Ultrafast XPCS

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HELMHOLTZ | ASSOCIATION

DESY

Ultrafast Phenomena of interest for ultrafast (sub-ns) XPCS

Atomic-scale dynamics in liquids and glasses on (sub-ns) timescales

fluctuations in glassforming liquids,.. water[1),…liquid metals[2) ,..

Surface fluctuations

capillary wave dynamics[3) at large Q (beyond equilibrium hydrodynamics)

Fluctuations in magnetic systems

ultrafast (de-)magnetization in rare earth systems (Gd[4) ,..)

Transient electronic structures and ultrafast dynamics in solids

* lattice relaxations in systems undergoing metal-insulator transitions (e.g. RTe_3 systems[5))
* coherent optical phonos (following photoexcitation): Bi[6) , Sb, Te,…

Non-equilibrium phenomena

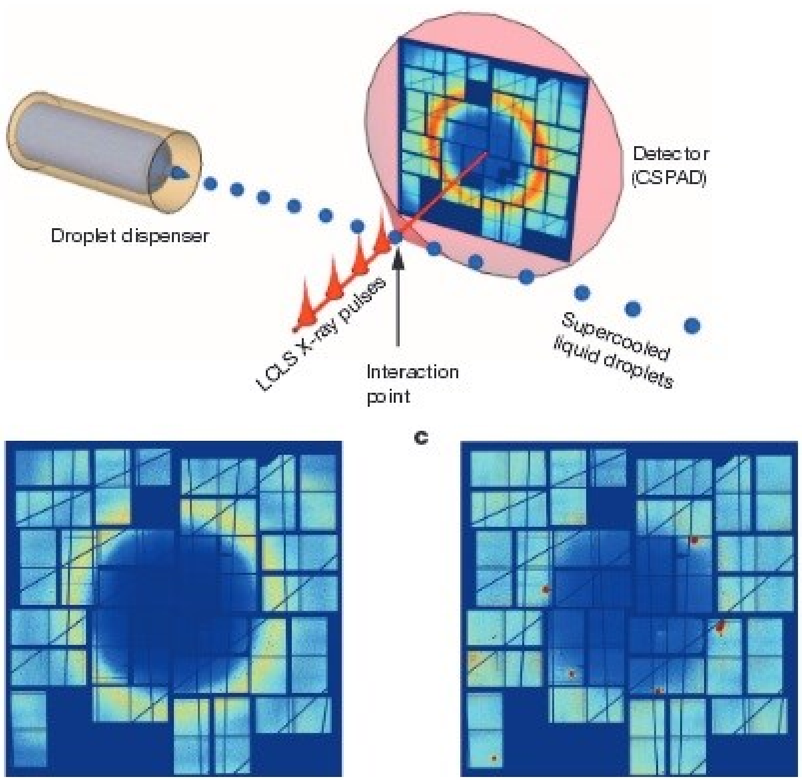
(x-ray) pump -- probe

1) Sellberg et al., Nature 510, 381 (2014)
2) Hruszkewycz et al., PRL 109,185(2012)
3) Gutt et al.,Thin Solid Films, 515, (2007)5532
4) Vaterlaus et al., PRL, 67, 3314 (1991)
5) Schmitt et al., Science 321, 5896 (2008)
6) Fritz et al., Science 315, 633 (2007)

Ultrafast Phenomena of interest for ultrafast XPCS

Atomic-scale dynamics in liquids and glasses on (sub) ns timescales

fluctuations in glassforming liquids….
water,…
liquid metals,..



towards undercooled regime:

- highfrequency dynamics ≈ 1 ps
- can reach 227 < T < 323 K in doplets
- local order

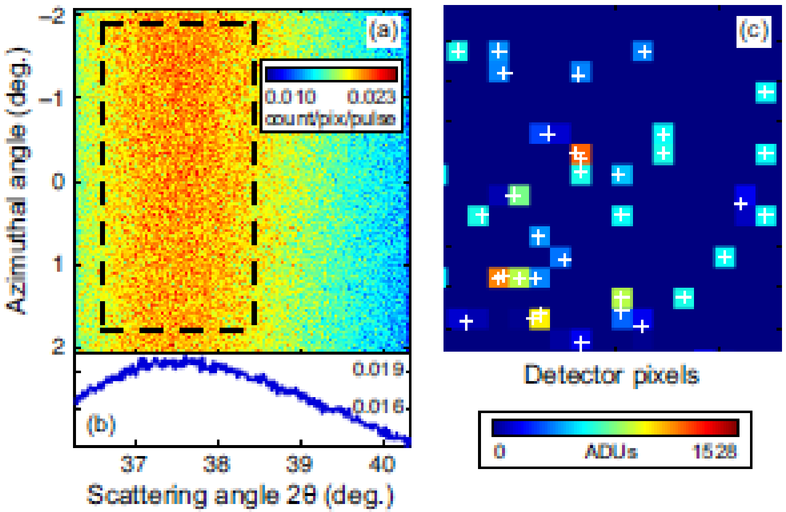
J. Sellberg et al., Nature 510, 381 (2014)

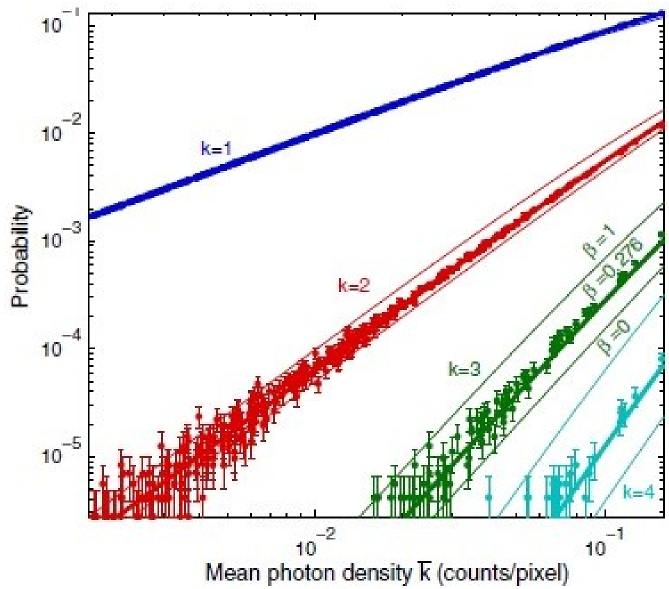
Ultrafast Phenomena of interest for ultrafast XPCS

Atomic-scale dynamics in liquids and glasses on (sub) ns timescales

fluctuations in glassforming liquids….
water,…
liquid metals,..

P(k) = P(k,M), M→ contrast ß





Liquid Gallium
(a) Average scattering integrated over 309 pulses. Det. at L = 37 cm from sample subtends angles including the main amorphous scattering ring at Q = 2.60 Å^{-1}.
(b) Average photon density vs. scattering angle.

ß = 0.276 ≈ ß^{calc}. = 0.307: Ga static on <70 fs timescale

S. O. Hruszkewycz, M. Sutton, P. H. Fuoss, B. Adams, S. Rosenkranz, K. F. Ludwig Jr., W. Roseker, D. Fritz, M. Cammarata, D. Zhu, S. Lee,, H. Lemke, C. Gutt, A. Robert, G. Grübel, and G. B. Stephenson, PRL 109(2012)1865502

Ultrafast Phenomena of interest for ultrafast XPCS

Surface fluctuations

capillary wave dynamics at large Q (beyond equilibrium hydrodynamics)

Table 1

Expected time constants and scattering intensities for capillary waves on liquid water using a delay line unit

k [$Å^{-1}$]	τ [s]	ΔE [meV]	Scattered photons per pulse	Scattered photons per second
10^{-3}	10^{-8}	$6.5\cdot10^{-5}$	4	$1.2\cdot10^{5}$
10^{-2}	$5\cdot10^{-10}$	$1.3\cdot10^{-3}$	$4\cdot10^{-2}$	$1.2\cdot10^{3}$
10^{-1}	$2.5\cdot10^{-11}$	$2\cdot10^{-2}$	$4\cdot10^{-4}$	$1.2\cdot10^{1}$
1	10^{-12}	$6.5\cdot10^{-1}$	$4\cdot10^{-6}$	$1.2\cdot10^{-1}$

Intensities are calculated for 50×50 μm^2 pixel size.

Water

≈ 1 ps

Expected time constants and scattering intensities for capillary waves on liquid mercury using a delay line unit

k [$Å^{-1}$]	τ [s]	ΔE [meV]	Scattered photons per pulse	Scattered photons per second
10^{-3}	$2\cdot10^{-10}$	$3.3\cdot10^{-3}$	$6\cdot10^{-2}$	$1.8\cdot10^{3}$
10^{-2}	$1\cdot10^{-11}$	$6.5\cdot10^{-2}$	$6\cdot10^{-4}$	$1.8\cdot10^{3}$
10^{-1}	$5\cdot10^{-13}$	1.3	$6\cdot10^{-6}$	$1.8\cdot10^{-1}$
1	$5\cdot10^{-14}$	1.3	$6\cdot10^{-8}$	$1.8\cdot10^{-3}$

Intensities are calculated for 50×50 μm^2 pixel size.

