Coherent Diffractive Imaging and Determining Structural Properties from Cross-correlation Analysis

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HELMHOLTZ | ASSOCIATION

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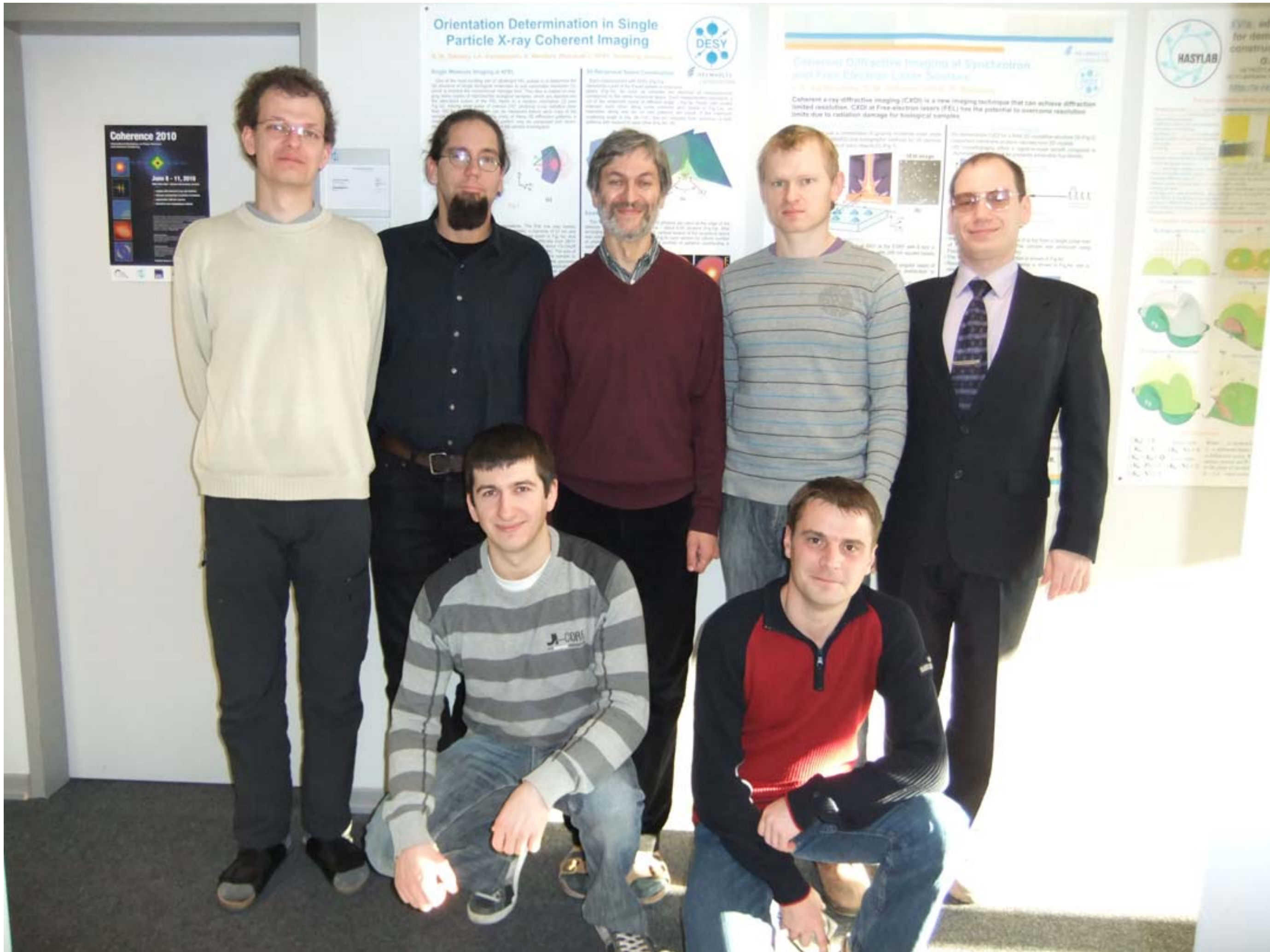
- R. Kurta
- U. Lorenz
- R. Dronyak



Coherence measurements at FEL sources

Measurements at LCLS (June 2010)

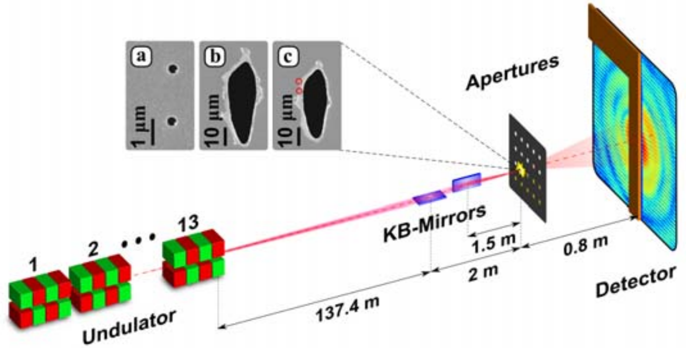
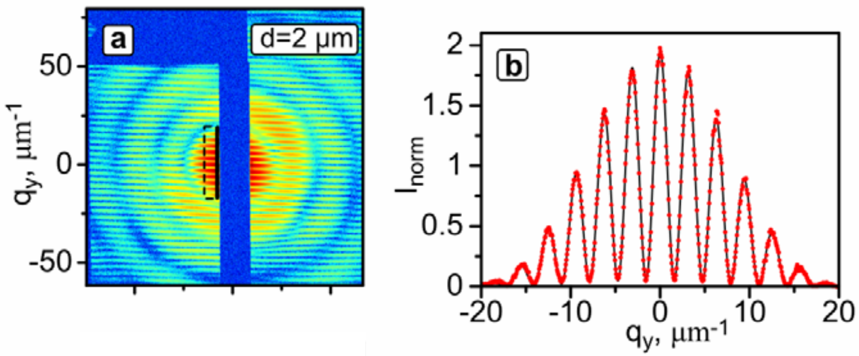
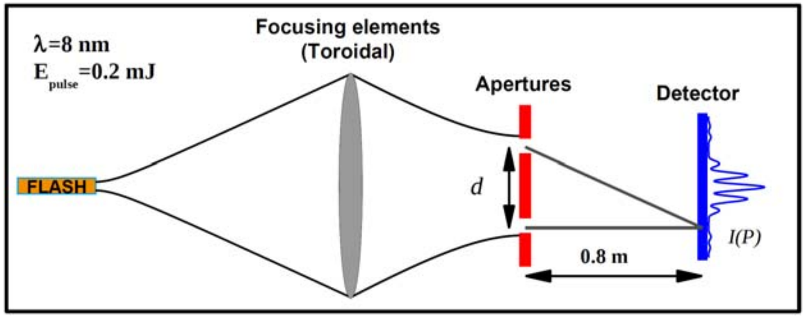
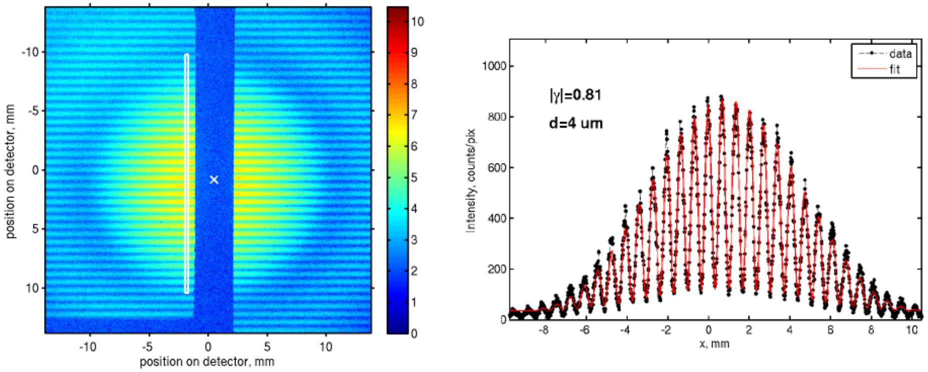


Fig. 1



Measurements at FLASH (October 2010)

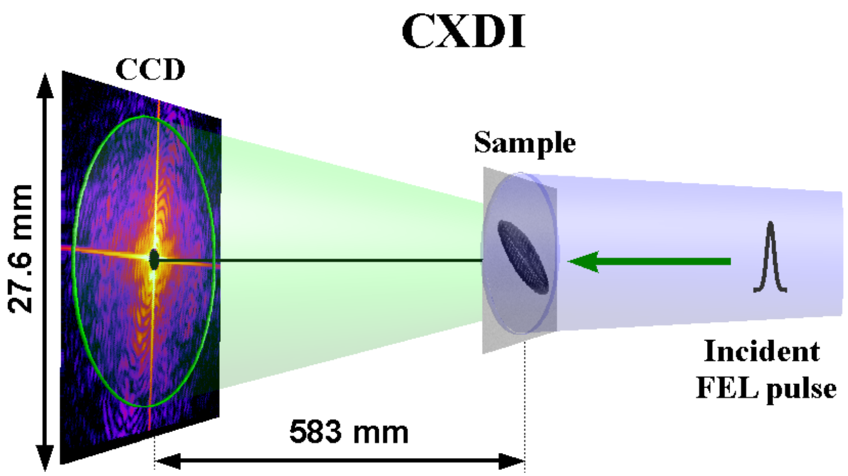


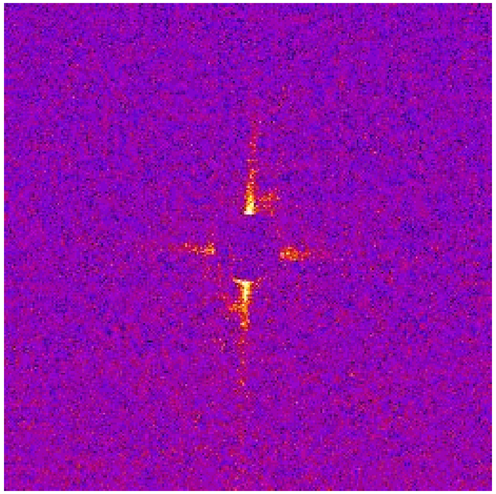


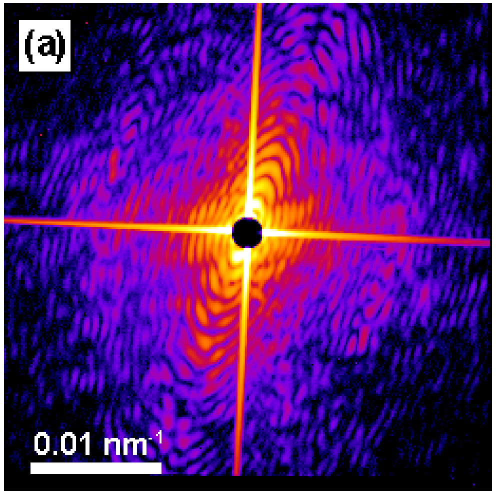
I. Vartanyants *et al.*, (2011)
(Phys. Rev. Lett. submitted)
available on ArXive:
http://arxiv.org/abs/1105.3898

