Nanoscale Dynamics in Condensed Matter

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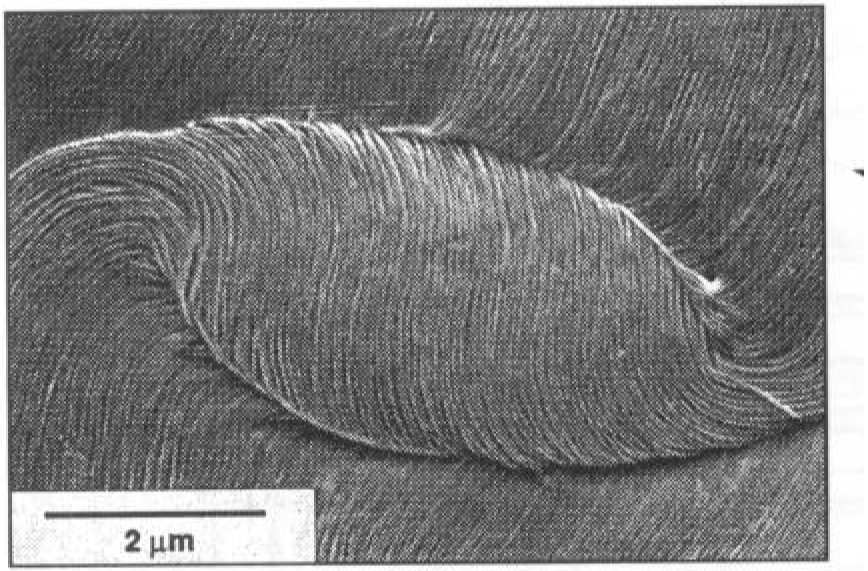
Why is Nanoscale Important in Dynamics?

- **Often determines mechanism of dynamics during materials processing**

Misfit-Strain-Induced Domain Walls in Ferroelectric Thin Film

C.M. Foster et al., *J. Applied Physics* **81**, 2349 (1997)

Magnetically-Induced Inversion Domain Wall in Nematic Liquid Crystal



M.J.E. O’Rourke and E.L. Thomas, *MRS Bulletin* **20** (9) 29 (1995)

Can be difficult to image during processing

Nanoscale Dynamics in Condensed Matter

Atomic- and nano-scale (<100nm) of great importance in dynamics

- Basic dynamic processes occur at atomic scale
- Overall dynamics mediated by defects and collective mechanisms at the nanoscale

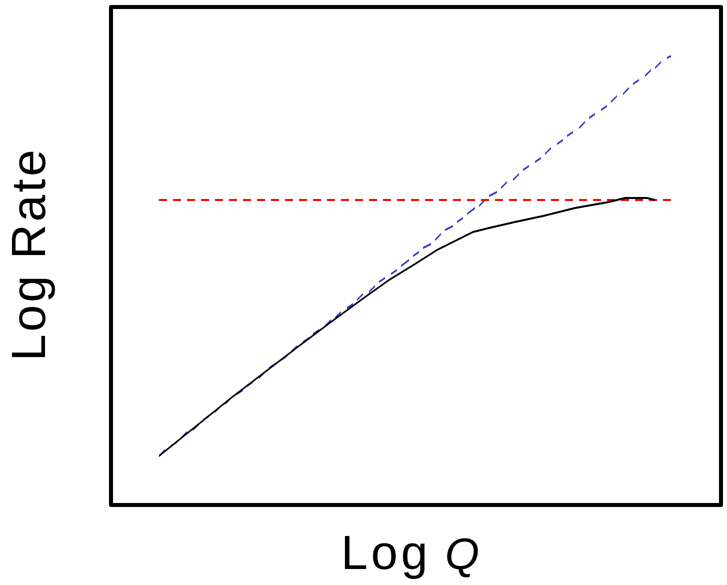
Would like to observe equilibrium dynamics:

- Non-equilibrium mechanisms are typically based on microscopic processes which occur, and are simpler to understand, at equilibrium
- Many useful properties are inherently dynamic

Advantages of Scattering Techniques

To understand dynamics, need *in-situ* techniques which resolve both *length* and *time*

Determining nature of rate-limiting step from wavenumber *(Q)* dependence of rate:



Rate $\propto Q^2$:
e.g. composition change
by diffusion
(conserved quantity)

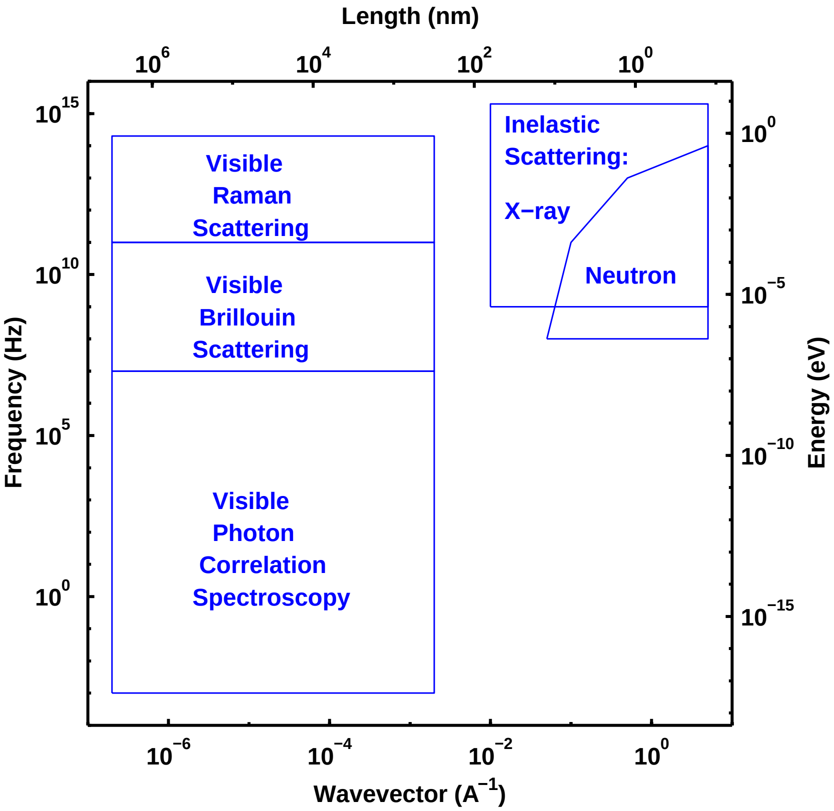
Rate indep. of Q :
e.g. deformation
by viscous flow
(non-conserved quantity)

Scattering Techniques for Equilibrium Dynamics

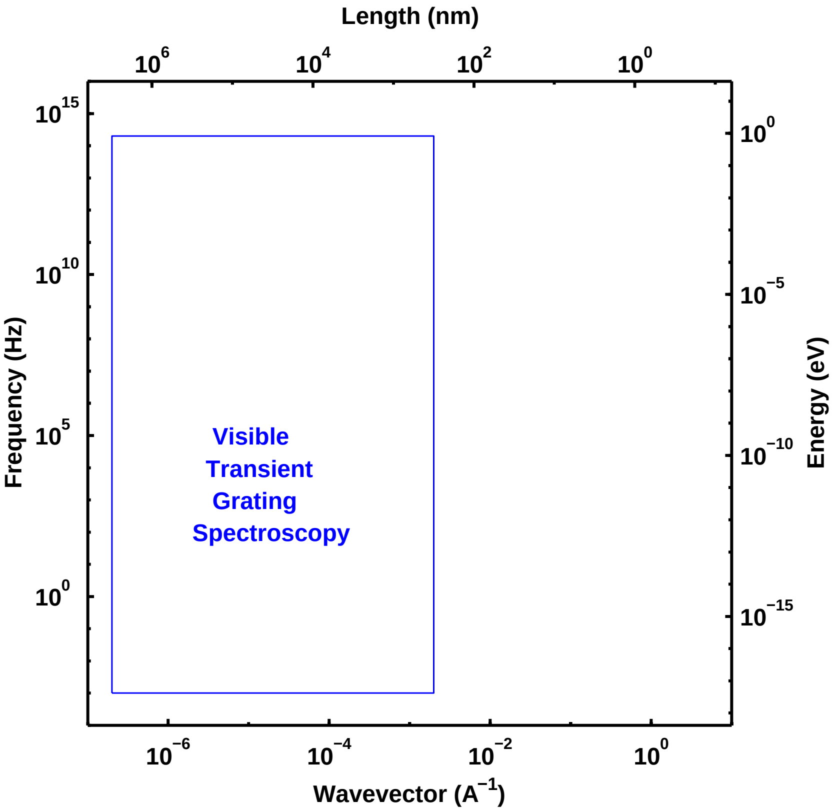
LCLS

Existing techniques

Probe thermal fluctuations:



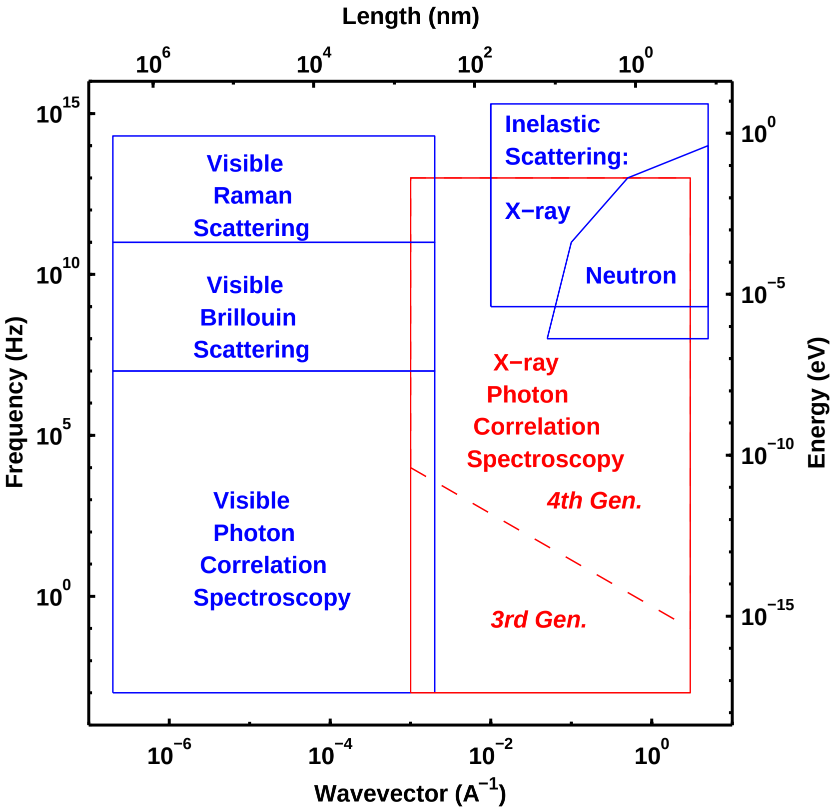
Excite and probe fluctuations:



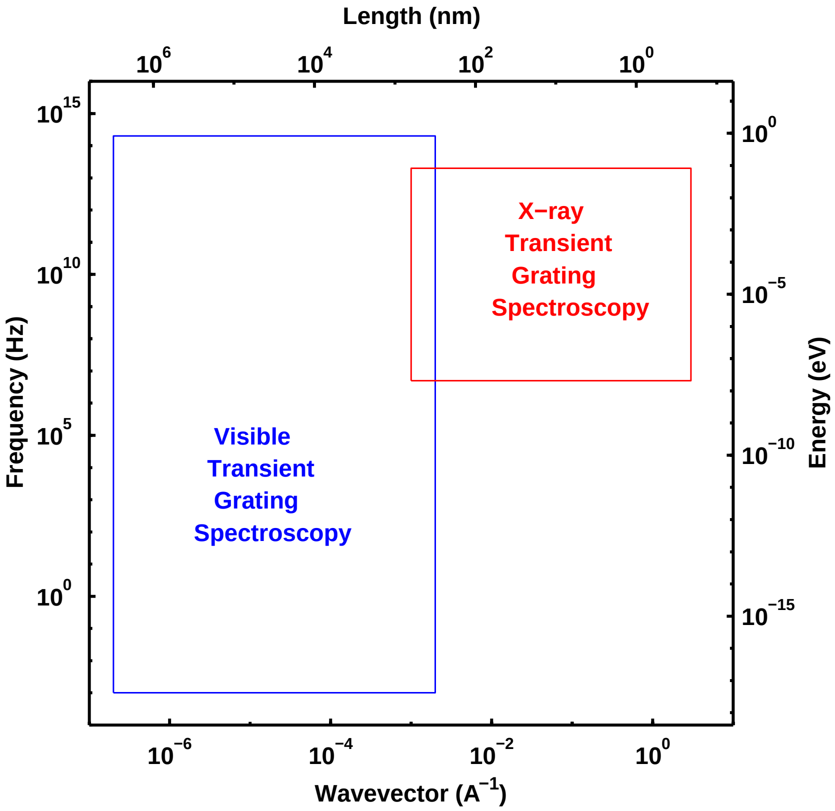
Scattering Techniques for Equilibrium Dynamics