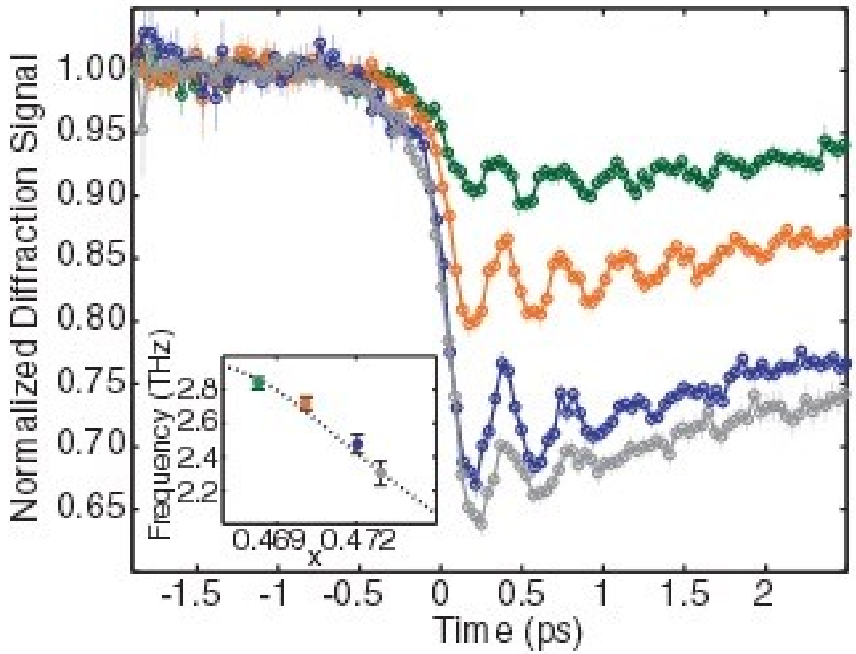
Mercury

≈ 50 fs

C. Gutt, O. Leupold, G. Grübel
Thin Solid Films, 515, (2007)5532

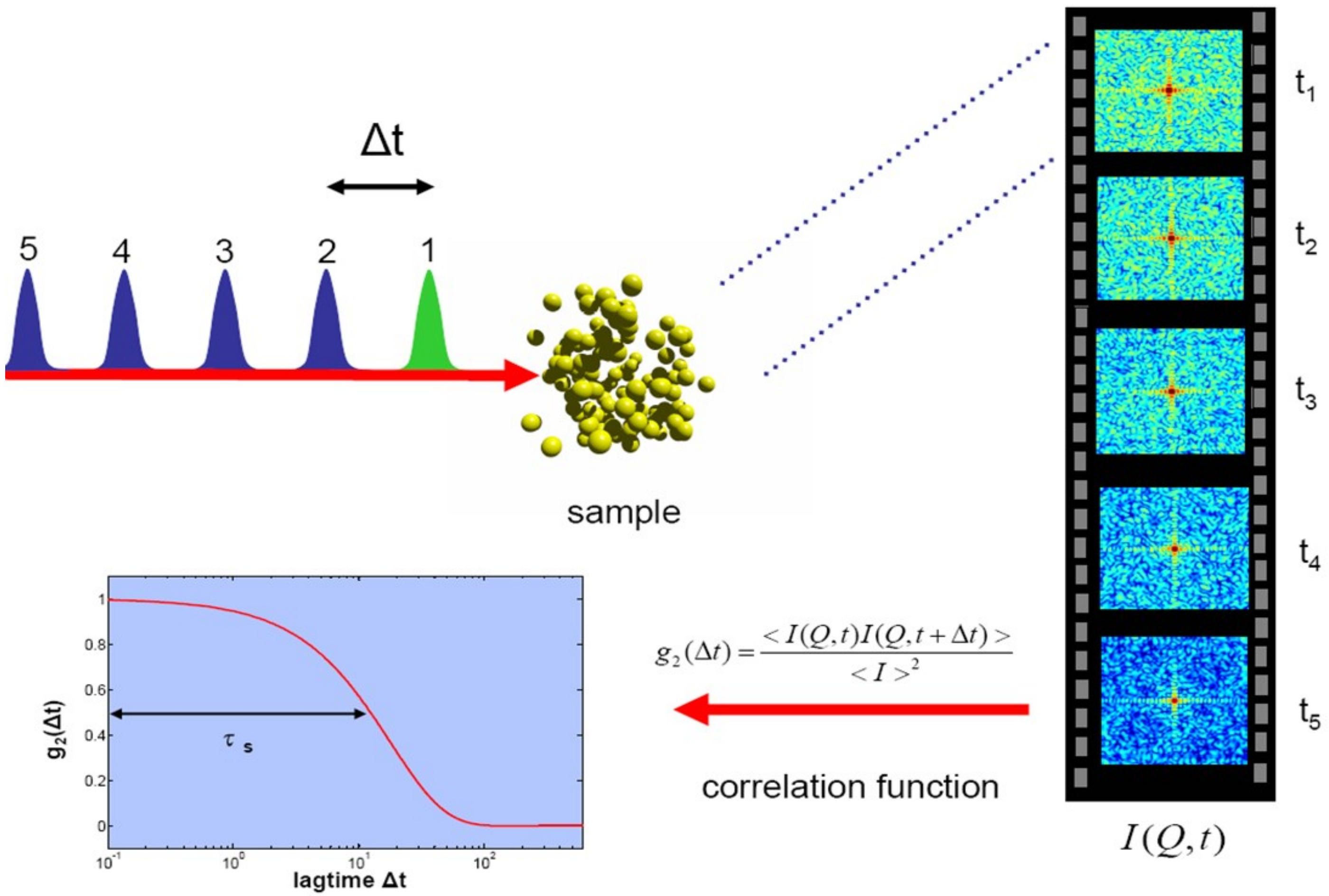
Coherent optical phonons in Bismuth

Fig. 1. Bismuth (111) x-ray diffraction efficiency as a function of time delay between the optical excitation pulse and x-ray probe for excitation fluences of 0.7 (green), 1.2 (red), 1.7 (blue), and 2.3 mJ/cm^2 (gray). The zero-delay point was set at the half maximum of the initial transient drop. The inset displays the optical phonon frequency as a function of the normalized atomic equilibrium position along the body diagonal of the unit cell *x* as measured by x-ray diffraction. The dotted curve represents the theoretical prediction obtained from DFT calculations of the excited-state potential-energy surface (*10*).



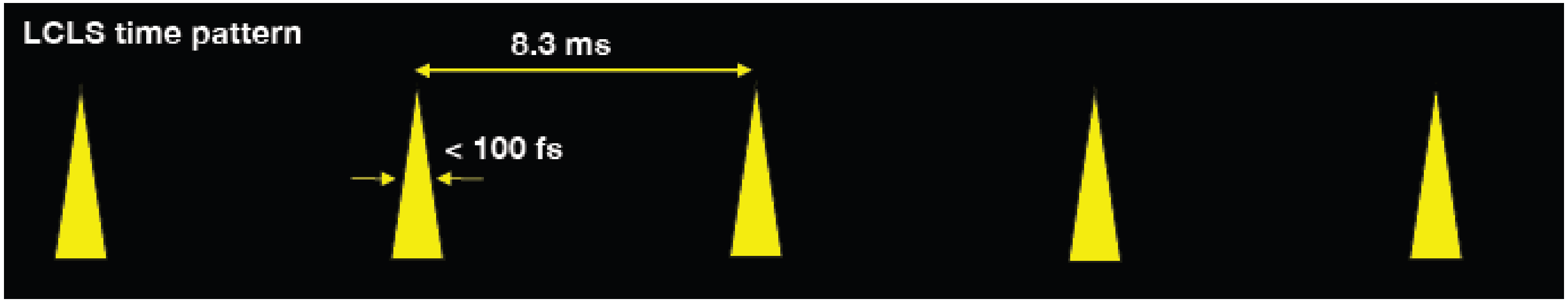
Fritz et al., Science 315, 633 (2007)

How to measure dynamics with an FEL?

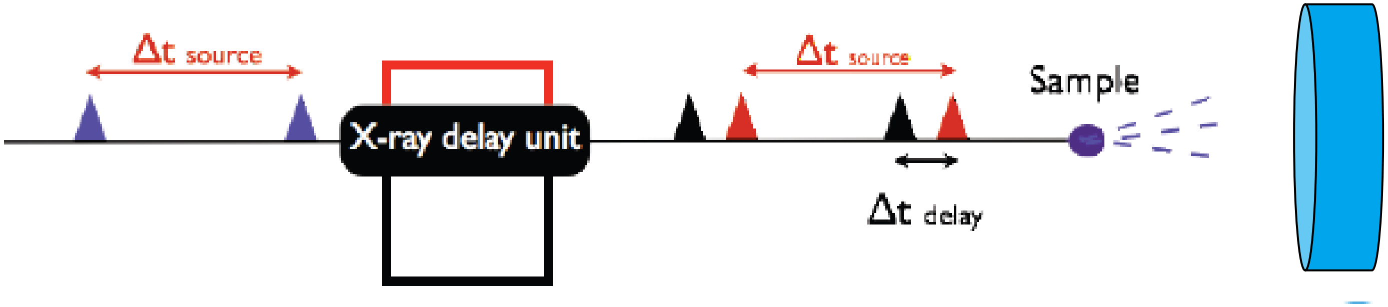


How to measure sub-ns dynamics with an FEL?

<u>Pulse spacing:</u> 8.3 ms (LCLS), 16.6 ms (SACLA), 200 ns during 0.6 ms (E-XFEL)



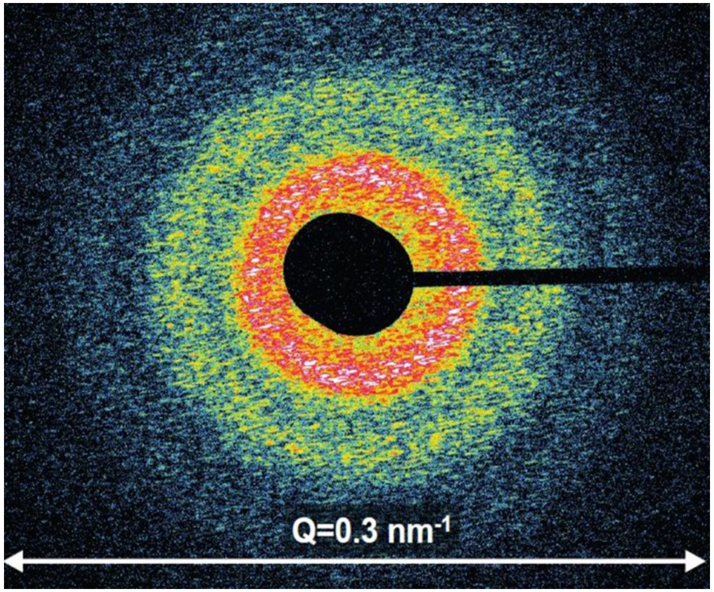
The solution is to split each single XFEL pulse into two and provide a suitable delay time between the photon pulses at the sample position independently of the time structure of the source.



permits also x-pump x-probe

Compatible 2D detector

The importance of contrast



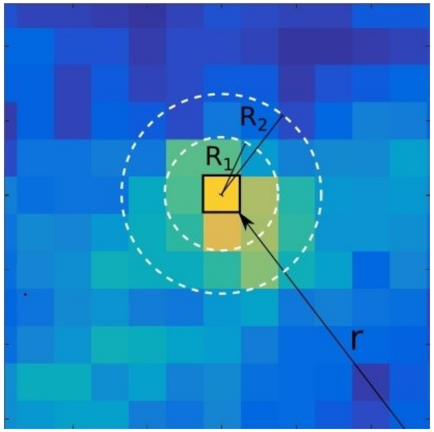
Compare n speckle patterns:

Speckle size: λ/d

Contrast: $\beta = \beta_t \ \beta_l(q)$

$\beta_t = \sqrt{(\mathrm{var}(I)/\langle I \rangle)} = 1/\sqrt{M} = \gamma(R=0)$

