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

## Part II Script 2

Vorlesung im GrK 1355

WS 2013

A. Hemmerich & G. Grübel

Location: SemRm 052, Gebäude 69, Bahrenfeld

Thursdays 12.15 – 13.45

G.Grübel (GR), A.Hemmerich (HE)

# 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://photon.physnet.uni-hamburg.de/fileadmin/user_upload/ILP/Hemmerich/teaching.html/Coherence.pdf***

Part II: **http://photon-science.desy.de/research/studentsteaching/lectures__seminars/ws_13_14/coherence_of_light_grk1355/…..**

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

## Part II: 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

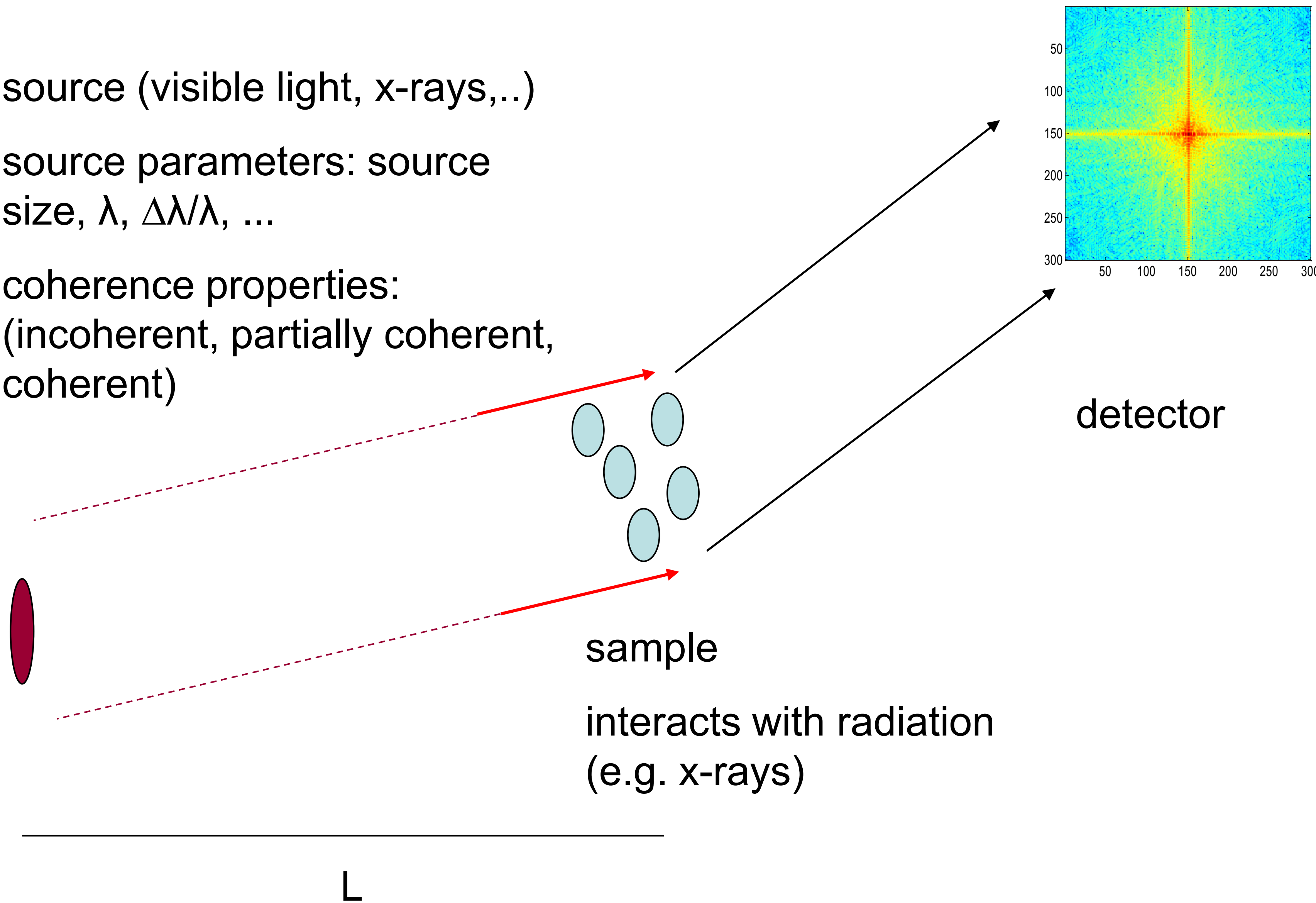
### Imaging and XPCS at FEL Sources

# Introduction: Experimental Set-Up

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

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

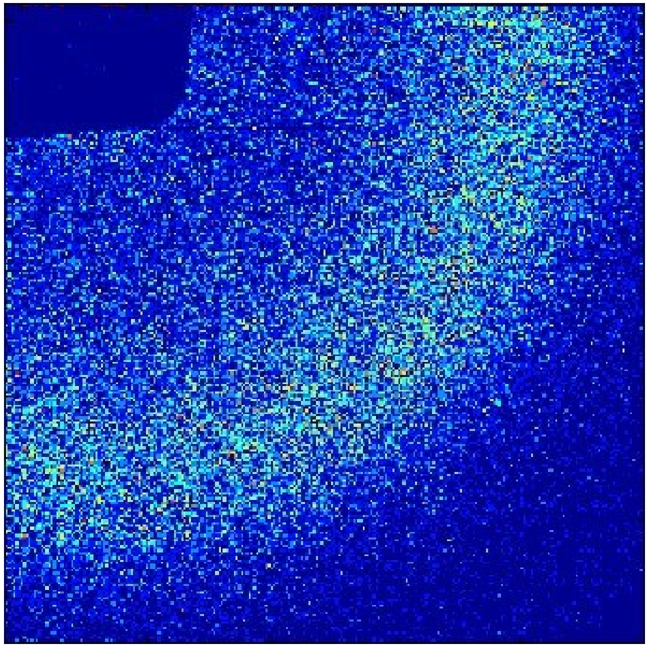
coherence properties: (incoherent, partially coherent, coherent)



