Coherence of light and matter: from basic concepts to modern applications

Part II Script 4

Vorlesung im GrK 1355

WS 2013

A. Hemmerich & G. Grübel

Location: SemRm 052, Gebäude 69, Bahrenfeld

Thursdays 12.15 – 13.45

G.Grübel (GR), A.Hemmerich (HE)

Literature

Basic concepts: The quantum theory of light
Rodney Loudon, Oxford University Press (1990)
Quantum Optics
Marlan O. Scully, M. Suhal Zubairy, Cambridge University Press (1997)
Dynamic Light Scattering with Applications
B.J. Berne and R. Pecora, John Wiley&Sons (1976)
Elements of Modern X-Ray Physics
J. A. Nielsen and D. McMorrow, J. Wiley&Sons (2001)

Matter Waves: Bose-Einstein Condensation in Dilute Gases
C. J. Pethick and H. Smith, Cambridge University Press (2002)

Lecture Notes

Part I: ***http://photon.physnet.uni-hamburg.de/fileadmin/user_upload/ILP/Hemmerich/teaching.html/Coherence.pdf***

Part II: **http://photon-science.desy.de/research/studentsteaching/lectures__seminars/ws_13_14/coherence_of_light_grk1355/…..**

▪ Coherence of light and matter: from basic concepts to modern applications

Part II: G. Grübel

Coherence based X-ray techniques

Overview, Introduction to X-ray Scattering, Sources of Coherent X-rays, Speckle pattern and their analysis

Imaging techniques

Phase Retrieval, Sampling Theory, Reconstruction of Oversampled Data, Fourier Transform Holography, Applications

X-ray Photon Correlation Spectroscopy (XPCS)

Introduction, Equilibrium Dynamics (Brownian Motion), Surface Dynamics, Non-Equilibrium Dynamics

Imaging and XPCS at FEL Sources

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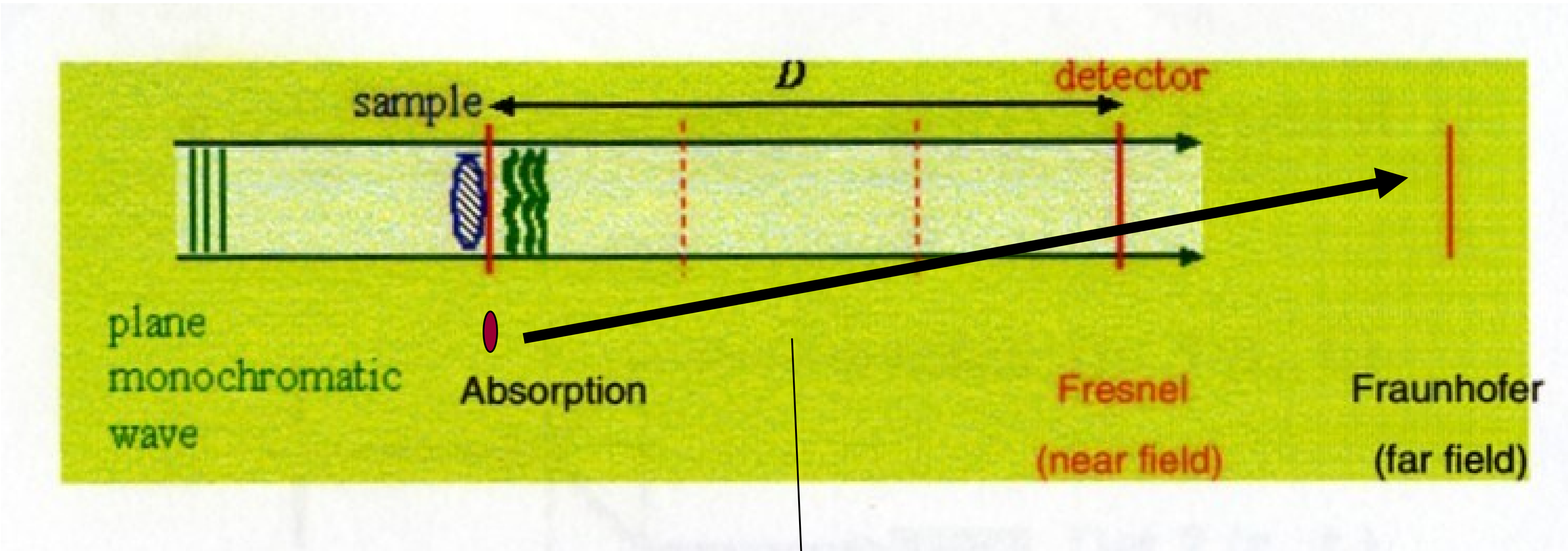
Imaging techniques:

Fourier Transform Holography

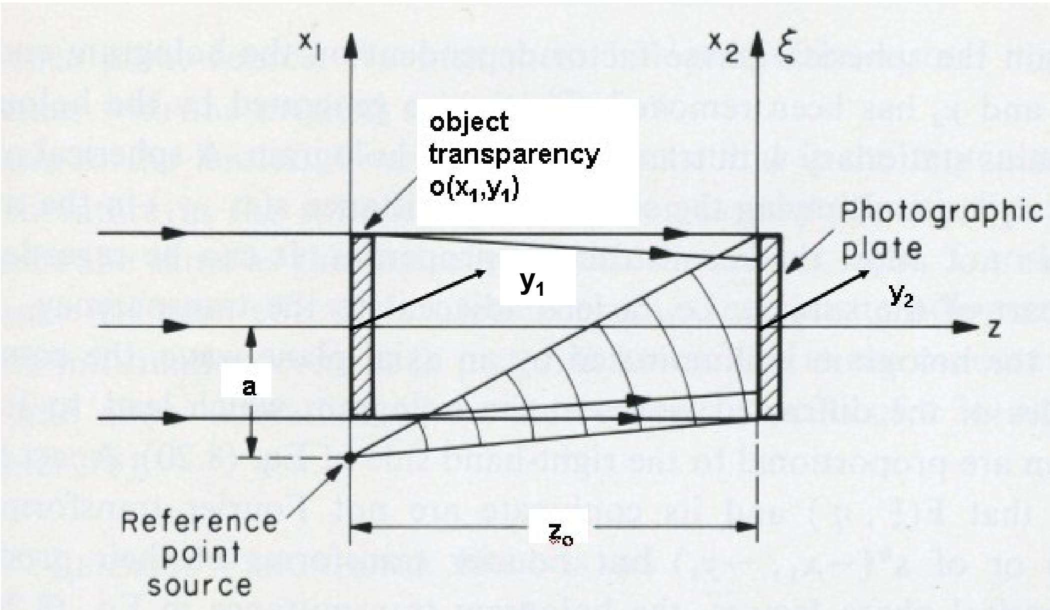
Imaging techniques:

Parallel beam, reference beam



reference beam: holography

Fourier Transform Holography – FTH (1)



$o(x_1,y_1)$: amplitude of the wave transmitted through object o

$r(x_1,y_1)$: reference wave

R.J. Collier, C.B. Burckhardt, L.H. Lin "Optical Holography", Academic Press (1971)

Fresnel Kirchhoff Theory

$o(x_2,y_2) = (i/\lambda z_o)\exp\{i\pi/\lambda z_o)(x_2^2+y_2^2)\}\ \bar{O}(\xi,\eta)$

$r(x_2,y_2) = (i/\lambda z_o)\exp\{i\pi/\lambda z_o)(x_2^2+y_2^2)\}\ \check{R}(\xi,\eta)\ \exp\{-2i\pi\xi a\}$

with $\bar{O}(\xi,\eta) = FT\{o(x_1,y_1)\}$, $\check{R}(\xi,\eta) = FT\{r(x_1,y_1)\}$, $\xi=x_2/\lambda z_o$, $\eta=y_2/\lambda z_o$

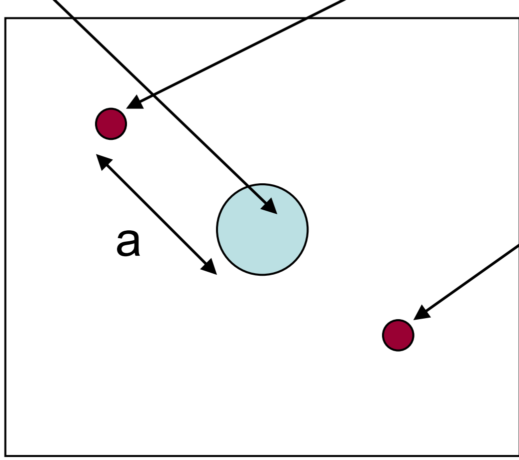
Fourier Transform Holography (2)

$$I(x_2,y_2) = | r(x_2,y_2) + o(x_2,y_2) |^2$$

$$I(x_2,y_2) = |r(x_2,y_2)|^2 + |o(x_2,y_2)|^2 + r^*(x_2,y_2)\, o(x_2,y_2) + r(x_2,y_2)\, o^*(x_2,y_2)$$

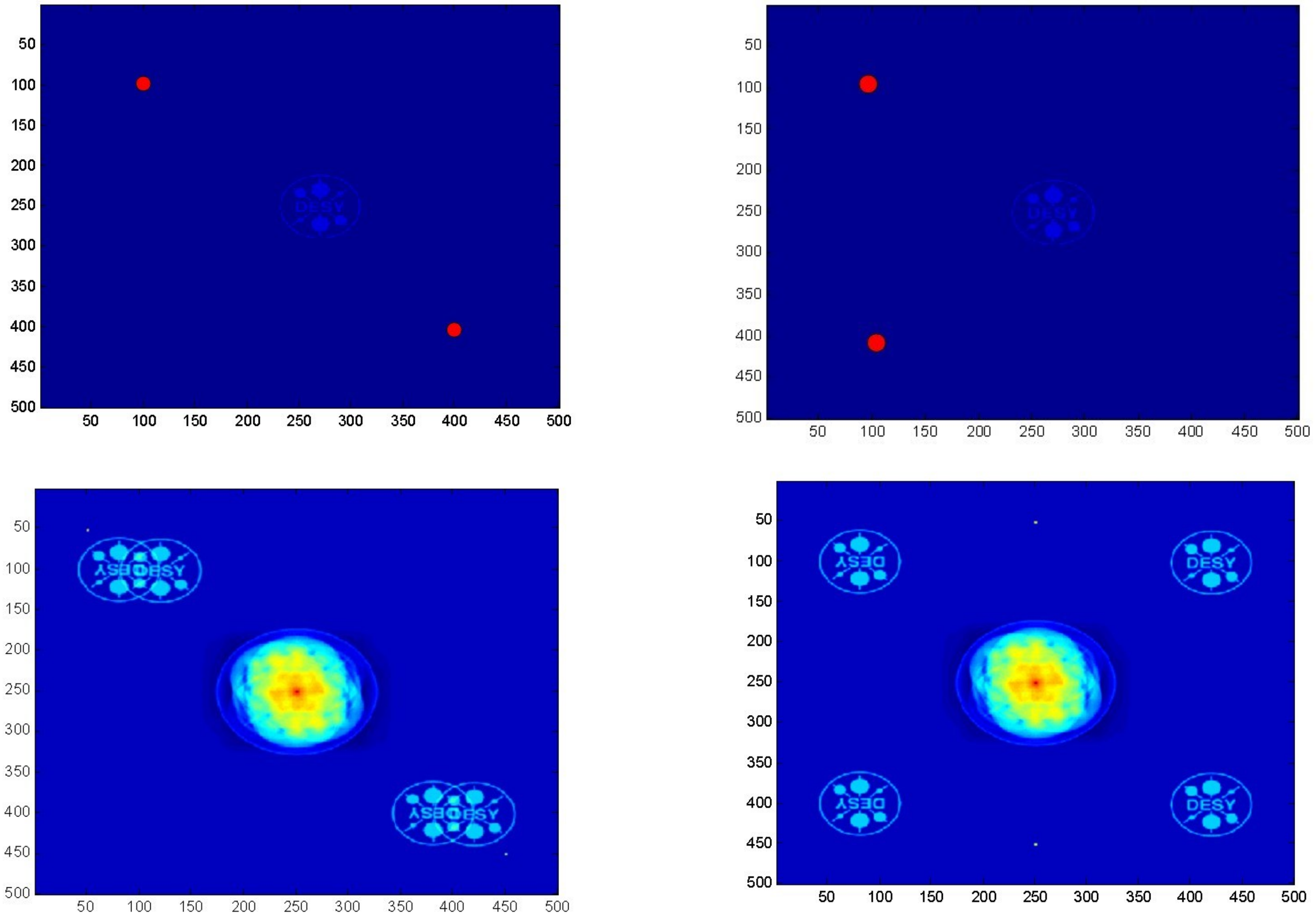
$$I(x_2,y_2) \approx |\check{R}(\xi,\eta)|^2 + |\bar{O}(\xi,\eta)|^2 + \check{R}^*(\xi,\eta)\, \bar{O}(\xi,\eta)\, e^{\,i\pi a\xi} + \check{R}(\xi,\eta)\, \bar{O}^*(\xi,\eta)\, e^{\,-i\pi a\xi}$$

