

Methoden moderner Röntgenphysik II: Streuung und Abbildung

Lecture 10	Vorlesung zum Haupt- oder Masterstudiengang Physik, SoSe 2015 G. Grübel, M. Martins, E. Weckert	
Location	Lecture hall AP, Physics, Jungiusstraße	
Date	Tuesday	12:45 - 14:15
	Thursday	8:30 - 10:00



Methoden moderner Röntgenphysik II: Streuung und Abbildung

Part I:

Basics of X-ray Physics

by Gerhard Grübel (GG)

Introduction

Overview, Introduction to X-ray Scattering

X-ray Scattering Primer

Elements of X-ray Scattering

Sources of X-rays, Synchrotron Radiation

Laboratory Sources, Accelerator Bases Sources

Reflection and Refraction from Interfaces

Snell's Law, Fresnel Equations

Kinematical Diffraction (I)

Diffraction from an Atom, a Molecule, from Liquids, Glasses, ...

Kinematical Diffraction (II)

Diffraction from a Crystal, Reciprocal Lattice, Structure Factor, ...



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. Suhail Zubairy,
Cambridge University Press (1997)

Courtesy: Andreas Hemmerich



• Reminder: First order coherence

Normalized autocorrelation function:

Correlation of amplitudes

$$g^{(1)}(\tau) \equiv \frac{\langle E(t + \tau)E^*(t) \rangle}{\langle E(t) \rangle^2}$$

Properties

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

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

Longitudinal coherence

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

Michelson-interferometer

Spatial coherence

$$d \cong \lambda/\theta$$



Second Order Coherence

Normalized autocorrelation function:

$$g^{(2)}(\tau) \equiv \frac{\langle I(t + \tau)I(t) \rangle}{\langle I(t) \rangle^2}$$

Correlation of intensities

degree of second order coherence

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

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

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

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

Proof (2):

$$\left(\frac{1}{N} \sum_{n=1}^N I_n \right)^2 = \frac{1}{N^2} \left(\sum_n I_n^2 + \sum_{n \neq m} I_n I_m \right) \leq \frac{1}{N^2} \left(\sum_n I_n^2 + \sum_{n \neq m} \frac{I_n^2 + I_m^2}{2} \right)$$

(inequality of arithmetic and geometric means)

$$= \frac{1}{N^2} \sum_{n,m} \frac{I_n^2 + I_m^2}{2} = \frac{1}{N} \sum_{n,m} I_n^2$$

$$\Rightarrow g^{(2)}(0) = \frac{\langle I(t)^2 \rangle}{\langle I(t) \rangle^2} = \frac{1}{N} \sum_{n,m} I_n^2 / \left(\frac{1}{N} \sum_{n=1}^N I_n \right)^2 \geq 1$$



Proof (3):

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

(Cauchy-Schwarz inequality)

Proof (4):

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

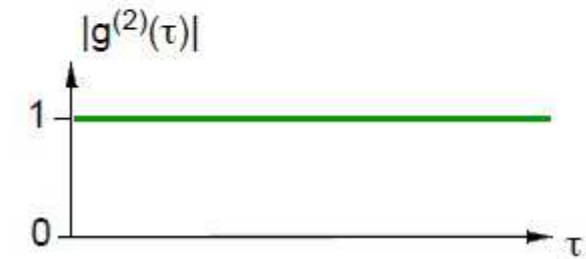
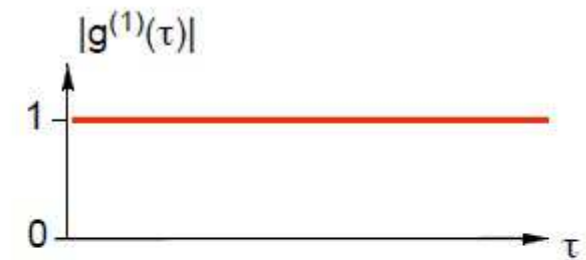
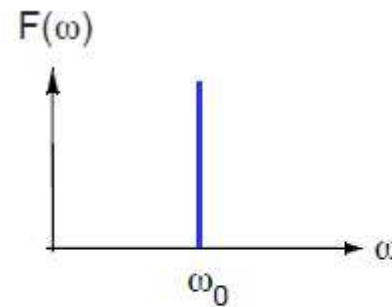
Example: monochromatic light

$$E(t) = E_0 e^{i(\omega_0 t + \phi)}$$

$$I(t) = E_0 E_0^*$$

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

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



Chaotic Light

$$E(t) = E_0 \sum_{n=1}^N e^{i\phi_n(t)}, \quad \phi_n(t) = \text{random phase, uniform at any time } t$$

$$g^{(1)}(\tau) = \sum_{n=1}^N \langle e^{i(\phi_n(t+\tau) - \phi_n(t))} \rangle$$

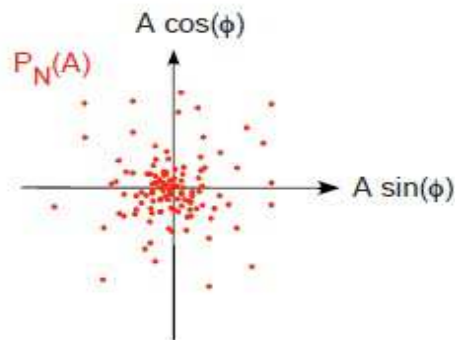
$$\langle e^{i(\phi_n(t+\tau) - \phi_n(t))} \rangle = 0 \quad \text{if } n \neq m,$$

Theory of stochastic processes:

Probability for $\sum_n^N e^{i\phi_n}$ to fall within unit areas at the point (A, Φ) in the complex plane:

$$P_N(A) = \frac{1}{N} \pi e^{-\frac{A^2}{N}}$$

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



$$\text{moments: } \langle I^n \rangle \equiv \int_0^\infty dI P(I) I^n = n! \langle I \rangle^n$$

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

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

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

$$\begin{aligned}
 \langle E(t + \tau)E(t)E^*(t)E(t + \tau)^* \rangle &= \sum_{n=1}^N \langle E_n(t + \tau)E_n(t)E_n^*(t)E_n^*(t + \tau) \rangle \\
 &+ \sum_{n \neq m}^N \langle E_n(t + \tau)E_n(t)E_m^*(t)E_m^*(t + \tau) \rangle \\
 &+ \sum_{n \neq m}^N \langle E_n(t + \tau)E_n^*(t + \tau)E_m^*(t)E_m(t) \rangle
 \end{aligned}$$