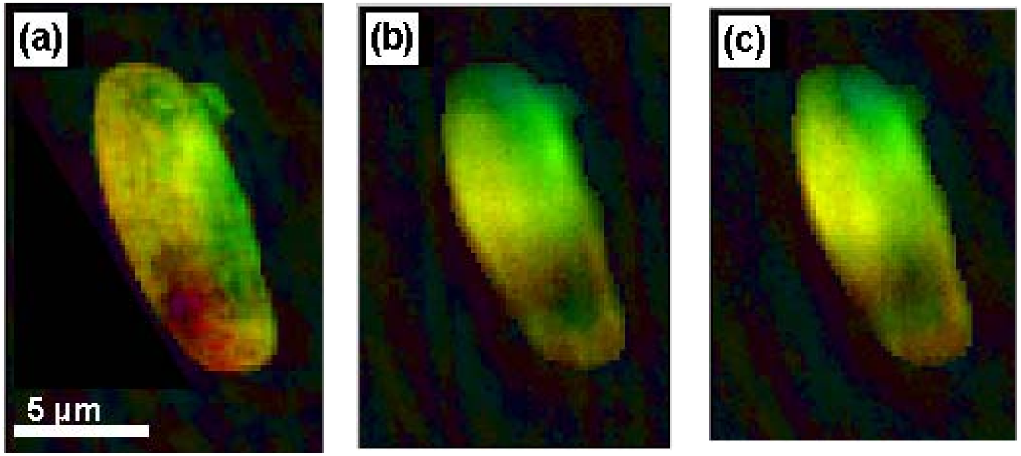
A. Singer, F. Sorgenfrey *et al.*, (2011)
(in preparation)

Femtosecond coherent imaging of biological samples at FLASH







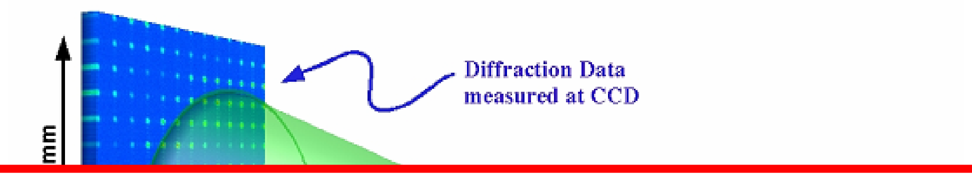


Best of 2010

A. Mancuso *et. al.* New J. Phys. Topical issue: Focus on coherent beams, **12** 035003 (2010)

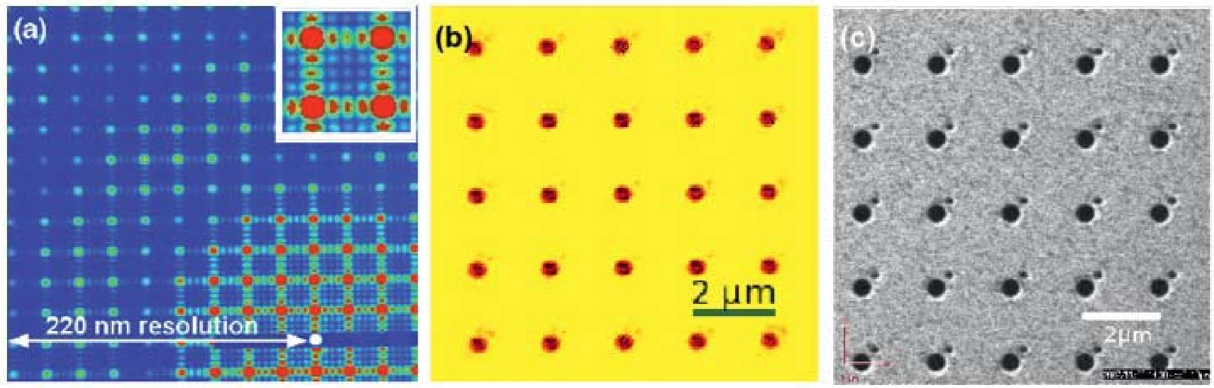
Coherent pulse 2D crystallography at FELs

CXDI experiment at FLASH



For an overview of our FLASH experiments see:

I. Vartanyants *et al.* Special issue: Intense x-ray science: The first 5 years of FLASH, J. Phys. B: At. Mol. Opt. Phys. 43, 194016 (2010)

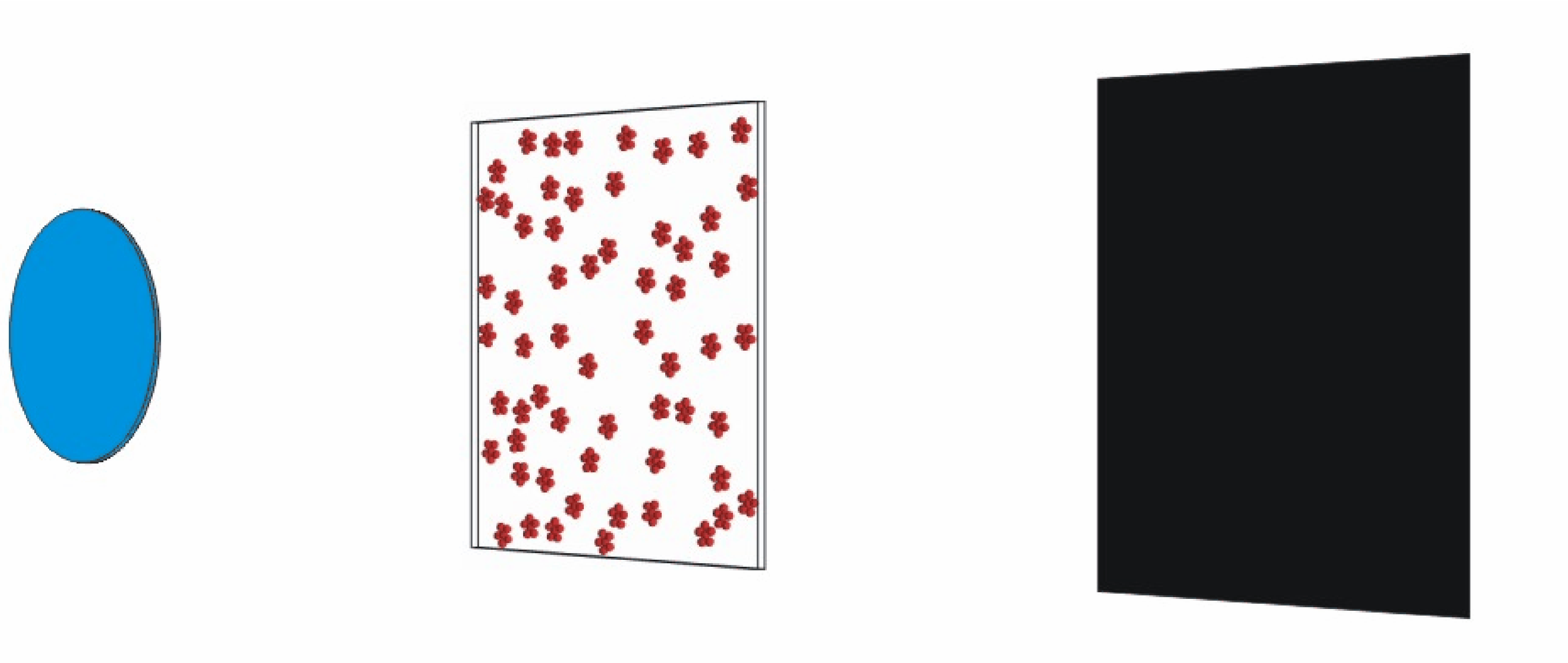


A. Mancuso, *et al.*, PRL **102,** 035502 (2009)

Single particle imaging

Main problem in a single particle imaging:
low scattered signal in a single pulse

Many particles in a coherent beam



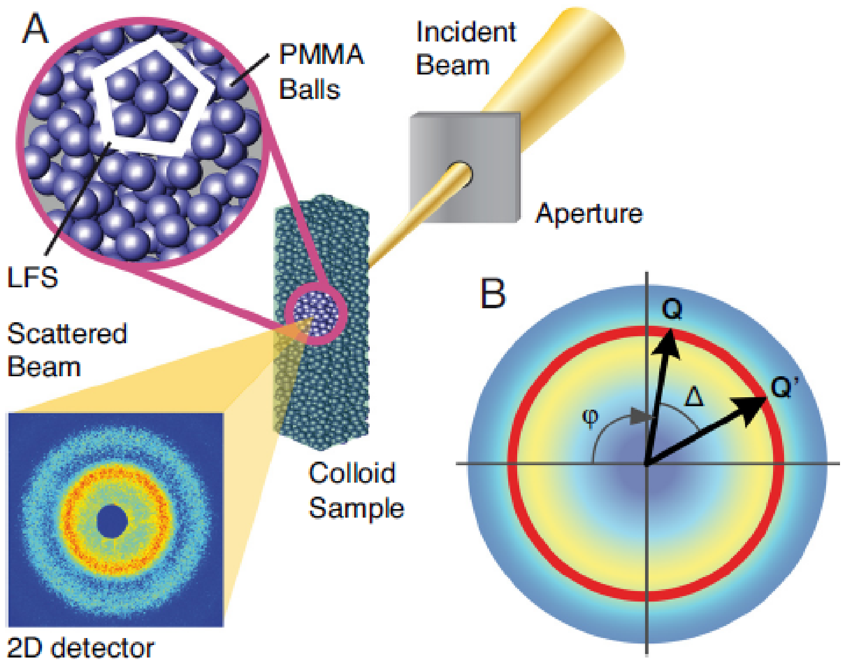
Can we determine the structure of individual particles in such experiment?

Is there any order in disordered systems ?