$$FT\{r^*(-x,-y) \otimes o(x+a,y)\} + FT\{r(x,y) \otimes o(-x-a,-y)\}$$



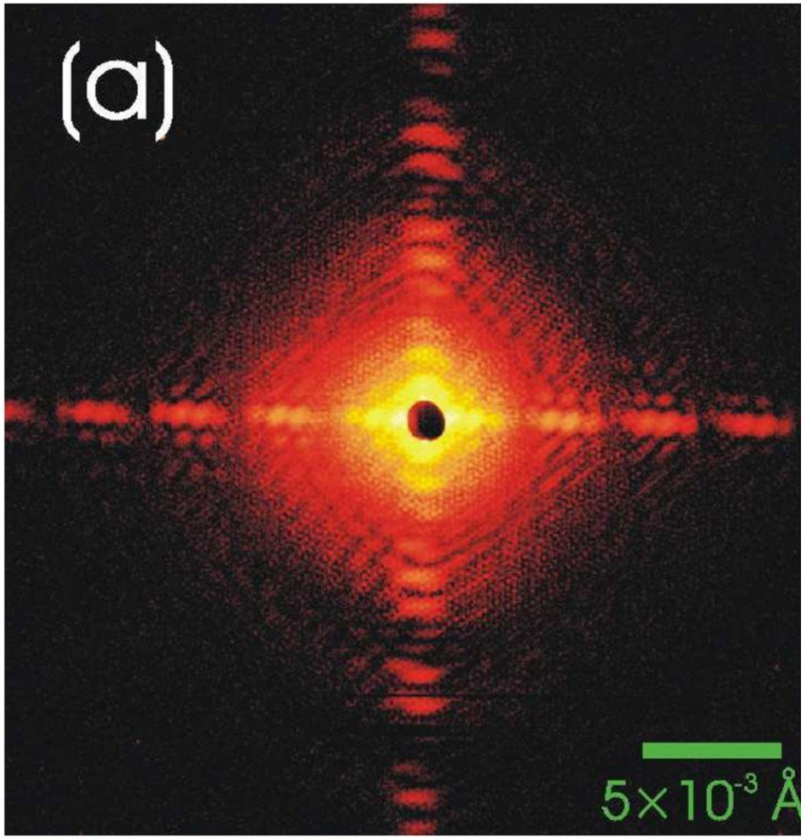
note: resolution determined by size of reference aperture

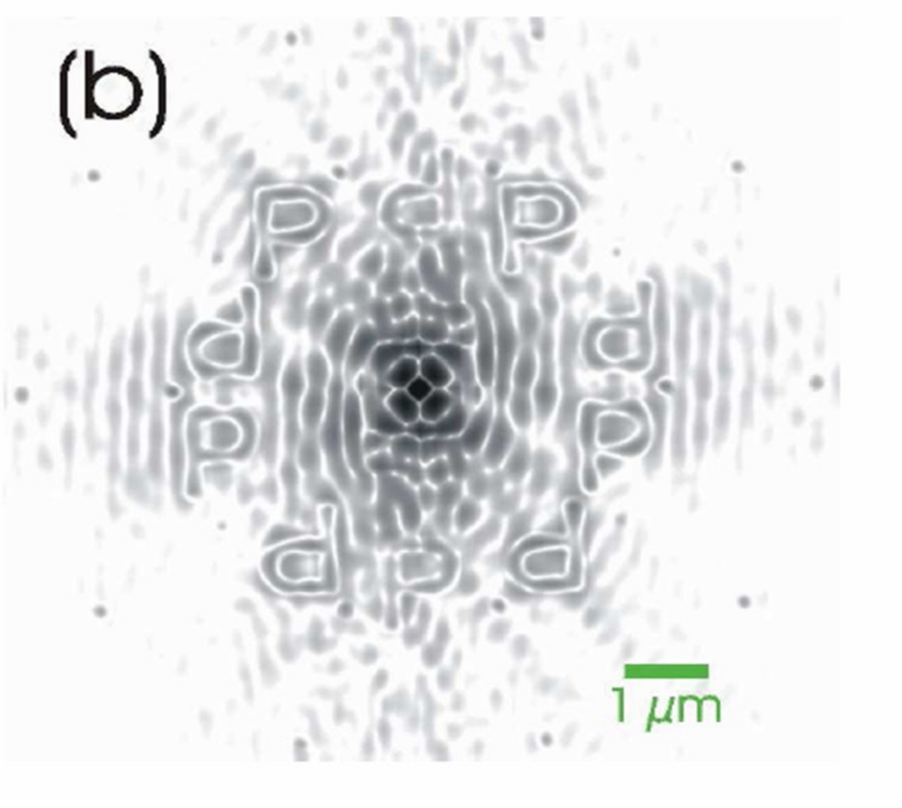
FTH (9): more than one reference hole



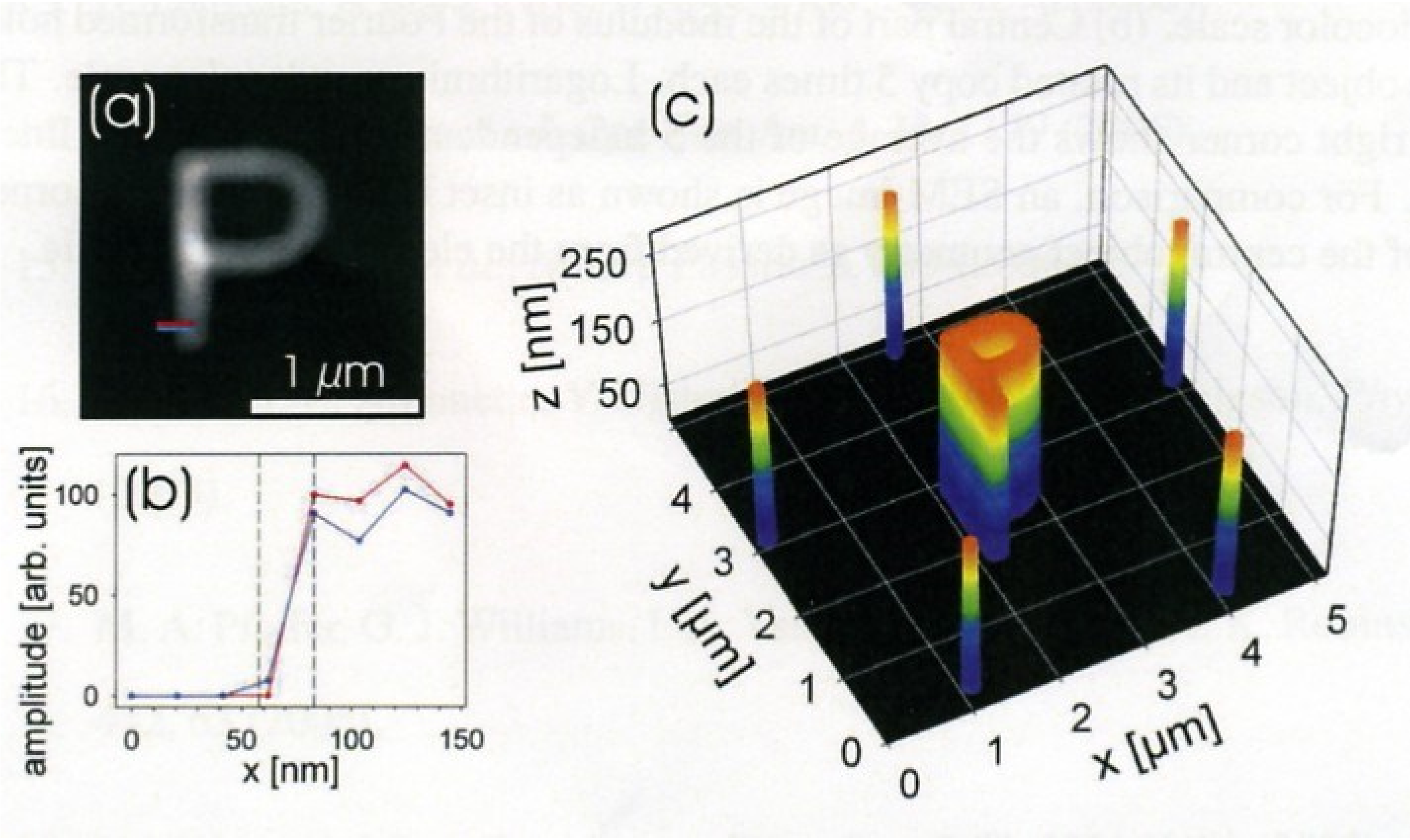
FTH (10)

Au reference structure (letter P): letter elements 200 nm in width and 220 nm heigh on 50 nm Si_3N_4-membrane. 5 Au reference dots of 175 nm diameter and 220 nm height on a circle of 2.5 μm around the sample. 200x 3s exposures (≈$1.4x10^8$ ph/s through $10x10μm^2$ at 8 keV.





