Methoden moderner Röntgenphysik: Streuung und Abbildung

Lecture 10	Vorlesung zum Haupt- oder Masterstudiengang Physik, SoSe 2019
	G. Grübel, L. Müller, O. Seeck, L. Frenzel, F. Lehmkühler, M. Martins, W. Wurth
Location	Lecture hall AP, Physics, Jungiusstraße

Date	Tuesdays	12:30 - 14:00	(starting 2.4.)
	Thursdays	8:30 - 10:00	(until 11.7.)

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, Applications of Coherent Scattering
Spatial Coherence, Second Order Coherence,
Imaging and Correlation Spectroscopy, ...

Second Order Coherence

Normalized autocorrelation function:

$$g^{(2)}(\tau) \equiv \frac{< I(t+\tau)I(t) >}{< I(t) >^2}$$

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

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

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

Proof (3):

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

(Cauchy-Schwarz inequality)

Proof (4):

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

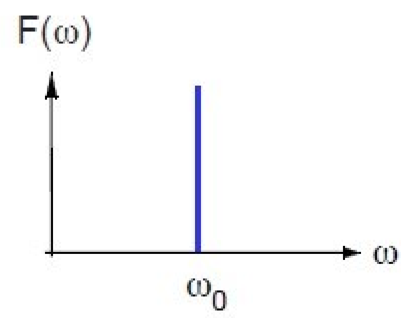
Example: **monochromatic light**

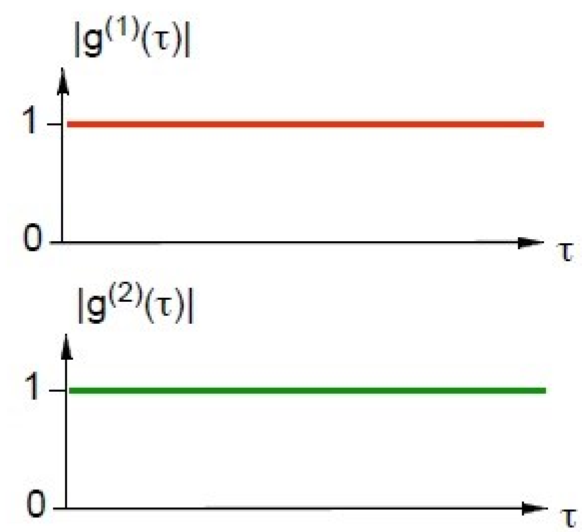
$$E(t) = E_0\, e^{i(\omega_0 t + \phi)}$$

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

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

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





Chaotic Light

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

$$< e^{i(\phi_n(t+\tau)-\phi_m(t))} > = 0 \qquad \text{if } n \neq m,$$

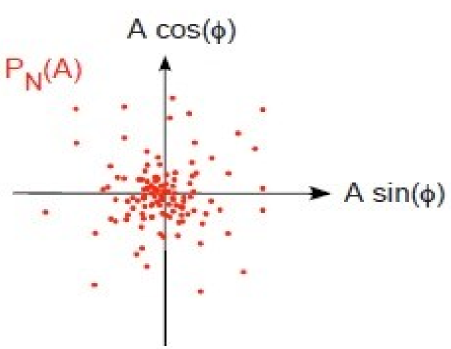
$$g^{(1)}(\tau) = \sum_{n=1}^{N} < e^{i(\phi_n(t+\tau)-\phi_n(t))} >$$

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



moments: $< I^n > \equiv \int_0^\infty dI\, P(I)\, I^n = n! < I >^n$

$$\Delta I \equiv (< I^2 > - < I >^2)^{\frac{1}{2}} = < I >$$

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 $n \neq m$:

$$< E(t+\tau)E(t)E^*(t)E(t+\tau)^* > = \sum_{n=1}^{N} < E_n(t+\tau)E_n(t)E_n^*(t)E_n^*(t+ \quad >$$

