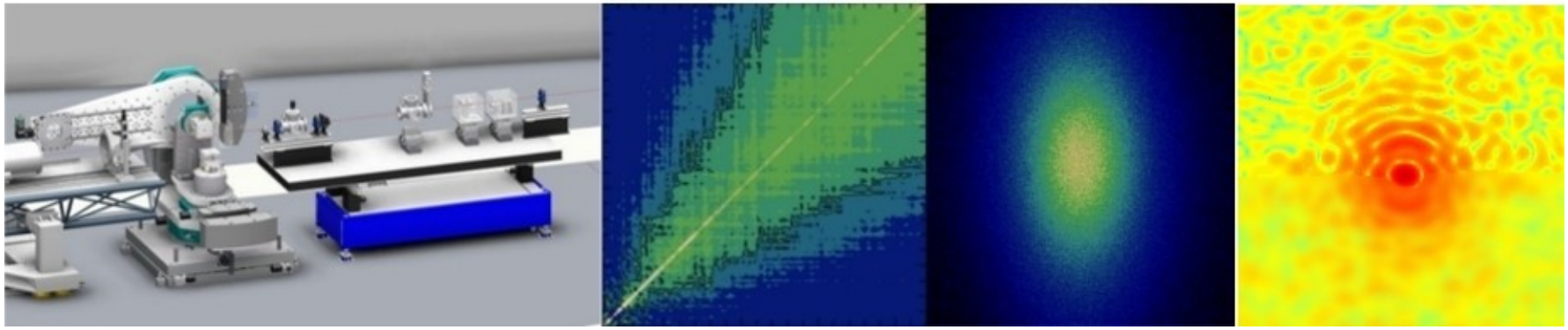
Dynamics with High Brightness Synchrotron Beams: Catching the Motion before it's to Late



Early Science Opportunities at CHX beamline NSLS-II

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Photon Sciences, BNL

NSLS, NSLS-II & CFN Users' Meeting

BNL, May 2014

CHX Beamline Overview

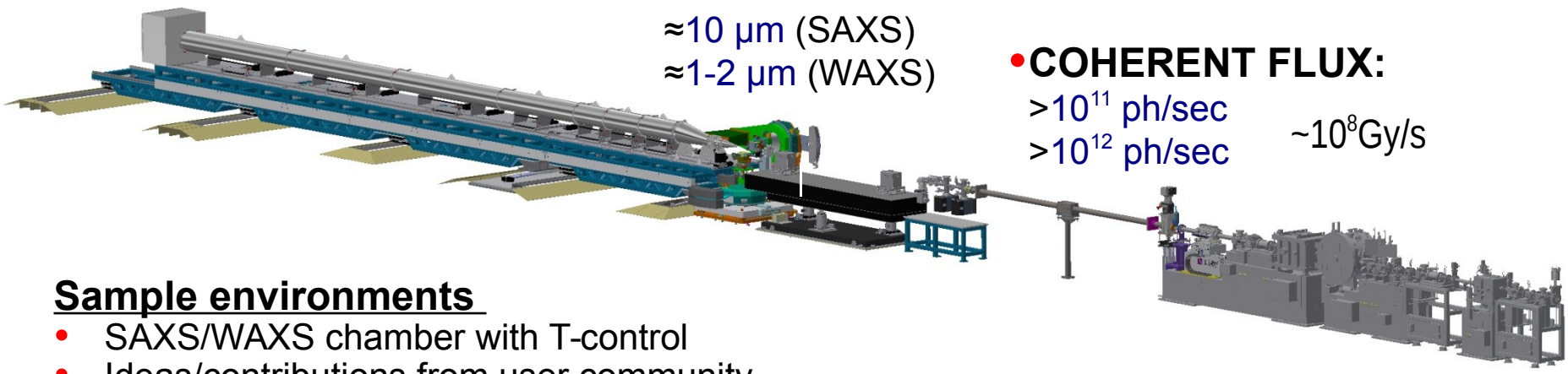
- **CHX optimized for Coherent X-ray Diffraction - *XPCS*, *(GI-)SAXS/WAXS*, *CDI***
  Unprecedented q-range available in-situ from Angstroms to Microns

- **Beamline Optics:** optimized for high stability & coherent wavefront preservation

- **BEAM SIZE**
  ≈10 μm (SAXS)
  ≈1-2 μm (WAXS)

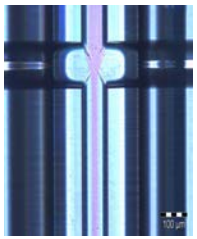
- **ENERGY RANGE:** 6–15 keV

- **COHERENT FLUX:**
  $>10^{11}$ ph/sec
  $>10^{12}$ ph/sec
  ~10^{8}Gy/s

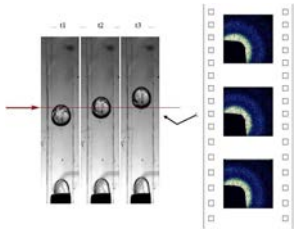


Sample environments

- SAXS/WAXS chamber with T-control
- Ideas/contributions from user community
  - Nano-Patterned substrates for GTSAXS
  - In-situ deposition chamber
  - etc
- Plans beyond project scope:
  - in-situ shear/flow
  - Langmuir trough
  - Etc.
- Direct observation (on-axis microscope)



A. Martel



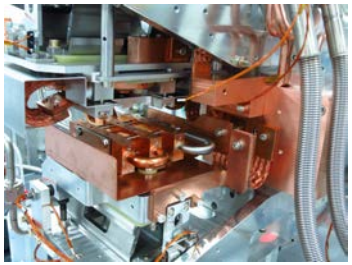
J.B.Salmon

L. Li

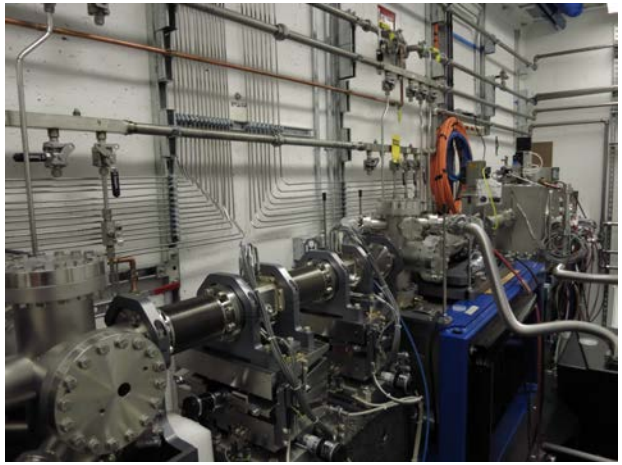
Beamline Readiness for First Experiments

X-ray Optics

- Designed to maximize stability and coherence preservation
- Installation is getting close to completion

DCM detail 

Transfocator

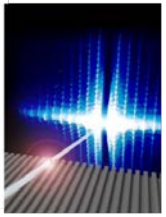


CHX FOE & beamline optics

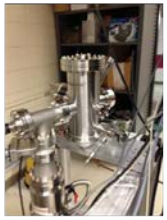
Sample Environments

- T controlled ("cryo-furnace") for SAXS/WAXS
- User contributed sample environments (i.e. from YOU :)

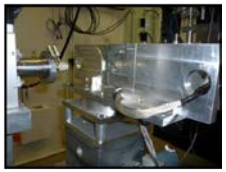
SAXS/WAXS



GT-SAXS



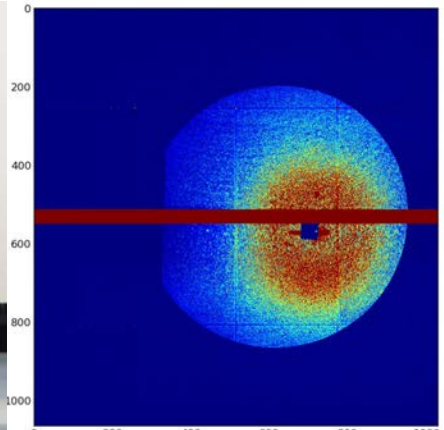
Thin Film deposition chamber



Shear Cell

Area Detectors

- c-WAXS: Eiger 1M (Dectris), 3 – 15 kHz
  - delivered: Mar 2014
- c-SAXS: Eiger 4M (Dectris)
  - delivery: Sept 2014



