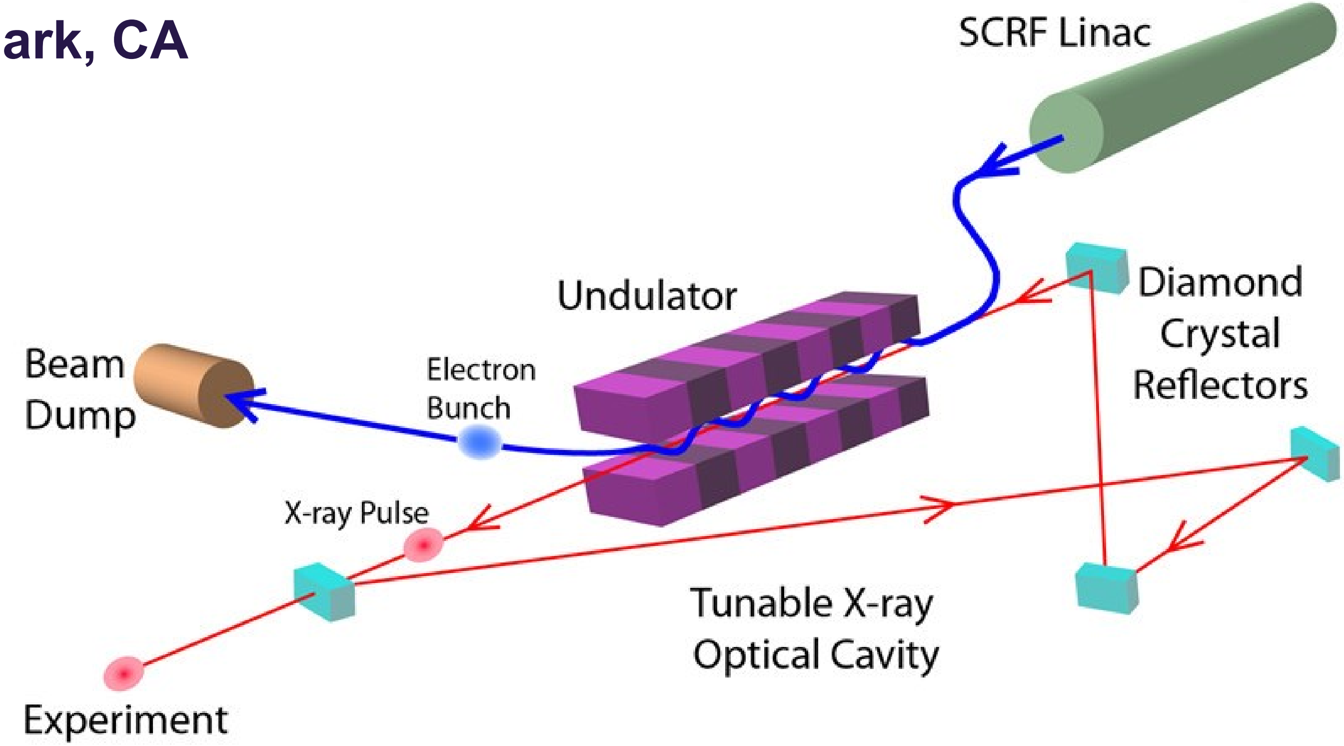
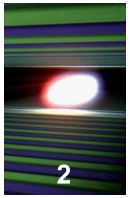
Thoughts about XPCS at an XFELO

XFELO Science Workshop
June 29 - July 1, 2016
SLAC National Accelerator Laboratory
Menlo Park, CA



A. Madsen, European XFEL, anders.madsen@xfel.eu

I had to look up what is an XFELO



Jerry said in the mail:

“5-25 keV photon energy range” (great, like European XFEL, but depends on C* reflections)

“3-10 meV bandwidth” (~10^{-6} bw, Wow! SASE is 10^{-3}. Fantastic possibilities for large NA optics, e.g. MLL, to reach nm focii)

“Full coherence” (transform limited? Then the pulse duration should be ~1ps – 400 fs)

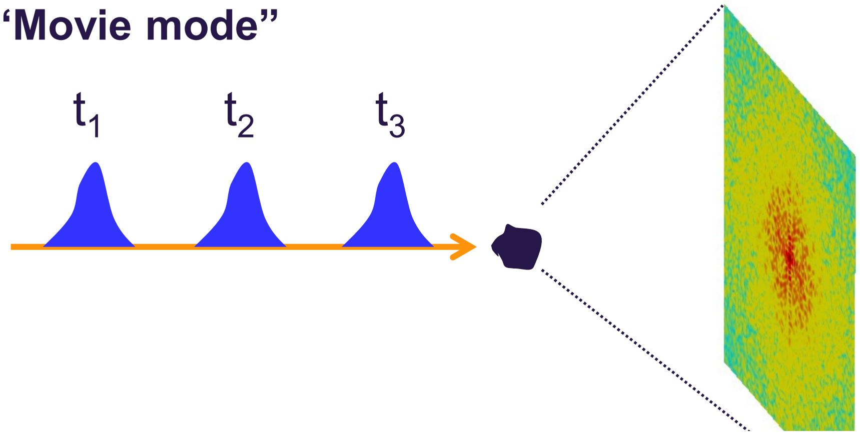
“Pulse length 200-500 fs” (OK, so like transform limited)

“10^8 – 10^9 ph/pulse” (not so much..)

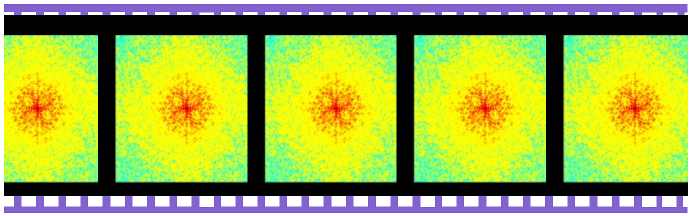
“1-2 MHz rep rate” (10^{14} - 10^{15} coherent photons/s, nice…)

