Coherent X-ray Scattering and X-ray Photon Correlation Spectroscopy

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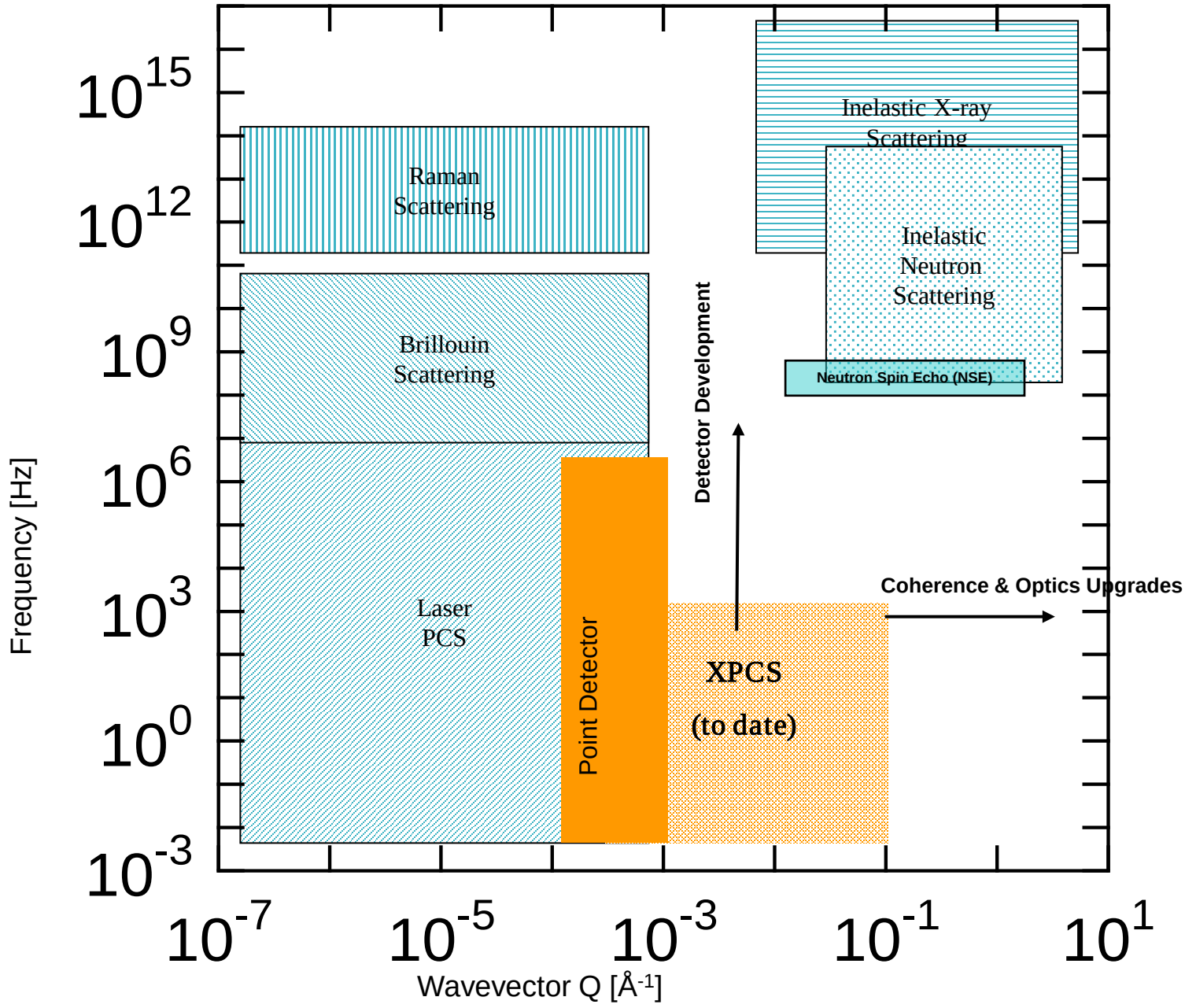
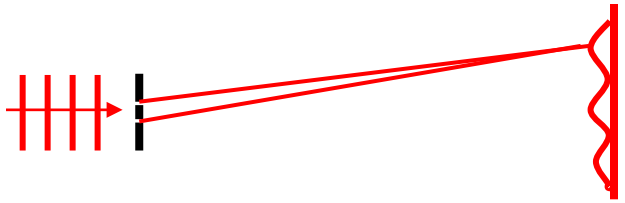
http://www.niu.edu/~llurio/coherence/


Frequency [Hz]
10^{15}
10^{12}
10^{9}
10^{6}
10^{3}
10^{0}
10^{-3}
10^{-7}
10^{-5}
10^{-3}
10^{-1}
10^{1}
Wavevector Q [Å^{-1}]
Raman Scattering
Brillouin Scattering
Laser PCS
Point Detector
XPCS
(to date)
Inelastic X-ray Scattering
Inelastic Neutron Scattering
Neutron Spin Echo (NSE)
Detector Development
Coherence & Optics Upgrades

What is Coherence?

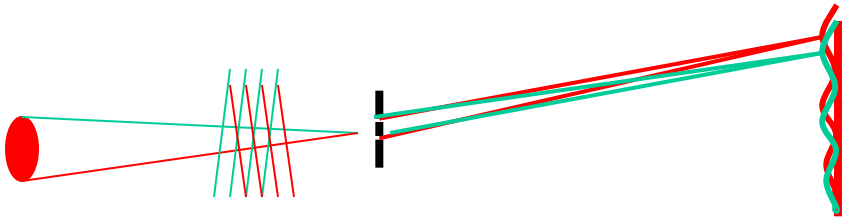
Ideal Young's double slit experiment



Intensity varies as

$$I = 2I_0\left[1+\cos\left(2\pi d\sin(\theta)/\lambda\right)\right]$$

Real Young's double slit experiment



Intensity varies as

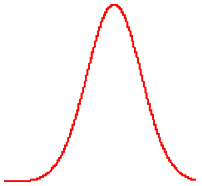
$$I = 2I_0\left[1+\beta\cos\left(2\pi d\sin(\theta)/\lambda\right)\right]$$

β is the contrast, determined by the angular size of the source

Coherence Length and Contrast

It is generally convenient to assume the source has a Gaussian intensity profile

$$I(x) = \frac{I_0}{\sqrt{2\pi}\xi} \exp\left[-\left(x - x_0 \right)^2 / 2\sigma^2 \right]$$



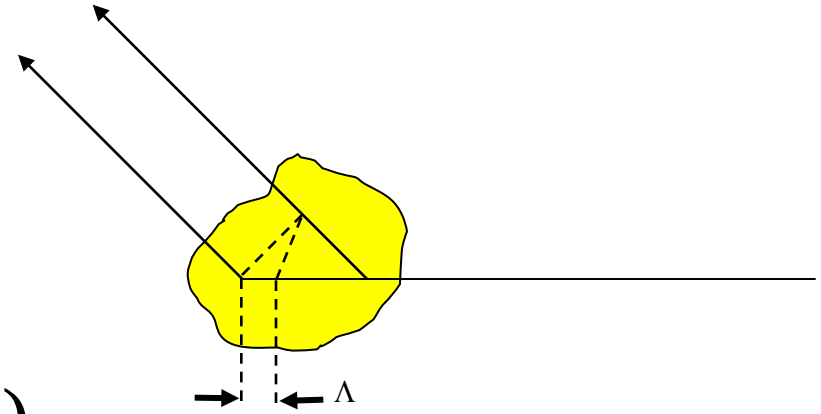
One can then define a coherence length

$$\xi = \frac{\lambda R}{2\sigma\sqrt{\pi}}$$

This characterizes the distance over which two slits would produce an interference pattern, or more generally the length scale over which any sample will produce interference effects.

A more rigorous theory can be found in e.g. Born and Wolf

Longitudinal coherence



$$\Lambda \approx \lambda(E / \Delta E)$$

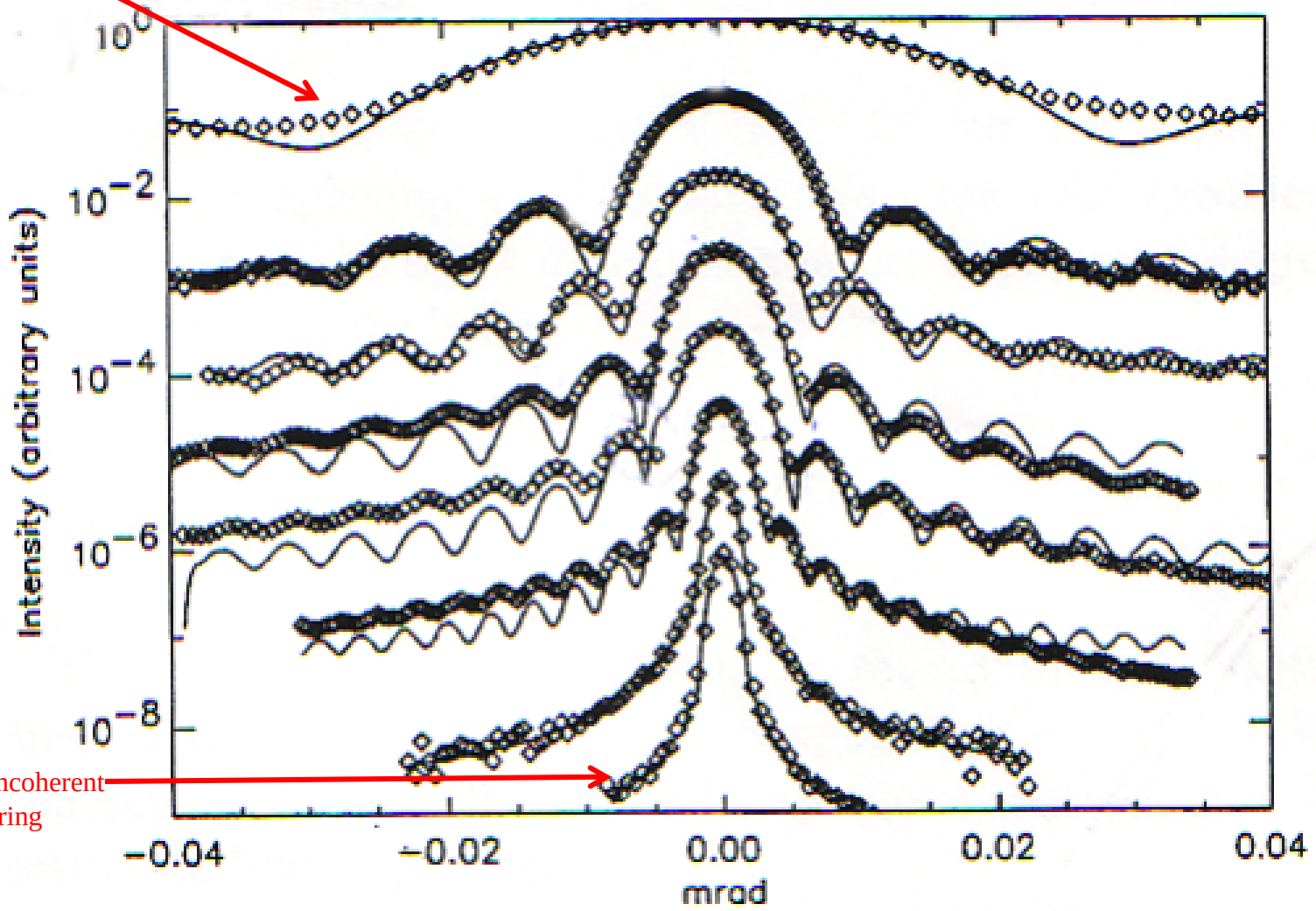
e.g. the number of wavelengths that can be added before the uncertainty adds up to a full wavelength.