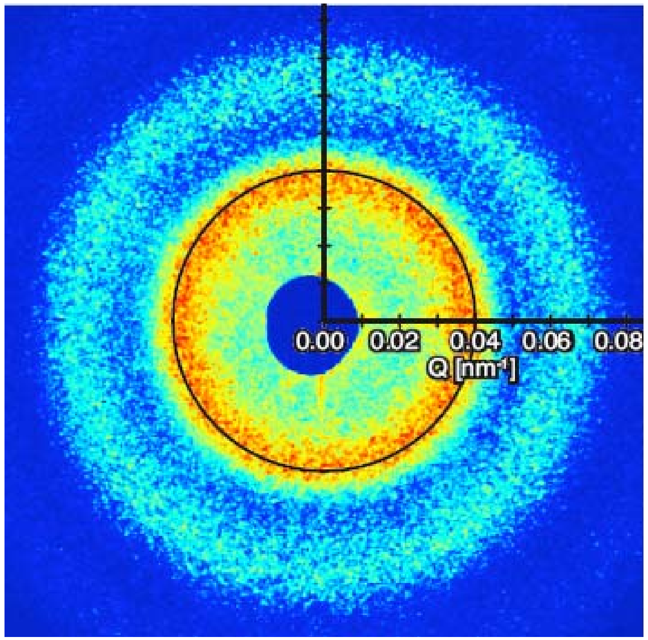
Are there means to observe this order using x-rays ?

Coherent scattering experiment on colloidal glass

Experiment



Diffraction pattern

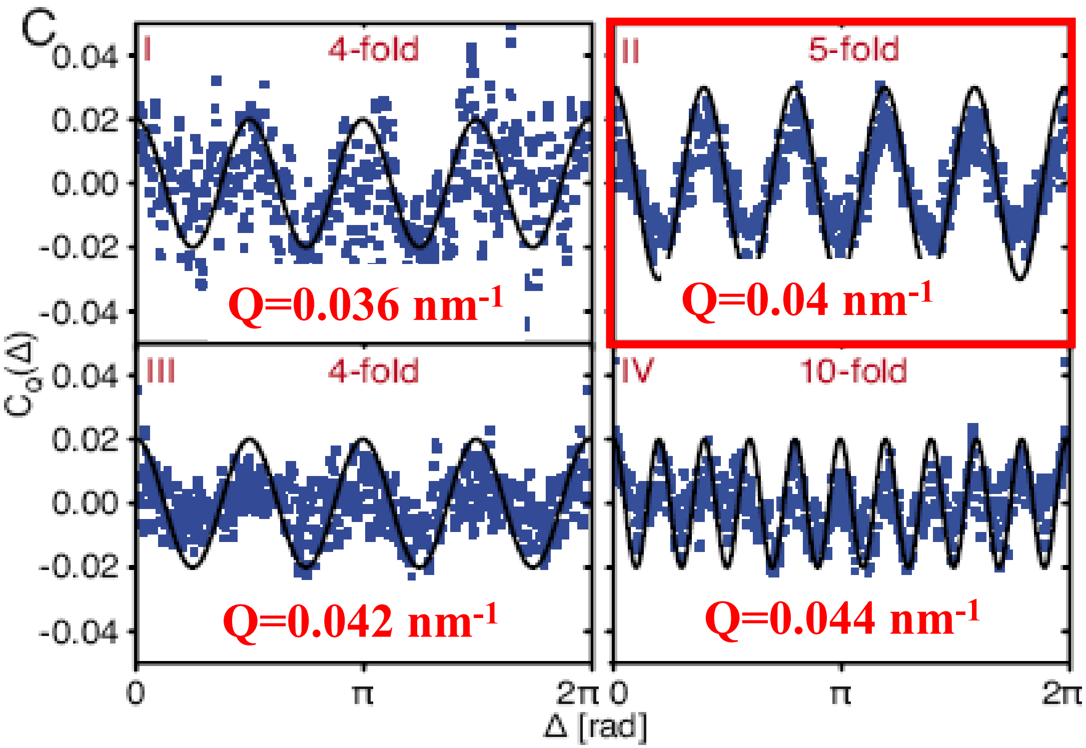


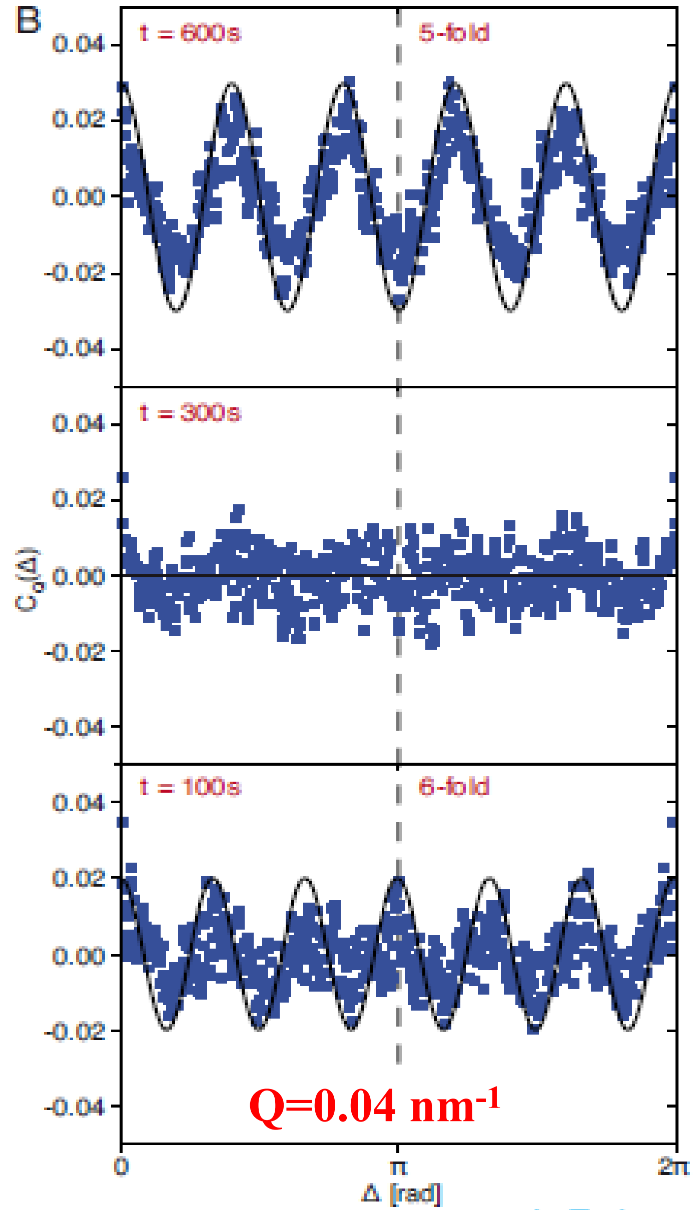
Wochner P *et al*., PNAS **106**, 11511 (2009)

Angular cross-correlation function

$$C_Q(\Delta) = \frac{\langle I(Q,\varphi)I(Q,\varphi+\Delta)\rangle_\varphi - \langle I(Q,\varphi)\rangle_\varphi^2}{\langle I(Q,\varphi)\rangle_\varphi^2}$$

X-Ray Cross Correlation Analysis

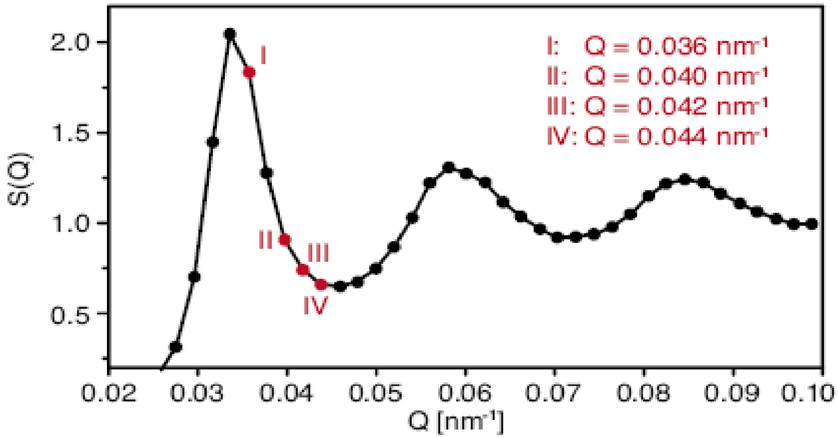




$$C_Q(\Delta) = \frac{\langle I(Q,\varphi)I(Q,\varphi+\Delta)\rangle_\varphi - \langle I(Q,\varphi)\rangle_\varphi^2}{\langle I(Q,\varphi)\rangle_\varphi^2}$$

Different type of analysis

SAXS analysis



Structure factor

$$S(Q)$$

$$g(r) \sim \langle \rho(\mathbf{r})\rho(\mathbf{0}) \rangle$$

$g(r)$ - pair correlation function;

$\rho(\mathbf{r})$ - electron density.

XCCA analysis

$$C_Q(\Delta) = \frac{\langle I(Q,\varphi)I(Q,\varphi+\Delta)\rangle_\varphi - \langle I(Q,\varphi)\rangle_\varphi^2}{\langle I(Q,\varphi)\rangle_\varphi^2}$$

Probe of high-order correlations:

$$\langle I(\mathbf{Q},t)I(\mathbf{Q}',t')\rangle \sim \int\int\int\int \mathrm{e}^{-i\mathbf{Q}(\mathbf{r}-\mathbf{s})-i\mathbf{Q}'(\mathbf{r}'-\mathbf{s}')}$$

$$\times g_4(\mathbf{r}',\mathbf{s}',t,\mathbf{r}',\mathbf{s}',t)d\mathbf{r}d\mathbf{s}d\mathbf{r}'d\mathbf{s}',$$

