XPCS at a DLSR

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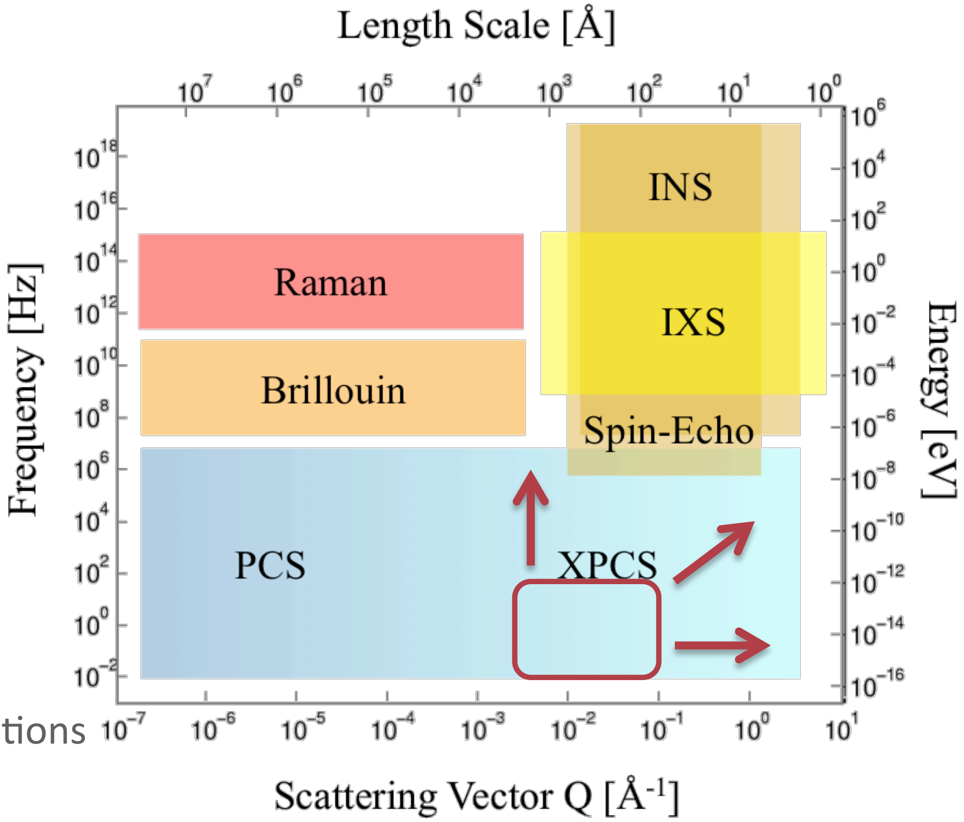
Outline

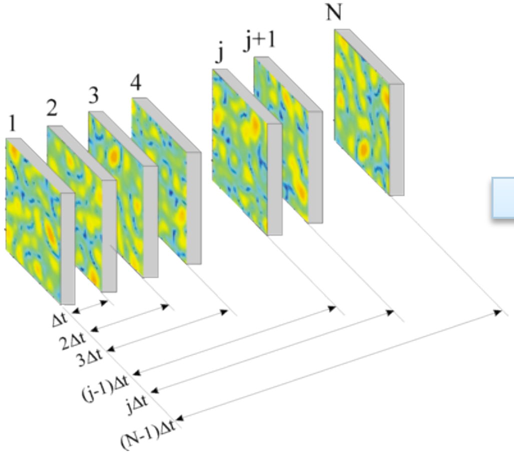
- XPCS Considerations
- XPCS at Storage Rings
- XPCS Detectors
- Science Opportunities
- Conclusions

XPCS Considerations

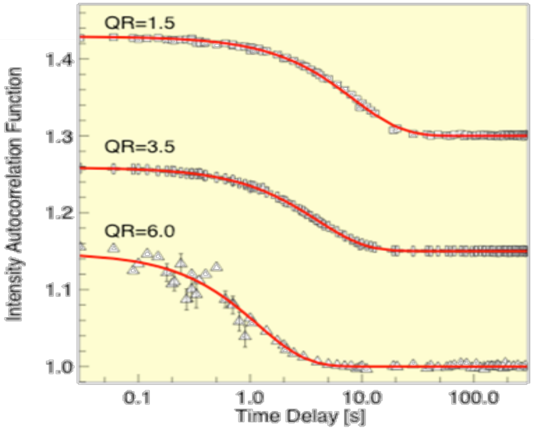
- XPCS at storage rings today is unique probe of relatively slow dynamics at the nanoscale

- Time autocorrelation function, $g_2(\tau)$, is typical quantity of interest
  - Visibility, higher-order correlation functions also interesting





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



XPCS Considerations

- Coherent flux: $F_c = B(\lambda/2)^2$
- Signal-to-noise (SNR) ratio of $g_2(\tau)$ guides improvements and enables new science

$$SNR = Contrast \times Intensity \times \sqrt{T\tau N}$$

- Contrast is speckle contrast or speckle visibility (0 → 1)
- (Coherent) Intensity ≈ Source Brilliance
- T = measurement duration (often limited by sample damage and/or long-time stability of measurement apparatus)
- τ = delay time ≈ (detector acquisition or frame rate)$^{-1}$
- N = number of detectors (pixels)

Fast 2-D detectors are the way to go!