$$+ \sum_{n \neq m}^{N} < E_n(t+\tau)E_n(t)E_m^*(t)E_m^*(t+ \quad >$$

$$+ \sum_{n}^{N} < E_n(t+\tau)E_n^*(t+\tau)E_m^*(t)E_m(t) >$$

Only fields for each atom contribute

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

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

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

N >>1

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

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

An example of chaotic light: collisional broadened source revisited

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

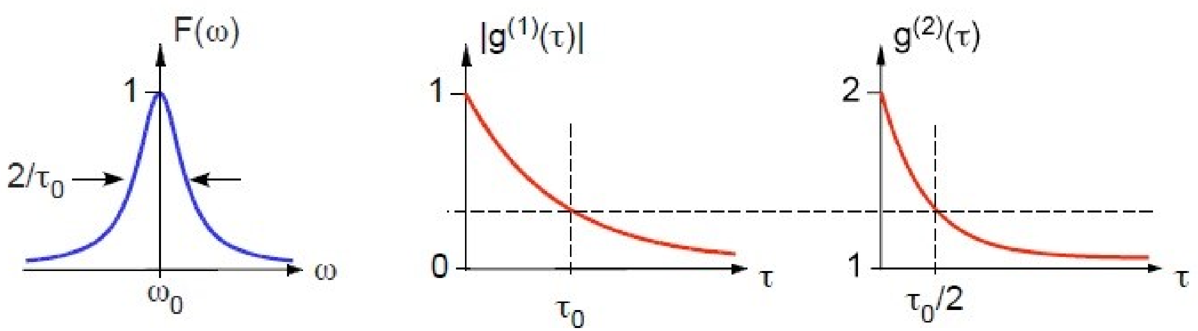
$$g^{(1)}(\tau) = \sum_{n=1}^{N} < e^{i(\phi_n(t+\tau)-\phi_n(t))} > = \sum_{n=1}^{N} < e^{i\omega_n \tau} > = \int_{-\infty}^{+\infty} d\omega\ e^{i\omega\tau} P(\omega)$$

Wiener-Khinchin

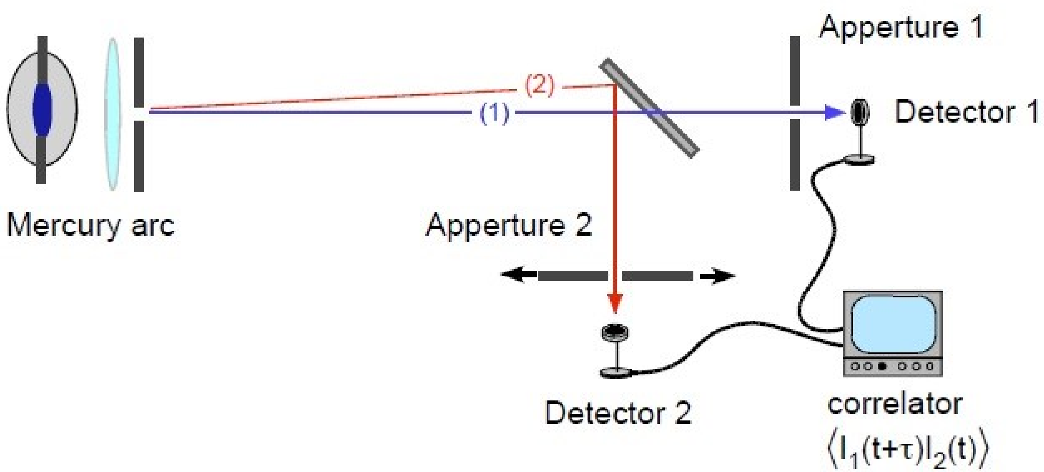
Example: assume Lorentzian spectrum (collision broadened light source)

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

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



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



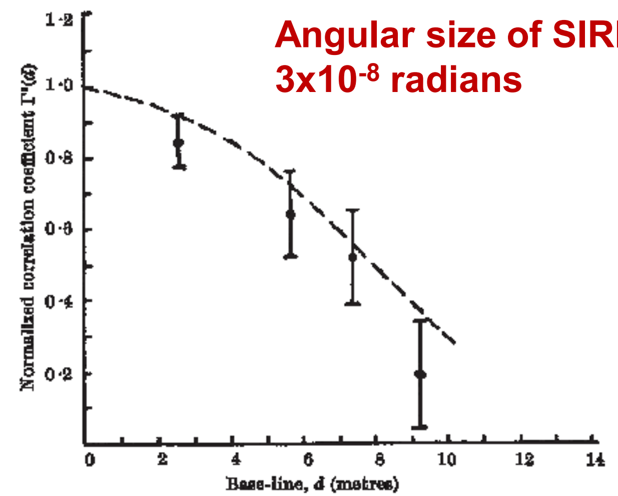


Fig. 2. Comparison between the values of the normalized correlation coefficient $\Gamma^2(d)$ observed from Sirius and the theoretical values for a star of angular diameter 0·0063″. The errors shown are the probable errors of the observations

