Methoden moderner Röntgenphysik: Streuung und Abbildung

Exercise 10	Vorlesung zum Haupt- oder Masterstudiengang Physik, SoSe 2021
	G. Grübel, O. Seeck, F. Lehmkühler, A. Philippi-Kobs, V. Markmann, M. Martins
Location	online

Date	Tuesday	12:30 - 14:00	(starting 6.4.)
	Thursday	8:30 - 10:00	(until 8.7.)

1. SPECKLE CONTRAST WITH DISTRIBUTION FUNCTIONS

The speckle contrast can be obtained from the distribution of 0-, 1-, 2- etc. photon events at low intensities. This distribution is modeled by a negative binomial distribution $P_{nb}(i) = \frac{\Gamma(i+M)}{\Gamma(M)\Gamma(i+1)}\left(1+\frac{M}{\langle i\rangle}\right)^{-i}\left(1+\frac{\langle i\rangle}{M}\right)^{-M}$. Here, i = 0, 1, 2, 3,… denotes the number of photon in a pixel, ⟨i⟩ the average count rate in the detector and M=1/β.

a) Determine the speckle contrast β from the ratio of 1 and 2 photon events.

c) Estimate the error of the contrast determination using an average count rate of $5 \cdot 10^{-3}$ photons per pixel and a megapixel detector chip. Consider a speckle contrast of 50 %.

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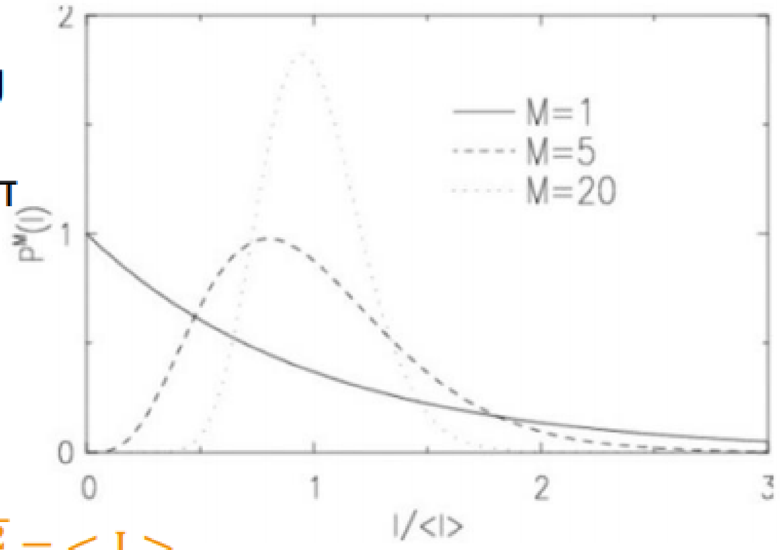
Speckle Statistics

If the source is fully coherent and the scattering amplitudes and phases of the scattering are statistically independent and distributed over 2π one finds for the probability amplitude of the intensities:

$$P(I) = \left(\frac{1}{<I>}\right) e^{\frac{-I}{<I>}}$$



Mean: $<I>$ **Std. Dev. σ:** $\sqrt{<I^2> - <I>^2} = <I>$

Contrast: $ß = \sigma^2 / <I>^2 = 1$

<u>Partially coherent illumination:</u> the speckle pattern is the sum of M independent speckle patterns

$$P_M(I) = M^M \bullet \frac{\left(\frac{1}{<I>}\right)^{M-1}}{\Gamma(M)<I>} \bullet e^{-\frac{MI}{<I>}}$$