FTH (10):

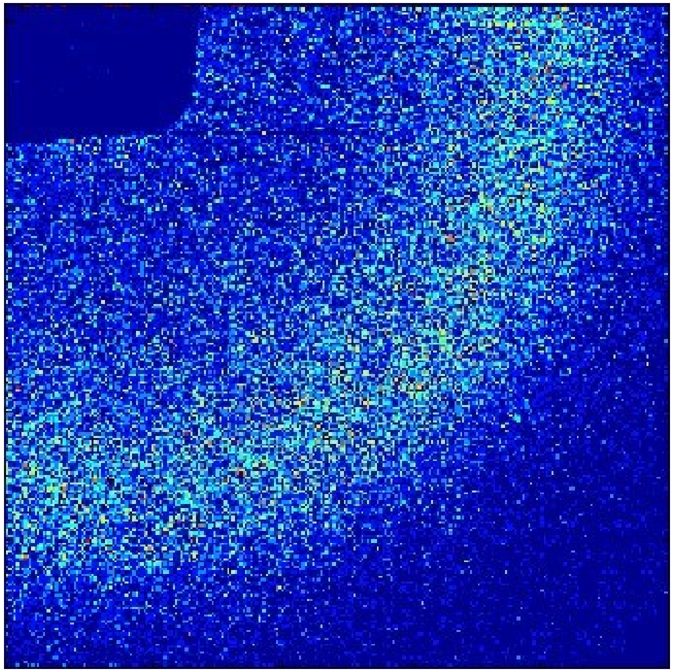


(a) average of 100 phase retrieval runs
(b) slices yielding a resolution of ≈ 25 nm
(c) visualization of the object as determined from the electron density profile

(Magnetic) Scattering with coherent X-rays

If coherent light is scattered from a disordered system it gives rise to a random (grainy) diffraction pattern, known as “speckle”. A speckle pattern is an interference pattern and related to the exact spatial arrangement of the scatterers in the disordered system.

$$I(Q,t) \sim S_c(Q,t) \sim \left| \Sigma \left(f_j^{charge} + f_j^{magnetic}\right) e^{iQRj(t)} \right|^2$$



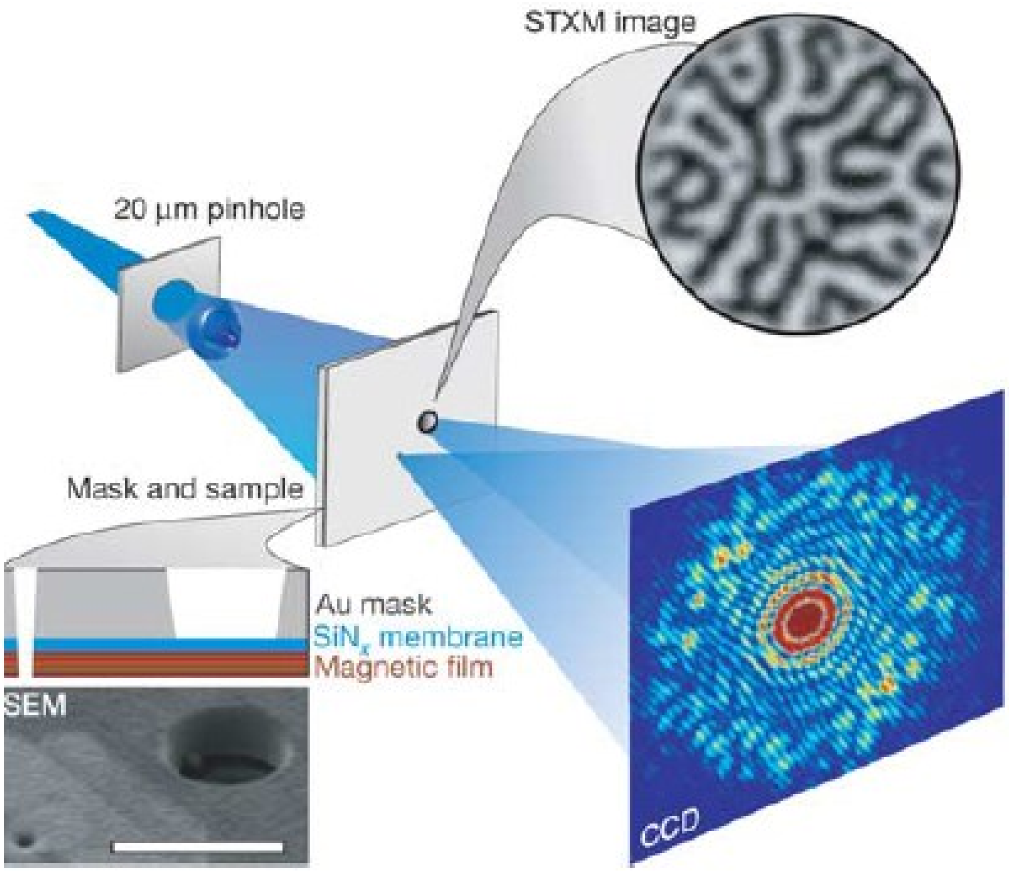
j in coherence volume $c = \xi_t^2 \xi_l$

Incoherent Light:

$$S(Q,t) = \langle S_c(Q,t) \rangle_{V>>c}$$

ensemble average

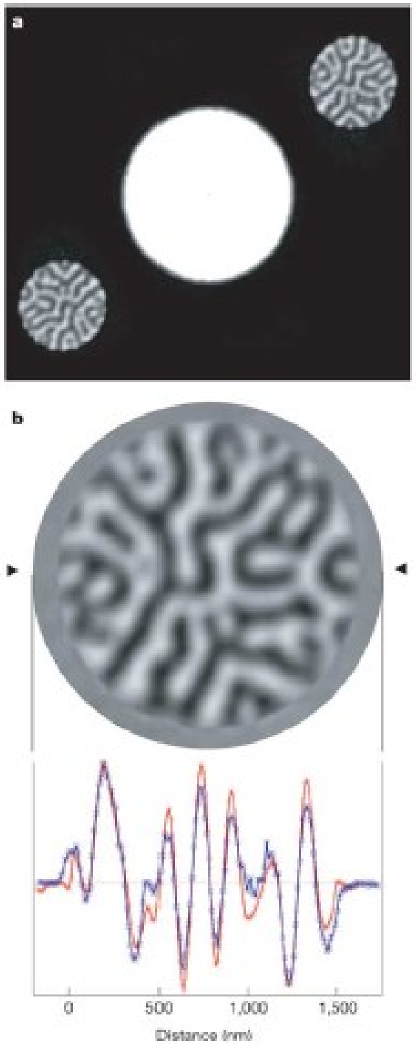
▪ Fourier Transform Holography (1)



Random magnetic (stripe) domains in a [Co(4)Pt(7)]50 ML sample, illuminated together with a reference aperature (1.5 µm) at the Co LIII edge absorption edge with a 778 eV (1.59 nm) 20 µm coherent soft x-ray beam.

S. Eisebitt, J. Lüning,W.F. Schlotter, M. Lörgen, O. Hellwig, W.Eberhardt and J. Stöhr,
NATURE, 432, 885 (2004)

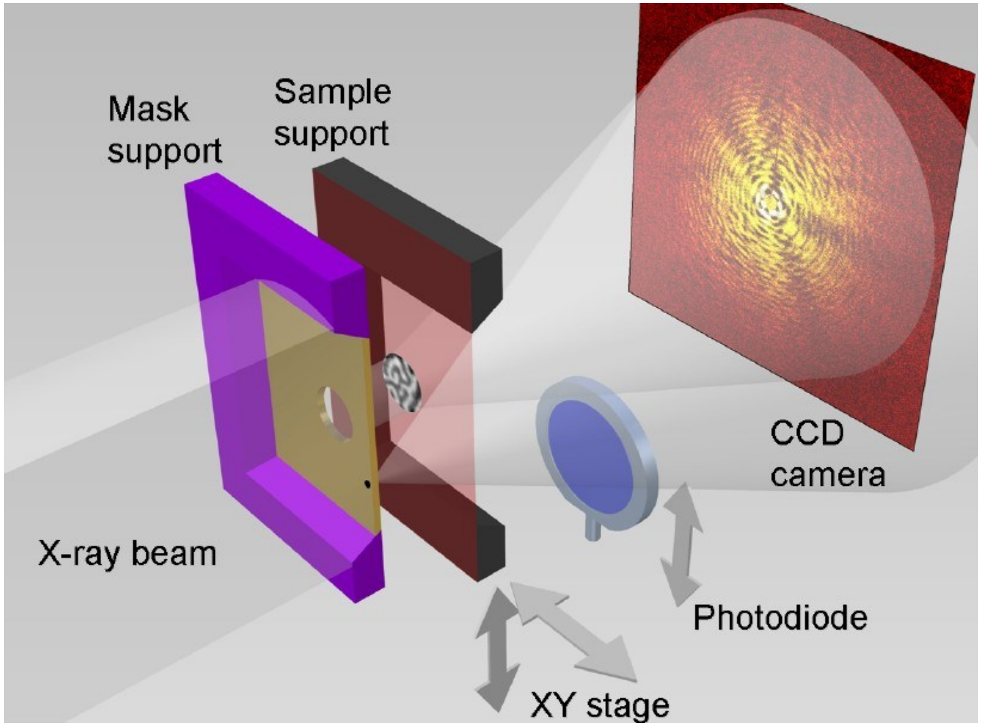
Fourier Transform Holography (2)

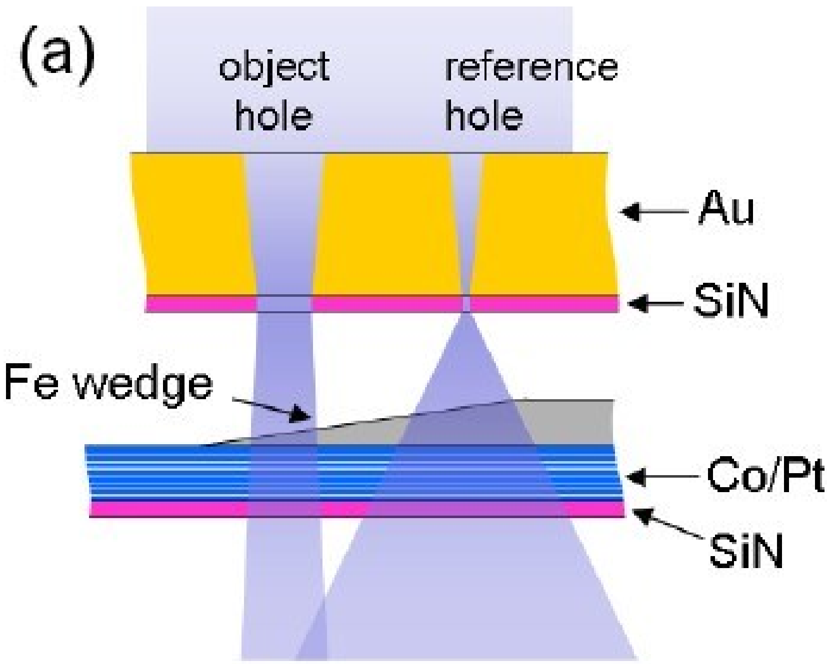


Reconstruction via direct 2-D Fourier transform of the [Co(4)Pt(7)]50 speckle pattern (top). Difference pattern obtained form two reconstructed images recorded with different helicity (bottom) compared to line profiles through the difference pattern (red) and an STXM image (blue) revealing 50 nm resolution.