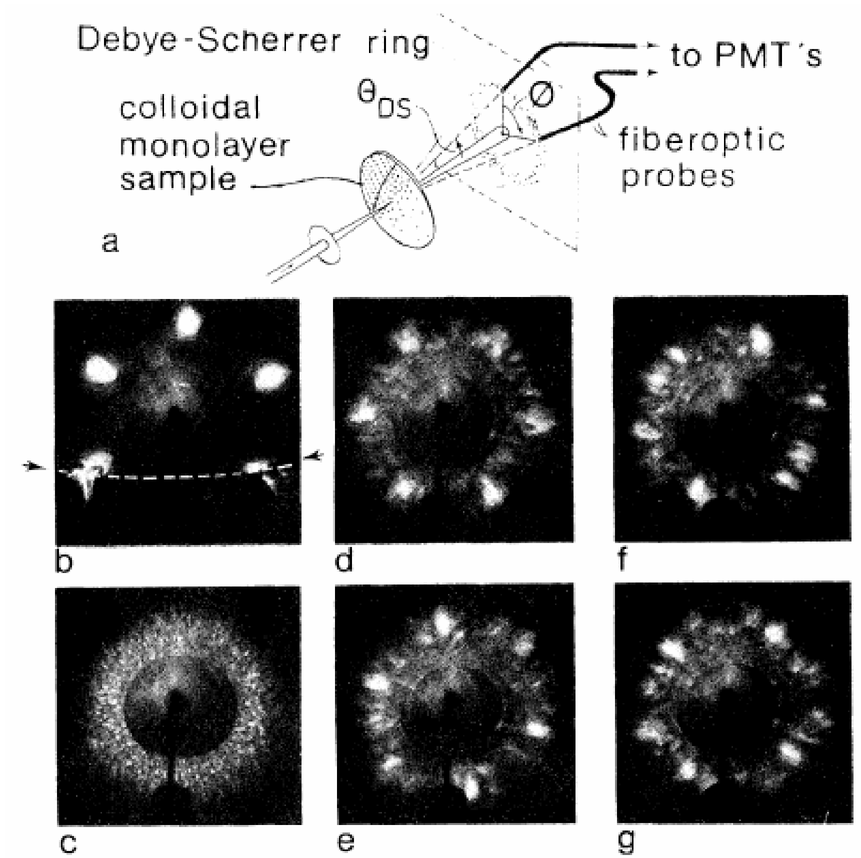
where

$$g_4(\mathbf{r}',\mathbf{s}',t,\mathbf{r}',\mathbf{s}',t) \sim \langle \rho(\mathbf{r},t)\rho(\mathbf{s},t)\rho(\mathbf{r}',t')\rho(\mathbf{s}',t')\rangle$$

4-point correlation function

Earlier use of intensity cross-correlation functions

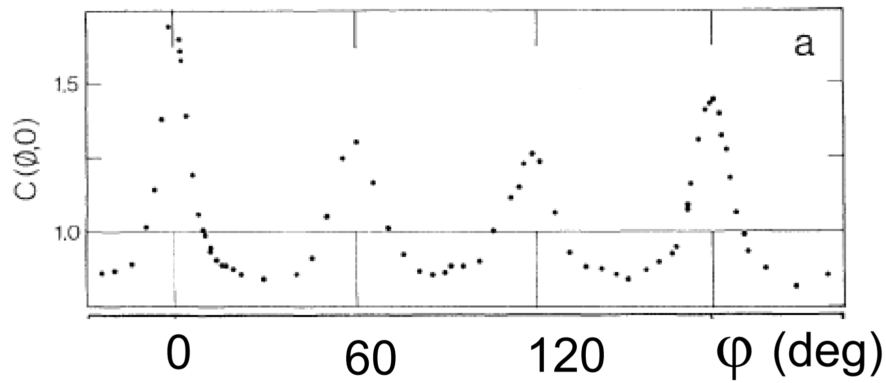
Scattering experiment on a charged polymer spheres in aqueous colloidal suspension



Photographs of typically observed scattered light distributions

Intensity cross-correlation function

$$C(\varphi,\tau=0)=\frac{\langle I(q_1,\varphi=0,t)I(q_2,\varphi,t)\rangle}{\langle I(q_1,t)\rangle\langle I(q_2,t)\rangle}$$



The measured intensity cross-correlation function in 2D liquid

N. Clark *et al.*, PRL **50**, 1459 (1983)

Questions

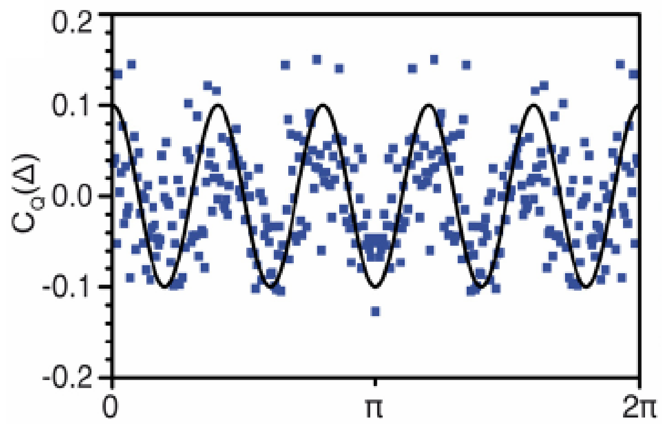
How angular CCFs are related to the structural properties of disordered systems?

Motivation for our work

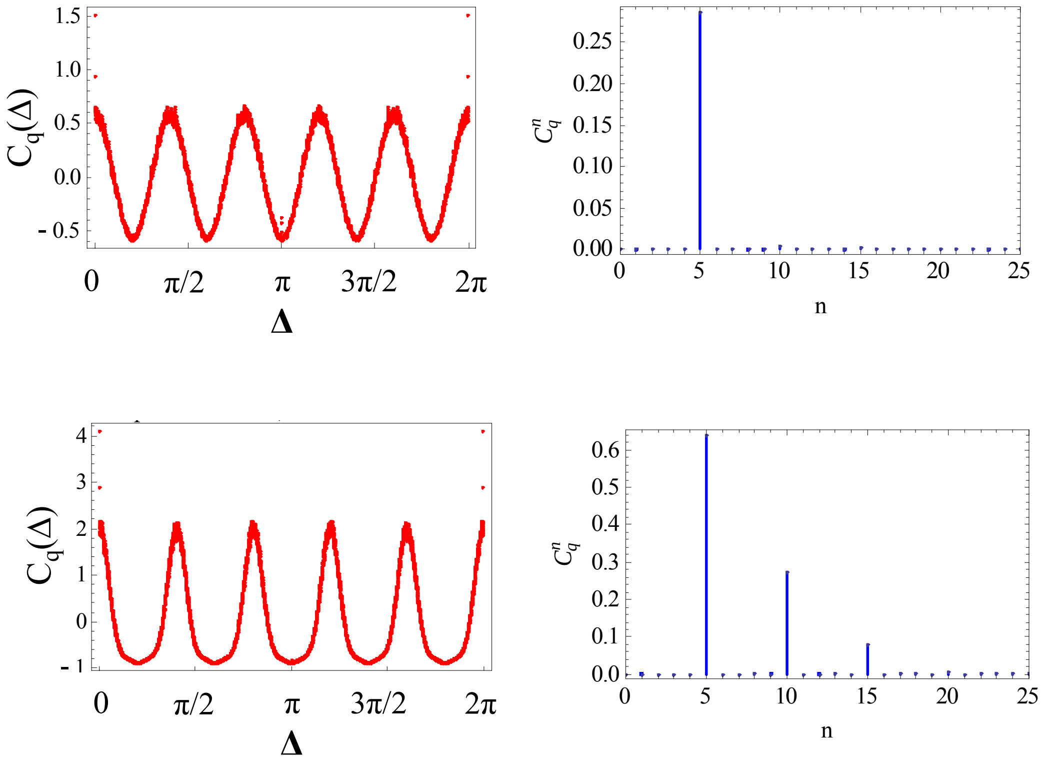
- **Provide a general theoretical background for the x-ray angular cross-correlation analysis**
- **Verify theoretical findings with model calculations**

- M. Altarelli, R. Kurta, and I. Vartanyants, Phys. Rev. B **82**, 104207 (2010)
- R. Kurta, M. Altarelli, E. Weckert, I. Vartanyants (2011) (in preparation)

Fourier series analysis of CCFs



CCFs Frequency spectra



What symmetry do you see in this Figure ? ? ?

Convenient way to study angular cross-correlations is to perform Fourier series analysis

Fourier series analysis of CCFs

Fourier series expansion:

$$C_q(\Delta) = \sum_{n=-\infty}^{\infty} C_q^n e^{in\Delta}$$

$$C_q^n = \frac{1}{2\pi}\int_0^{2\pi} C_q(\Delta) e^{-in\Delta} d\Delta$$

Convolution theorem:

$$C_q(\Delta) = 2\sum_{n=1}^{\infty} C_q^n \cos(n\Delta), \quad C_q^n = \left|\frac{I^n(q)}{I^0(q)}\right|^2$$

Fourier analysis of intensity:

$$I_q^n = \frac{1}{2\pi}\int_0^{2\pi} I(q,\varphi) e^{-in\varphi} d\varphi$$

- This result explains a **single** ~cos(nΔ) behavior of CCF obtained in Wochner *et al.* paper

- Fourier analysis of the CCF **does not** contain additional information with respect to Fourier analysis of the intensity

Questions

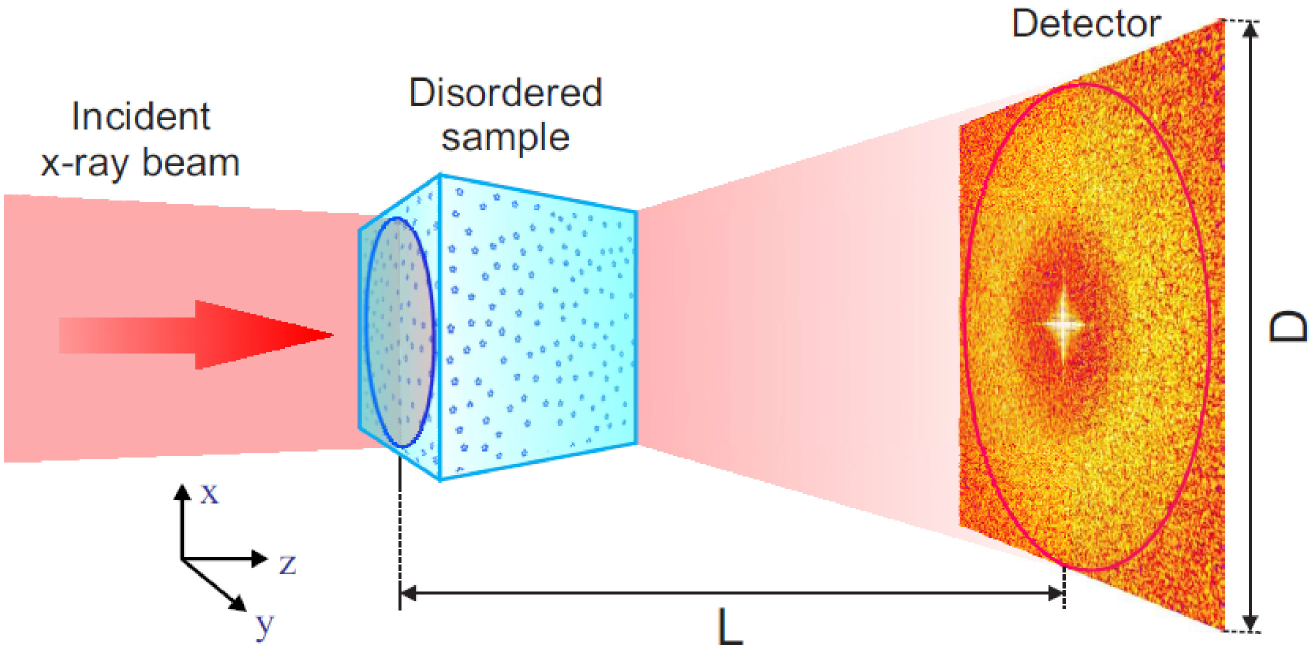
How angular CCFs are related to structural properties of disordered systems?

