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

Part III

Vorlesung im GrK 1355

SS 2011

A. Hemmerich & G. Grübel

Location: SemRm 052, Gebäude 69, Bahrenfeld

Tuesdays 10.15 – 11.45

G.Grübel (GR), A.Hemmerich (HE), M.Yurkov (YU)

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

12.4.	Introduction	(HE)
19.4+3.5.	Coherence of classical light , basic concepts and examples	(HE)
10.+17.5.	Coherence of quantized light, basic concepts and examples	(HE)
24.5.	Coherence of matter waves	(HE)
31.5.	Coherence properties of the radiation from x-ray free electron lasers	(YU)
7.6.	Coherence based X-ray techniques: Introduction	(GR)
21+28.6.	Imaging techniques	(GR)
5.+12.7.	X-ray Photon Correlation Spectroscopy	(GR)

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://www.photon.physnet.uni-hamburg.de/ilp/ hemmerich/teaching.html**

Part II+III: **http://hasylab.desy.de/science/studentsteaching/lectures/ss11/ graduiertenkolleg/……**

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

Part III: G. Grübel Script 1

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

▪

Coherence based X-ray techniques:

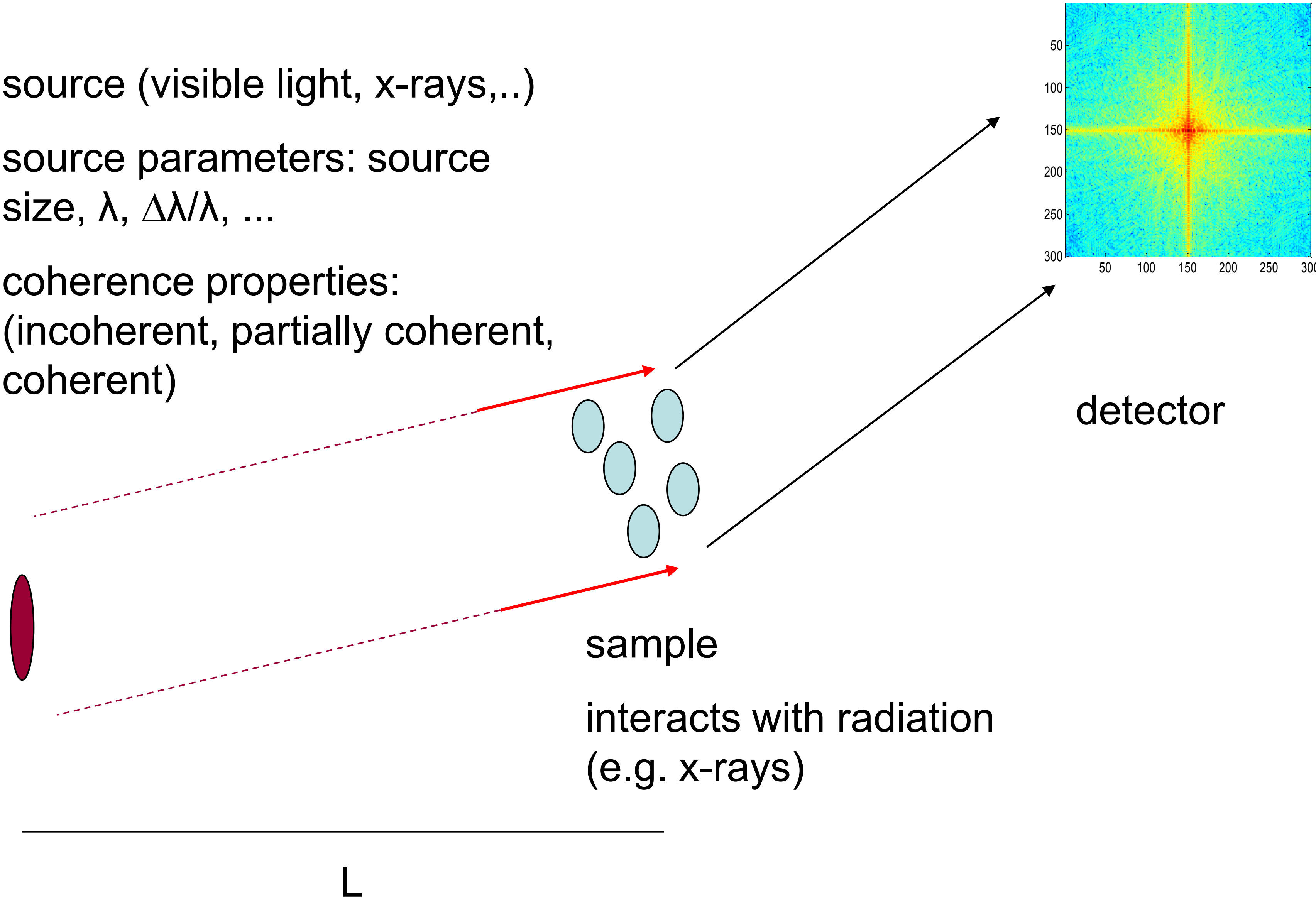
Introduction

Introduction: Experimental Set-Up

source (visible light, x-rays,..)

source parameters: source size, λ, Δλ/λ, ...

coherence properties: (incoherent, partially coherent, coherent)



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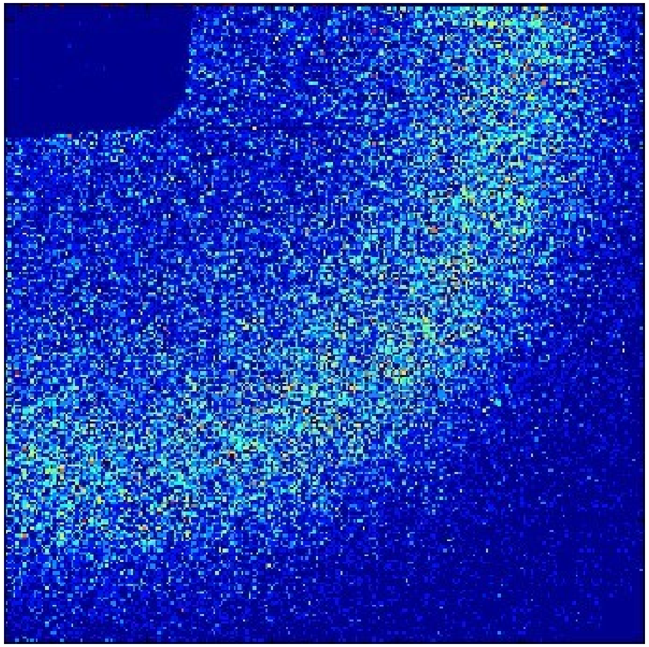
Introduction: Scattering with coherent X-rays

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:

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

Introduction: Speckle Pattern

A speckle pattern contains information on both, the source and sample that produced it.