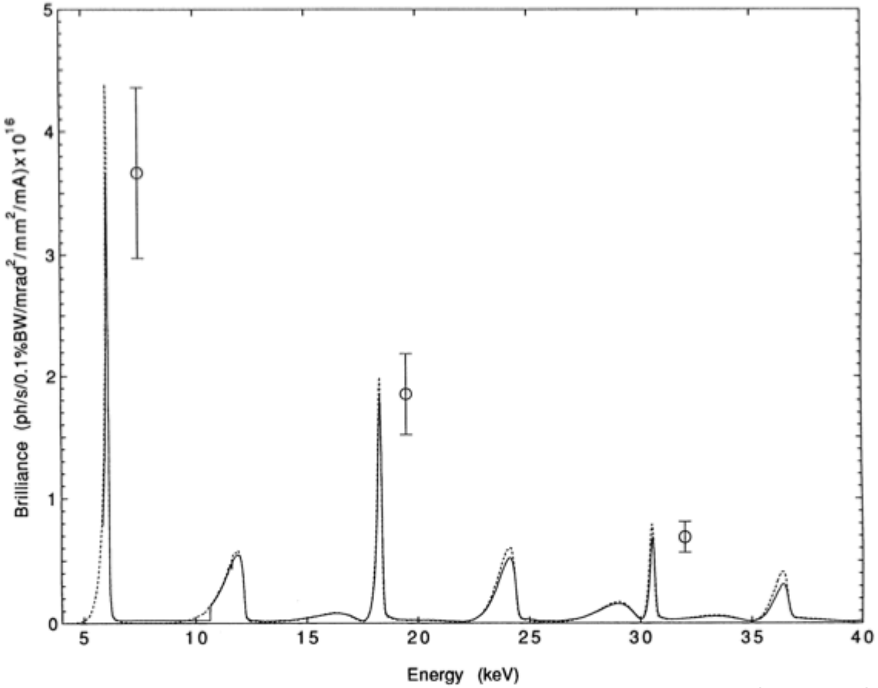
Accessible delay times go like the square of the coherent intensity!

XPCS at Storage Rings

- Modest changes in storage rings, beamline design and detectors have produced a remarkable increase in XPCS capabilities and science over the past 15-20 years
  - Time scales from 10-100’s of seconds to 2-10 ms with 10X more brilliance
  - Quantitative measures of unusual nanoscale dynamics