First answers

- **The problem is reduced to calculation of Fourier coefficients of angular intensity distribution in the conditions of coherent illumination**

- **XCCA gives an access to a 4-point correlation function in the form of a product of two 2-point correlation functions**

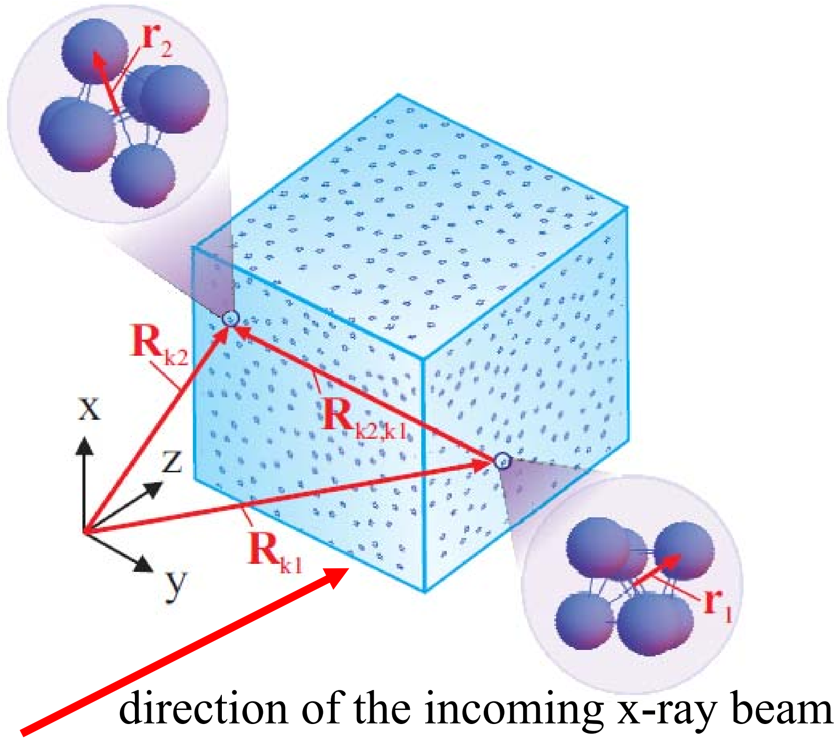
Scattering geometry



Assumptions:

- Kinematical scattering
- Coherent illumination of the sample
- Far-field scattering conditions
- Finite size sample

Sample: 3D disordered system with n-fold local symmetry



3D sample that consists of clusters of a certain symmetry that are spatially and orientationally disordered

Kinematical scattering

$$A(\mathbf{q}) = \int \rho(\mathbf{r}) e^{i\mathbf{q}\mathbf{r}} d\mathbf{r}$$

Electron density of the sample

$$\rho(\mathbf{r}) = \sum_{k=1}^{N} \rho_k(\mathbf{r} - \mathbf{R}_k)$$

The scattered amplitude

$$A(\mathbf{q}) = \sum_{k=1}^{N} e^{i\mathbf{q}\cdot\mathbf{R}_k} A_k(\mathbf{q})$$

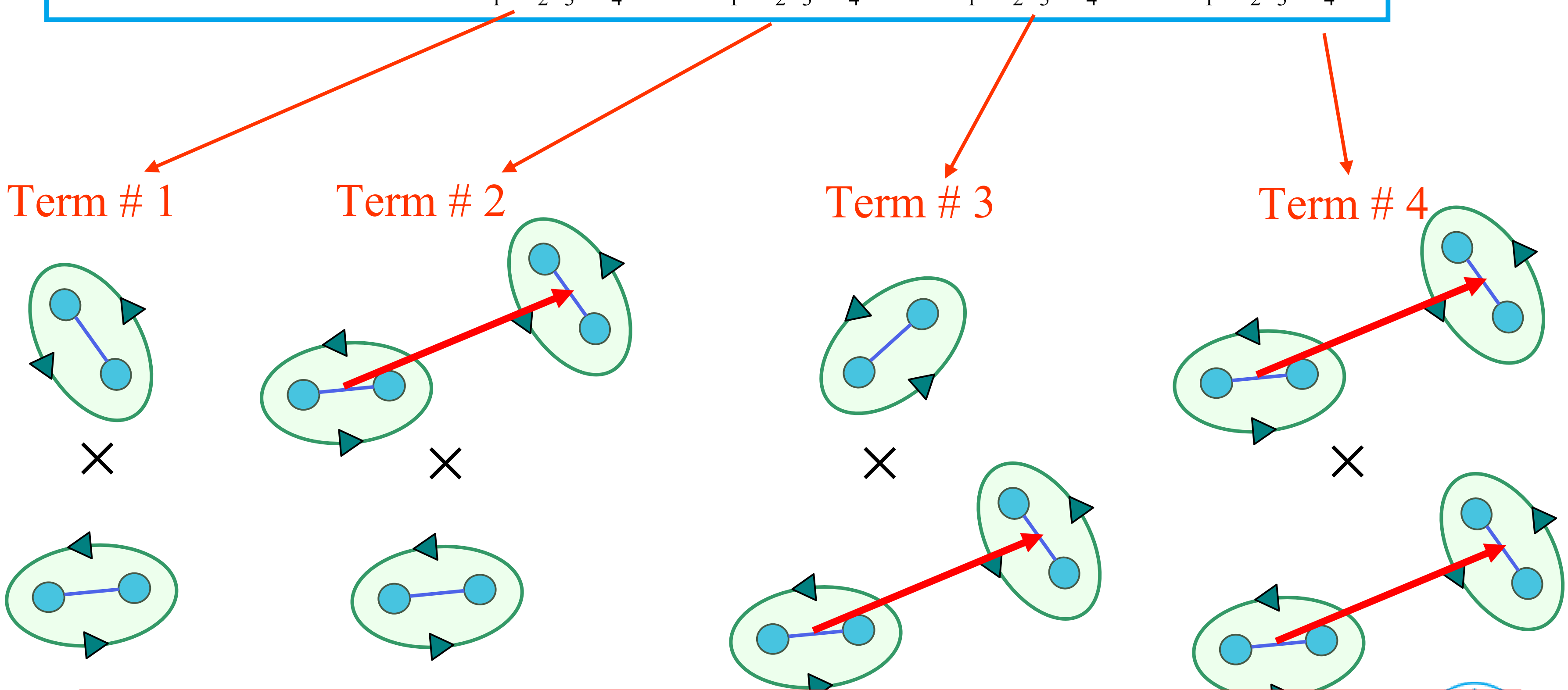
where $A_k(\mathbf{q})$ is the amplitude scattered by one LS

$$A_k(\mathbf{q}) = \int \rho_k(\mathbf{r}) e^{i\mathbf{q}\mathbf{r}} d\mathbf{r}$$

Contribution of different terms to Fourier coefficients C^n_q

$$I^n(q) \propto \sum_{k_1=k_2=k}^{N} L^n_k(q) + \sum_{k_1 \neq k_2}^{N} L^n_{k_1,k_2}(q),$$

$$C^n_q \propto I^{n*}(q) I^n(q) \propto \sum_{k_1=k_2}^{N}\sum_{k_3=k_4}^{N} \ldots + \sum_{k_1=k_2}^{N}\sum_{k_3 \neq k_4}^{N} \ldots + \sum_{k_1 \neq k_2}^{N}\sum_{k_3=k_4}^{N} \ldots + \sum_{k_1 \neq k_2}^{N}\sum_{k_3 \neq k_4}^{N} \ldots$$



Term 1: correlations within the same cluster → local structure

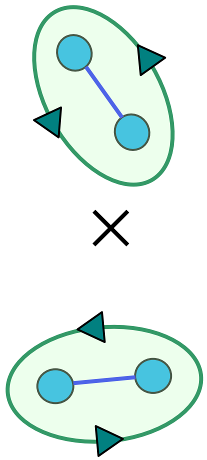
Term 2 –Term 4: correlations between different clusters → medium range order.

2D dilute systems

Dilute systems: R >> d

$$C^{n}_{q_1,q_2} \propto \sum_{k_1=k_2}^{N}\sum_{k_3=k_4}^{N}\ldots + \sum_{k_1=k_2}^{N}\sum_{k_3\neq k_4}^{N}\ldots + \sum_{k_1\neq k_2}^{N}\sum_{k_3=k_4}^{N}\ldots + \sum_{k_1\neq k_2}^{N}\sum_{k_3\neq k_4}^{N}\ldots$$

Term # 1



2D dilute systems

$$C_q^n \propto \Lambda_q^n \cdot \left| \left\langle e^{in\phi} \right\rangle_\phi \right|^2$$