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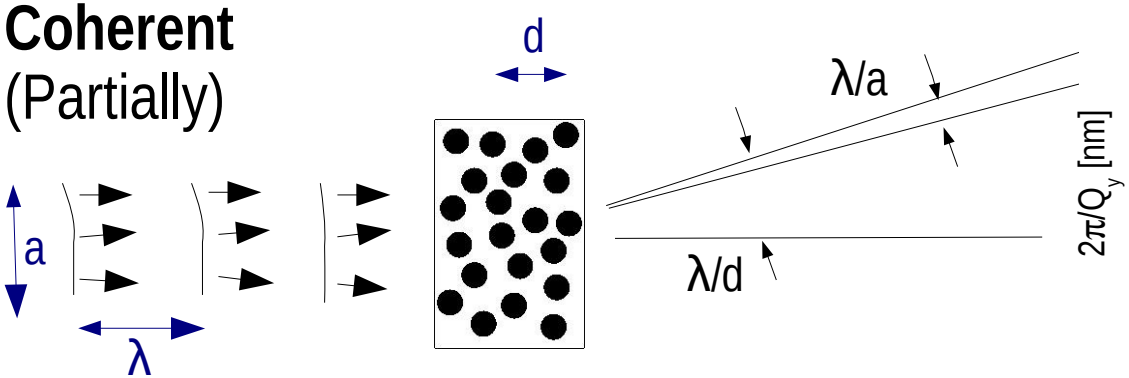
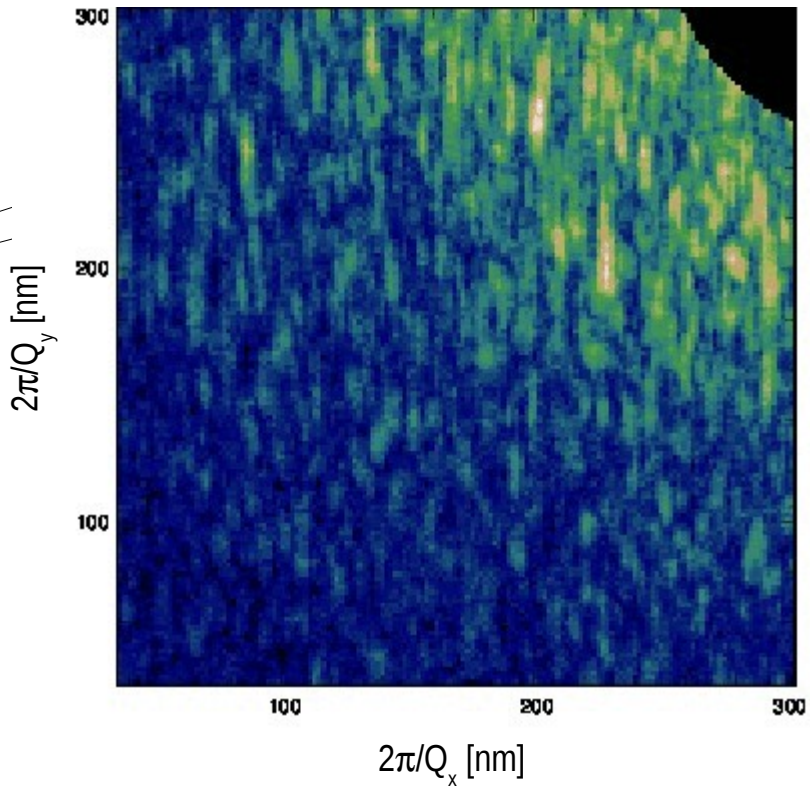
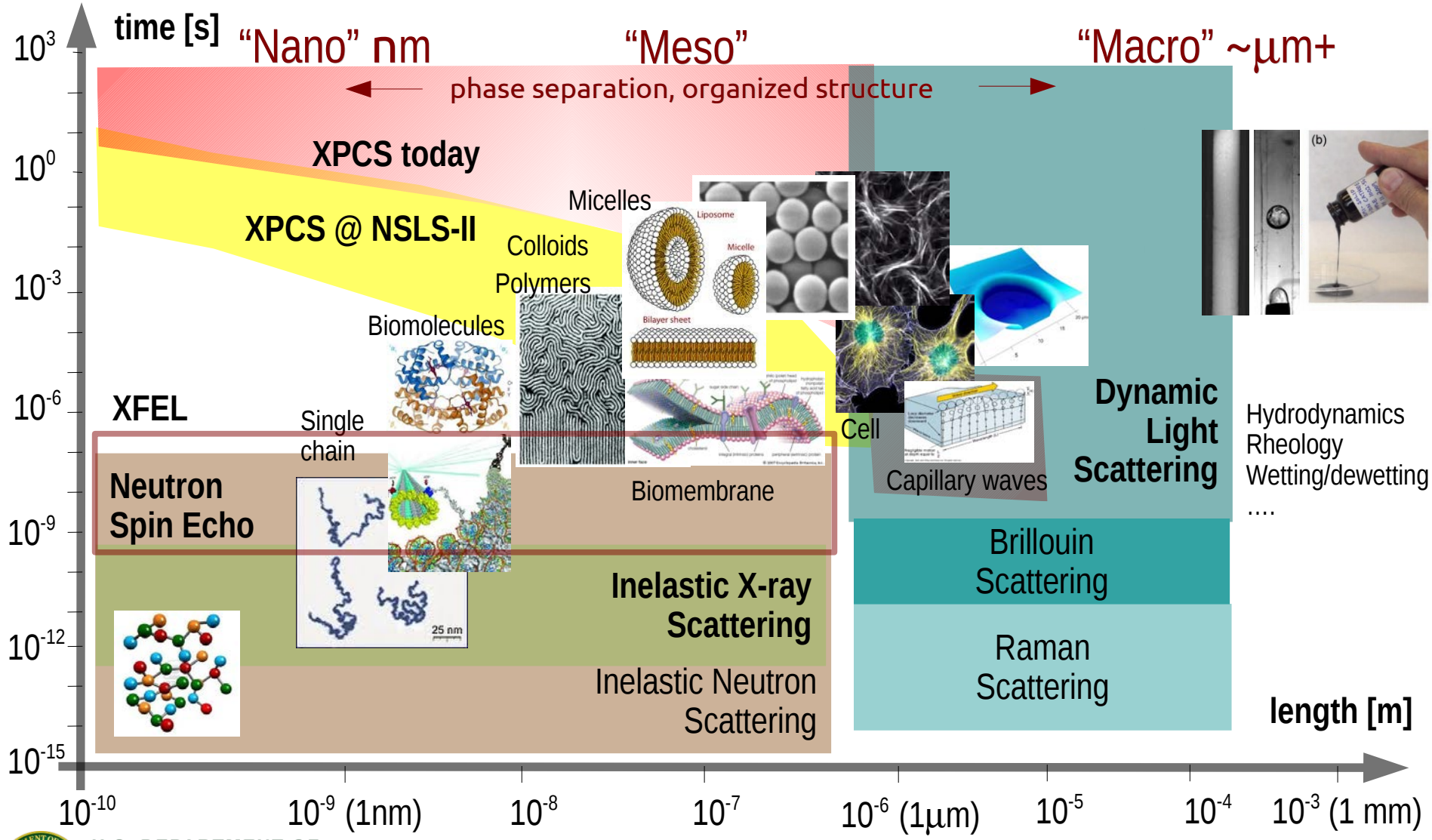
X-ray Photon Correlation Spectroscopy (XPCS)

- CHX: a beamline dedicated to measuring dynamics of materials by recording “speckle movies”





The future is Bright @ NSLS-II

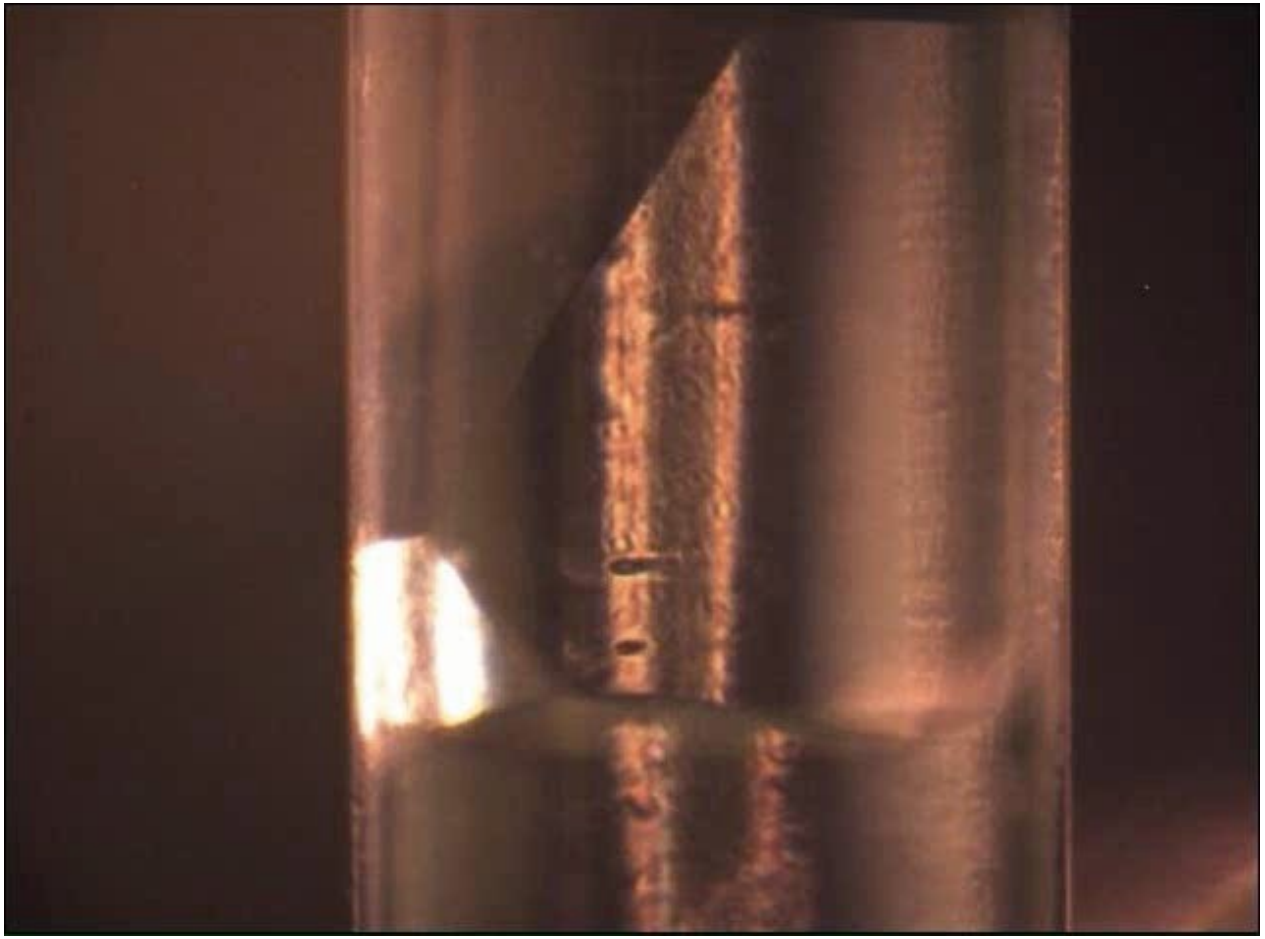


Function Require Motion!