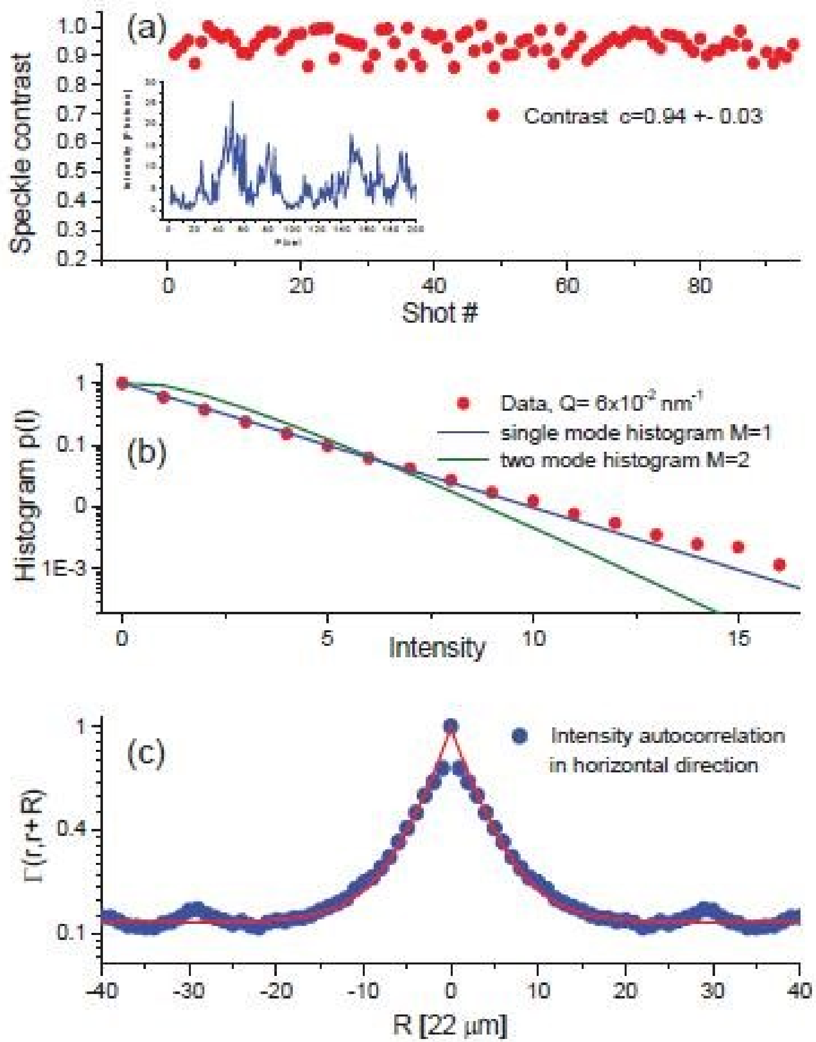
M mode-number
γ mutual coherence function



$$P(I) = \frac{\Gamma(I+M)}{\Gamma(M)\Gamma(I+1)} \ (1+M/\langle I \rangle)^{-I} \ (1+\langle I \rangle/M)^{-M}$$

$$g(R) = \langle I(r+R)\ I(r) \rangle / \langle I(r) \rangle^2 = |\ \gamma(R)\ |^2$$

The LCLS case (monochromatic)



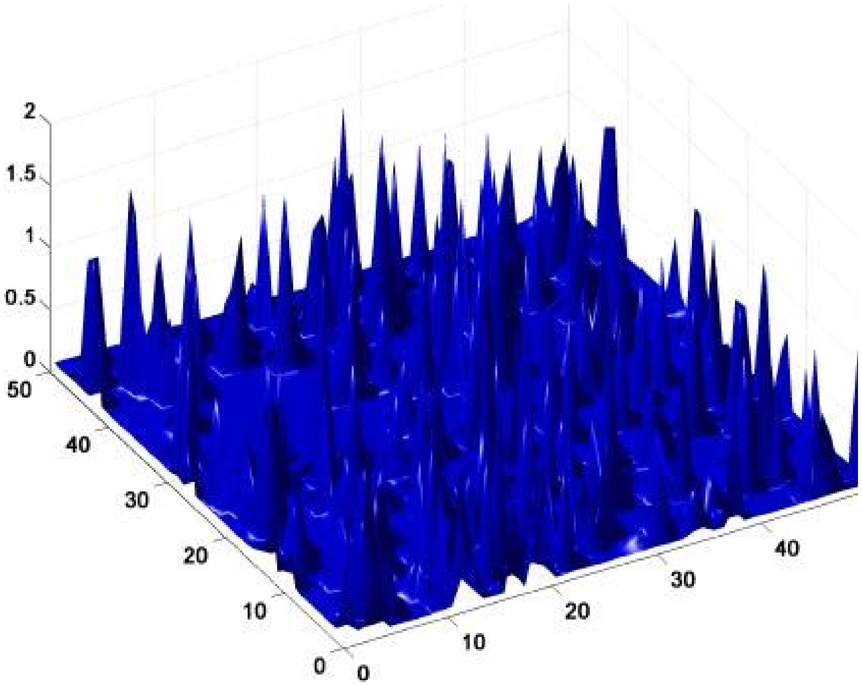
$ß_t$ = 0.94(3)

$$P_M(I) = \frac{M^M I^{M-1}}{\Gamma(M) < I >^M} e^{-i\frac{MI}{<I>}}$$

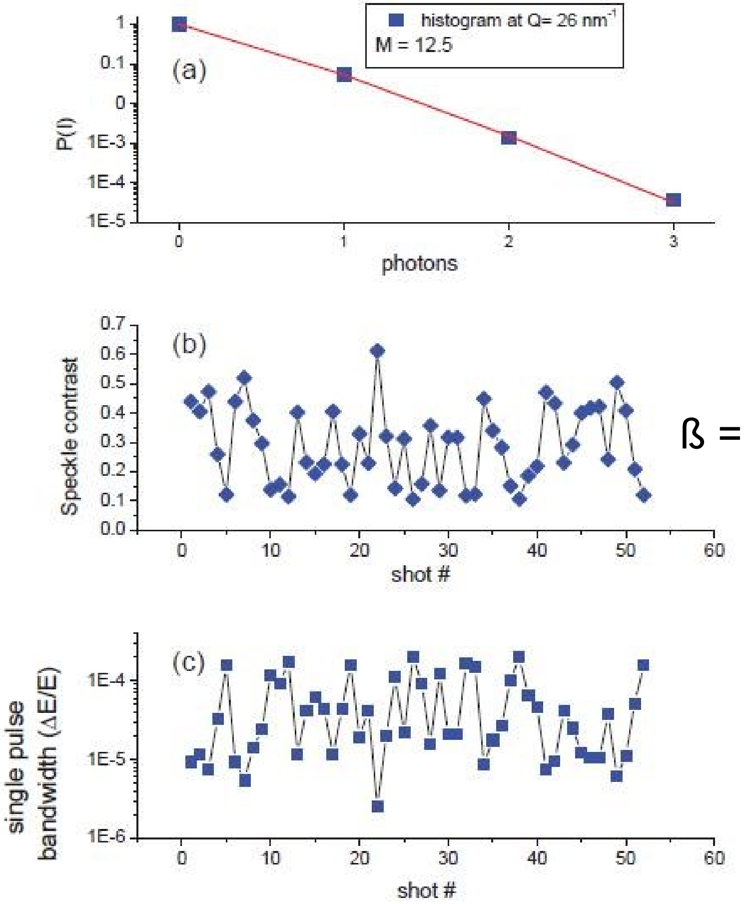
9 x 3.3 μm^2 beam

Gutt et al., PRL 108, 024801 (2012)

The first large Q attempt

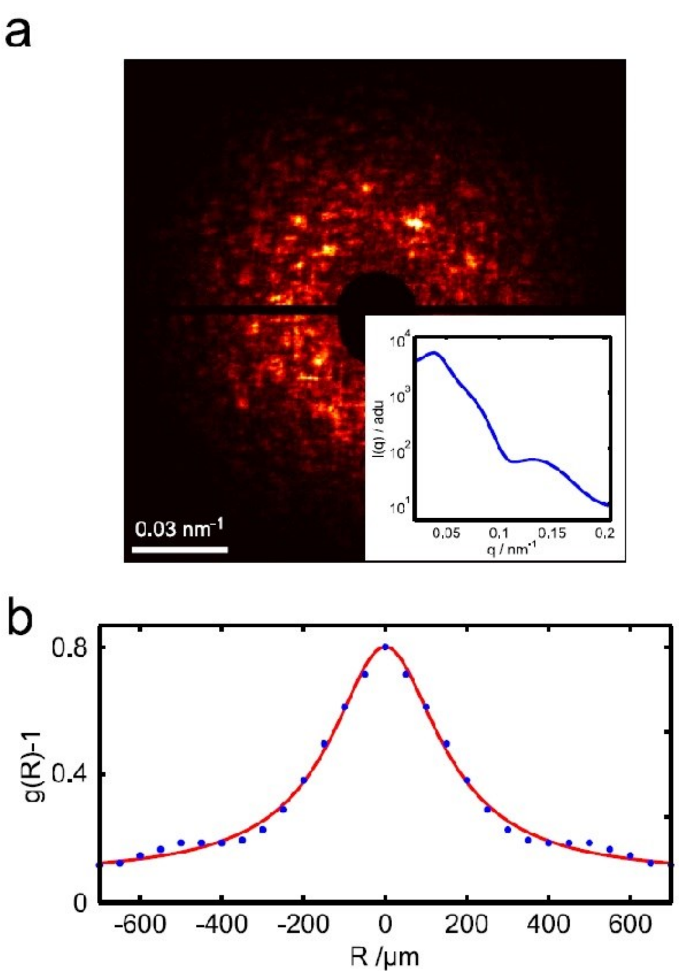


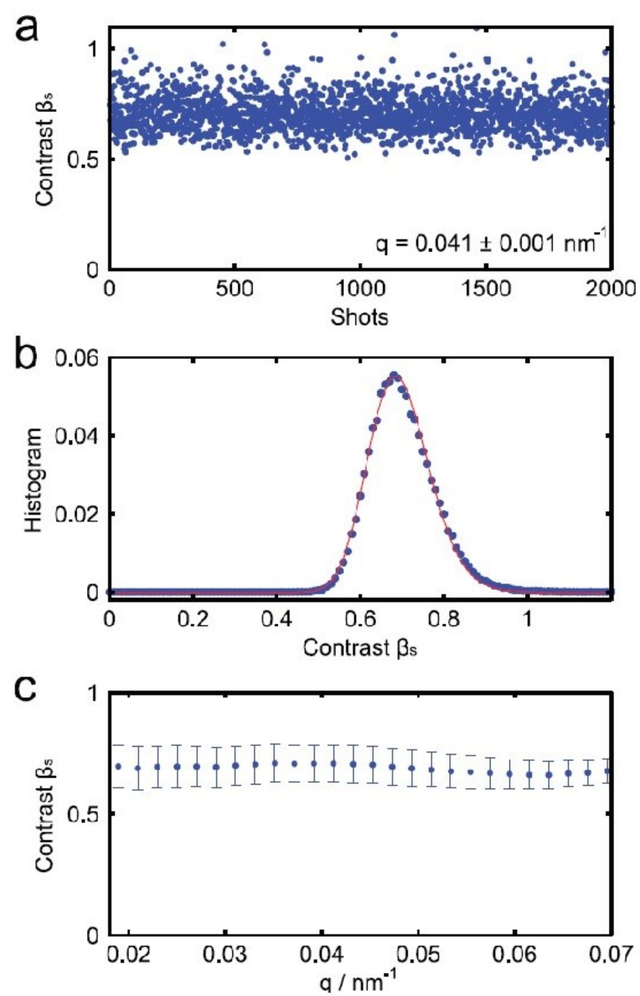
Single pulse speckle pattern from a Au nano-powder recorded at the Au(111) diffraction peak at Q=26 nm^{-1}