Only fields for each atom contribute

$$\begin{aligned}
 &= N \langle E_n(t + \tau)E_n(t)E_n^*(t)E_n^*(t + \tau) \rangle \\
 &+ N(N - 1) \langle E_n(t + \tau)E_n^*(t) \rangle \langle E_m(t)E_m^*(t + \tau) \rangle \\
 &+ N(N - 1) \langle E_n(t + \tau)E_n^*(t + \tau) \rangle \langle E_m^*(t)E_m(t) \rangle
 \end{aligned}$$

$N \gg 1$

$$\cong N^2 |\langle E_n(t + \tau)E_n^*(t) \rangle|^2 + N^2 \langle E_m^*(t)E_m(t) \rangle^2 = N^2 \langle E_m^*(t)E_m(t) \rangle^2 (|g^{(1)}(\tau)|^2 + 1)$$

$$= \langle I \rangle (|g^{(1)}(\tau)|^2 + 1)$$



An example of chaotic light: collisional broadened source revisited

$$E(t) = E_0 \sum_{n=1}^N e^{i\phi_n(t)}, \quad \phi_n(t) = -\omega_n t + \phi_n, \quad \phi_n = \text{random phase} \Rightarrow$$

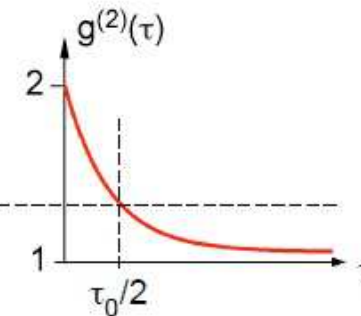
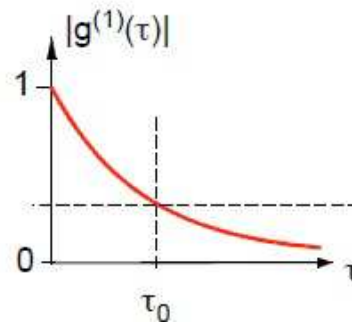
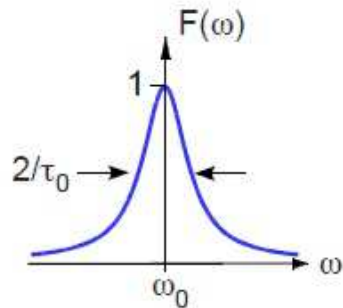
$$g^{(1)}(\tau) = \sum_{n=1}^N \langle e^{i(\phi_n(t+\tau) - \phi_n(t))} \rangle = \sum_{n=1}^N \langle e^{i\omega_n \tau} \rangle = \int_{-\infty}^{+\infty} d\omega e^{i\omega \tau} P(\omega)$$

Wiener-Khinchin

Example: assume Lorentzian spectrum (collision broadened light source)

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

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



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

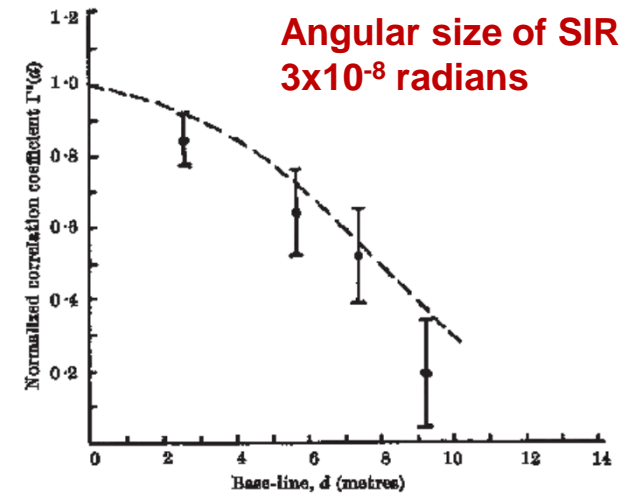
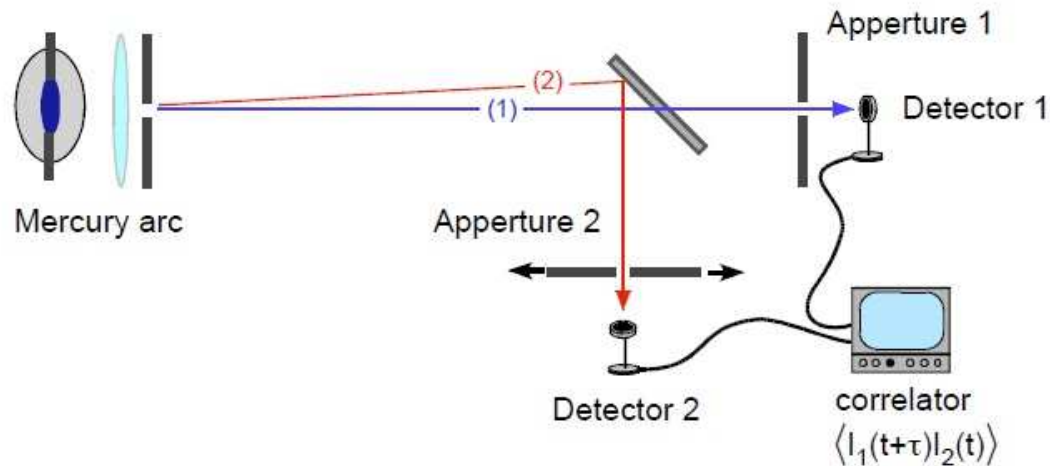


Fig. 2. Comparison between the values of the normalized correlation coefficient $I^u(d)$ observed from Sirius and the theoretical values for a star of angular diameter $0.0045''$. The errors shown are the probable errors of the observations