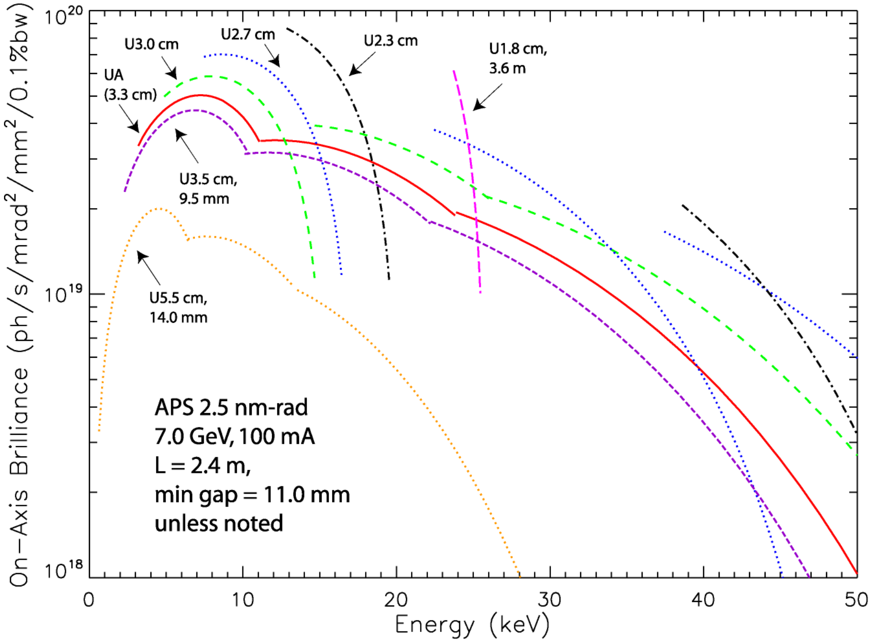
APS – Undulator A – circa 1996

10^{18}



Z. Cai et al., Rev. Sci. Instrum. **67**, 3348 (1996)

APS – Undulator A – circa 2010

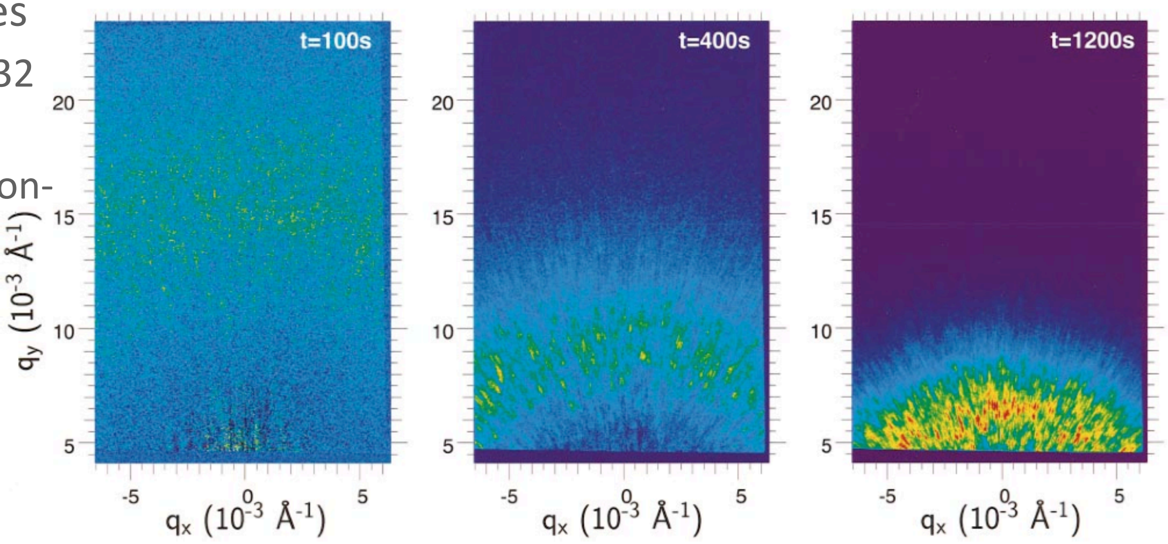


APS 2.5 nm-rad
7.0 GeV, 100 mA
L = 2.4 m,
min gap = 11.0 mm
unless noted

XPCS at Storage Rings

1998

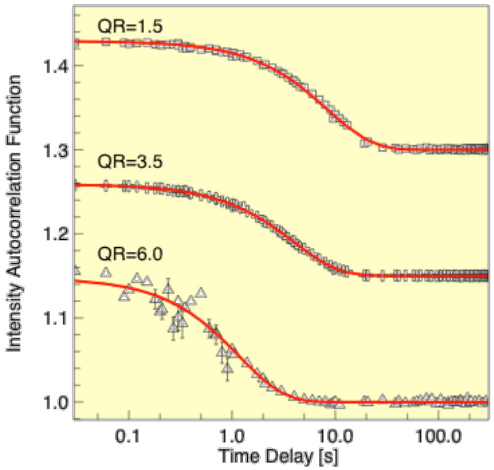
- Domain coarsening in glasses
  - [A. Malik et al., PRL 81, 5832 (1998)]
  - Dynamics measured in a non-equilibrium system
  - Time scales: > 50-100 s



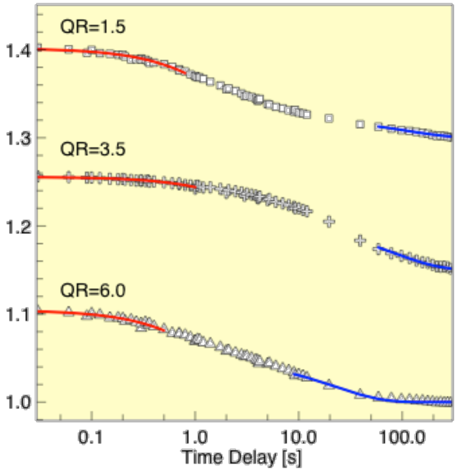
2000

- Hydrodynamic corrections to diffusion in concentrated hard-sphere diffusion
  - L. B. Lurio et al., PRL 84, 785
  - Time scales > 50 ms

