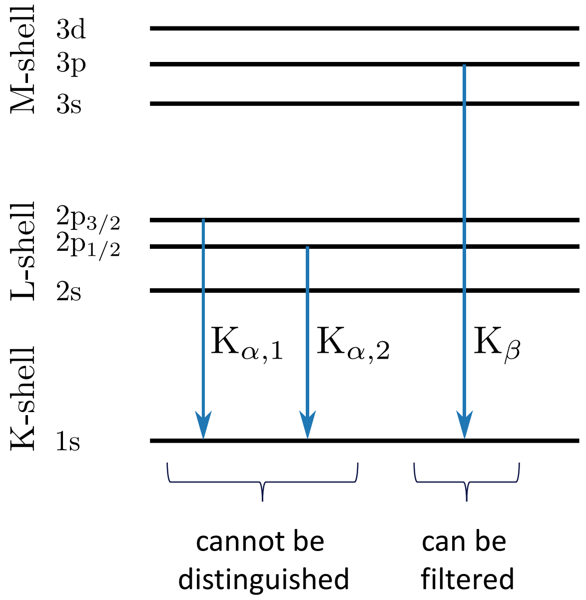
Usable signal - visibility factor β

Not all fluorescence photons can interfere. → independent coherent modes M.

→visibility factor $\beta = {}^{1}/_{M}$ modulates the "usable signal".

$$g^{(2)}(\vec{q}) = 1 + \beta \left|g^{(1)}(\vec{q})\right|^2$$

offset (1)   usable signal ($\left|g^{(1)}(\vec{q})\right|^2$)

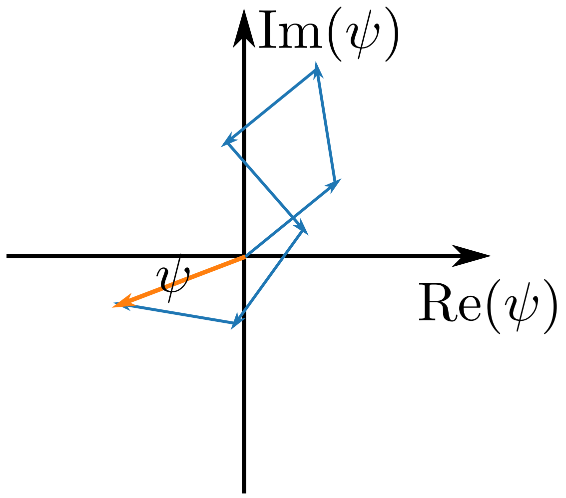


- Fluorescence is unpolarized → $\beta_{\text{pol}} = \frac{1}{2}$.
- We can not distinguish $K_{\alpha,1}$ and $K_{\alpha,2}$ → $\beta_{\text{lines}} \approx \frac{5}{9}$.
- Finite excitation pulse length → $\beta_{\text{pulse}} \approx \frac{\tau_c}{1.5\,T}$.

→ Expected visibility $\beta \approx \frac{5\,\tau_c}{27\,T}$.

- Further reading:
  Trost et al. J. Synchrotron Rad. (2023)

Signal-to-noise ratio - photon statistics



- Consider many (classical) emitters with random phases.
  Theirs intensity follow an exponential distribution

  $$P_{\mathrm{Exp}}(I|\mu) = \frac{1}{\mu}\, e^{-\frac{I}{\mu}} .$$

- We measure discrete photons (Poisson distributed) → Bose-Einstein distribution $P_{\mathrm{BE}}(n|\mu) = \frac{1}{1+\mu} + \left(\frac{\mu}{1+\mu}\right)^n$.

- Independent modes $M = \beta^{-1}$ yields a negative binomial distribution

  $$P_{\mathrm{NB}}(n|\mu, M) = \frac{M^M\, \mu^n\, \Gamma(M+n)}{(M+\mu)^{M+n}\, n!\, \Gamma(M)} ,$$

  with $\mathbb{E}(n) = \mu = \langle I \rangle$ and $\mathrm{Var}(n) = \mu + \beta\mu^2$.

Signal-to-noise ratio - photon statistics

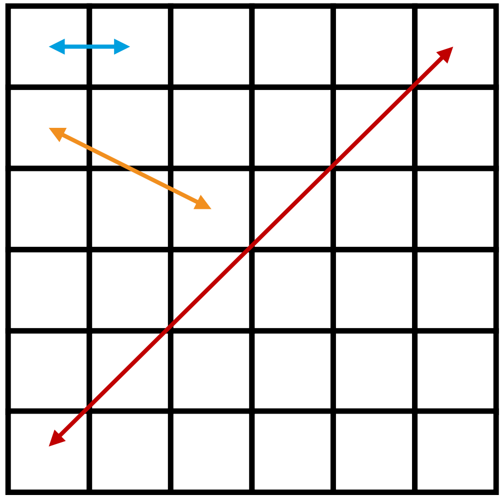
Photon statistics: negative binomial

- $\mathbb{E}(n) = \mu = \langle I \rangle$, $\mathrm{Var}(n) = \mu + \beta\mu^2$.