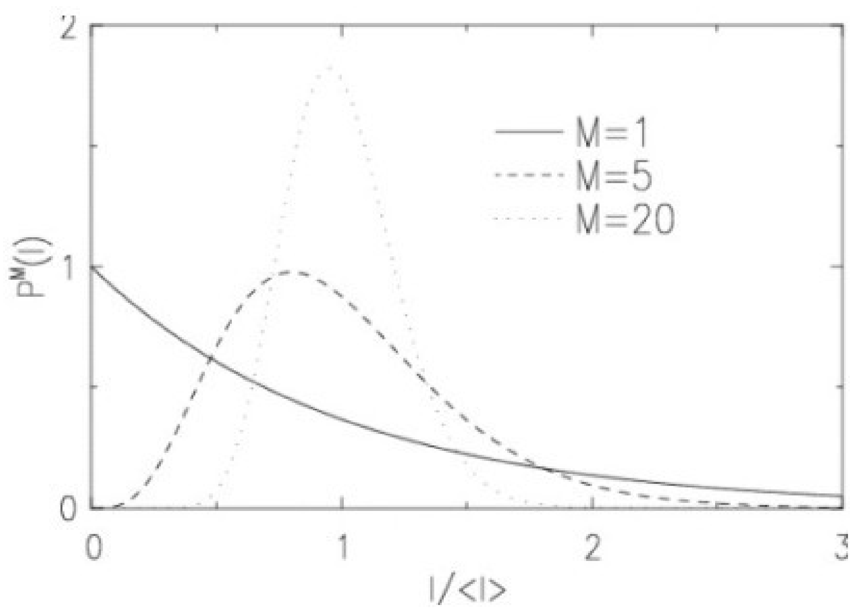
If the source is fully coherent and the scattering amplitudes and phases of the scattering are statistically independent and distributed over 2π one finds for the probability amplitude of the intensities:

$$P(I) = (1/\langle I \rangle) \exp(- I/\langle I \rangle)$$

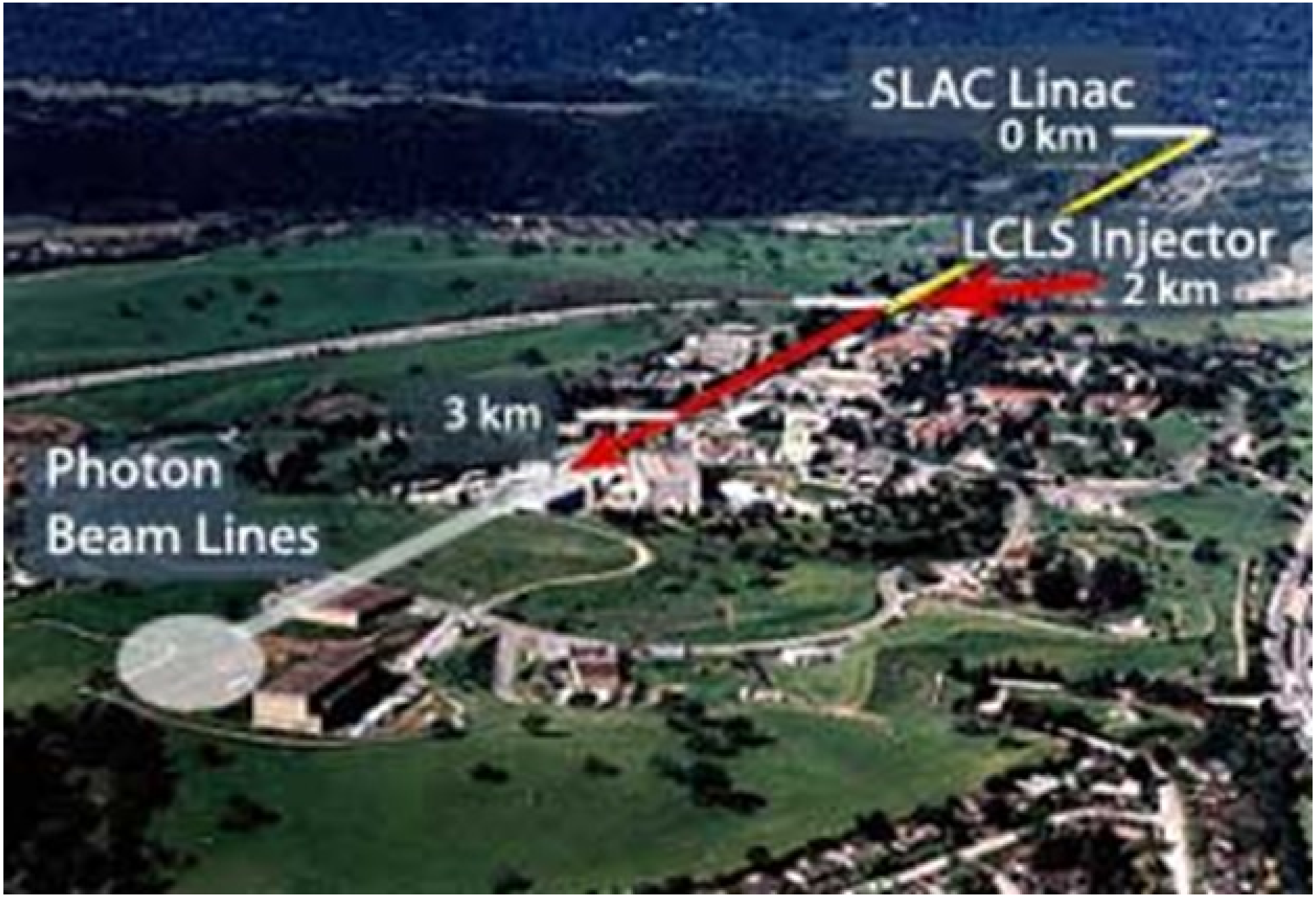
Mean: $\langle I \rangle$

Std.Dev. σ: $\text{sqrt}(\langle I^2 \rangle - \langle I \rangle^2) = \langle I \rangle$

Contrast: $ß = \sigma^2/\langle I \rangle^2 = 1$



▪ The Linac Coherent Light Source (LCLS)



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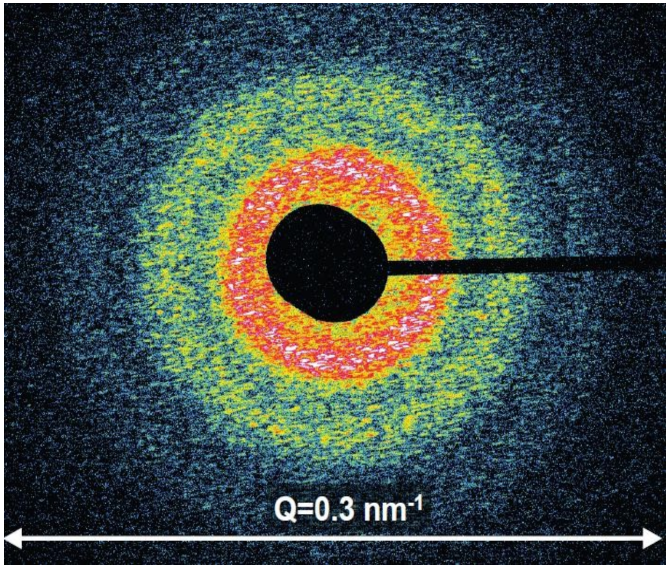