Angular size of SIRIUS: $3\text{x}10^{-8}$ radians

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

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

Coherence: Applications

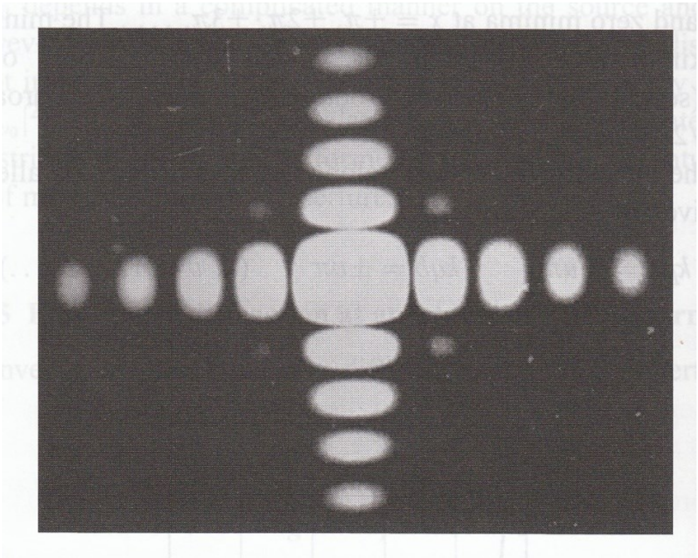
Interference Patterns

X-ray Speckle

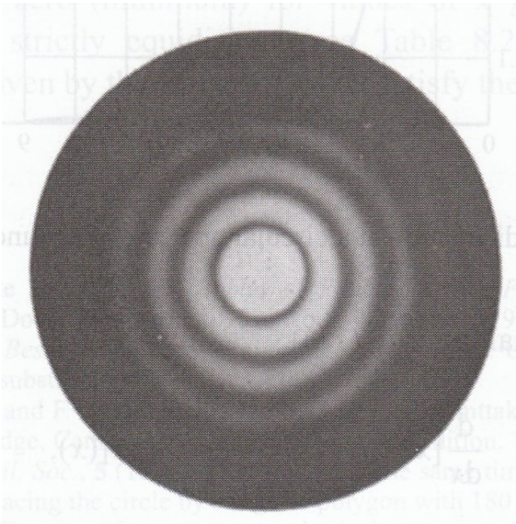
(Imaging)

X-Ray Photon Correlation Spectroscopy (XPCS)

Fraunhofer Diffraction

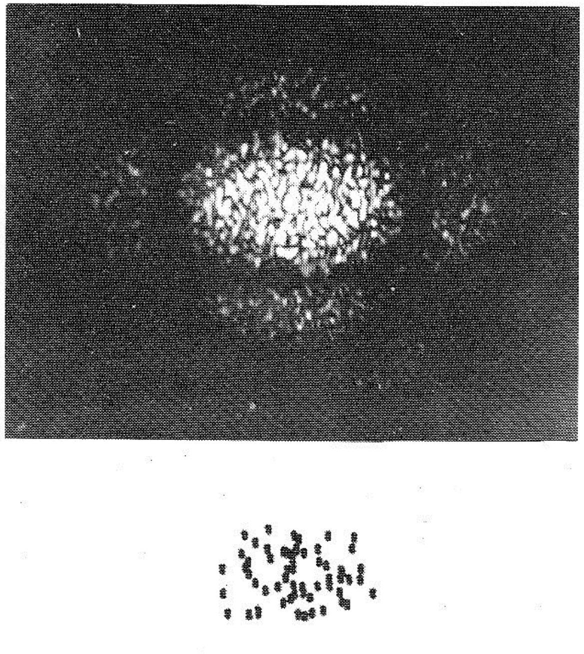


Fraunhofer diffraction of a rectangular aperture 8 x 7 mm^2, taken with mercury light λ=579nm
(from Born&Wolf, chap. 8)

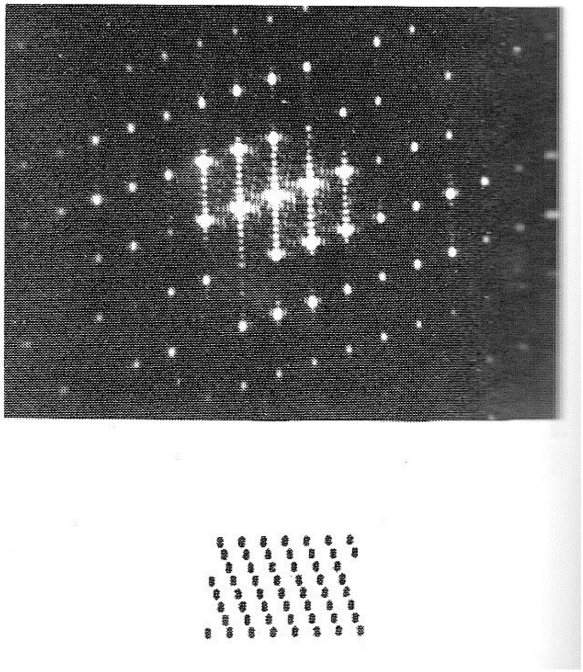


Fraunhofer diffraction of a circular aperture, taken with mercury light λ=579nm
(from Born&Wolf, chap. 8)