Mean: $<I>$; $\sigma = \frac{<I>}{\sqrt{M}}$ $ß = 1/M$

DESY

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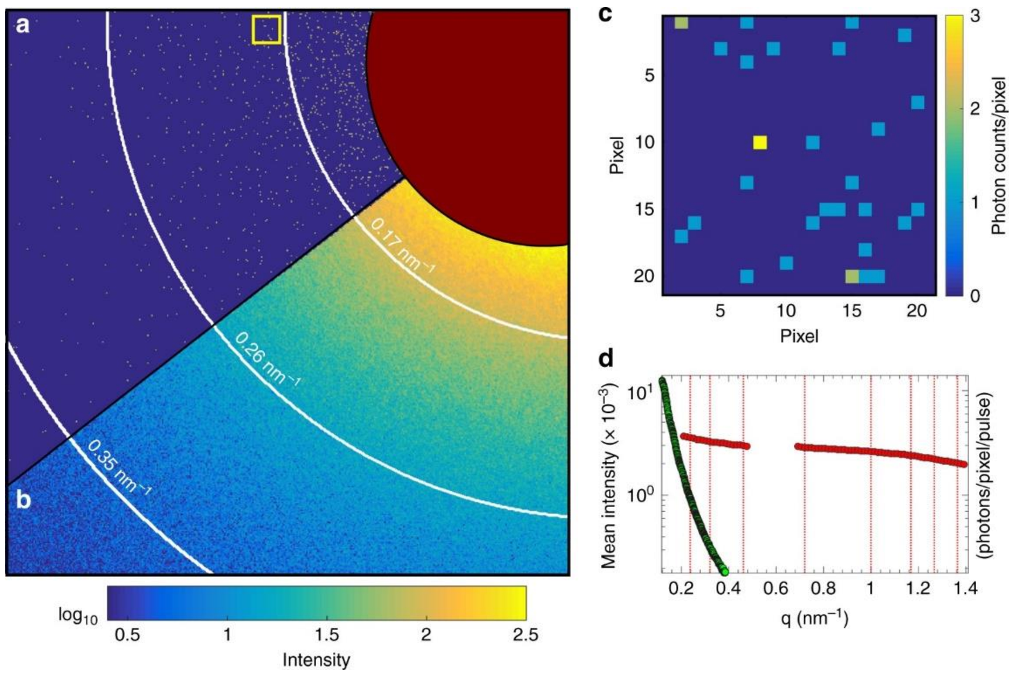
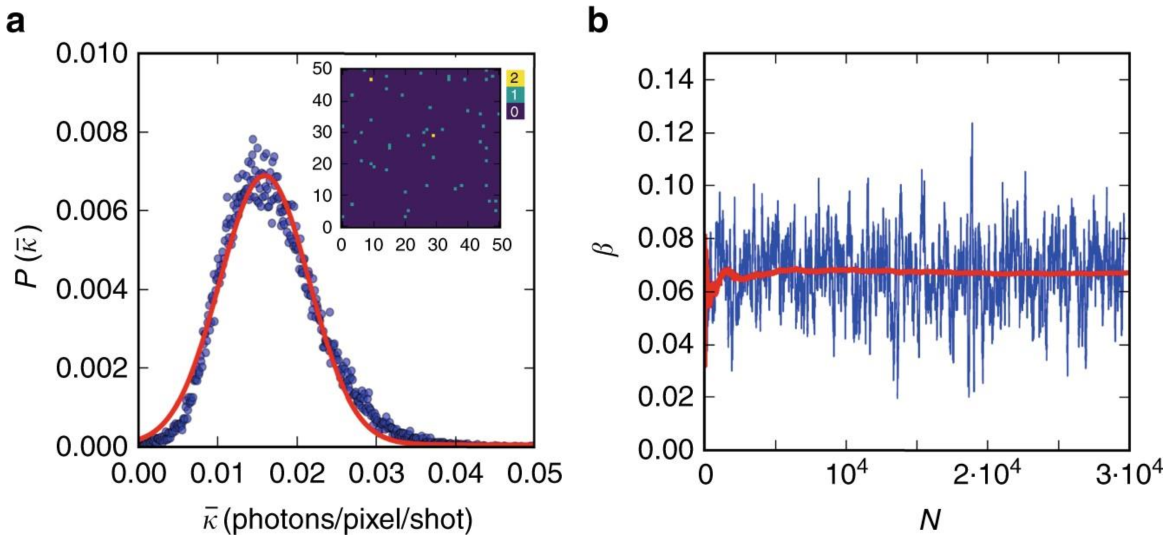


Nature Comm. 9, 1704 (2018)