Experimental modes

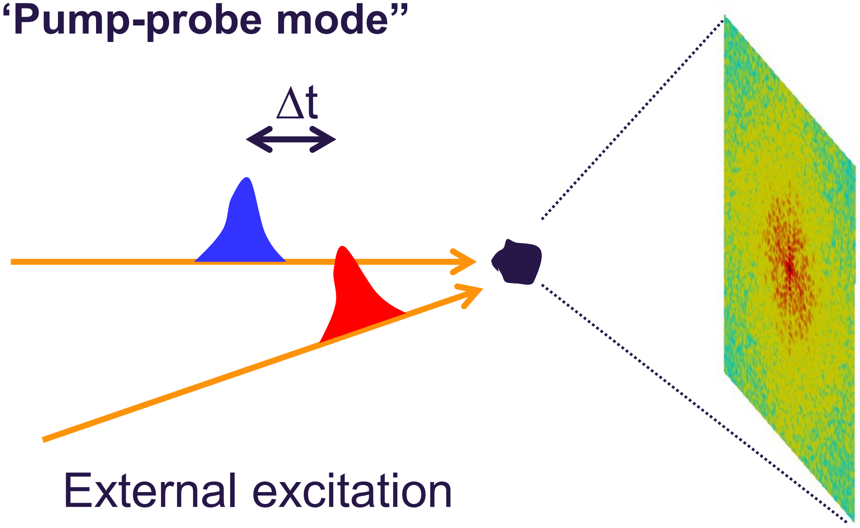
"Movie mode"



Movie mode at MHz rates, defined by XFEL rep rate and detector speed



"Pump-probe mode"



External excitation
(optical laser, B, E, T,…)

Δt can be very small, fs, ps, ns possible. Time encoded in every image.

Experiments can be destructive (diffract & destroy) but not necessarily (CDI).

Movie mode

10^{15} coherent photons/s for XPCS. What would it bring?

SNR XPCS $\propto (t_m)^{1/2} \times I_{coherent}$

Today: I ~ 10^{10} gives access to ms dynamics

XFELO: I ~ 10^{15} would give access to sub-ps dynamics

BUT:

No 2d detectors can run at sub-ps frame rates
Pulses only arriving at MHz rate (machine rep rate)

→ traditional XPCS at an XFELO will not bring us further than μs regime where there is **competition from MBA SR** (single shot damage with XFELO…)

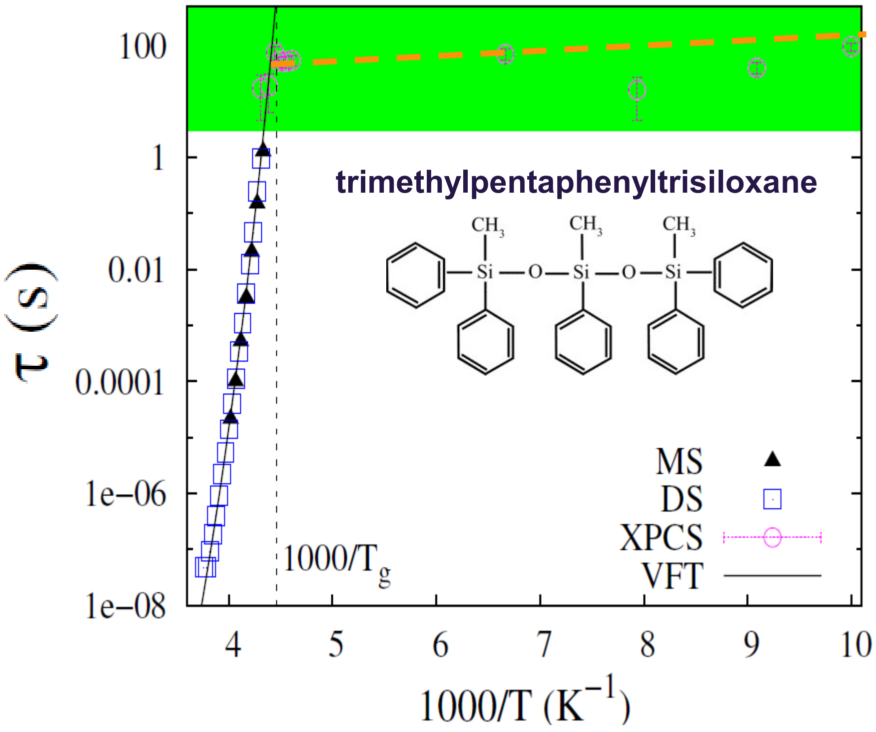
→ ultrafast dynamics studies at an XFELO still requires advanced optical schemes (**e.g. split-delay line**,…)

Possibilities for MHz Movie Mode XPCS

Study of supercooled liquid glass formers at high Q

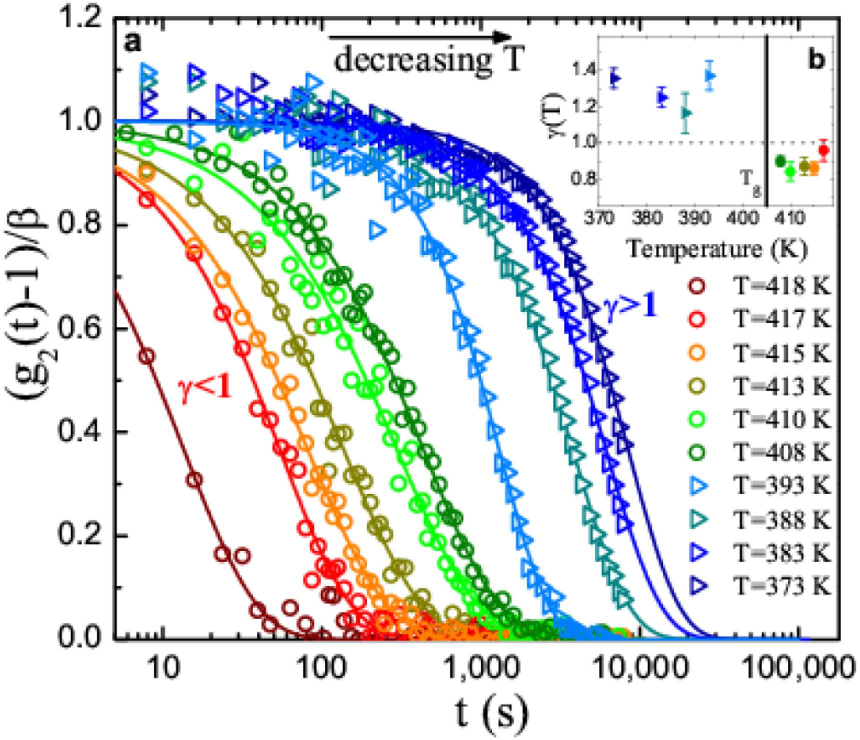
XPCS data on DC705, Q=7.5 nm^{-1}

Chushkin et al., JAC **45**, 807 (2012)