Dilute



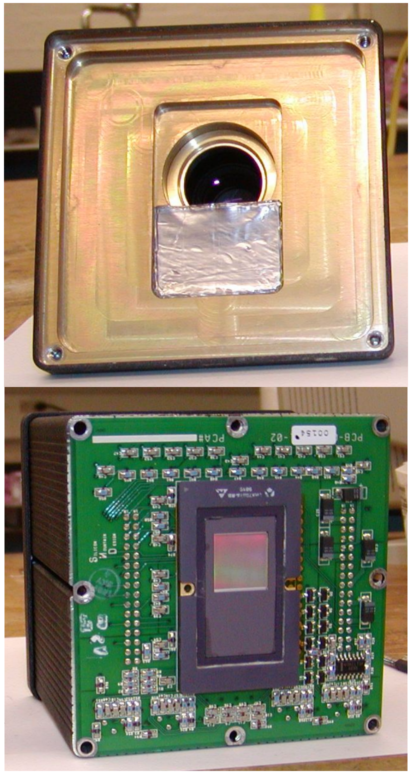
Concentrated

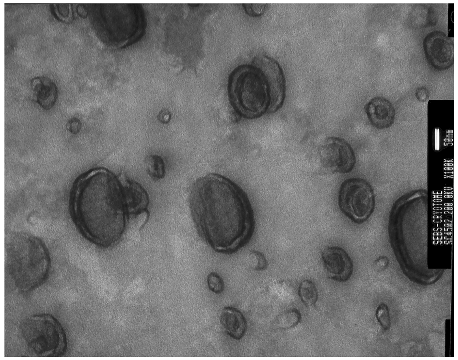


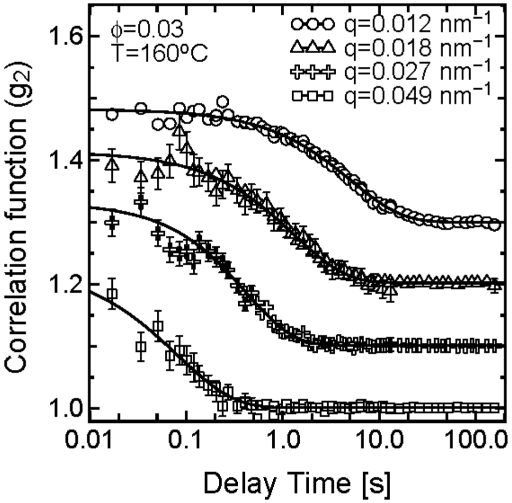
XPCS at Storage Rings

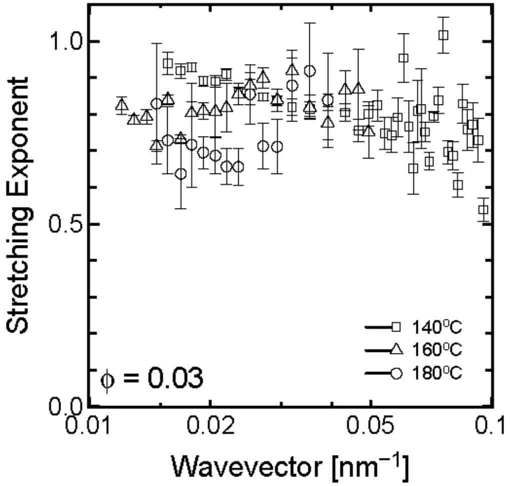
2005

- Dynamics of block copolymer vesicles
  - Membrane fluctuations in a tri-block homopolymer mixture
    - Rapidly fluctuating speckle patterns
    - Correlation functions of sufficient quality to see stretched (2/3) exponential decays
  - Home-built SMD/Dalsa fast direct-detection detector
  - [P. Falus et al., PRL **94**, 016105 (2005) and P. Falus et al., RSI **75**, 4383 (2004)]
  - Time scales: > 10 ms