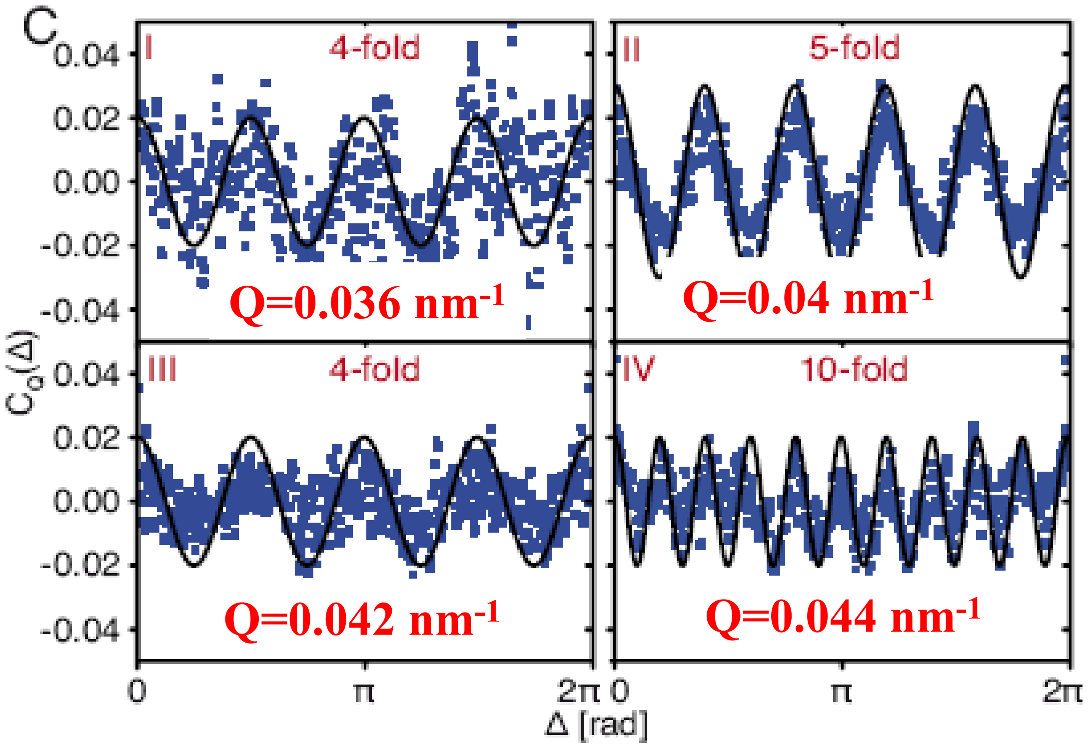
1. Structure term: Λ_q^n

For a cluster with a certain symmetry has non-zero values only for selected values of *n* (selection rules)

2. Statistical term: $A^2 = \left\langle e^{in\phi} \right\rangle_\phi = \frac{1}{N} \sum_{k=1}^{N} e^{in\phi_k}$

For a statistically disordered system with the angular distribution $p(\phi)$ depends on a concrete realization of the system (random phasor sum)

X-Ray Cross Correlation Analysis

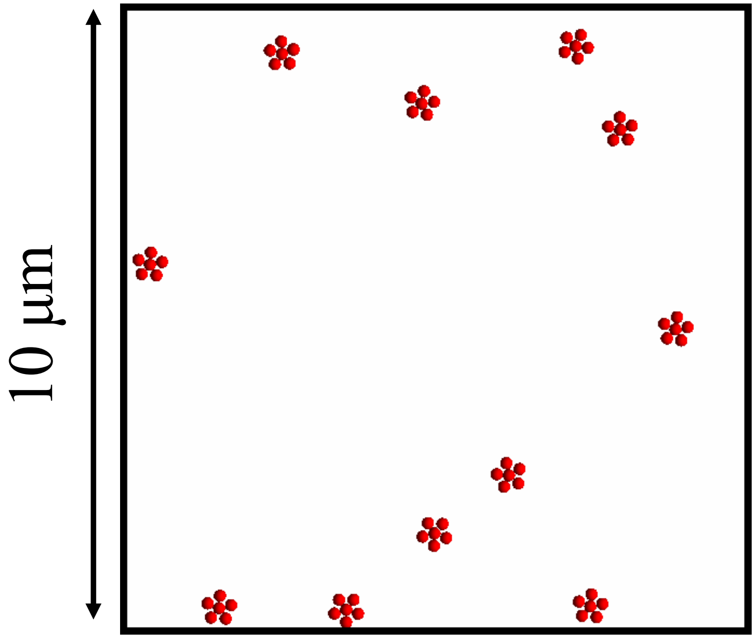


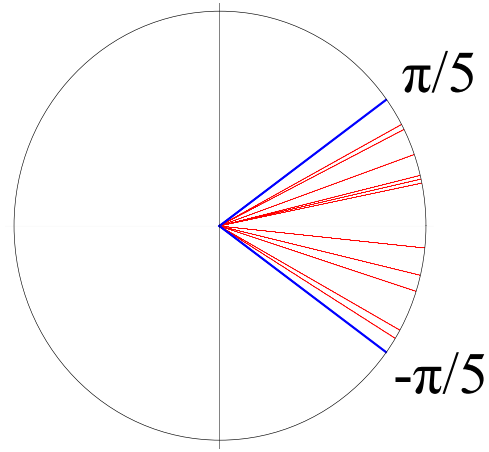
This could be a possible explanation of fast changes of the Fourier components of CCFs with small changes of ***Q***

Statistical term (small number of clusters)

Statistical term:

$$A^2 = \left| \left\langle e^{in\phi} \right\rangle_\phi \right|^2 = \frac{1}{N} \left| \sum_{k=1}^{N} e^{in\phi_k} \right|^2$$



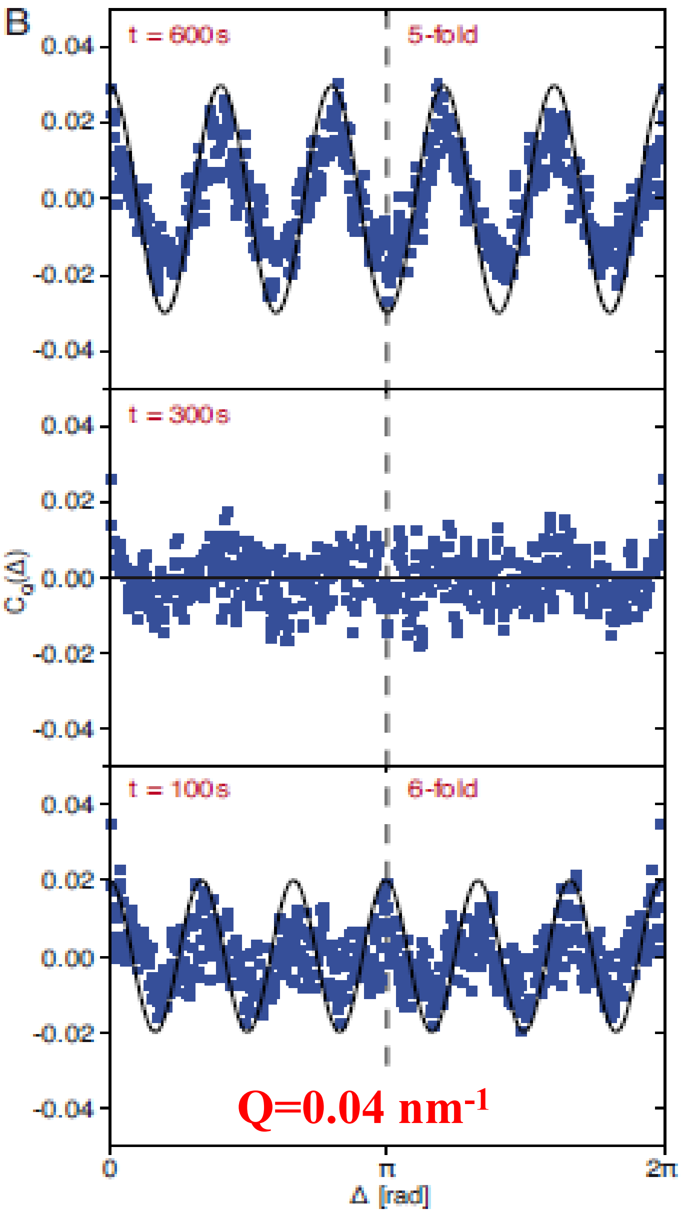


Mean value: $\langle A^2 \rangle = 1/N$

Variance: $\sigma^2_{A^2} = 1/N^2$

- Sample consists of **11 pentagonal clusters**
- Clusters are spatially disordered and have different orientations (uniform distribution, **11 orientations**)

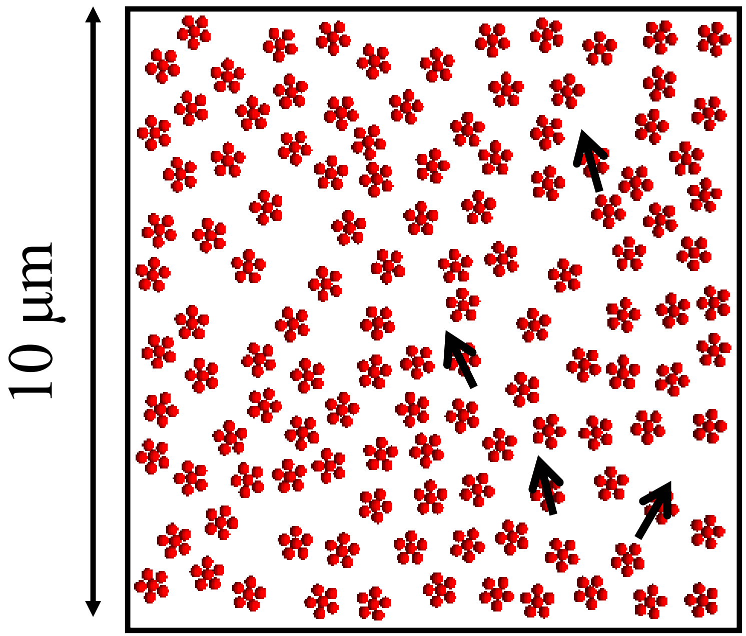
X-Ray Cross Correlation Analysis

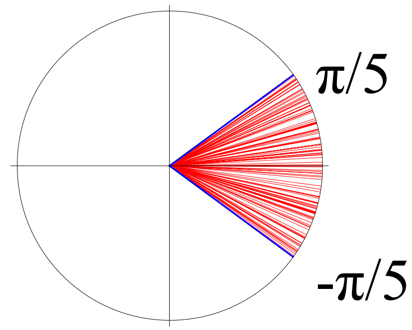


This could be a possible explanation of the dynamics observed in this experiment

Statistical term (big number of clusters)

$$A^2 = \left| \left\langle e^{in\phi} \right\rangle_\phi \right|^2 = \frac{1}{N} \left| \sum_{k=1}^{N} e^{in\phi_k} \right|^2$$