- At CHX, we wish to measure complex collective dynamics of biological macromolecules and other bio- systems

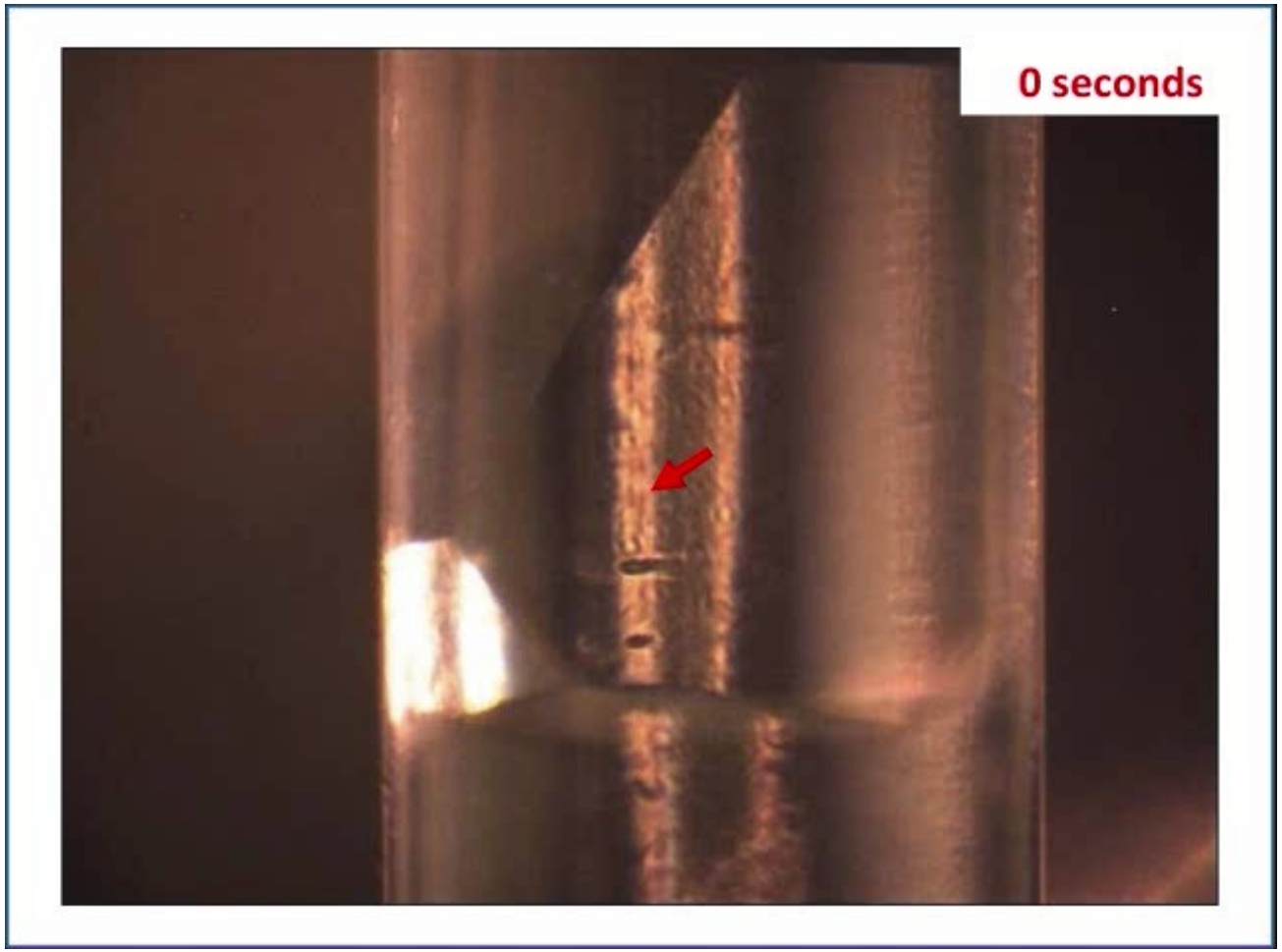
Houston we have a problem ...

- PXs @ room temperature



Houston we have a problem ...

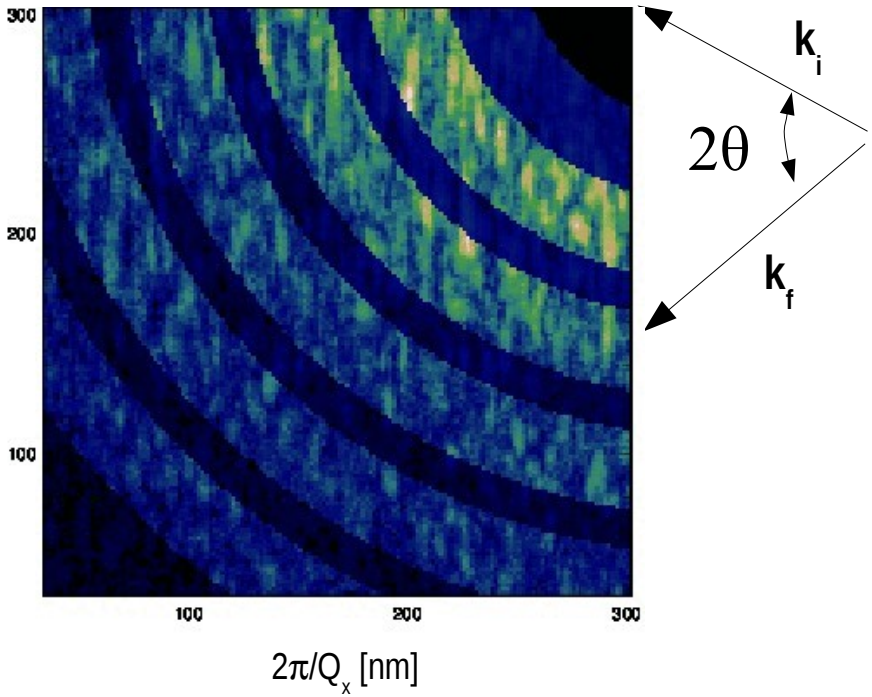
- PXs @ room temperature



Outline

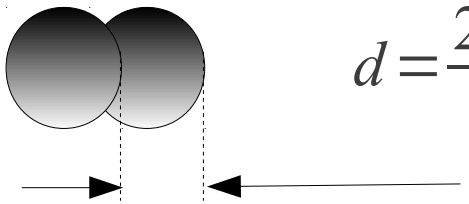
- Opportunities for *Early Science*
  Coherent Hard X-ray Beamline @ NSLS-II
- Brief intro to coherence – *X-ray Speckles*
  X-ray Photon Correlation Spectroscopy
  - Dynamics in "crowded" colloidal suspensions
- Mitigating beam damage
  - XPCS under flow
  - Speckle Visibility Spectroscopy
- X-ray speckles come to life: Proteins
- Outlook & Conclusions

Probing the Colloidal Dynamics with XPCS



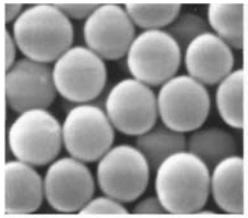
$$q = \frac{4\pi}{\lambda}\sin\left(\frac{2\theta}{2}\right)$$

- Measures time scale associated with displacement of colloids



$$d = \frac{2\pi}{q}$$

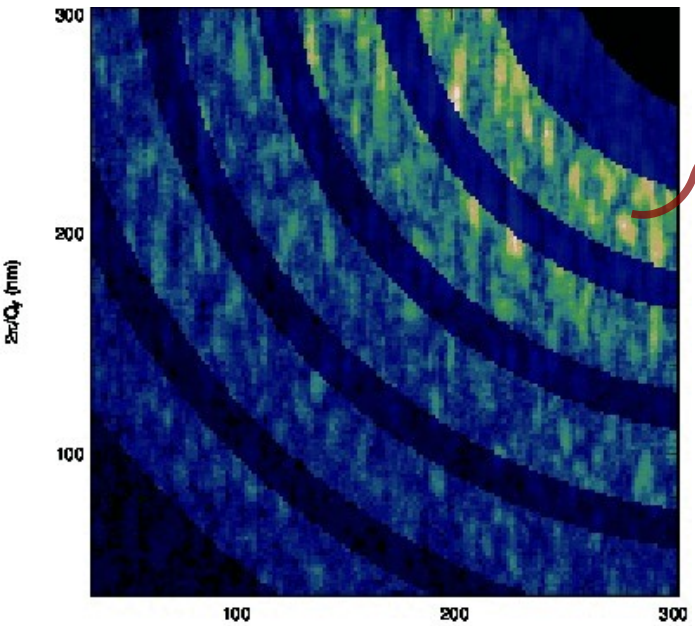
- i.e. measures dynamic structure factor *S(q,t)*
- By averaging over ~10^{11} particles