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

⇒ Determination of the opening angle of the source

Coherence: Applications

Interference Patterns

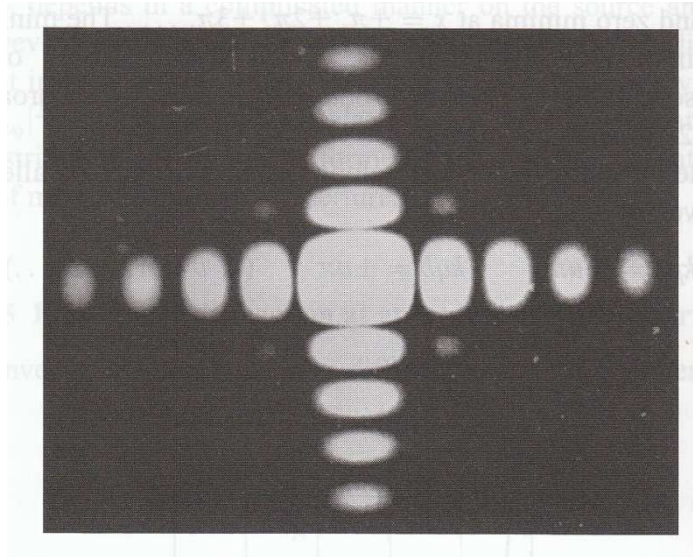
X-ray Speckle

Imaging

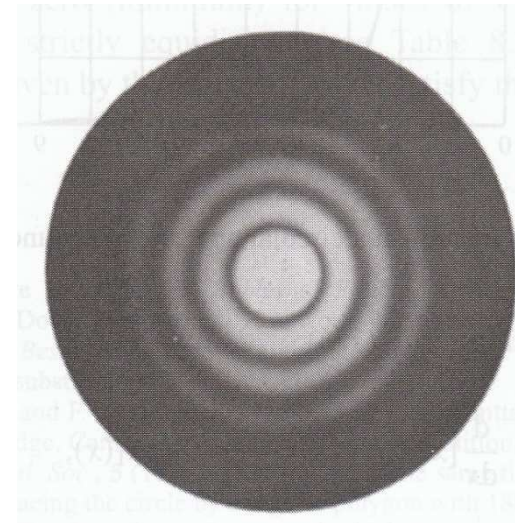
X-Ray Photon Correlation Spectroscopy (XPCS)



Fraunhofer Diffraction

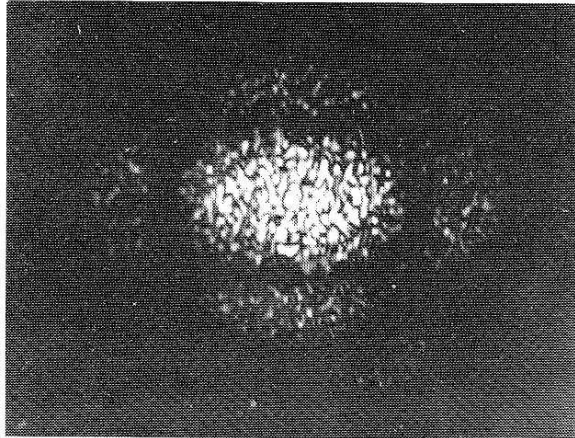


Fraunhofer diffraction of a rectangular aperture $8 \times 7 \text{ mm}^2$, taken with mercury light $\lambda=579\text{nm}$ (from Born&Wolf, chap. 8)

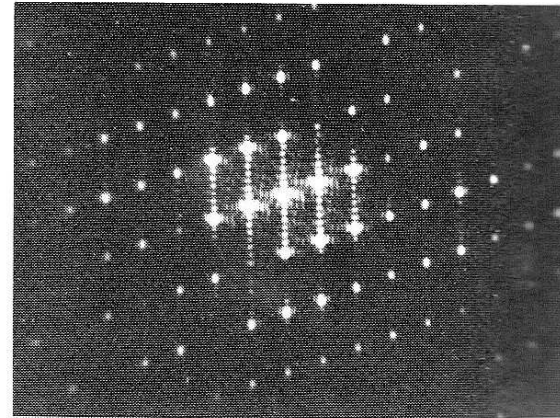


Fraunhofer diffraction of a circular aperture, taken with mercury light $\lambda=579\text{nm}$ (from Born&Wolf, chap. 8)

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 :

$$\xi_l = \left(\frac{\lambda}{2}\right) \left(\frac{\lambda}{\Delta\lambda}\right)$$

$$\lambda = 0.1 \text{ nm} \quad \frac{\Delta\lambda}{\lambda} = 10^{-4} \quad \Rightarrow \xi_l \approx 1 \text{ } \mu\text{m}$$

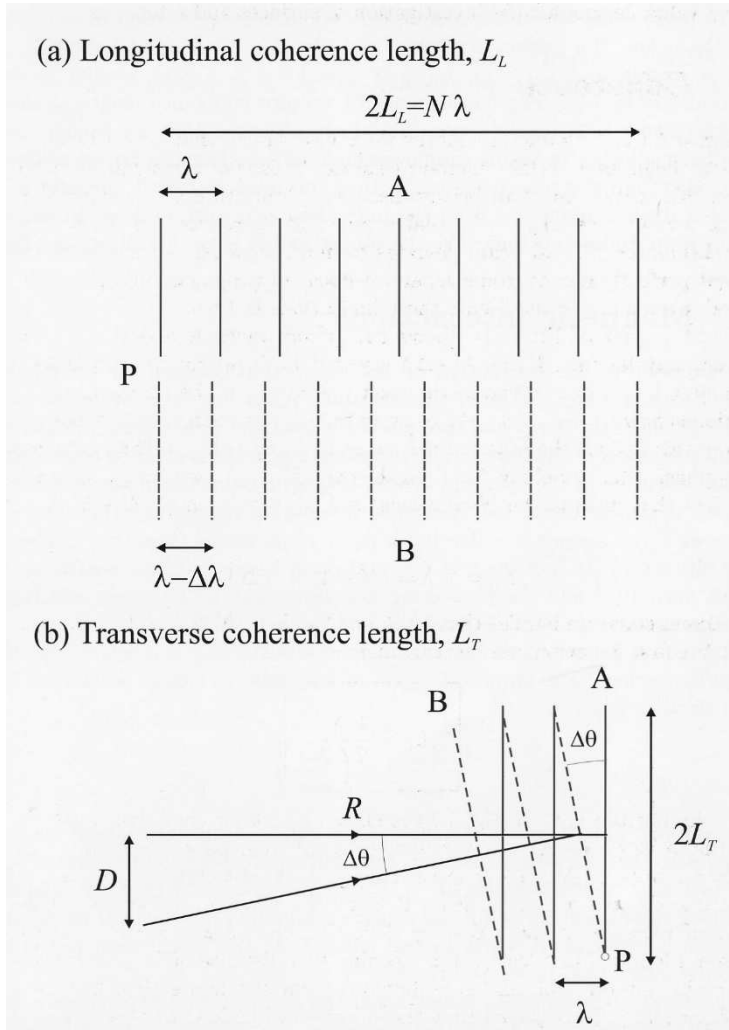
Transverse coherence:

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

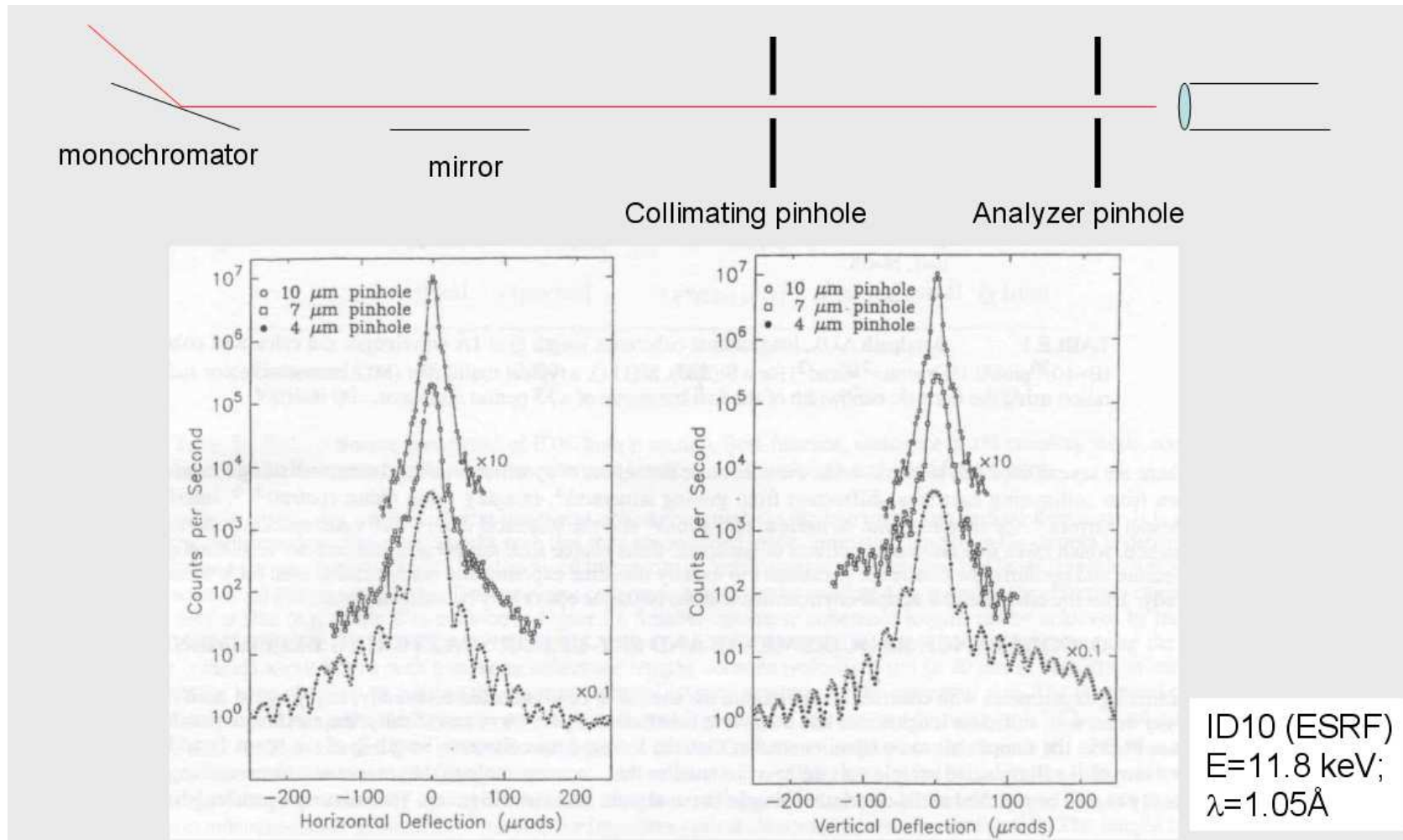
$$2\xi_t \Delta\theta = \lambda$$

$$\xi_t = \left(\frac{\lambda}{2}\right) \left(\frac{R}{D}\right)$$

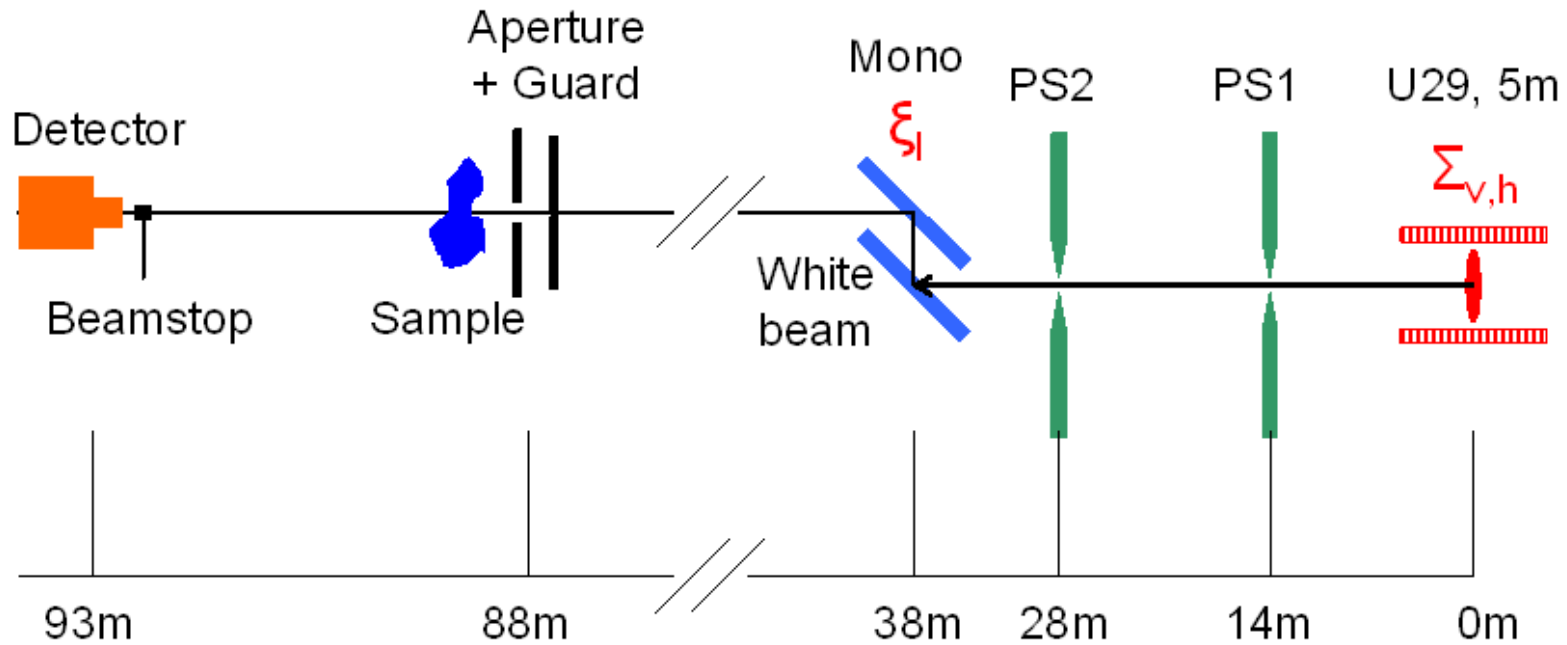
$$\lambda = 0.1 \text{ nm}, R = 100 \text{ m}, D = 20 - 150 \text{ } \mu\text{m} \Rightarrow \xi_t \approx 100 \text{ } \mu\text{m}$$