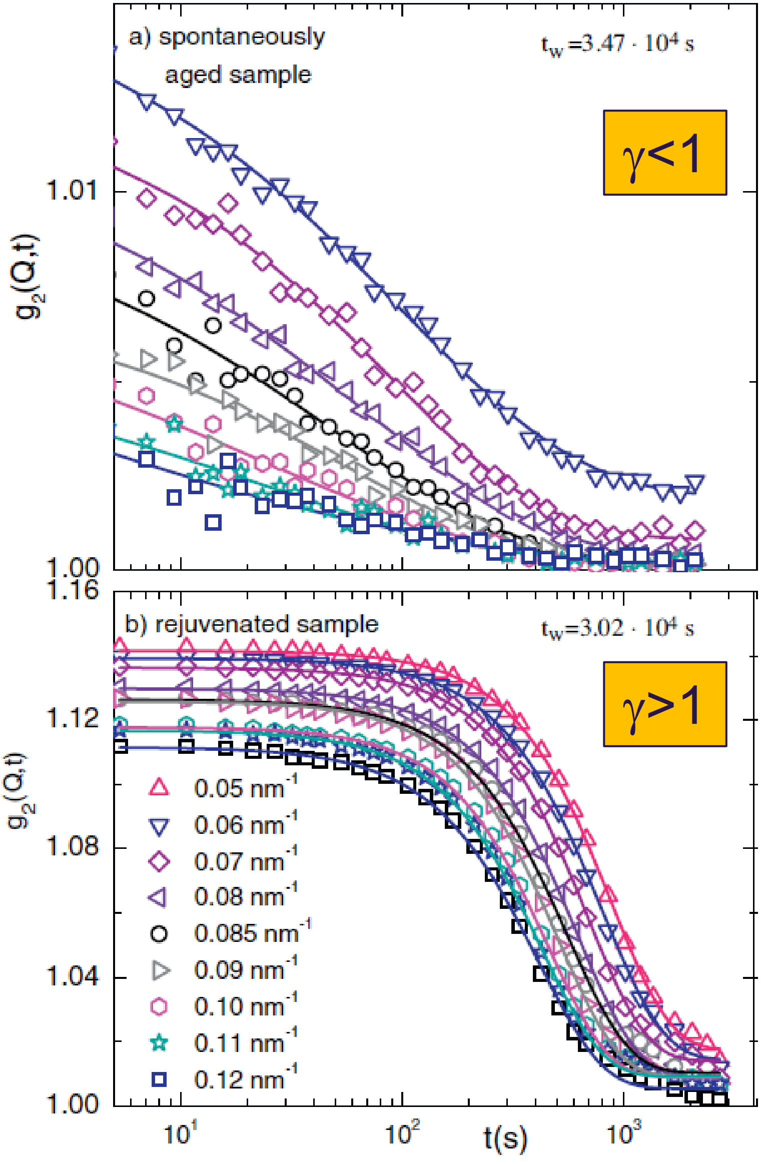


XPCS in metallic glass at Q=25 nm^{-1}

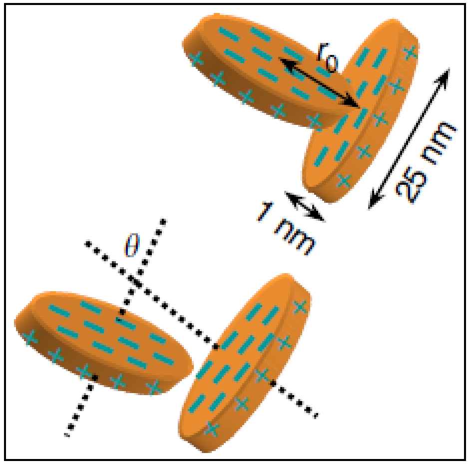
Ruta et al., PRL **109**, 165701 (2012)