Low-intensity diffraction patterns

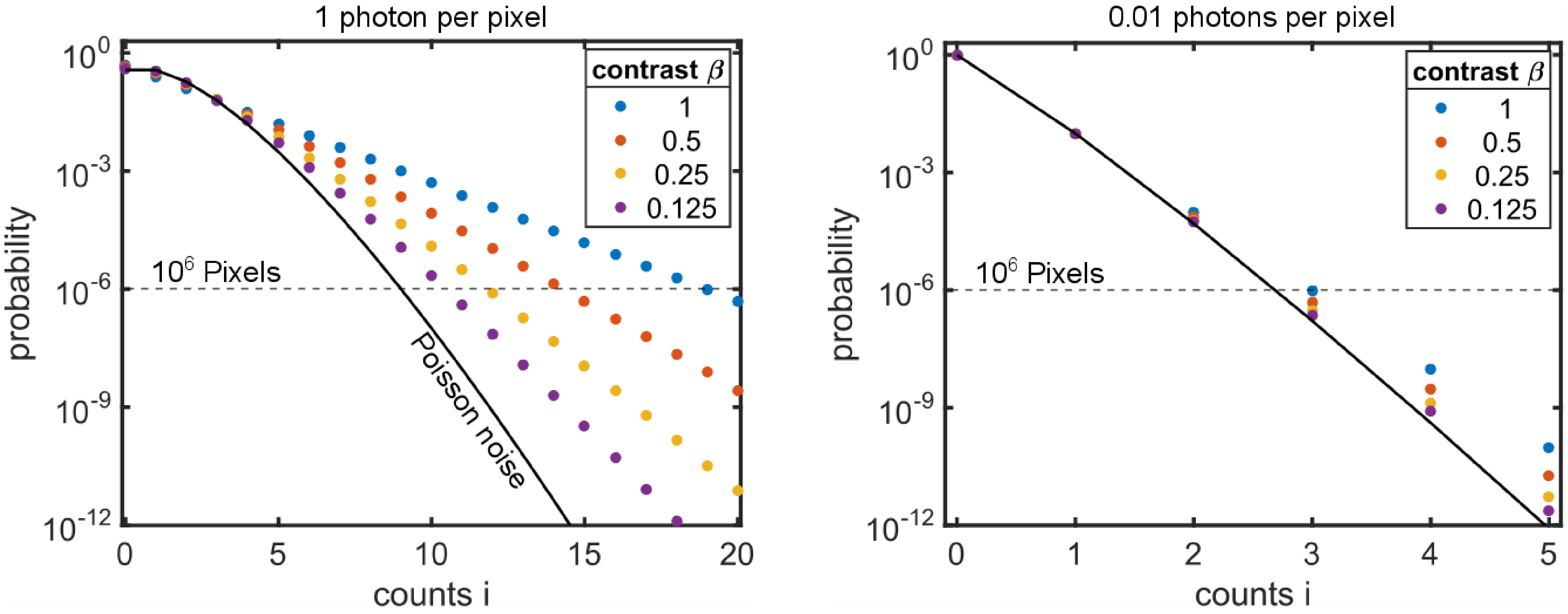
→ Dynamics via XSVS at FELs



Nature Comm. 9, 1917 (2018)

In experiments:

- Intensity histograms from whole detector (treated as one q value) or certain region of interests (e.g. particular q)
- Model distribution with negative binomial function $P_{nb}(i) = \frac{\Gamma(i+M)}{\Gamma(M)\Gamma(i+1)}\left(1+\frac{M}{\langle i\rangle}\right)^{-i}\left(1+\frac{\langle i\rangle}{M}\right)^{-M}$



a. Contrast calculation at low count rates

$$P_{nb}(i) = \frac{\Gamma(i+M)}{\Gamma(M)\Gamma(i+1)}\left(1+\frac{M}{\langle i\rangle}\right)^{-i}\left(1+\frac{\langle i\rangle}{M}\right)^{-M}$$