Can also be viewed as a coherence time $T_c = \Lambda/c$

Fraunhofer X-ray Diffraction from a Slit

Narrow slit, coherent scattering

B. Lin et. al. RSI 67, (9) (1996)



Wide slit, incoherent scattering

How Practical is it to Make X-rays Coherent?

Consider a point 65 meters downstream of an APS Undulator A

$$\lambda = 0.2\text{nm}, \quad \Delta\lambda/\lambda = 3\times10^{-4}$$

Ge 111

$$\sigma_x = 254\mu\text{m}, \sigma_y = 12\mu\text{m}$$

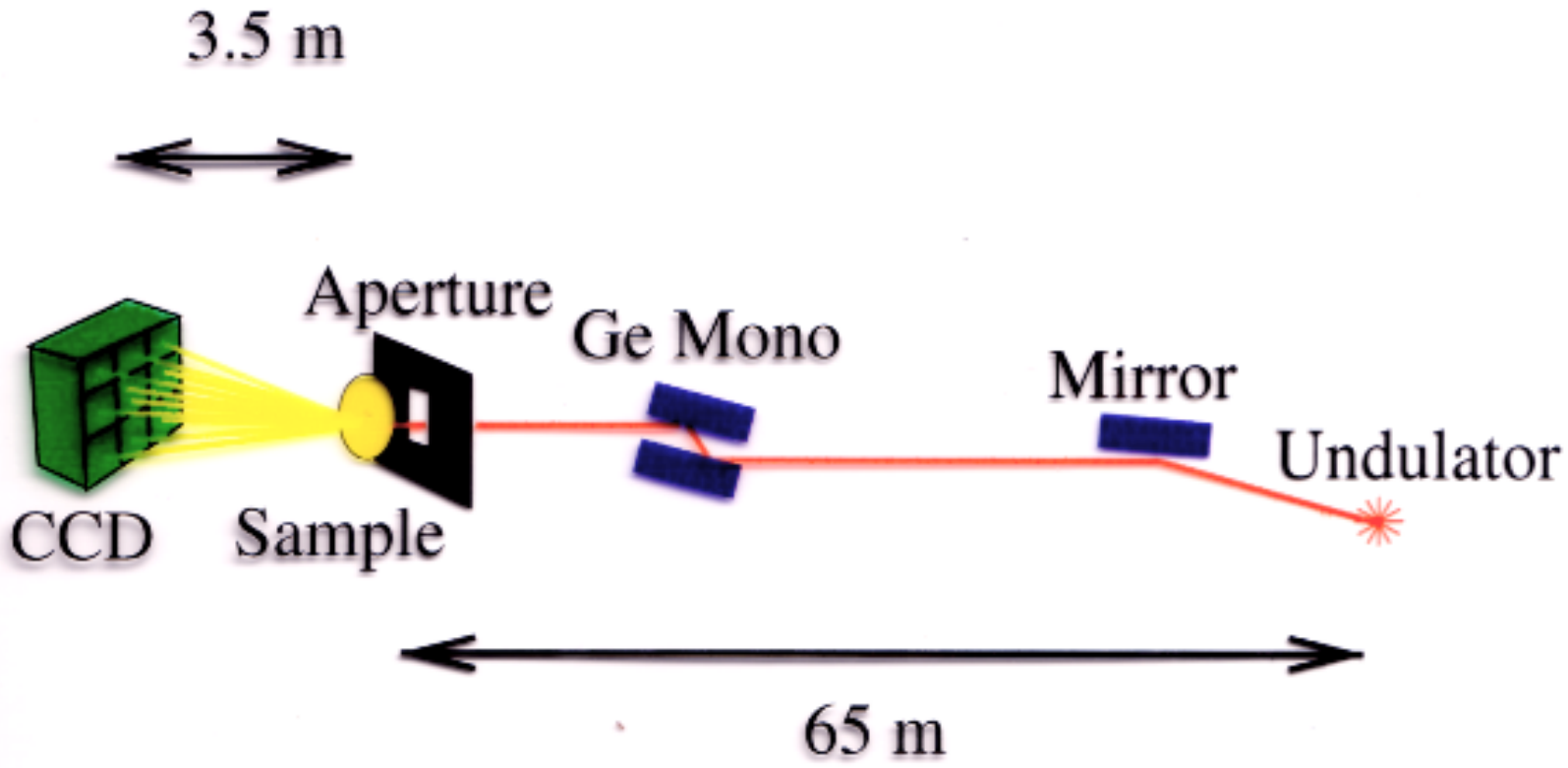
$$\xi_x = \frac{\lambda R}{2\sigma_x\sqrt{\pi}} = 14\mu\text{m}$$

$$\xi_y = \frac{\lambda R}{2\sigma y\sqrt{\pi}} = 306\mu\text{m}$$

$$\Lambda = 0.66\mu\text{m}$$

$\sim 3\times10^{10}$ Photons/Coherence Area

Setup for XPCS at Sector 8 of the APS



Optics must preserve coherence

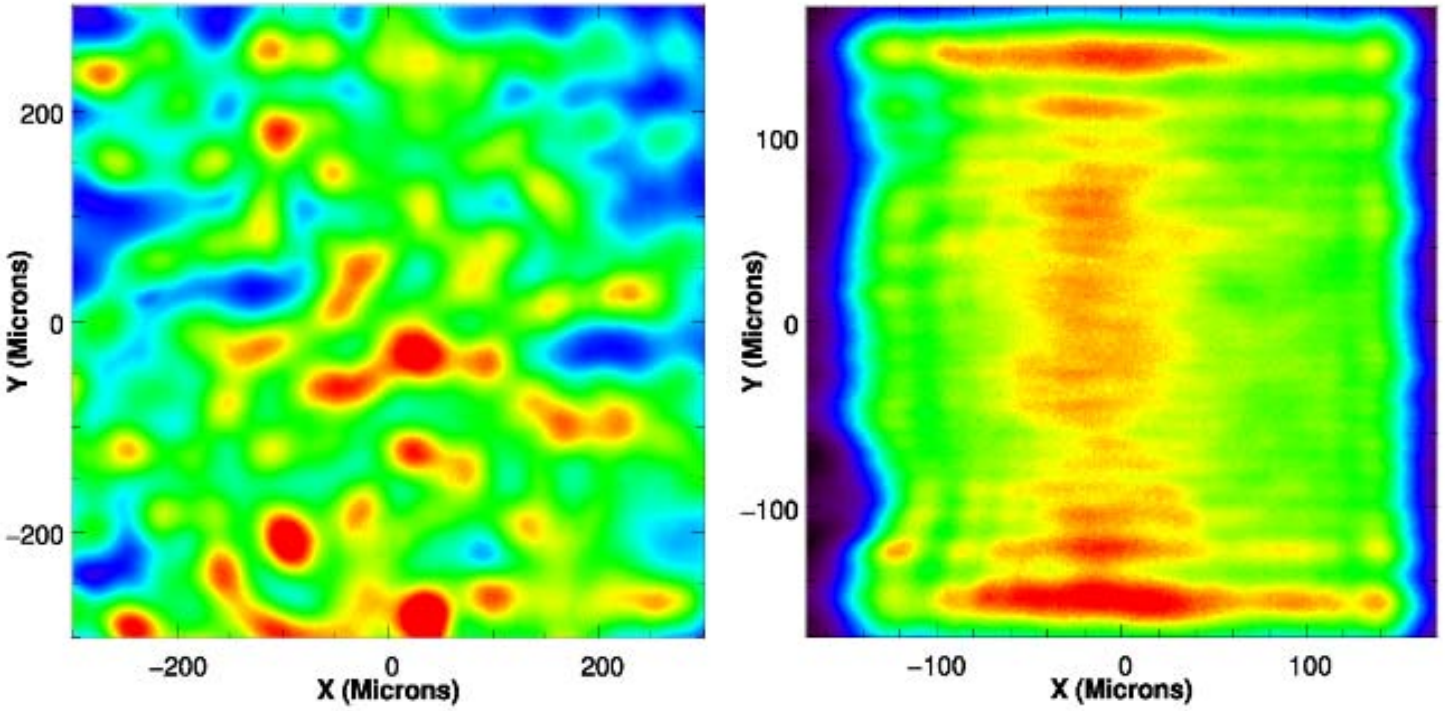


Image of x-ray beam reflected from channel cut monochromator (left) vs. artificial channel cut which allows better polish of interior faces.