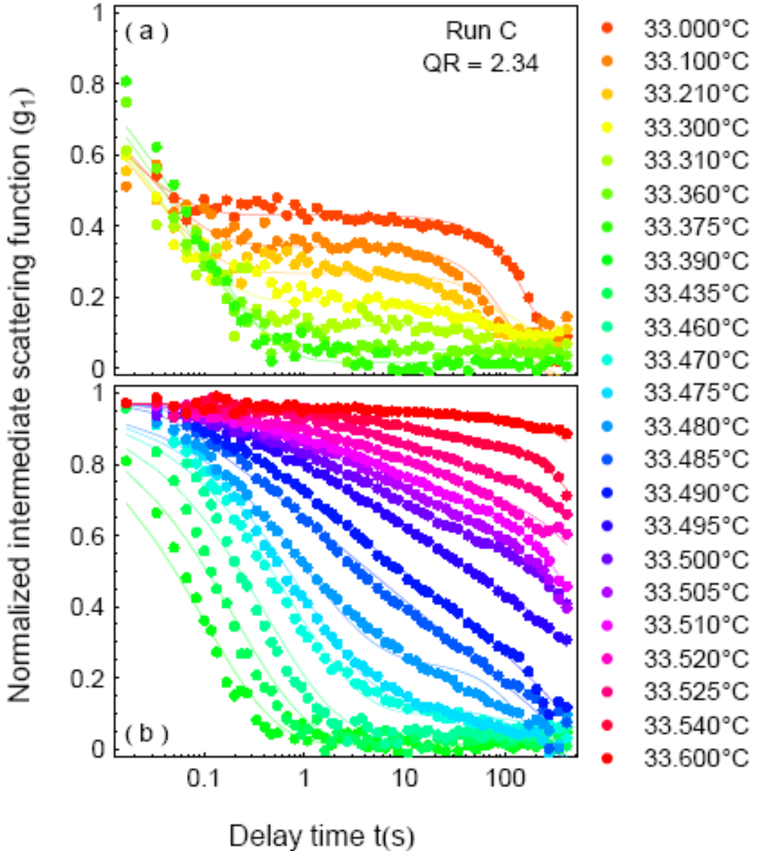


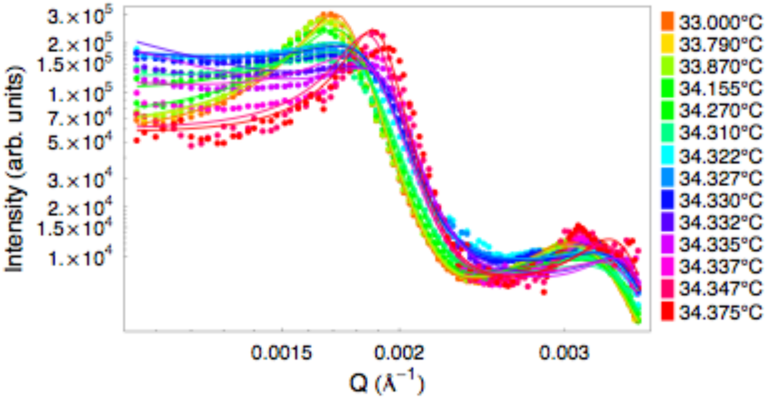




XPCS at Storage Rings

~ 2008





Dynamics in a re-entrant glass
X. Lu et al., PRL **100**, 045701

Time scales > 2 ms
Unusual dynamics over many decades in delay time

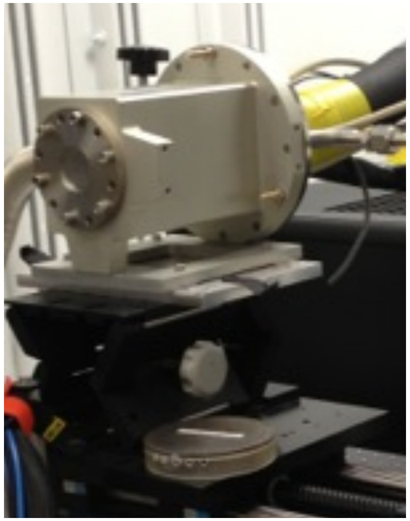
- Modest changes in storage rings and beamline design and somewhat larger changes in detectors have produced a remarkable increase in XPCS capabilities and science over the past 15-20 years

XPCS at Storage Rings

- Recap
  - Increased time-averaged brightness from storage rings has (and will be!) leveraged
    - Accessible delay times at the APS have decreased by more than 3 orders of magnitude with 10X increase in brightness, heroic but limited detector efforts and very modest improvements in beamline design
- 2-D detector development must parallel DLSR developments!

XPCS Detectors for a DLSR

- Encouraging recent advances but considerable room for improvement
  - Current detector characteristics (Maxipix, Eiger, SMD, Frame store FastCCD)
    - Frame rates ~ 100-1,000 s^{-1}
    - Pixel sizes ~ 30-100 um^2
    - Frame sizes ~ 0.1 – 1 Mpix
    - ~ 100% efficiency to ~ 12 keV
    - Isolated ad hoc firmware compression and off-line calculation of correlation functions



XPCS Detectors for a DLSR

- Greatly improved (XPCS) 2-D detectors are required to leverage the tremendous brilliance gains provided by a DLSR
  - Ideal XPCS detector characteristics:
    - Fast
      - Framing time to 1 µs or less
    - Efficient
      - Operation to ~ 20 keV
        - Mitigate damage in softer materials
        - Penetration of diverse sample environments
    - Single-photon sensitivity (low dynamic range)
    - Large area
      - Signal-to-noise increases as square root of number of “detectors”
    - Pixel sizes of 50-100 µm
      - Focusing and sample-to-detector distance mitigates the need for really small pixels
    - Counting or integrating vis-à-vis charge sharing?
    - Upstream intelligence
      - Charge sharing
      - Compression/Sparse readout
      - Short-time correlations

Microstructural Avalanches

Opportunity

- *Industrially relevant steel and shape memory alloys undergo diffusionless (martensitic) transformations*
- *Martensitic transformations involve cooperative motion of large groups of atoms*

Challenge

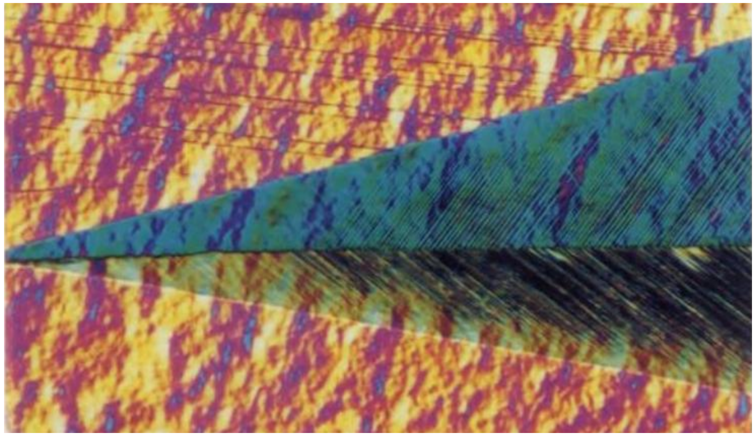
- Events are intermittent and localized
- Current observation techniques provide before and after snapshots

DLSR Strength

- Coherent beam provides localized information even with a large illumination area
- Microsecond probe of avalanche event dynamics
- Measurements relevant to fatigue mechanism in materials

Collective Avalanche Events

Time resolution to resolve dynamics of diffusionless transformations



Spear of martensite (low T phase) penetrating the austenite (high T phase) during transformation.