Fraunhofer Diffraction ($\lambda = 0.1 \text{ nm}$)



Coherence Lengths of a Storage Ring Beamline



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

$$\Sigma_v \approx 5 - 10\mu\text{m}$$

$$\Sigma_h \approx 100 - 200\mu\text{m}$$

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.

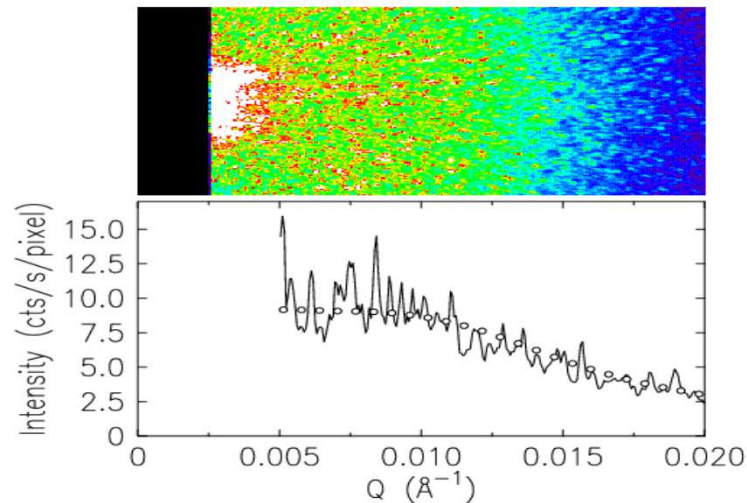
$$I(Q, t) \propto S_c(Q, t) \propto \left| \sum e^{iQR_j(t)} \right|^2$$

j in coherence volume $V_c = \xi_t^2 \xi_s$

Incoherent Light:

$$S(Q, t) = \langle S_c(Q, t) \rangle_{V \gg V_c} \text{ ensemble average}$$

Aerogel
 $\lambda = 1 \text{ \AA}$
 CCD (22 μm)

