Methoden moderner Röntgenphysik II: Streuung und Abbildung

Lecture 10

Vorlesung zum Haupt/Masterstudiengang Physik

SS 2014

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Location: Hörs AP, Physik, Jungiusstrasse

Tuesdays 12.45 – 14.15

Thursdays 8:30 – 10.00

Methoden moderner Röntgenphysik II: Streuung und Abbildung

Small Angle Scattering, and Soft Matter
Introduction, form factor, structure factor, applications, ..

Anomalous Diffraction
Introduction into anomalous scattering,..

Introduction into Coherence
Concept, First order coherence, ..

Coherent Scattering
Spatial coherence, second order coherence,..

Applications of coherent Scattering
Imaging and Correlation spectroscopy,..

The concept of coherence: classical light

First order coherence

Coherence and emission spectrum

Spatial coherence

Second order coherence

Chaotic light

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)

Courtesy: Andreas Hemmerich

• Reminder: First order coherence

Normalized autocorrelation function: $g^{(1)}(\tau) \equiv \langle E(t+\tau)E^*(t)\rangle / \langle I\rangle$

Correlation of amplitudes

Properties

(1) $g^{(1)}(0)=1$

(2) $g^{(1)}(-\tau)=g^{(1)*}(\tau)$

Longitudinal coherence

$\xi_l = \lambda/2\ \lambda/\Delta\lambda$

Michelson-interferometer

Spatial coherence

$d \cong \lambda/\theta$

Young double slit

▪ Second Order Coherence

Normalized autocorrelation function: $g^{(2)}(\tau) \equiv \langle I(t+\tau)I^*(t)\rangle / \langle I(t)\rangle^2$

Correlation of intensities — degree of second order coherence

(1) $g^{(2)}(-\tau) = g^{(2)}(\tau)$

(2) $g^{(2)}(0) \geq 1$

(3) $g^{(2)}(\tau) \leq g^{(2)}(0)$

(4) $g^{(2)}(\tau \rightarrow \infty) = 1$ if correlations vanish

Proof (2): $(1/N \, {}_{n=1}\sum^N I_n)^2 =$

$$1/N^2 \left({}_n\sum I_n^2 + {}_{n\neq m}\sum I_n I_m\right)$$

$$\leq 1/N^2\left({}_n\sum I_n^2 + {}_{n\neq m}\sum (I_n^2+I_m^2)/2\right)$$

(*inequality of arithmetic and geometric means*)

$$= 1/N^2 \, {}_{n,m}\sum (I_n^2+I_m^2)/2$$

$$= 1/N \, {}_{n,m}\sum I_n^2$$

$$\Rightarrow g^{(2)}(0) = \langle I(t)^2\rangle / \langle (I(t)\rangle^2 = 1/N_{n,m}\sum I_n^2 \,/\, (1/N \, {}_{n=1}\sum^N I_n)^2 \geq 1$$

Proof (3):

$$(\langle I(t+\tau)I(t)\rangle)^2 = \left(1/N \sum_{n=1}^{N} I(t_n+\tau)I(t_n)\right)^2 \qquad \textit{(Cauchy-Schwarz inequality)}$$

$$\leq \left(1/N \sum_{n=1}^{N} I(t_n+\tau)^2\right)\left(1/N \sum_{n=1}^{N} I(t_n)^2\right) = (\langle I(t)^2\rangle)^2$$

Proof (4):

$$\tau \rightarrow \infty \quad \Rightarrow \quad \langle I(t+\tau)I^*(t)\rangle = \langle I(t+\tau)\rangle\langle I(t)\rangle = \langle I(t)\rangle^2$$

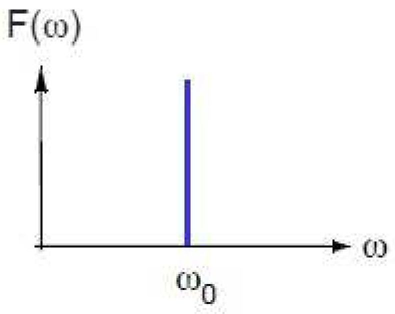
Example: monochromatic light

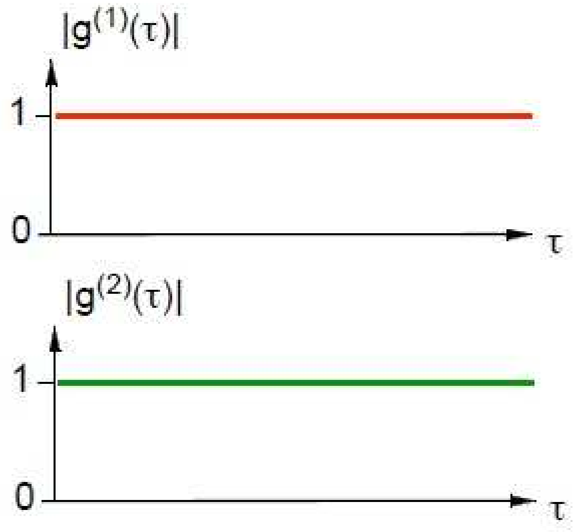
$E(t) = E_0 \exp[i(\omega_0 t + \phi)]$

$I(t) = E_0 E_0^*$

$|g^{(1)}(\tau)| = 1$

$g^{(2)}(\tau) \equiv \langle I(t+\tau)I^*(t)\rangle / \langle I(t)\rangle^2 = 1$





Chaotic Light:

$E(t) = E_0 \, {}_{n-1}\Sigma^N \exp[i\phi_n(t)]$, $\phi_n(t)$ = random phase, uniform at any time t

$\langle \exp[i\phi_n(t+\tau) - \phi_m(t)] \rangle = 0$ if $n \neq m$,

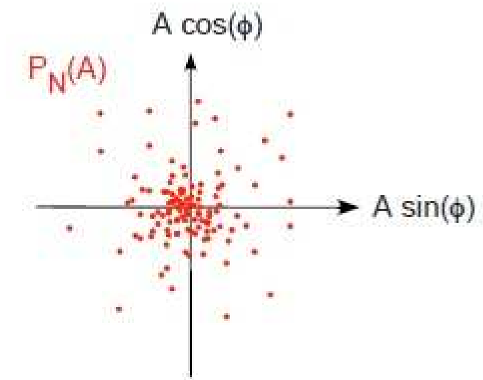
$$g^{(1)}(\tau) = {}_{n-1}\Sigma^N \langle \exp[i(\phi_n(t+\tau) - \phi_n(t))] \rangle$$

Theory of stochastic processes:

Probability for ${}_n\Sigma^N \exp[i\phi_n]$ to fall within unit areas at the point (A,Φ) in the complex plane:

$$P_N(A) = 1/N\pi \exp(-A^2/N)$$

Probability for measuring an intensity $\in$ [I, I+dI]: $P(I)dI = 1/\langle I \rangle \exp(-I/\langle I \rangle)dI$



moments: $\langle I_n \rangle \equiv {}_0\int^\infty dI \, P(I) \, I^N = n! \langle N \rangle^n$

$$\Delta I \equiv (\langle I^2 \rangle - \langle I \rangle^2)^{1/2} = \langle I \rangle$$

▪

Note: for chaotic light: $g^{(2)}(\tau) = 1 + |g^{(1)}(\tau)|^2$

$E(t) = {}_{n=1}\sum^{N} E_n(t)$, with $E_n(t)$, $E_m(t)$ uncorrelated for $n \neq m$:

$$<E(t+\tau)E(t)E^*(t)\,E(t+\tau)^*> = {}_{n=1}\sum^{N} <E_n(t+\tau)E_n(t)E_n^*(t)\,E_n(t+\tau)^*>$$

$$+ {}_{n\neq m}\sum^{N} <E_n(t+\tau)E_m(t)E_n^*(t)\,E_m(t+\tau)^*>$$

$$+ {}_{n\neq m}\sum^{N} <E_n(t+\tau)E_n(t+\tau)^*E_m^*(t)\,E_m(t)>$$

$$= N <E_n(t+\tau)E_n(t)E_n^*(t)\,E_n(t+\tau)^*>$$

$$+ N(N-1) <E_n(t+\tau)E^*_n(t)><E_m(t)\,E_m(t+\tau)^*> + N(N-1) <E_n(t+\tau)E_n(t+\tau)^*><E_m^*(t)\,E_m(t)>$$

$$\cong N^2 |<E_n(t+\tau)E^*_n(t)>|^2 + N^2<E_m^*(t)\,E_m(t)>^2 = N^2<E_m^*(t)\,E_m(t)>^2 (|g^{(1)}(\tau)|^2 + 1)$$

$$= <I>^2 (|g^{(1)}(\tau)|^2 + 1)$$

▪

An example of chaotic light: collisional broadened source revisited

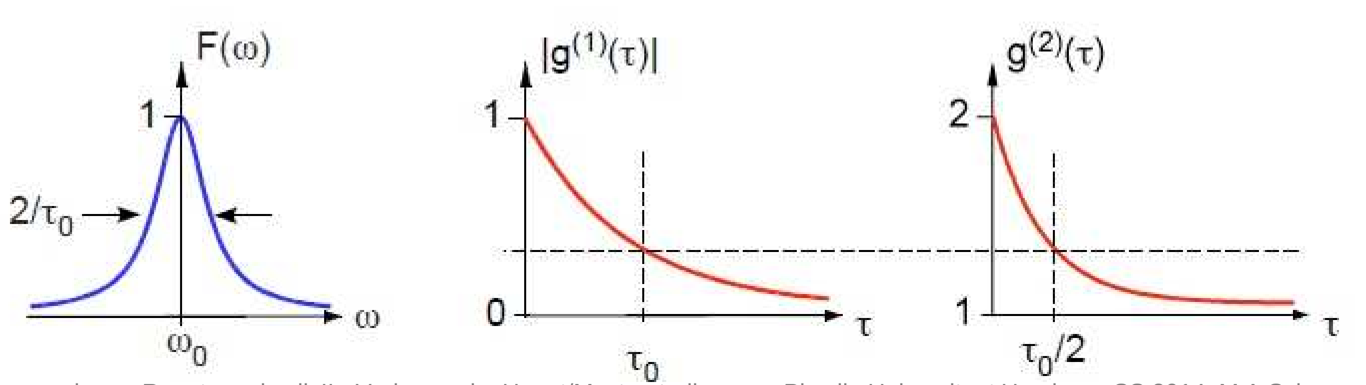
$E(t) = E_0 \sum_{n=1}^{N} \exp[i\phi_n(t)]$, $\phi_n(t) = -\omega_n t + \phi_n$, ϕ_n = random phase $\Rightarrow$

$$g^{(1)}(\tau) = \sum_{n=1}^{N} \langle \exp[i(\phi_n(t+\tau) - \phi_n(t))] \rangle = \sum_{n=1}^{N} \langle \exp(i\omega_n \tau) \rangle = \int_{-\infty}^{\infty} d\omega \exp(i\omega\tau)\, P(\omega)$$

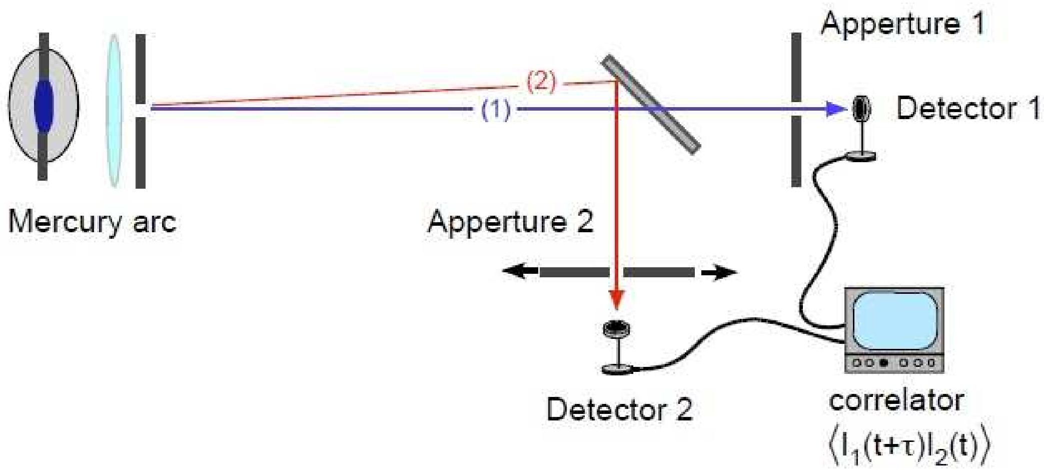
Example: assume Lorentzian spectrum (collision broadened light source)

$$P(\omega) = \tau_0/\pi \;\; 1 / [1+(\omega_0-\omega)^2\tau_0^2] \;\Rightarrow\; g^{(1)}(\tau) = \exp(-i\omega_0\tau - |\tau|/\tau_0)$$

$$g^{(2)}(\tau) = 1 + \exp(-2|\tau|/\tau_0)$$



Measurement of $g^{(2)}(\tau)$: Hanbury Brown & Twiss (1956)