ß = 0.26(2)

The SACLA case (pink)





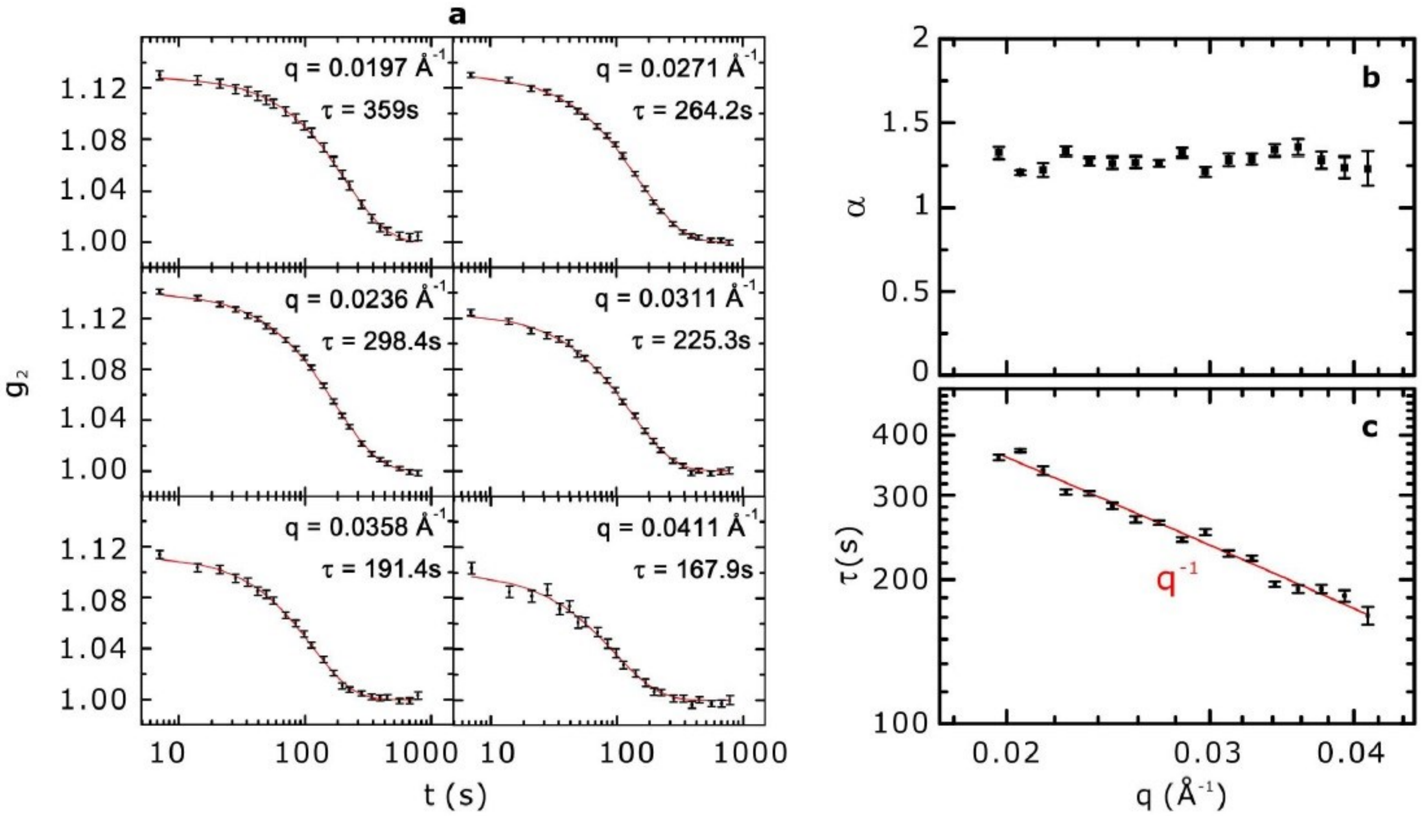
$\beta = 0.70(8)$

$\beta_{corr} =$
$\beta_t = 0.79(9)$

Lehmkuehler et al., Scientific Reports, 4, 5234; DOI:10.1038/srep05234 (2014)

Movie Mode @ LCLS

Dynamics of nanoparticles in polymer melt



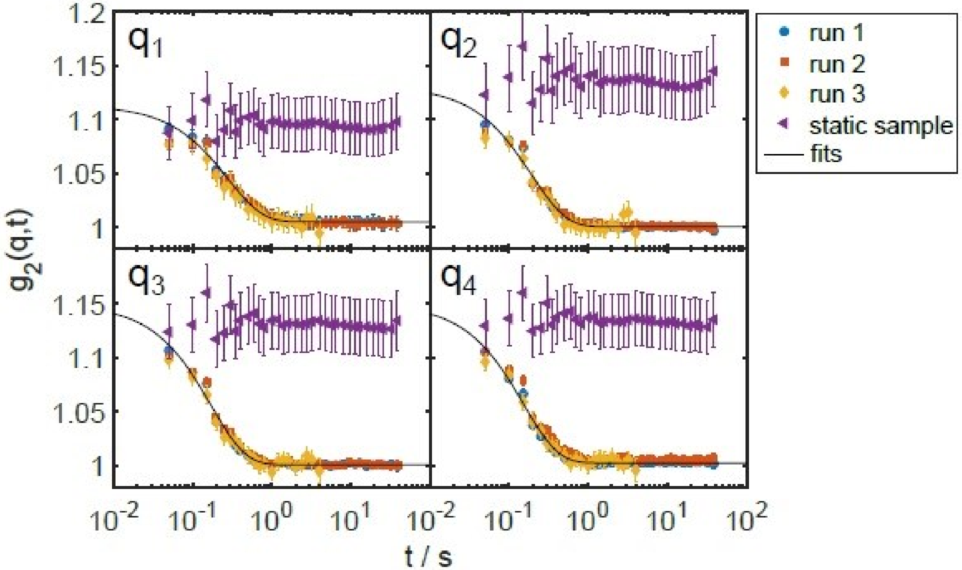
PI CCD with 7 s readout time

ß^{dyn}: 0.08 – 0.12

Carnis et al., Scientific Reports, 4, 6017;
DOI 10.1038/srep0617 (2014)

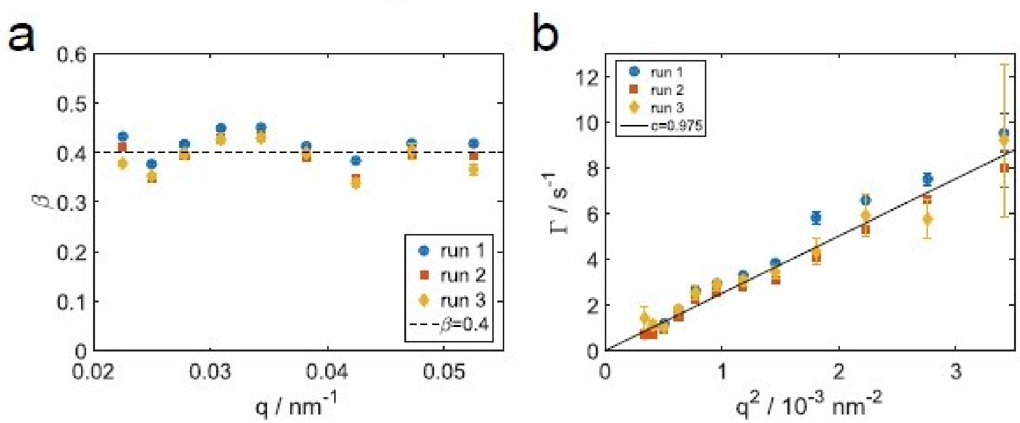
Movie mode @ SACLA

Dynamics of nanoparticles in glycerol/water



MPCCD in 20 Hz mode

ß^{dyn} : 0.09 – 0.13



Lehmkuehler et al., to be published

Static vs. dynamic contrast

LCLS

8.96 keV
monochromatic $\Delta E/E = 1.4 \times 10^{-4}$

ß = 0.94(3)