Fluctuations in Strongly Correlated Materials

Opportunity

- *Competing order leads to frustrated ground states in strongly correlated materials*
- *Fluctuations between equivalent ground states provide fundamental information on the competing interactions in such materials and are relevant to their stability*

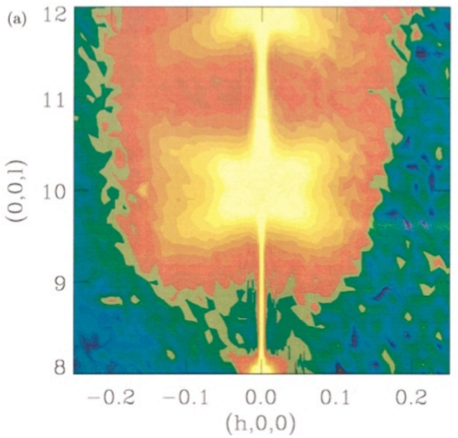
Challenge

- Environments such as low temperatures and strong magnetic and electric fields thwart measurements
- Small samples and surface effects complicate experiments

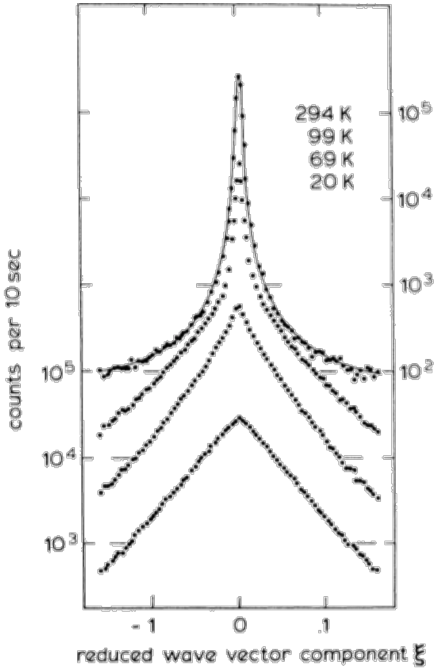
DLSR Strength

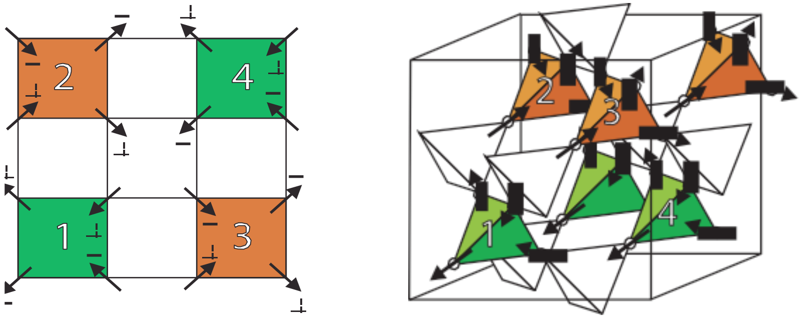
- Access through complex sample environments and to bulk properties
- Rapid dynamics

Competing Ground States in Orientational Glasses (Loidl et al.)



"Polaron" Hopping Dynamics





"Spin Ice" Frustrated Ground State

Fluctuations in Hierarchically Ordered Materials

Opportunity

- *Hierarchically ordered materials can provide a suite of engineered functionality such as artificial photosynthesis, media separation and emulsion stability (oil recovery) but the activity and stability of such materials is key to their use*
- *Microscale structure and stability of permafrost regions has significant environmental and industrial impact*
- *Engineered jamming and unjamming to control optical properties and chemical activity*

Challenge

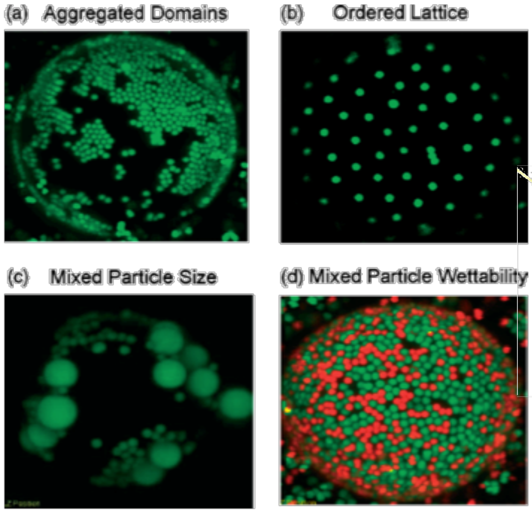
- Dynamics over a broad span of timescales
- Dynamics in dense environments