Mean value: $\langle A^2 \rangle = 1/N$

Variance: $\sigma^2_{A^2} = 1/N^2$

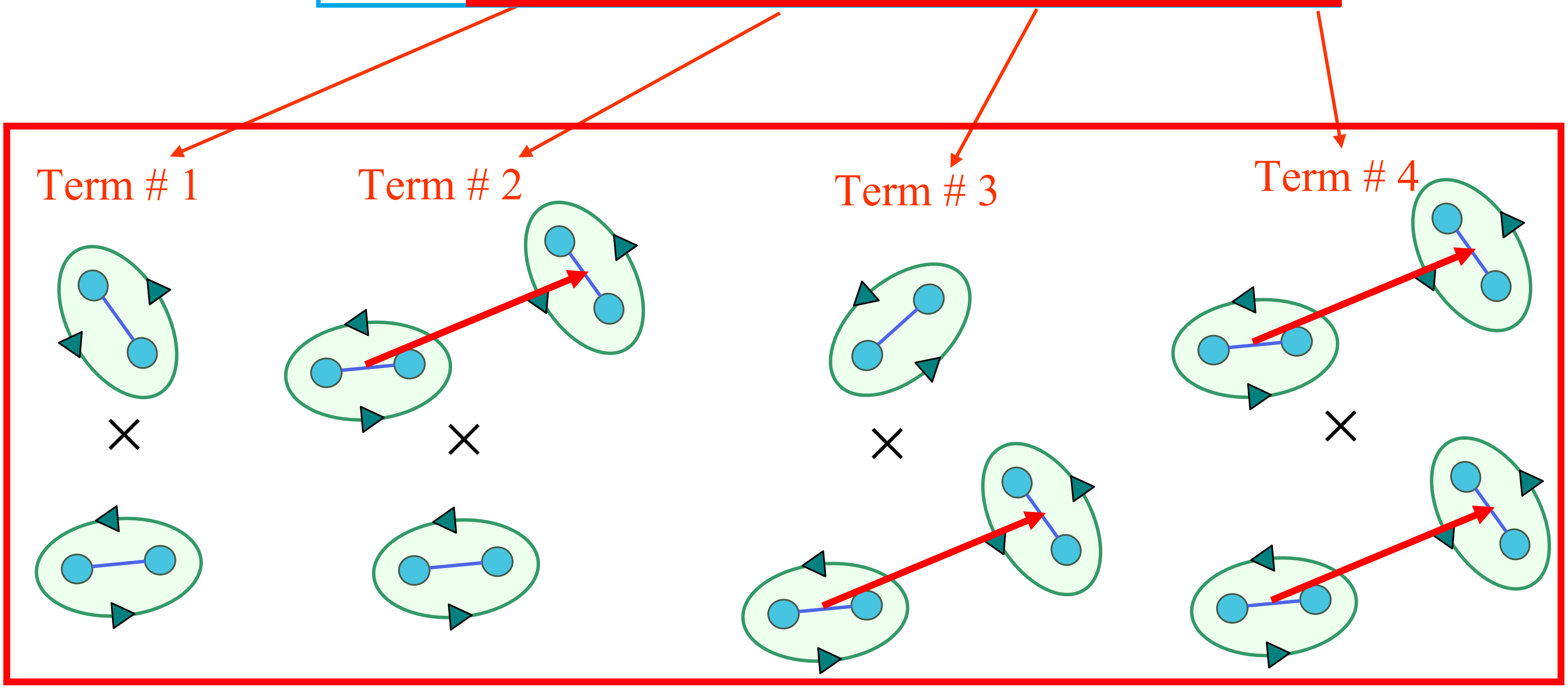
- Sample consists of **121** pentagonal **clusters**
- Clusters are spatially disordered and have a **uniform** distribution of angles

It means that the values of Fourier coefficients of CCF will fluctuate around the mean value ~1/N

Closed packed systems

Close packed systems: R ~ d

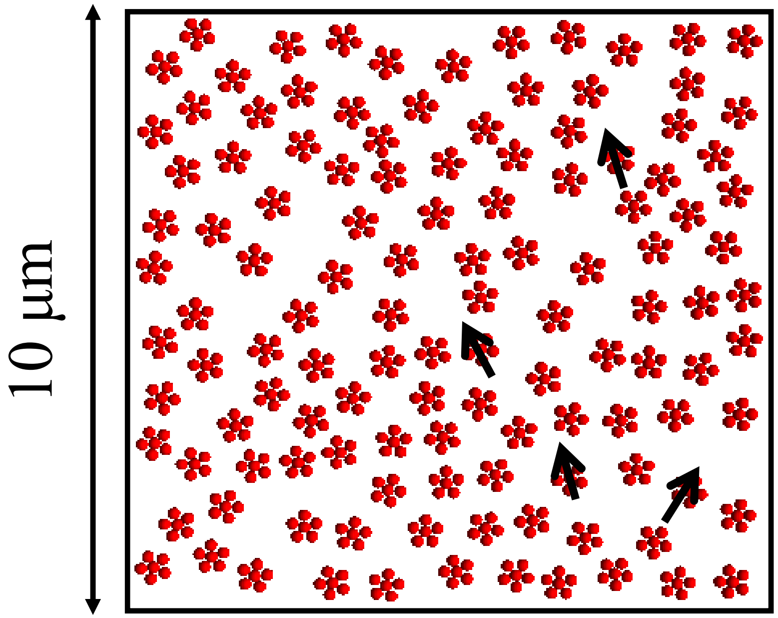
$$C^n_{q_1,q_2} \propto \sum_{k_1=k_2}^{N} \sum_{k_3=k_4}^{N} \ldots + \sum_{k_1=k_2}^{N} \sum_{k_3 \neq k_4}^{N} \ldots + \sum_{k_1 \neq k_2}^{N} \sum_{k_3=k_4}^{N} \ldots + \sum_{k_1 \neq k_2}^{N} \sum_{k_3 \neq k_4}^{N} \ldots$$

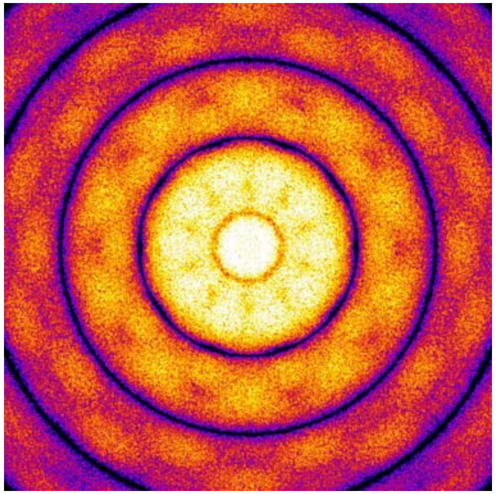


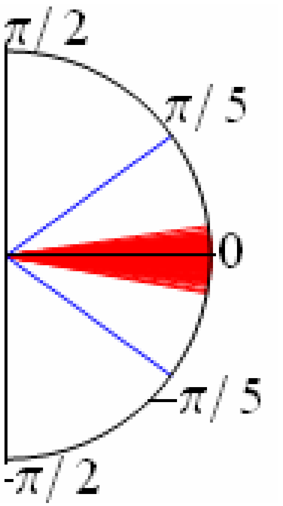
For closed packed systems **interparticle correlations** could not be neglected

Oriented systems (big number of clusters)

Disordered 2D sample





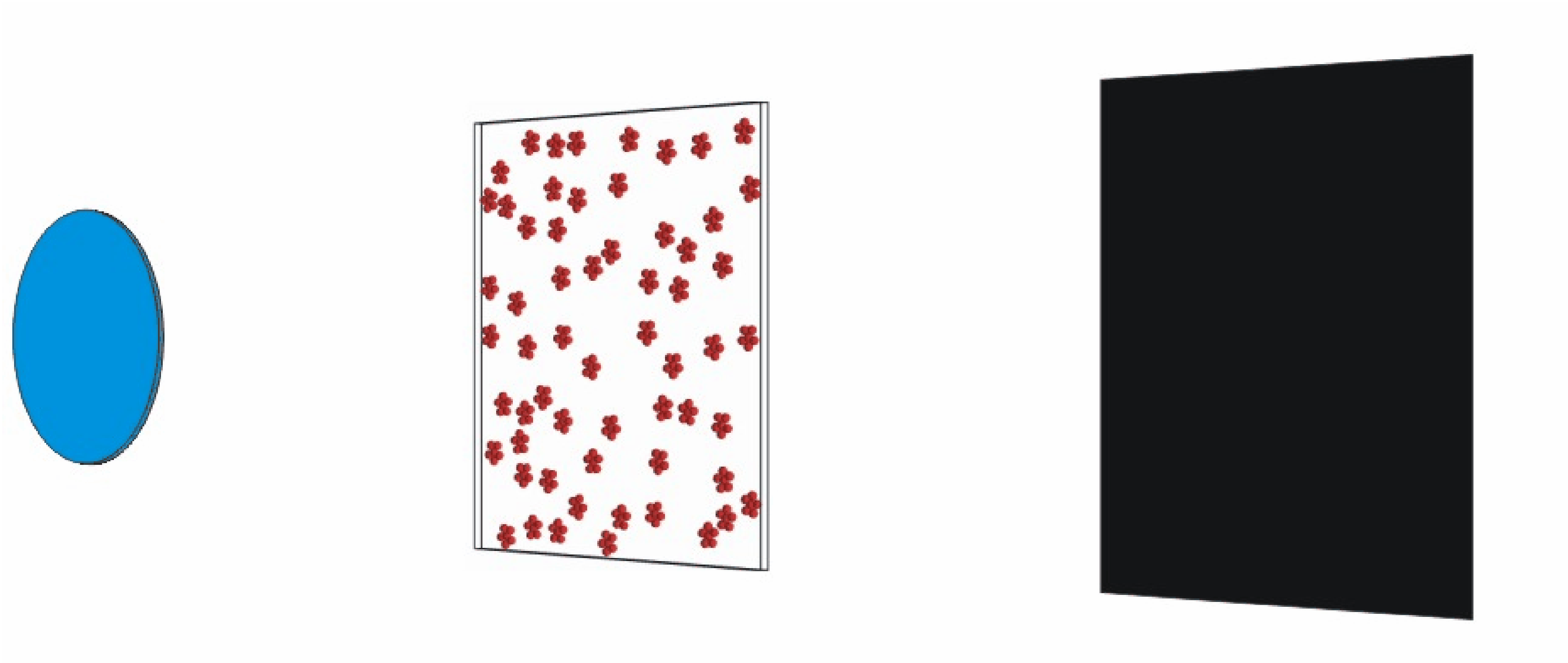


- Sample consists of **121** pentagonal **clusters**
- Clusters are spatially disordered and have a **narrow Gaussian** distribution of angles

Gaussian distribution of cluster orientations, $\sigma_\varphi=0.02\pi$

Mean value: $\langle A^2 \rangle \sim \exp(-n^2\sigma_\varphi^2)$

Many particles in a coherent beam



Analysis of averaged CCFs

Z. Kam, Macromolecules **10**, 927 (1977)
D. K. Saldin et al., Phys. Rev. B **81** (2010)

Analysis of averaged CCFs

CCF averaged over a sufficiently large number M of diffraction patterns

$$\left\langle C_q(\Delta)\right\rangle = \frac{1}{M}\sum_{i=1}^{M} C_q^i(\Delta)$$