XPCS and XTGS

Probe thermal fluctuations:

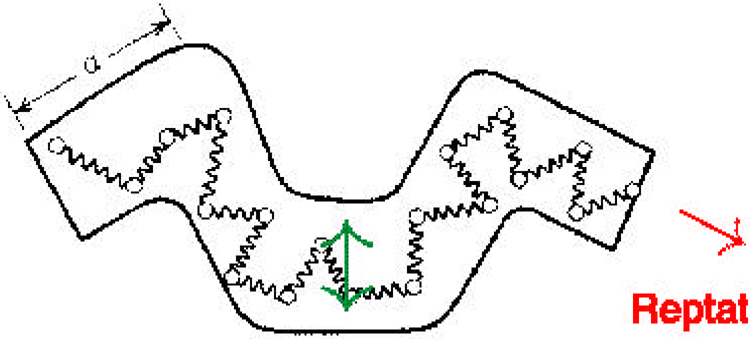


Excite and probe fluctuations:



Example: Test of Reptation Model

Dynamics of Long-Chain Polymers



Reptation

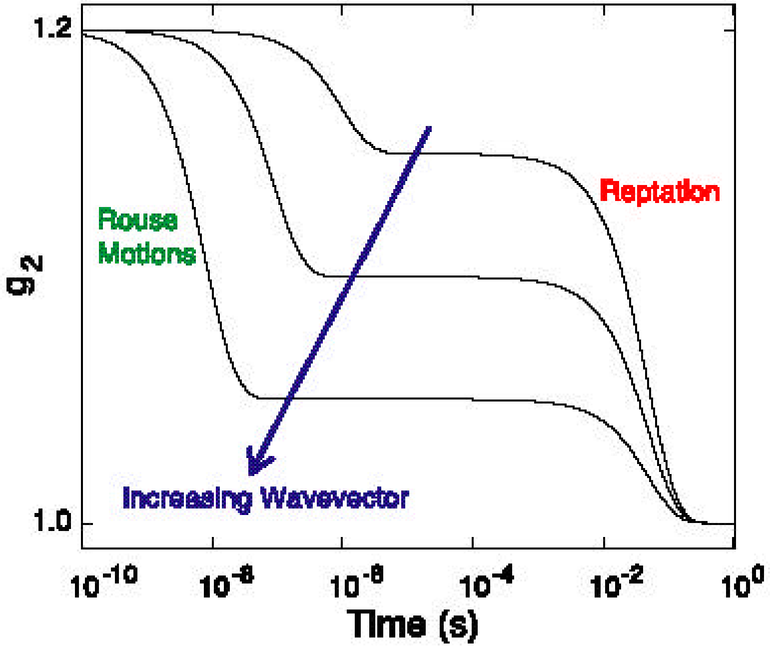
$t =$ “disentanglement time”