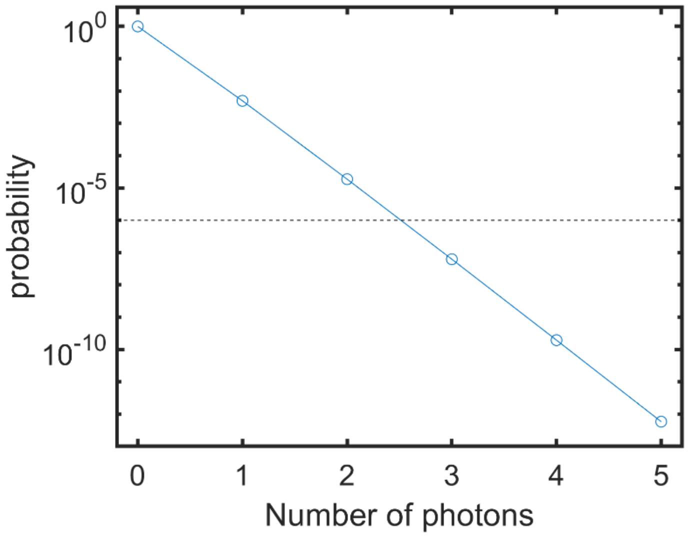
Gamma function:

- $\Gamma(1) = 1$
- $\Gamma(x+1) = x\Gamma(x)$, in particular for $n \in \mathbb{N}: \Gamma(n) = (n-1)!$

- $P_{nb}(1) = \frac{\Gamma(1+M)}{\Gamma(M)\Gamma(2)}\left(1+\frac{M}{\langle i\rangle}\right)^{-1}\left(1+\frac{\langle i\rangle}{M}\right)^{-M} = M\left(1+\frac{M}{\langle i\rangle}\right)^{-1}\left(1+\frac{\langle i\rangle}{M}\right)^{-M}$, using $\Gamma(1+M) = M\Gamma(M)$ and $\Gamma(2) = 1$
- $P_{nb}(2) = \frac{\Gamma(2+M)}{\Gamma(M)\Gamma(3)}\left(1+\frac{M}{\langle i\rangle}\right)^{-2}\left(1+\frac{\langle i\rangle}{M}\right)^{-M} = \frac{(M+1)M}{2}\left(1+\frac{M}{\langle i\rangle}\right)^{-2}\left(1+\frac{\langle i\rangle}{M}\right)^{-M}$, using $\Gamma(2+M) = (1+M)M\Gamma(M)$
- $r = \frac{P_{nb}(2)}{P_{nb}(1)} = \frac{M+1}{2}\left(1+\frac{M}{\langle i\rangle}\right)^{-1} \Rightarrow \ldots \Rightarrow \beta = \frac{1}{M} = \frac{2r-\langle i\rangle}{\langle i\rangle(1-2r)}$

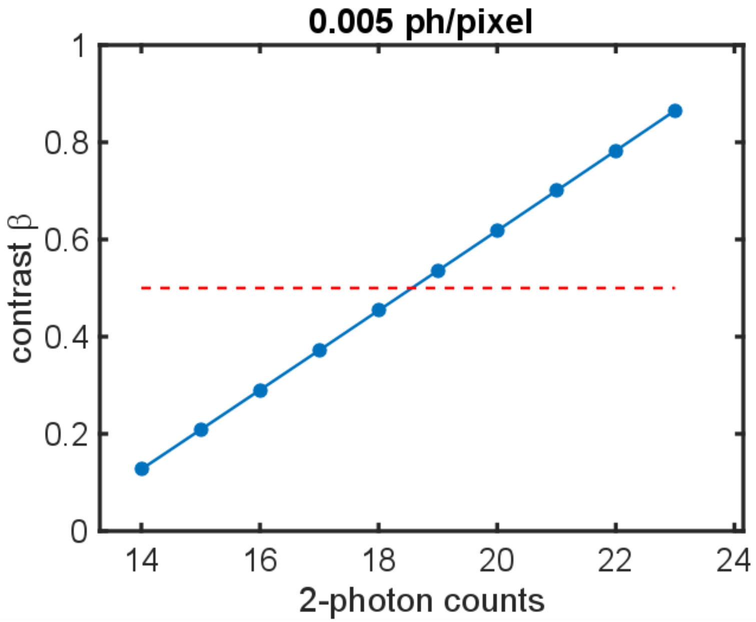
b. Assume 1 Megapixel Detector & $\langle i \rangle = 5 \cdot 10^{-3}$ ph/pixel at $\beta = 0.5 \Rightarrow M = 2$