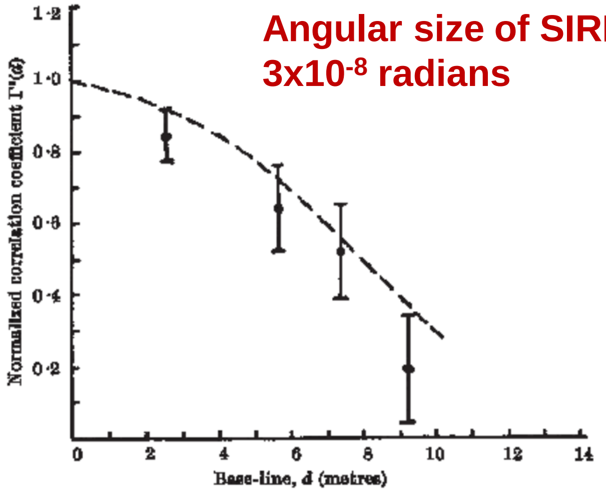


Fig. 2. Comparison between the values of the normalized correlation coefficient $\Gamma^2(d)$ observed from Sirius and the theoretical values for a star of angular diameter 0·0063". The errors shown are the probable errors of the observations

Angular size of SIRIUS:
3×10^{-8} radians

Variation of aperture 2 allows a measurement of the transverse coherence length

$\Rightarrow$ Determination of the opening angle of the source

Coherence: Applications

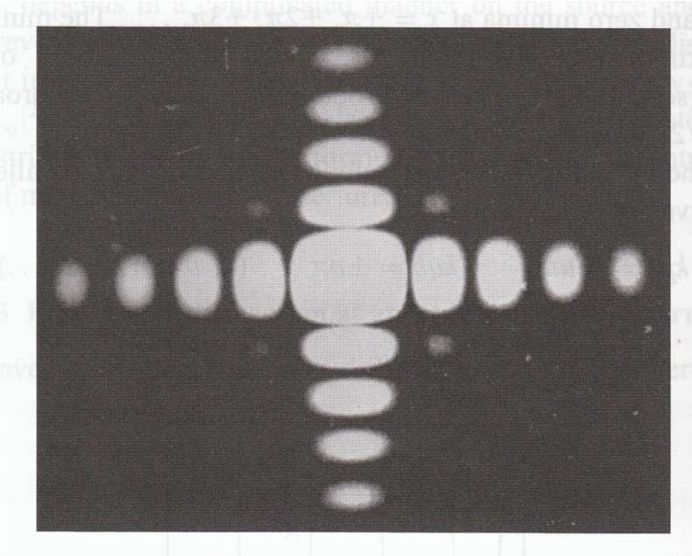
Interference patterns

X-ray speckle

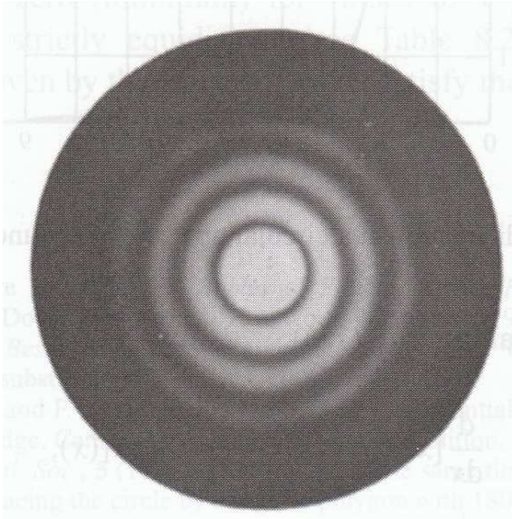
Imaging

X-Ray Photon Correlation Spectroscopy (XPCS)

Fraunhofer Diffraction

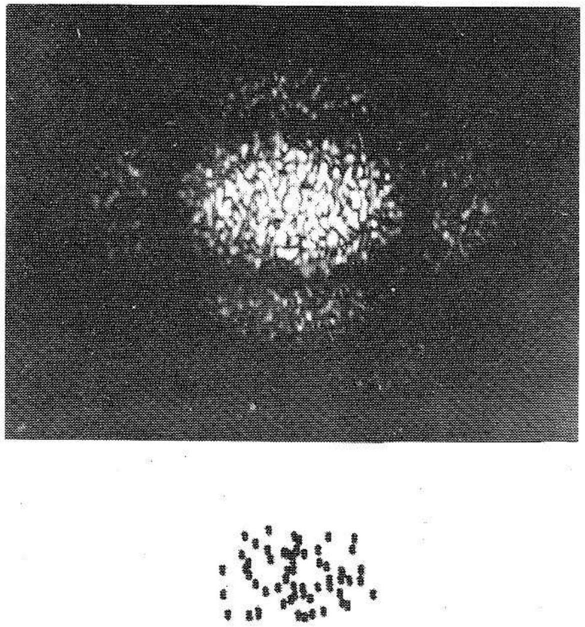


Fraunhofer diffraction of a rectangular aperture 8 x 7 mm^2, taken with mercury light λ=579nm
(from Born&Wolf, chap. 8)

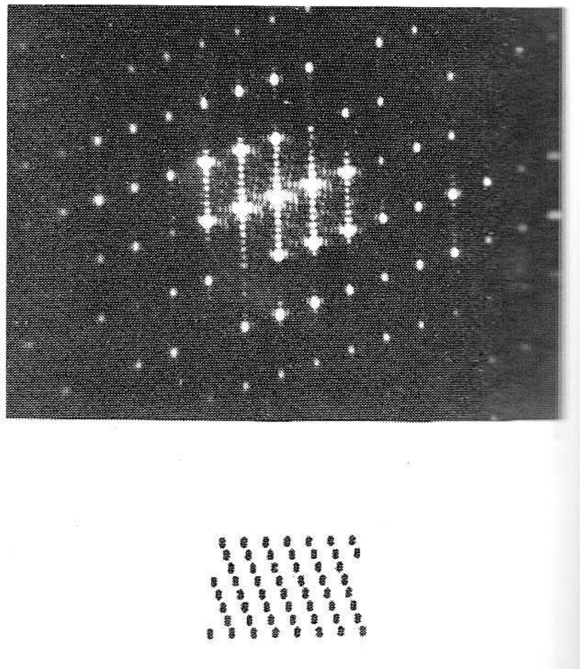


Fraunhofer diffraction of a circular aperture, taken with mercury light λ=579nm
(from Born&Wolf, chap. 8)

