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

Part III: 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
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.

\[ I(Q,t) \sim S_c(Q,t) \sim \left| \sum j \ e^{iQRj(t)} \right|^2 \]

\( j \) in coherence volume \( c = \xi_l^2 \xi_t \)

Incoherent Light:

\[ S(Q,t) = \langle S_c(Q,t) \rangle_{V>>c} \text{ ensemble average} \]
quantify dynamics in terms of the intensity correlation function $g_2(Q,t)$:

$$I(Q,t) = |E(Q,t)|^2 = |\sum b_n(Q) \exp[iQ \cdot r_n(t)]|^2$$

*Note: $E(Q,t) = \int \rho(r') \exp[iQ \cdot r'](t)\,dr'$*

$$g_2(Q,t) = \frac{\langle I(Q,0) \cdot I(Q,t) \rangle}{\langle I(Q) \rangle^2}$$

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

$$g_2(Q,t) = 1 + \beta(Q) \frac{\langle E(Q,0) E^*(Q,t) \rangle^2}{\langle I(Q) \rangle^2}$$

<> ensemble av.; $\beta(Q)$ contrast

$$g_2(Q,t) = 1 + \beta(Q) |f(Q,t)|^2$$

with $f(Q,t) = F(Q,t) / F(Q,0)$

$F(Q,0)$: static structure factor

$N$: number of scatterers

$$F(Q,t) = \left[1/N\{b^2(Q)\}\right] \sum_{m=1}^{N} \sum_{n=1}^{N} \langle b_n(Q)b_m(Q) \cdot \exp{iQ[r_n(0)-r_m(t)]} \rangle$$
A time correlation function $g_2(Q, \tau)$

$$g_2(Q,t) = 1 + \beta(Q) |f(Q,t)|^2 \quad \text{and} \quad f(Q,t) = \exp(-\Gamma t) = \exp(-t/\tau)$$
Surface Sensitivity of X-Rays

refractive index:

\[ n = 1 - \delta + i \beta \]

= 1.5 - 1.8 visible light

X-rays: \( \delta = 10^{-5} \) (solid matter)

\( \beta \ll \delta \)

\( n < 1 \)

\[ \cos \alpha = n \cdot \cos \alpha' \]

total external reflection for: \( \alpha < \alpha_c \)

for \( \alpha' = 0 \) (and \( \beta = 0 \)): \( \alpha_c = \sqrt{2\delta} \) [mrad]

\( \alpha < \alpha_c \): evanescent wave with nanometer penetration
Dynamics at Surfaces: Capillary Waves

Thermally excited capillary waves decorate the surfaces of all liquids depending on the surface tension $\sigma$ and the viscosity $\eta$.

**Capillary wave:**

$$\zeta(r,t) = \zeta_0 \exp(iq_||r - \omega t)$$

$$\omega = \omega_0 + i\Gamma$$

The linear Navier-Stokes equation for simple liquids yields:

**Low viscosity:**

$$\frac{\sigma \rho}{4\eta^2 q_||^2} > 1$$

$$\Gamma = \frac{2\eta}{\rho} q_||^2$$

$$\omega_0 = \sqrt{\frac{\sigma}{\rho}} q_||^{3/2}$$

propagating wave

**High viscosity:**

$$\frac{\sigma \rho}{4\eta^2 q_||^2} < 1$$

$$\Gamma = \frac{\sigma}{\eta} q_||$$

overdamped wave
Dynamics at Surfaces: Capillary Waves

X-Ray Photon Correlation Spectroscopy in Surface Geometry

Glycerol: a „prototypical“ glassformer
Seydel, Madsen, Tolan, Grübel, Press, PRB63, 73409 (2001)

\[
\alpha = \beta : \text{reflectivity} \quad q_z = (2\pi/\lambda) \ 2 \sin \alpha
\]

\[
\alpha_i = \alpha_c : 
\quad 50 - 100\text{Å penetration surface sensitivity}
\]

\[
\alpha \neq \beta : 
\quad q_{||} = 2\pi/\lambda(\cos \beta - \cos \alpha) 
\quad \text{in-plane correlations}
\]

overdamped regime:

\[
\Gamma = (\gamma/2\eta)k; \quad \tau_0 = \{\eta(T)/\pi \ \gamma(T)\}x_0
\]
- **Viscosity of a liquid crystal near the nematic to smecticA transition**
  

\[
T >> T_{NA} \quad T \rightarrow T_{NA} = 337 \text{ K}
\]

\[
\xi_{||} \sim \{(T - T_{NA})/T_{NA}\}^{V_{||}} = \xi_{||}^{S}\]

\[
\xi_{\perp} \sim \{(T - T_{NA})/T_{NA}\}^{V_{\perp}}
\]

\[
v_{||} = 0.71; \quad v_{\perp} = 0.58
\]