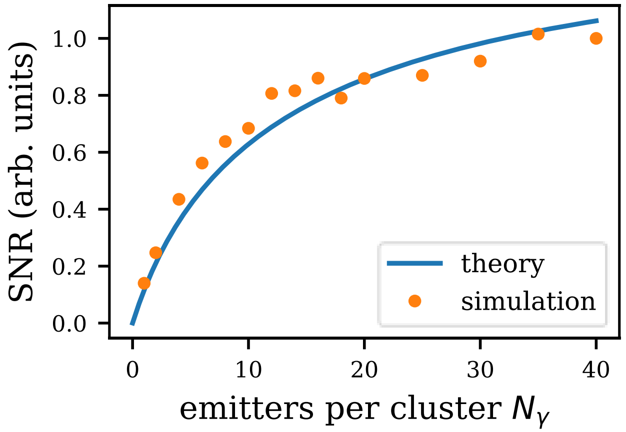
- Assuming uncorrelated intensities:

  → $\mathrm{Var}_{\mathrm{AC}}(\mu,\beta) = (\beta^2 + 4\beta)\mu^4 + (4 + 2\beta)\mu^3 + \mu^2$.

- $$\mathrm{SNR} = \frac{\beta \left| g^{(1)}(\vec{q}) \right|^2 \mu^2 \sqrt{N_\mathrm{p}\, C(\vec{q})}}{\sqrt{(\beta^2+4\beta)\mu^4 + (4+2\beta)\mu^3 + \mu^2}}$$

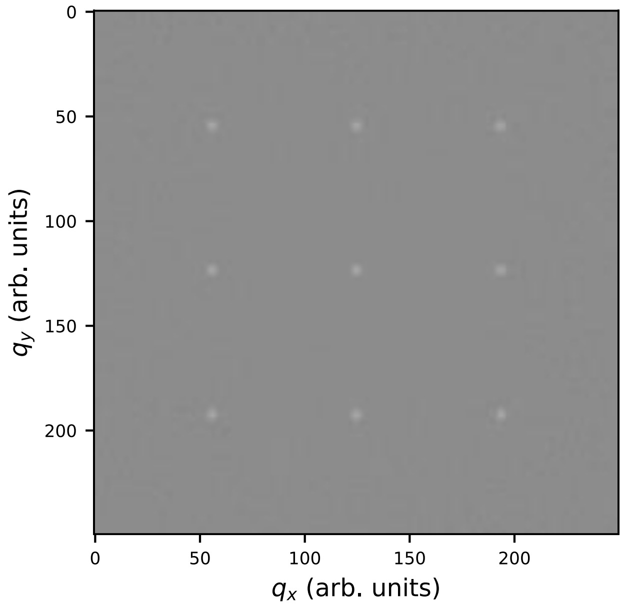


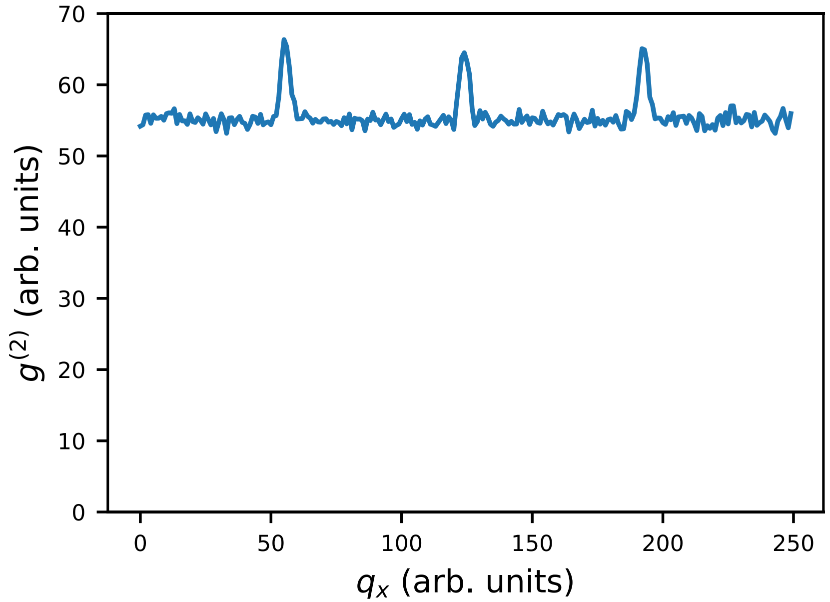
$C(\vec{q}) = 30$
$C(\vec{q}) = 20$
$C(\vec{q}) = 1$