S. Naryanan, A. Sandy, M. Sprung, D. Shu and J. Sullivan

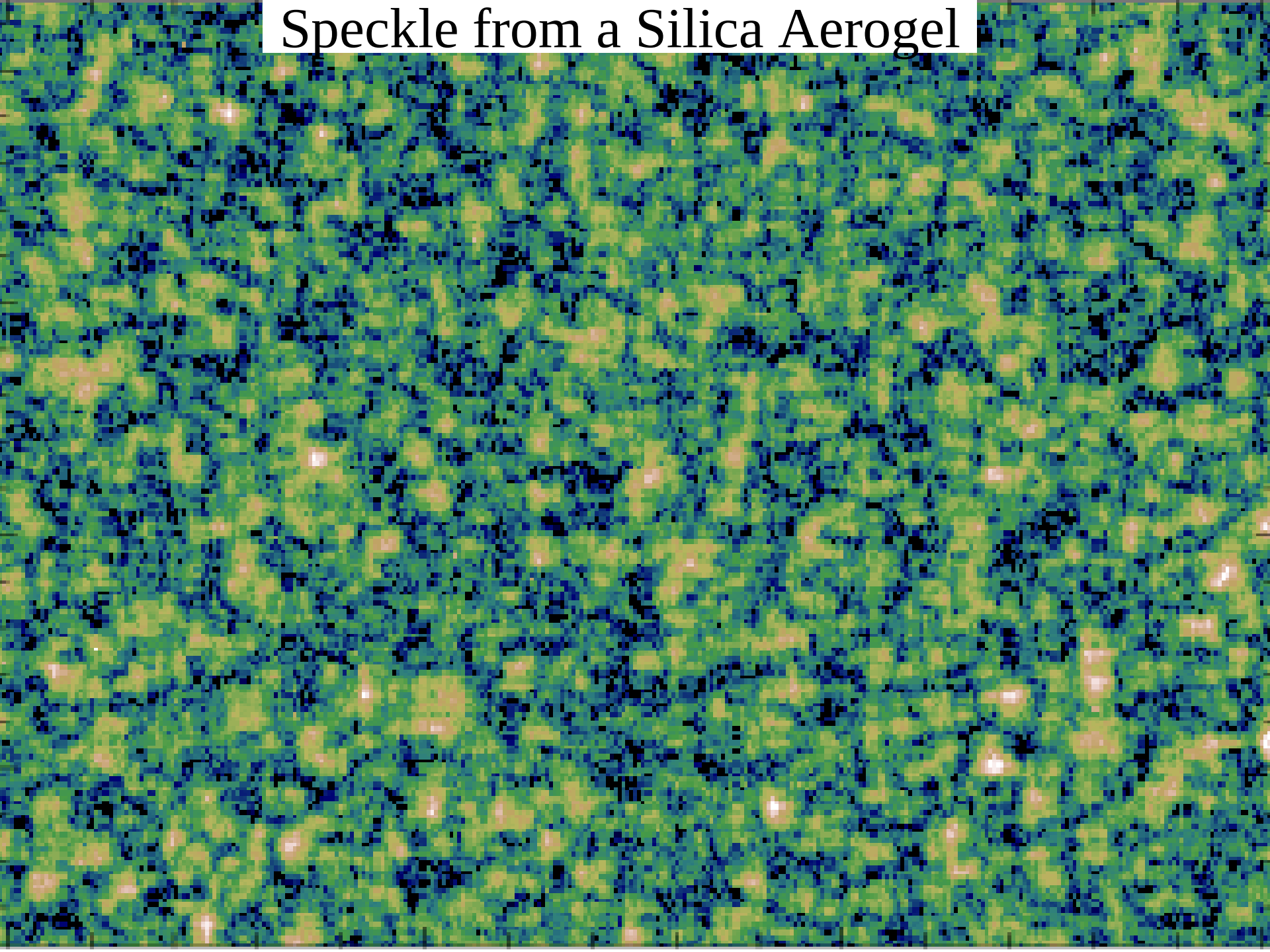
Scattering of Coherent X-rays

$$I(Q) \propto \int\int e^{i\vec{Q}\cdot\vec{r''}} \rho_e\left(\vec{r}\right)\rho_e\left(\vec{r}-\vec{r''}\right)d\vec{r}d\vec{r''}$$

For incoherent x-rays the actual scattering represents a statistical average over many incoherent regions within the sample and one obtains:

$$\rho_e\left(\vec{r}\right)\rho_e\left(\vec{r}-\vec{r''}\right) \approx \left\langle \rho_e\left(\vec{r}\right)\rho_e\left(\vec{r}-\vec{r''}\right)\right\rangle \equiv g\left(\vec{r}\right)$$

For coherent x-rays one measures the Fourier transform of the exact density distribution, not the average. What one observes is a speckle pattern superposed on the average scattering pattern.

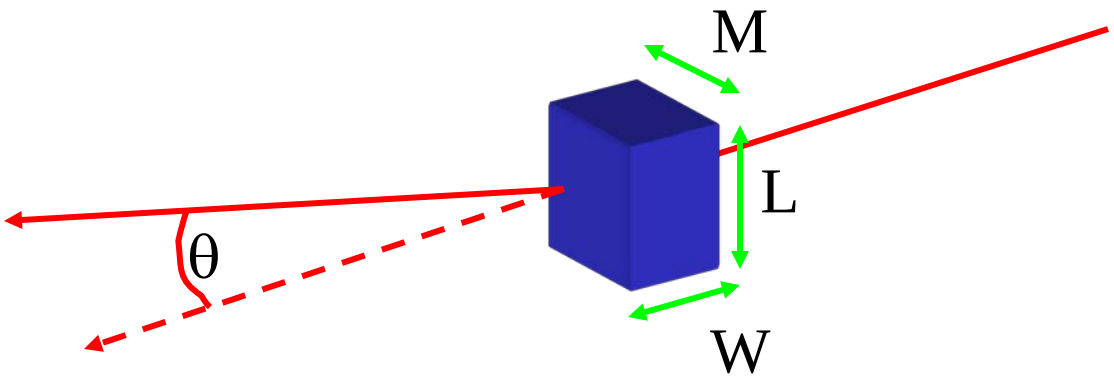

Speckle from a Silica Aerogel

Speckle Size and Contrast

The speckle widths are approximately the size of the diffraction pattern from a slit the size of the sample:

$$\Delta\theta \approx \lambda / L$$

The contrast is given by the ratio of the scattering volume to the coherence volume, $\Lambda\xi_x\xi_y/MLW\sin(\theta)$



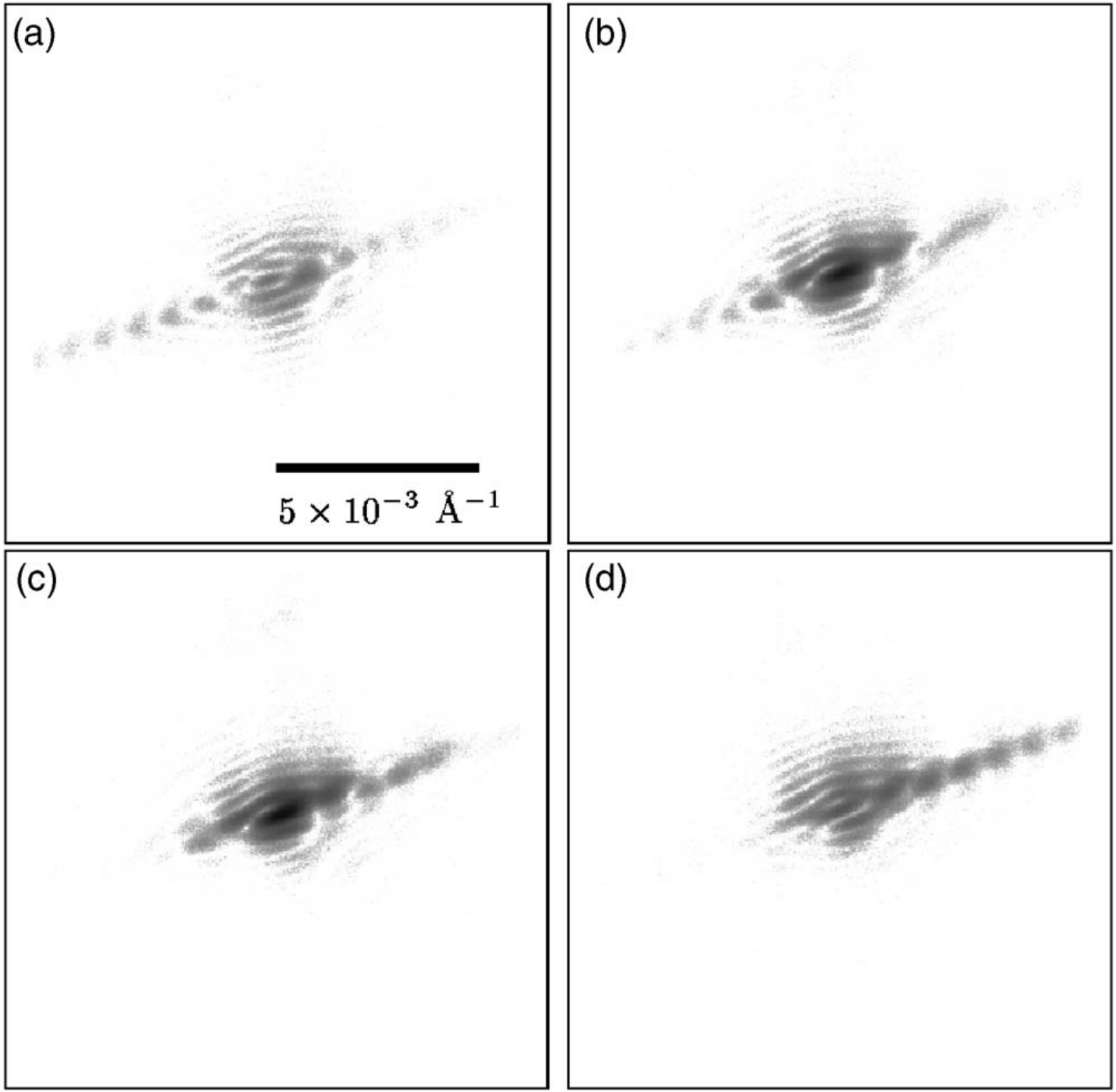
Exact numbers require integrals over the sample volume and electric field spatial correlation function. For small angles, the scattering volume is much smaller than the sample volume.

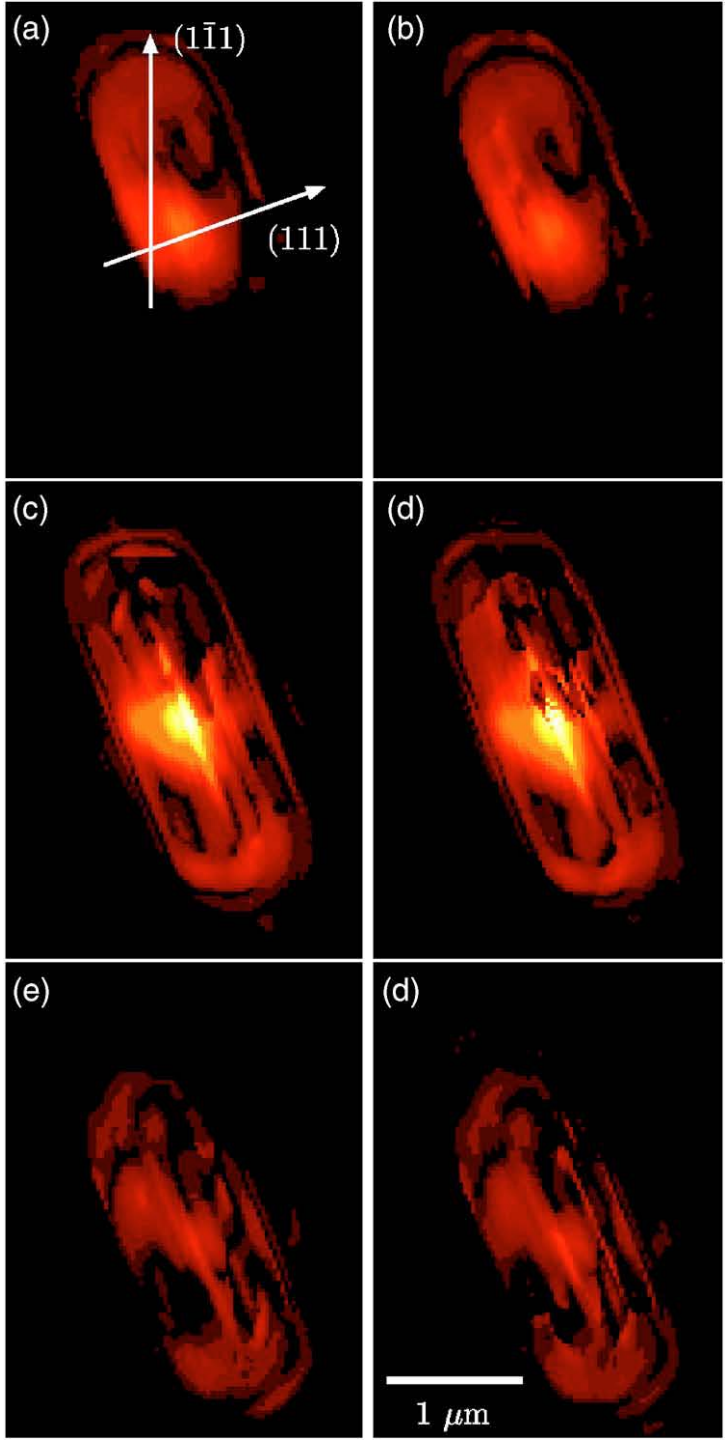
What to do with coherent x-rays?