**References:**
- Pershan and Als-Nielsen, PRL 52, 759 (1984)
Viscosity of a Liquid Crystal near the Nematic-SmecticA Transition

Dynamics:

viscosity is anisotropic: \( \eta_1, \eta_2, \eta_3 \)
depending on the relative orientations: \( n, v, \nabla \cdot v \)
described by Leslie coefficients \( \alpha_1, \ldots, \alpha_5 \),
or parameters \( v_1, \ldots, v_5 \) (Harvard notation)
\( (v_4=v_5=0 \text{ for incompressible fluids}) \)

Predictions:

\[ \beta = 3v_{||} - 2v_{\perp} \quad [1] \]
\[ \beta = 1/3 \quad [2] \]
\[ \beta = 1/2 \quad [3] \]


deGennes, Sol. State Comm.,10,753 (1972)


Theory: (N-SmA transition)

\[ \eta_1 \sim \exp(E_A/kT) \]
\[ \eta_2 \sim \exp(E'_A/kT) \]
\[ \eta_3 = c(T-T_{NA}/T_{NA})^{-\beta} + \text{non.div.} \]
Viscosity of a Liquid Crystal near the Nematic-SmecticA Transition

\[ \eta_3 \sim (T - T_{NA}/T_{NA})^{-\beta} \]
\[ \beta = 0.95(2) \]

in agreement with theory [1]:
\[ \beta = 3\nu_\parallel - 2\nu_\perp = 0.94 \]
\[ \nu_\parallel = 0.70; \quad \nu_\perp = 0.58 \] (static data)

\( \eta / \sigma \) diverging
\( \sigma \) constant [1]; data reflect temperature dependence of viscosity

- Out-plane movements (\( \zeta \parallel n \)) within the smectic layers are strongly damped (\( \eta_3 \) critical).
- The smectic layers remain viscous in-plane (\( \eta_2 \) non-critical)
Non-Equilibrium Dynamics

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

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

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

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

Phase – separating Glass
Malik et al., PRL 81, 5832, 1998

$< R(t) > \sim t^{n}$

$\chi(q,t)/\chi_{\text{max}} = F (q/q_{\text{max}})$

$\chi(q) = F (q/q_{\text{max}})$

$T=1033K$

$(\text{Na}_2\text{O})_{0.07}(\text{B}_2\text{O}_3)_{0.22}(\text{SiO}_2)_{0.71}$

$943K<T<963K$

$\chi_{\text{max}}$
Two time correlation function:

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

Fluctuations \( \tau = \tau (q,t) \)

Prediction: \[ \tau(q,t) = \left[ t_{\text{max}}(q) - t_0 \right] \left\{ a \frac{t-t_0}{t_{\text{max}}(q) - t_0} \right\}^{(1-n)} \]

\[ \sim 1/q \ t^{2/3} \]

\( \Delta t = t_1 - t_2 \)

\[ t = \frac{t_1 + t_2}{2} \]

\[ \tau = \tau (t) \]

\[ a = 0.72(2) \quad (1-n) = 0.65(4) \quad = 1 - \frac{1}{3} \]
**Phase-Ordering in Cu$_3$Au**

- high $T$: fcc sites occupied by either Cu or Au
- $T \leq T_c = 383 \, ^\circ C$: ordering with Au on corner and Cu on face sites
- 4-fold degenerate ground-state

(corners can be chosen in 4 different ways)

groundstates separated by domain walls

domain walls give rise to ellipse shaped superlattice reflections of type [100]

quench: domain formation and growth in disordered phase

domain coarsening with $R \sim t^{1/2}$

[100] superlattice reflection after quench from 425 $^\circ C$ to 370 $^\circ C$

Fluerasu & Sutton, PRL94(2005)55501
Study fluctuations about the average behaviour: XPCS characterize by two-time correlation function

\[ C(q,t_1,t_2) = \frac{<l(t_1) l(t_2)> - <l(t_1)> <l(t_1)>}{[<l^2(t_1)> - <l(t_1)>^2]^{1/2} [<l^2(t_2)> - <l(t_1)>^2]^{1/2}} \]
rescaled correlation time $\tau \sim t_{\text{mean}}$

$\sim t_{\text{mean}}^{1/2}$

in the low $t_{\text{mean}}$ limit

in the high $t_{\text{mean}}$ limit

with $dq = Q - [100]$
XPCS – operating range

- access to large momentum transfers \((Q_{\text{max}}=2\pi \cdot \sin \theta / \lambda)\) or short lengthscales
- not subject to multiple scattering
- can be combined with the surface sensitivity of X-rays
Imaging and XPCS at an FEL source

\[ F_c = \left(\frac{\lambda}{2}\right)^2 \cdot B(\text{rlliance}) \]

\[ B(\text{storage ring}) \approx 10^{20} - 10^{21} \]

Length of a storage ring pulse \( \approx 50-100 \text{ ps} \)
(X)FEL key parameters

- X-ray FEL radiation (0.2 - 14.4 keV)
  - ultrashort pulse duration: 100 fs
  - extreme pulse intensities: $10^{12}$-$10^{14}$ ph
  - coherent radiation: $10^9$
  - average brilliance: $10^4$
LINAC driven SASE free-electron laser
The radiation emitted by a single electron in subsequent oscillations in an undulator is in phase. Radiation from different electrons is NOT (positional disorder in bunch).