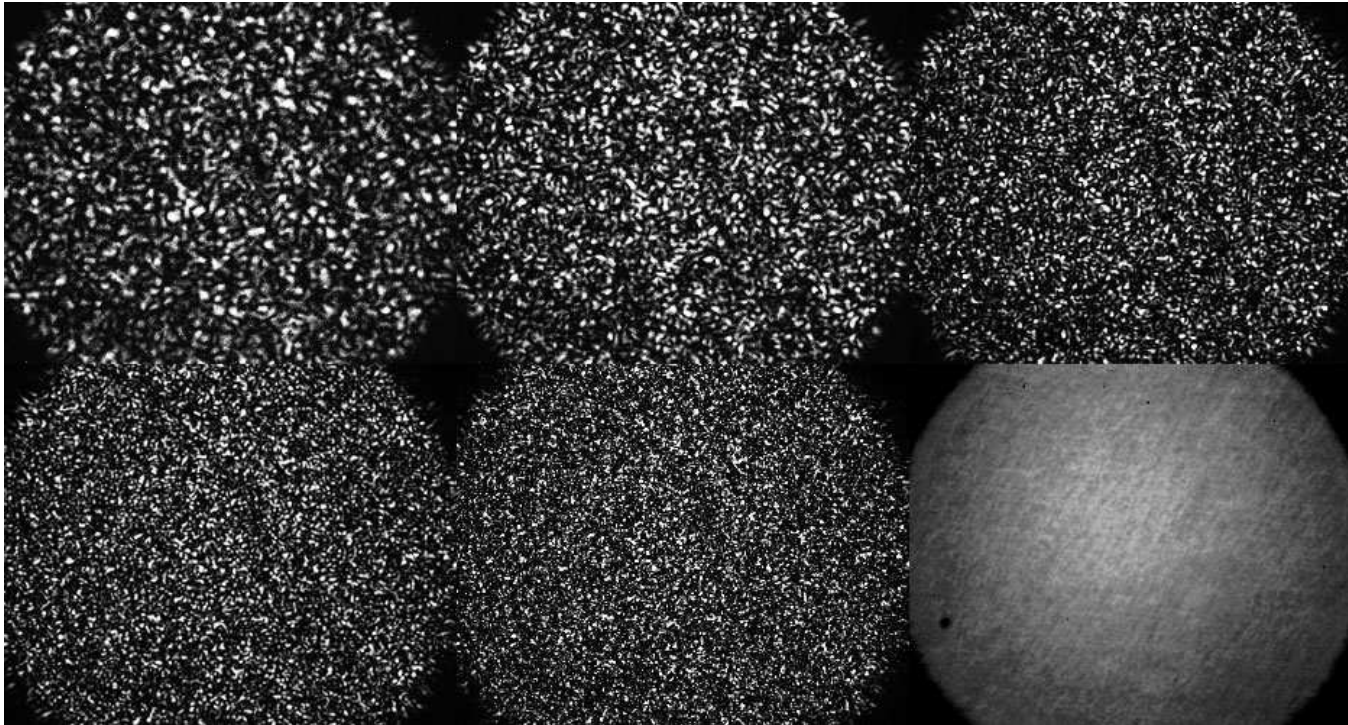


Abernathy, Grübel, et al.
 J. Synchrotron Rad. 5, 37,
 1998



Speckle size

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



Speckle size s as a function of different beam diameter d

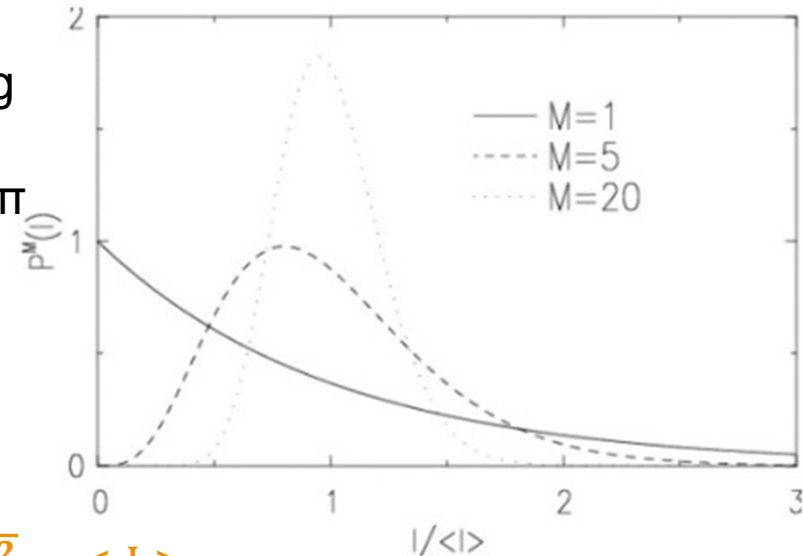
$$s \approx \frac{\lambda L}{d}$$

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:

$$P(I) = \left(\frac{1}{\langle I \rangle} \right) e^{-\frac{I}{\langle I \rangle}}$$



Mean: $\langle I \rangle$ **Std. Dev. σ :** $\sqrt{\langle I^2 \rangle - \langle I \rangle^2} = \langle I \rangle$

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

Partially coherent illumination: the speckle pattern is the sum of M independent speckle patterns

$$P_M(I) = M^M \cdot \frac{\left(\frac{1}{\langle I \rangle} \right)^{M-1}}{\Gamma(M) \langle I \rangle} \cdot e^{-\frac{MI}{\langle I \rangle}}$$

Mean: $\langle I \rangle$; $\sigma = \frac{\langle I \rangle}{\sqrt{M}}$ $\beta = \frac{1}{M}$

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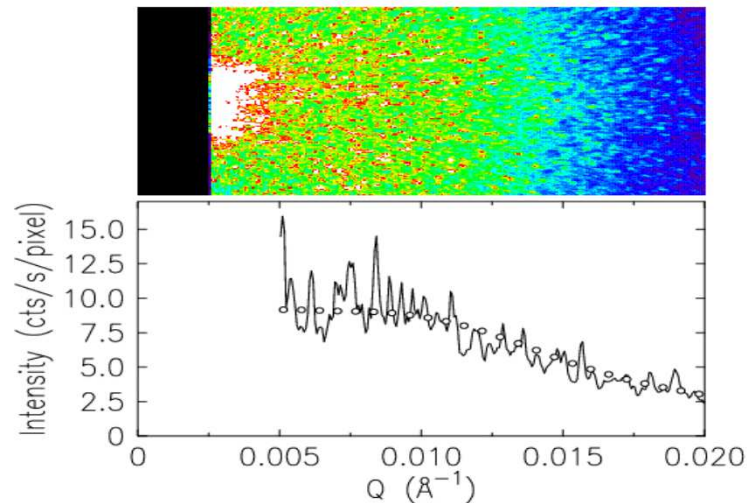
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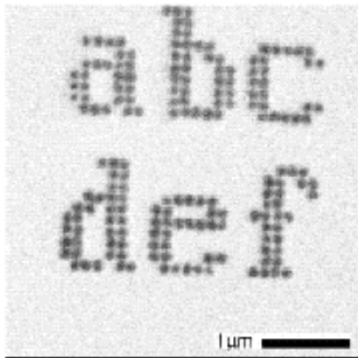


Abernathy, Grübel, et al.
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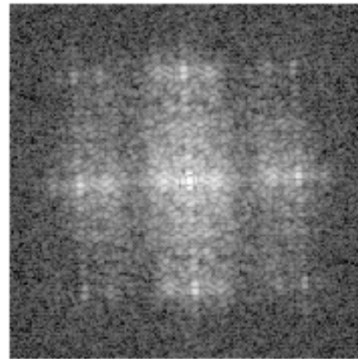


Speckle Reconstruction

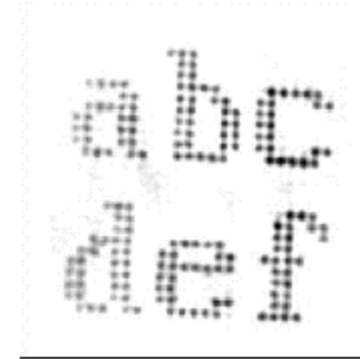
Reconstruction (phasing) of a speckle pattern: “oversampling” technique



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



$\lambda=17\text{\AA}$ coherent beam at X1A
(NSLS), $1.3 \cdot 10^9$ ph/s 10 µm pinhole
24 µm x 24 µm pixel CCD



reconstruction
“oversampling” technique

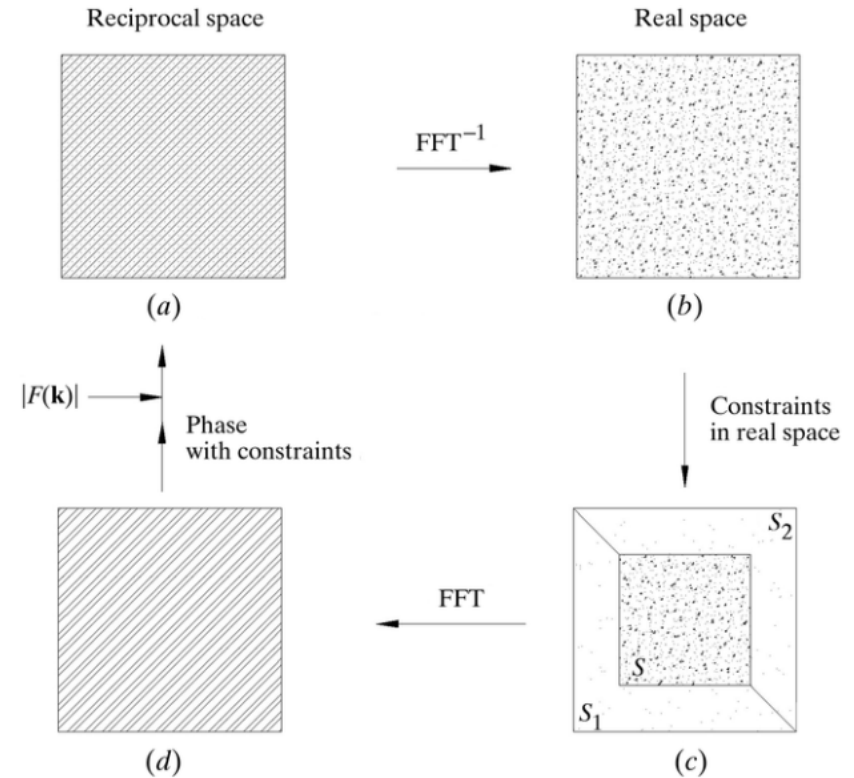
Miao, Charalambous, Kirz, Sayre, Nature, 400, July 1999

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

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
- ...