Speckle Pattern

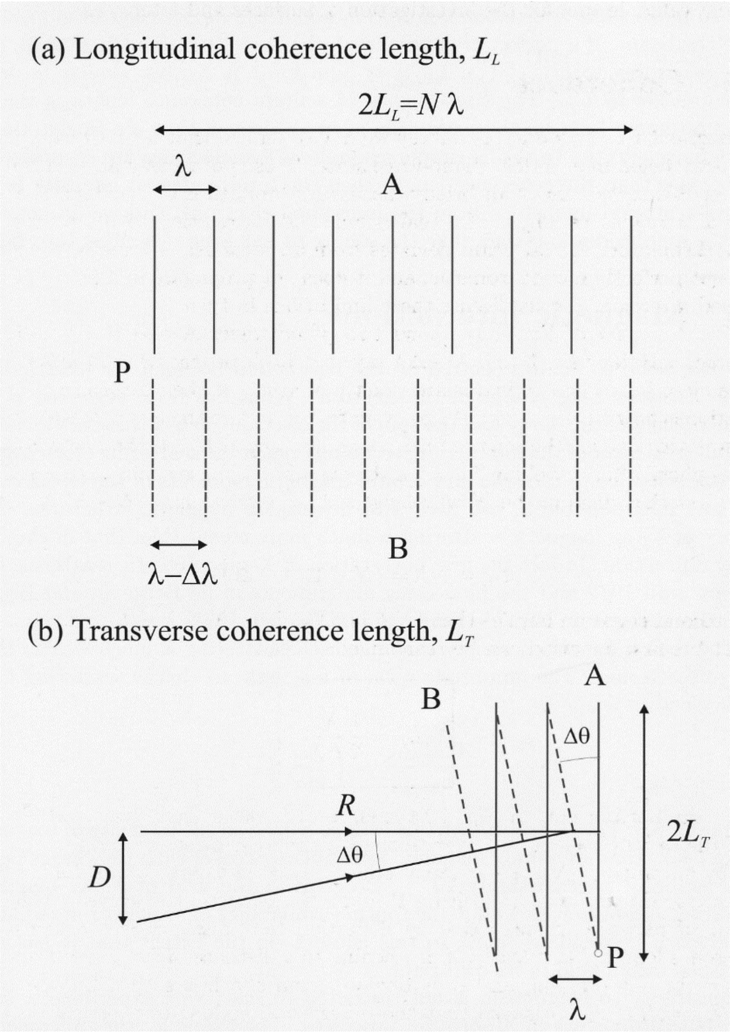


Random arrangement of apertures: speckle



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:

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

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

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:

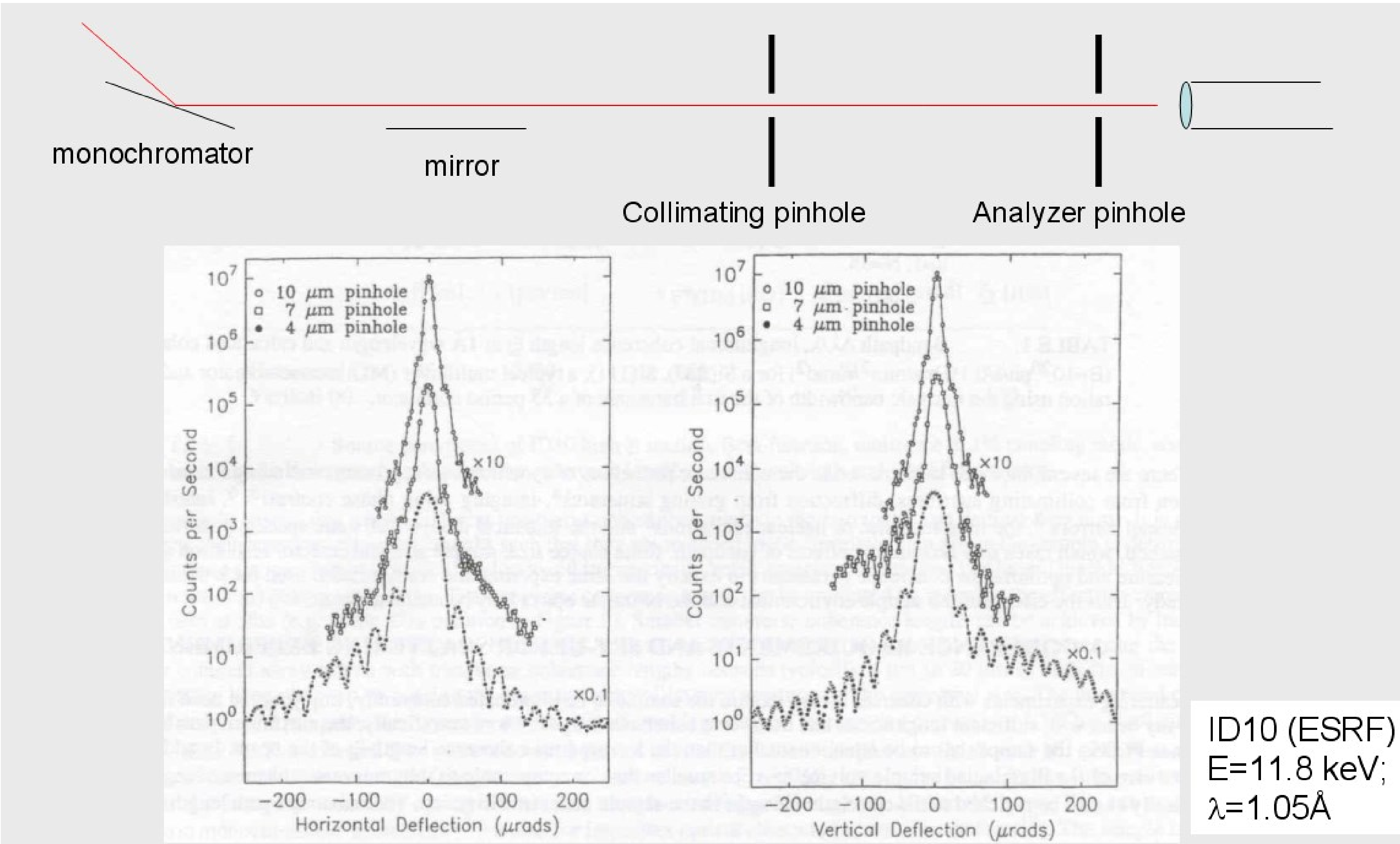
$2\xi_t\,\Delta\theta = \lambda$

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

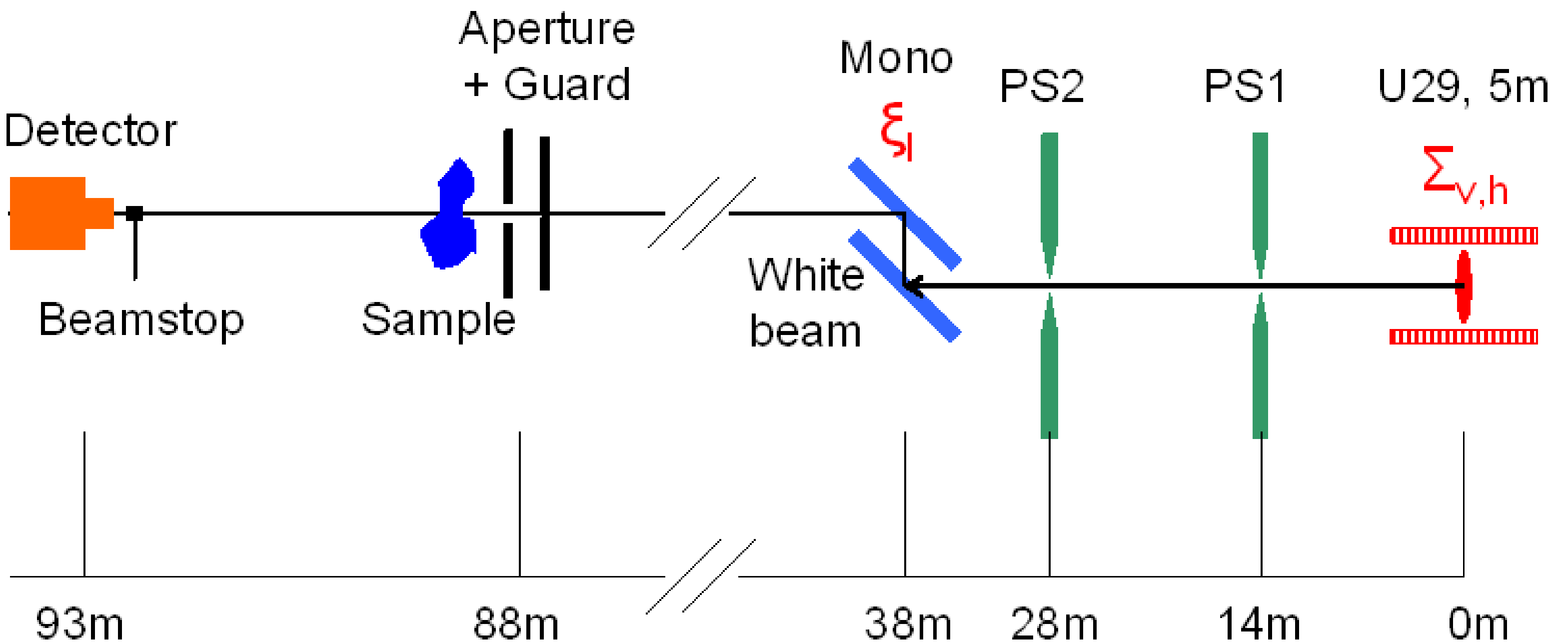
$\lambda = 0.1\text{nm}, R = 100\ \text{m}, D = 20 - 150\ \mu\text{m}$

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

Fraunhofer Diffraction (λ = 0.1nm)