“Phasing” is achieved via positional order in the bunch (micro-bunching) with a period equal to the x-ray wavelength.
SASE (self-amplified spontaneous emission)

Idea: Send a perfect electron beam through a very long undulator using the spontaneous radiation as a seed

Generation of coherent radiation by a relativistic electron beam in an undulator
Collective instabilities and high-gain regime in a free electron laser
Simulation

GENESIS – simulation for TTF parameters

Courtesy Sven Reiche (UCLA)
Time Structure and Coherence Properties of XFEL radiation

Get single spatially and temporally coherent wave packet carrying about $10^9$ photons.

#ph/mode $\approx 10^9 = \frac{#ph/bunch}{M(#\text{ modes})}$

$= 10^{12} / 1000$

$M < 1000$

Pulse-to-pulse statistics:

M=1: single mode
strong pulse-to-pulse fluctuations

M>1: multi-mode

for $\Delta\lambda/\lambda / N(=1000) = 10^{-6}$:

$0 \quad 1 \quad 2 \quad 3$  
$E/\langle E \rangle$

$P_M(E)$
The FLASH facility

\[ \lambda \geq 4.5 \text{ nm} \]

Commissioning: 2004/5
User experiments: 2005
FLASH Overview

- Jan 2005: first lasing at 32 nm
- Aug 2005: first user exper.
FLASH Performance

FLASH performance:

- Pulse duration $\leq 20$ fs
- 1st: 13.7 nm $B_{\text{peak}} = 6 \times 10^{29}$
- 3rd: 4.6 nm $B_{\text{peak}} = 2 \times 10^{28}$
- 5th: 2.75 nm $B_{\text{peak}} = 2 \times 10^{27}$
Double Slit Diffraction at FLASH

$\lambda = 100 \text{ nm}$

- $d=0.5\text{mm}$
- $d=1\text{mm}$
- $d=2\text{mm}$
- $d=3\text{mm}$

Thesis Rasmus Ischebeck
Double Slit Diffraction at FLASH

Wavelength $\lambda = 25.6$ nm, Slits separation $0.15$ $\mu$m, Image is a sum of 10 FEL pulses
Imaging at a FEL
Incident FEL pulse: 25 fs, 32 nm, $4 \times 10^{14}$ W cm$^{-2}$ ($10^{12}$ ph/pulse)


Model structure in 20 nm SiN membrane

Speckle pattern recorded with a single (25 fs) pulse

Reconstructed image
First FEL pulse:
25 fs, 32 nm, 
$4 \times 10^{14} \text{ W cm}^{-2}$
$(10^{12} \text{ ph/pulse})$

“destroyed” model structure

Second FEL pulse:
25 fs, 32 nm, 
$4 \times 10^{14} \text{ W cm}^{-2}$
$(10^{12} \text{ ph/pulse})$
An approach to three-dimensional structures of biomolecules by using single-molecule diffraction images: A simulation

3-D structure (2.5 Å resolution) of rubisco molecule.

(106 kDa)

Top view of a section (kz=0) of 3-D scattering pattern from $10^6$ single molecules (of known relative orientation) each “exposed” by a single 10 fs XFEL pulse ($\lambda=1.5\text{Å}$, 0.1μm beamsize) containing $2 \times 10^{12}$ photons.

Reconstructed 3-D pattern (from 250 2-D projections). Phasing by “oversampling” technique.


**NOTE: Radiation Damage**
Beam – Sample Interaction

Coulomb explosion of a small protein (lysozyme)

- 50 fs
- $3 \times 10^{12}$ photons/100 nm spot
- 12 keV

Radiation damage interferes with atomic scattering factors and atomic positions

Magnetic Small Angle Scattering at FLASH (1)

Magnetic Small Angle Scattering:

50 [Co(4Å) / Pt(7Å)] sputtered on 20nm Pt layer on Si₃N₄ membrane, capped with 2nm Pt

FLASH operation 19.12.2007:
SASE on 5th harmonic of 7.97 nm = 1.59 nm = 778 eV
Magnetic Small Angle Scattering at FLASH (2)

meandering magnetic stripe domain of a CoPt multilayer

Magnetic SAXS pattern:

E = 778.1 eV

ON (Co L_{III}) resonance
Time-dependent Imaging and XPCS at a FEL
Time delay holography (1)

25 fs long single pulses ($\approx 10^{12}$ ph/pulse) of 32.5 nm light from FLASH impinging on a particle coated (140 nm polystyrene PS particles) Si$_3$N$_4$ membrane.