- Try to invert the speckle to get information about the exact structure factor.
- Ignore the details of the exact structure factor, but use the time fluctuations of the pattern to study dynamics of the material (XPCS)

Example: G. J. Williams, M. A. Pfeifer, I. A. Vartanyants, and I. K. Robinson, PRB 73, 94112

- Mathematically invert pattern using constraints of finite support, real positive density and over sampling of image.
- Look at pattern around a diffraction peak. Sensitive to crystal strain, avoids scatter around direct beam.
- Iterative technique.
  - Guess random phases
  - Transform data back to real space and impose constraints.
  - Transform data back to reciprocal space and impose amplitudes.
  - Cycle until convergence (not always easy)


(a)
(b)
(c)
(d)
5×10^{-3} Å^{-1}

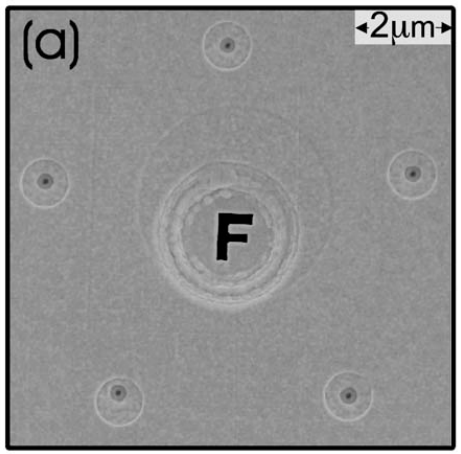


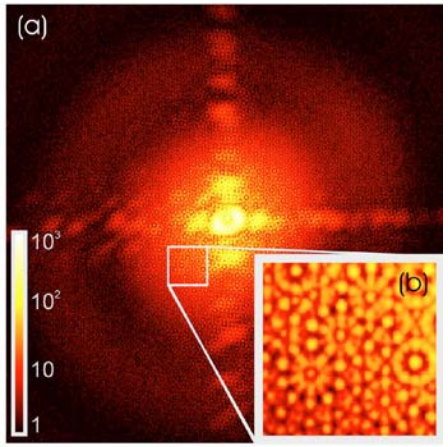
2D slices through the reconstructed 3D density are shown for two fits. The slices are separated by 1.35 μm in the third direction, oriented out of the page. Both fits possess a bright region in the center of the real space reconstruction, seen in c and d, which is likely an artifact due to the partial coherence of the beam.

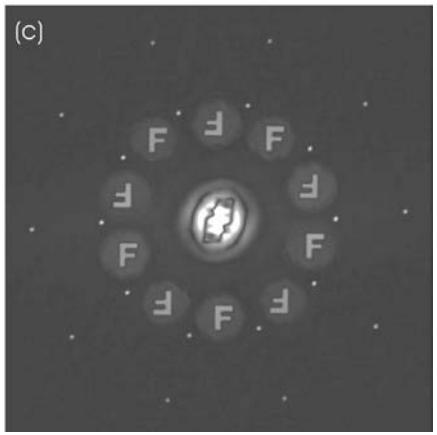
Another approach: Supply Reference Beam to Obtain the Phase.

Example: W. F. Schlotter,a R. Rick, K. Chen A. Scherz, J. Stöhr, J. Lüning S. Eisebitt, Ch. Günther, W. Eberhardt O. Hellwig and I. McNulty, APL, 89, 163112

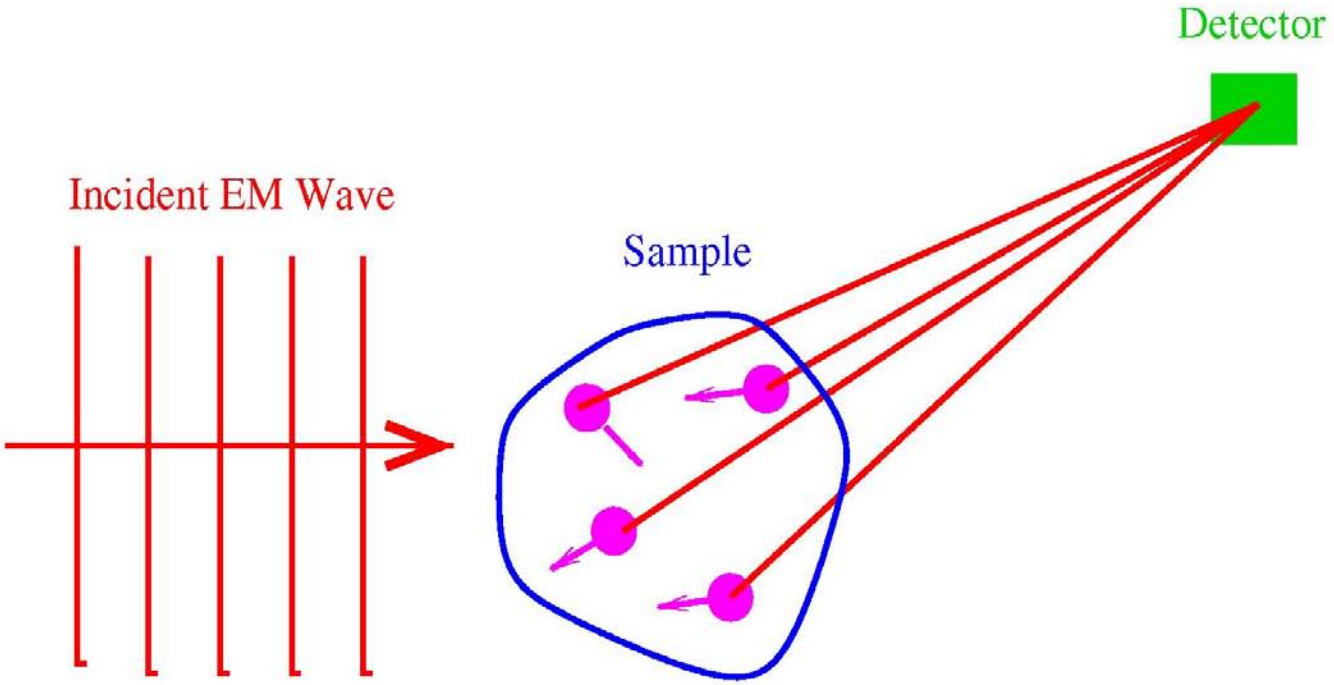
- Experiment is done in soft x-rays (1.58 nm) in SAXS







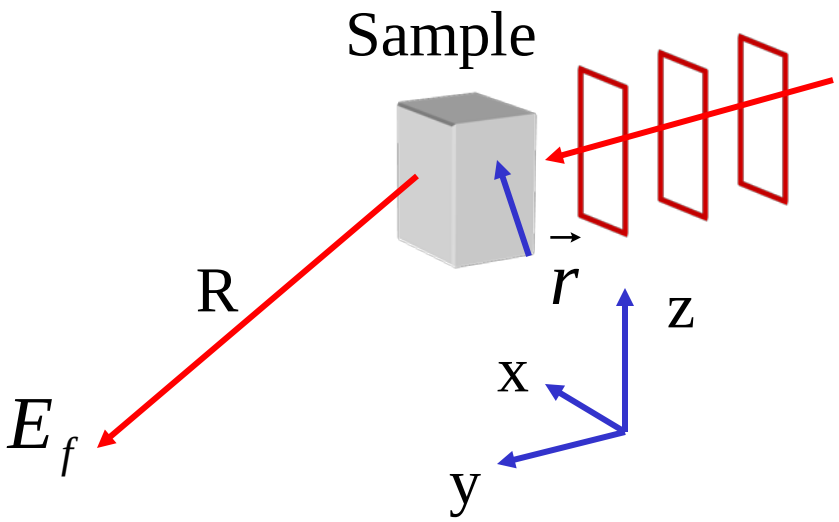
Measuring Dynamics



Typical applications are where the average structure is constant, but the local structure fluctuates.

- Diffusion of particles in solution
- Concentration fluctuations in binary liquids
- Fluctuations of order parameter in a crystal
- Thermally driven surface height fluctuations in a viscous fluid
- Vibrations of a membrane
- Aging: evolution of the “equilibrium” dynamics with time.

The Intensity-Intensity Correlation Function



$$g_2(Q,\tau) \equiv \frac{\langle I(Q,t)\, I(Q,t+\tau)\rangle}{\langle I\rangle^2}$$

How to calculate g_2

- Calculate electric field intensity correlation function at the observation point:

$$G_2(Q,\tau)=\int\exp(iQ\cdot r')\langle E_f^2(r,t)\,E_f^2(r+r',t+t)\rangle_{r,t}dr'$$

- The fourth order correlations in E, can be reduced to pairs of second order correlation functions

- Assume correlation lengths are smaller than sample size, and the scattering can be factored into independent space and time parts.

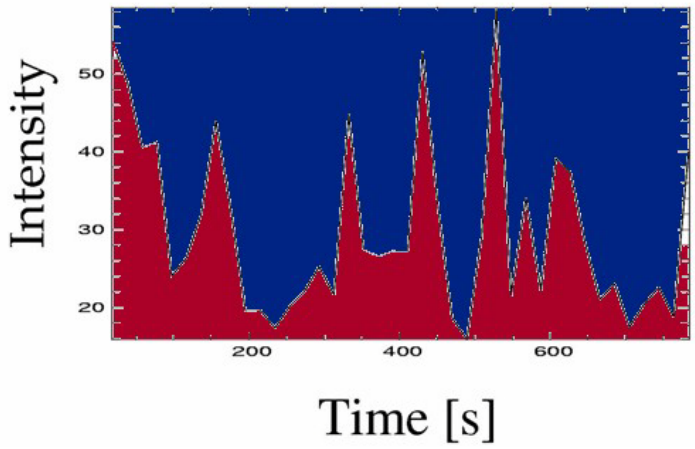
Final Result

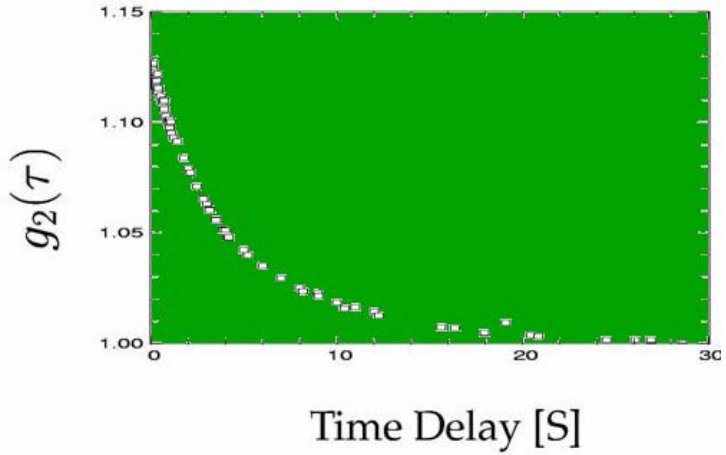
$$G_2(\vec{Q},\tau) = \langle I \rangle^2 \left[1 + \beta f(Q,\tau)^2 \right]$$