- Speckle pattern

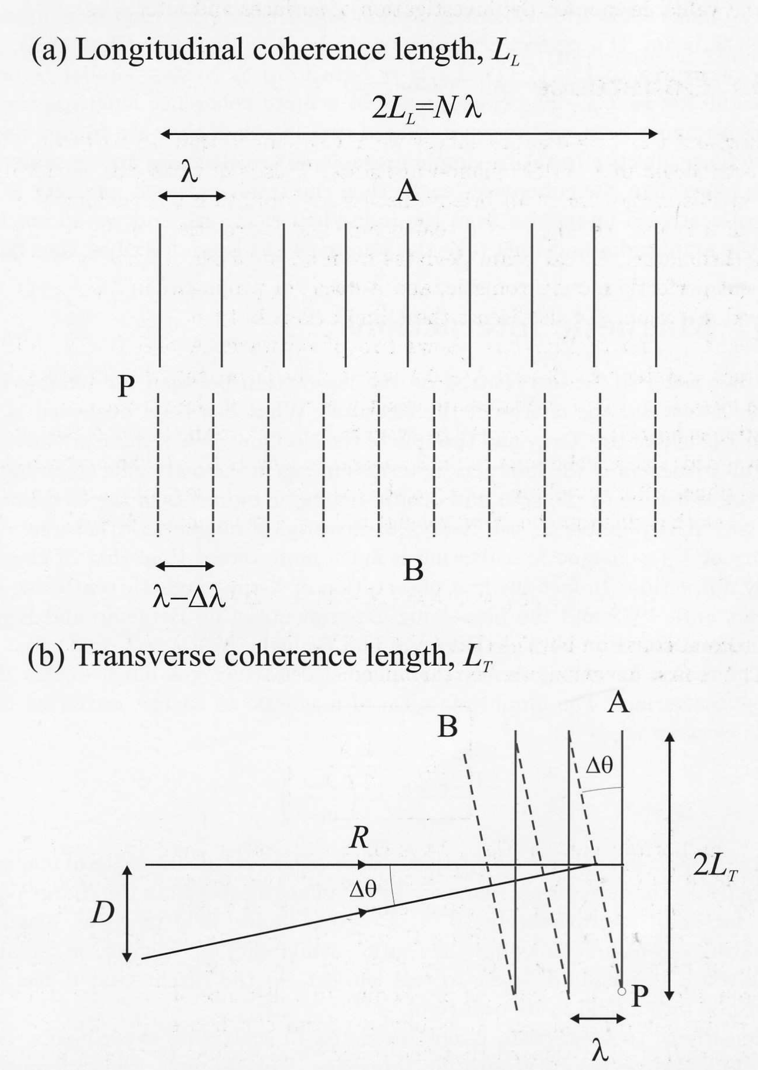


random arrangement of apertures: speckle



regular arrangement of apertures

▪ # Coherence Lengths (0.1 nm X-Rays)



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:

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

$\lambda = 0.1\text{nm}$ $\Delta\lambda/\lambda = 10^{-4}$

$\xi_l \approx 1\ \mu\text{m}$

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:

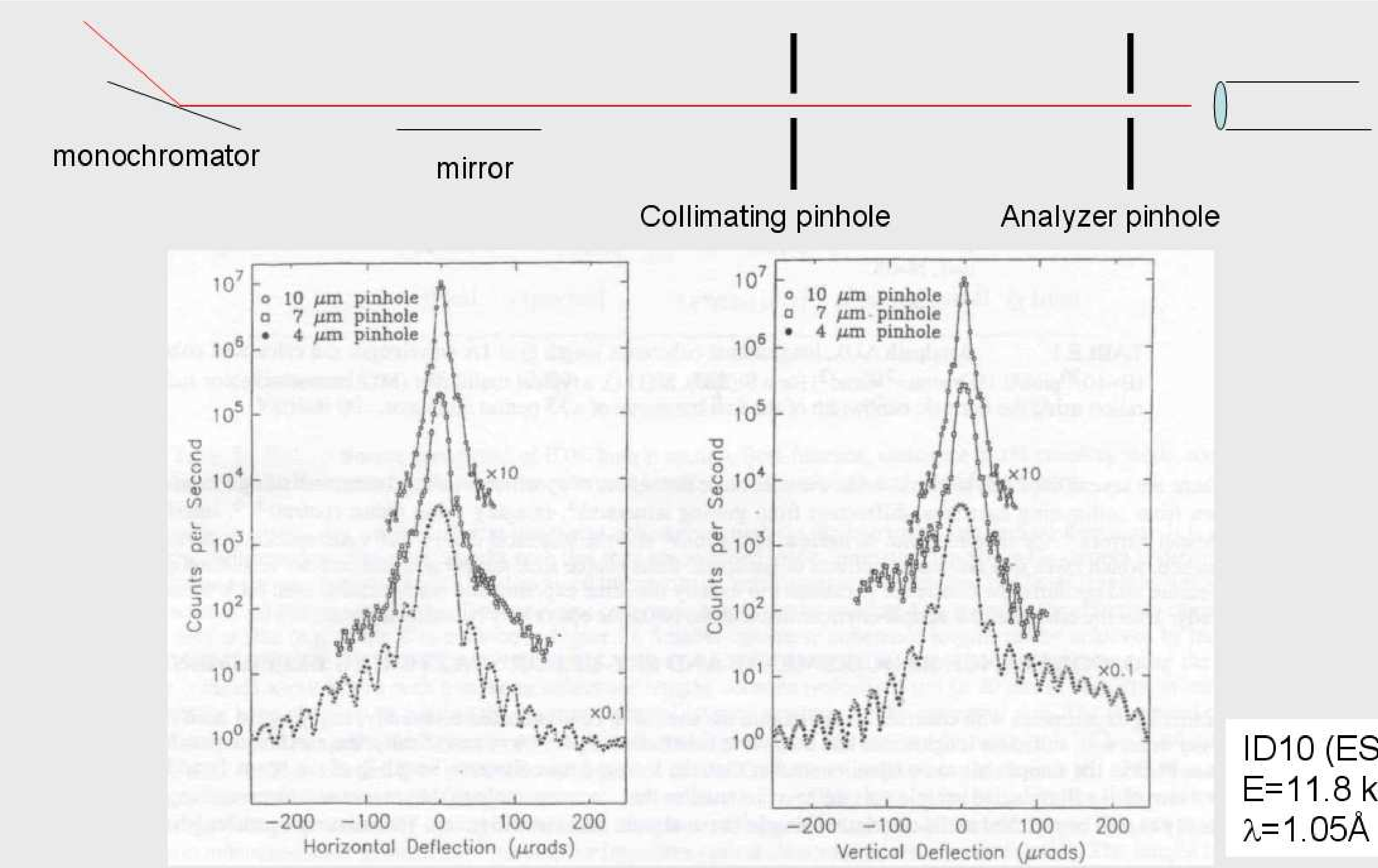
$2\xi_t\,\Delta\theta = \lambda$

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

$\lambda = 0.1\text{nm}$, $R = 100$ m, $D = 20\text{-}150\ \mu\text{m}$

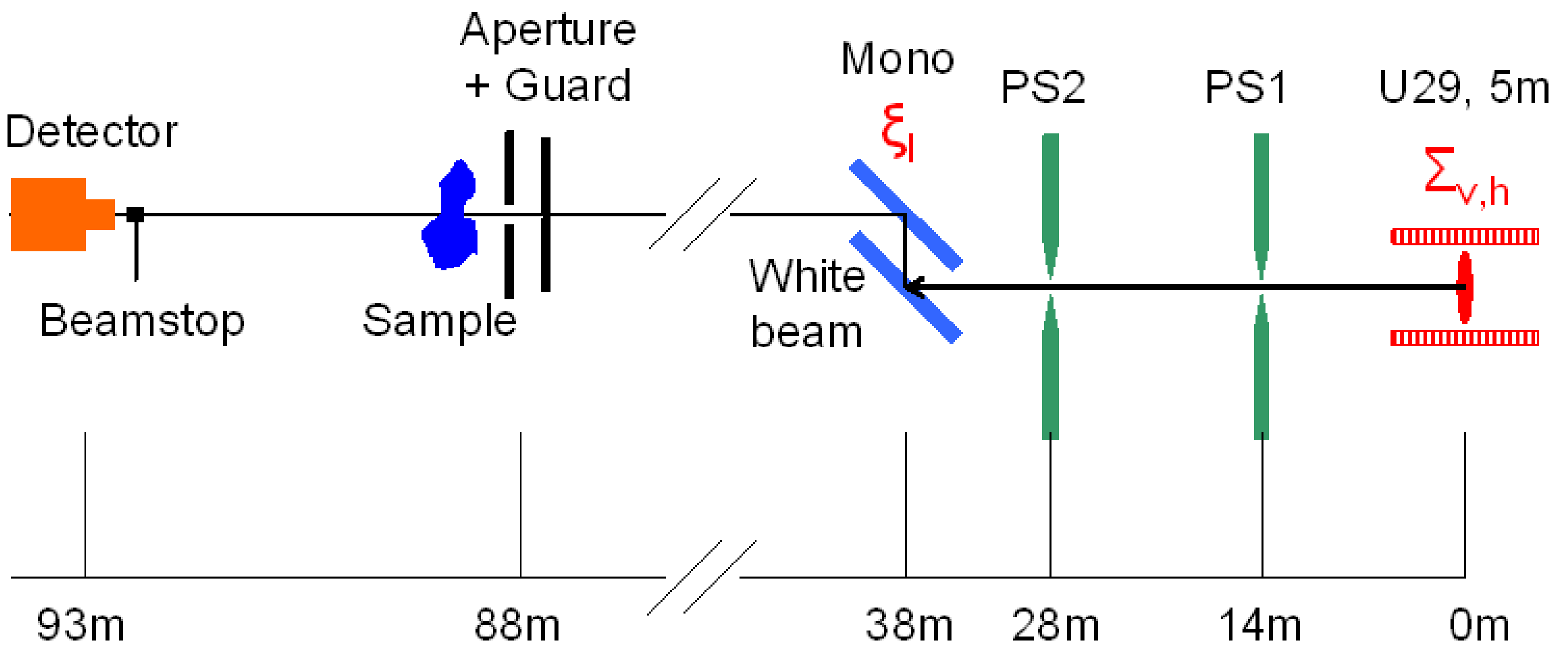
$\xi_t \approx 100\ \mu\text{m}$

▪ Fraunhofer Diffraction (λ=0.1nm)



ID10 (ESRF)
E=11.8 keV;
λ=1.05Å

- Coherence lengths of a storage ring beamline



$\Delta\lambda/\lambda = 10^{-4}$

$\Sigma_v \approx$ 5-10µm

$\Sigma_h \approx$ 100-200µm

- # Speckle

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.

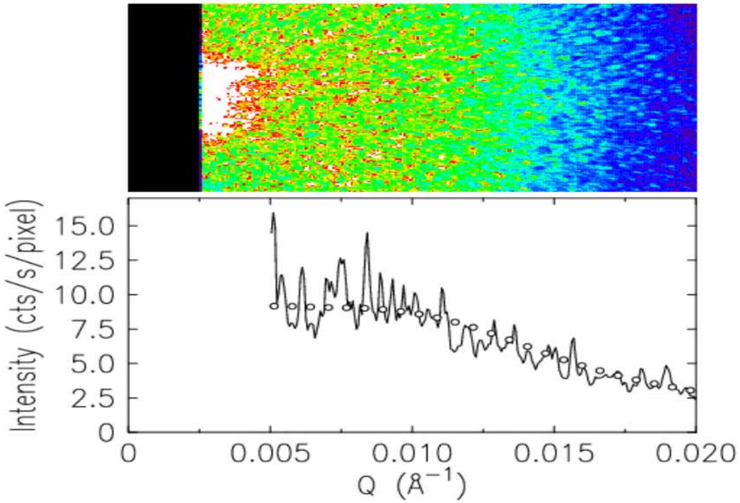
$$I(Q,t) \propto S_c(Q,t) \propto \left| \sum e^{iQR_j(t)} \right|^2$$

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

Aerogel
λ=1Å
CCD (22 μm)