Fourier analysis

$$\left\langle C_q^n\right\rangle = \frac{1}{M}\sum^{M} C_q^{ni}$$

For di

neglect

$$\cdot\left\langle A_n^2\right\rangle$$

For dilute systems this approach gives direct access to Fourier components of individual clusters

For a uniform distribution of orientations

$$\left\langle A_n^2\right\rangle \Rightarrow 1/N$$

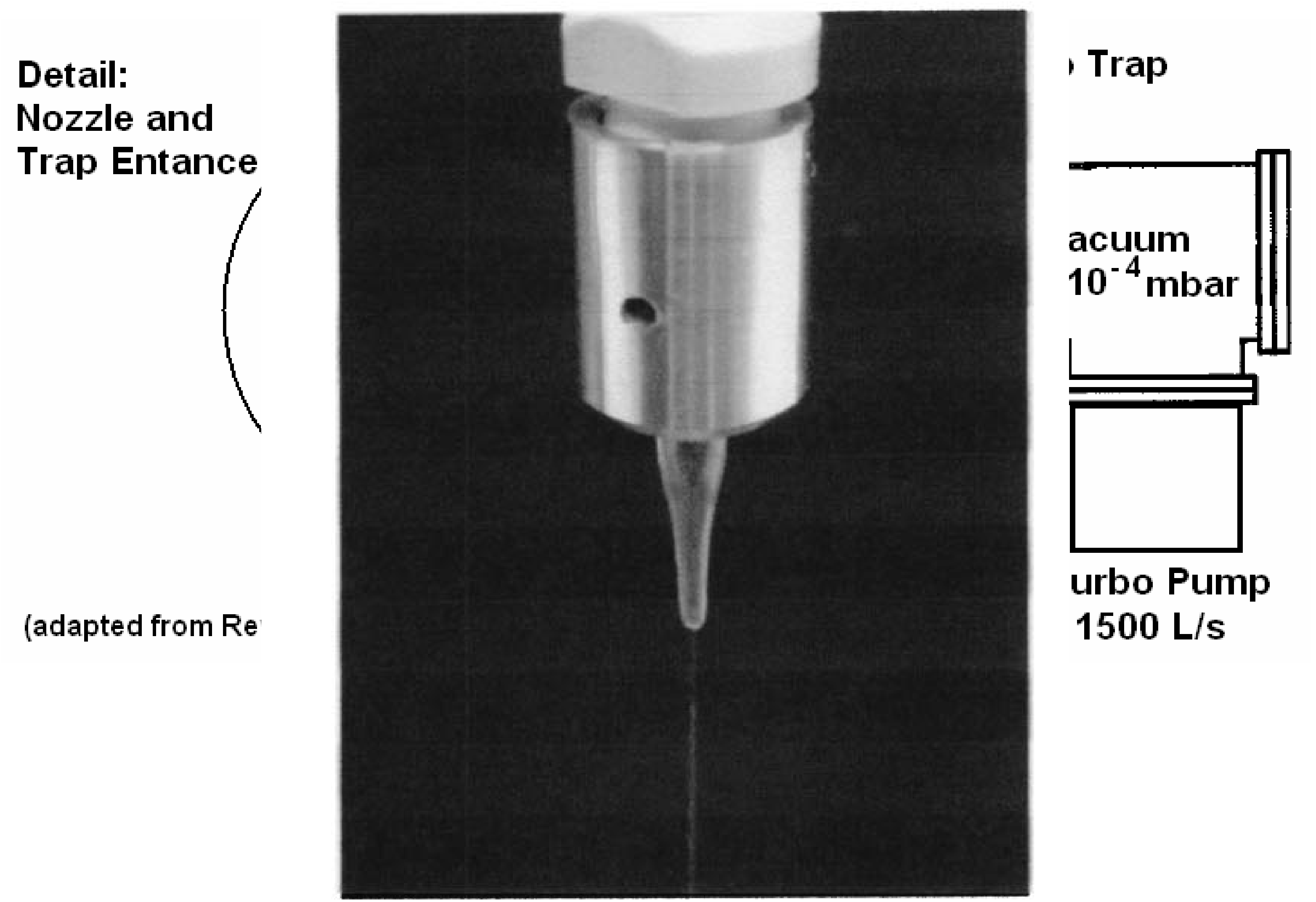
For a Gaussian distribution of orientations

$$\left\langle A_n^2\right\rangle \Rightarrow e^{-n^2\sigma_\varphi^2}\left(1-1/N\right)+1/N$$

Questions

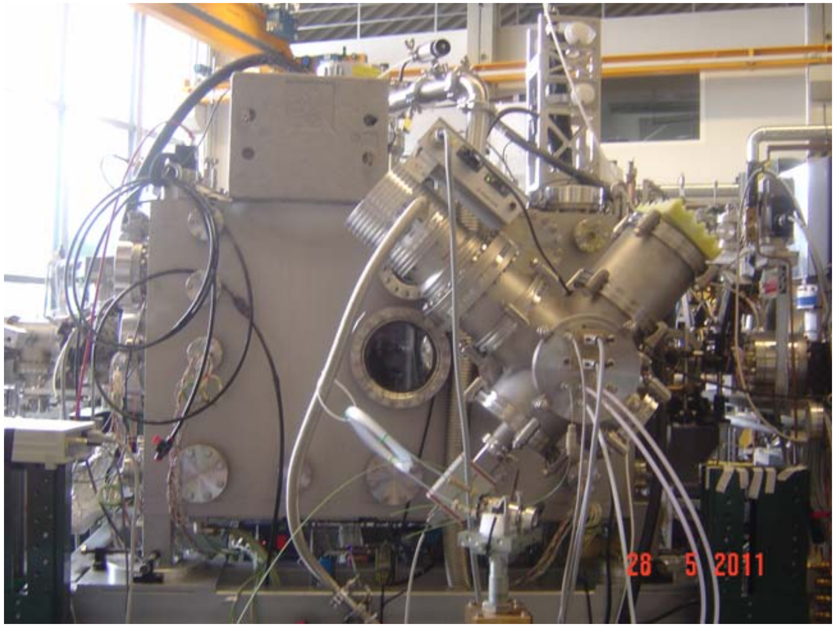
How ERL sources can be used for these type of experiments?

FLASH experiment on a liquid jet

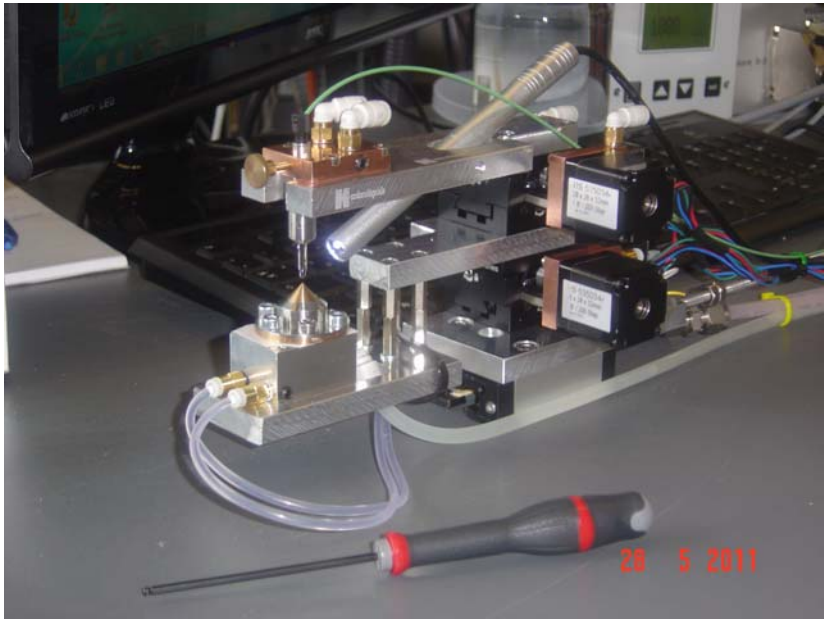


M. Faubel
MPI für Strömungsforschung,Göttingen, Germany

FLASH experiment on a liquid jet



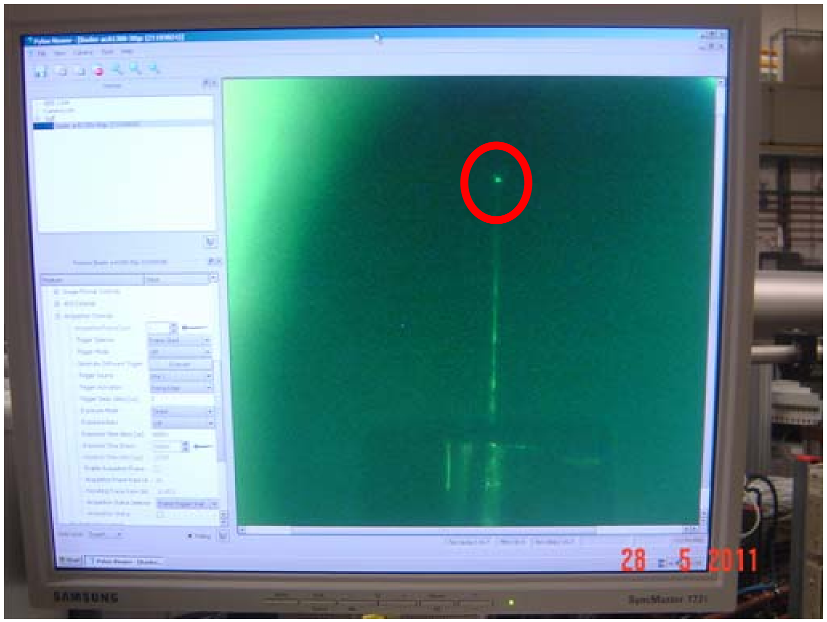
HORST chamber



Liquid jet

Collaboration with: T. Saditt (Göttingen)
A. Rosenhahn (Heidelberg)
A. Mancuso (European XFEL)

FLASH experiment on a liquid jet



FLASH beam
superimposed with the jet

FLASH Parameters

- Wavelength: 8 nm
- 3rd harmonic: 2.66 nm
- Pulse duration: 100 fs
- Pulse energy: 99.5 μJ
- FWHM Spectrum: 0.1 nm

- Water jet: 25 μm nozzle

Conclusions

- A convenient way to study angular CCFs is to analyze their **Fourier coefficients**
- In a general case CCFs deliver a complex information on the **internal symmetry** of clusters and their **spatial correlations** (medium-range order)
- In **dilute systems** the main contribution to CCFs is determined by a local structure **symmetry** “selection rules” and by the **statistical distribution** of different orientations
- In **close-packed systems** correlations **between clusters** become important

Thank you for your attention