The contrast factor, β, is related to the degree of coherence and can be between 0 and 1

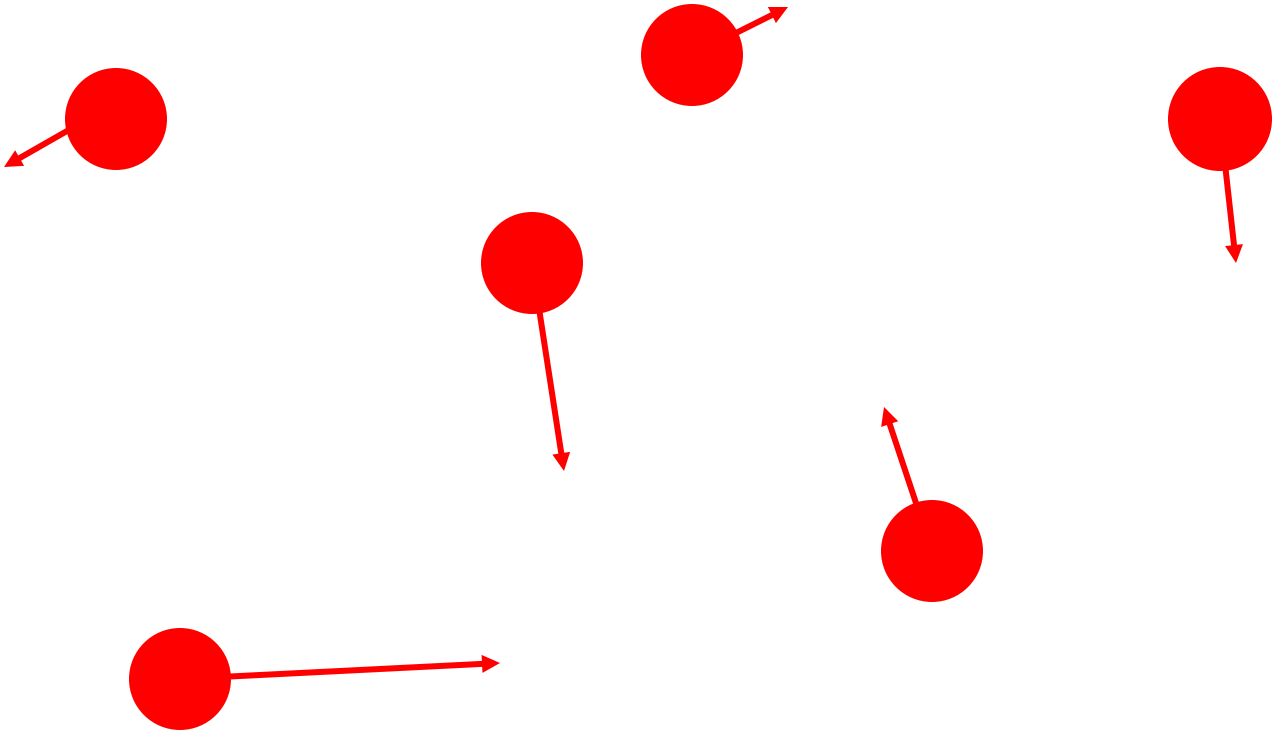
$$f(\vec{Q},\tau) = S(\vec{Q},\tau) / S(\vec{Q},0)$$

$$S(\vec{Q},\tau) = \left\langle \int e^{i\vec{Q}\cdot\vec{r}} \rho_e(0,0)\rho_e\left(\vec{r},\tau\right) d\vec{r} \right\rangle$$





A dilute colloidal suspension (71 nm Latex in Glycerol)



To theoretically calculate the dynamic scattering factor one has to take the correlation functions for a collection of point scatterers diffusing in the liquid.

$$f(\vec{Q},\tau) = \frac{1}{N}\sum_{i,j}\left\langle \exp(i\vec{Q}\cdot\left(\vec{r}_i(t) - \vec{r}_j(t+\tau)\right)\right\rangle$$

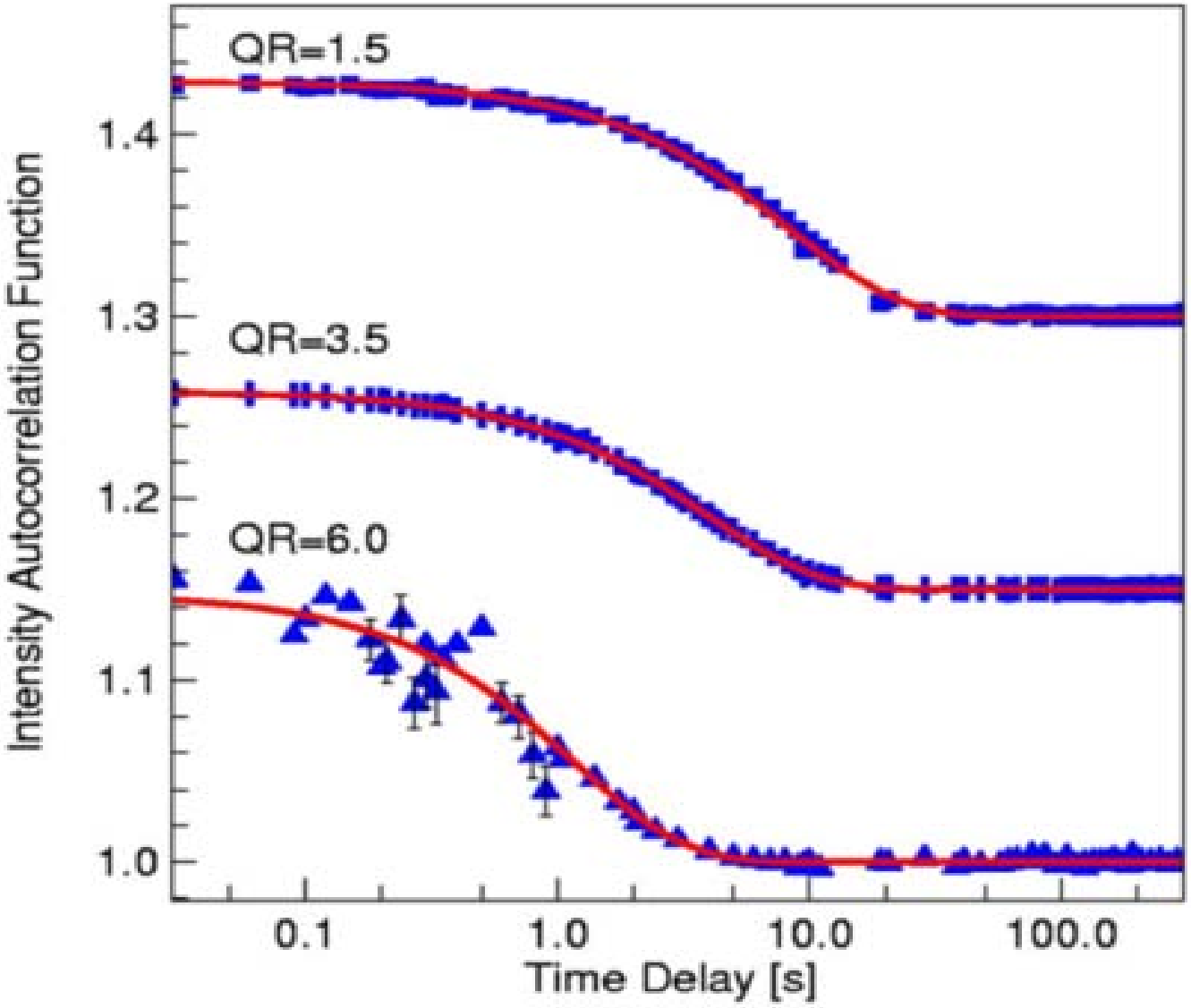
For Brownian motion, this can be reduced to an exponential decay proportional to the diffusion coefficient.

$$f(Q,\tau) = e^{-DQ^2\tau}$$

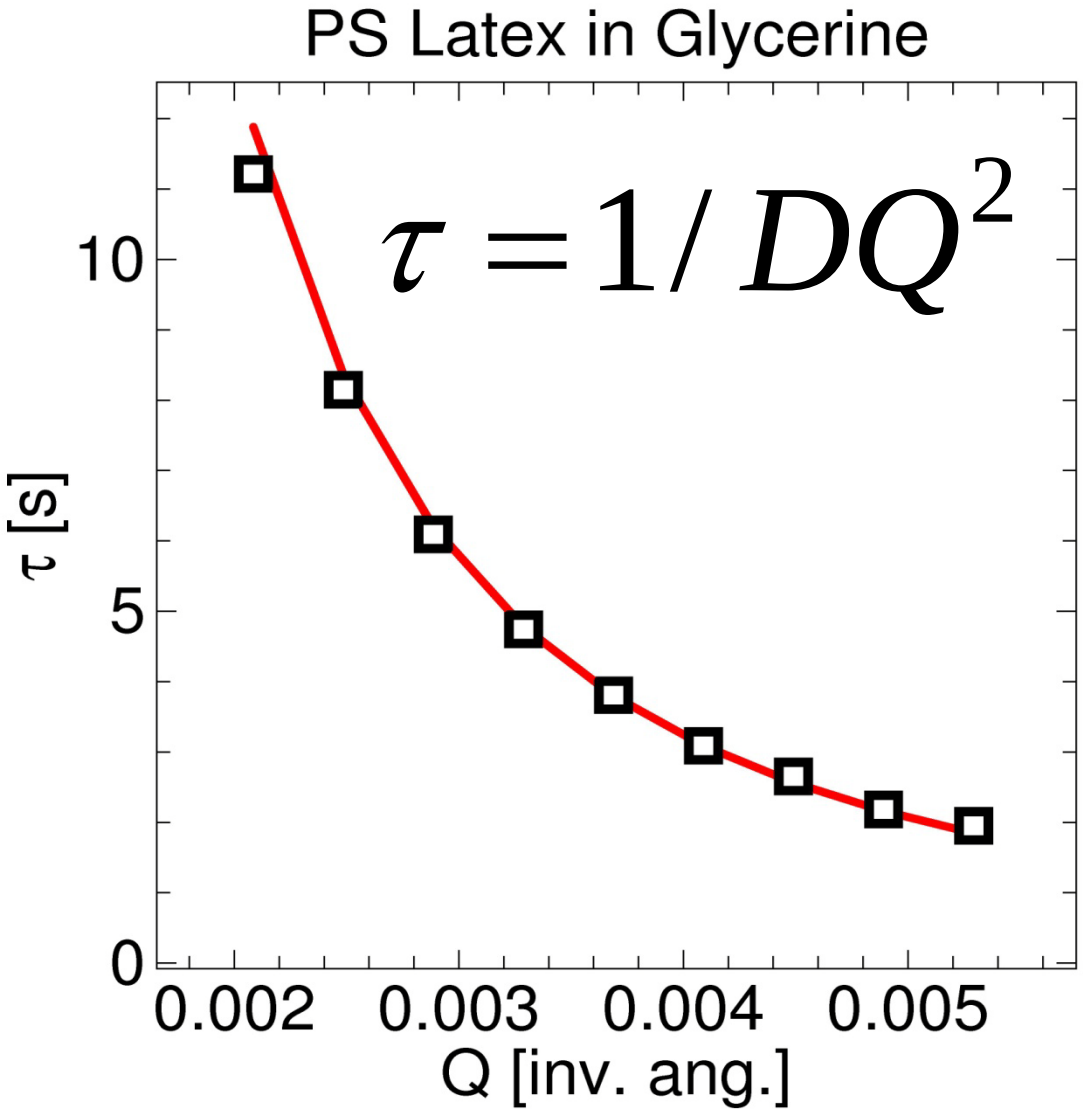
Here, the diffusion coefficient is related to the viscosity, η and the radius a, via the Stoke-Einstein relation:

$$D = k_B T \,/\, 6\pi\eta a$$

Correlation Functions



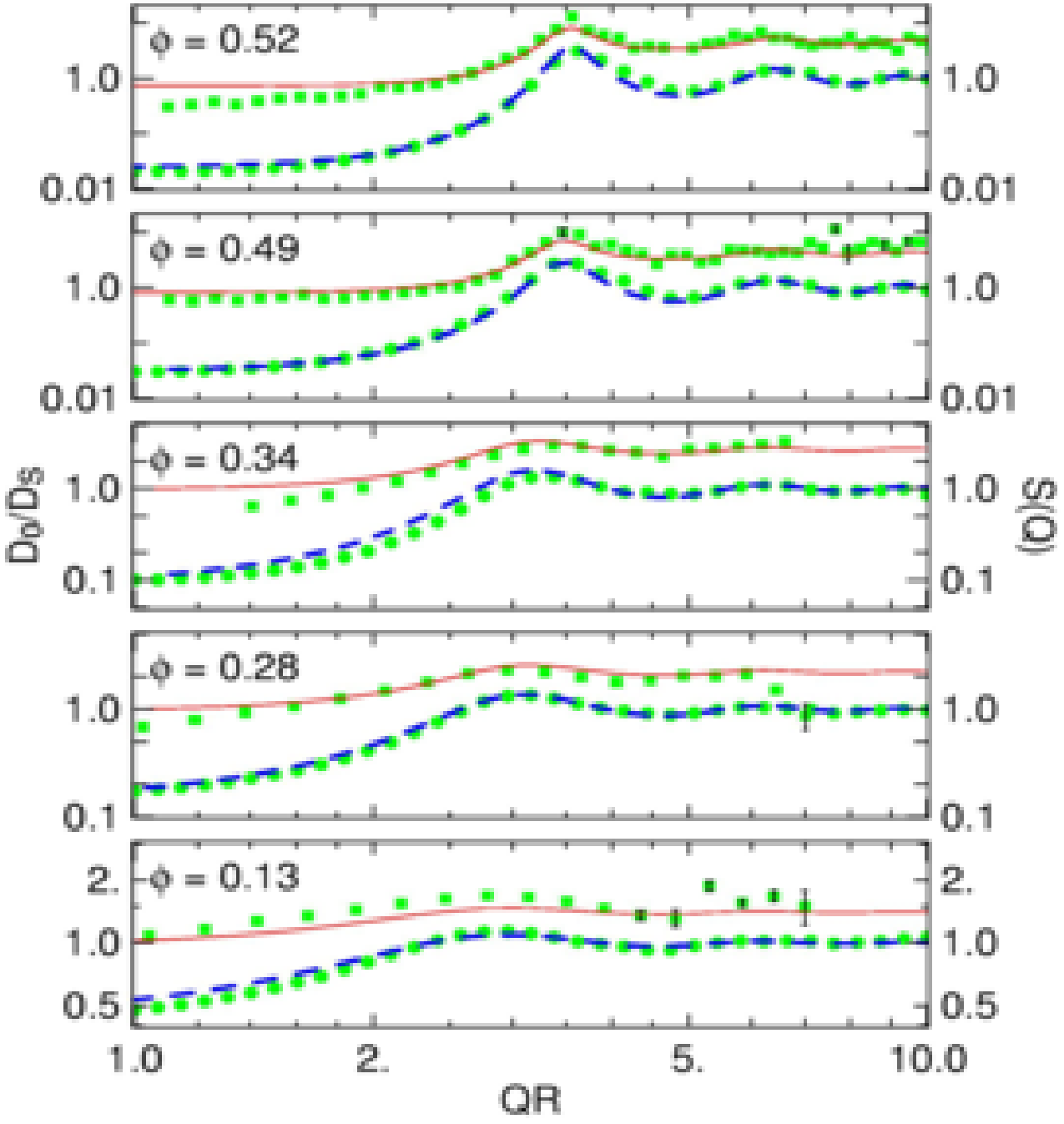
Wavevector Dependence



Short Time Diffusion Constants in Concentrated Suspension

1. Structural correlations lead to a slowing down of dynamics
2. Hydrodynamic interactions further modify the dynamics at high concentration
3. These effects can be calculated for the initial decay rate of the correlation function, but the f(Q,t) will not generally be an exponential at long times.

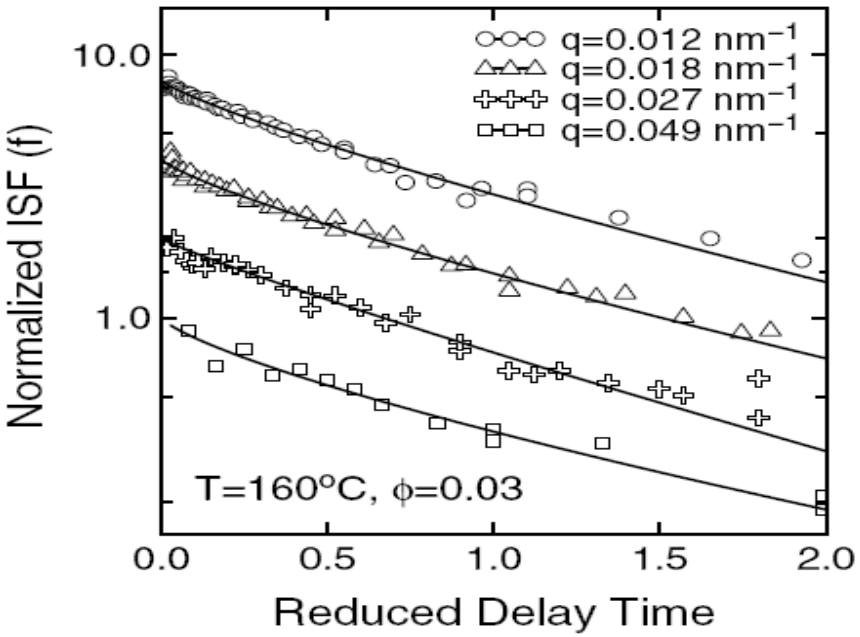
$$D(Q) = D_0 H(Q) / S(Q)$$


ϕ = 0.52
ϕ = 0.49
ϕ = 0.34
ϕ = 0.28
ϕ = 0.13
1.0
0.01
0.1
2.
0.5
D_ϕ/D_S
S(Q)
1.0
2.
5.
10.0
QR

Example 2 Concentrated Polymeric Vesicles

Concentrated Block Copolymer Vesicles

Falus et. al. PRL 94, 16105, 2005



In Liquid State, Near Tg Exponential Decays Become Stretched

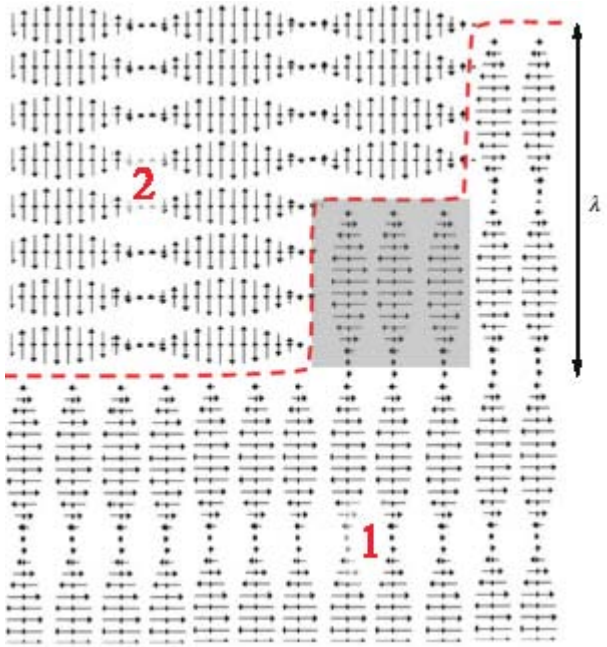
$$f(q,t) = \exp\left[-(\Gamma t)^{\beta}\right]$$