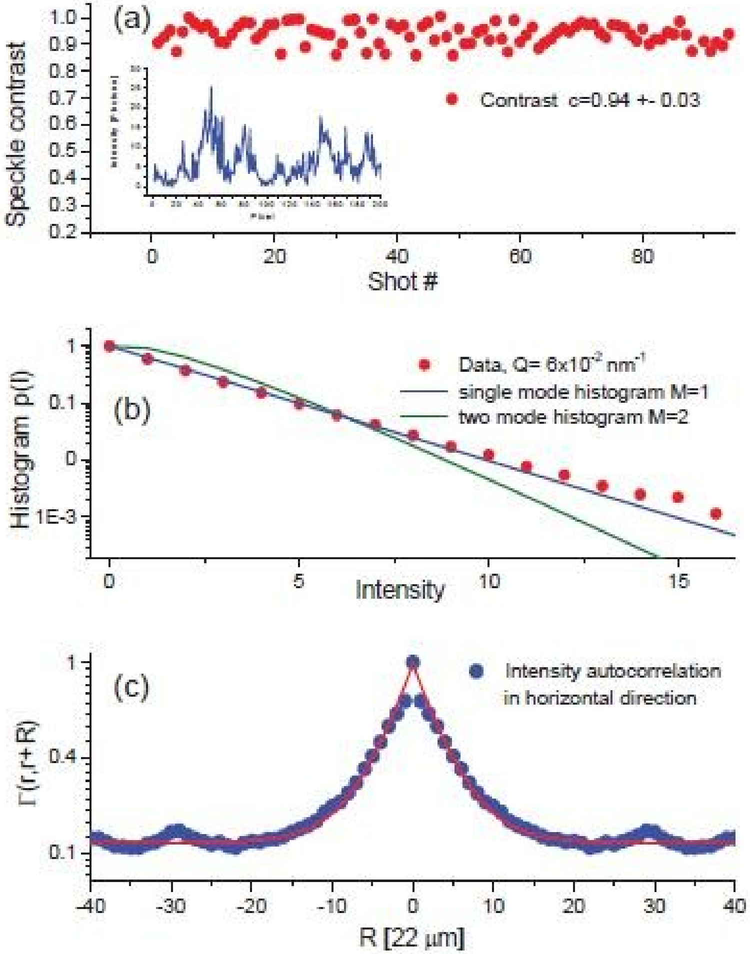

Single pulse hard X-ray speckle pattern captured from nano-particles in a colloidal liquid (photon wave-length λ= 1.37 Å

▪

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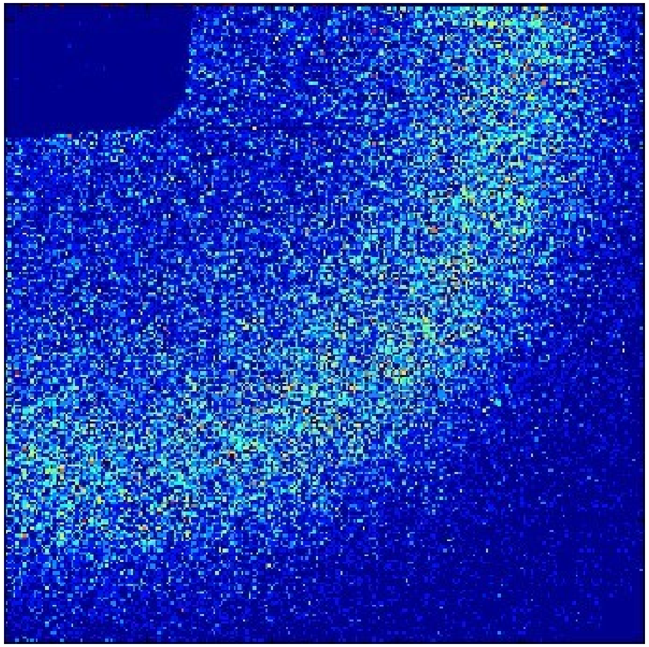


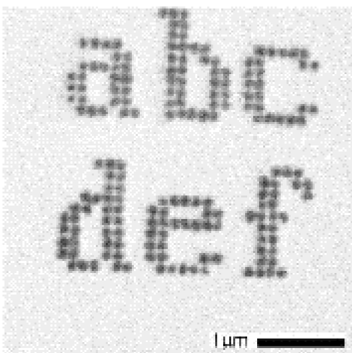
j in coherence volume $c = \xi_t^2 \xi_l$

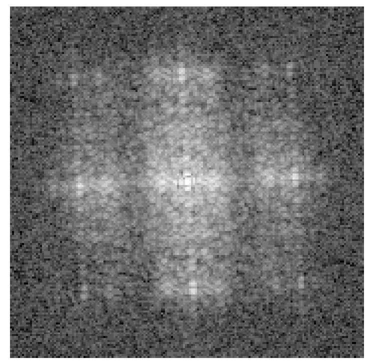
<u>Incoherent Light:</u>

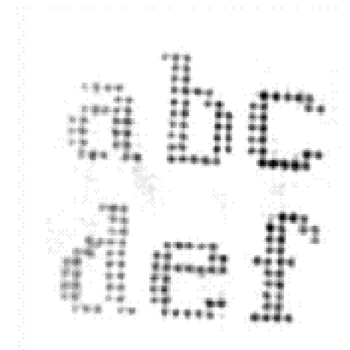
$S(Q,t) = \langle S_c(Q,t) \rangle_{V>>c}$ ensemble average

Introduction: Speckle Reconstruction

Reconstruction (phasing) of a speckle pattern: **"oversampling" technique**







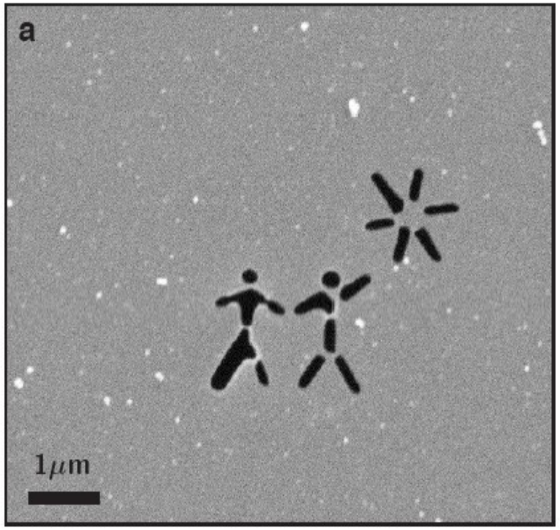
gold dots on SiN membrane (0.1 µm diameter, 80 nm thick)

λ=17Å coherent beam at X1A (NSLS), $1.3 \cdot 10^9$ ph/s 10µm pinhole 24 µm x 24 µm pixel CCD