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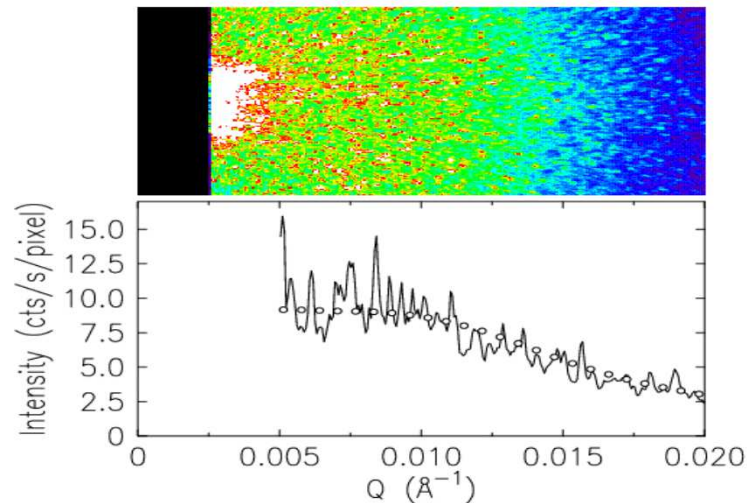
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 $\lambda = 1 \text{ \AA}$
 CCD (22 μm)

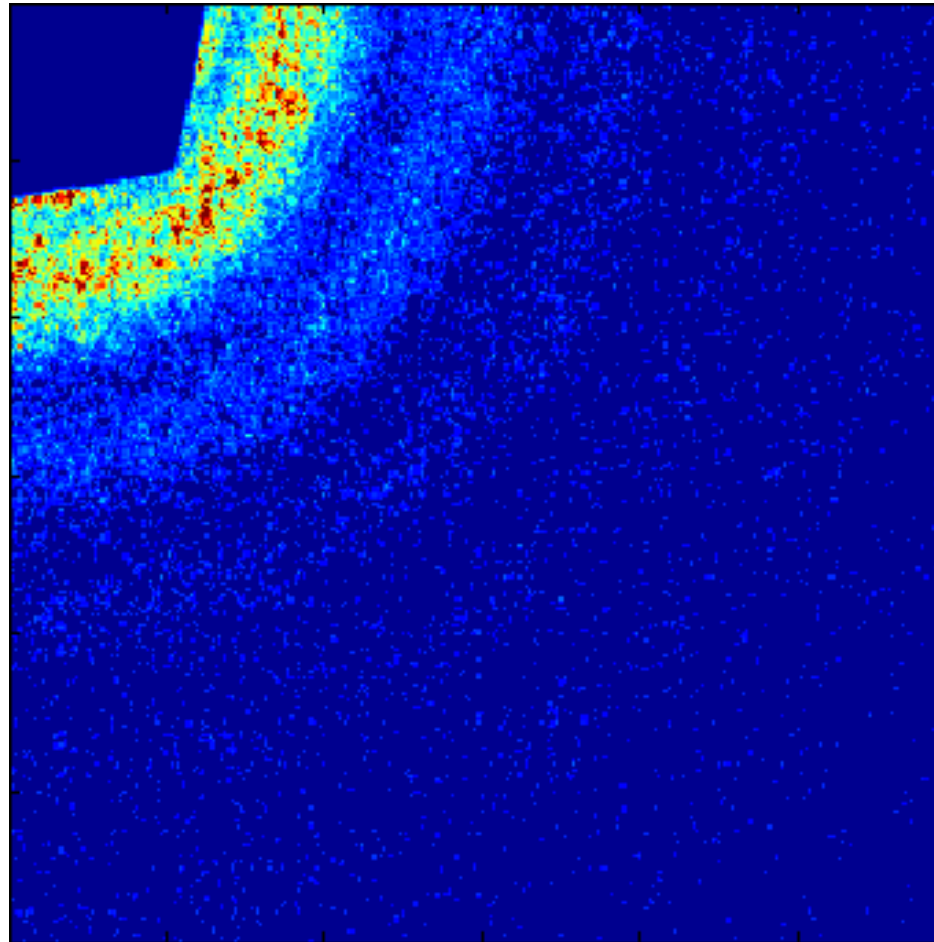


Abernathy, Grübel, et al.
 J. Synchrotron Rad. 5, 37,
 1998



Fluctuating Speckle Patterns

Silica: 2610 Å, $\frac{\Delta R}{R} = 0.03$, 10 vol% in glycerol, $T = -13.6$ °C, $\eta \approx 56000$ cp



V. Trappe
& A. Robert

X-Ray Photon Correlation Spectroscopy (XPCS)

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

$$I(Q, t) = |E(Q, t)|^2 = \left| \sum b_n(Q) e^{iQ \cdot r_n(t)} \right|^2$$

Note: $E(Q, t) = \int dr' \rho(r') e^{iQ \cdot r'(t)}$ $\rho(r')$: charge density

If $E(Q, t)$ is a zero mean, complex Gaussian variable:

$$g_2(Q, t) = 1 + \beta(Q) \frac{\langle E(Q, 0) E^*(Q, t) \rangle^2}{\langle I(Q) \rangle^2} \quad \langle \rangle: \text{ensemble av.}; \beta(Q): \text{contrast}$$

$$g_2(Q, t) = 1 + \beta(Q) |f(Q, t)|^2 \quad \text{with } f(Q, t) = S(Q, t)/S(Q, 0)$$