Signal-to-noise ratio - complexity

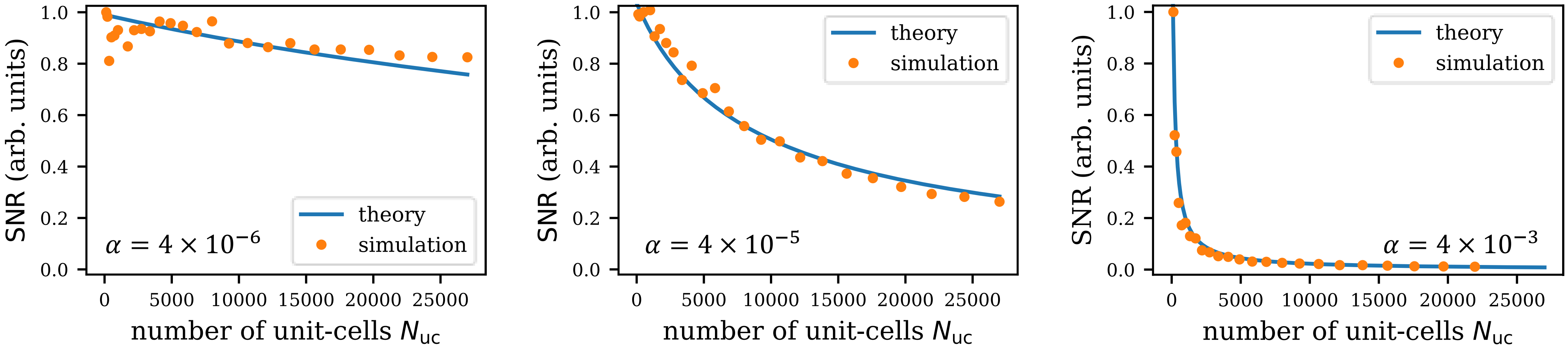
- The "usable signal" is on top of an offset $g^{(2)}(\vec{q}) = 1 + \beta\left|g^{(1)}(\vec{q})\right|^2$.
- However, the offset is noisy.
  Remember $\sigma_{AC} = \sqrt{(\beta^2 + 4\beta)\mu^4 + (4 + 2\beta)\mu^3 + \mu^2}$.
- For example crystals:
  - Bragg peaks are on top of the offset.
  - Offset is proportional to N_{uc}^2.
  - "Usable signal" is proportional to N_{uc}.
    ► Signal-to-offset ratio is $\propto 1/N_{\mathrm{uc}}$.





Signal-to-noise ratio - complexity

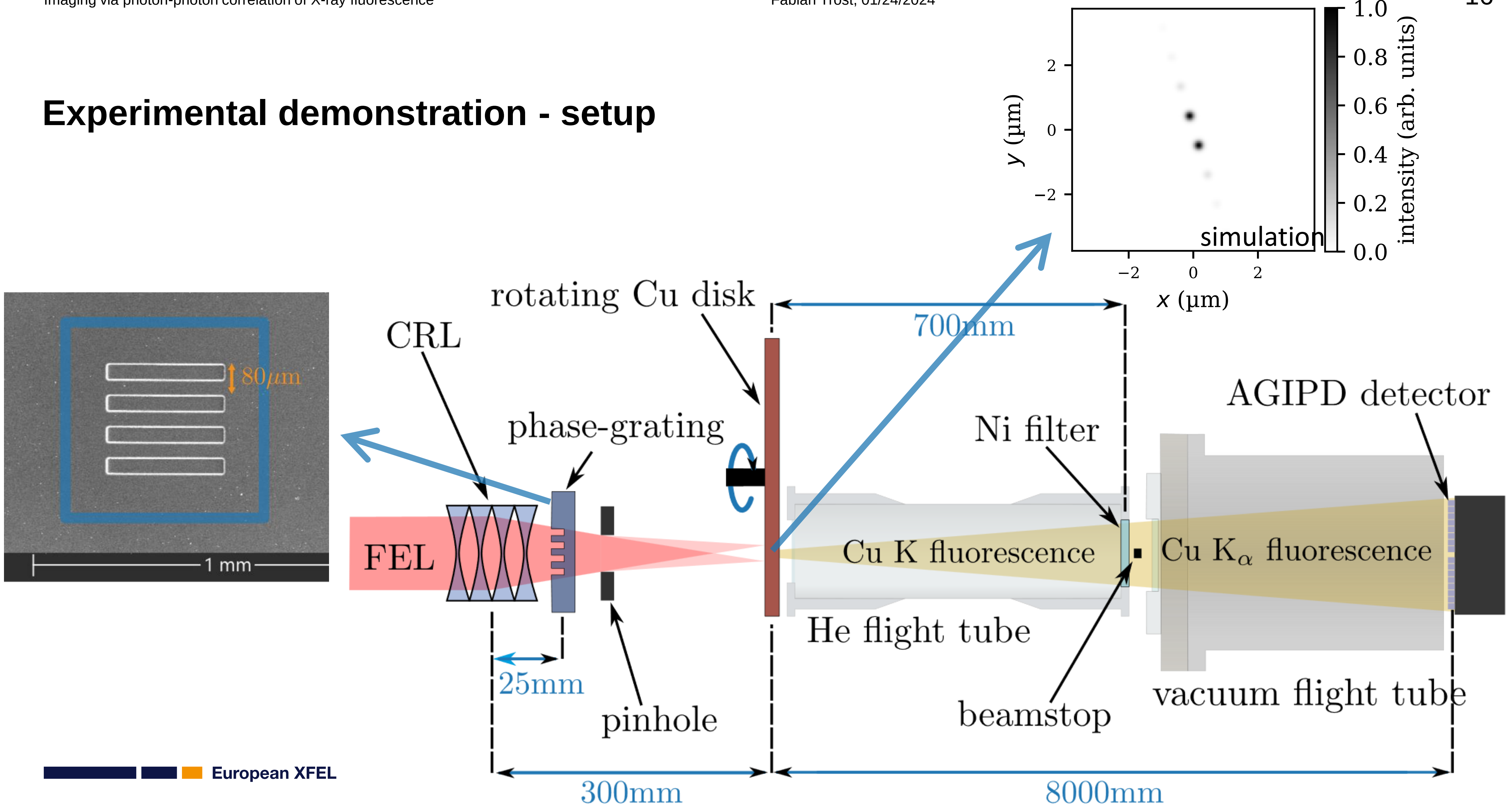
- Crystals: more unit cells lead to a poorer SNR, always!