S. Eisebitt, J. Lüning,W.F. Schlotter, M. Lörgen, O. Hellwig, W.Eberhardt and J. Stöhr, NATURE, 432, 885 (2004)

Soft X-ray holographic microscopy (I)



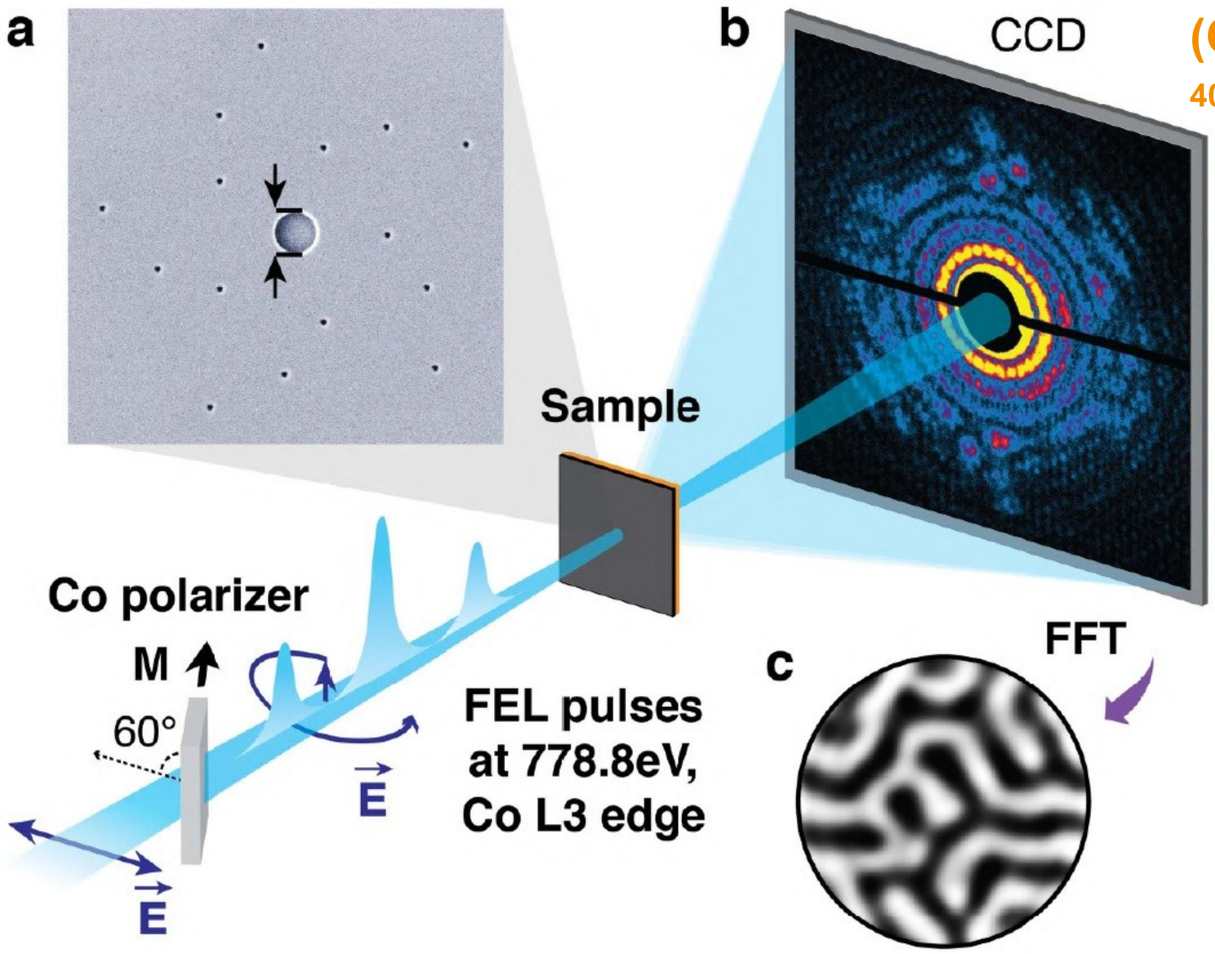


D. Stickler et al., APL **96**, 042501 (2010)

Resonant Magnetic Imaging at SXR (LCLS)

Sample: ($Co_{0.5nm}Pd_{0.7nm})_{40}$

10^9 ph/pulse
58% circular



T. Wang et al., Phys. Rev. Lett. 108, 267403 (2012)

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X-Ray Photon Correlation Spectroscopy (XPCS)

analogous to Dynamic Light Scattering (DLS) or Photon Correlation Spectroscopy (PCS) with visible light

- gives access to larger momentum transfers ($Q_{max}=2\pi\bullet\sin\theta/\lambda$) or shorter lengthscales
- not subject to multiple scattering
- can be combined with the surface sensitivity of X-rays

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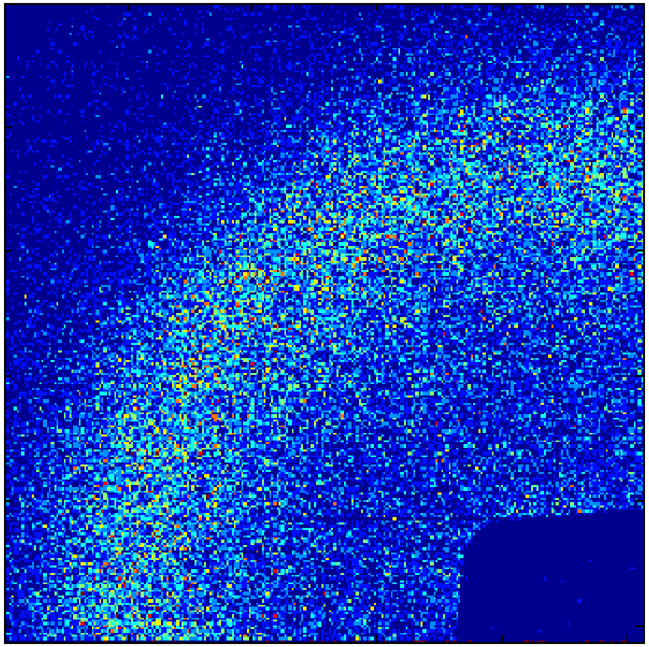
Introduction: Scattering with coherent X-rays

If coherent light is scattered from a disordered system it gives rise to a random (grainy) diffraction pattern, known as “speckle”. A speckle pattern is an interference pattern and related to the exact spatial arrangement of the scatterers in the disordered system.

$$I(Q,t) \sim S_c(Q,t) \sim | \sum e^{iQRj(t)} |^2$$



j in coherence volume $c = \xi_t^2 \xi_l$

Incoherent Light:

$S(Q,t) = < S_c(Q,t)>_{V>>c}$

ensemble average

- quantify dynamics in terms of the intensity correlation function $g_2(Q,t)$:

$I(\mathbf{Q},t) = |\mathbf{E}(\mathbf{Q},t)|^2 = |\sum b_n(\mathbf{Q}) \exp[i\mathbf{Q}\bullet\mathbf{r}_n(t)|^2$

Note: $\mathbf{E}(\mathbf{Q},t) = \int d\mathbf{r}' \rho(\mathbf{r}') \exp[i\mathbf{Q}\bullet\mathbf{r}'(t)]$ $\rho(\mathbf{r}')$: *charge density*

$g_2(\mathbf{Q},t) = \langle I(\mathbf{Q},0) \bullet I(\mathbf{Q},t)\rangle / \langle I(\mathbf{Q})\rangle^2$

if $\mathbf{E}(\mathbf{Q},t)$ is a zero mean, complex gaussian variable:

$g_2(\mathbf{Q},t) = 1 + ß(\mathbf{Q}) \langle\mathbf{E}(\mathbf{Q},0)\mathbf{E}^*(\mathbf{Q},t)\rangle^2 / \langle I(\mathbf{Q})\rangle^2$ <> ensemble av.;ß($\mathbf{Q}$) contrast

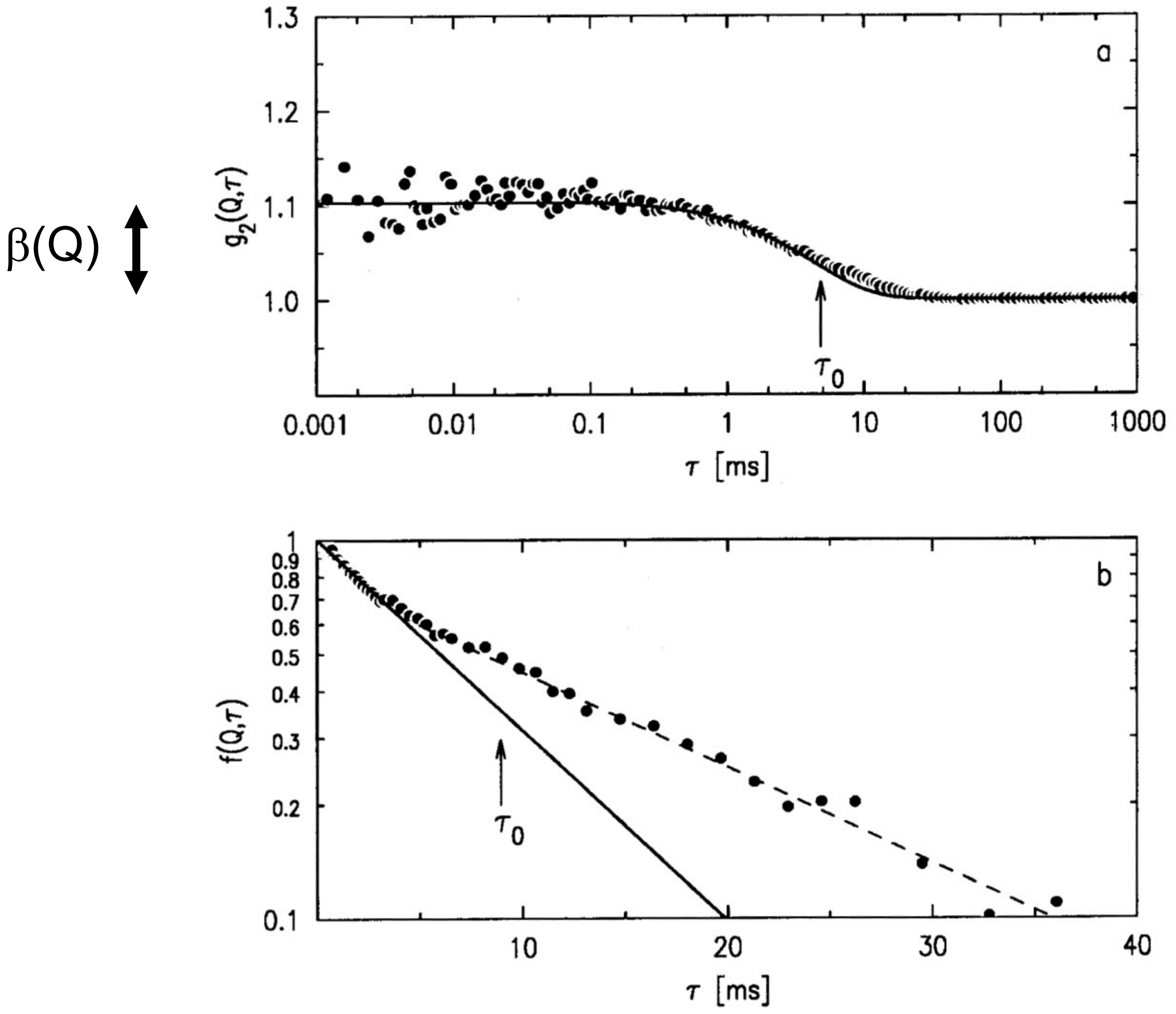
$g_2(\mathbf{Q},t) = 1 + ß(\mathbf{Q}) |f(\mathbf{Q},t)|^2$ with $f(\mathbf{Q},t) = F(\mathbf{Q},t) / F(\mathbf{Q},0)$

F(Q,0): static structure factor N: number of scatterers

$F(\mathbf{Q},t) = [1/N\{b^2(\mathbf{Q})\} |\sum^N_{m=1}\sum^N_{n=1} \langle b_n(\mathbf{Q})b_m(\mathbf{Q})\bullet\exp\{i\mathbf{Q}[\mathbf{r}_n(0)-\mathbf{r}_m(t)]\}\rangle$

A time correlation function $g_2(Q,\tau)$

$g_2(\mathbf{Q},t) = 1 + \beta(\mathbf{Q})\,|f(\mathbf{Q},t)|^2$ and $f(Q,t) = \exp(-\Gamma t) = \exp(-t/\tau)$



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Examples:

Dynamics of interacting colloidal particles

Dynamics in heterodyne mode detection

Dynamics at surfaces/interfaces

Dynamics (of a liquid crystalline system) near a phase transition

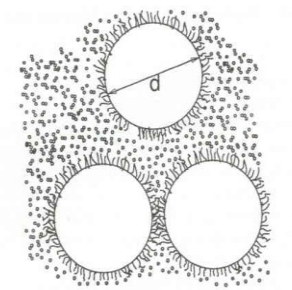
Non-equilibrium dynamics

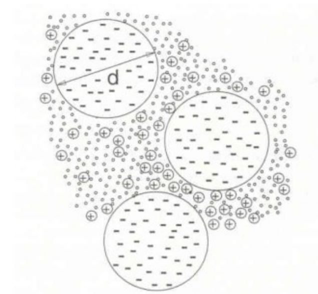
…

Dynamics of interacting Colloidal Particles

Colloidal particles (paints, ink, clays, ……silica,…..) suspended in a solvent (molecular fluid, …). Stabilised against van der Waals“ attraction.