Independent of Q

Rouse Motions

$t \propto Q^{-3}$

Neutron spin-echo has been used to observe Rouse motions

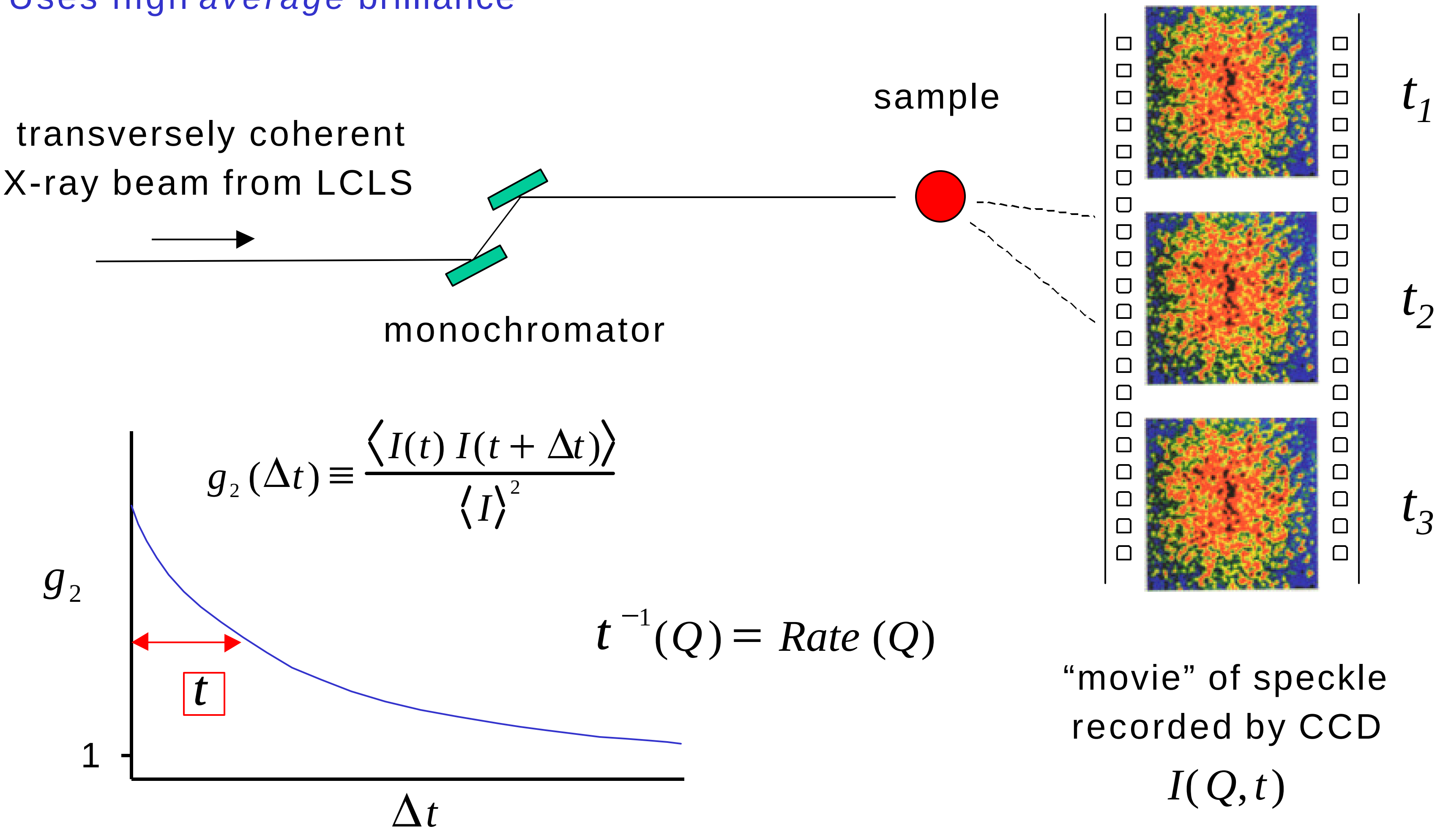


XPCS at LCLS would allow test of reptation model

Experiment 1:
X-ray Photon Correlation Spectroscopy (XPCS)