$ß_{large\ Q} = 0.26(2)$

$\tau_c = 2(1)$ fs

$g_2(t)$ contrast: 0.08 – 0.12

$\mathbf{ß}^{\mathbf{dyn}} = \mathrm{sqrt}(g_2(t)) = \mathbf{0.28\text{-}0.34}$

$\mathbf{ß}^{\mathbf{dyn}} \approx \mathbf{0.3}$

$\mathbf{ß}^{\mathbf{dyn}} / \mathbf{ß} \approx \mathbf{3}$

SACLA

8 keV
pink $\Delta E/E = 5 \times 10^{-3}$

ß = 0.70(8) $\mathbf{ß_{corr} = 0.79(9)}$

$\tau_c = 0.1$ fs

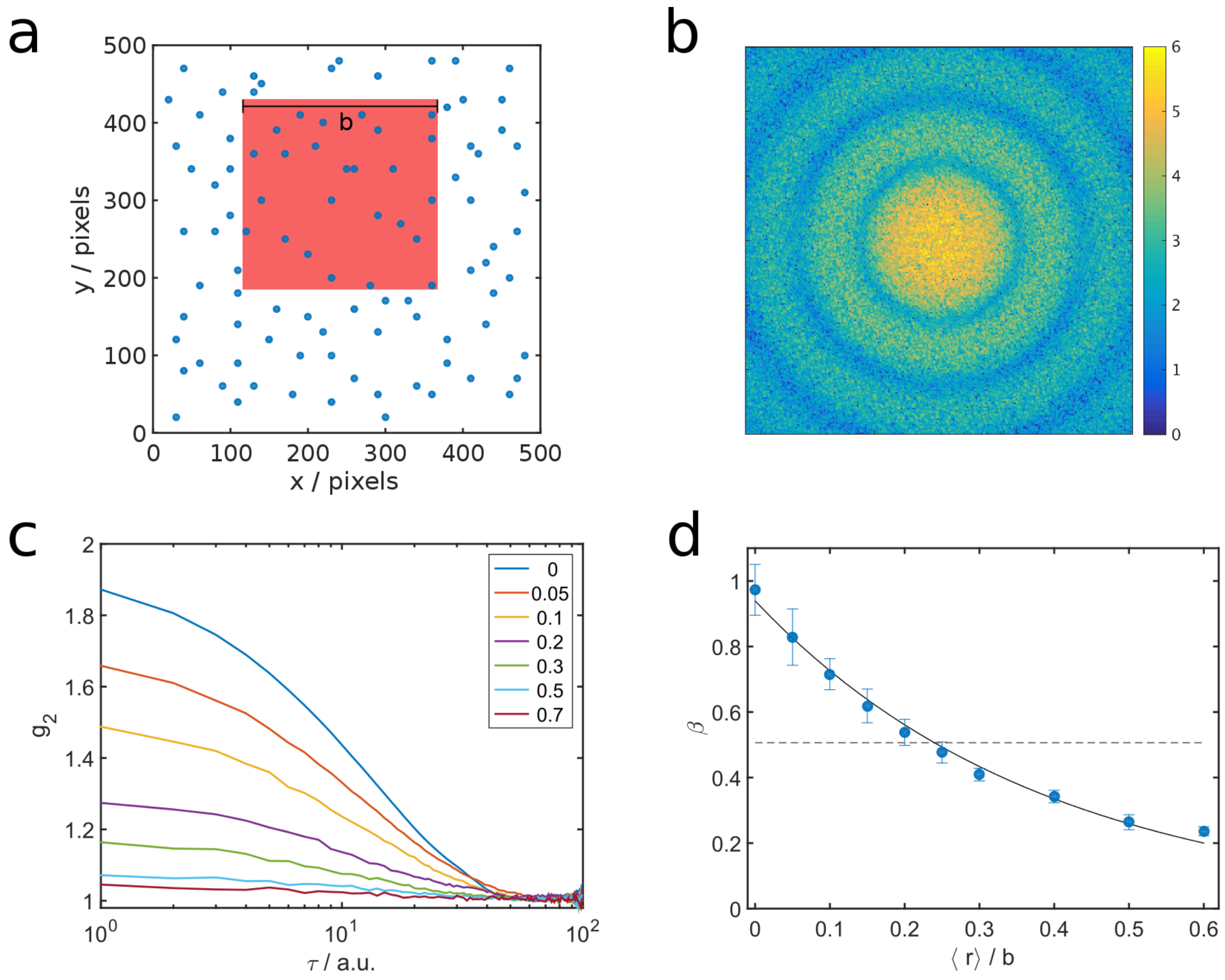
$g_2(t)$ contrast: 0.09 – 0.13

$\mathbf{ß}^{\mathbf{dyn}} = \mathrm{sqrt}(g_2(t)) = \mathbf{0.28\text{-}0.36}$

$\mathbf{ß}^{\mathbf{dyn}}_{\mathbf{corr}} = \mathbf{0.40}$

$\mathbf{ß}^{\mathbf{dyn}} / \mathbf{ß} \approx \mathbf{2}$

Simulations: shot-to-shot movements ?



contrast as a function of

mean displacements <r> in units of the beamsize b

Questions I:

How is a superconducting machine doing in terms of shot-to-shot contrast ?

NOTE: Single shot contrast looks good.

Is movie-mode feasible?

Detectors for movie mode?

How to measure sub-ns dynamics with an FEL?

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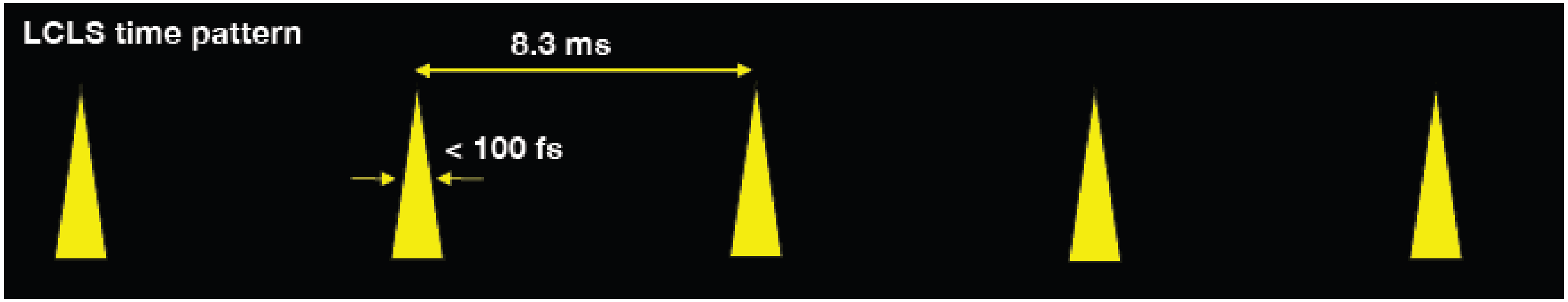

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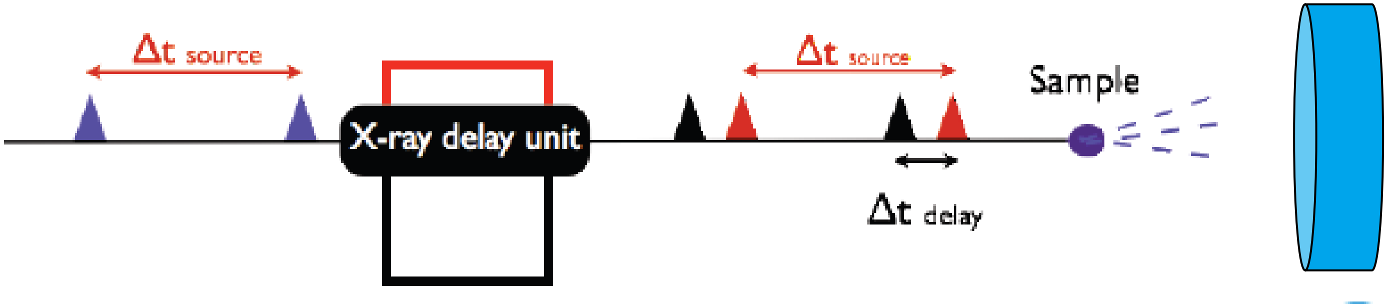
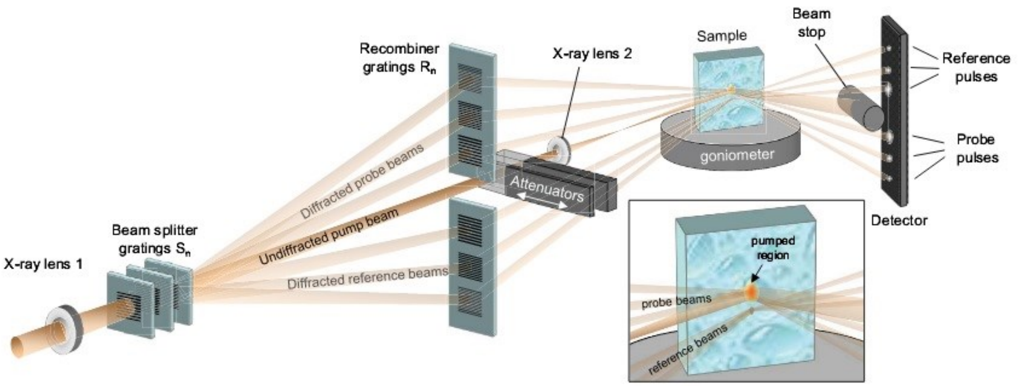


permits also x-pump x-probe

Compatible 2D detector

Split-and Delay concepts

- **Wavefront splitting (used in soft x-ray regime)**
- **Diffraction gratings (Δt < ps)**



David et al., Sci. Rep. 5, 7644; DOI:10,1038/srep0764(2015)

- **Amplitude splitting (0 < Δt < 3 ns)**

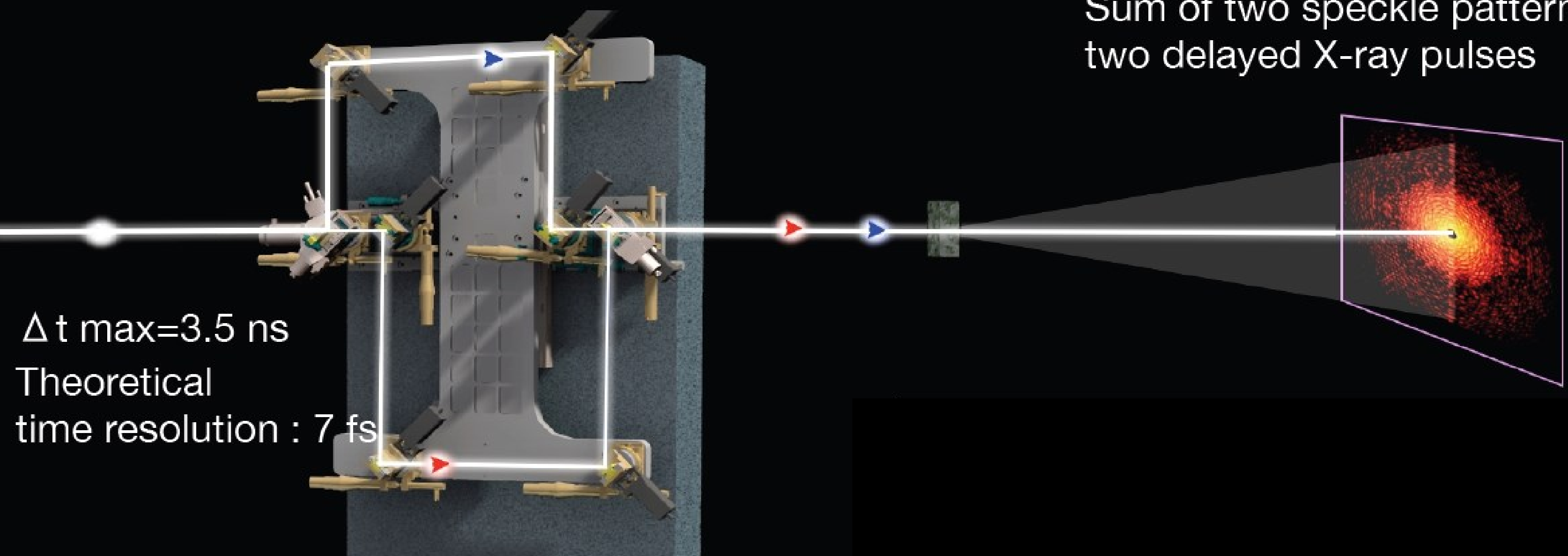
Roseker et al. Optics Letters, 34/12, 1768 (2009); J. Synchr. Rad. 18, 481 (2011); *Proc. SPIE* 8504, X-Ray Free-Electron Lasers: 85040I (October 17, 2012); doi:10.1117/12.929759