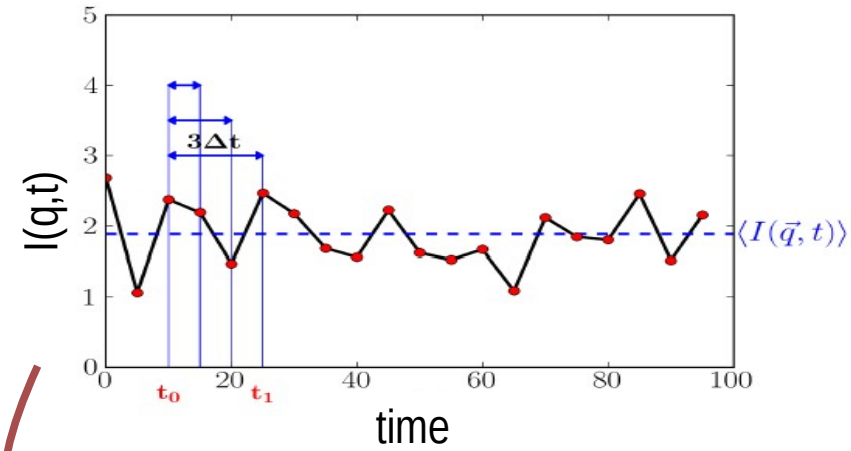
- For different *q* values

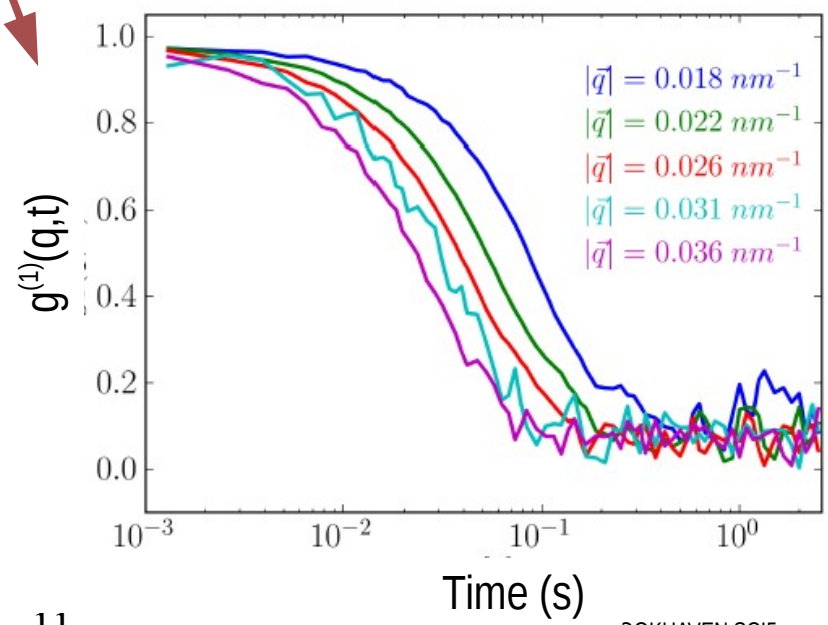
Probing the Colloidal Dynamics with XPCS

- Correlation functions



$2\pi/Q_x$ [nm]

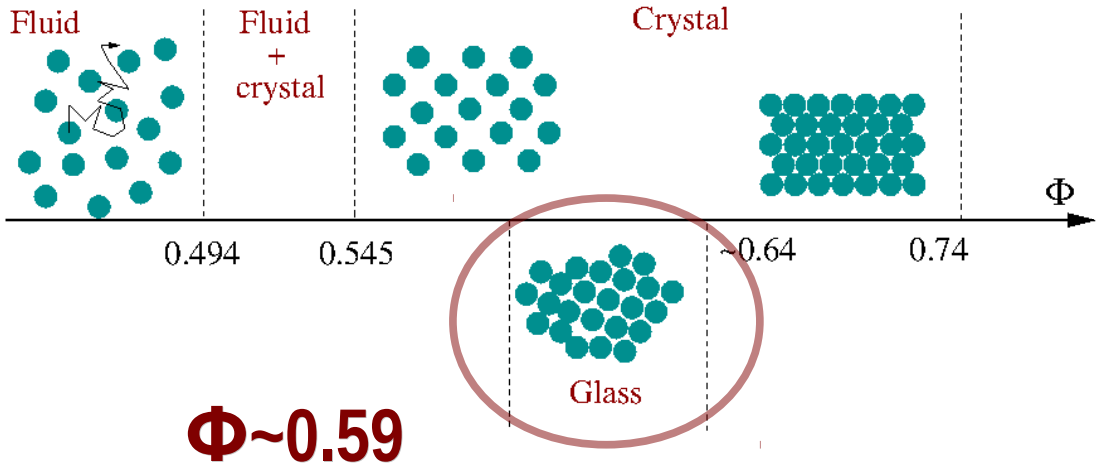


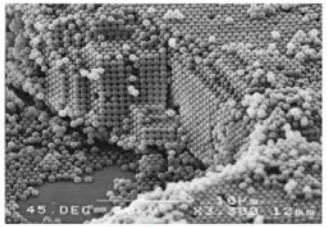


The Colloidal Glass Transition

- What happens here?

$$\Phi = \frac{N\,V_{colloid}}{V_{total}}$$

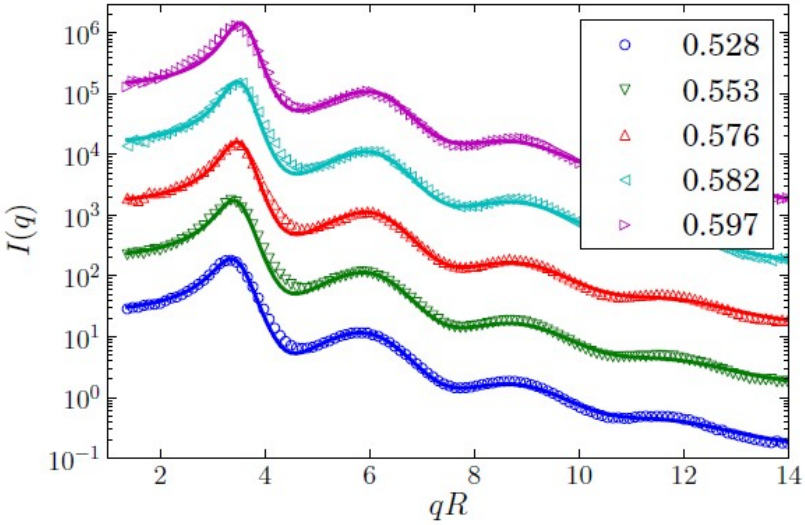




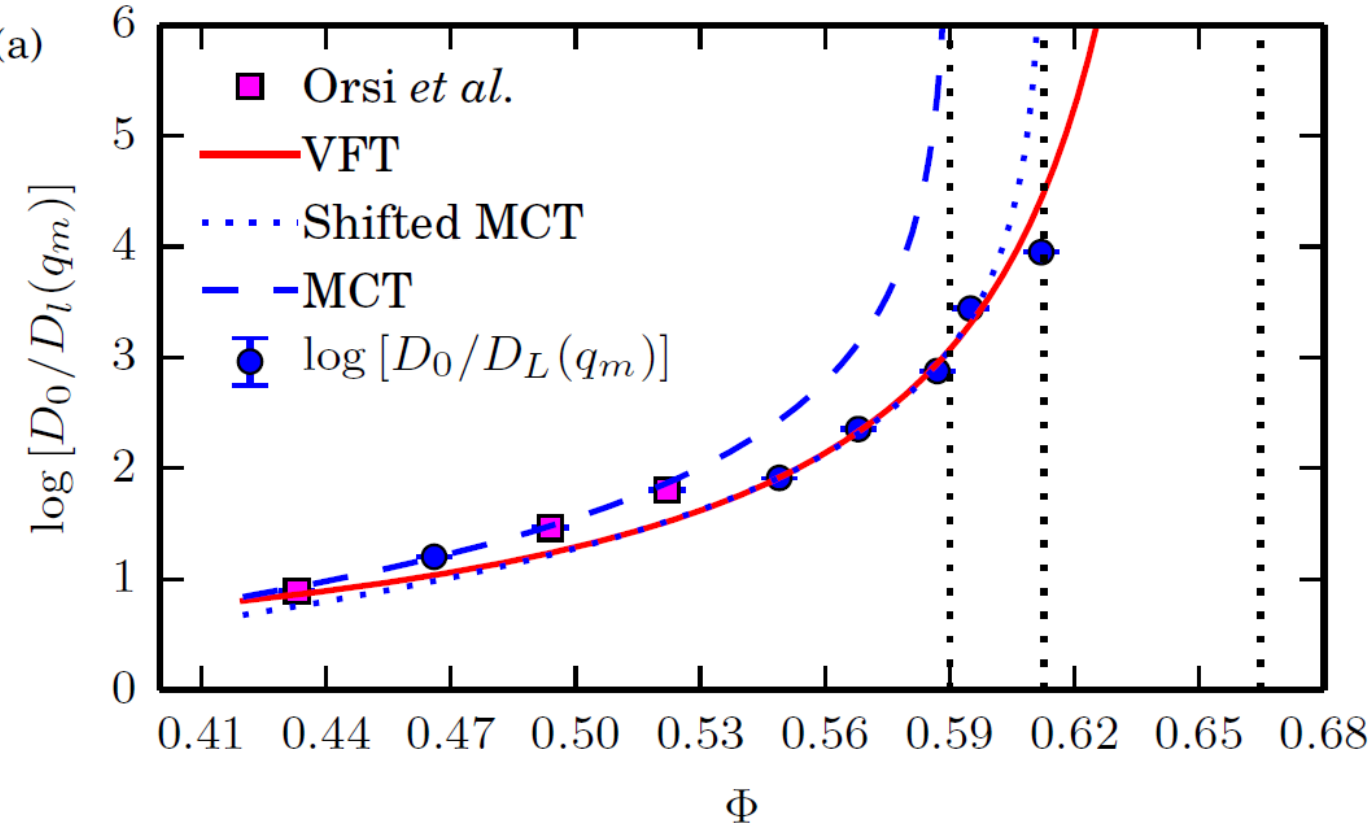
(b) R.M. Amos et al., PRE **61**, 2929 (2000)

Φ~0.59

- From SAXS / static scattering: pretty much *nothing* ...