„Hard Spheres“





„Soft Spheres“

Structure: $S(Q)=1+4\pi\rho \int [g\{r\}-1] (\sin(Qr)/Qr) r2dr$; $g\{r\}=\exp[-V(r)/kT]$ ρ: number density

$\Phi << 1\%$: $S(Q)=1$ $D(Q) = Do\ (=kT/6\pi\eta R_H)$

$\Phi > 1\%$:

$$V(r)= \begin{cases} 0 & r \geq d \\ \infty & r < d \end{cases}$$

$S=S(Q,\Phi)$ (Percus-Yevick)

weak interaction: DLVO

$V(r)$ ∞ $(eZeff)^2/r \exp(-\kappa r)$

$S=S(Q,\Phi,Zeff,\kappa)$ (MSA, RMSA)

Dynamics: Interaction: colloid-solvent, colloid-colloid, hydrodynamics

Smoluchowski (many particle) diffusion equation:

$$D_{short}(Q) = Do/S(Q) * H(Q) \quad t<<R^2/Do$$

H(Q) (Beenakker and Mazur)

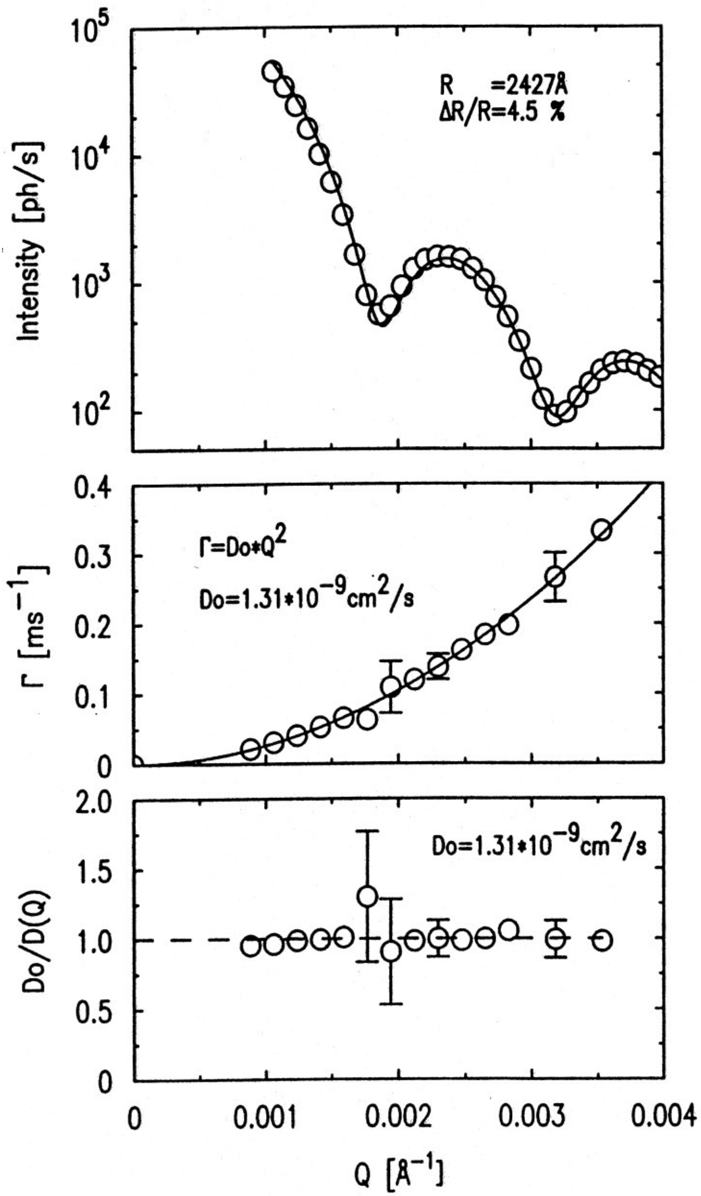
H(Q) (via Do, S(Q), D(Q))

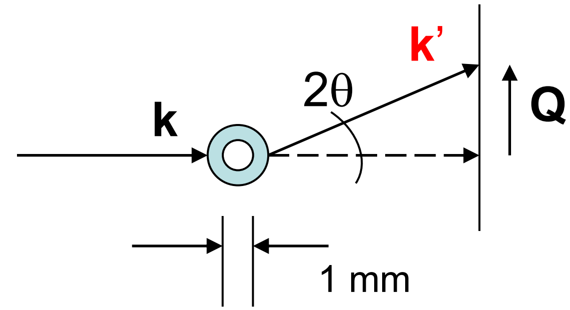
The dilute case

I~|F(Q)|^2 S(Q)

~[(sinQR-QRcosQR)/(QR)^3]^2

Γ = Do Q^2





Q = k'- k

Q = 2k sinθ

k = 2π/λ

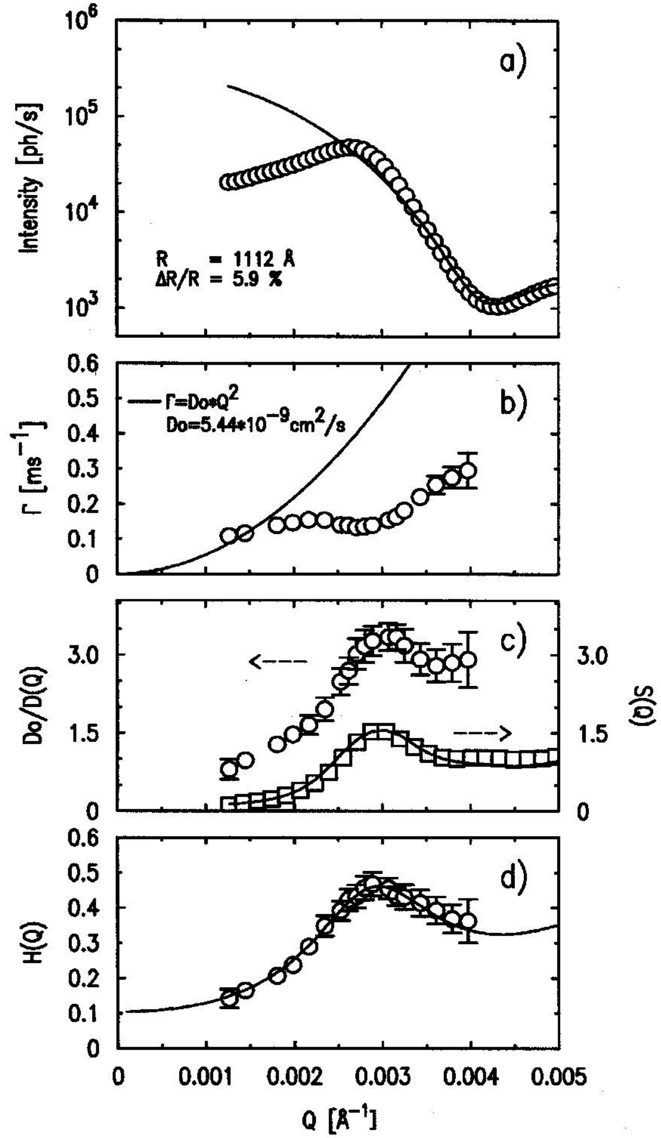
G. Grübel, A. Robert, D. Abernathy
8th Tohwa University International Symposium on "Slow Dynamics in Complex Systems", 1998, Fukuoka, Japan

▪ The Hard Sphere Case

Poly-
methylmetacrylate

37% volume fraction
in cis-decaline

sterically stabilized
(**hard-spheres**)



--- F(Q)

--- $\Gamma = D_0Q^2$

$S(Q) = I(Q)/F(Q)$

caging (deGennes
narrowing)

$H(Q) = S(Q)/ [D_0 /D(Q)]$

--- δ-γ expansion

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Examples:

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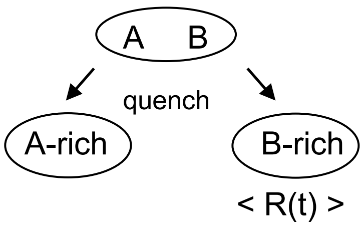
Dynamics (of a liquid crystalline system) near a phase transition

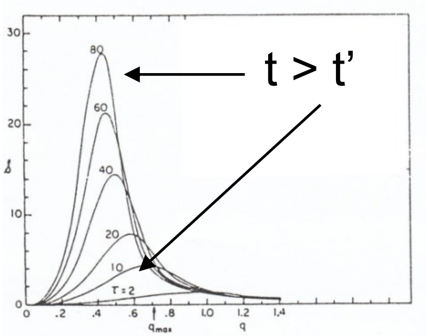
Non-equilibrium dynamics

…

Non-Equilibrium Dynamics

Domain coarsening in phase separating systems (glasses, alloys,…) e.g. after quenching, aging…