Coherence Lengths of a Storage Ring Beamline



$$\Sigma_v \approx 5 - 10\mu m$$

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

$$\Sigma_h \approx 100 - 200\mu m$$

Speckle

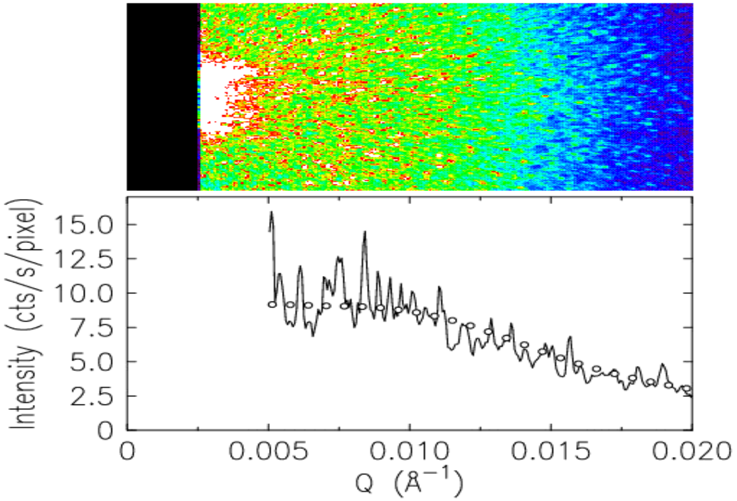
If coherent light is scattered from a disordered system it gives rise to a random (grainy) diffraction pattern, known as "**speckle**". A speckle pattern is an interference pattern and related to the exact spatial arrangement of the scatterers in the disordered system.

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

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

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

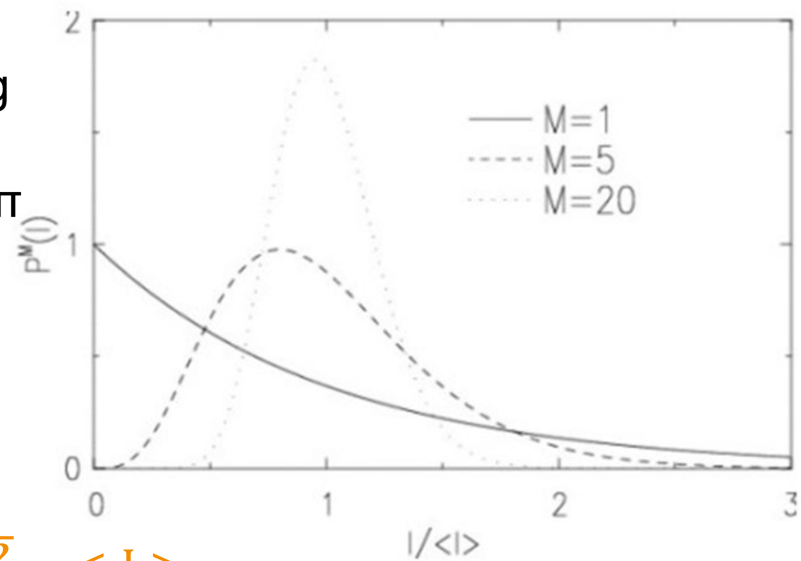
Aerogel
λ=1Å
CCD (22 μm)



Abernathy, Grübel, et al.

J. Synchroton Rad. 5, 37, 1998

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:

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

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

Contrast: ß = σ²/ $< I >^2$ **= 1**

<u>Partially coherent illumination:</u> the speckle pattern is the sum of M independent speckle patterns

$$\mathbf{P_M(I) = M^M \bullet \frac{\left(\frac{1}{<I>}\right)^{M-1}}{\Gamma(M)<I>} \bullet e^{-\frac{MI}{<I>}}}$$

Mean: $< \mathbf{I} >; \ \boldsymbol{\sigma} = \frac{<\mathbf{I}>}{\sqrt{\mathbf{M}}}$ $ß = 1/M$

Speckle

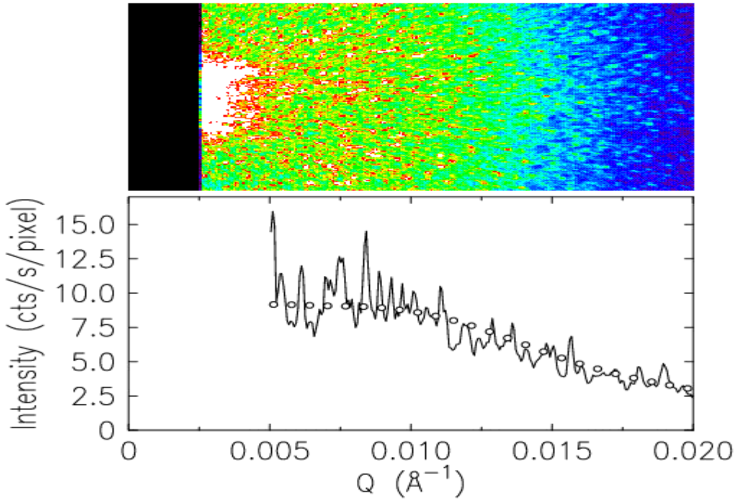
If coherent light is scattered from a disordered system it gives rise to a random (grainy) diffraction pattern, known as “speckle”. A speckle pattern is an interference pattern and related to the exact spatial arrangement of the scatterers in the disordered system.