Structural Relaxations near the Colloidal Glass Transition



MCT:

$$\frac{D_0}{D_l(q_m)} \propto \left|\frac{\Phi_g - \Phi}{\Phi_g}\right|^{-\gamma}$$

γ~2.58

VFT:

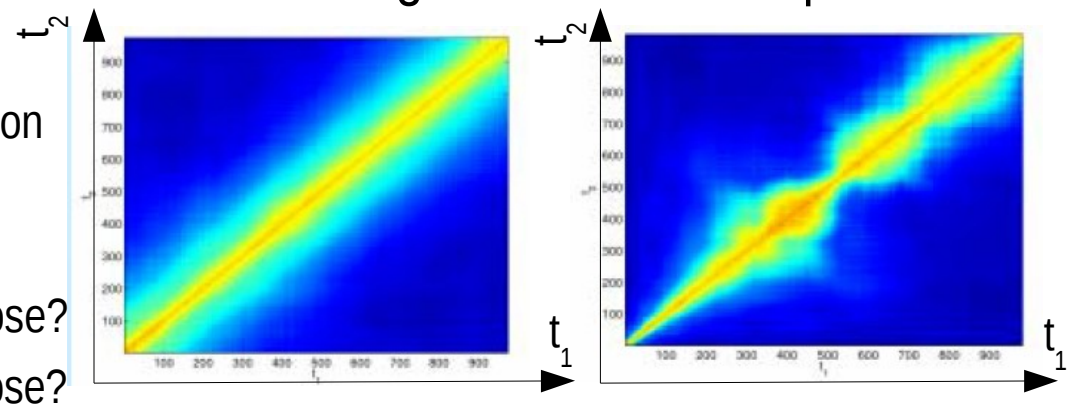
$$\frac{D_0}{D_l(q_m)} = \tau_\infty \exp\left[\frac{F}{(\Phi_0 - \Phi)^\delta}\right]$$

- relaxations follow an unexpected functional (VFT) form suggesting a kinetic arrest near the "random close packing concentration $\boldsymbol{\Phi_{RCP}}$***~0.67*** (~10% polydispersity)
- Suggests connection with ***Jamming***

P. Kwasniewski, AF, A. Madsen, *in review*; D. Orsi, AF *et al.* PRE 12
See also; Brambilla, Cipelletti *et al.*, *Phys. Rev. Lett.* 104, 169602 (2010)

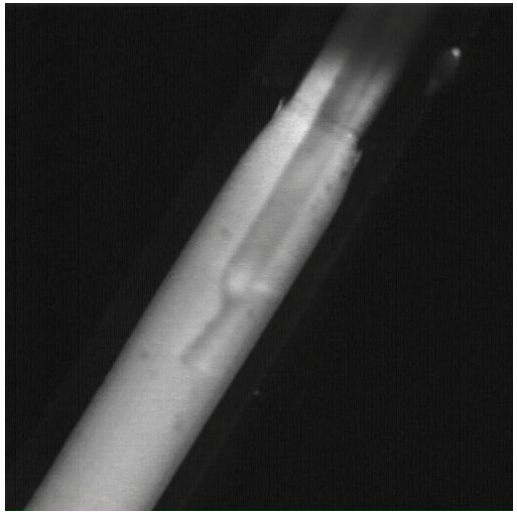
Beam Damage

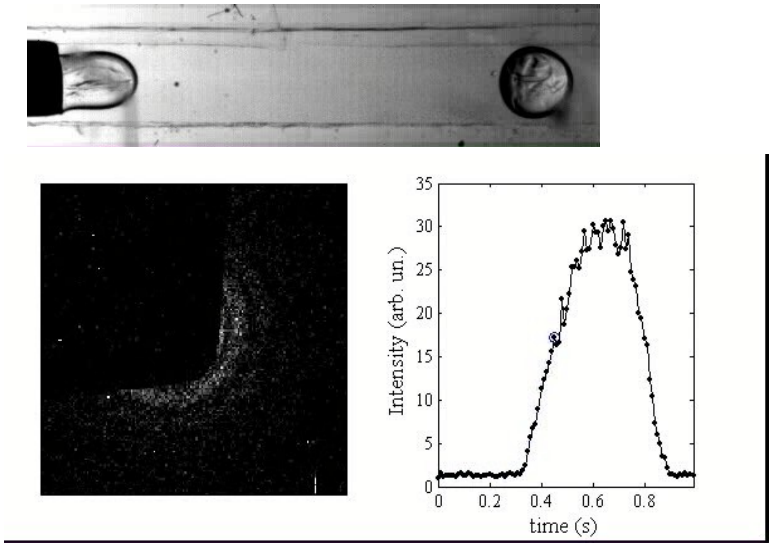
- Is the most common “bottle neck” with soft- and bio- materials
- We're certainly sensitive well below the Henderson limit because:
  - Room temperature
  - (Subtle) dynamical artefacts may preceed the beam-damage signatures in the static scattering
  - Limit: Dose < 1-10 MGy (?)
- To be able to avoid beam damage we need to recognize it in the first place
  - “Self-consistency checks”
  - Is there a q dependence on the relaxation rates?
  - Does the q dependence make sense?
  - Is the static scattering changing with dose?
  - Are the relaxation times changing w. dose?
  - Any “unexpected” dynamical heterogeneities developing with time?
  - ….



Mitigating Beam Damage

- Use 2D detectors, choose the 'right' energy, limit unnecessary dose – e.g. use a (fast) shutter before the sample (!), etc.
- Use better sample handling techniques – e.g. XPCS under continuous flow

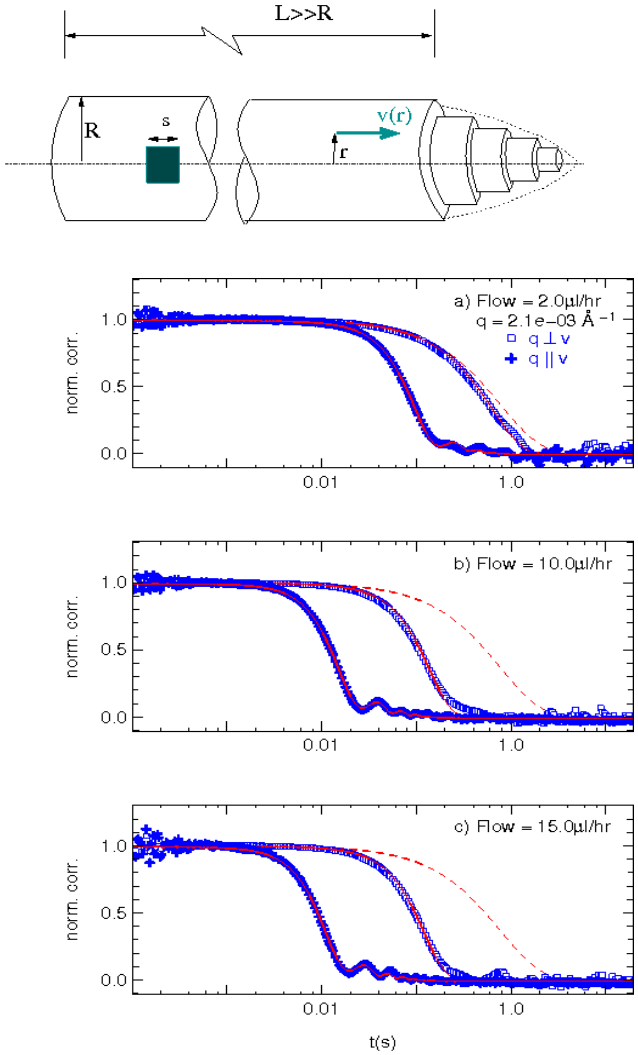




F. Destremaut, J.B. Salmon, AF et al., unpublished

- Change experimental strategy if required – speckle visibility spectroscopy

XPCS under continuous shear flow



The importance of scale[1]

Diffusion - $\Gamma = Dq^2$

Shear rate: $\dot{\gamma} = -\frac{dv(r)}{dr}$ (T^{-1})

$\mathrm{Wi} = \dot{\gamma}/\Gamma \ll 1$

Transit time effects - $\Gamma_{tr} = \frac{v_{flow}}{s}$

(s - transverse beam size)

Shear-induced effects $\Gamma_s = \mathbf{v}\cdot\mathbf{q}$

Péclet number:

$\mathrm{Pe} = \frac{\Gamma_s}{D} \gg 1$!!!!

Shear number:

$\mathrm{S} = \frac{\mathbf{q}\cdot\mathbf{v}}{Dq^2} \gg 1$!!!!

Shear-induced correlation:

$G_{shear}(q,t) = \int\int \cos[(\mathbf{q}\cdot\delta\mathbf{v})t]dr_1 dr_2$

XPCS: Can measure, and study the interplay between, *dissipative dynamics* and the *advective response* to flow/shear

[1]Note: assuming *homodyne* detection
heterodyne detection – M. Sutton *et al.*

AF et al., J. Synchrotron Rad. (2008)S. Busch, AF *et al.*, *Eur. Phys. J. E.* **26**, 55 (2008)
AF *et al.*, *New Journal of Phys.* **12**, 035023 (2010)
G.G.Fuller *et al.*, *J. Fluid Mech.* **100**, 555 (1980)

Speckle Visibility Spectroscopy

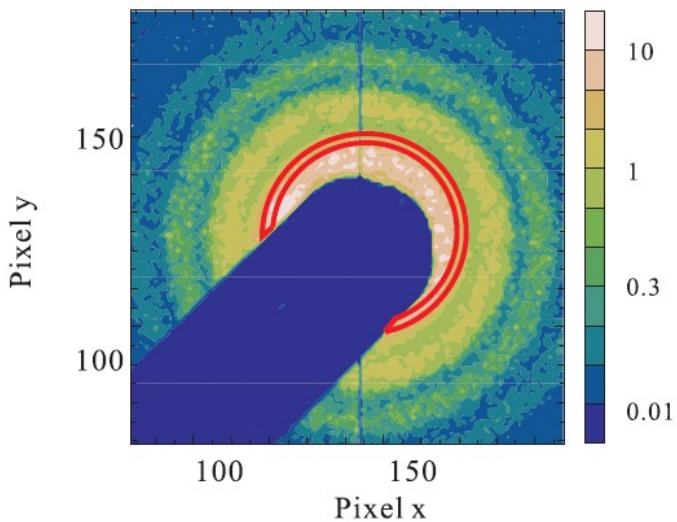
- Main idea (D.J. Durian et al., Phys. Rev. Lett. 2003, Rev. Sci. Inst 2005):
  speckle contrast are blured by internal motions if the integration time is increased
- Speckle contrast is determined by how coherne tis the beam (M - # of coherent modes) but also affected by Poisson statistics in the detection system

$$v_2 = \frac{1}{M} + \frac{1}{\langle K \rangle}$$

- In the low scattering limit (i.e. always with X-rays :) this is given by the Negative binomial (or 'Poison-Gamma') distribution:

$$P(K, M) = \frac{\Gamma(K+M)}{\Gamma(K+1)\Gamma(M)} \left(\frac{\bar{K}}{\bar{K}+M}\right)^K \left(\frac{M}{\bar{K}+M}\right)^M$$