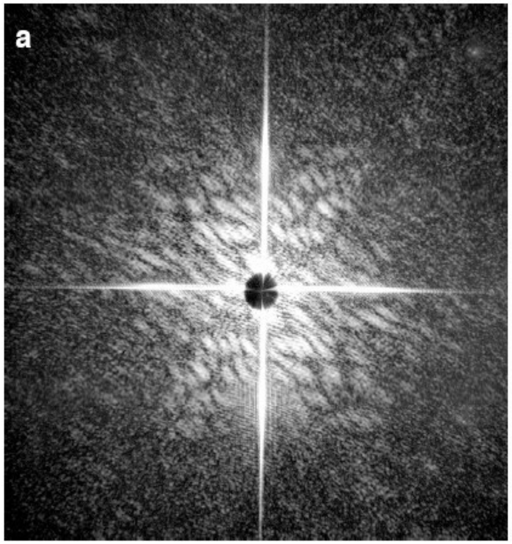
reconstruction "oversampling" technique

Miao, Charalambous, Kirz, Sayre, Nature, 400, July 1999

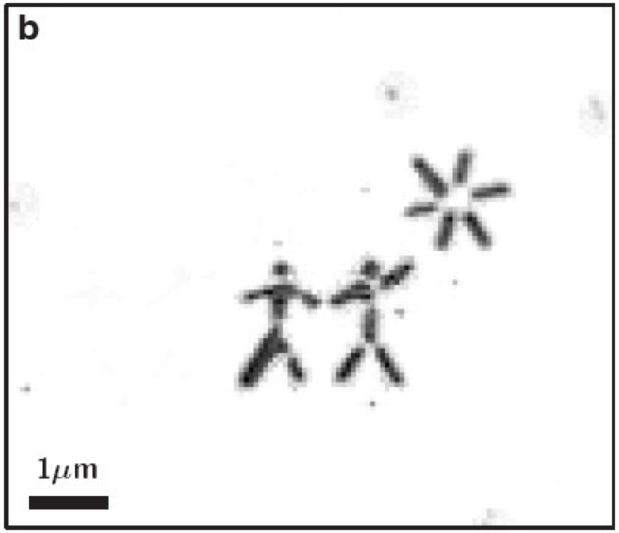
Reconstruction of “oversampled” data



Model structure in 20 nm SiN membrane

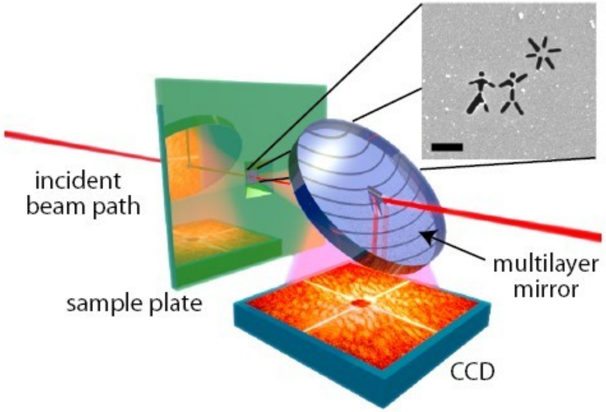


Speckle pattern recorded with a single (25 fs) pulse



Reconstructed image

Incident FEL pulse:
25 fs, 32 nm,
4 x 10^{14} W cm^{-2} (10^{12} ph/pulse)



H. Chapman et al., Nature Physics, 2,839 (2006)

Introduction: Scattering with coherent X-rays

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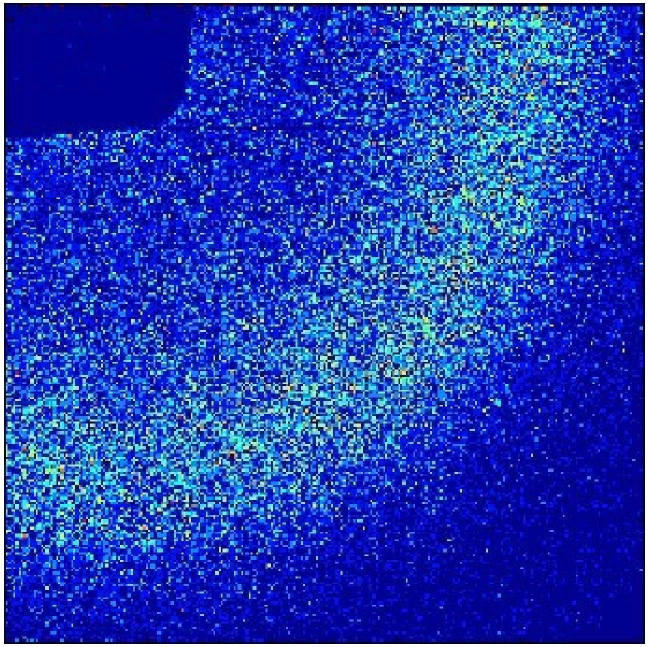

j in coherence volume $c = \xi_t^2 \xi_l$

Incoherent Light:

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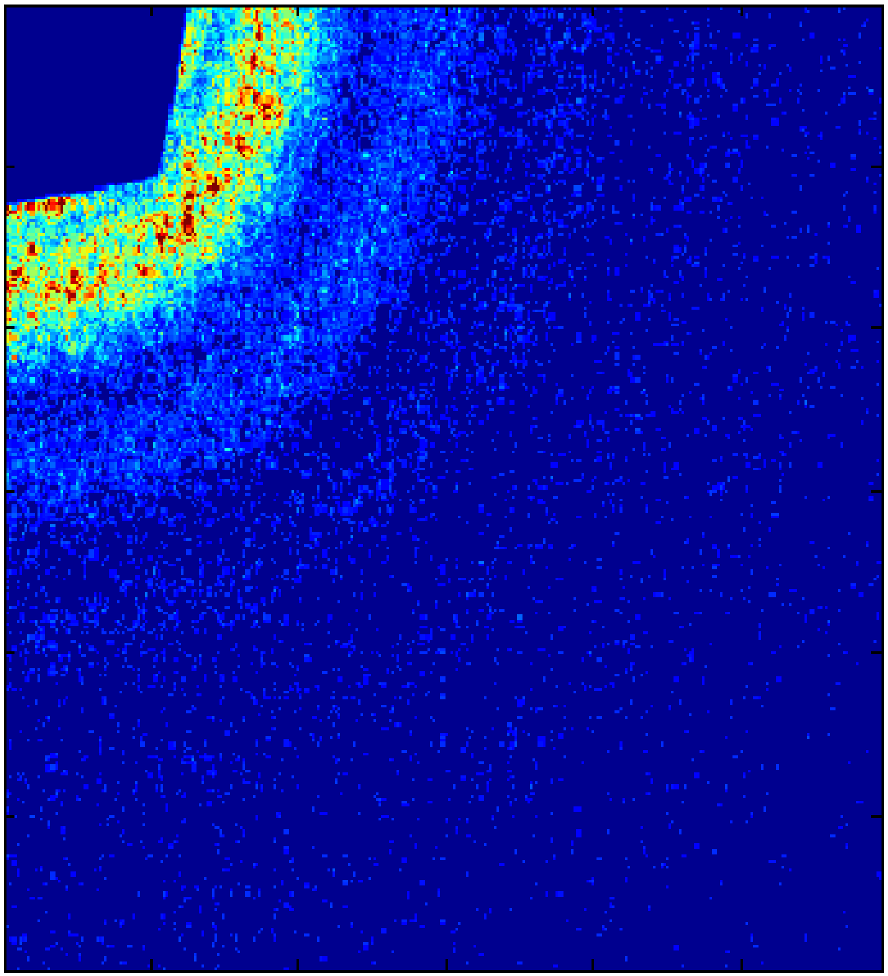
 Coherence of light and matter: from basic concepts to modern applications, Vorlesung im GrK 1355, SS 2011 A. Hemmerich & G. Gruebel

Introduction: X-ray Photon Correlation



colloidal silica particles undergoing Brownian motion in high viscosity glycerol

V. Trappe and A. Robert

- 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:

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

$\langle\rangle$ 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

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

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Coherence based X-ray techniques:

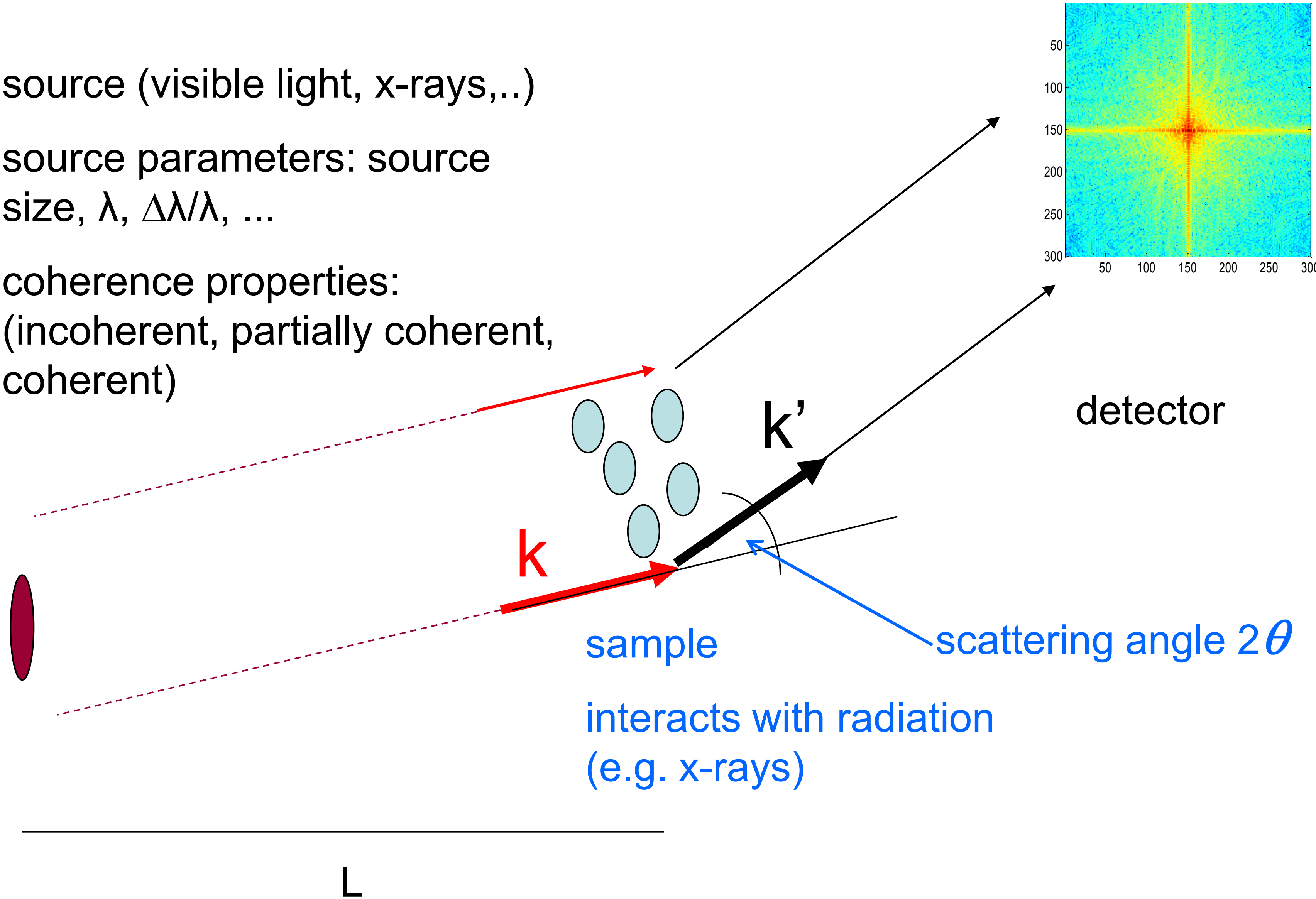
An X-ray Scattering Primer

Experimental Set-Up for Scattering Experiments

source (visible light, x-rays,..)

source parameters: source size, λ, Δλ/λ, ...

coherence properties: (incoherent, partially coherent, coherent)



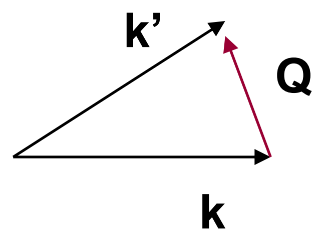
Scattering of X-rays: A primer

consider a monochromatic plane (electromagnetic) wave with wavevector k:

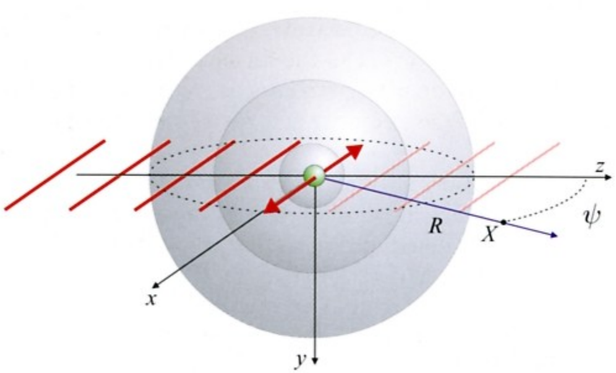
$\mathbf{E}(\mathbf{r},t) = \boldsymbol{\varepsilon}\, Eo\, \exp\{i(\mathbf{kr}-\omega t)\}$ with $|\mathbf{k}|=2\pi/\lambda$, $\lambda[Å]=hc/E$, $\omega=2\pi/\nu$

elastic scattering:

$\hbar\, \mathbf{k'} = \hbar\, \mathbf{k} + \hbar \mathbf{Q}$



Scattering by a single electron:



$E_{rad}(R,t)/E_{in} =$

$-(e^2/4\pi\varepsilon_o mc^2)\exp(ikR)/R \cos\psi$

spherical wave

thomson scattering length r_o

$(=2.82 \cdot 10^{-5}$ Å)

▪

scattered intensity:

$I_S/I_o = |E_{rad}|^2 R^2\Delta\Omega \,/\, |E_{In}|^2 A_o$

$R^2\Delta\Omega$: solid angle seen by detector
A_o incident beam size

$$I_s = (d\sigma/d\Omega)\,(I_o/A_o)\,\Delta\Omega$$

with the differential cross section (for Thomson scattering)

$$(d\sigma/d\Omega) = r_o^2 P \qquad P = \begin{cases} 1 & \text{vertical} \\ \cos^2\psi & \text{horizontal} \\ \tfrac{1}{2}(1+\cos^2\psi) & \text{unpolarized} \end{cases}$$

note: $\sigma_{total} = \int (d\sigma/d\Omega) = (8\pi/3)\, r_o^2$

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scattering by a single atom:

phase factor

scattering amplitude by an ensemble of electrons

$$-r_o f^o(Q) = -r_o \sum_{r_j} \exp(iQ \cdot r_j)$$

(atomic) formfactor — position of scatterers

$$f^o(Q \to 0) = Z, \quad f^o(Q \to \infty) = 0$$

form factor of an atom:

$$f(Q,\hbar\omega) = f^o(Q) + f'(\hbar\omega) + i f''(\hbar\omega)$$

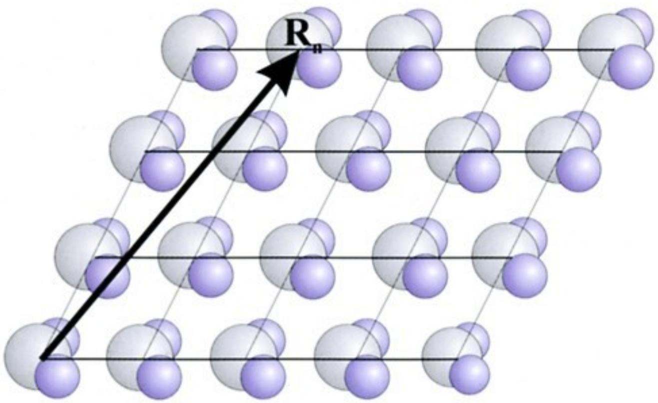
dispersion corrections: level structure — absorption effects

scattering intensity:

$$I_s = r_o^2 f(Q) f^*(Q) P$$

▪

scattering by a crystal:



$r_{j'} = R_n + r_j$

lattice vector + atomic position in lattice

$F^{crystal}(Q) = \Sigma_{rj}\, f_j(Q)\exp(iQr_j)\Sigma_{Rn}\exp(iQR_n)$

unit cell structure factor — lattice sum

$I_s = r_o^2\, F(Q)\, F^*(Q)\, P$

lattice sum ≡ phase factor of order unity or N (number of unit cells) if

$Q \bullet R_n = 2\pi \text{ x integer}$ ($)

▪

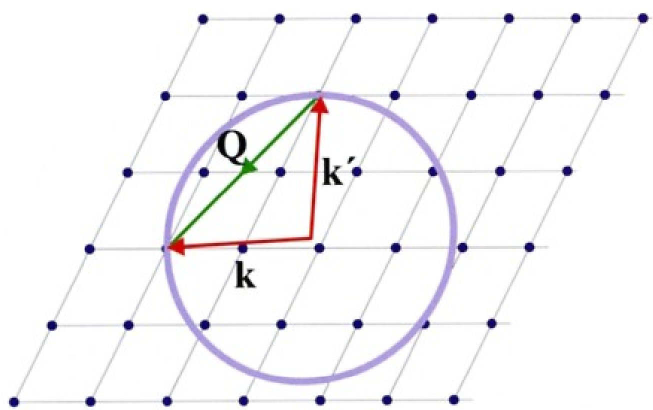
evaluation of lattice sums:

construct reciprocal space such that: $a_i \bullet a_j^* = 2\pi\ \delta ij$

with a_i defining a

reciprocal lattice *such that* $G = h\ a_1^* + k\ a_2^* + l\ a_3^*$

and G fullfills ($) for Q = G (Laue condition)



$k + Q = k'$

Ewald sphere

$\sin(\theta/2) = (Q/2) / k$

Laue condition ≡ Bragg's law

lattice sum: $|\Sigma_{Rn} \exp(iQR_n)|^2 \rightarrow N\ v_c^*\ \delta\ (Q\text{-}G)$

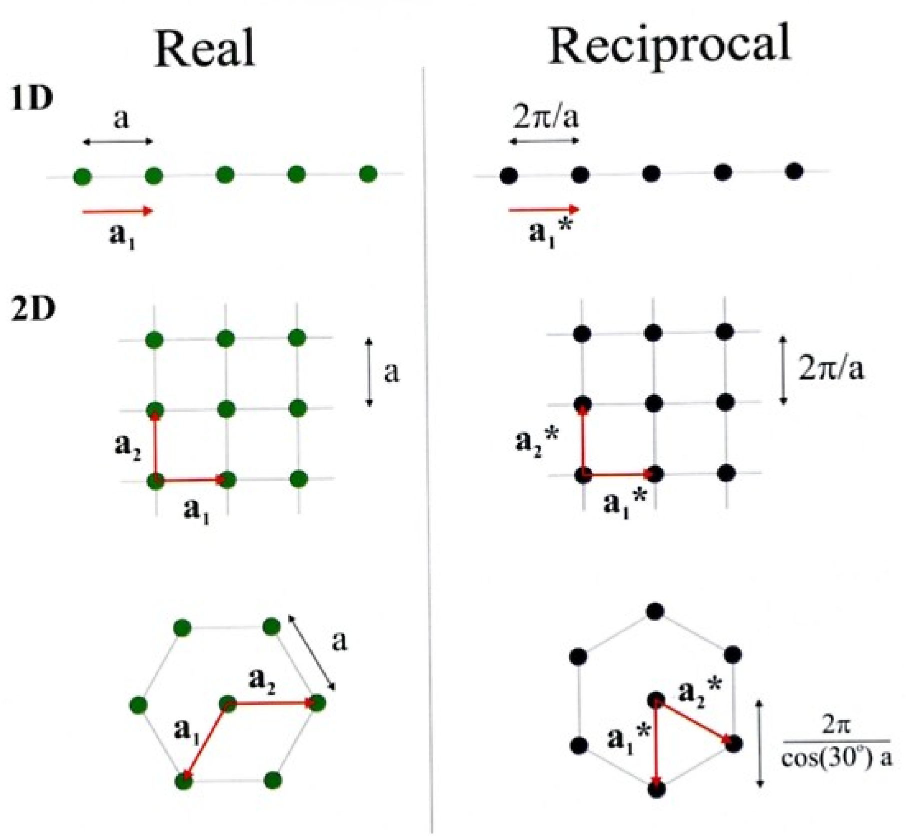
N number of unit cells; v_c^* unit cell volume in reciprocal space

▪

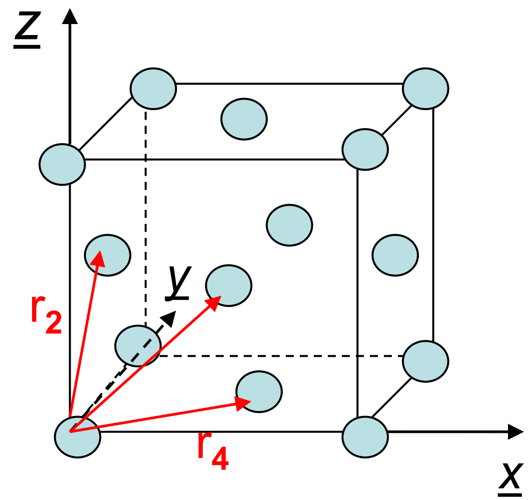
construction of reciprocal space:

(real space lattice constants a_1, a_2, a_3); $v_c = a_1 \bullet (a_2 \times a_3)$

$a_1^* = 2\pi/v_c\ (a_2 \times a_3)$ $a_2^* = 2\pi/v_c\ (a_3 \times a_1)$ $a_3^* = 2\pi/v_c\ (a_1 \times a_2)$



The unit cell structure factor



$$F^{uc}(Q) = \sum_{r_j} F_j^{mol}(Q) \exp(iQr_j)$$

<u>example:</u> fcc lattice *(use conventional cubic unit cell)*

$r_1 = 0$, $r_2 = ½\, a\, (\underline{y} + \underline{z})$, $r_3 = ½\, a\, (\underline{z} + \underline{x})$, $r_4 = ½\, a(\underline{x} + \underline{y})$

$$G = ha_1^* + ka_2^* + la_3^*$$

$$a_1^* = 2\pi/v_c\, (a_2 x a_3) = 2\pi/a^3\, [a\underline{y} \times a\underline{z}] = 2\pi/a\, [\underline{y} \times \underline{z}] = 2\pi/a\, \underline{x}$$

$$a_2^* = 2\pi/v_c\, (a_2 x a_3) = 2\pi/a^3\, [a\underline{z} \times a\underline{x}] = 2\pi/a\, [\underline{z} \times \underline{x}] = 2\pi/a\, \underline{y}$$

$$a_3^* = 2\pi/v_c\, (a_2 x a_3) = 2\pi/a^3\, [a\underline{x} \times a\underline{y}] = 2\pi/a\, [\underline{x} \times \underline{y}] = 2\pi/a\, \underline{z}$$

$$v_c = a1 \bullet (a_2 \times a_3)$$

$G \bullet r_1 = 2\pi/a\, (\, hx + ky + lz) \bullet 0 \quad = 0$

$G \bullet r_2 = 2\pi/a\, (\, h\underline{x} + k\underline{y} + l\underline{z}) \bullet 1/2a(\underline{y} + \underline{z}) \quad = \pi\, (k+l)$