- The offset grows usually faster than the usable signal with more resolvable emitters.
  $|\mathfrak{F}[\rho(\vec{r})](0)| \geq |\mathfrak{F}[\rho(\vec{r})](\vec{q})|$ if $\rho(\vec{r}) \geq 0 \ \forall \vec{r} \Rightarrow \left|g^{(1)}(\vec{q})\right|^2 \leq 1.$

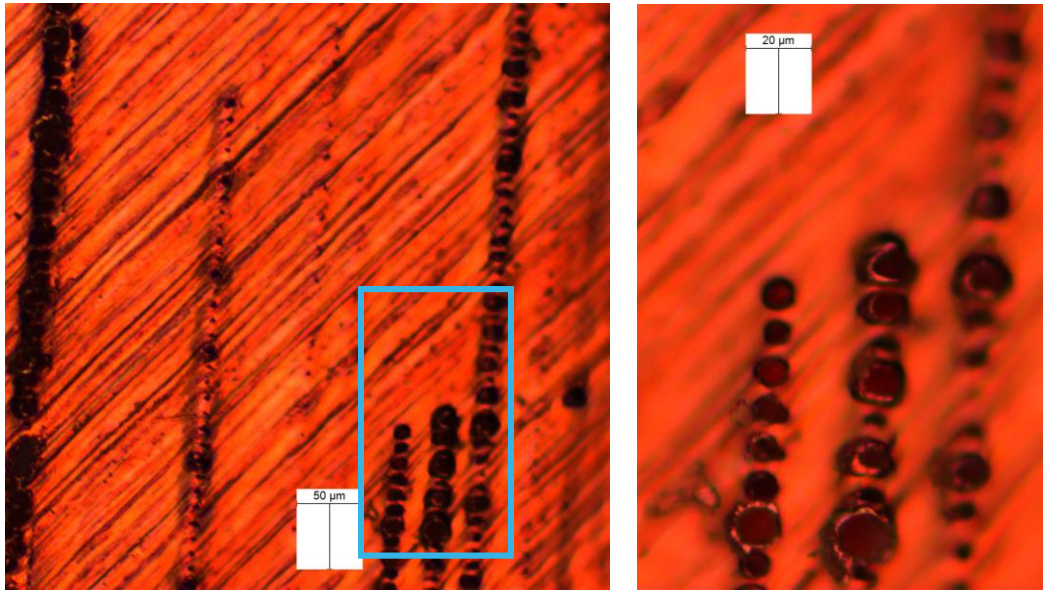
- Further reading: *Trost et al. 2020 New J. Phys. 22 083070*