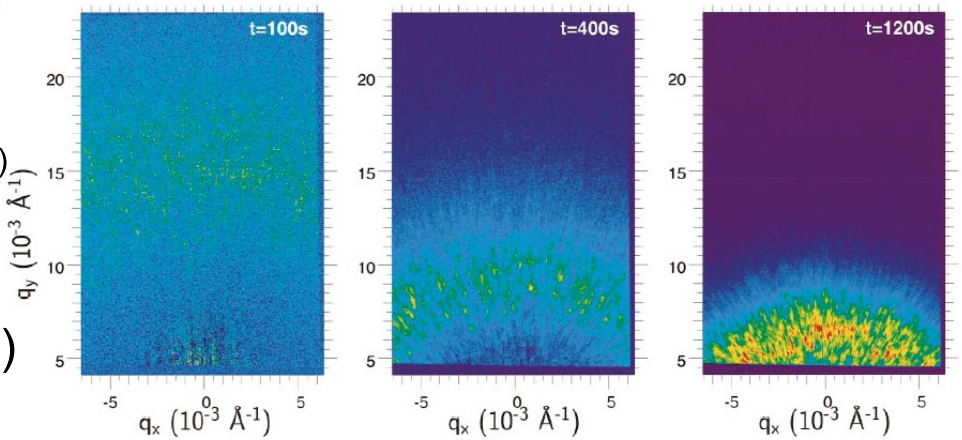
Phase – separating Glass

Malik et al., PRL 81, 5832, 1998

$(Na_2O)_{0.07}(B_2O_3)_{0.22}(SiO_2)_{0.71}$ T=1033K

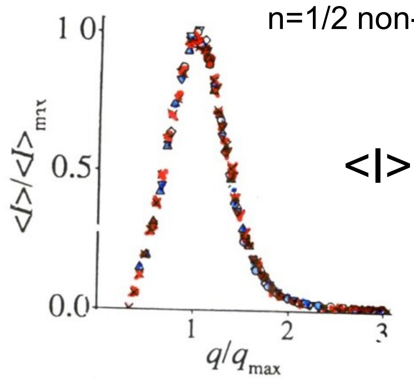
quench

(B_2O_3)-rich (SiO_2)-rich 943K<T<963K



Dynamic Scaling: $< R(t) > \sim t^{\,n}$

n=1/3 conserved order parameter (model B)
n=1/2 non-cons. order parameter (model A)



$$<I>(q,t)/<I>_{max} = F\,(q/q_{max})$$

$< R(t) > \sim t^{\,n}$ n=1/3

XPCS: investigate fluctuations around the average scaling behaviour: $\tau = \tau\,(q,t)$

Phase-Ordering in Cu_3Au

high T: fcc sites occupied by either Cu or Au

T≤Tc=383 C: ordering with Au on corner and Cu on face sites

4-fold degenerate ground-state s

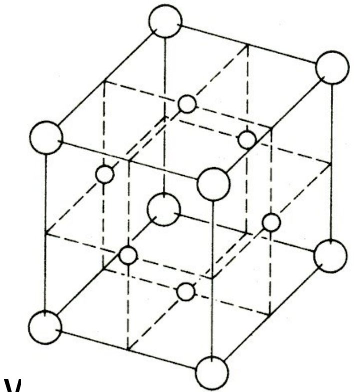
can be choosen in 4 different ways) v

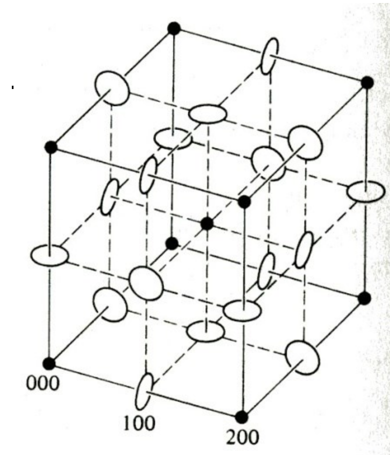
groundstates separated by domain walls

walls give rise to ellipse shaped superlattice reflections of

quench: domain formation and growth in disordered phase

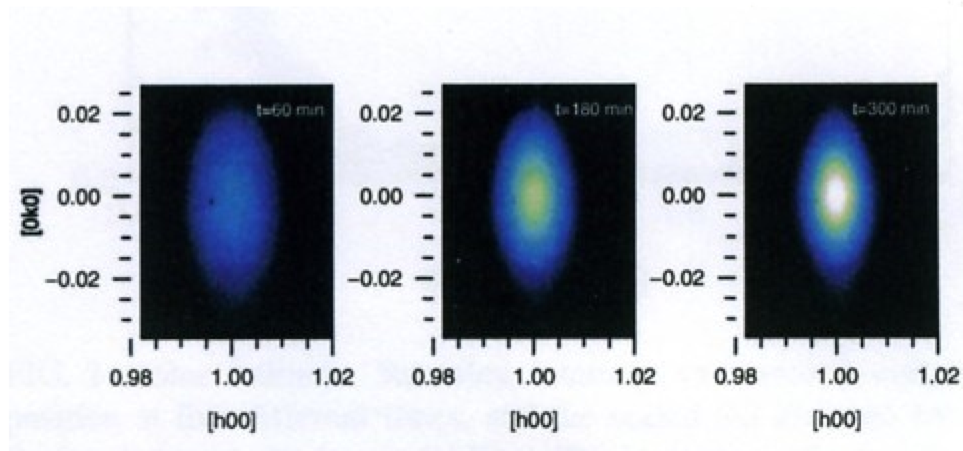
coarsening with R ~ $t^{1/2}$





[100] superlattice reflection after quench from 425 C to 370 C

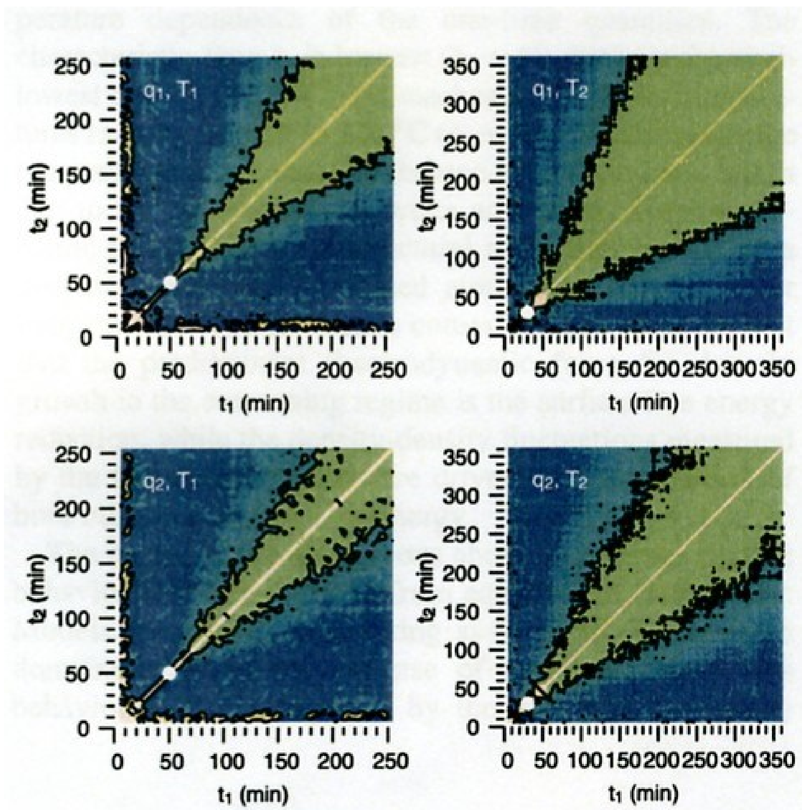
Fluerasu&Sutton, PRL94(2005)55501



- Study fluctuations about the average behaviour: XPCS

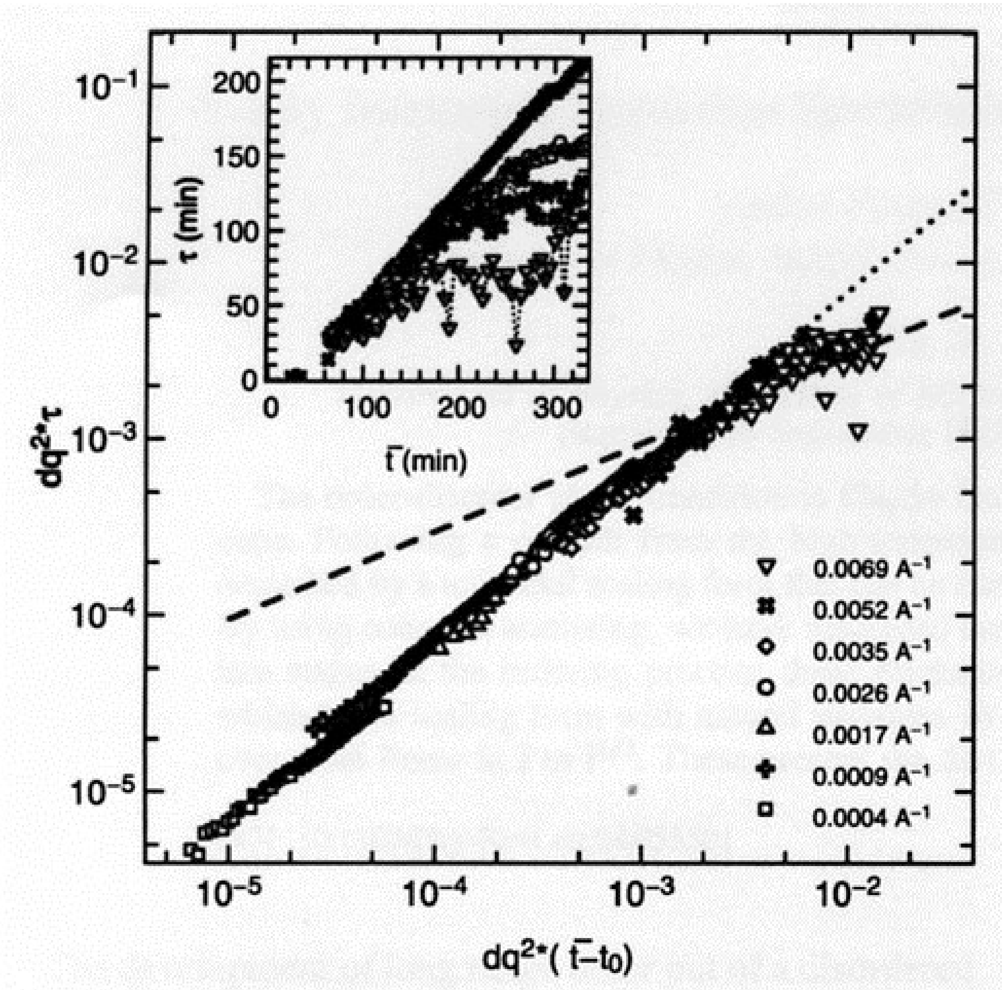
  characterize by two-time correlation function

$$C(q,t_1,t_2) = \frac{\langle I(t_1)\, I(t_2)\rangle - \langle I(t_1)\rangle \langle I(t_1)\rangle}{[\langle I^2(t_1)\rangle - \langle I(t_1)\rangle^2]^{1/2}\, [\langle I^2(t_2)\rangle - \langle I(t_1)\rangle^2]^{1/2}}$$



- rescaled correlation time $\tau \sim t_{mean}$ in the low t_{mean} limit

  $\sim t_{mean}^{1/2}$ in the high t_{mean} limit



= Q - [100]