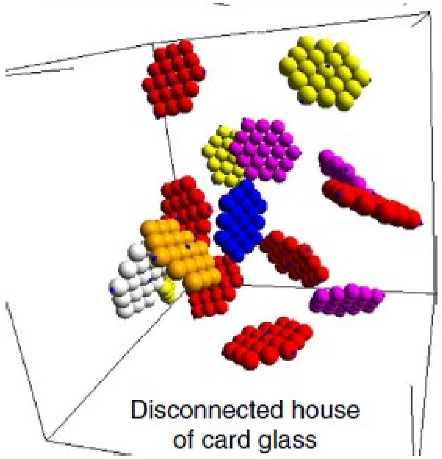


Possibilities for MHz Movie Mode XPCS



$$g^{(2)}(Q,t) = 1 + \exp(-2[t/\tau]^{\gamma})$$



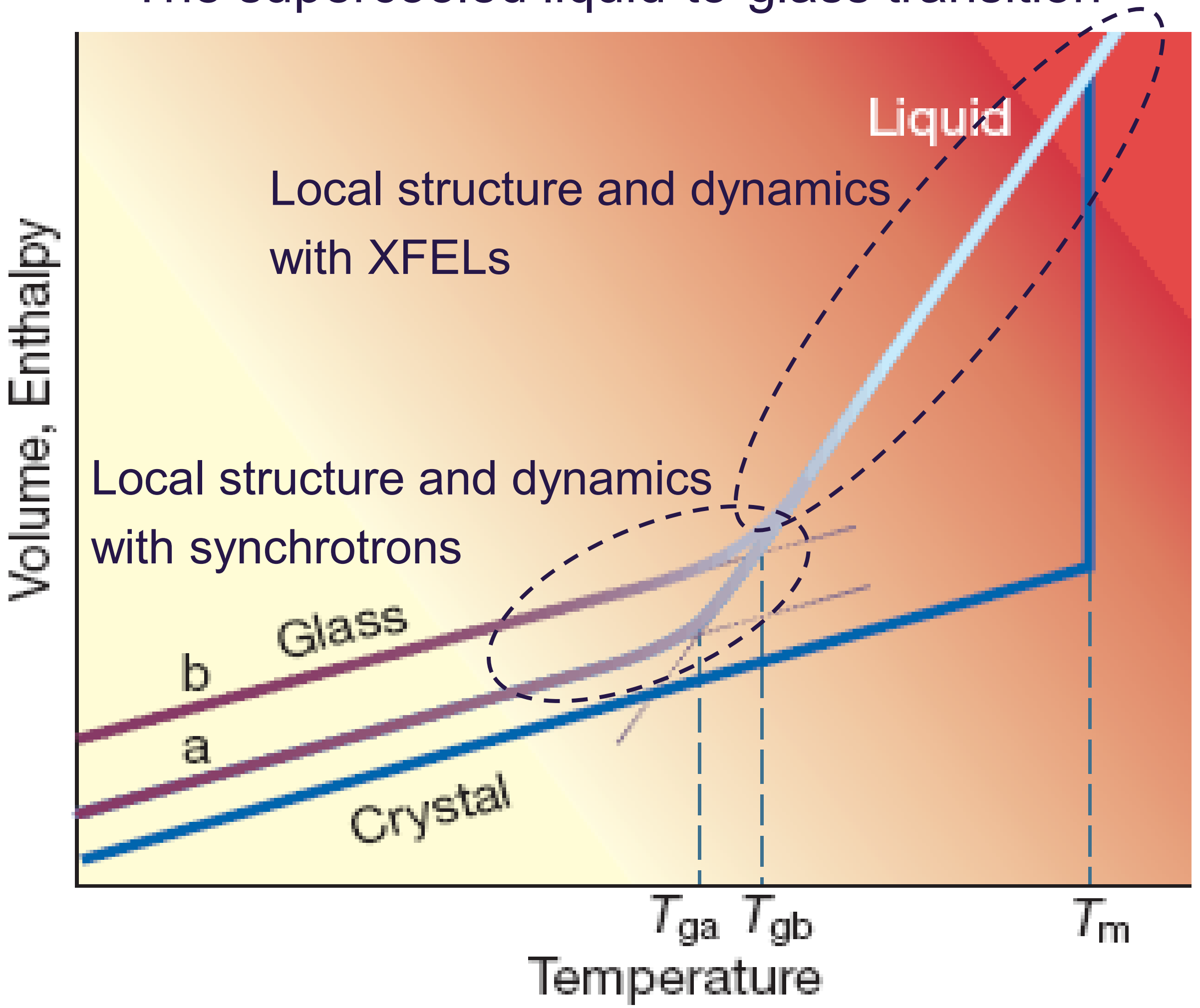

Disconnected house of card glass

Aging dynamics of a Laponite glass:

R. Angelini *et al*., Soft Matter **9**, 10955 (2013)

Possibilities for MHz Movie Mode XPCS

The supercooled liquid-to-glass transition



Local structure and dynamics with XFELs

Local structure and dynamics with synchrotrons

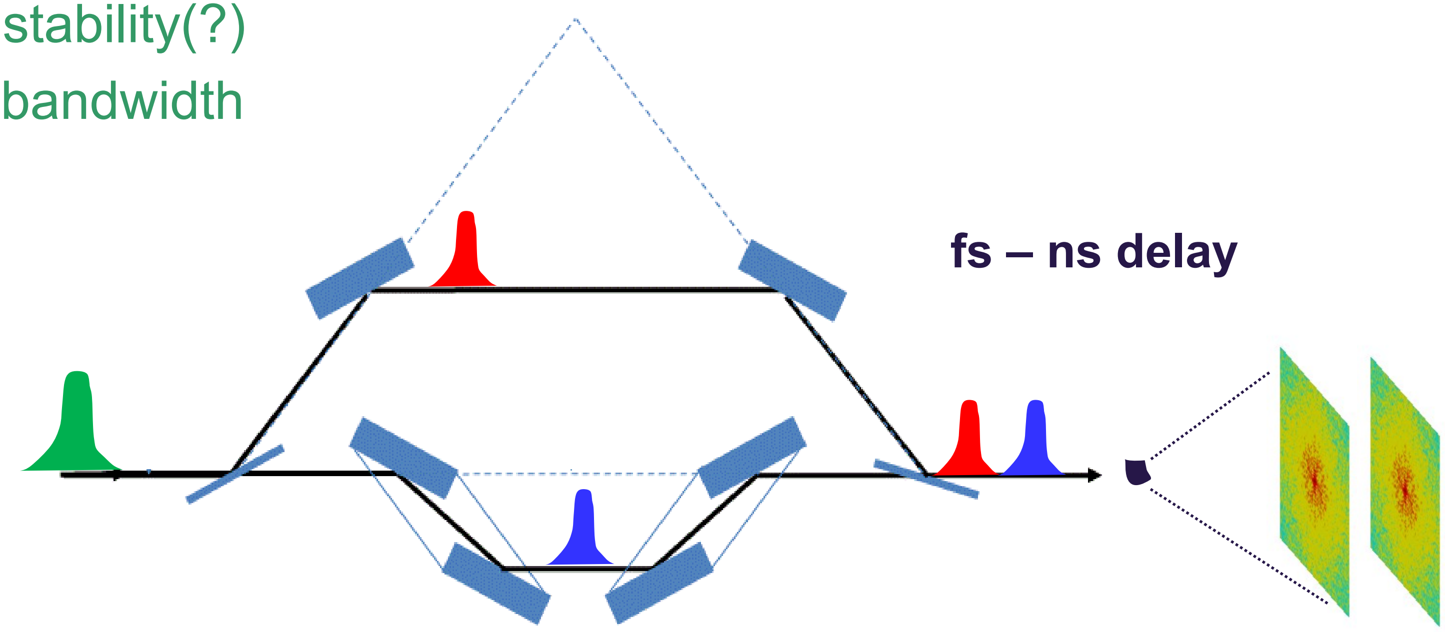
Pablo G. Debenedetti and Frank H. Stillinger, Nature (2001)

Split-delay line (SDL) at an XFELO

Better than with regular XFELs?