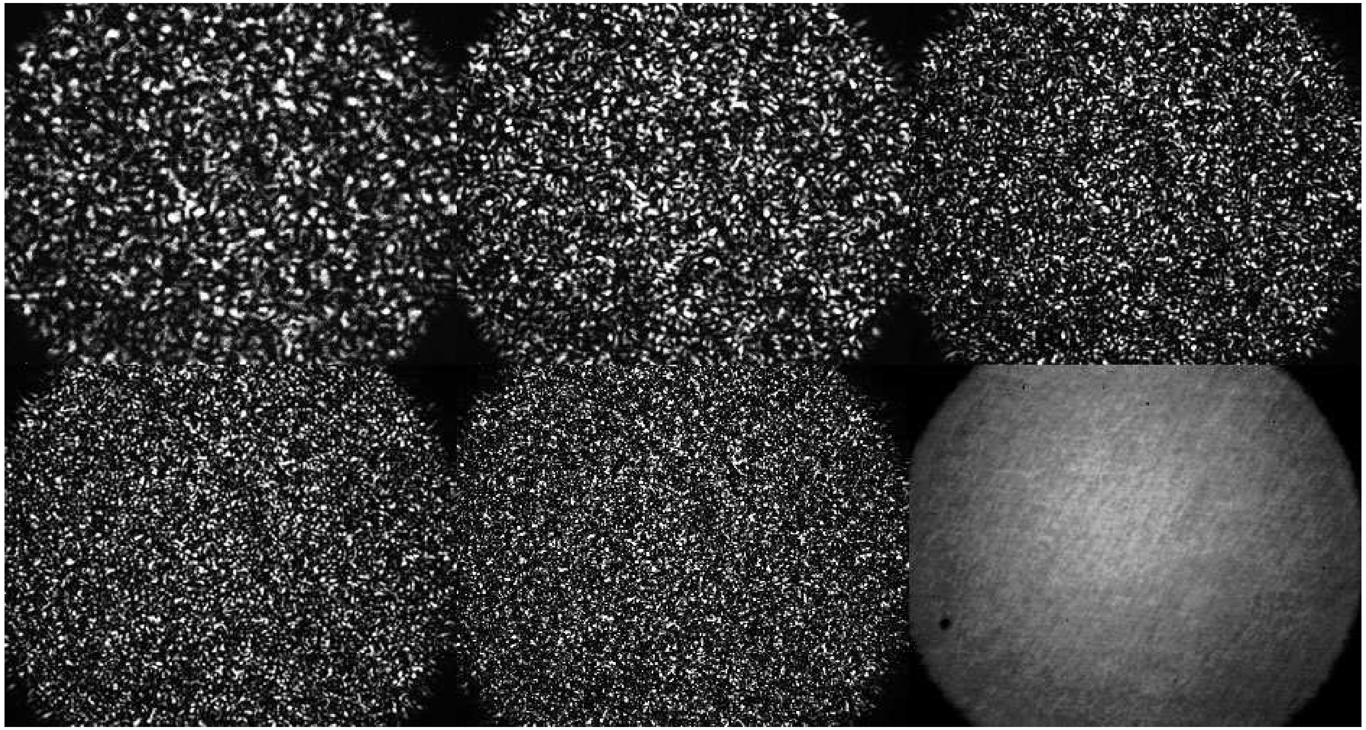


Abernathy, Grübel, et al.

J. Synchroton Rad. 5, 37, 1998

Speckle size

Taken from: Photonic Crystal group, Instituto deCiencia de Materiales de Madrid



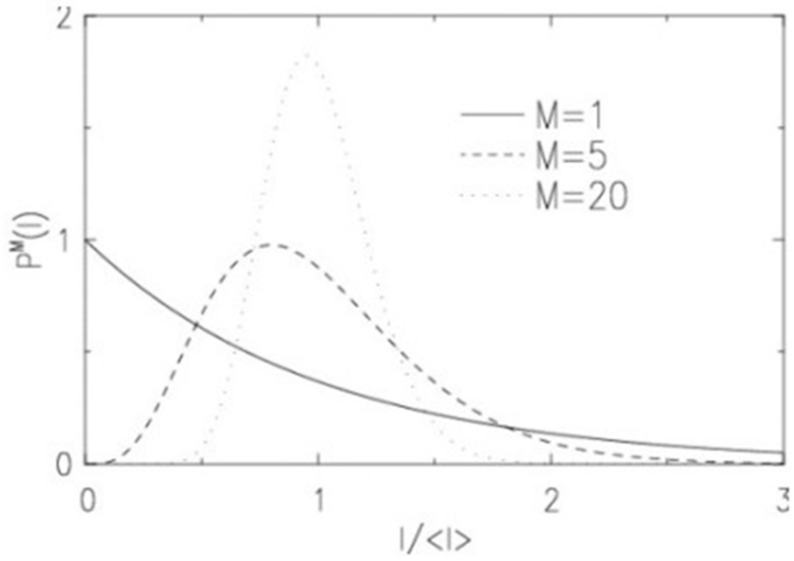
Speckle size *s* as a function of different beam diameter d

$$s \approx \frac{\lambda L}{d}$$

L: sample-detector distance
d: beam diameter

Speckle Statistics

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: $\mathrm{sqrt}(\langle I^2\rangle - \langle I\rangle^2) = \langle I\rangle$

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

<u>partially coherent illumination:</u>

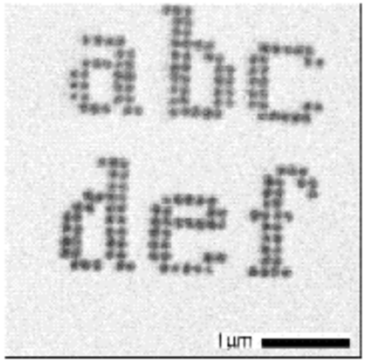
the speckle pattern is the sum of M independent speckle pattern

$P_M(I) = M^M \bullet (I/\langle I\rangle)^{M-1}/(\Gamma(M)\langle I\rangle) \bullet \exp(-MI/\langle I\rangle)$

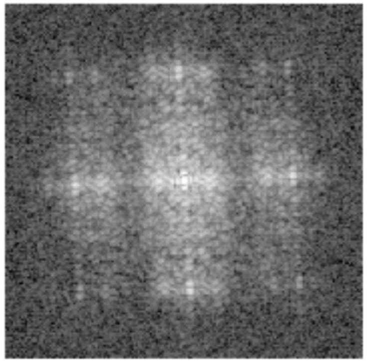
Mean: $\langle I\rangle$; $\sigma = \langle I\rangle/M^{1/2}$; $ß = 1/M$

Speckle Reconstruction

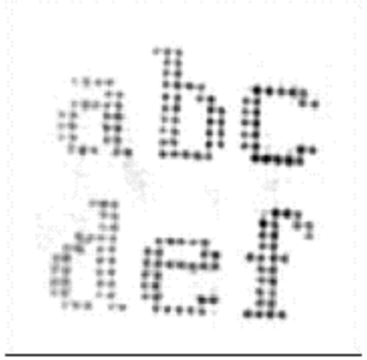
Reconstruction (phasing) of a speckle pattern: "oversampling" technique



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

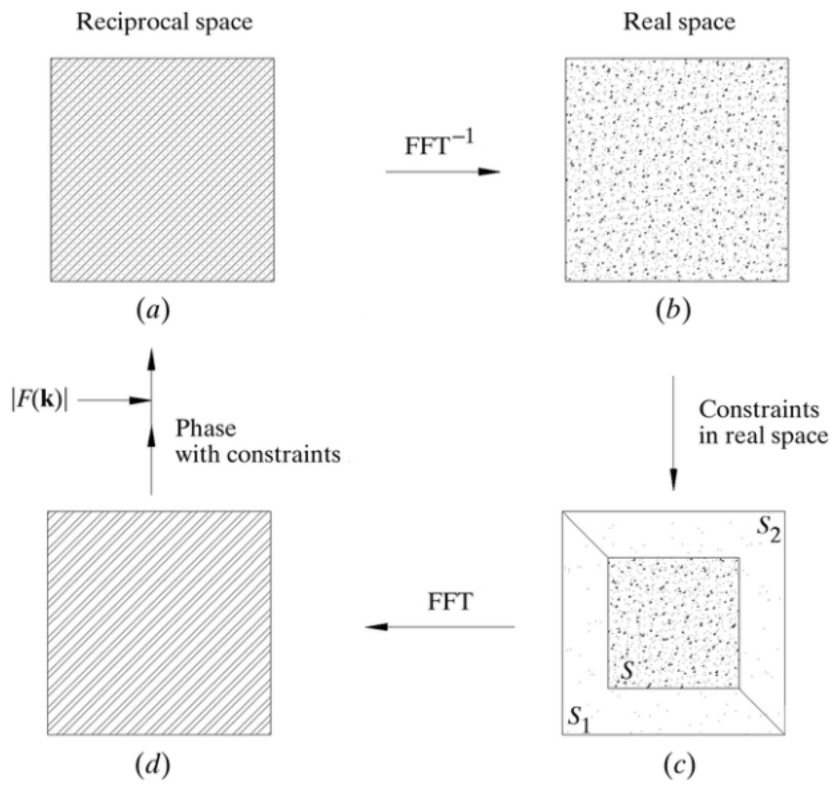
other examples: Nanocrystalline Materials

(Williams et al., PRL90,175501,2003; He et al.,PRB67,174114,2003 , Robinson et al., PRL87,195505-1)

Lensless or Coherent Diffraction Imaging (CDI)

Lensless imaging (coherent diffractive imaging) techniques aim to reconstruct the real-space structure of objects from its diffraction pattern (or hologram) by the use of constraints and phase-retrieval algorithms (e.g. Gerchberg-Saxton-Fienup) or by holographic reconstruction using Fresnel back propagation.