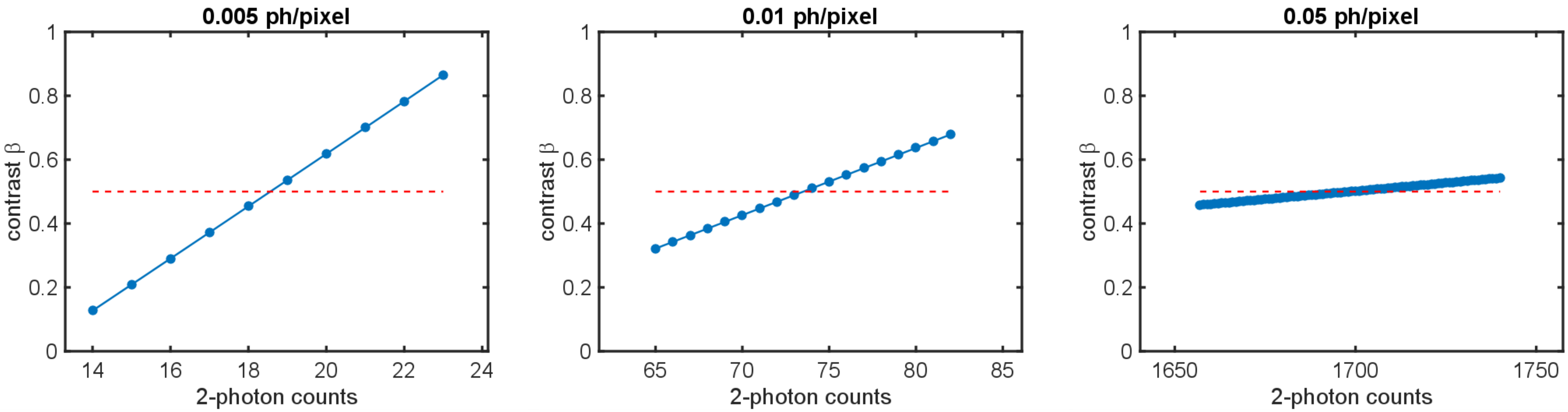
$$P_{nb}(i) = \frac{\Gamma(i+2)}{\Gamma(2)\Gamma(i+1)}\left(1+\frac{2}{5\cdot 10^{-3}}\right)^{-i}\left(1+\frac{5\cdot 10^{-3}}{2}\right)^{-2} = \frac{0.995(i+1)}{400^i}$$



- One photon counts: $P_{nb}(1) \cdot 10^6 \sim 4975$ (following distribution function)
- Two photon counts: $P_{nb}(2) \cdot 10^6 \sim 18.6$

- Consider counting statistics, e.g., detector counts $18.6 \pm \sqrt{18.6} \approx 18.6 \pm 4.3$ two photon events
- This results in a contrast variation between 0.1 and 1!
- Need to measure many speckle patterns.




0.005 ph/pixel
contrast β
2-photon counts
0
0.2
0.4
0.6
0.8
1
14
16
18
20
22
24
0.01 ph/pixel
contrast β
2-photon counts
65
70
75
80
85
0.05 ph/pixel
contrast β
2-photon counts
1650
1700
1750