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

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

<u>Incoherent Light:</u> $S(Q,t) = \langle S_c(Q,t) \rangle_{V \gg Vc}$ ensemble average

Aerogel
λ=1Å
CCD (22 μm)

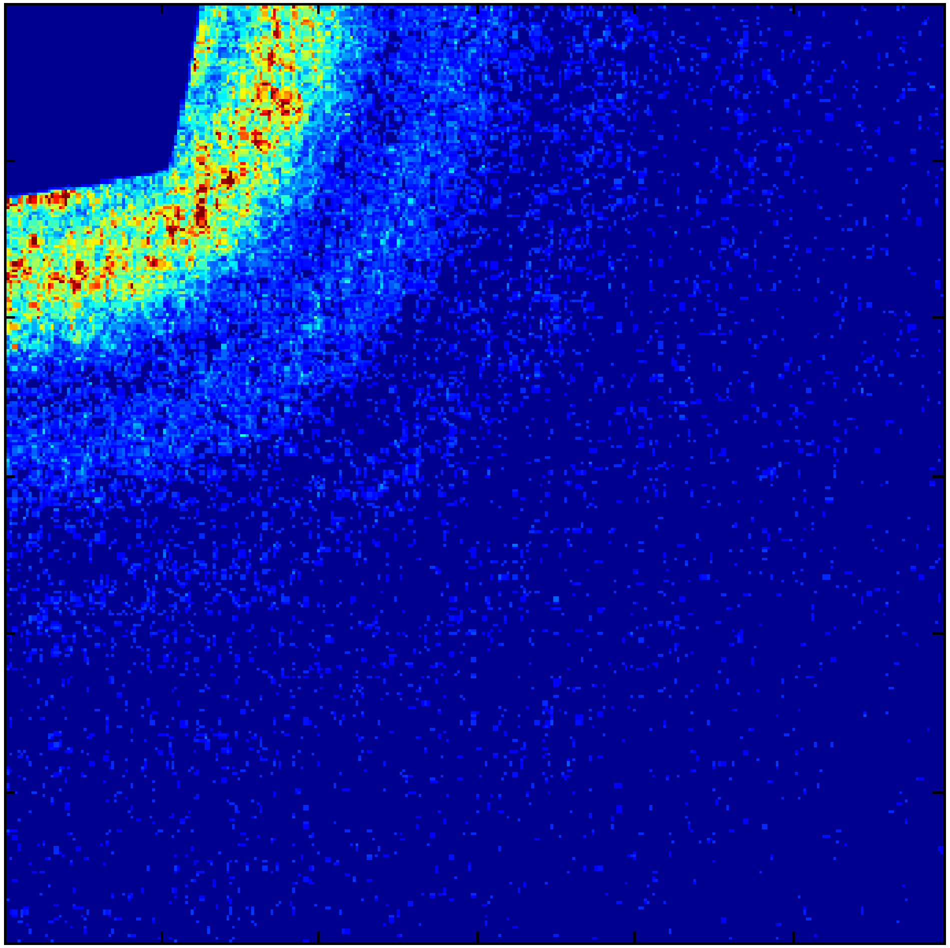


Abernathy, Grübel, et al.