LCLS

In **milliseconds - seconds** range:
Uses high *average* brilliance



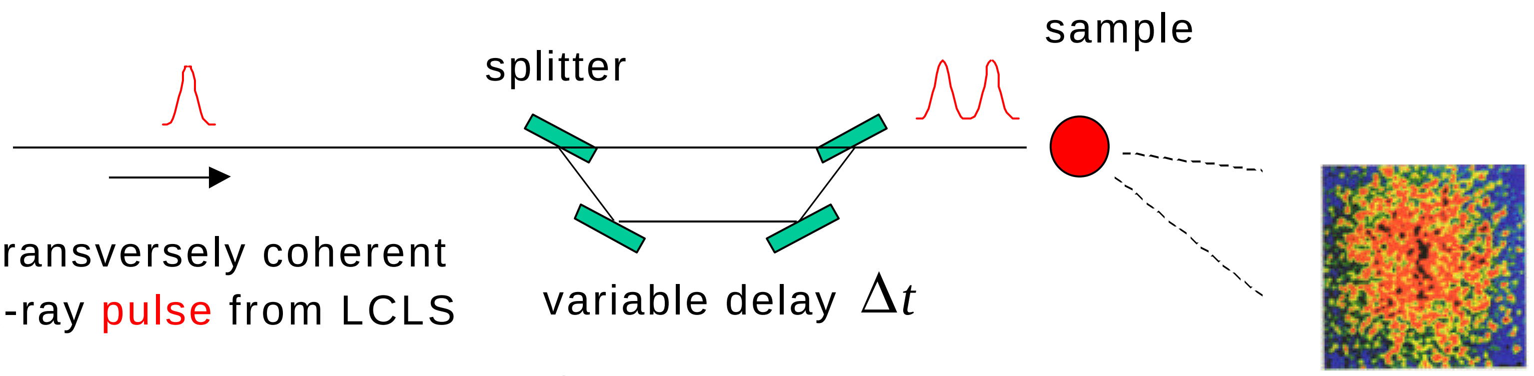
$$g_2(\Delta t) \equiv \frac{\langle I(t)\, I(t+\Delta t)\rangle}{\langle I\rangle^2}$$

$$t^{-1}(Q) = Rate\ (Q)$$

“movie” of speckle
recorded by CCD
$I(Q,t)$

Experiment 2: XPCS Using Split Pulse

In **picoseconds - nanoseconds** range:
Uses high *peak* brilliance



sample

splitter

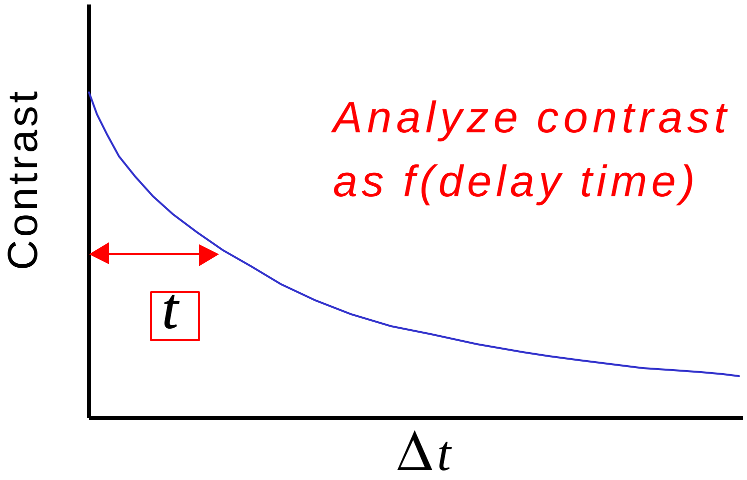
transversely coherent
X-ray pulse from LCLS