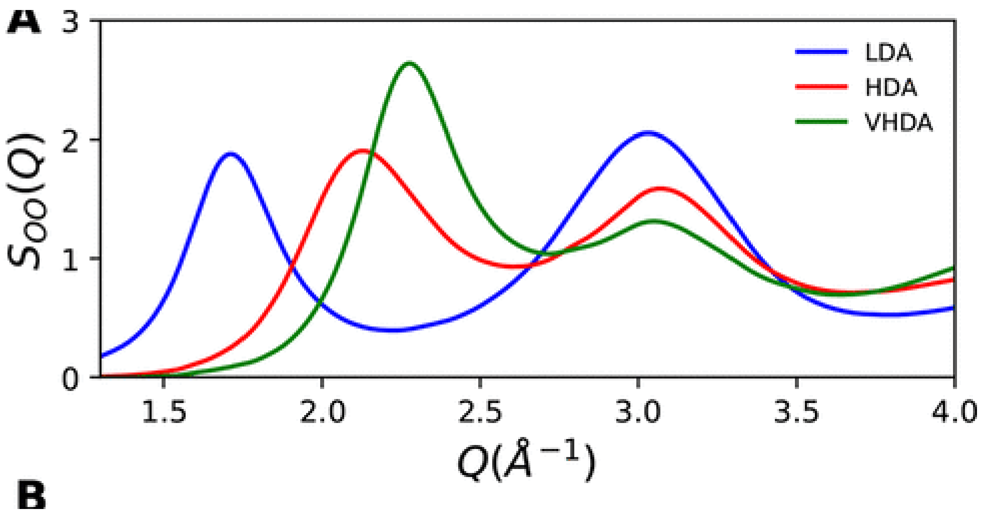
2. XCCA ON LIQUID WATER

To determine the local order in the vicinity of the next-neighbour distance in amorphous Ice, an XCCA experiment should be performed at an XFEL source. Water microdroplets will be injected into the sample chamber by a liquid jet. The next-neighbour distance (O-O distance) is approximately 2.75 Å.

1. Which q-range needs to be covered in the experiment?
2. As a coherent scattering experiment is performed, speckles should be resolved. The X-ray energy is 9 keV, an AGIPD 1M is used as detector (1024 x 1024 pixels with 200 µm x 200 µm size each).
   i. Can speckles be resolved in the q-range found in a)? Consider using a focused beam of 500 nm size.
   ii. What size is needed for an AGIPD-type detector and where should it be placed to measure a speckle signal with 1:1: speckle-to-pixel size?
3. The water freezes during the experiment and forms hexagonal ice. The first four Bragg peak ({100}, {002}, {101}, and {102}) are found at q-values of 1.608 $Å^{-1}$, 1.711 $Å^{-1}$, 1.822 $Å^{-1}$, und 2.348 $Å^{-1}$. Hexagonal ice has the space group P6_3/mmc, i.e. a hexagonal lattice with $a=b<c$ and $\alpha=\beta=90°$ and $\gamma=120°$.
   i. Determine the lattice constant a and c from the measured peak positions.
   ii. Which symmetries would be measured in an XCCA experiment for the first three peaks? Hint: Correlations can only be found between equivalent Bragg reflections, that originate from the same crystallite (see e.g. http://journals.iucr.org/j/issues/2016/06/00/zg5001/index.html). Maxima of the correlation function $C(\Delta)$ can only be found for particular correlation angles Δ at q-values of the Bragg peaks.

1. Which q-range needs to be covered?

Structure factor peak expected in the vicinity of $q \approx \frac{2\pi}{2.75\ \text{Å}} = 2.285\ \text{Å}^{-1}$



J. Phys. Chem. B 2018, 122, 30, 7616−7624

2. i. Can speckles be resolved in the q-range found in 1.?

Speckle size: $s \approx \lambda \cdot \frac{D}{b}$, with $b = 500$ nm as beam size. Speckle should match pixel size: $s = 200$ µm.

Wave length λ is obtained from the energy: $E = h\nu = \frac{hc}{\lambda} \;\Rightarrow\; E\,[\mathrm{keV}] = \frac{12.398}{\lambda[\mathrm{Å}]} \;\Rightarrow\; \lambda = 1.378$ Å.

So we can obtain a sample-detector distance of $D = b \cdot \frac{s}{\lambda} \approx 0.73$ m that fullfils the needs.