- Ptychography
- Plain-Wave CDI
- Holographic imaging
- Keyhole imaging
- ...



Speckle

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

Incoherent Light: $S(Q,t) = < S_c(Q,t)>_{V>>c}$ ensemble average

Aerogel
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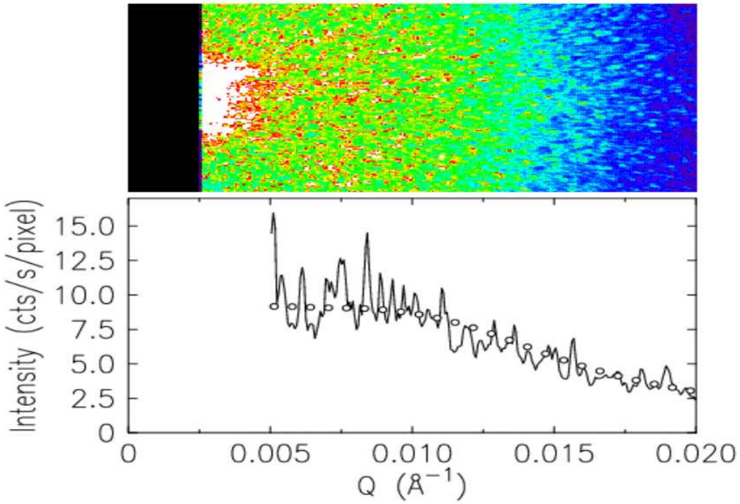
CCD (22 µm)

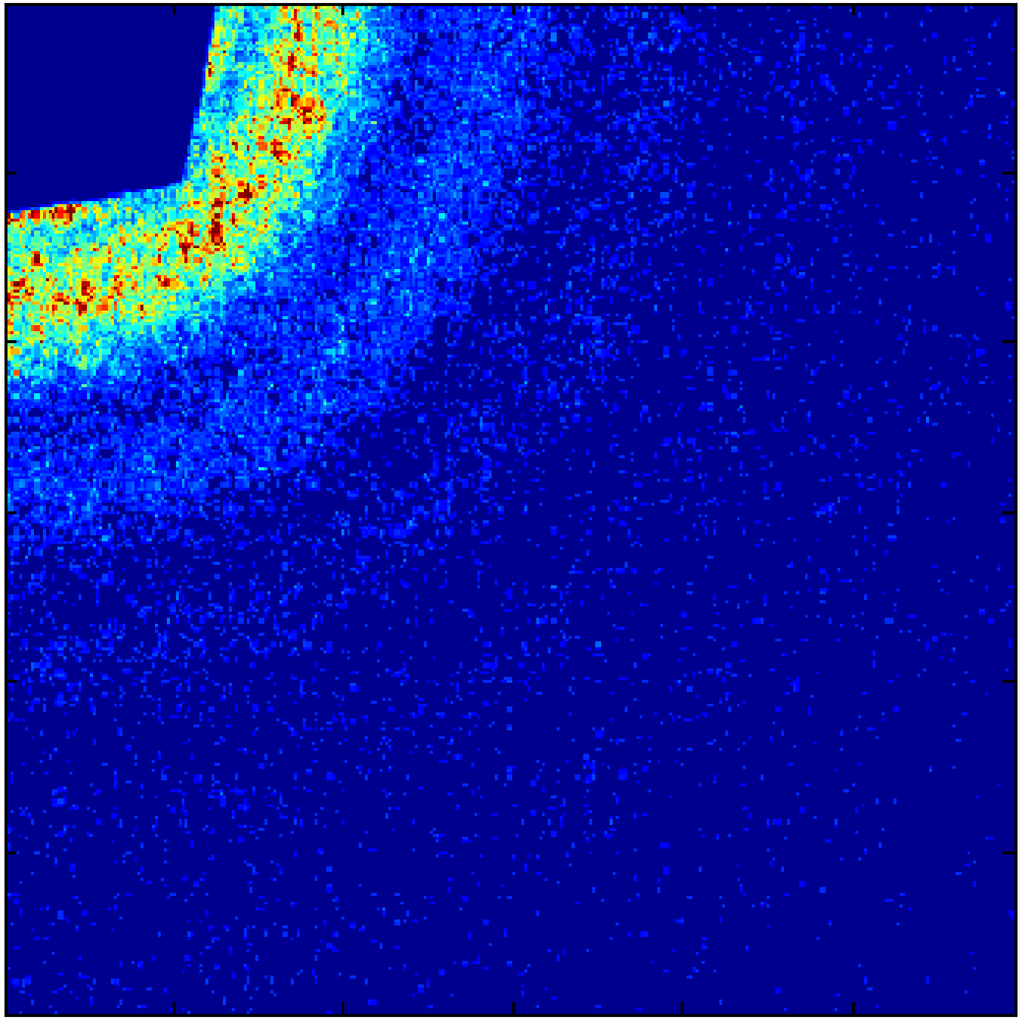


Abernathy, Grübel, et al.

J. Synchroton Rad. 5, 37, 1998

- # Fluctuating Speckle Patterns

Silica: 261.0 nm, ΔR/R = 0.03, 10 vol% in glycerol, T=-13.6℃, η ≈ 56000 cp



V. Trappe
& A. Robert

X-Ray Photon Correlation Spectroscopy(XPCS)