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

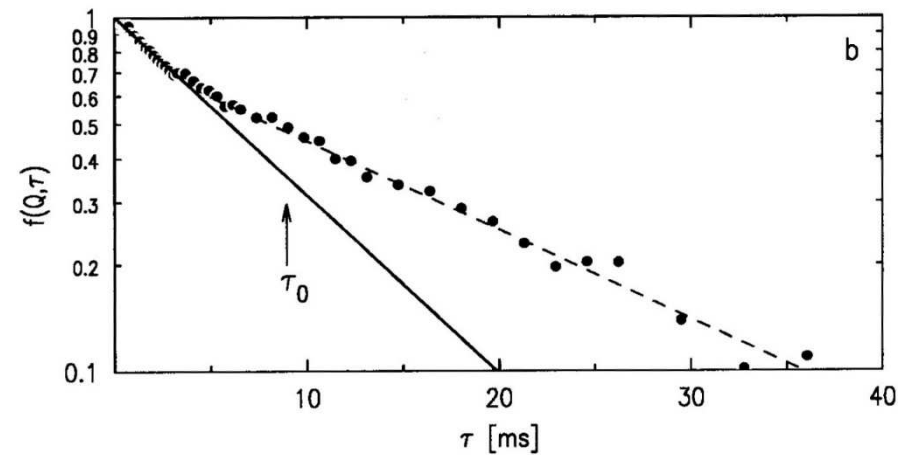
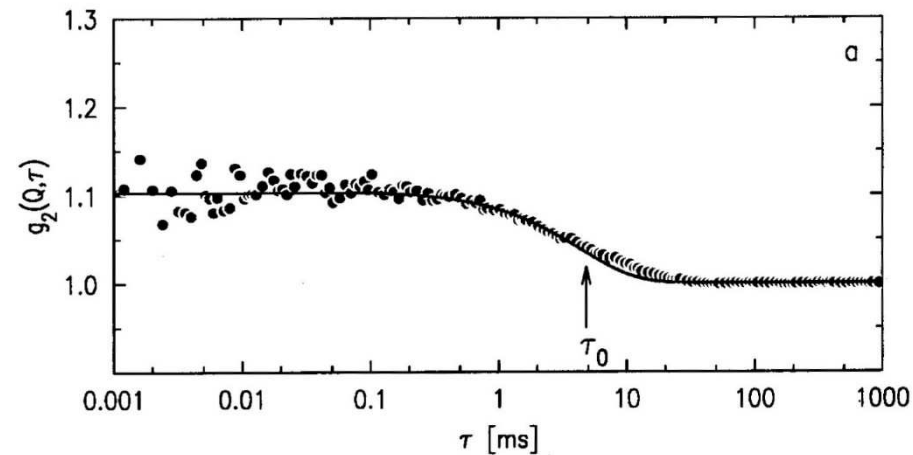
N : number of scatterers

$$S(Q, t) = \frac{1}{N \{b^2(Q)\}} \sum_{m=1}^N \sum_{n=1}^N \langle b_n(Q) b_m(Q) e^{iQ[r_n(0) - r_m(t)]} \rangle$$



Time Correlation Function $g_2(Q,t)$

$$g_2(Q,t) = 1 + \beta(Q)|f(Q,t)|^2 \text{ and } f(Q,t) = e^{(-\Gamma t)} = e^{\left(\frac{-t}{\tau_0}\right)}$$



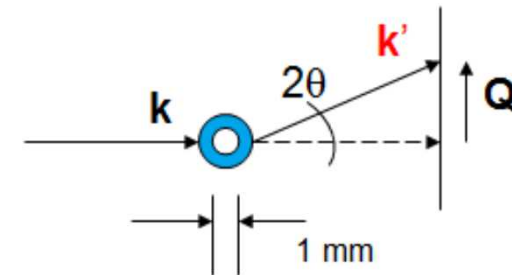
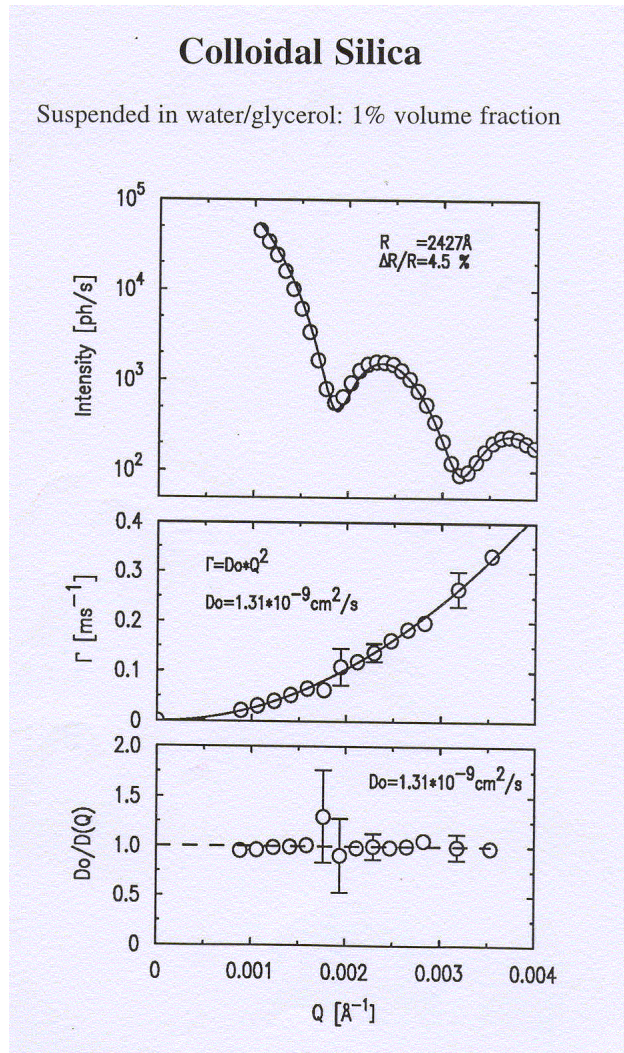
Dynamics in a Dilute, Non-interacting System

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

$$\sim \left[\frac{\sin QR - QR \cos QR}{(QR)^3} \right]^2$$

$$\Gamma = D_0 Q^2$$

$$D_0 = \frac{k_B T}{6\pi\eta R}$$



$$\mathbf{Q} = \mathbf{k}' - \mathbf{k}$$

$$Q = 2k \sin\theta$$

$$k = 2\pi/\lambda$$

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

Outlook

Imaging Holographic Imaging, Ptychography,....

Impact of FEL sources.....

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