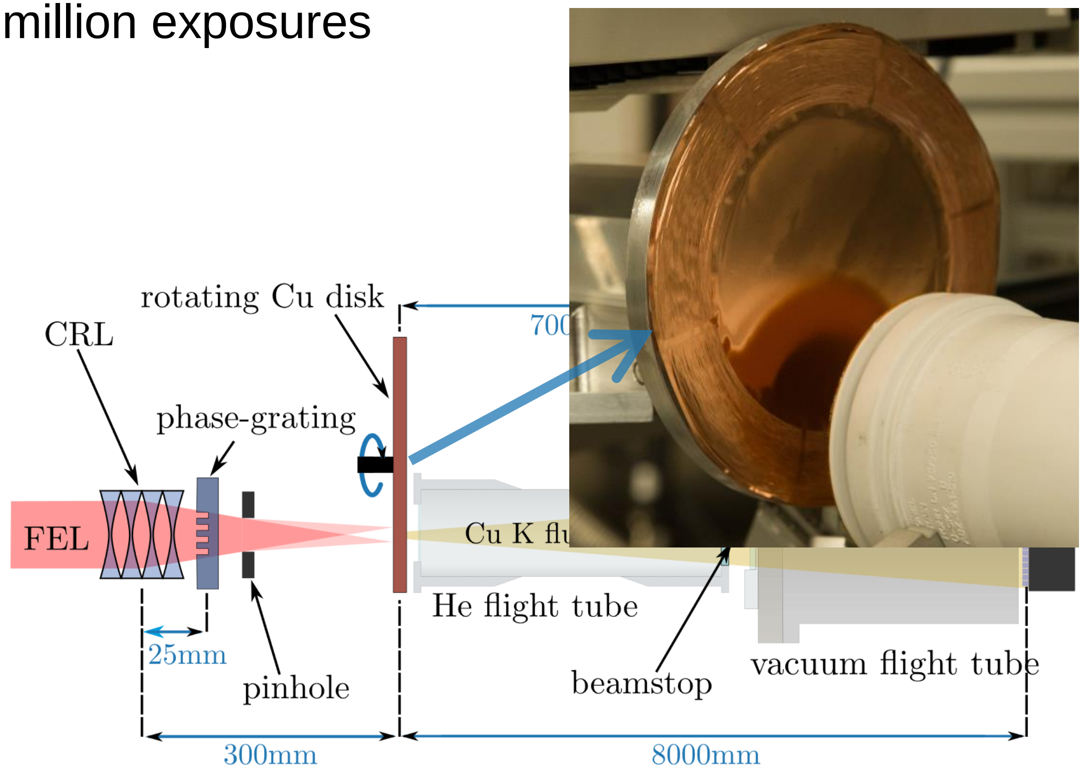
Experimental demonstration - setup



Experimental demonstration – Cu-target

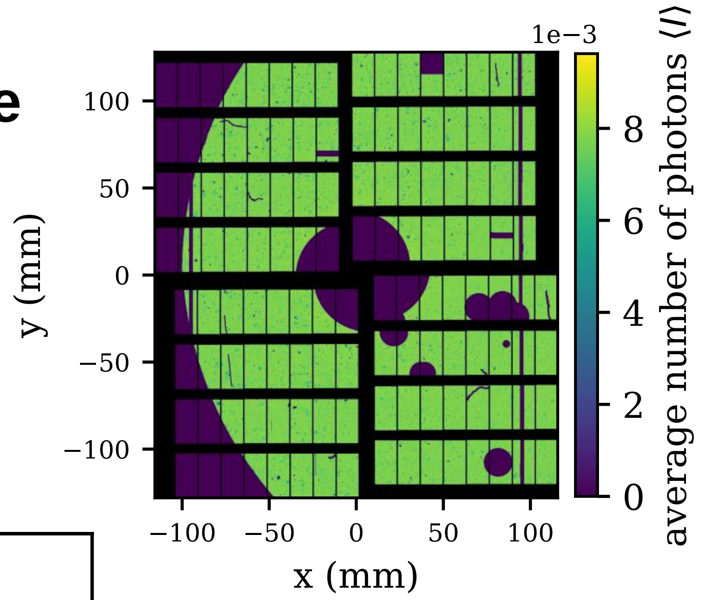
- EuXFEL provided trains with 444ns spaced pulses, each powerful enough to drill through the Cu foil.
- The Cu target is replaced after each pulse, by spinning the disk at around 4500 rpm.
- We were able to take a total of approximately 600 million exposures during the beamtime (3.1PB raw data)