DLSR Strength

- Broad dynamic range

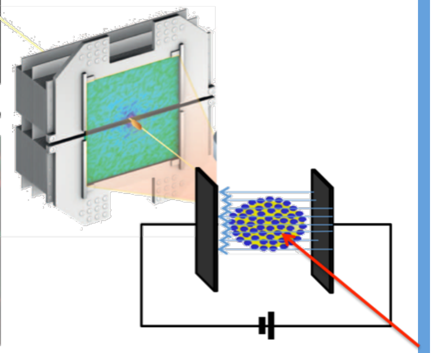
Particle Dynamics at Interfaces

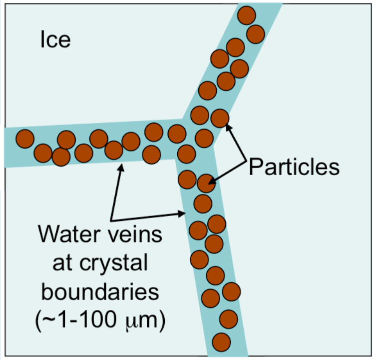
Engineered Functionality



L. Dai et al., Scanning **30**, 87 (2008)

Controlled Jamming





Thawing Permafrost

Origin of Dynamics in Glassy Materials

Opportunity

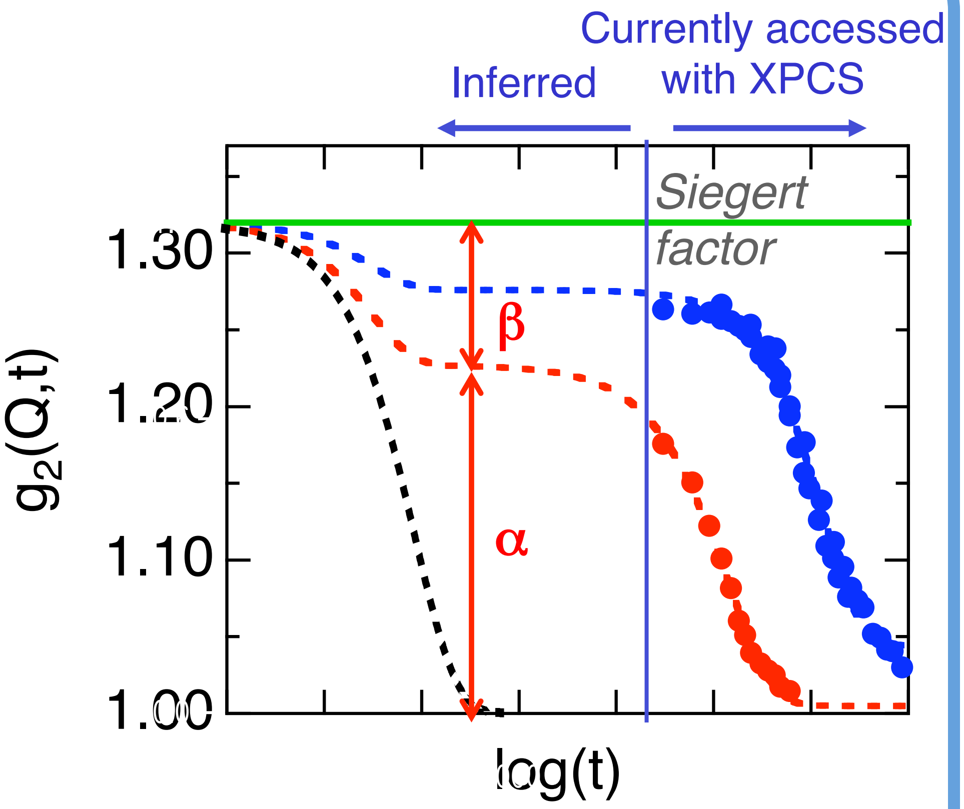
- Hierarchical dynamic phenomena in glassy materials: colloidal suspensions, gels, nanoemulsions, polymers. Microscopic origin of high viscosity in glassy materials.
- Ability to probe both the alpha and the beta relaxation at the nanoscale

Challenge

- Limited coherent flux to access micro second time scales
- Limited coherent fraction to access nanometer length scales

DLSR Strength

• 100x brilliance will expand the accessible dynamic range to microseconds, enabling probing the full dynamic evolution and fast localized dynamics (beta relaxations)



Sub-Micron Flow Profiles

Opportunity

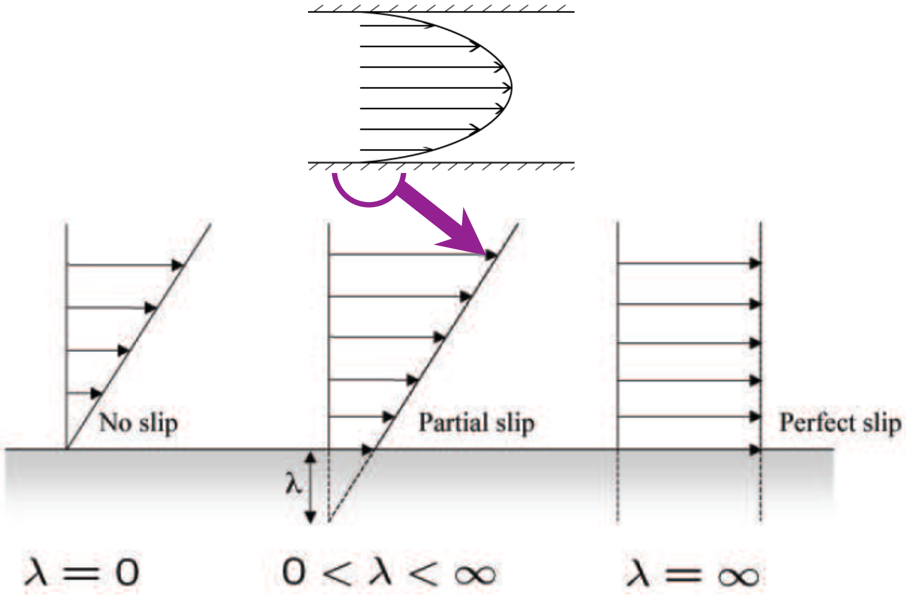
- Measure flow velocity profiles with sub-micron spatial resolution. Nanometer flow profiles are important to understand dynamics in materials
- Demonstration experiment: studying velocity boundary conditions at fluid-solid interface