+ stability(?)
  bandwidth

- less ph/pulse



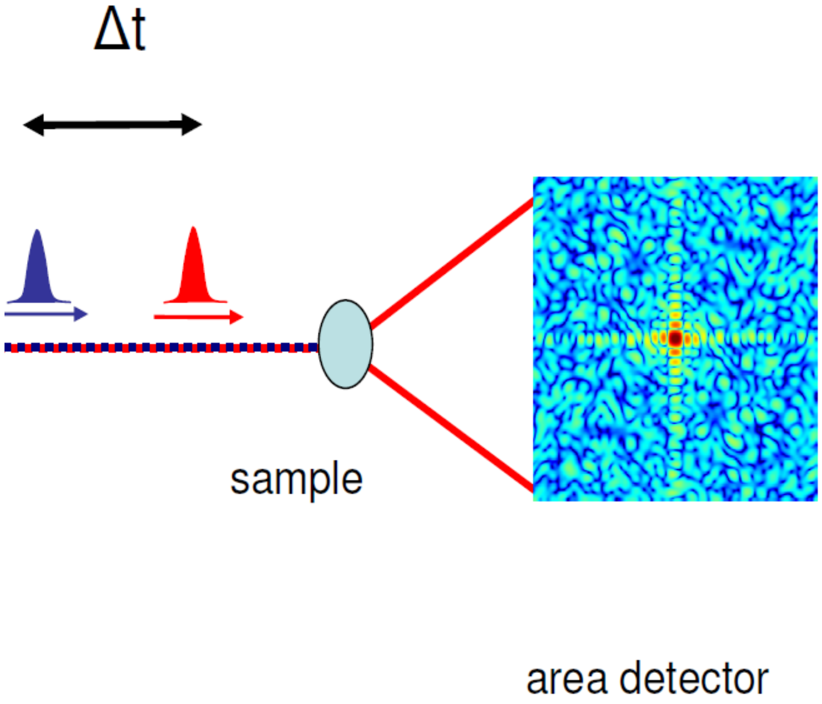
fs – ns delay

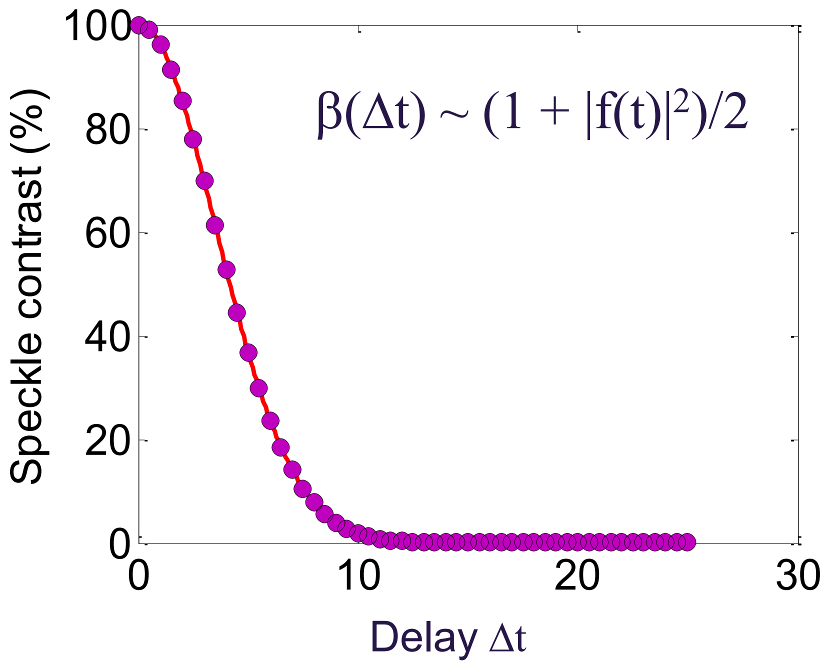
Detector cannot discriminate images → sum recorded

Dynamics via double pulse exposures

Image analysis (normalized variance) yields the speckle contrast b of summed images.

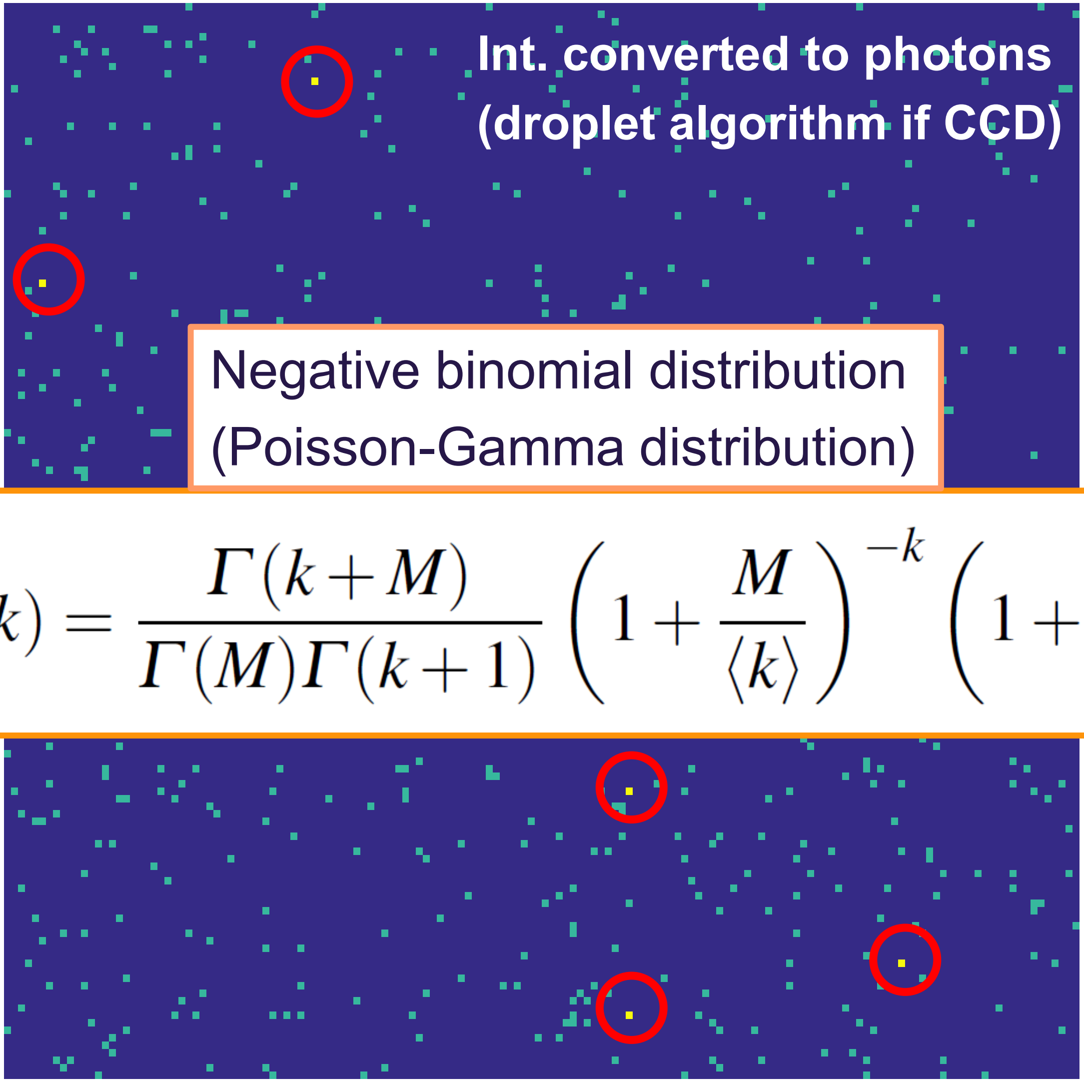
$\beta(\Delta t)$ can be mapped out




$$\beta(\Delta t) \sim (1 + |f(t)|^2)/2$$

C. Gutt *et al.*, Optics Express **17**, 55 (2009)

Homeopathic speckle sum-images



0 ph/pix

1 ph/pix

2 ph/pix

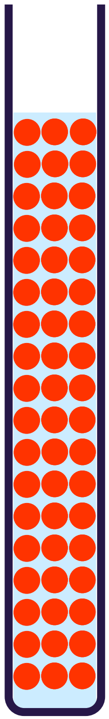
Int. converted to photons (droplet algorithm if CCD)