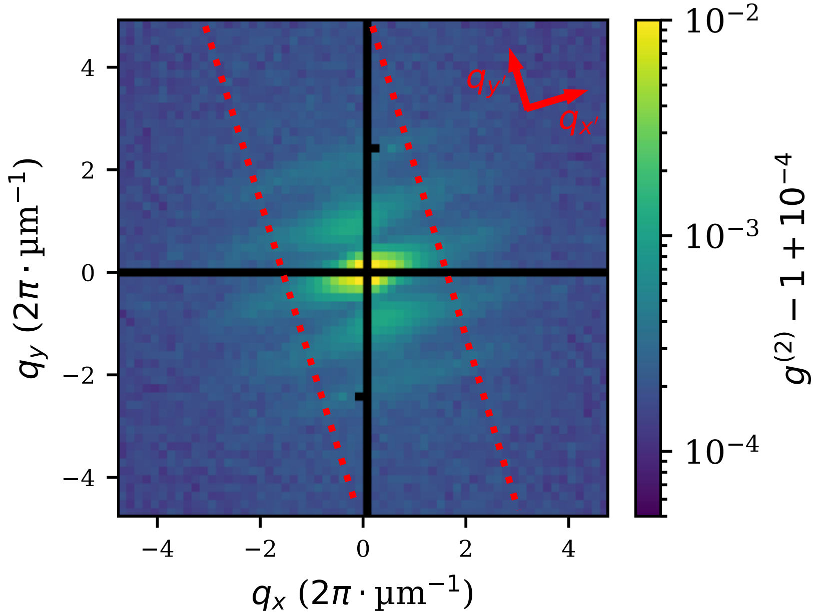


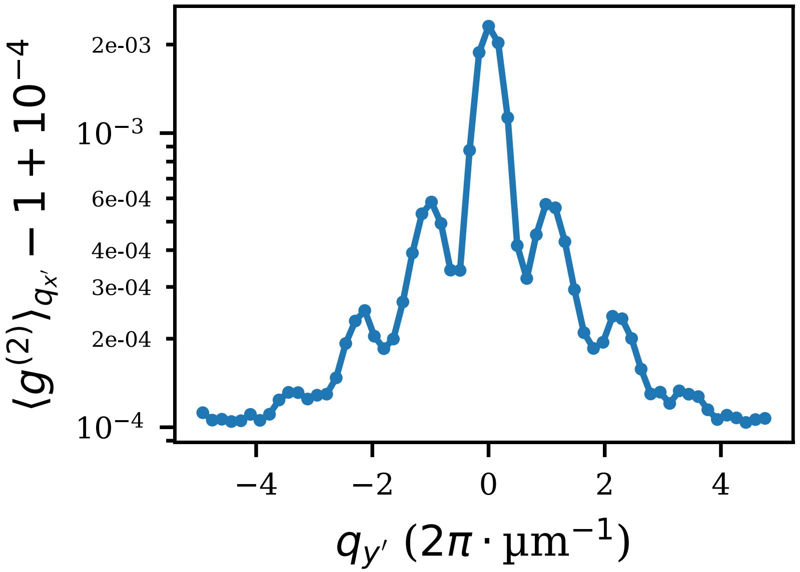


Experimental demonstration –
imaging via photon-photon correlation of X-ray fluorescence

- We measured 58 million patterns with the phase-grating.
- Averaged photon counts shows a flat intensity distribution. $\langle I \rangle = 0.0077$.
- Pattern-wise autocorrelation reveals fringes.