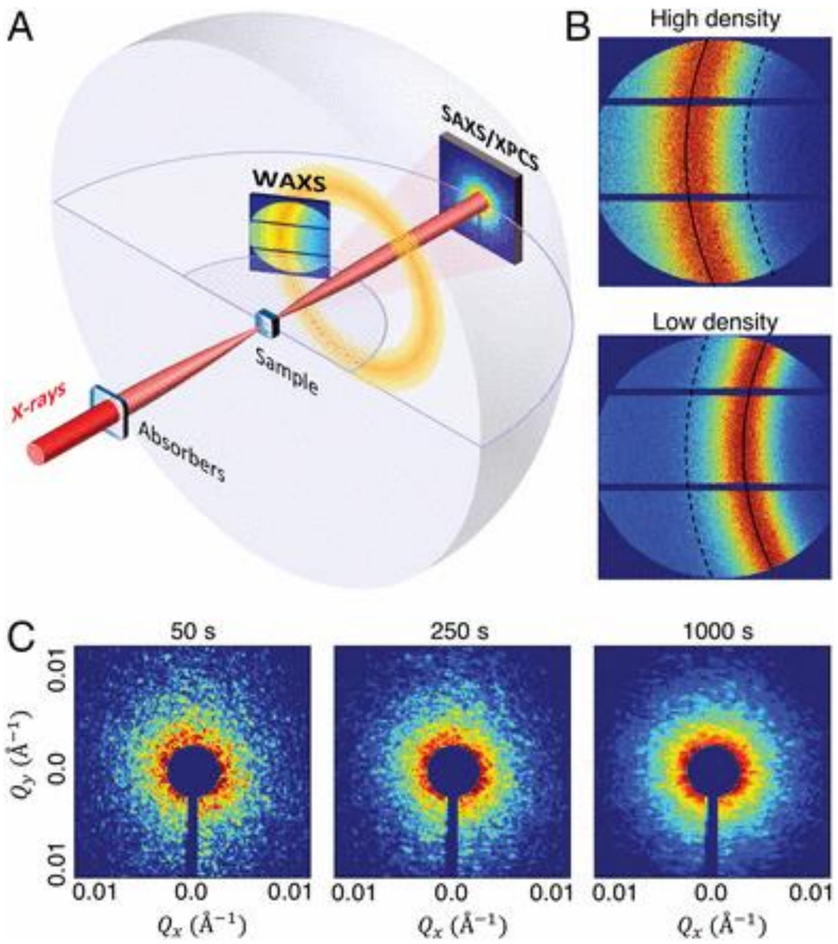
BUT: will the q-range be covered by the detector? Consider to measure full diffraction rings.
Obtain the scattering angle via $q = \frac{4\pi}{\lambda}\sin\left(\frac{\theta}{2}\right) \;\Rightarrow\; \theta = 2 \cdot \mathrm{asin}\left(\frac{q\lambda}{4\pi}\right) \approx 29°$.
Placing the AGIPD at 0.73 m distance, a maximum scattering angle of $\theta_{AGIPD} = \mathrm{atan}\left(\frac{500 \cdot 200\ \mu m}{0.73\ m}\right) \approx 7.8°$ can be achieved. As consequence, the detector would be needed to be placed to a shorter distance: $D_{new} \approx \frac{500 \cdot 200\ \mu m}{\tan(29°)} \approx 0.18$ m → Speckles cannot be resolved! → Need to reduce beam size by a factor of $\frac{0.73}{0.18} \approx 4$

2. ii. What size of an detector would be needed to keep 1:1 speckle to pixel size

We found a sample-detector distance of $D \approx 0.73$ m that fullfils the needs.

Assume again to hit the detector in the centre and that we would resolve at least 29° - or better 40° to have the full ring.

The detector would need to be $p = 0.73 \cdot \tan(40°) \cdot 2 \approx 1.225$ m → very large, corresponds to about 38 Megapixels



PNAS 114, 8793 (2017)

3. i. Determine lattice constant a and c

Hexagonal ice → hexagonal lattice.

Distance of lattice planes for hexagonal lattice: $\frac{1}{d_{hkl}^2} = \frac{4}{3a^2}(h^2 + hk + k^2) + \frac{l^2}{c^2}$

Q-value of Bragg reflections ($q_{hkl} = \frac{2\pi}{d_{hkl}}$) : $q_{hkl} = 2\pi\sqrt{\frac{4}{3a^2}(h^2 + hk + k^2) + \frac{l^2}{c^2}}$

- {100}: $q_{100} = 2\pi\sqrt{\frac{4}{3a^2}(1^2 + 1 \cdot 0 + 0^2) + \frac{0^2}{c^2}} = \frac{2\pi}{a}\sqrt{\frac{4}{3}} = 1.608$ Å^{-1} → $a = 4.512$ Å
- {002}: $q_{002} = 2\pi\sqrt{\frac{4}{3a^2}(0^2 + 1 \cdot 0 + 0^2) + \frac{2^2}{c^2}} = \frac{4\pi}{c} = 1.711$ Å^{-1} → $c = 7.345$ Å
- Check with the others

DESY.