- i.e. we can measure the speckle contrast by measuring the probablity of recording 0,1,2,3... photons in a pixel



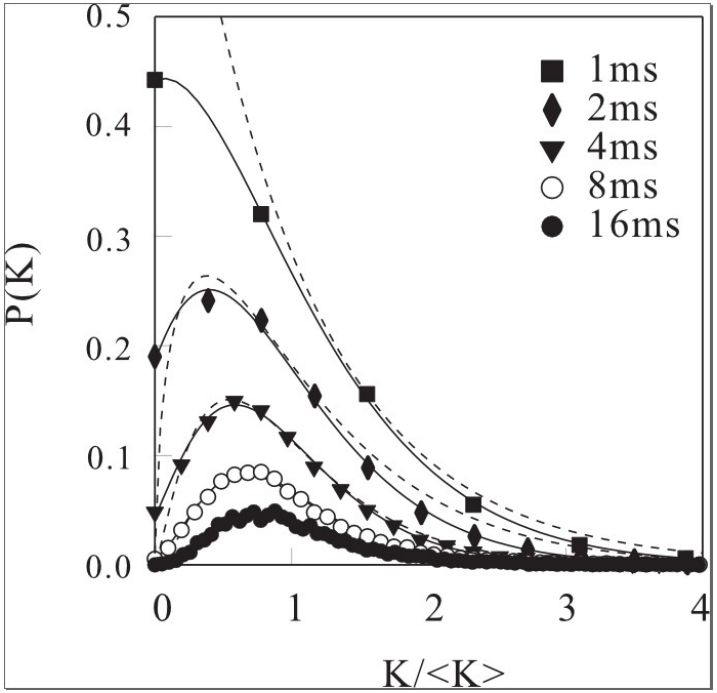
Texp	M	<K>
4ms	2.66	1.55
8ms	3.45	3.10
16ms	4.08	5.76
64ms	4.49	13.02

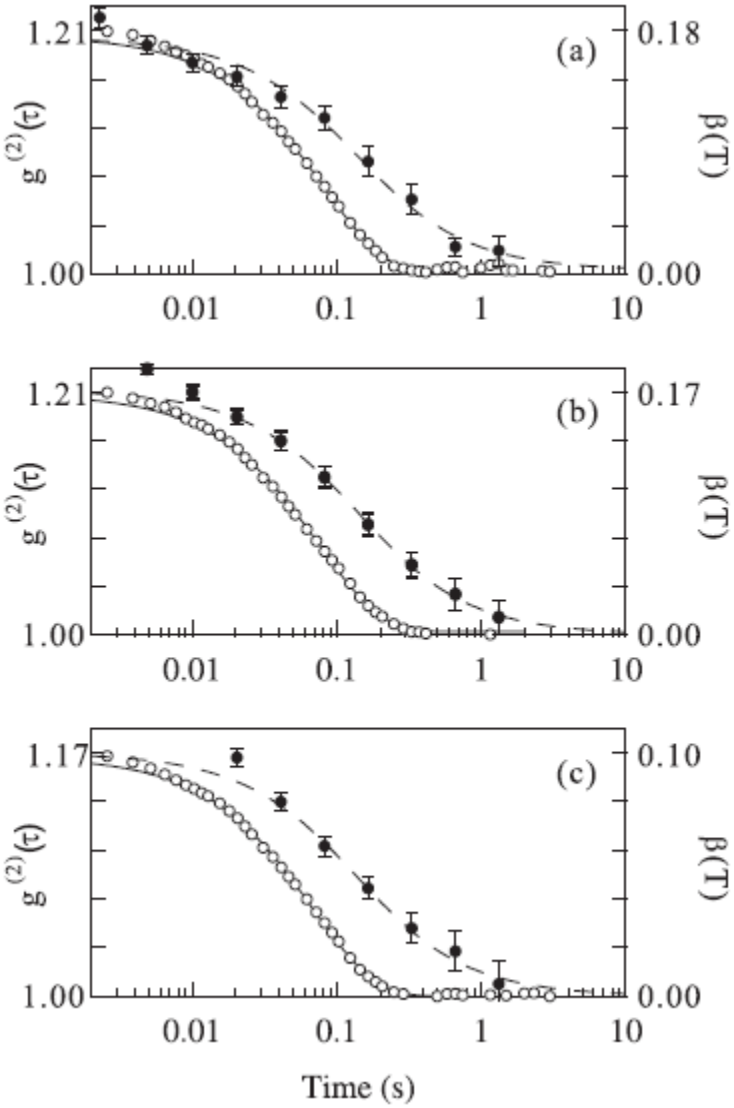
L. Li, AF *et al. submitted*

S. Hruszkewycz *et al. Phys. Rev. Lett. 2012*

Speckle Visibility Spectroscopy

- Photon statistics (NBD) → speckle contrast (visibility)
- Visibility decay with integration time → sample dynamics

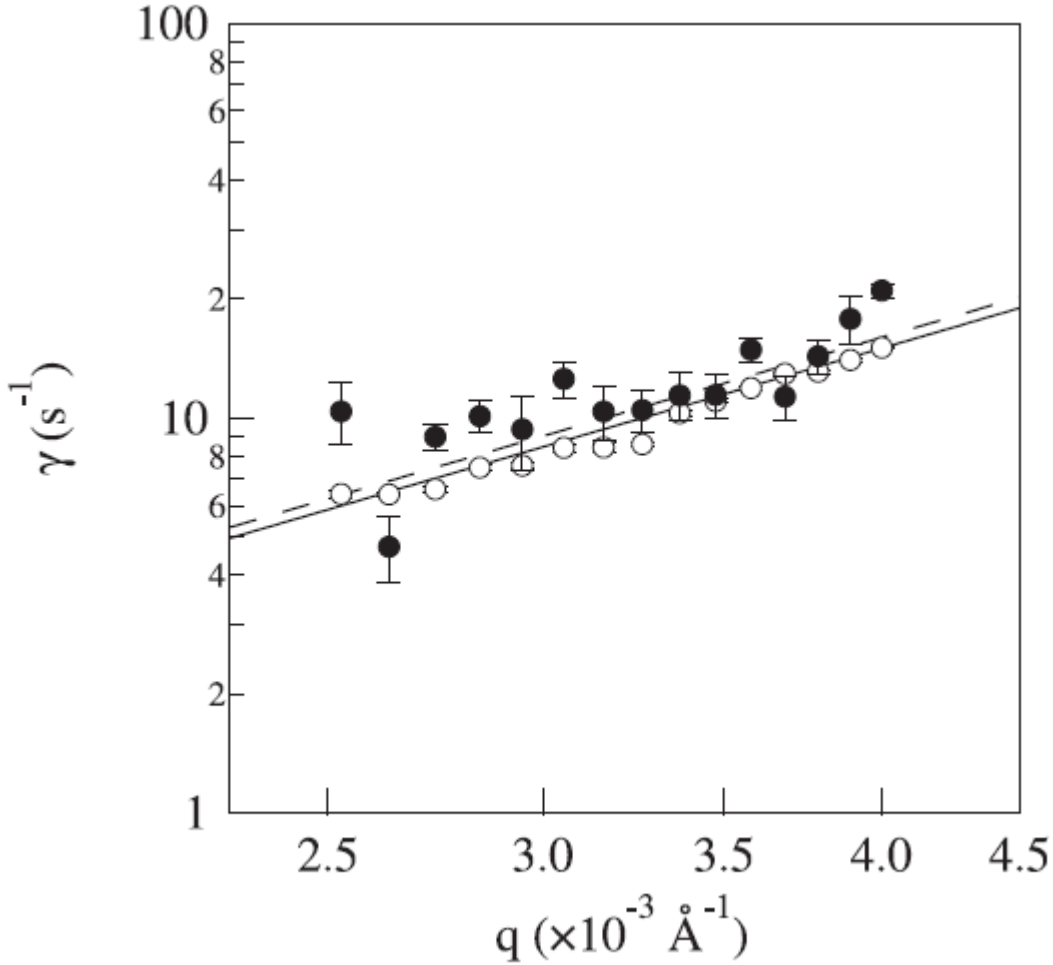




L. Li, AF *et al. submitted*

Speckle Visibility Spectroscopy

- Relaxation rate ~q^2
  → free Brownian diffusion
- SNR in XSVS can be increas by repeating the experiment (requires an ensemble of nominally identical samples .
- Other major advantage of XSVS: can measure dynamic on time scales faster than the rep rate of the detector (one need to control the exp time)



L. Li, AF *et al. submitted*

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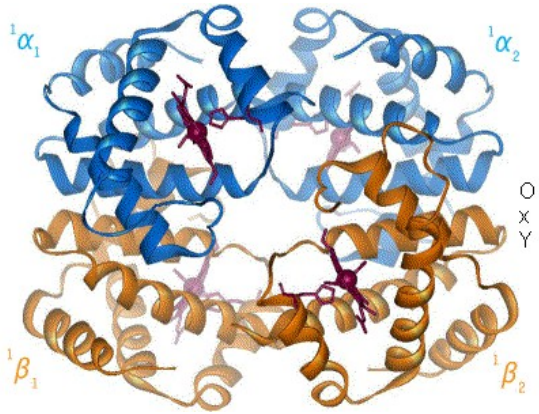
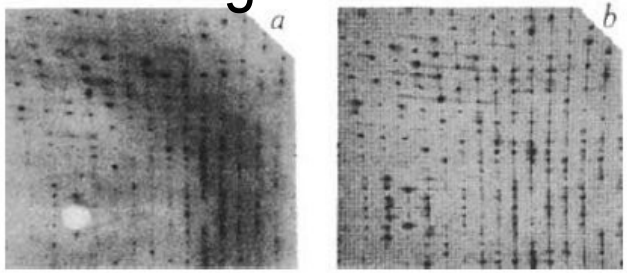
BROOKHAVEN SCIENCE ASSOCIATES