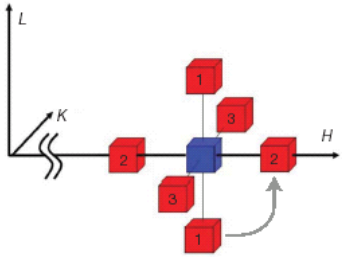
$$\beta < 1$$

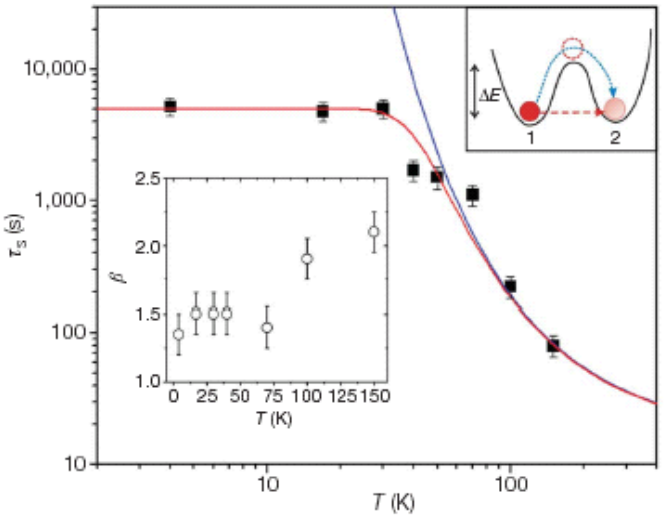
Example 3

Antiferromagnetic Domain Fluctuations

- O. G. Shpyrko *et al.*, *Nature* ***447, 68 (2007).***

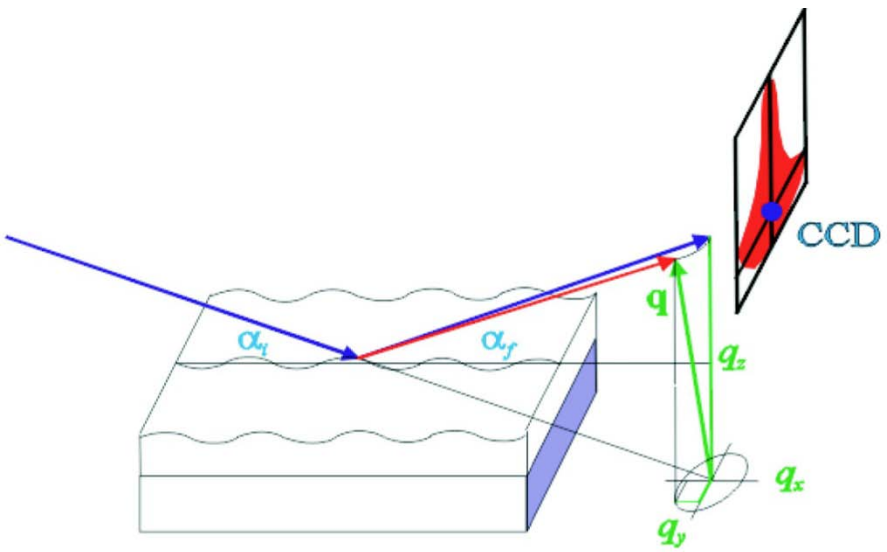


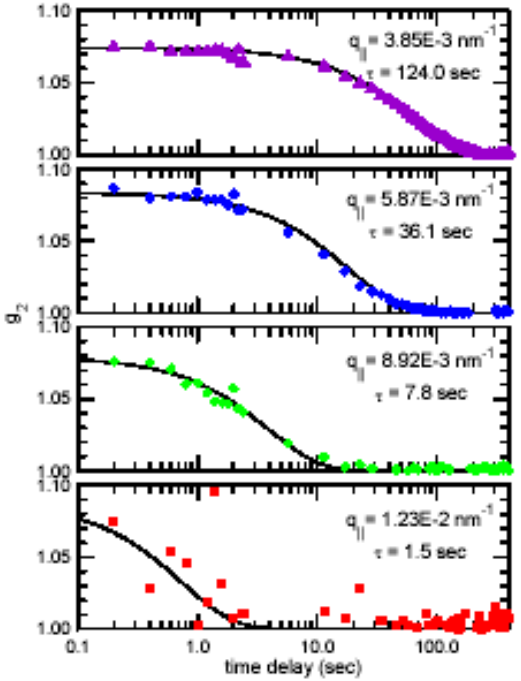


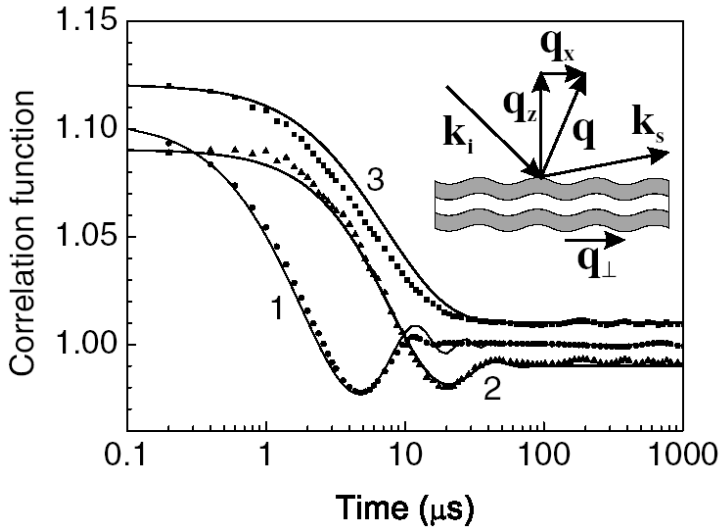


Note that although the length scale of the fluctuations is large (>10 nm) they require x-rays with wavelengths ~0.1 nm in order to be visible.

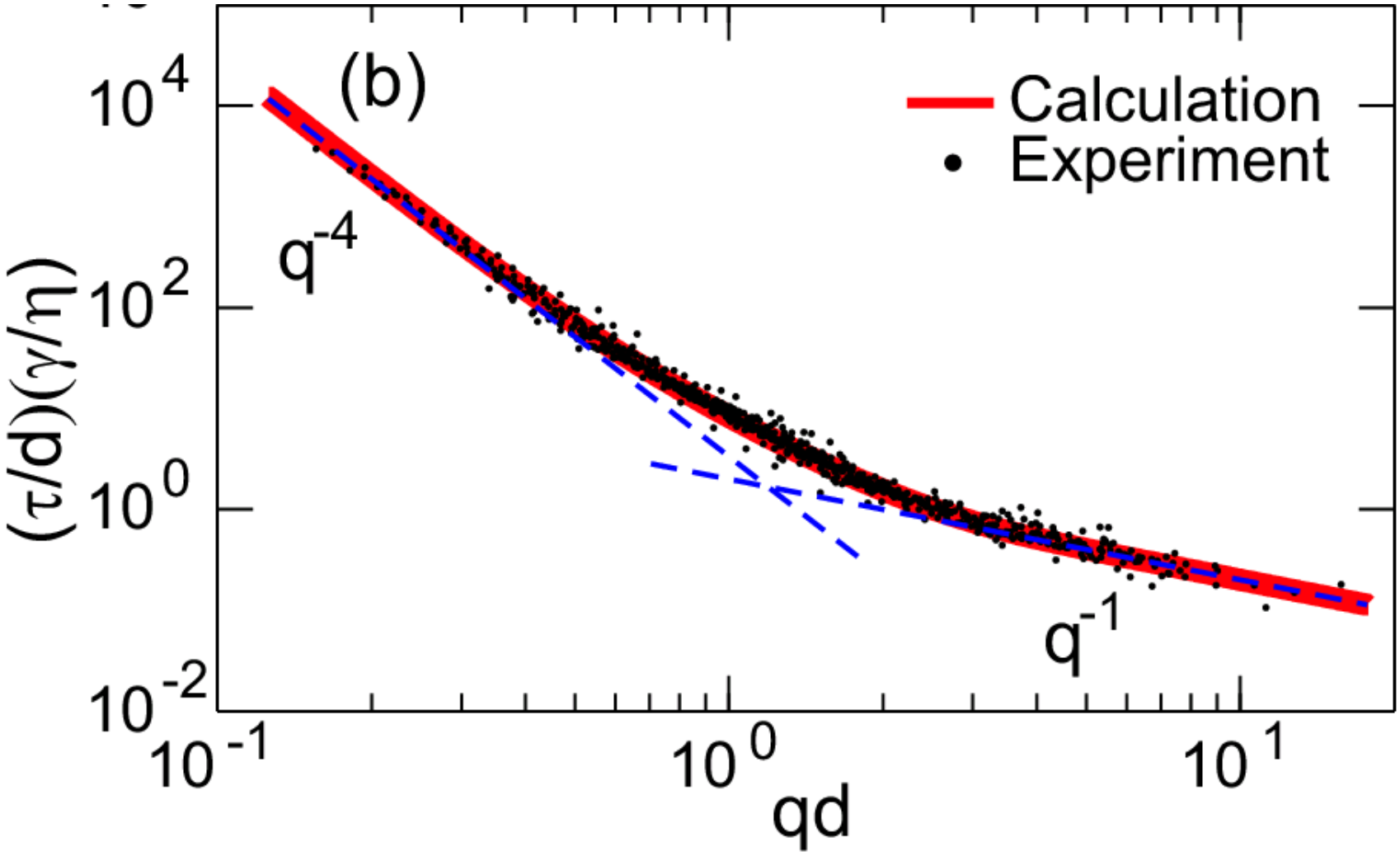
Example 4: Dynamics of Surfaces







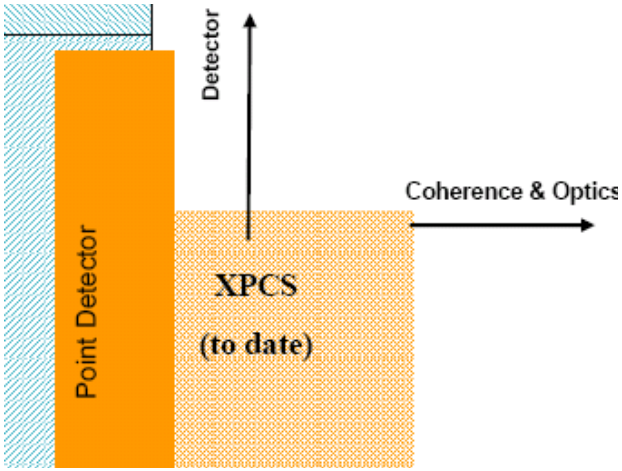
- Polystyrene films on Si wafers (Kim, PRL 2003) are highly viscous and show exponential decay
- Thin liquid crystal membranes (Sikharulidze, PRL 2002) show transition from oscillatory to overdamped behavior.



From Jiang et. al. PRL, 246104, 2008

Limitations of XPCS

- Too much flux (X-ray damage)
  - Radiation can cause cross linking of polymers and charging of colloids.
  - A reasonably radiation resistant polymer (PS) in vacuum can absorb around ~10^7 Gy. (10 min in surface reflection geometry)
  - Protein in water can absorb ~10^5 Gy (10 sec in transmission geometry)
- Too little flux (Poor signal to noise)



Some tricks of the trade …

Multispeckle Detection

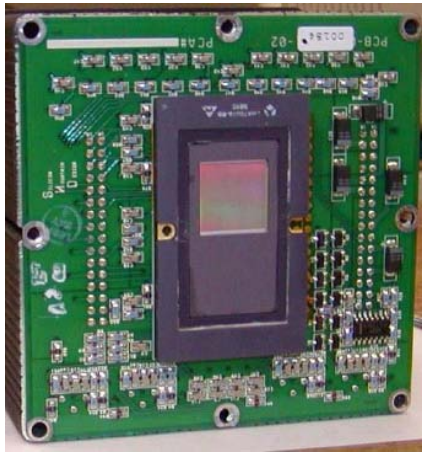
•Falus, Borthwick, Mochrie, RSI (2004)

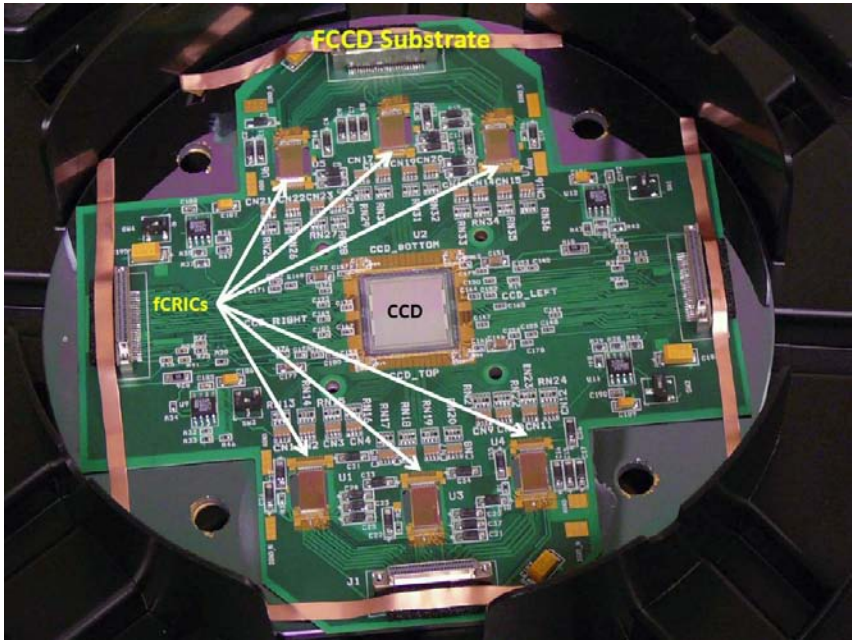
•SNR increases as $\sqrt{N}$

•Limitations:

- •Readout rate presently around 60 Hz
- •Efficiency < 50%

•New Camera under development at LBL and APS ~500 Hz at 100% efficiency. (John Weizeorick and Alec Sandy) Should see first light this cycle.