3. ii. Symmetries in an XCCA experiment

Correlations only between equivalent Bragg reflections. Find equivalent planes via (hkil) notation, with $i = -(h+k)$. Then permutation of hki give equivalent planes.

- {100}: (100) → $(10\bar{1}0)$ is equivalent to e.g. $(01\bar{1}0)$ → (010).

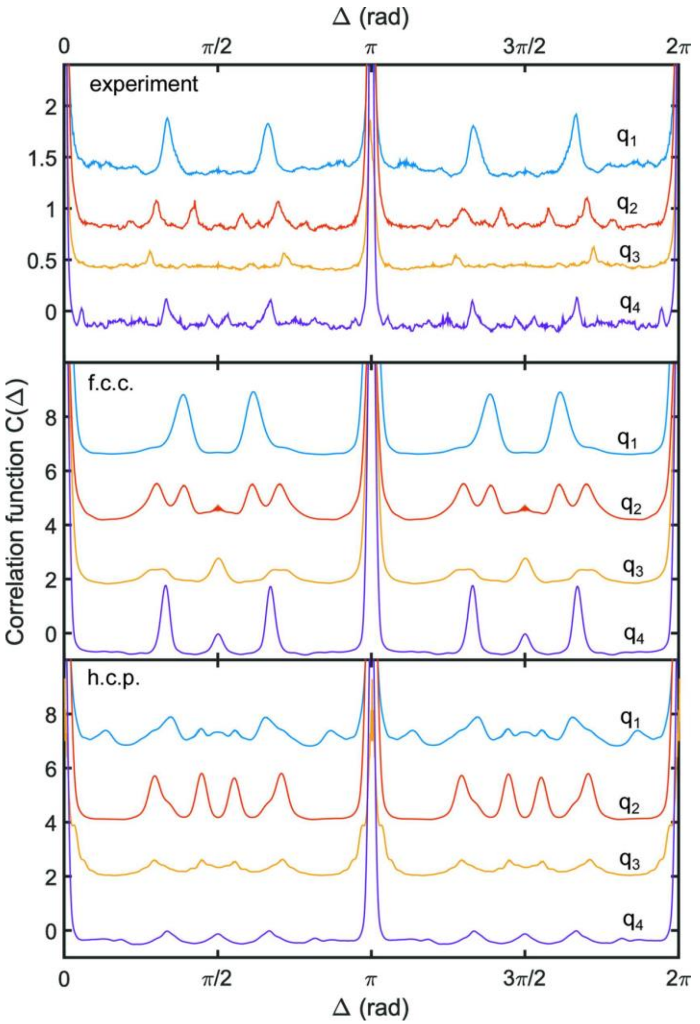
Consider basis vector of hexagonal lattice system: $a_1 = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} a,\ a_2 = \begin{pmatrix} -0.5 \\ \sqrt{3}/2 \\ 0 \end{pmatrix} a,\ a_3 = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} c,$

So we have $(100) = a_1,\ (010) = a_2$.

Correlation angle is then $\cos\omega = \frac{a_1 \cdot a_2}{|a_1||a_2|} = -0.5 \Rightarrow \omega = 120°$

- {002}: Only third direction contributes → $\omega = 180°$
- {101}: similar ideas for {100} → (101) and (011) are equivalent. $(101) = a_1 + a_3 = \begin{pmatrix} a \\ 0 \\ c \end{pmatrix},\ (011) =$

$$a_2 + a_3 = \begin{pmatrix} 0 \\ a \\ c \end{pmatrix} \quad \Rightarrow \quad \cos\omega = \frac{a_1 \cdot a_2}{|a_1||a_2|} = \frac{c^2}{a^2+c^2} \approx 0.726 \quad \Rightarrow \omega = 43.45°$$



	q (nm^{-1})	f.c.c. index	f.c.c. label	h.c.p. index	h.c.p label
q_1	0.027	111	i	002	b
q_2	0.029	111/200	i and ii	011	c
q_3	0.031	200	ii	–	–
q_4	0.044	220	iii	110	e