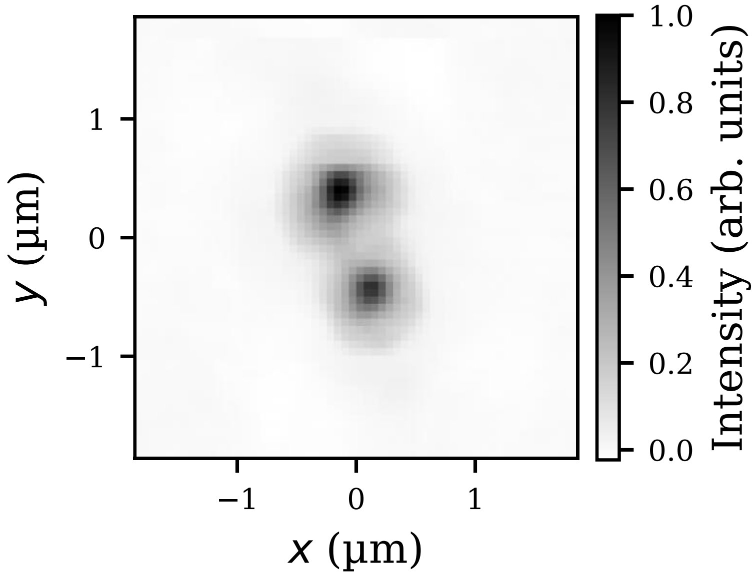


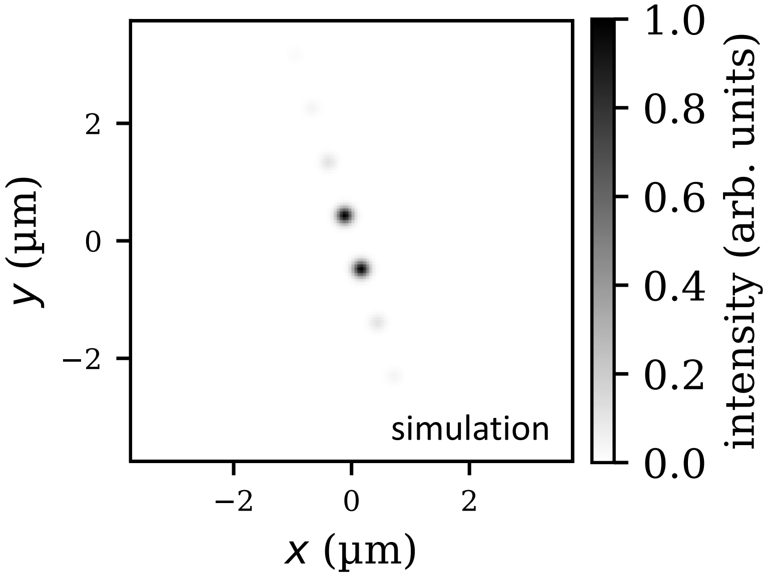


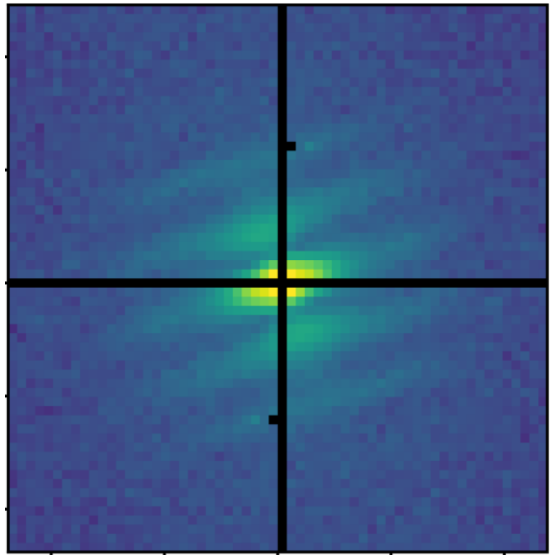
[Trost et al. Phys. Rev. Lett. 130, 173201]

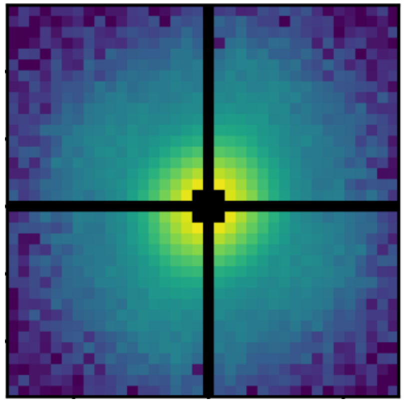
Experimental demonstration – imaging via photon-photon correlation of X-ray fluorescence

- Phasing reveals the emitter distribution.







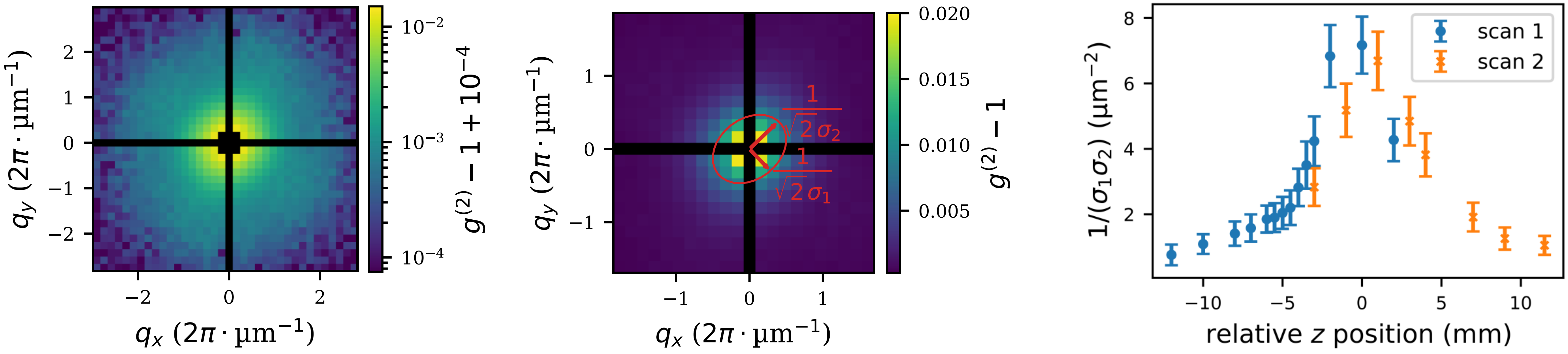


$g^{(2)} - 1$ without phase grating

- Integrated usable signal was around 33% with the grating in comparison to the case without grating.

FEL pulse and focus characterization

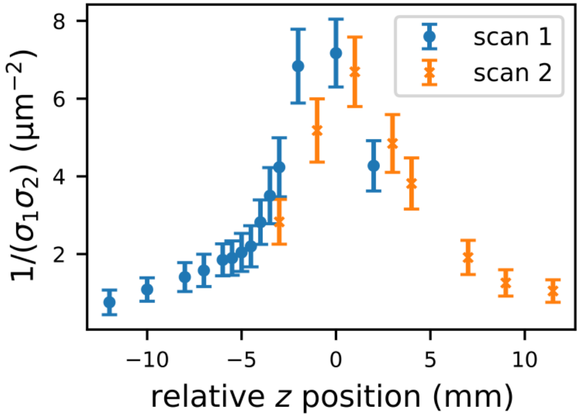
- Photon-photon correlation can be used for focus finding and characterization.