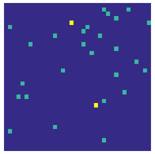
Negative binomial distribution
(Poisson-Gamma distribution)

$$P_M^{P-\Gamma}(k) = \frac{\Gamma(k+M)}{\Gamma(M)\Gamma(k+1)}\left(1+\frac{M}{\langle k\rangle}\right)^{-k}\left(1+\frac{\langle k\rangle}{M}\right)^{-M}$$

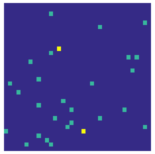
$$\beta = 1/M$$

Double pulse experiments at a synchrotron





Sum of images taken at same spot with delay time Δt } correlated images



Sum of images taken at different spots or with Δt >> τ } uncorrelated images

Poisson-Gamma distribution

$$\frac{P(0)}{P(1)} = \frac{1}{\langle k \rangle} + \beta$$

$$\frac{P(1)}{P(2)} = \frac{2\beta + 2/\langle k \rangle}{1 + \beta}$$

Need:

Precise identification of 0, 1, and 2 (droplet algo.)
Precise measure of <k> (average photon number)

Double pulse experiments at a synchrotron

$$\beta = \frac{P(0)}{P(1)} - \frac{1}{\langle k \rangle}$$

GeO_2 glass, T=20°C

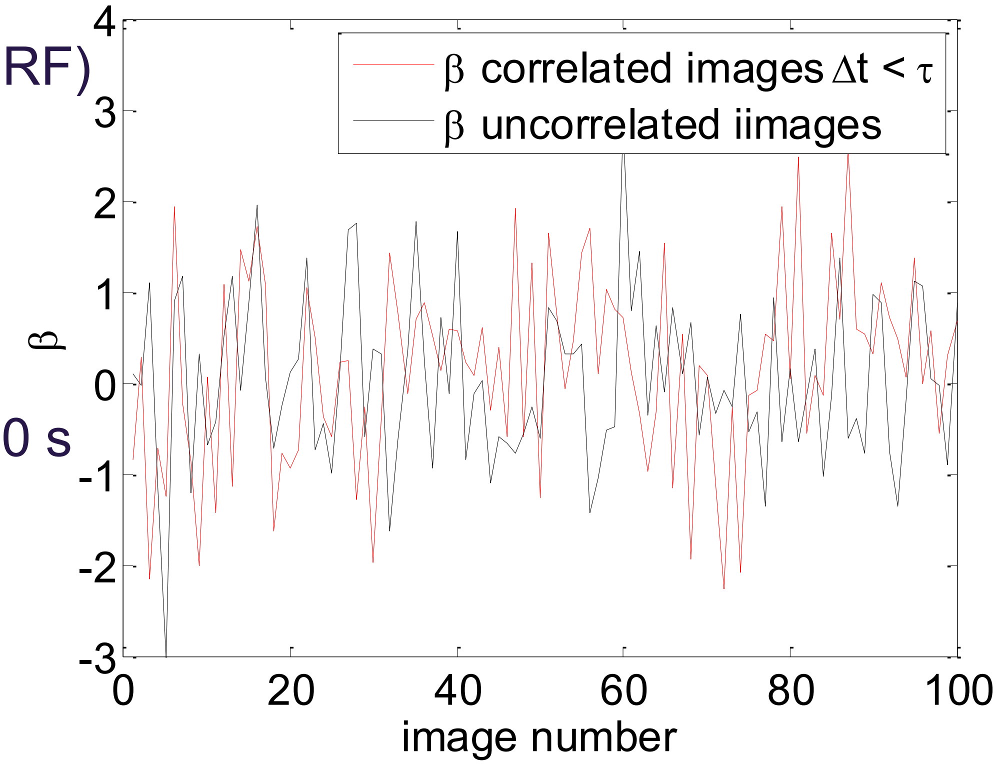
Data taken at Q ~2.5 Å^{-1} (ID10, ESRF)

Andor CCD data, 13 μm pixels, dropletized

Correlated images: Δt = 6 s; τ ~ 60 s

Uncorrelated images:

Images separated by Δt = 600 s

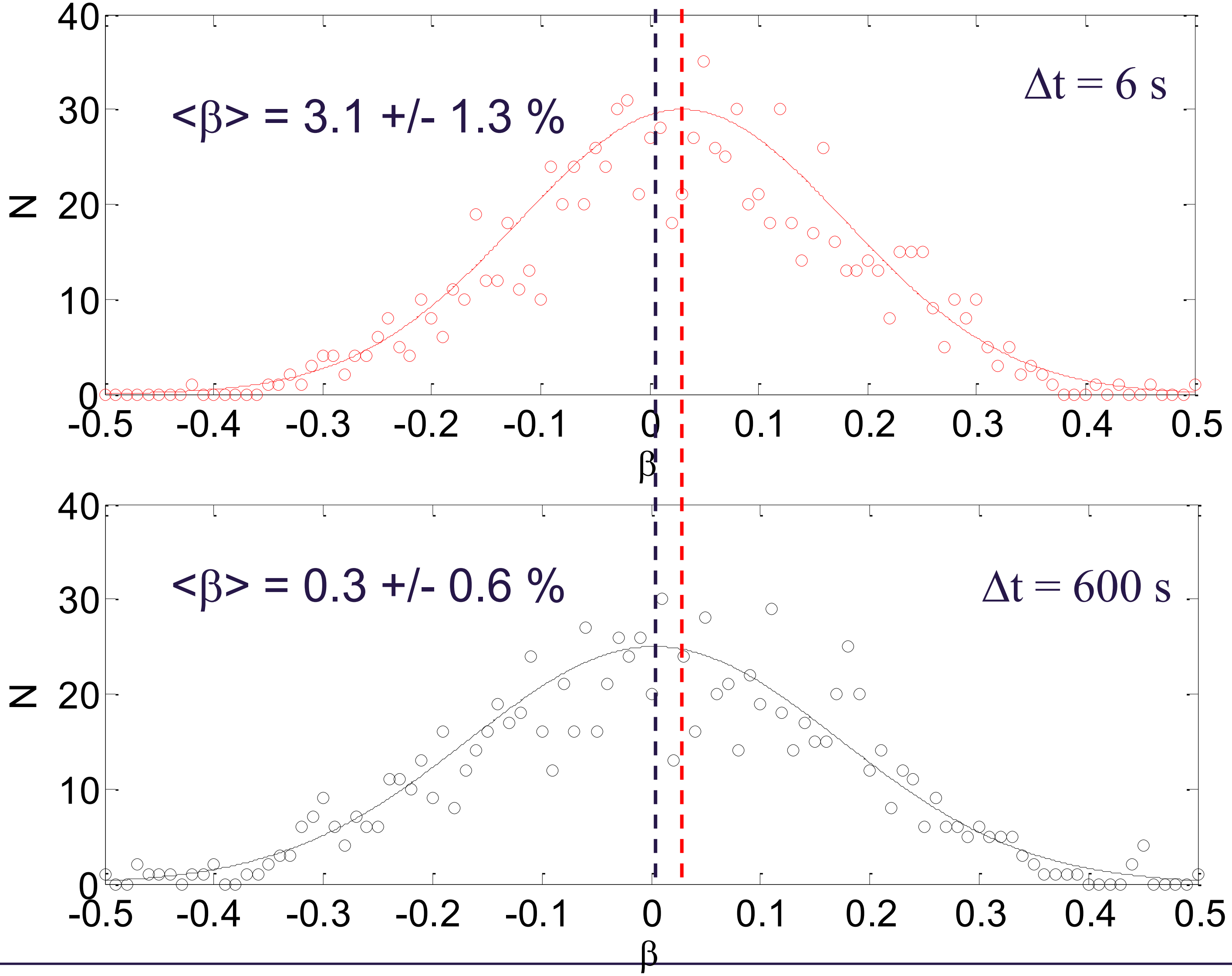


Double pulse experiments at a synchrotron

Analysis of
700 images

$$\beta = \frac{P(0)}{P(1)} - \frac{1}{\langle k \rangle}$$

$$\Delta\beta = 2.8\ \%$$



Double pulse experiments at a synchrotron

Data on GeO_2 glass, Q ~2.5 $Å^{-1}$ (ID10, ESRF)

ESRF

SORRY, picture removed ☺

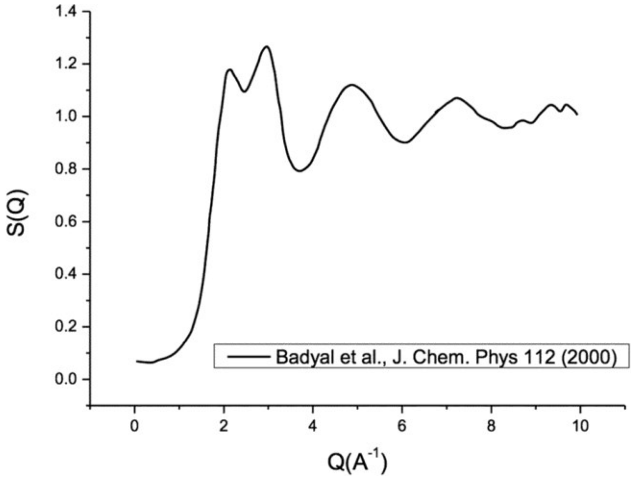
Hallmann et al, unpublished

High stability, or at least precise measurement of I_0 for every individual pulse, is mandatory for the sum technique to work.

Advantage of XFELO stability?

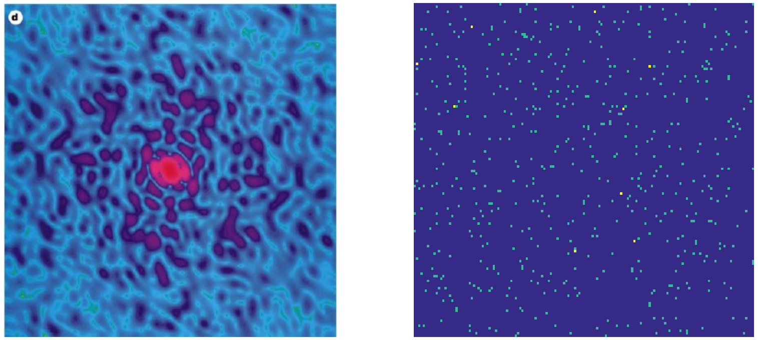
Photons/pulse drawback

Differential scattering cross section $H_2O \sim 3 \times 10^{-24}$ cm^2/(rad)^2/atom (1st peak at ~ 2 Å^{-1})



Scattering from (10 μm)^3 cube of H_2O:
~100 ph per 10^9 incident photons in a 10 × 10 cm^2 detector at 5 m distance

100 ph in 1M pixels → **1×10^{-4} ph/pixels/pulse**
(difficult conditions for speckle patterns)

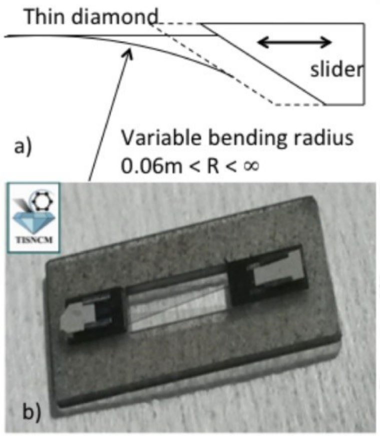


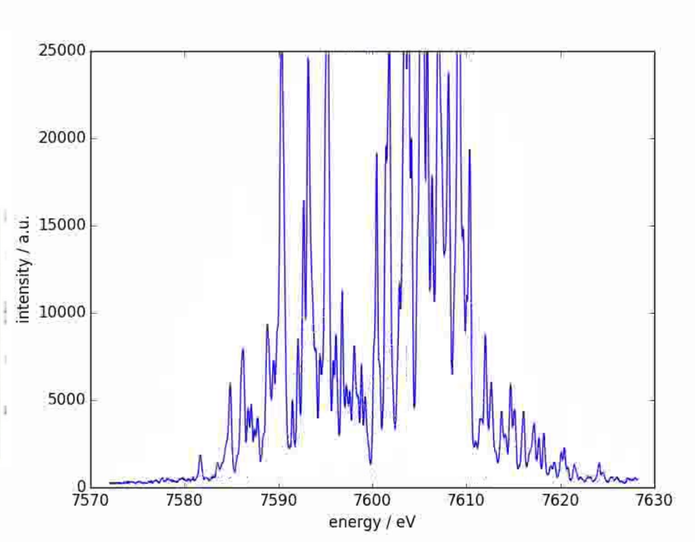
Hard to distinguish speckle from poison statistics & noise