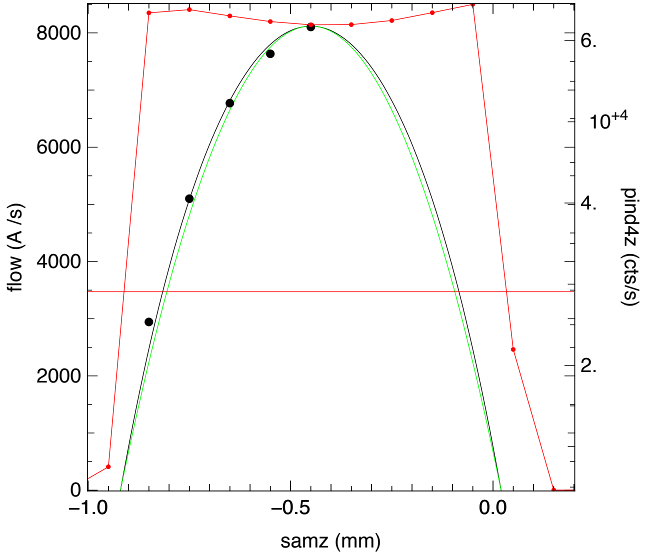
Challenge

- Limited coherent flux and coherent fraction to probe nanoscale dynamics

DLSR Strength

- Sub-micron focusing with increased coherent flux will enable probing faster velocities and/or smaller sample volumes such as in micro and nano-channels





Nanoscale Dynamics and Macroscale Rheology

Opportunity

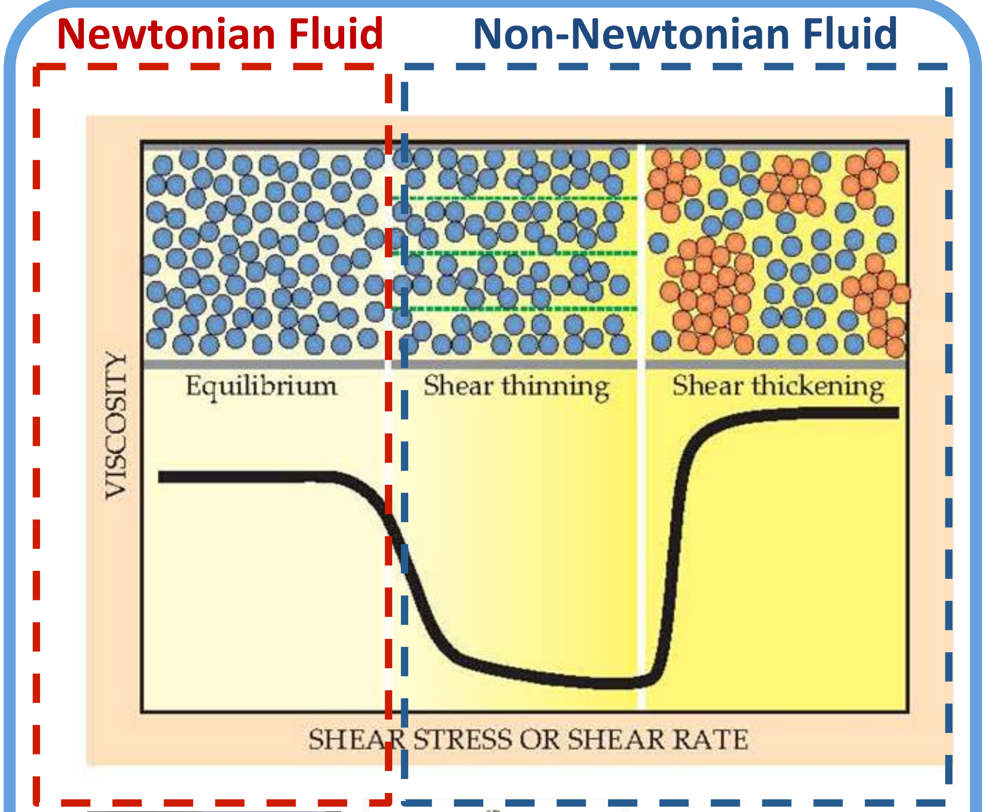
- Linear and non-linear bulk rheology and its relation to nanoscale dynamics in complex fluids
- Shear induced velocity banding and flow instabilities in complex fluids
- Probe of stress relaxation at the nanoscale as manifested in the non-eq. dynamics

Challenge

- Limited coherent fraction at high energies (20-30 keV) for environment penetration
- Limited coherent flux and coherent fraction to probe nanoscale dynamics at micro-second time scales

DLSR Strength

- High coherent fraction and coherent flux at high energies will enable probing real materials in real shear flow conditions



Rheo-SAXS-XPCS

Conclusions

- Modest improvements in 3rd generation SR's, XPCS detectors and beamline design are in line with XPCS SNR considerations and indicate many realizable future opportunities
- Suitable advanced 2-D detectors are absolutely required to utilize the gains provided by a DLSR
- Many science opportunities