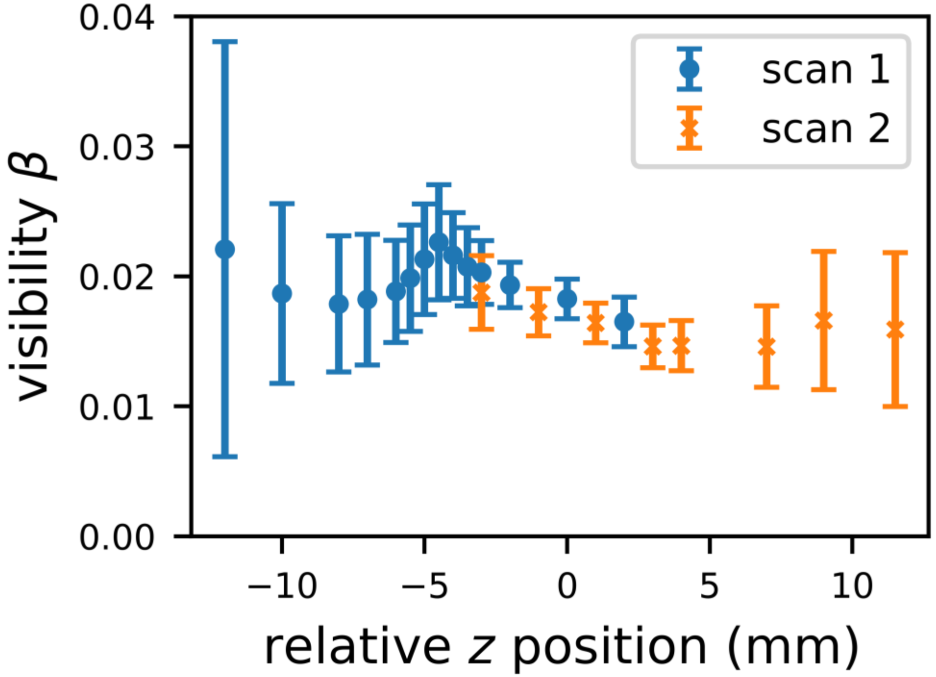


- Retrieved focus was $(640 \pm 40)\mathrm{nm} \times (480 \pm 30)\mathrm{nm}$ FWHM.
- Each data point consist of between 750 000 and 3 000 000 patterns, acquired within 5 - 20 min.
  ► Could be reduced to ~10 000 patterns for 10 times more photons →less than 5 sec.
- Insensitive to beam jitter.

FEL pulse and focus characterization

- Signal fitting also yields the visibility factor.



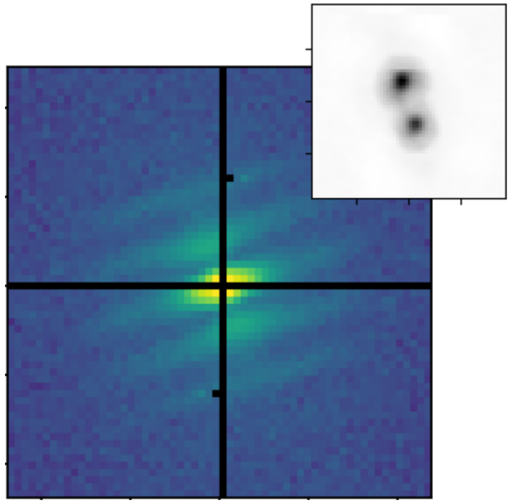


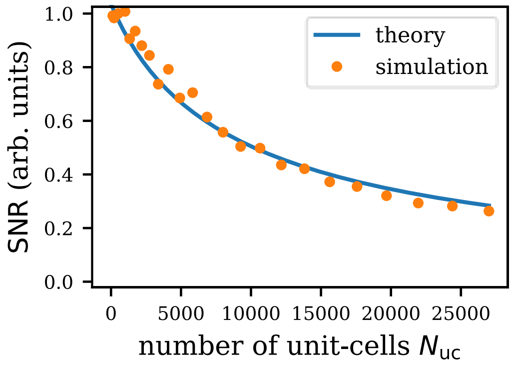
- We have determined $\beta = (0.018\ \pm 0.002)$, with $\beta \approx 0.185\frac{0.6\text{fs}}{T}$:
  - ▶ retrieved pulse duration $T = (6.2 \pm 0.8)\text{fs}$ (FWHM).
- Further reading: *Trost et al. Phys. Rev. Lett. 130, 173201*

[also demonstrated by Inoue et al., see J. Synchrotron Rad. 26, 2050–205 (2019).]

Summary and Outlook

- Imaging via correlation of X-ray fluorescence is possible.
  - can be used for robust focus measurements.
- Imaging of complex emitter distributions is complicated by a loss in SNR.
- Visibility is dependent on the excitation pulse duration.
  - can be used to determine the pulse length.





Next steps:

- Demonstrate single particle imaging via photon-photon correlation.
- Implement photon-photon correlation imaging as a robust tool for beam characterization.
- Advent of attosecond pulses on FELs will open new possibilities for X-ray fluorescence correlation imaging.

Thank you!

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