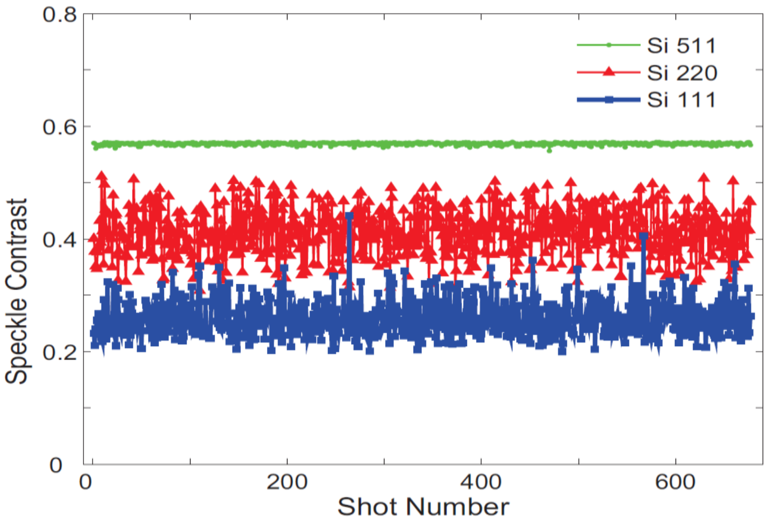
Bragg peaks will help but maybe difficult to compete with high gain XFEL

Bandwidth advantage

XPCS is a time-domain technique, i.e. small $\Delta E/E$ not so important. However, there is an **improvement of the speckle contrast at high Q**, depending on beam size and sample thickness.







S. Lee *et al.*
Opt. Express **20**, 9790 (2012)

But **Fourier transform spectroscopy** may become possible in the hard X-ray domain. $L_{coh} \sim \lambda/(\Delta\lambda/\lambda) \sim 150\ \mu m \rightarrow t_{coh} = L_{coh}/c = 0.5$ ps

XFELO application: Fourier Transform X-ray Spectroscopy (FTXS)

Based on measurements of the temporal/spatial coherence of light

Analogous to FTIR spectroscopy and the Michelson interferometer

<u>FTXS recipe:</u>

1) Scan the pulse overlap and read out the images
2) Interferogram I(Δt) collected for every pixel
3) Group pixels, maybe averaging to get I(Δt,q)
4) FT to get **S(ω) vs q**, or **g(r) vs Δt**

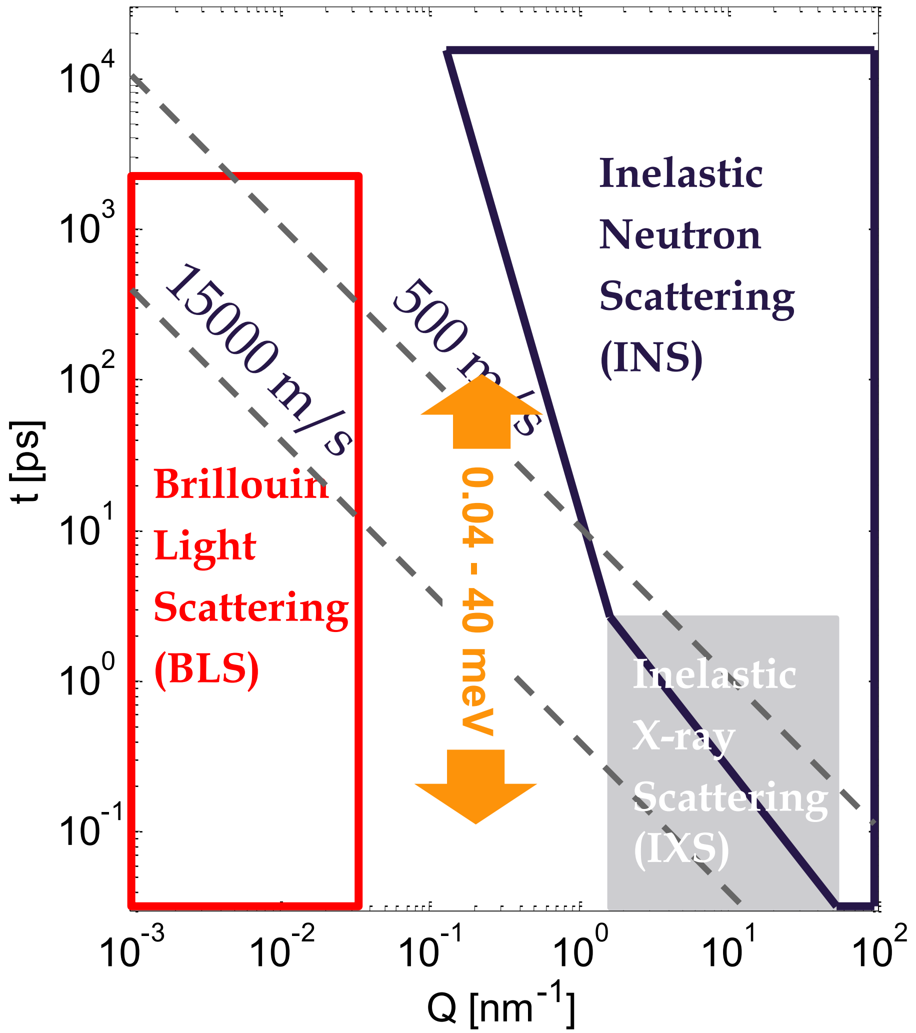
Possible to use for broadband lensless imaging (CDI) as well

Already done with broadband (HHG) sources

(S. Witte *et al., talk at Coherence 2016*)

European XFEL

FTXS Science



Possible science examples:

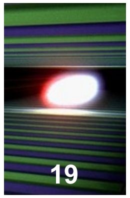
High Tc SC, cuprates, ferropnictides
SC bandgap, nesting, nm-μm corr.

Correlated electrons, Charge-spin correlations, SDW, CDW dynamics

Electron-phonon interactions

Correlated dynamics in disordered systems, supercooled liquids, glasses polymers…

Summary



- MHz rep rate, high stability together with 10^{15} ph/s (coherent) may give unique options for μs dynamics (single-shot rad damage?)
- SDL techniques will benefit from the XFELO stability but suffer from the low ph/pulse number; combined effect hard to quantify
- High spectral purity biggest asset (and distinct feature) as the longitudinal coherence is better defined, important for high-Q scattering from disorder
- Split-delay-interfere experiments possible: FTXS
  Previous suggestion: D. Reis, Next talk: M. Sutton
- Compact SDL design for FTXS as limited scanning range is required (double pendulum, Si monolith?)
- Sparse (homeopathic) data can (almost) always be expected. Need conversion to photons. Excessive detector requirements…
- New possibilities for high NA focusing