$G \bullet r_3 = 2\pi/a\, (\, h\underline{x} + k\underline{y} + l\underline{z}) \bullet 1/2a(\underline{z} + \underline{x}) \quad = \pi\, (h+l)$

$G \bullet r_4 = 2\pi/a\, (\, h\underline{x} + k\underline{y} + l\underline{z}) \bullet 1/2a(\underline{x} + \underline{y}) \quad = \pi\, (h+k)$

- # The unit cell structure factor for a fcc lattice

$$F_{hkl}^{fcc}(\mathbf{Q}) = \sum_{j=1-4} f(Q)\exp(i\mathbf{Q}r_j) = f(Q)\,[\exp(i\mathbf{Gr_1}) + \dots \exp(i\mathbf{Gr_4})]$$

$$F_{hkl}^{fcc}(Q) = f(Q)\,[1 + \exp(i\pi(k+l)) + \exp(i\pi(h+l)) + \exp(i\pi(h+k))]$$

$$= \begin{cases} 4 & \text{if h,k,l are all even or odd} \\ 0 & \text{otherwise} \end{cases}$$

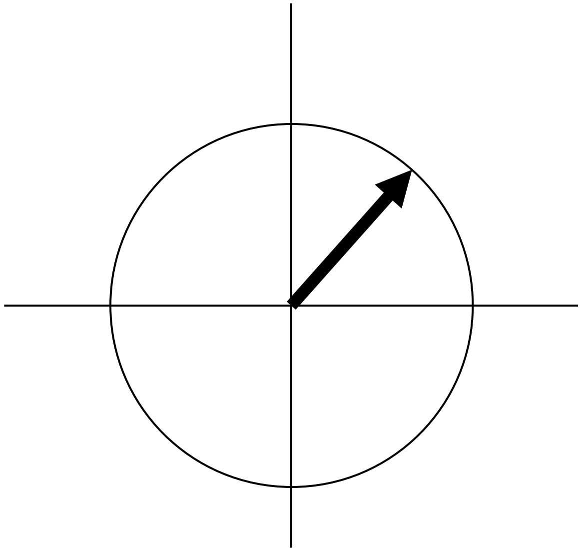
$$I_{hkl}^{fcc}(\mathbf{Q}) = F(\mathbf{Q}) \bullet F^*(\mathbf{Q})$$

<u>Reflections:</u>

100 forbidden

111 allowed

200 allowed



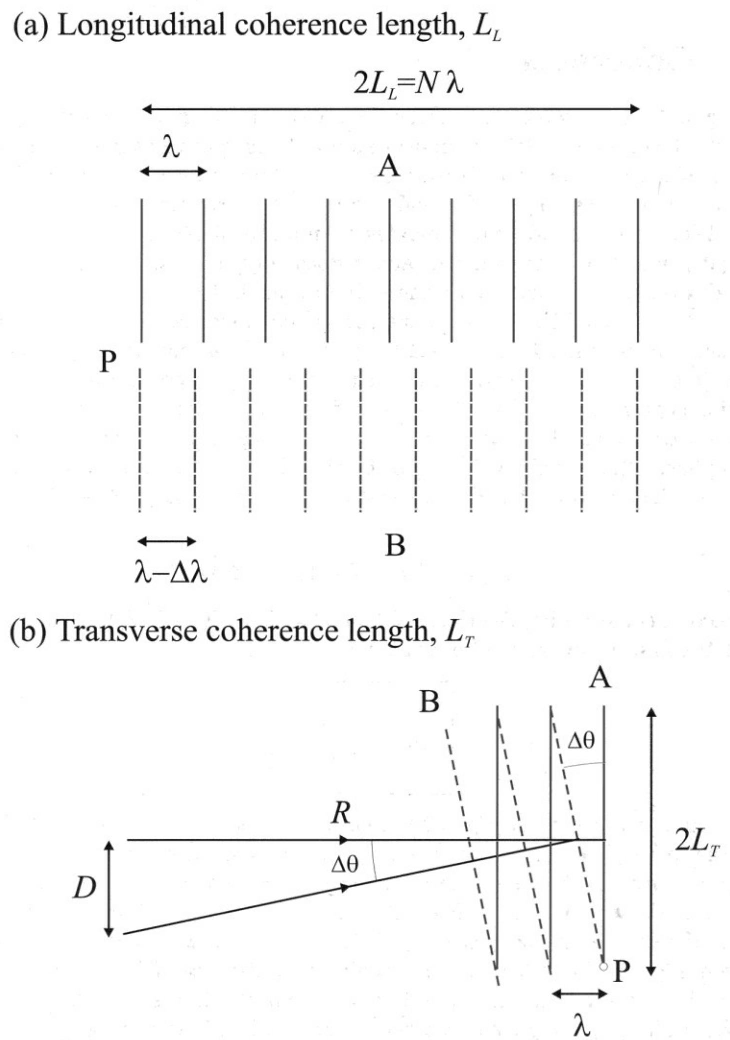
- From a measurement of a (large) set of crystal reflections $|F_{hkl}|^2$ it is possible to deduce the positions of the atoms in the unit cell.

Limitations:

phaseproblem: $| F(Q)| = | F(-Q) |$

$| F(Q) | = | F(Q)e^{i\Phi} |$

Coherence



Longitudinal coherence:

Two waves are in phase at point P. How far can one proceed until the two waves have a phase difference of π:

$$\xi_l = (\lambda/2)\,(\lambda/\Delta\lambda)$$

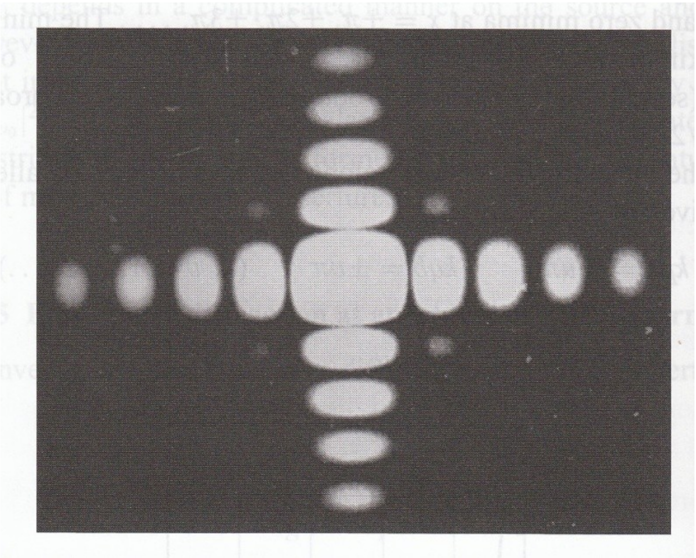
Transverse coherence:

Two waves are in phase at P. How far does one have to proceed along A to produce a phase difference of π:

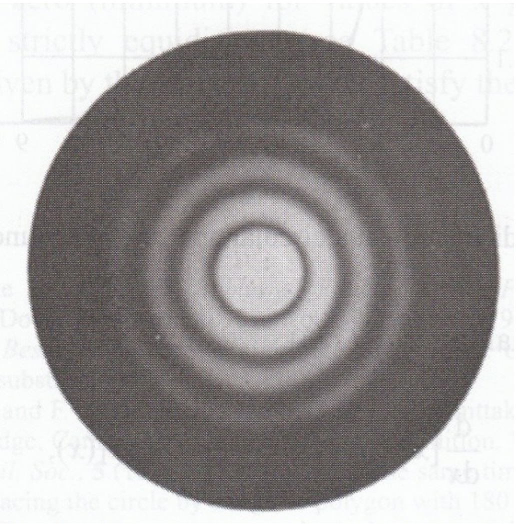
$$2\xi_t\,\Delta\theta = \lambda$$

$$\xi_t = (\lambda/2)\,(R/D)$$

Fraunhofer Diffraction

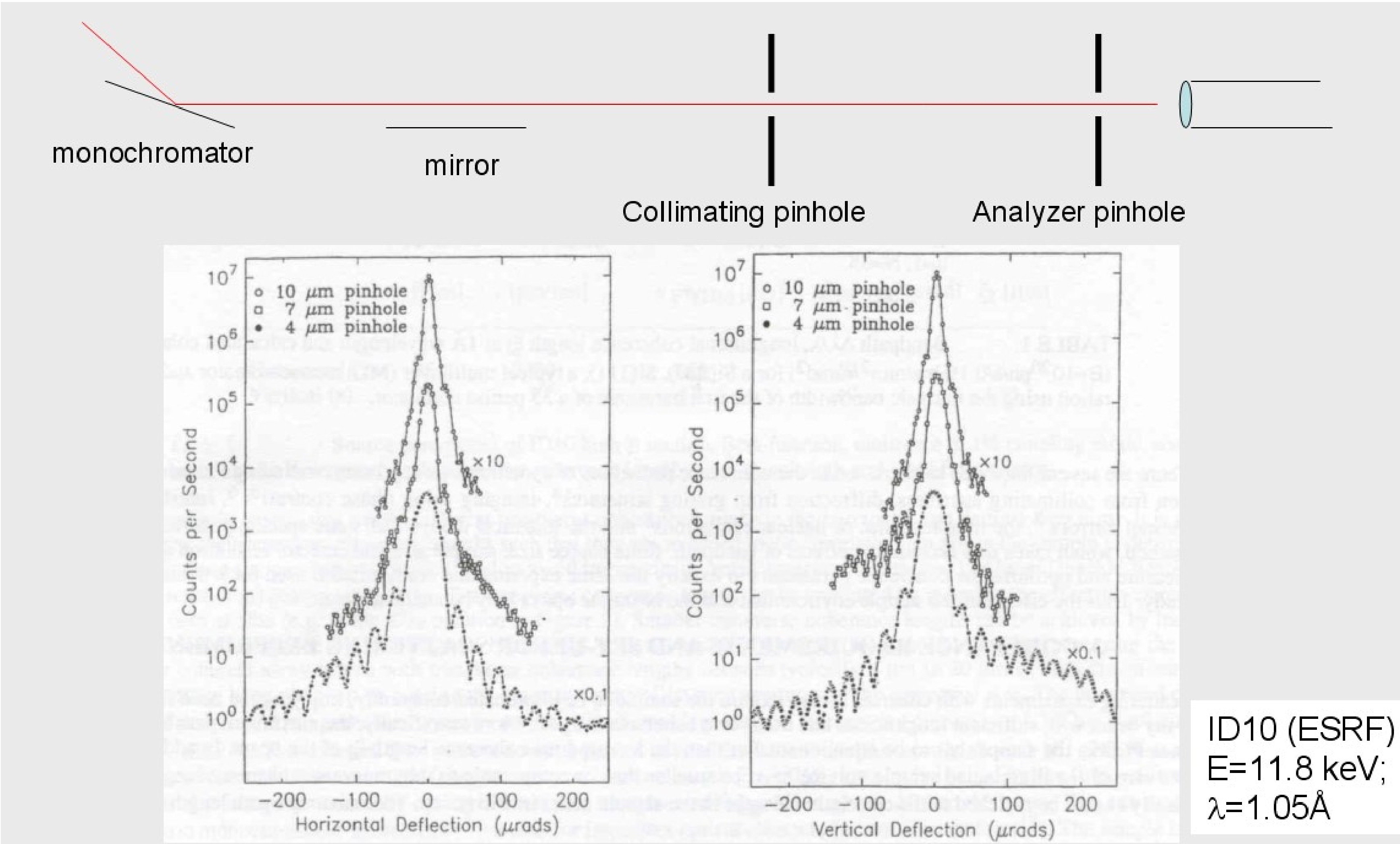


Fraunhofer diffraction of a rectangular aperture 8 x 7 mm^2, taken with mercury light λ=579nm (from Born&Wolf, chap. 8)

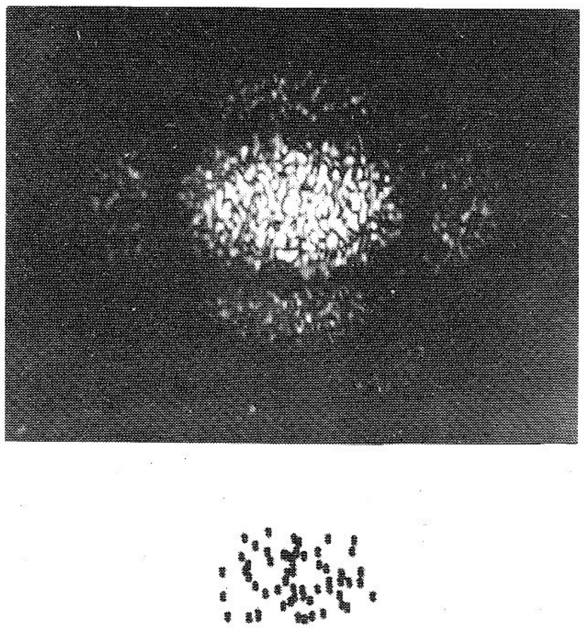


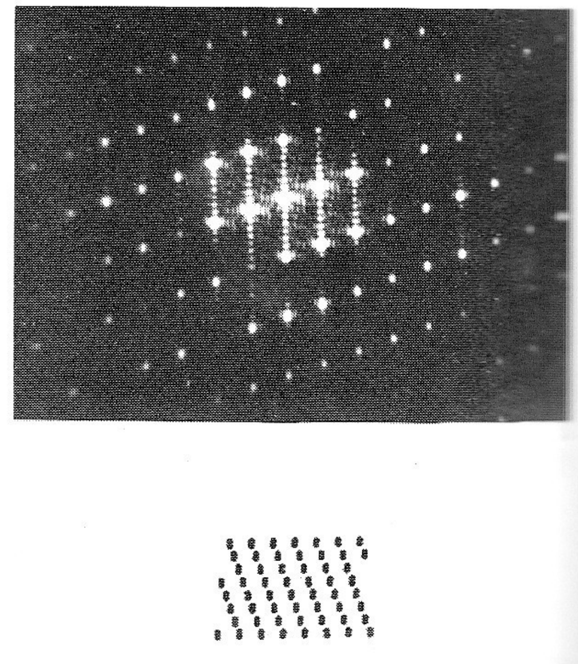
Fraunhofer diffraction of a circular aperture, taken with mercury light λ=579nm (from Born&Wolf, chap. 8)

Fraunhofer Diffraction (λ=0.1nm)



▪ Speckle pattern





random arrangement of apertures: speckle

regular arrangement of apertures