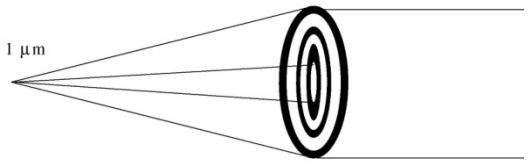




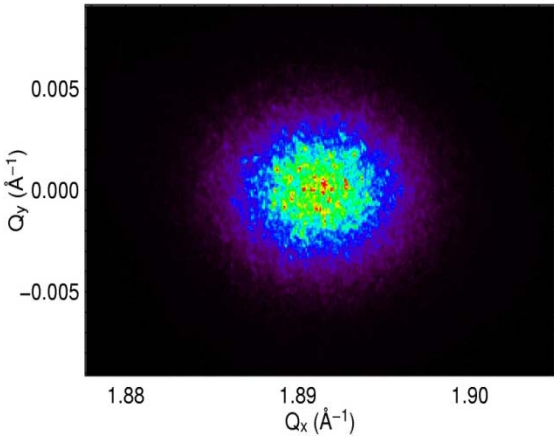
Focusing

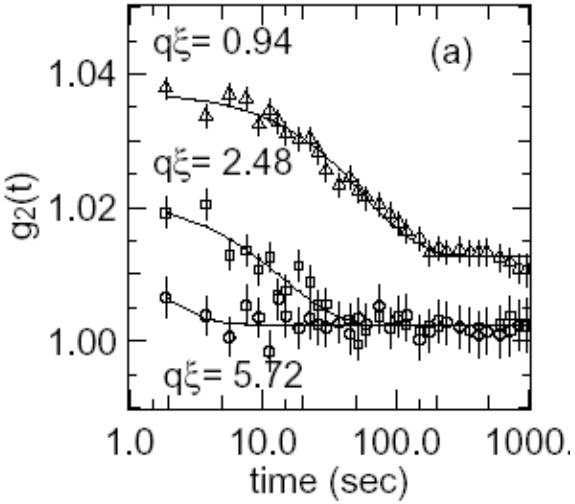
Khalid Laaziri and Mark Sutton

- Focusing the x-ray beam down to a small spot (~1 μm) maintains flux, but increases the speckle size. SNR ~ photons/speckle, so SNR goes up. Disadvantage is that radiation damage goes up as well.
- Fluctuations in Fe_3Al



Fresnel Zone Plate

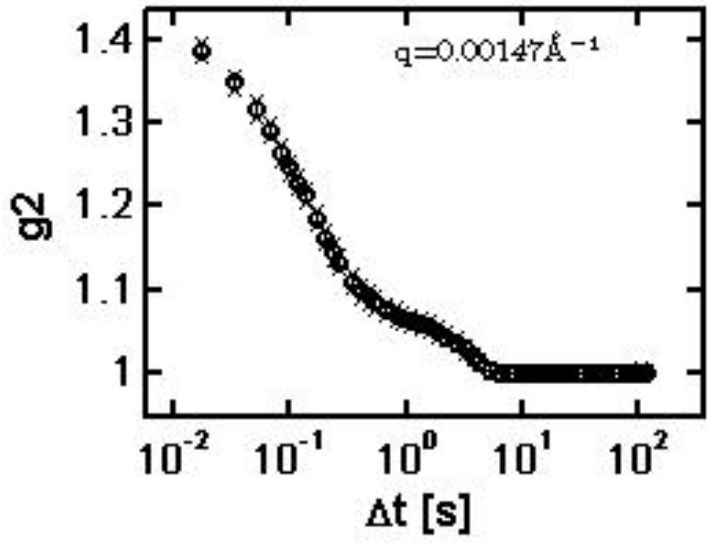




Flowing Sample

Marchand, Mukhopadhyay, Lurio

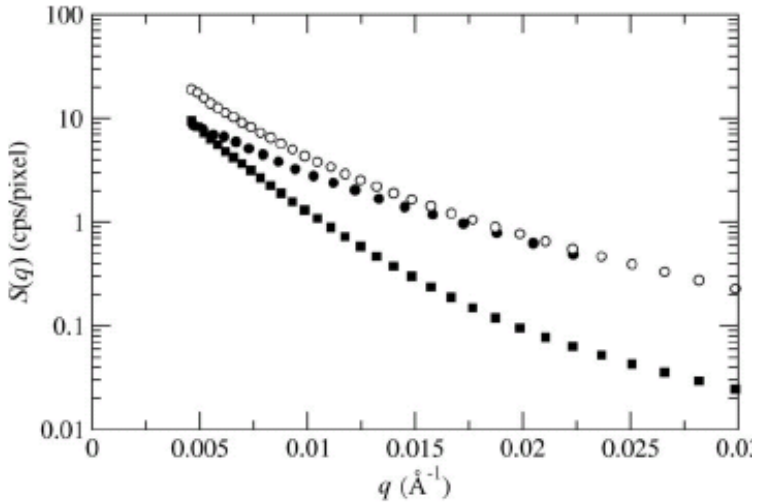
- Continuously flow sample so as to reduce damage.
- Need to separate flow dynamics from sample dynamics.

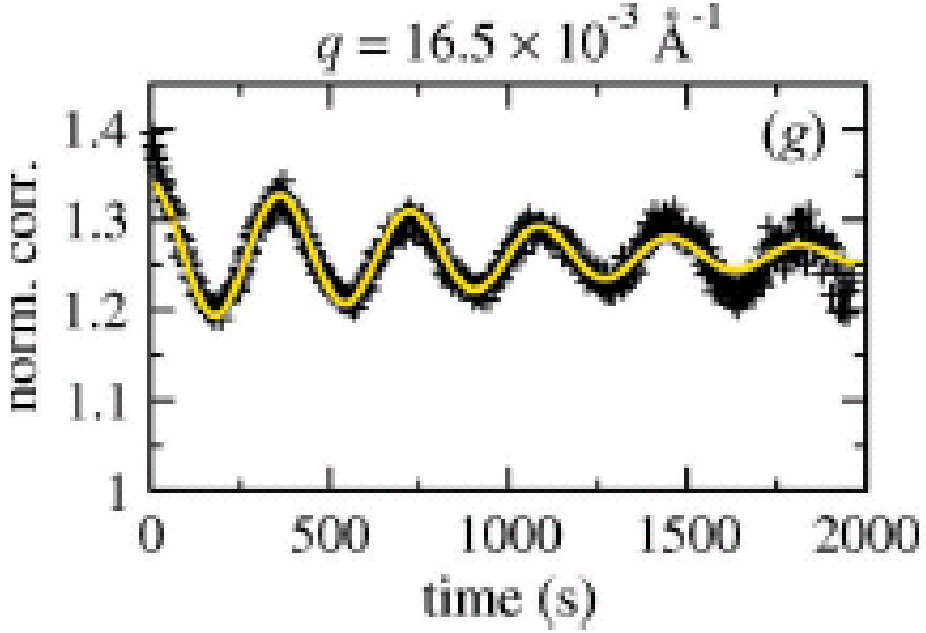


Heterodyne Detection

Livet. et. al. JSR (2006)

- Mix a static signal with a weaker dynamic signal.
- For diffusive motion relaxation times are longer: exp(-Γt/) instead of exp(-2Γt).
- Constant flow can be detected, which is invisible to homodyne. (example aerosil + carbon-black rubber)

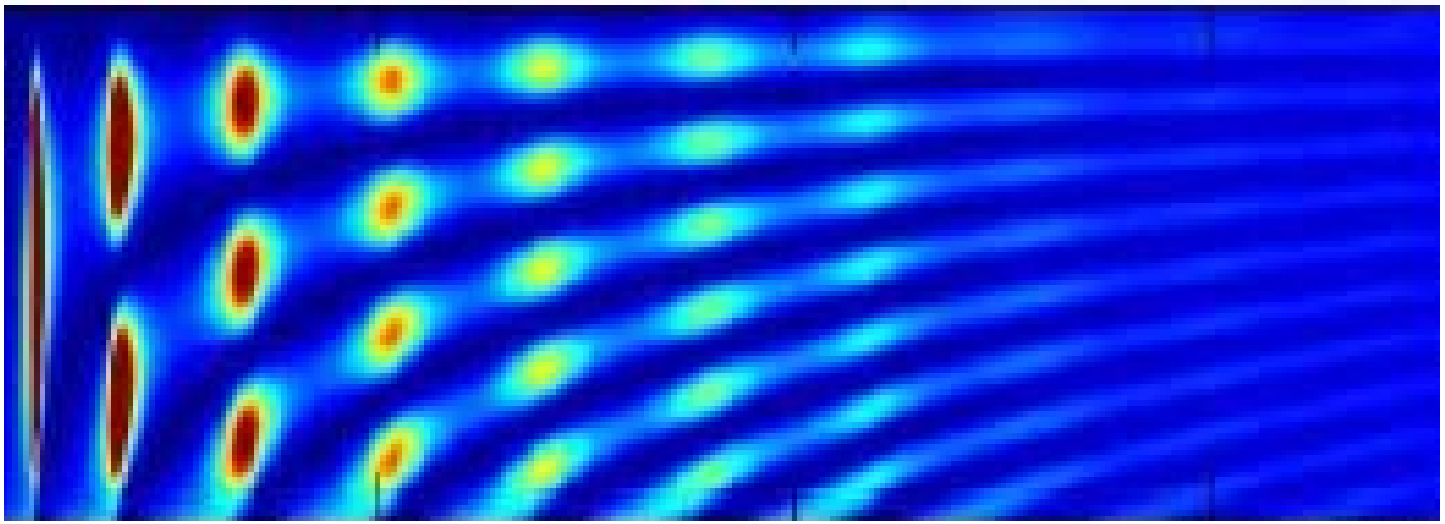




Wave Guides

Hu et. al. PRE, 010602R, 2006 and S. Narayanan et. al PRL, 185506, 2007

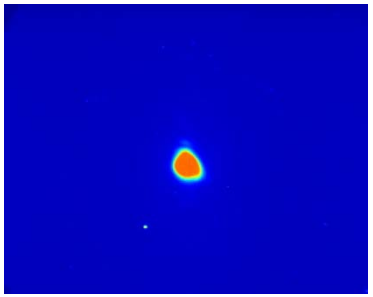
- Use a thin film (typically the sample!) as an x-ray wave guide.
- Standing wave intensities can be increased by over an order of magnitude.



Future Developments

- Better Detectors: Faster detectors with higher efficiencies would yield immediate improvement in XPCS with no commensurate increase in x-ray damage
- Better Optics: Preserve coherence, taylor coherence lengths and speckle size. Tradeoff of flux damage with focussing.
- Better X-ray sources: Longer undulators, Energy Recovery Linacs. (NSLS II and APS Refresh)
- The LCLS will require a whole new way of thinking about dynamics measurements.

April 11, 2009, LCLS first lasing at 1.5 Angstroms



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