$g_2(Q,t) = <I(Q,0) \bullet I(Q,t)> / <I(Q)>^2$

$I(Q,t) = |E(Q,t)|^2 = |\Sigma\, b_n(Q) \exp[iQ \bullet r_n(t)|^2$

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

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

$g_2(Q,t) = 1 + ß(Q) <E(Q,0)E^*(Q,t)>^2 / <I(Q)>^2$

< >: ensemble average; ß(Q): contrast

$g_2(Q,t) = 1 + ß(Q) |f(Q,t)|^2$

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

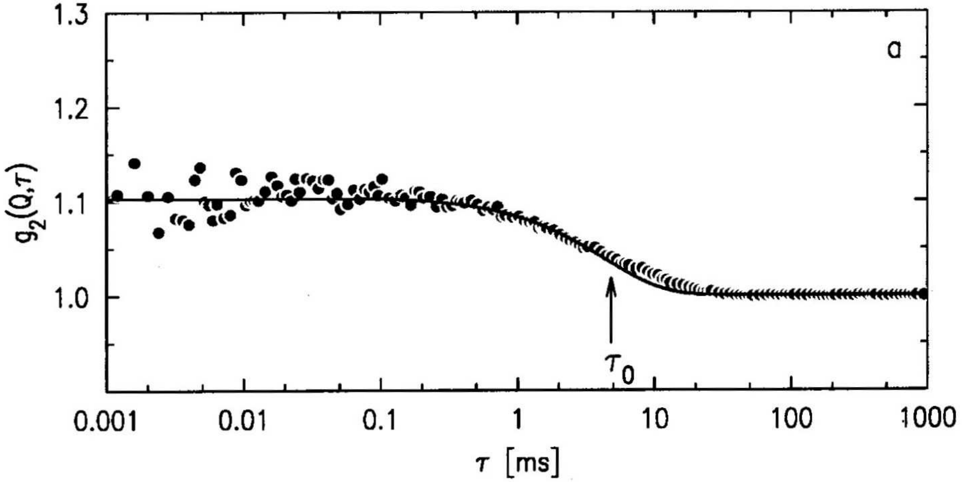
F(Q,0): static structure factor

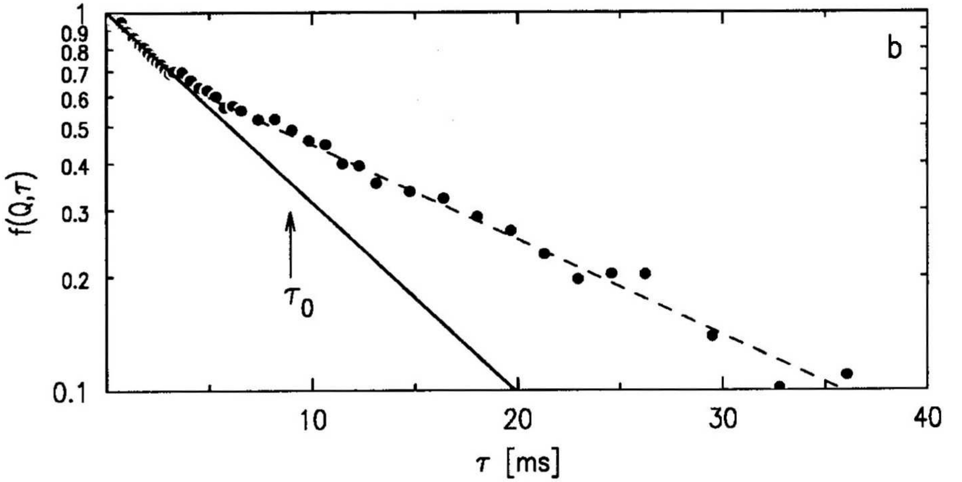
N: number of scatterers

$F(Q,t) = [1/N\{b^2(Q)\}]\, \Sigma^N_{m=1} \Sigma^N_{n=1} <b_n(Q) b_m(Q) \bullet \exp\{iQ[r_n(0)-r_m(t)]\}>$

Time correlation function $g_2(Q,t)$

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

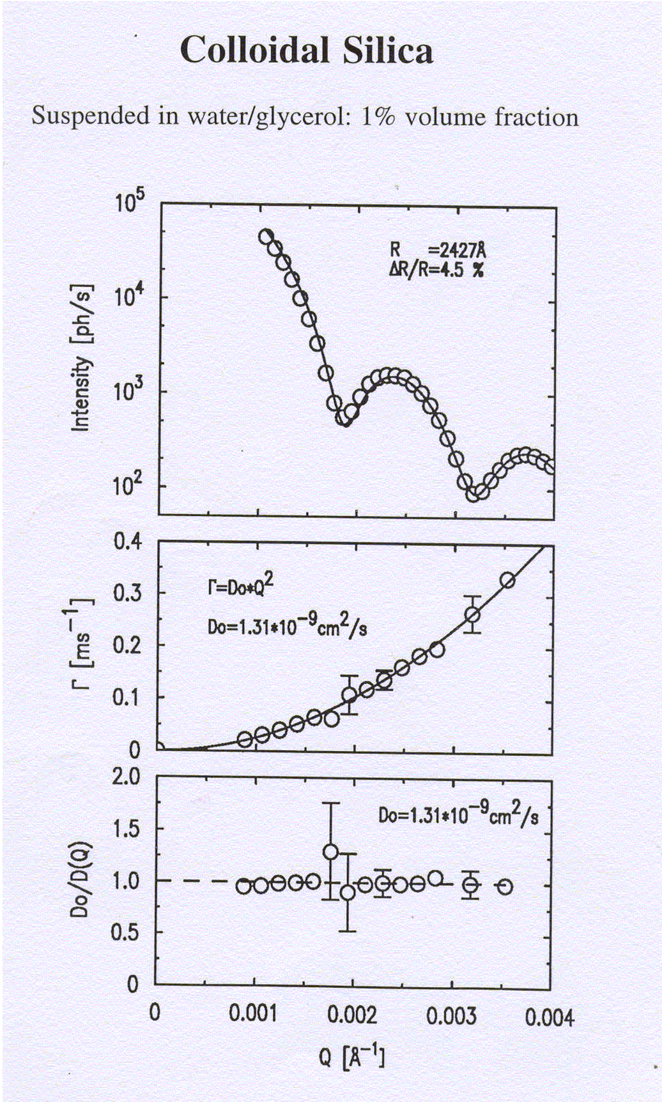




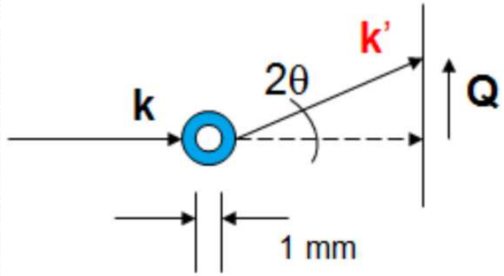
▪ Dynamics in a dilute, non-interacting system

$I \sim |F(Q)|^2\, S(Q)$

$\sim [(\sin QR - QR\cos QR)/(QR)^3]^2$



$\Gamma = D_0\, Q^2$



$Q = k' - k$

$Q = 2k \sin\theta$

$k = 2\pi/\lambda$

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

$D_0 = k_B T/(6\pi\eta R_H)$

(*Stokes-Einstein*)

R_H: hydrodynamic radius

- # Outlook

Imaging Holographic Imaging, Ptychography,….
impact of FEL sources
……

XPCS Equilibrium, non-equilibrium dynamics delay line techniques at FEL sources
…..