Split-and-delay XPCS

E = 7.9 keV
E = 8.4 keV
E = 12.4 keV

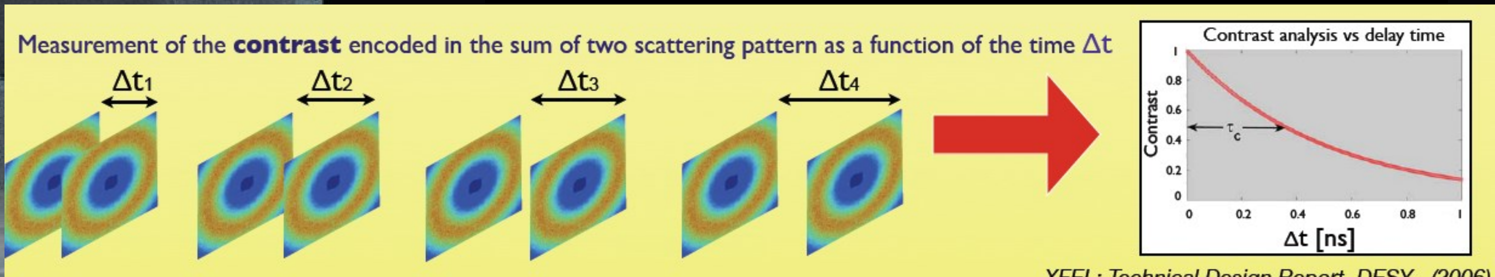
Eight Silicon perfect crystals arranged in 90° geometry

Sum of two speckle patterns from the two delayed X-ray pulses



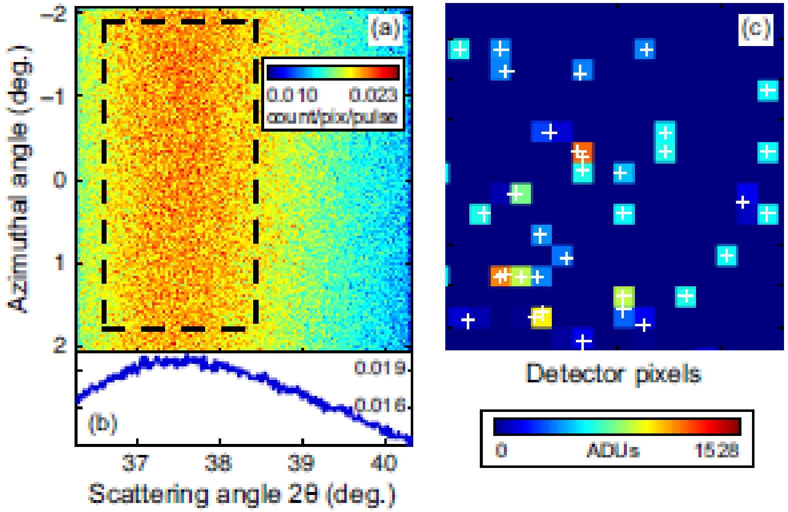
Δt max=3.5 ns
Theoretical time resolution : 7 fs

Beam splitting is accomplished using 20 um thin Si crystals



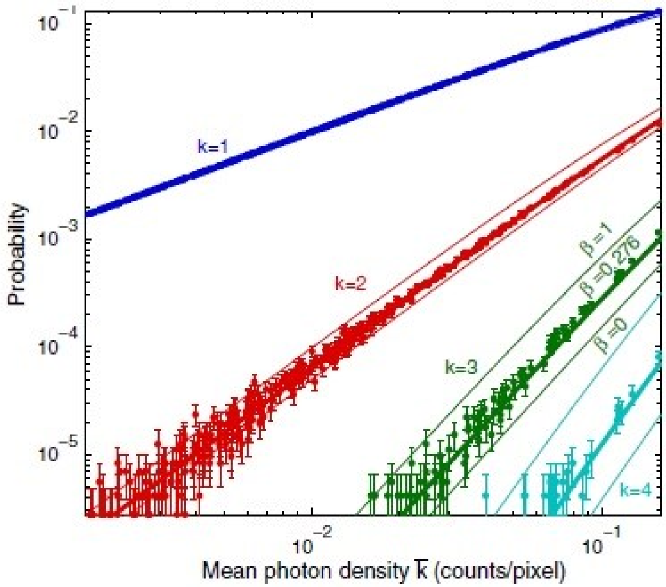
XFEL: Technical Design Report, DESY (2006)

Contrast Analysis in the low intensity limit



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

k	number of photons per pixel
<k>	mean photon density/pixel
M	number of modes
ß = 1/M	contrast



Liquid Gallium

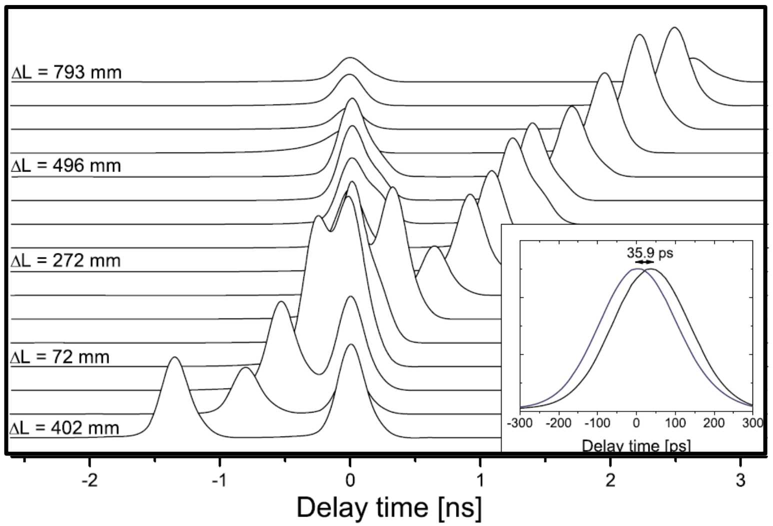
(a) Average scattering integrated over 309 pulses. Det. at L = 37 cm from sample subtends angles including the main amorphous scattering ring at Q = 2.60 $Å^{-1}$.

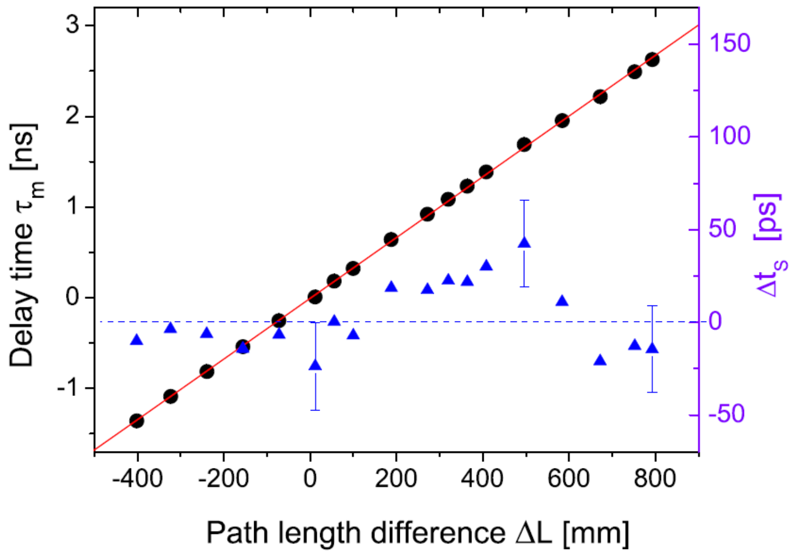
(b) Average photon density vs. scattering angle.

S. O. Hruszkewycz, M. Sutton, P. H. Fuoss, B. Adams, S. Rosenkranz, K. F. Ludwig Jr., W. Roseker, D. Fritz, M. Cammarata, D. Zhu, S. Lee,, H. Lemke, C. Gutt, A. Robert, G. Grübel, and G. B. Stephenson, PRL 109(2012)1865502

Delay time measurements (ESRF)

Measure delayed pulses vs storage ring bunch clock



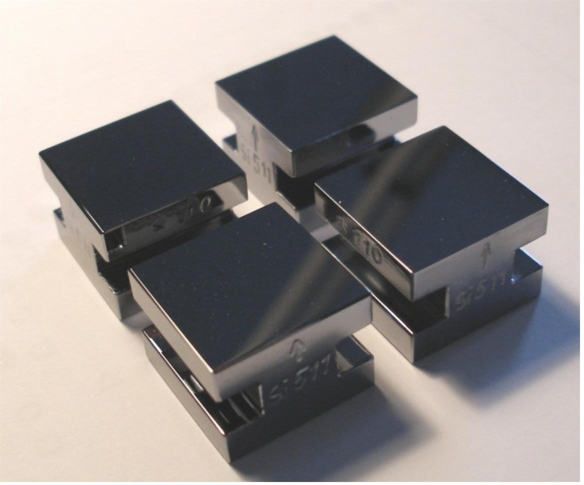


Roseker et al. Optics Letters, 34/12, 1768 (2009);
J. Synchr. Rad. 18, 481 (2011)