J. Synchroton 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)

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

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

<u>Note:</u> E(Q,t) = ∫ dr' ρ(r') $e^{iQ \bullet r'(t)}$ ρ(r'): charge density

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

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

<>: ensemble av.; ß(Q): contrast

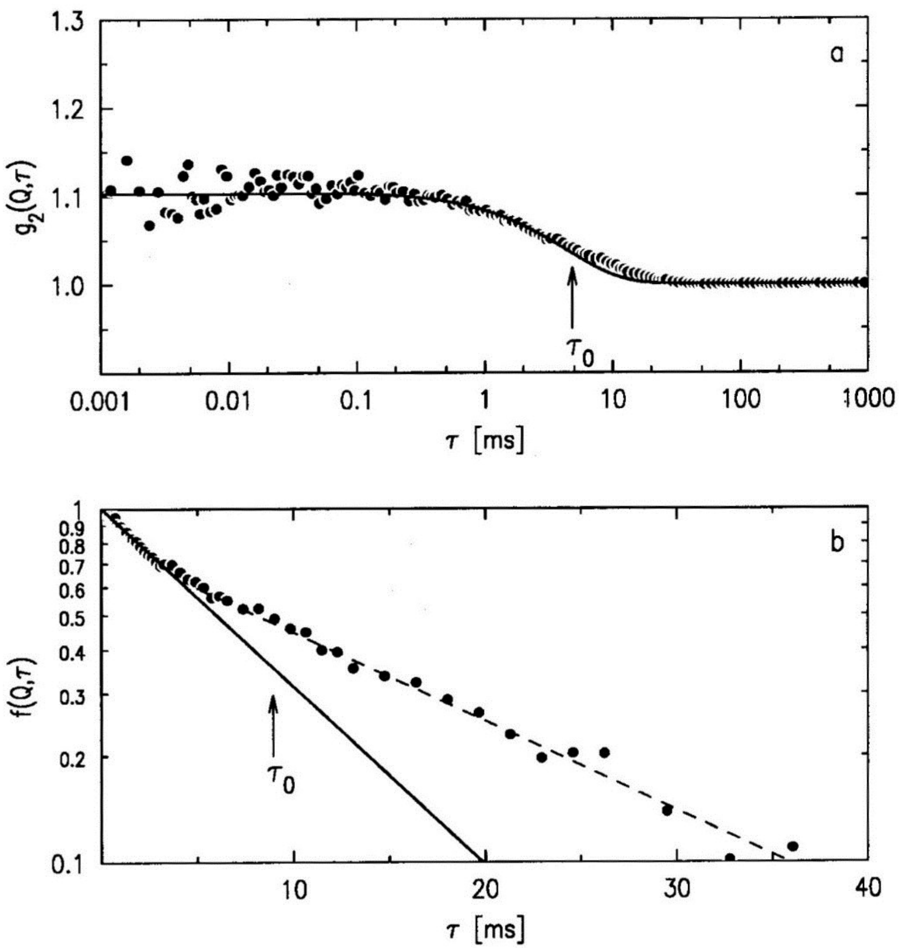
$g_2(Q,t) = 1 + ß(Q)\, |f(Q,t)|^2$ with f(Q,t) = S(Q,t)/S(Q,0)

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

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

Time Correlation Function $g_2(Q,t)$

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



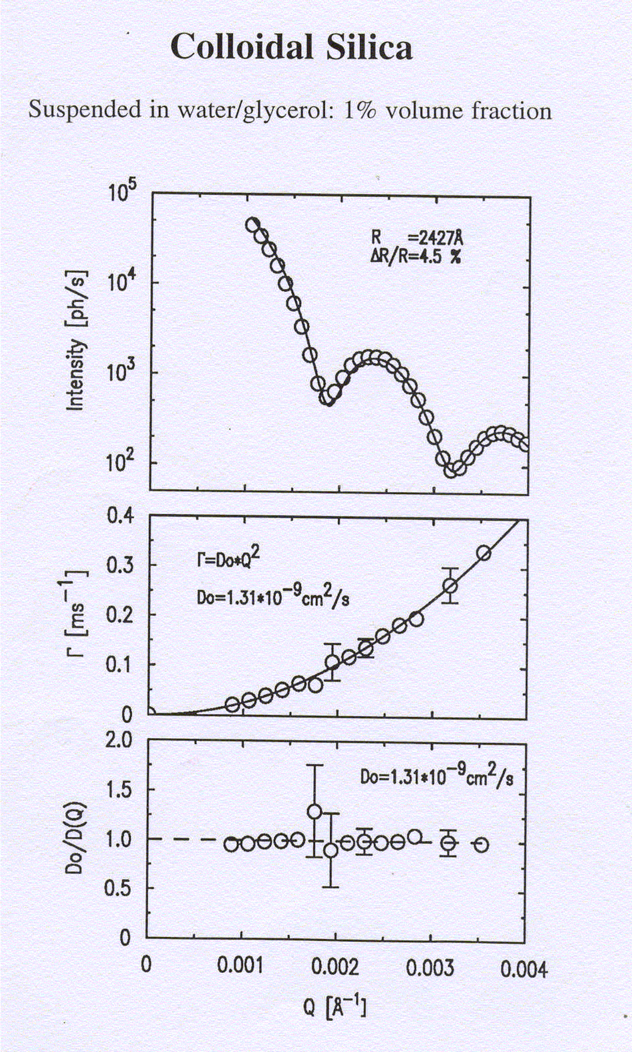
Dynamics in a Dilute, Non-interacting System

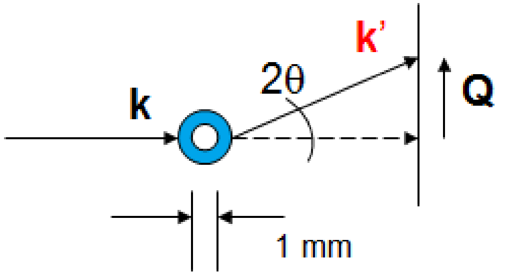
$$I \sim |F(Q)|^2 S(Q)$$

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

$$\Gamma = D_0\, Q^2$$

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





$Q = k' - k$

$Q = 2k \sin\theta$

$k = 2\pi/\lambda$

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

Outlook

Imaging Holographic Imaging, Ptychography,….

Impact of FEL sources……

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