Collective Dynamics of Proteins

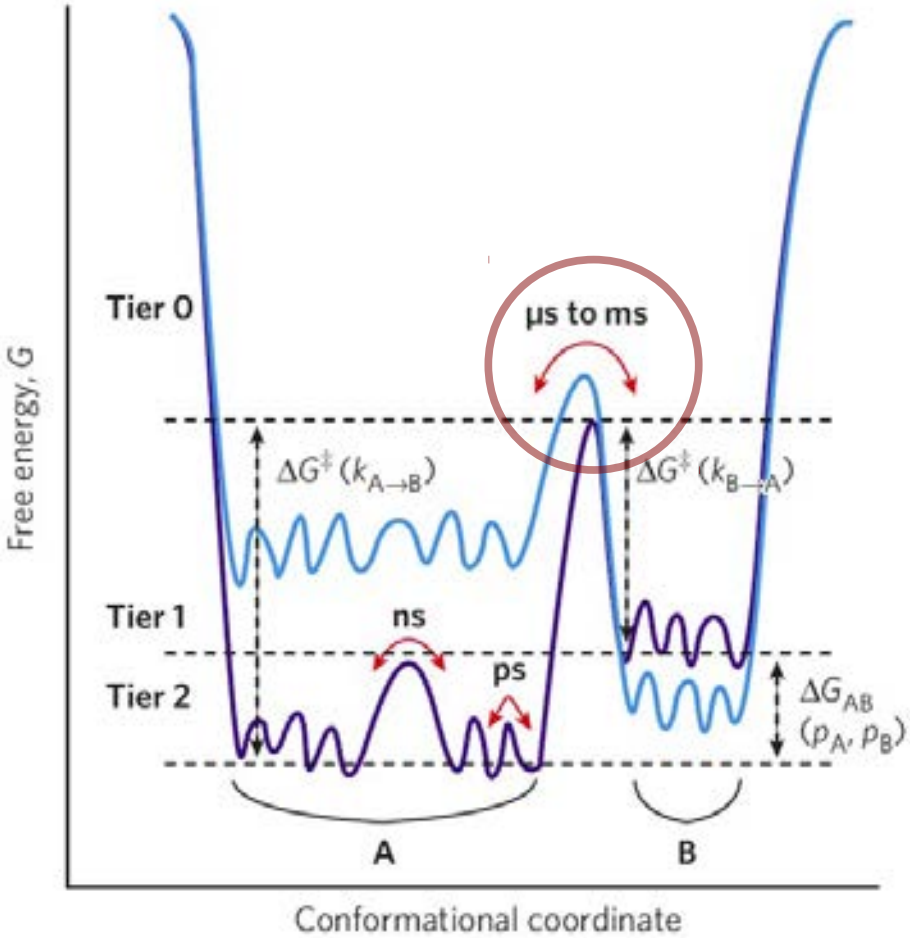
- Proteins - “Glassy Systems”
- Actions are usually described in terms of static structure, but *function requires motion*.



- Diffuse Scattering



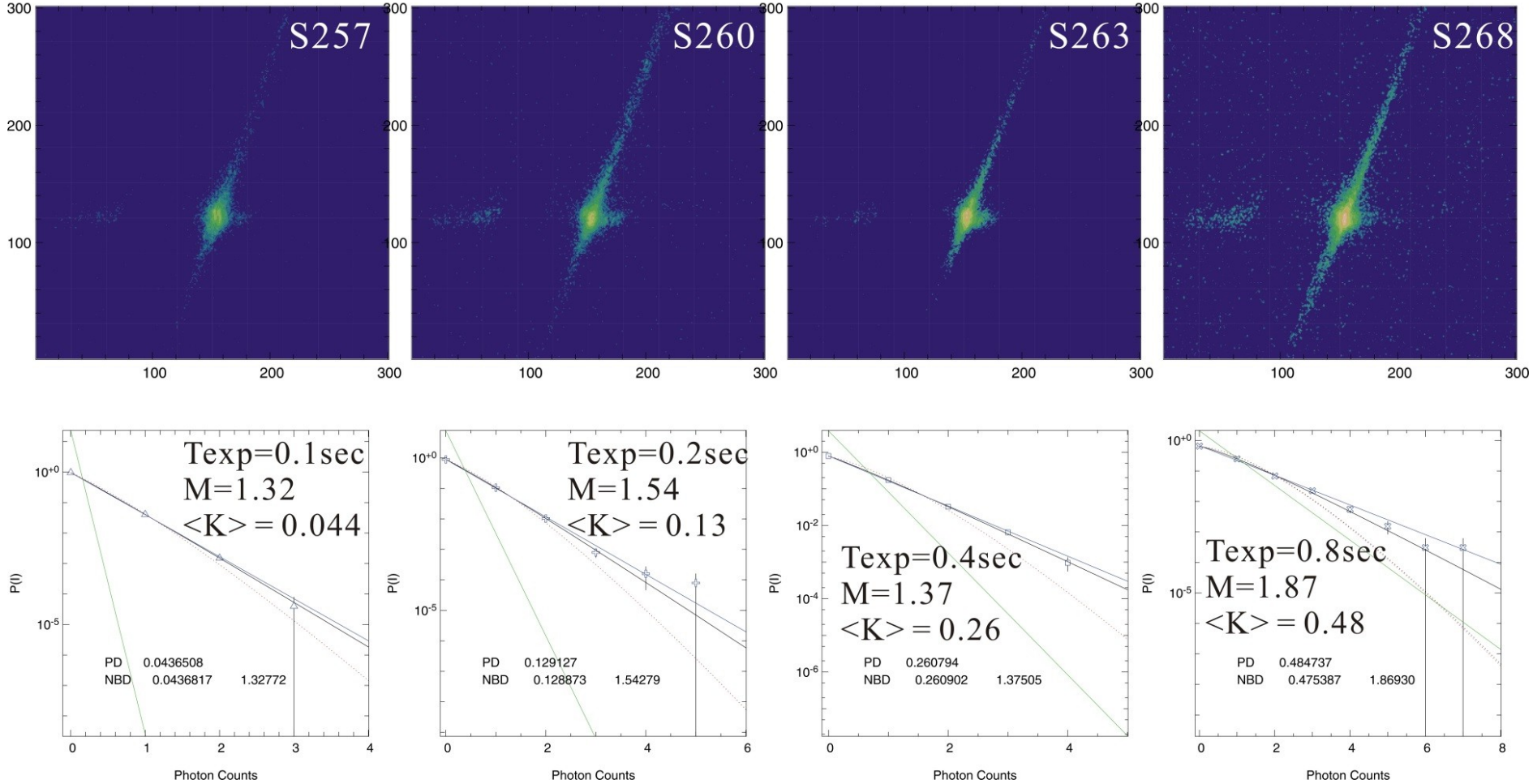
Doucet & Benoit, Nature 325, 643 (1987).



H. Frauenfelder *et al.*

X-Ray Speckle Come to Life

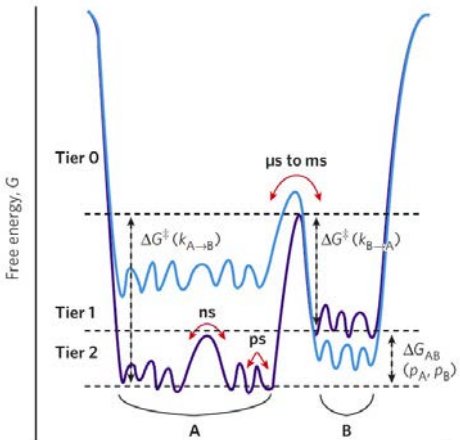
- Speckles from Concanavalin A (Con A) from Jack Bean

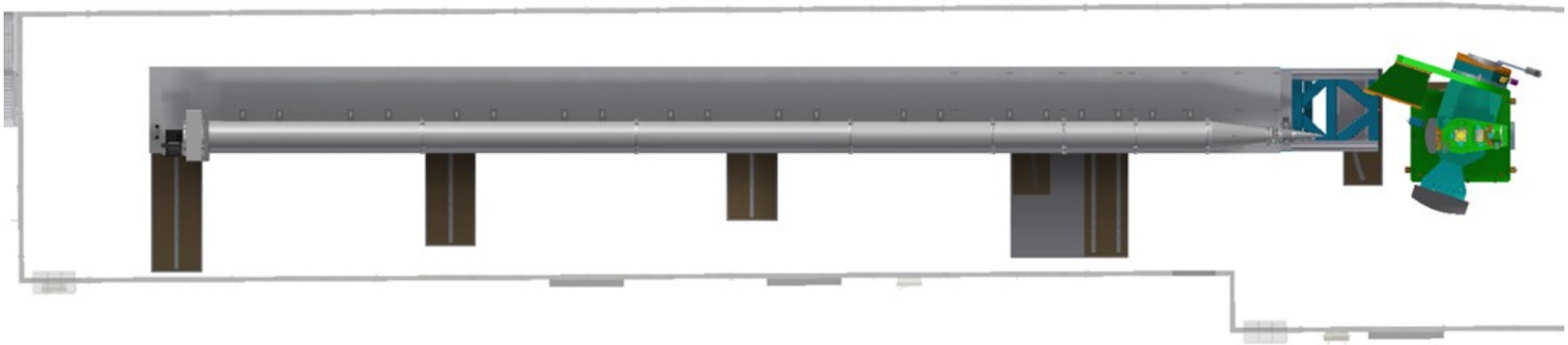


L. Li et al, in preparation.

Instead of Conclusions: a wish-list

- NSLS-II
  - Here we showed that we can detect static speckles (e.g. along a truncation rod) without beam damage for ~0.5 s (probably <1MGy)
  - Even with a factor of 100 in increase dose, the same experiment shouldbe feasible in ~10-100 ms at NSLS-II
  - This opens the possibility of measuring low-tier collective modes in various bio-materials
  - CHX is uniquely adapted for this type of experiments!