Δt_{max} = 2.62 ns
Δt_{res} = 16.7 ps

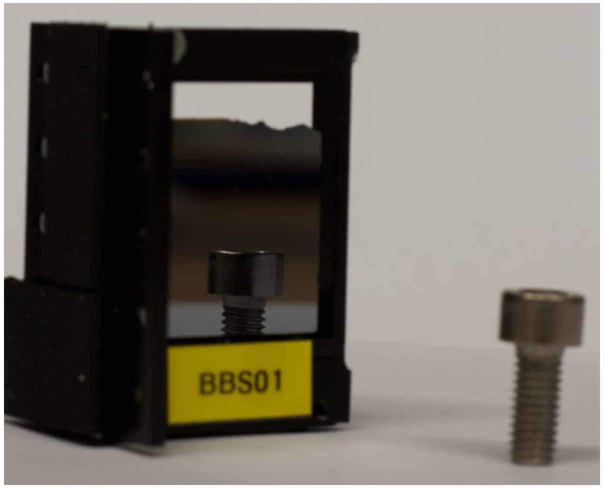
Crystal optics

Bragg optics employed in the delay line



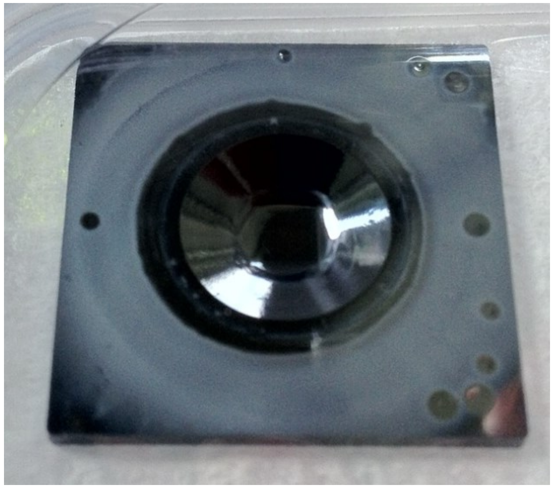
Thickness: 5mm

Beam splitter developed at HASYLAB



Wedge shape: 20-200 μm

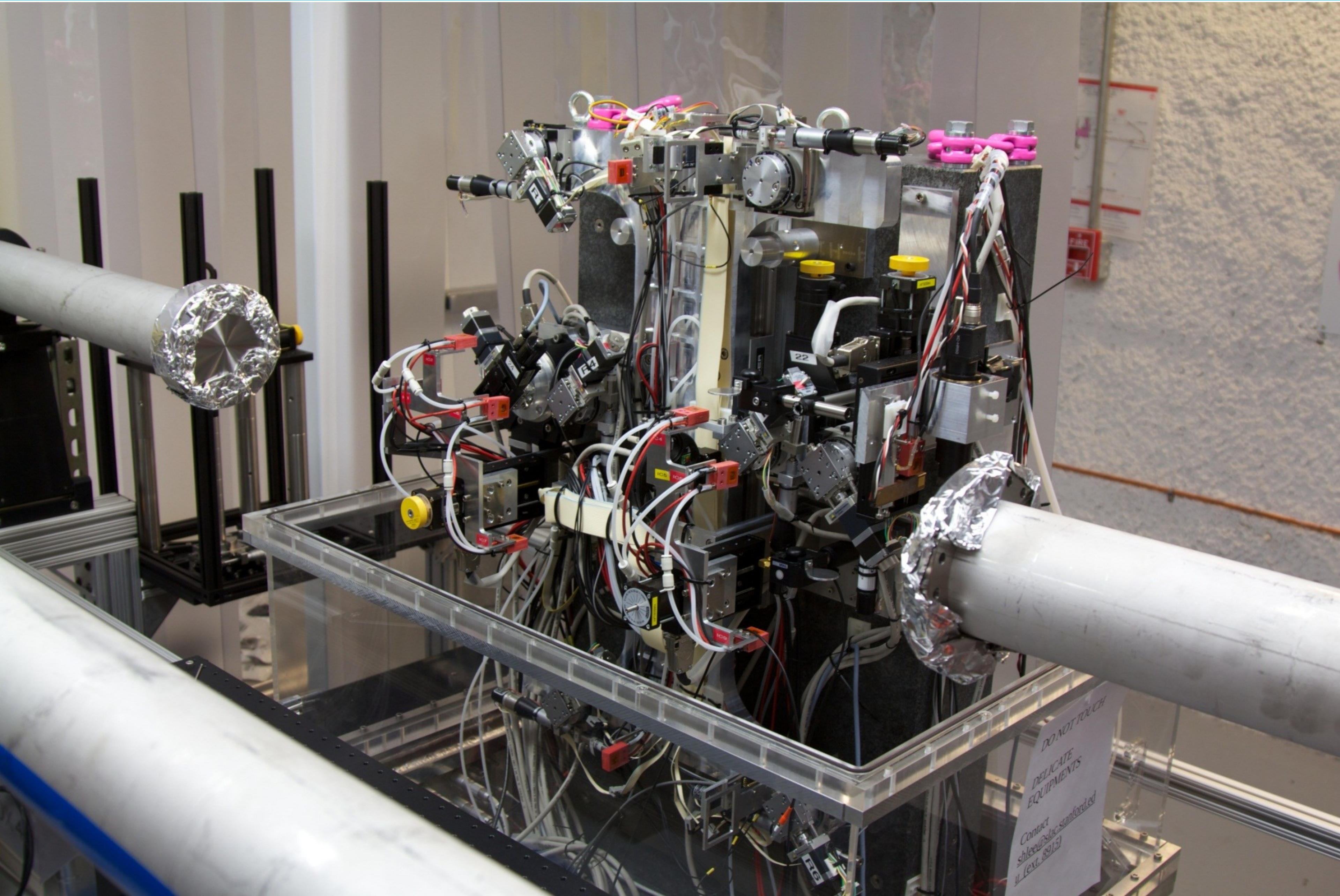
Beam splitter developed at SPring-8



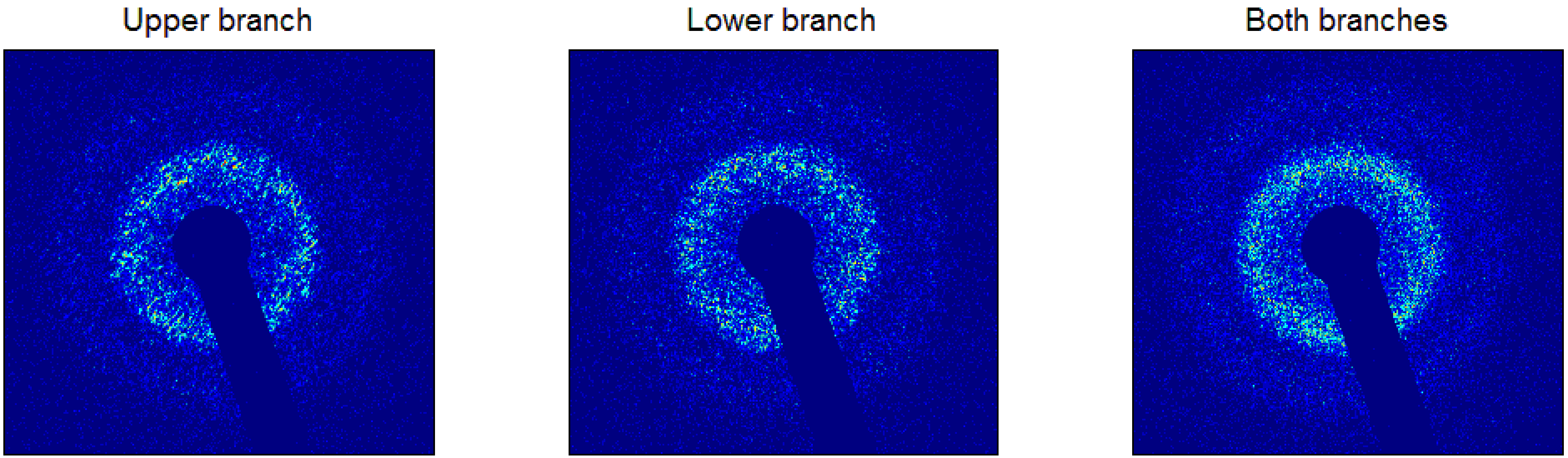
10-15 μm membrane

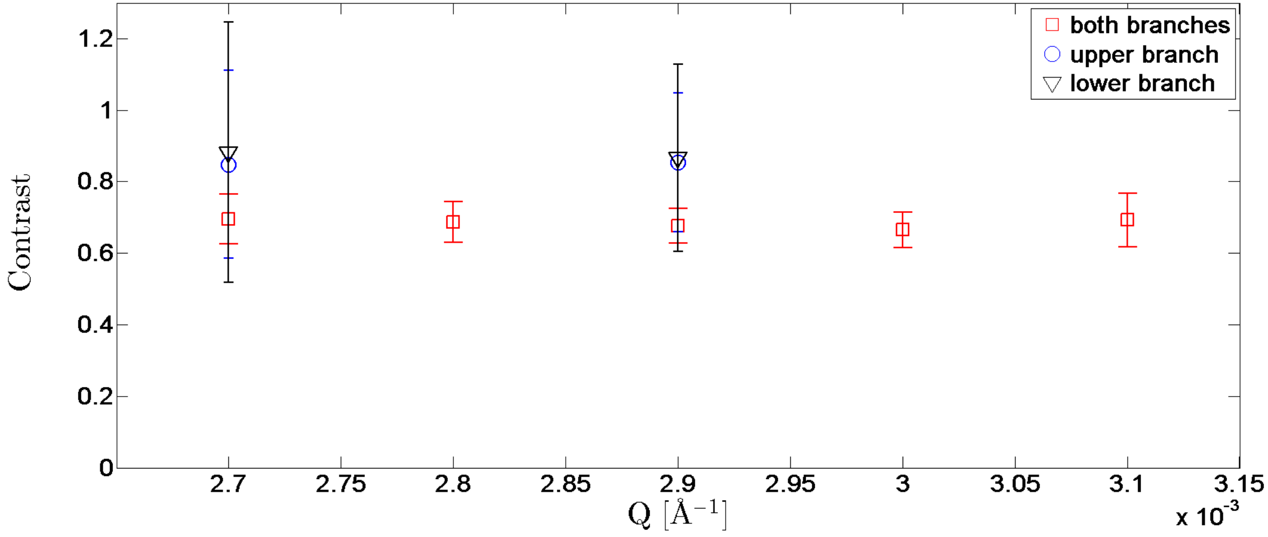
Courtesy: Makina YABASHI

Delay Line at XCS - LCLS



(Single shot) speckle images and contrast

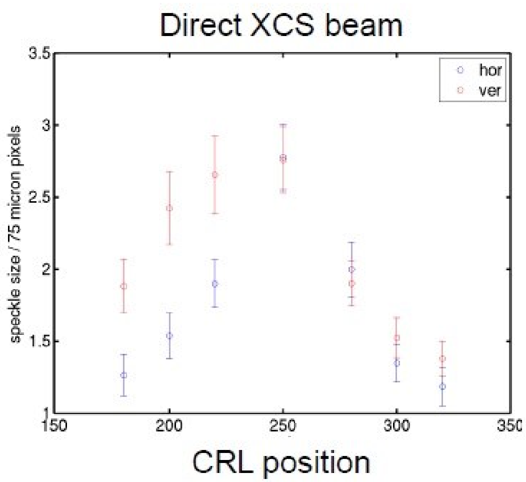


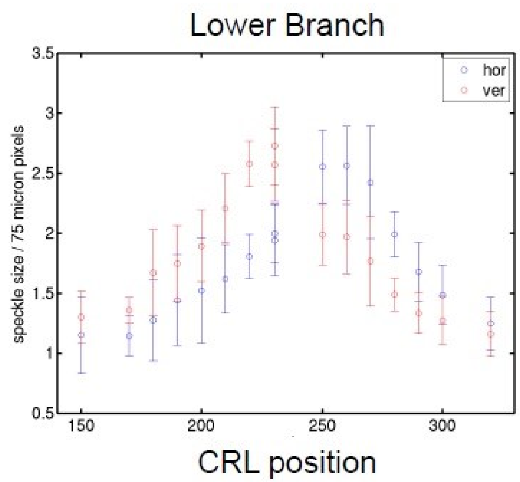


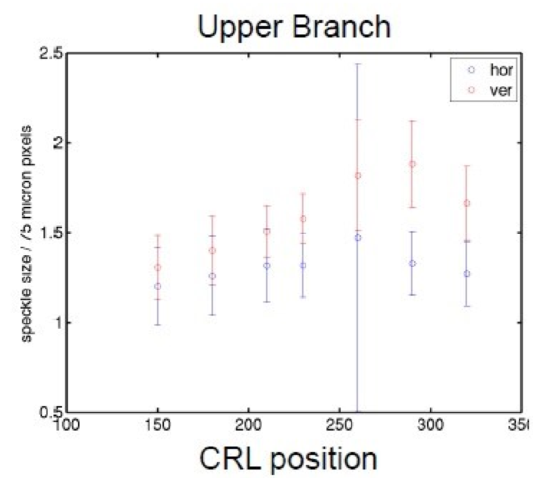
Dried PMMA colloids

Focussing

Be aware of astigmatism !!