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XPCS at a FEL source

Magnetization Dynamics

Ferroelectrics

Ultrafast dynamics at surfaces and interfaces of liquids

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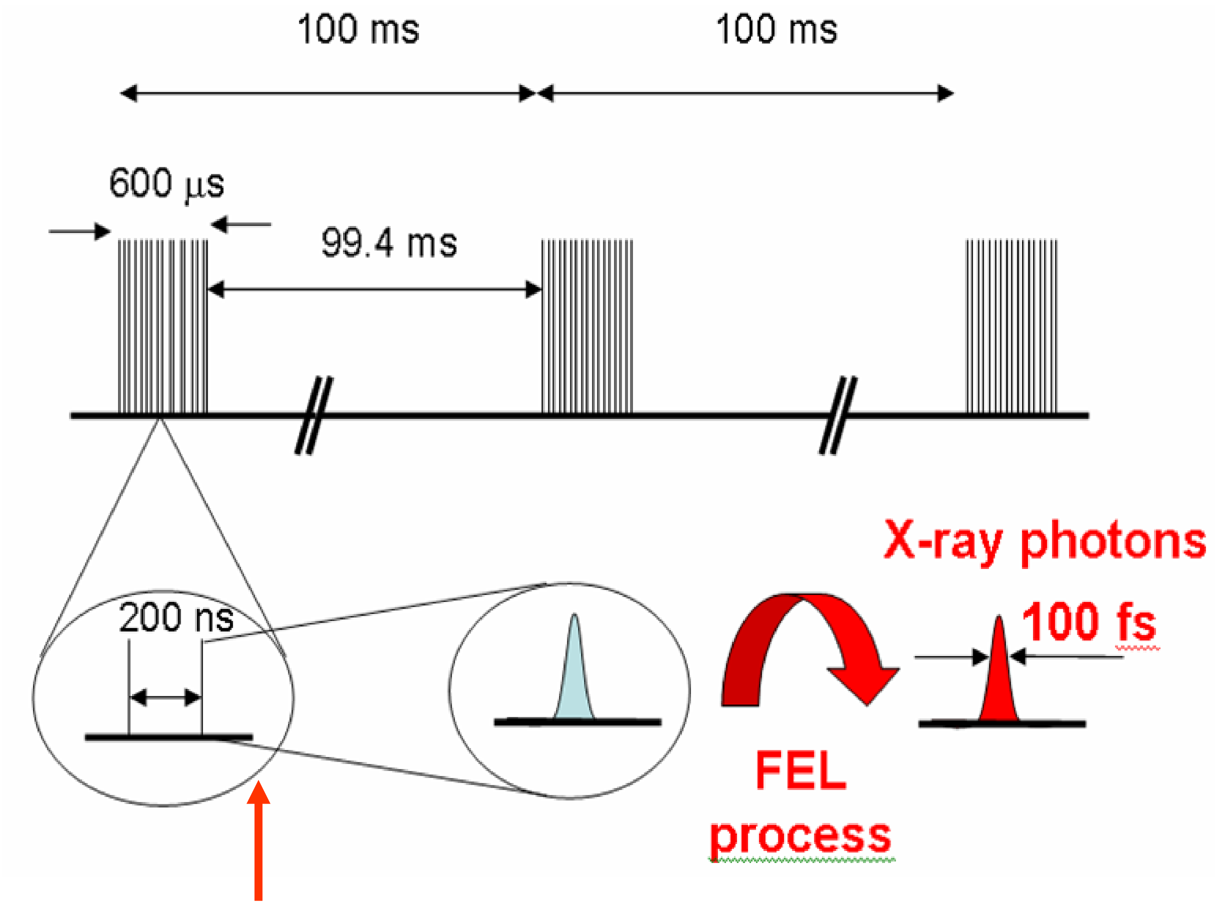
XPCS at a XFEL source:



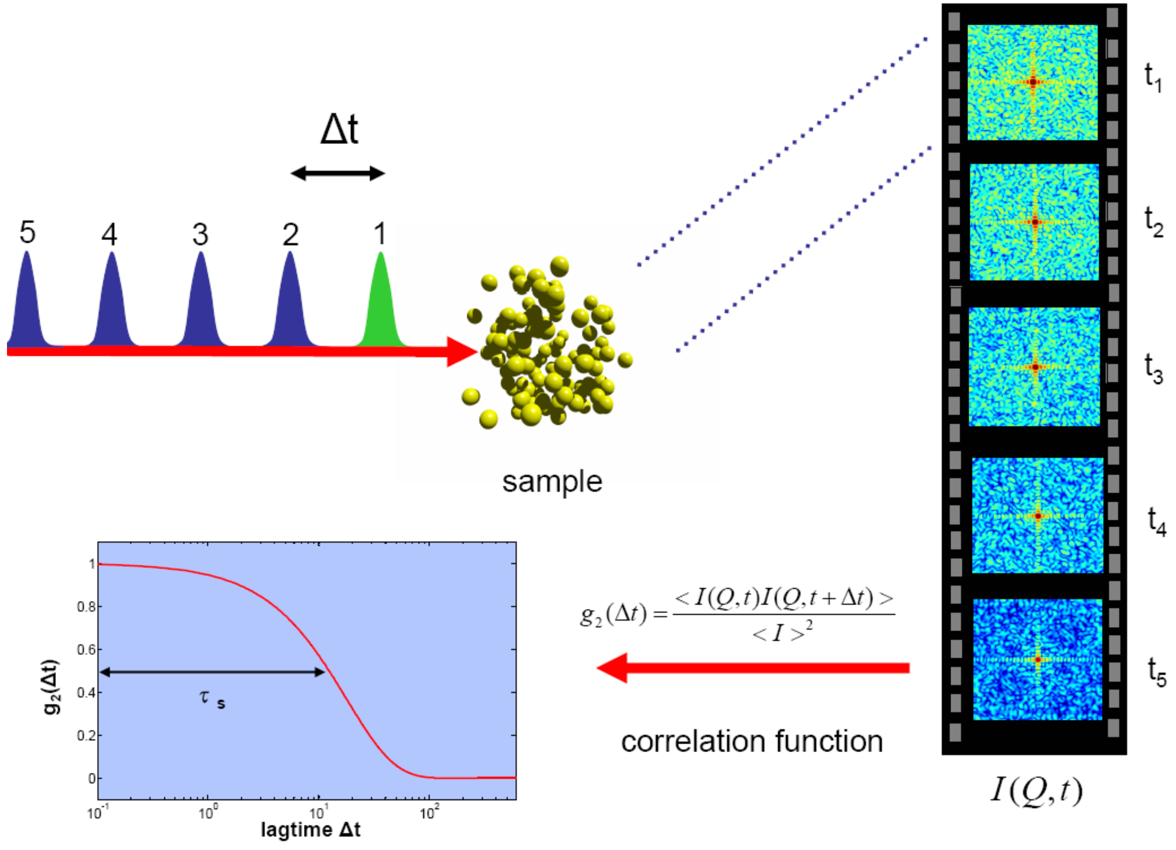
t > 0.1 s
200ns < t < 600 µs:

<u>" movie" mode</u>

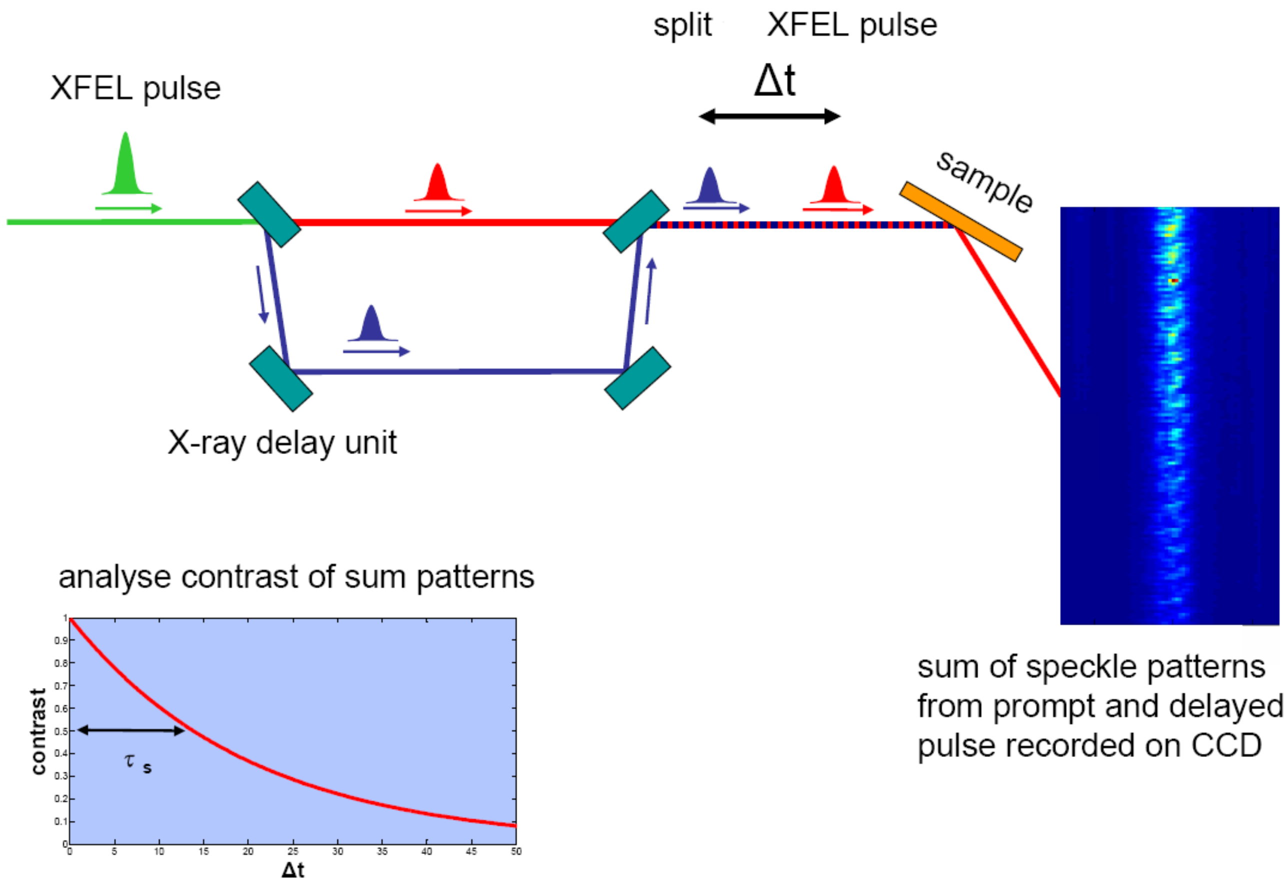
1ps < t< 10 ns: **<u>"delay-line" mode</u>**