Acknowledgements

Luxi Li *(NSLS-II) – loking for a job*

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Anders Madsen (European XFEL)

CHX
Lutz Wiegart, M. Carlucci-Dayton, D. Chabot,
J. Sullivan, K. Roy

Beamlines
ID 10 ESRF - Y. Chushkin
34-ID APS - R. Harder
8-ID APS - A. Sandy, S. Narayanan

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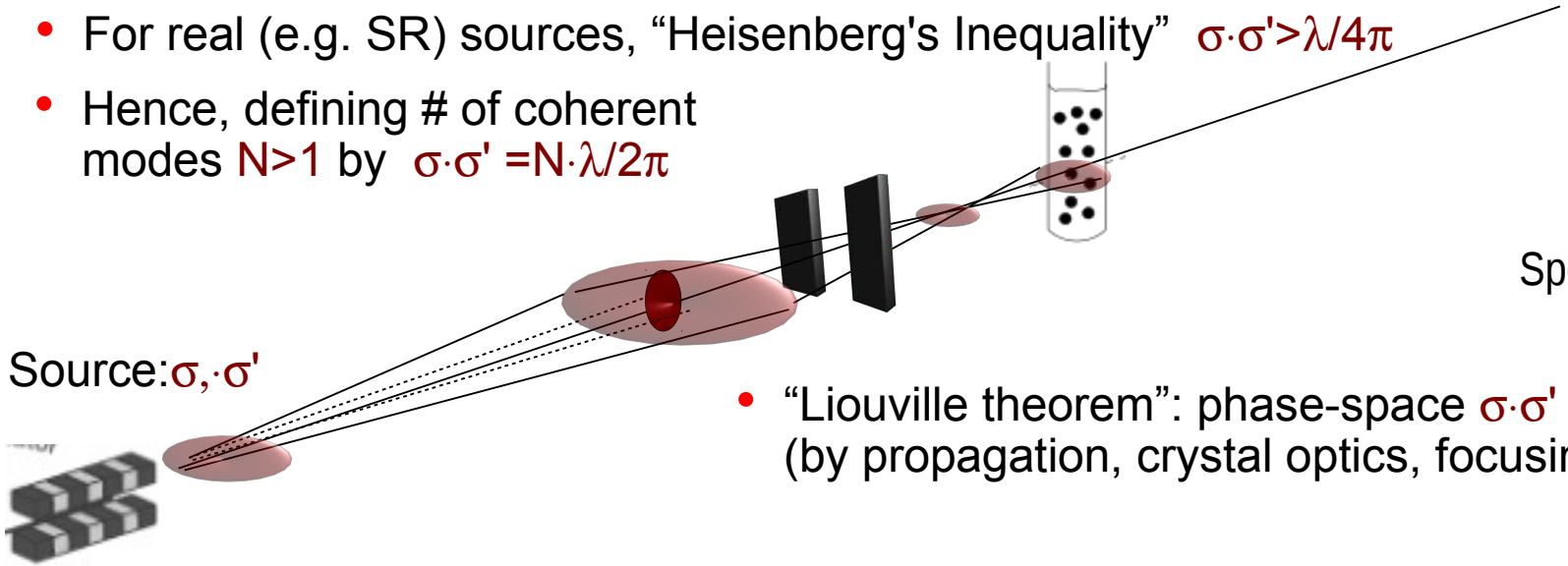
NSLS-II project: DOE# E-AC02-98CH10886
BNL LDRD 11-025

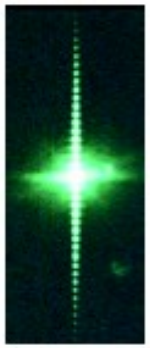
Coherence with a chaotic SR Source

- Degree of coherence given by "phase-space" volume $\sigma \cdot \sigma'$
- "Ideal" coherent (point) source $\sigma \cdot \sigma' \sim \lambda/2\pi$
- For real (e.g. SR) sources, "Heisenberg's Inequality" $\sigma \cdot \sigma' > \lambda/4\pi$
- Hence, defining # of coherent modes $N>1$ by $\sigma \cdot \sigma' = N \cdot \lambda/2\pi$



- "Liouville theorem": phase-space $\sigma \cdot \sigma'$ is conserved (by propagation, crystal optics, focusing, etc)

- The phase-space volume can never be increased, but can be reduced by e.g. closing a "beam-defining" slit – i.e. increasing coherence at the expense of flux
- Transverse coherence length $\rho = \cdot \lambda L/2\pi\sigma$
  see also complex coherent factor,
  Van Cittert-Zernike theorem (Van Cittert 1934, Zernike 1938)
- Also – Longitudinal Coherence length $\sim \Delta\lambda/\lambda$
  (= monochromaticity)

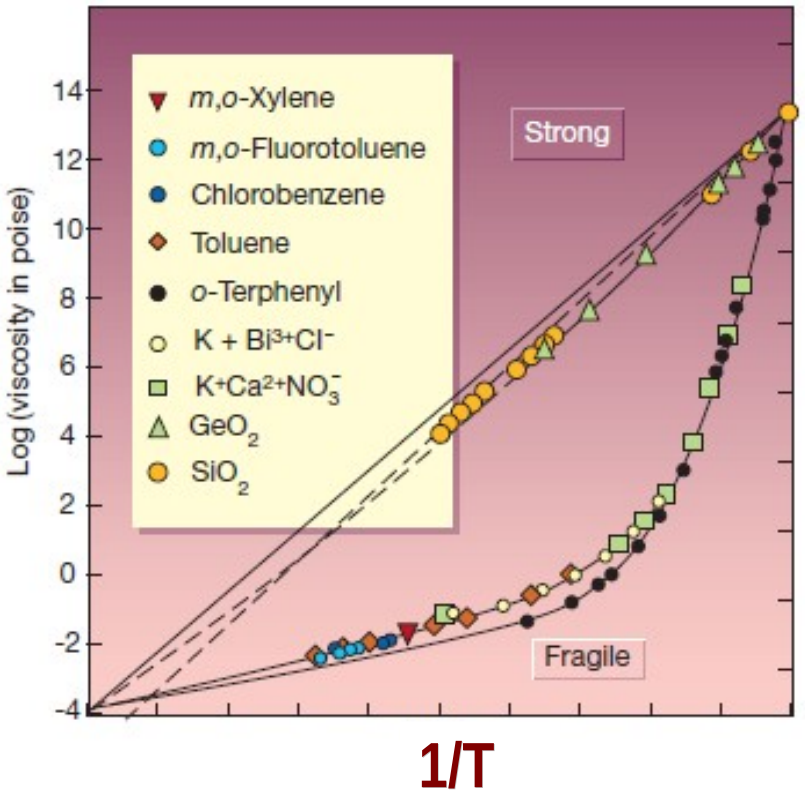


Young double-slit experiment with SR X-rays

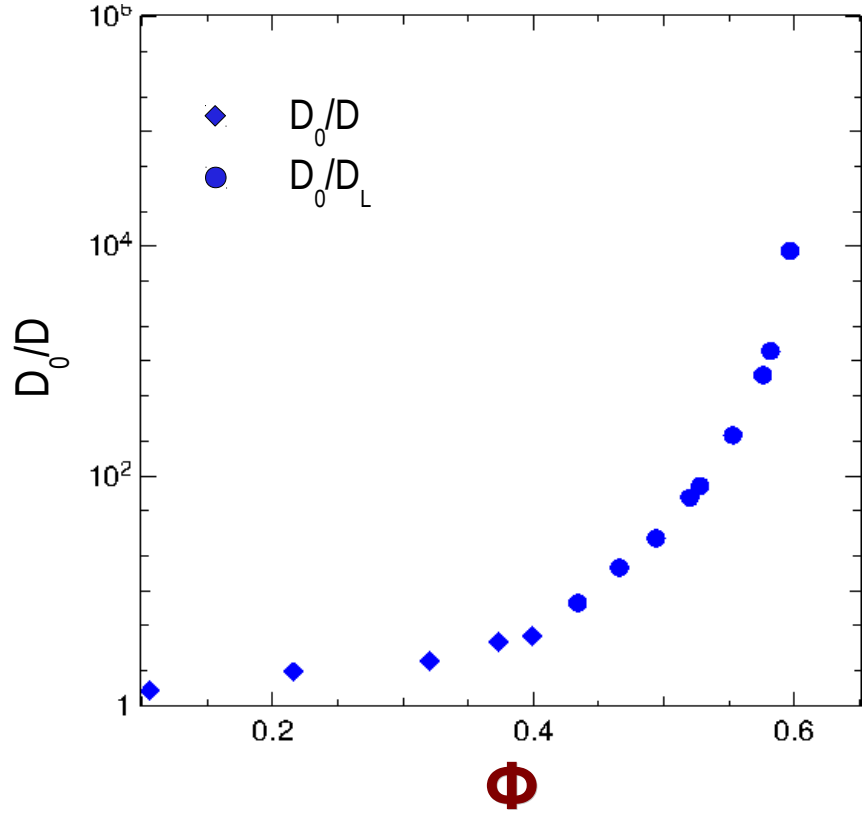
W. Leitenberger *et al.*, *Physica B* **336**, 36 (2003)

Supercooled Liquids vs. Hard Sphere Colloids

- In addition to being interesting/useful in their own right, colloids are an excellent model system for supercooled liquids and molecular glassformers



Denenedetti, Stillinger, *Nature* 2001



D. Orsi, AF et al. *Phys. Rev. E* 2012
P. Kwasniewski, AF, A. Madsen, in preparation 2013
$\eta/\eta_0 \rightarrow D_0/D_L$ (Segre *et al.*, *Phys. Rev. Lett* 2001)