The primary diffraction pattern (blue) interferes with the secondary (red) diffraction pattern arising from the (incident) beam being backreflected by a mirror and diffracted after a time delay $\Delta t$ from the “exploding” PS particles.
• Time delay holography (2)

\[ \lambda = 32.5 \text{ nm} \]
\[ 25 \text{ fs duration} \]
\[ 0.5 \times 10^{14} \text{ W/cm}^2 \]

\[ \Delta t = 348 \text{ fs} \]

\[ \Delta t = 733 \text{ fs} \]

The intensity envelope of the interference pattern evolves to lower q indicating increasing radii of the particles.
XPCS at a XFEL source:

100 ms  

100 ms

600 μs

99.4 ms

X-ray photons

FEL process

100 fs

1ps < t< 10 ns:  “delay-line” mode

for “all” times:  “pump-probe” mode

200ns < t < 600 μs:

” movie” mode

t > 0.1 s
XPCS at a FEL source: Movie Mode

\[ g_2(\Delta t) = \frac{\langle I(Q,t)I(Q,t+\Delta t) \rangle}{\langle I \rangle^2} \]

\[ \text{correlation function} \]

\[ I(Q,t) \]
The Quest for fast 2-D Detectors

Pilatus detector module: 172 µm pixel size
Silica particles suspended in PPG
sum of 5000 frames with 30 ms exposure
data taken at cSAXS/SLS
The Quest for fast 2-D Detectors

Data acquisition time with 0-D detector: \( \approx 2-3 \) days

Pilatus: sum of 5000 frames with 30 ms exposure separated by a 20 ms delay btw. 2 frames:

Total data acquisition time: \( 5000 \times 50 \) ms = 250 s

Estimated data acq. at XFEL:

\( 3000 \times (100 \text{fs} + 200 \text{ns}) \approx 600 \mu \text{s} \)
Delay Line Mode

“Delay Line” Mode: \( 1 \text{ps} < \Delta t < 10\text{ns} \) (1 ps \( \approx \) 0.3 mm; 1 ns \( \approx \) 300 mm) “luminosity limited.”
Delay Line for “hard” X-Rays

Si(511) at 8.38 keV

\[ \Delta t_{\text{max}} = 2.8 \text{ ns} \]

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
XPCS at a FEL source

Magnetization Dynamics

Ferroelectrics

Ultrafast dynamics at surfaces and interfaces of liquids

......
Ultrafast demagnetization

Optical Excitation

Electrons

Phonons

Electron-phonon relaxation time

Electron-spin relaxation time

Spin-lattice relaxation time

Magnetization

- spin lattice excitations: domains, vortices, spin waves & their non-linear interactions
- optical pump – x ray scattering probe


Coherence of light and matter: from basic concepts to modern applications, Vorlesung im GrK 1355, SS 2011A. Hemmerich & G. Gruebel
Magnetic SAXS data – time dependant

Time-delay scan for 18 mJ/cm² pumping power, $\Delta t = -1.2 \text{ ps...34.5 ps}$
Ultra-fast demagnetization from SAXS data

High pump fluence, characteristic demagnetization time below 1 ps
Polarization Switching in Ferroelectrics

Domain wall motion during polarization switching in ferroelectric Pb(Zr,Ti)O$_3$

PZT (002) reflection
(0.1 $\mu$m, 10 keV, APS)

polarization switching:
Nucleation of domains with reversed polarity, followed by domain growth with domain wall velocities of here about 40m/s.

Study fluctuations and domain growth via XPCS.

XPCS at a XFEL source: An example

Ultrafast dynamics at surfaces and interfaces in:
- liquids,
- membranes,
- ...

Today: \( Q_{\text{max}} \approx 10^{-4} \text{ nm}^{-1} \) (water)

XFEL:
- Onset of non-classical behaviour \((Q > 1 \text{ nm}^{-1}, \text{ beyond continuum hydrodynamics})\)
- Study capillary wave dynamics at high \( Q \) in model systems \((\lambda=1\text{Å}, Q=1 \text{ nm}^{-1})\)

\[ \tau [\text{s}] \text{ countrate (FEL)} \]

Water \( \approx 25 \text{ ps} \quad 20 \)
Mercury \( \approx 0.5 \text{ ps} \quad 0.3^* \)
Glycerol \( \approx 1 \text{ µs} \quad 3000 \)

* expect important deviations due to layering at the surface

G. Grübel et al., TDR XFEL, DESY (2006)
G. Grübel et al., NIM B, submitted

C. Gutt et al., PRL 91 (2003)76104

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The XFEL

www.xfel.eu
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