for "all" times: **<u>"pump-probe" mode</u>**

XPCS at a FEL source: Movie Mode



Delay Line Mode

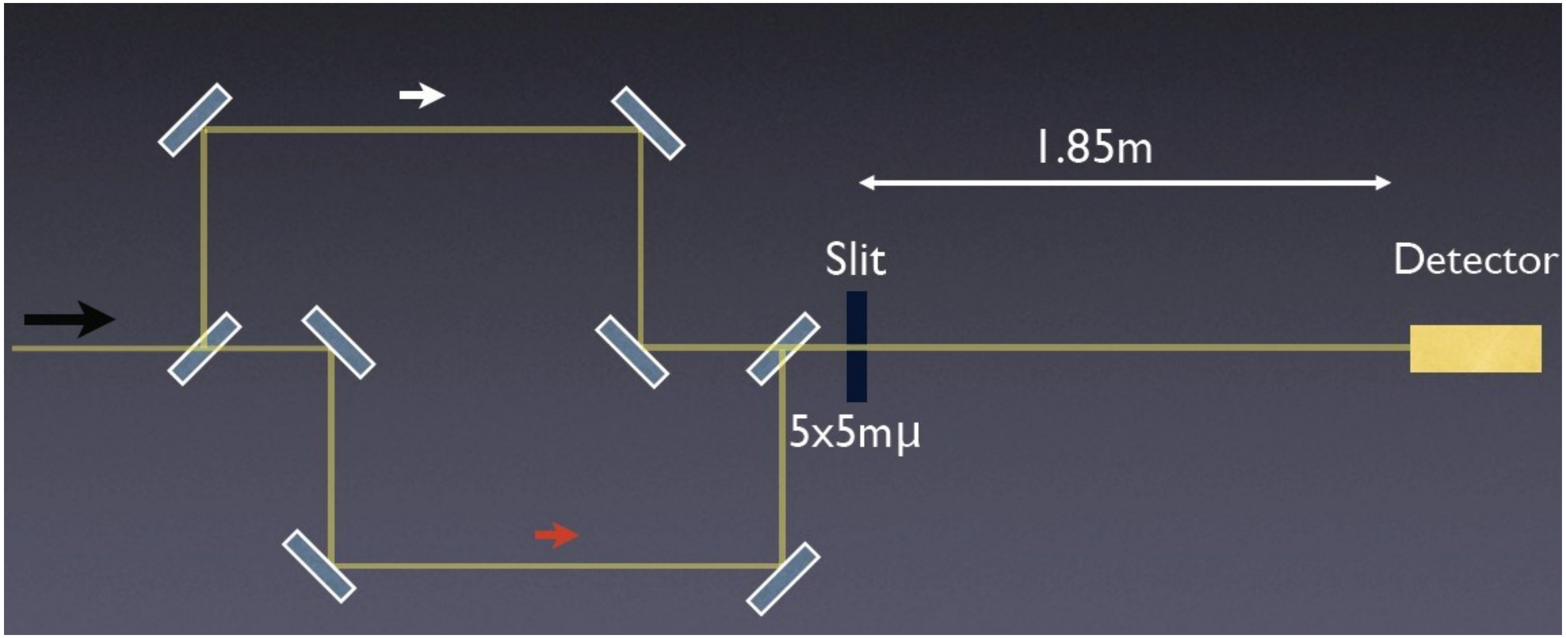


<u>"Delay Line" Mode:</u> 1ps < Δt < 10ns (1 ps ⇔ 0.3 mm; 1ns ⇔ 300 mm) "luminosity limited".

Delay Line for “hard” X-Rays

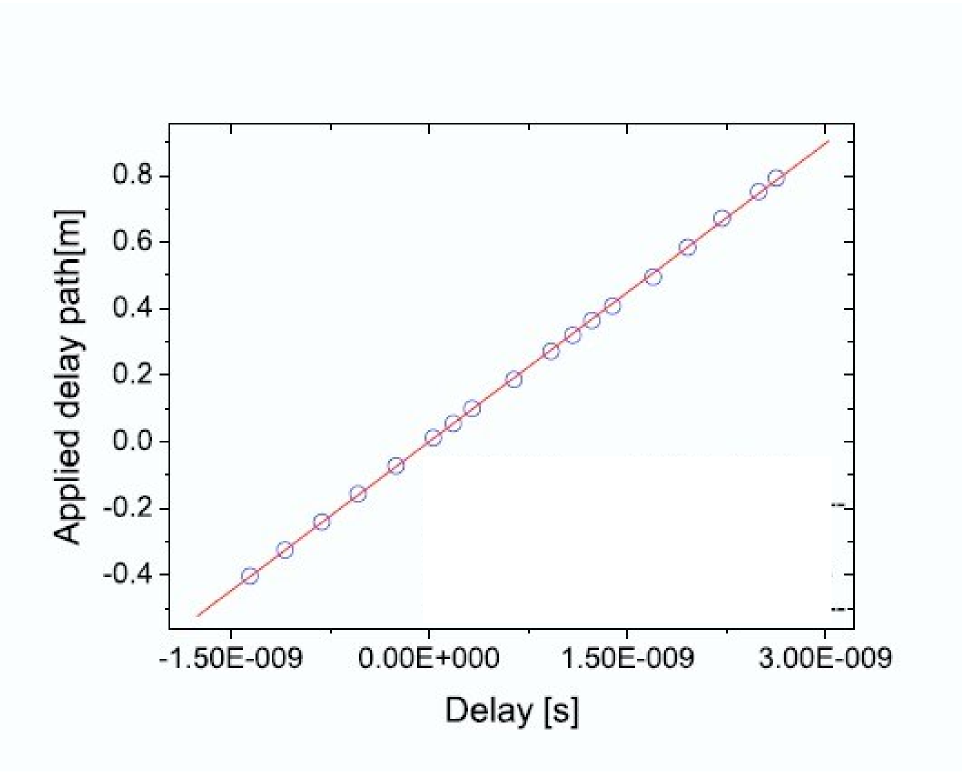
Si(511) at 8.38 keV

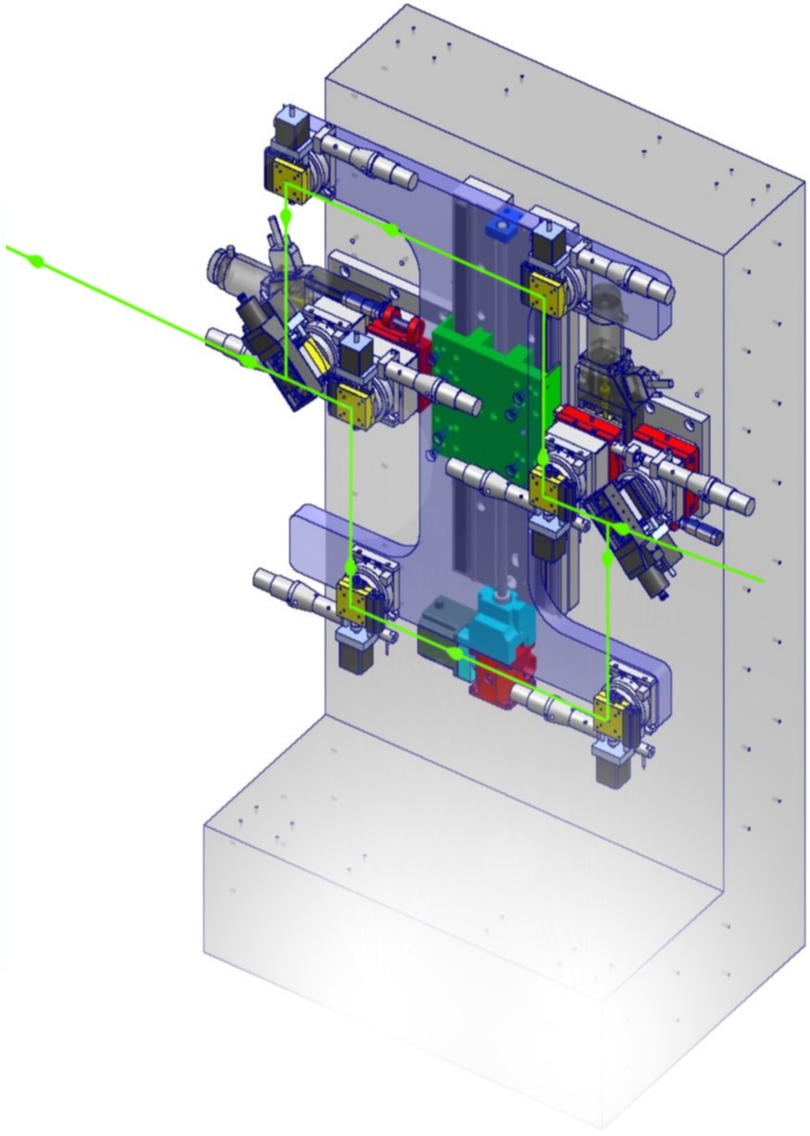
Δt_{max} = 2.8 ns



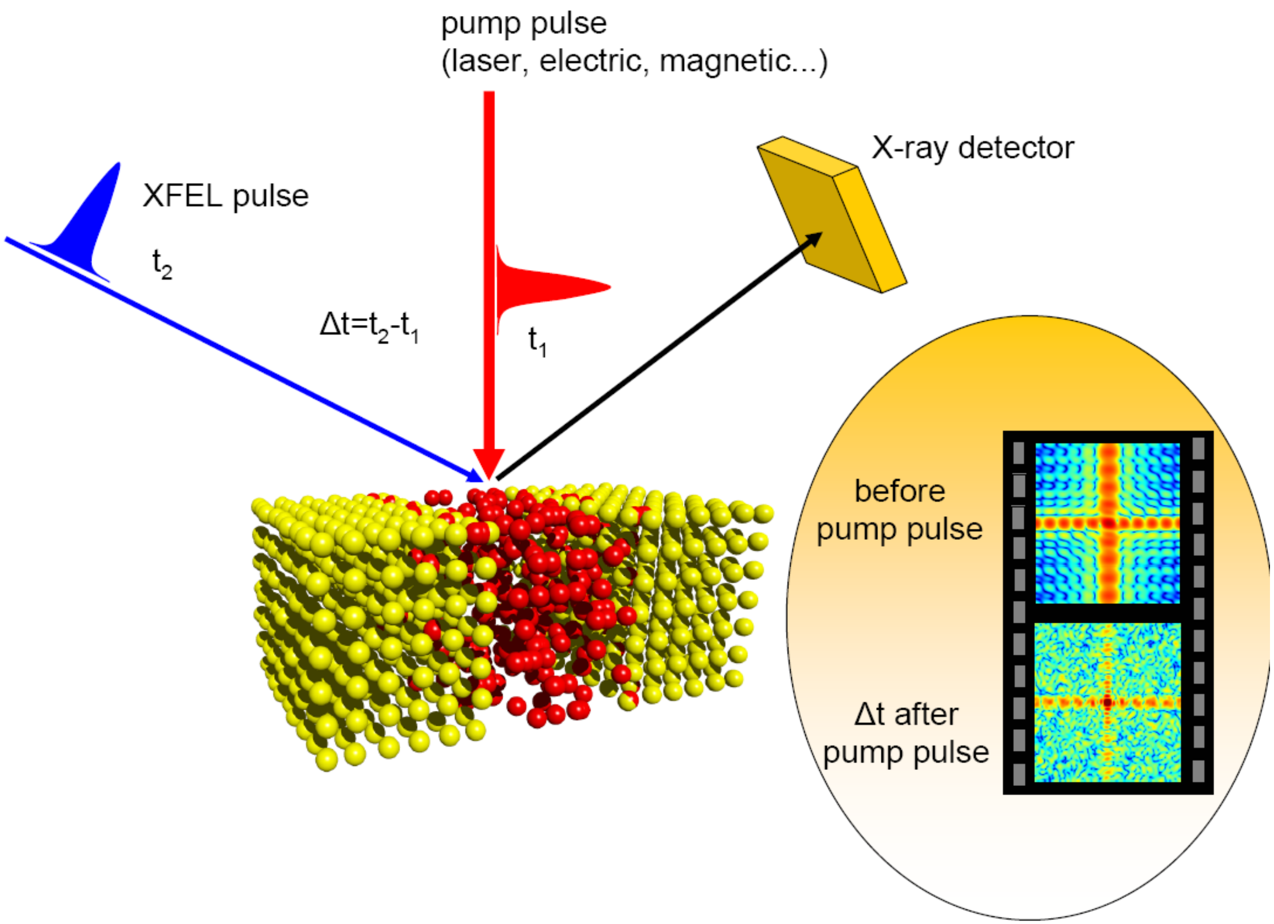
W. Roseker, H. Schulte-Schrepping,
A. Ehnes, H. Franz, O. Leupold and
G. Grübel

X-Ray delay line





XPCS at a FEL source: pump-probe mode



▪

The XFEL

www.xfel.eu

3.4km