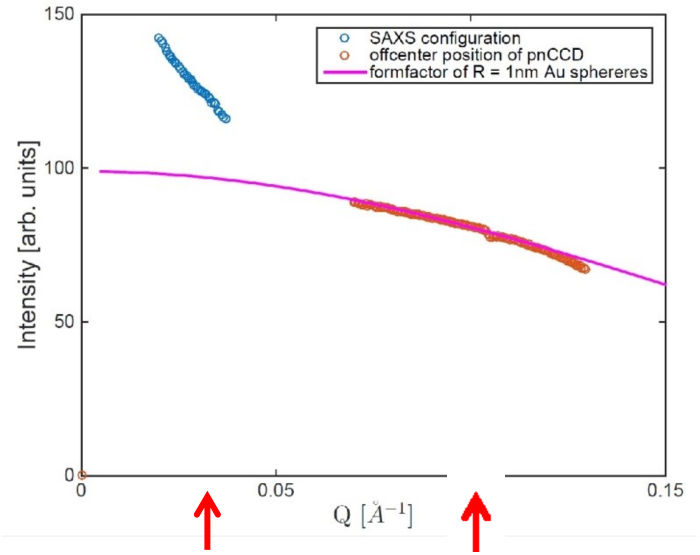


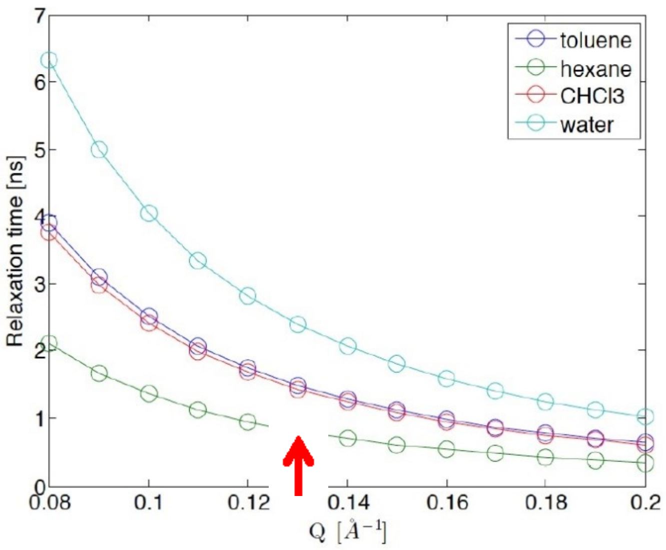




A prototype experiment (XCS/LCLS)

Colloidal gold particles (R=1nm) in hexane
Set delay line to Δt = 1.3 ns



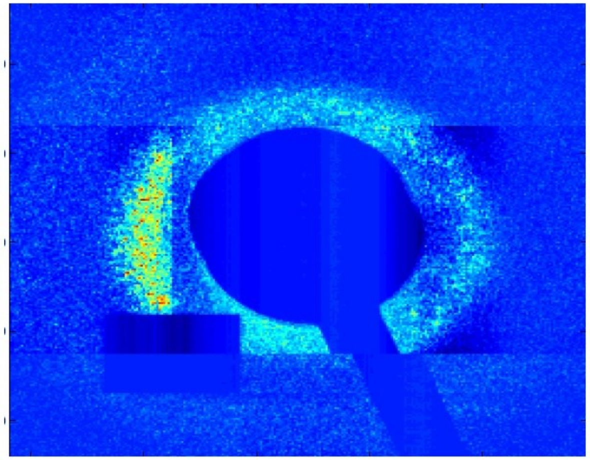


Take data at 0.035 $Å^{-1}$ (static) and 0.13 $Å^{-1}$ (dynamic)

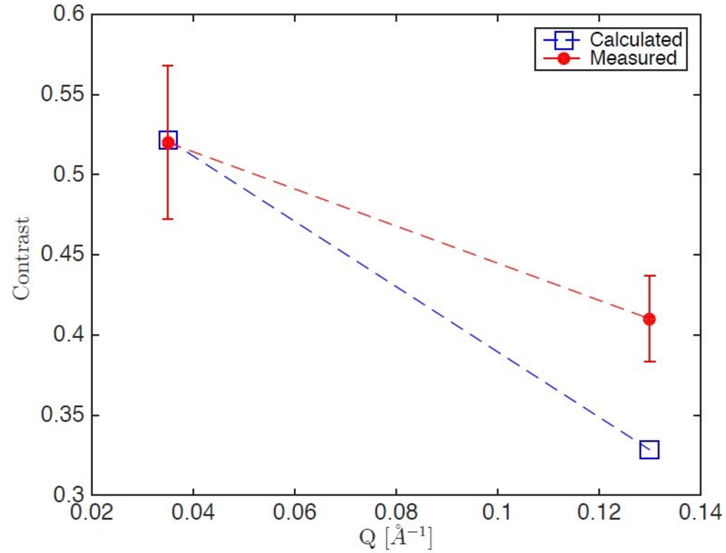
Take 21×10^4 frames and 27×10^4 frames
About 10% of the frames show 1:1 splitting ratio

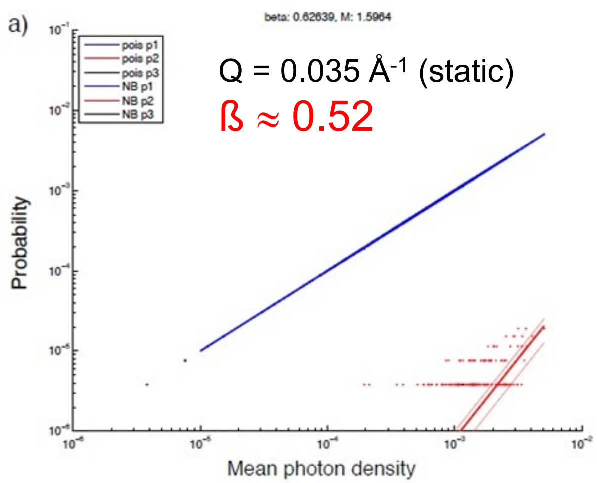
Expect 0.05 ph/pulse/speckle (@5×10^7 ph/pulse)
Get 0.003 ph/pulse/speckle

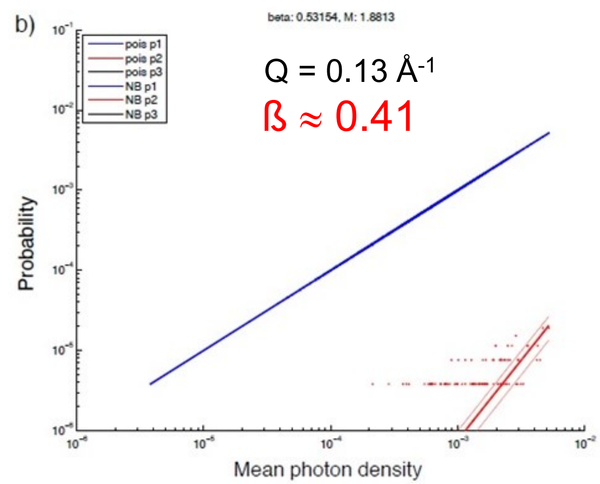
A prototype analysis: the first 2 points (Δt=1.3 ns)



Use droplet algorithm on 1:1 split frames
pnCCD (1024x2024); 4.3 m, 75 micron pixel







Split-and Delay:

The concept "works"

Split- and delay device is a pre-requisite for fast XPCS at E-XFEL

overcome limitations of 1st generation device (variable energy)
need optics effort (thin X-tals)
commissioning time
implementation in control system
delicate analysis of week data sets

Need performing beamline optics and diagnostics

non bw related losses are inacceptable
reliable focussing scheme
avoid pointing instabilities

Need performing pulse-picking schemes

combine with beam multiplexing

Need XPCS detector

the specs have been discussed before !

Need performing DAC system with rapid user access

Plan for “day 1” experiments:

Coherence characterization

single shot contrast (ΔE/E), flux, focus, positional stability, bunch length…

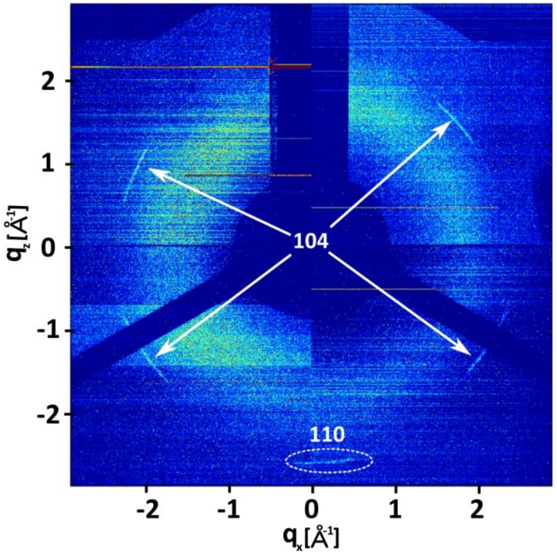
Movie mode experiment

Prototype (x-ray pump) - (x-ray probe)

will design small local setup for “day 1”

Early single shot jet experiment

can provide jet for “day 1+”



Hematite spindles in water droplets (sum of 2000 single frames)

The end