variable delay Δt

$10\ ps \Leftrightarrow 3\,mm$

sum of speckle patterns
from prompt and delayed pulses
recorded on CCD

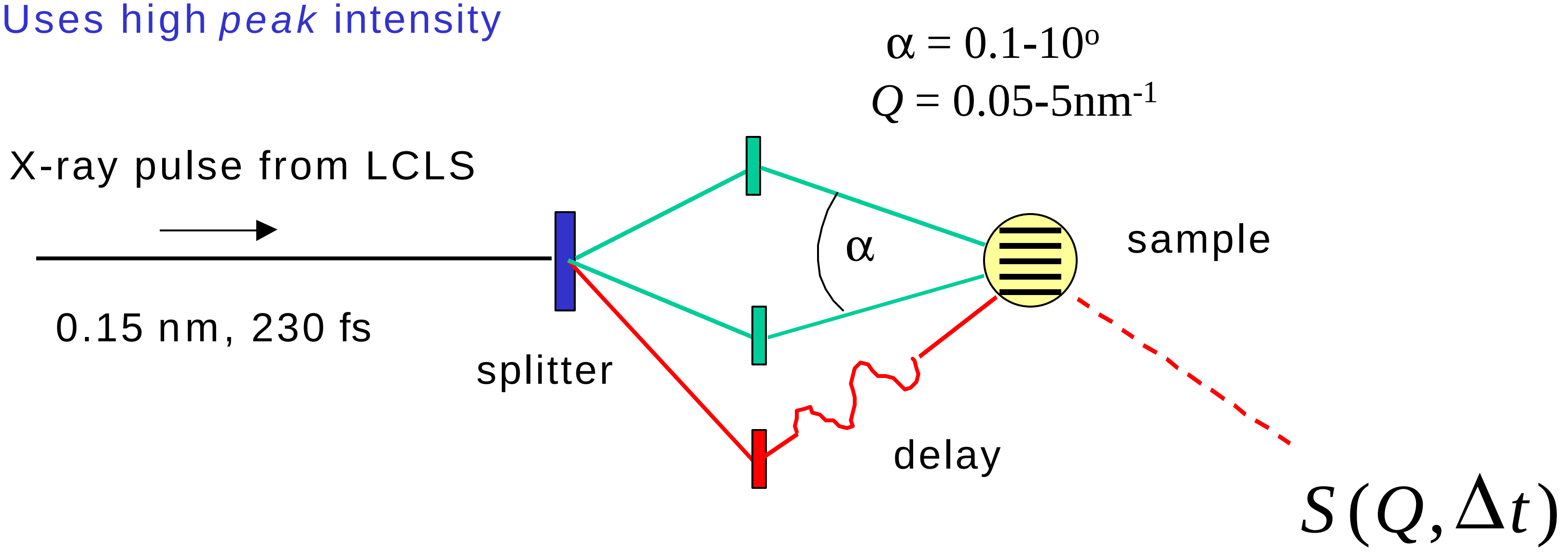
$I(Q,\Delta t)$



Analyze contrast
as f(delay time)

Experiment 3:
X-Ray Transient Grating Spectroscopy

In **picoseconds - nanoseconds** range:
Uses high *peak* intensity



$\alpha = 0.1\text{-}10^{o}$
$Q = 0.05\text{-}5\text{nm}^{-1}$

X-ray pulse from LCLS

0.15 nm, 230 fs

splitter

α

sample

delay

$S(Q, \Delta t)$

Drive system with chosen Q ,
observe response as f(delay time)

Feasibility Issues

Is there enough signal from a single LCLS pulse?
Is sample heating by x-ray beam a problem?

Available photons per pulse:

$$N_{AVAIL} = f(E, \Delta E, A)$$

Minimum photons per pulse to give sufficient signal:

$$N_{MIN} = \frac{2\pi A E^2 \sigma_{abs}}{h^2 c^2 \sigma_{el} M_{corr}} N_{MIN}^{SPECKLE}$$

Maximum photons per pulse to give 1° temperature rise:

$$N_{MAX} = \frac{3 k_B A}{E \sigma_{abs}} \Delta T_{MAX}$$

Heating and Signal from Unfocused Pulse

LCLS

Applications of XPCS and XTGS

- ***Simple Liquids*** **–** Transition from the hydrodynamic to the kinetic regime.
- ***Complex Liquids*** **–** Effect of the local structure on the collective dynamics.
- ***Polymers*** **–** Entanglement and reptative dynamics.
- ***Glasses*** **–** Vibrational and relaxational modes in the mesoscopic space-time region.
- ***Dynamic Critical Phenomena*** **–** Order fluctuations in alloys, liquid crystals, *etc.*
- ***Charge Density Waves*** **–** Direct observation of sliding dynamics.
- ***Quasicrystals*** **–** Nature of phason and phonon dynamics.
- ***Surfaces*** **–** Dynamics of adatoms, islands, and steps during growth and etching.
- ***Defects in Crystals*** **–** Diffusion, dislocation glide, domain dynamics.
- ***Ferroelectrics*** **–** Order-disorder *vs.* displacive nature; correlations and size effects.