# 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.

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



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

Incoherent Light:

$S(Q,t) = < S_c(Q,t)>_{V>>c}$

ensemble average

▪

# Coherence based X-ray techniques:

# An X-ray Scattering Primer

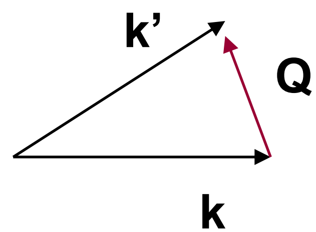
# Scattering of X-rays: A primer

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

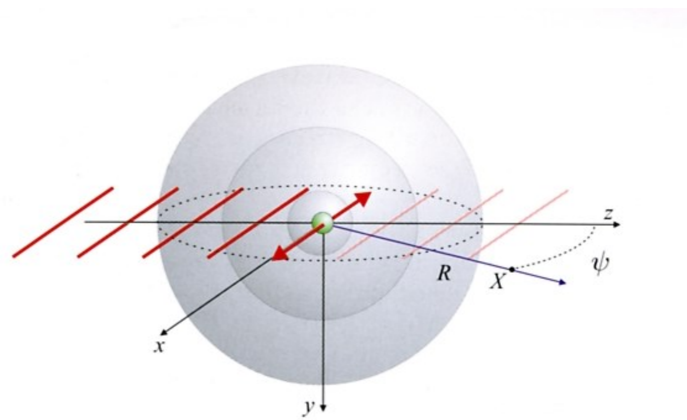
$\mathbf{E}(\mathbf{r},t) = \boldsymbol{\varepsilon}\, Eo \exp\{i(\mathbf{kr}-\omega t)\}$ with $|\mathbf{k}|=2\pi/\lambda$, $\lambda[Å]=hc/E$, $\omega=2\pi/\nu$

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 $r_o$

($=2.82*10^{-5}$ Å)

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## scattering by a single atom:

phase factor

scattering amplitude by an ensemble of electrons

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

(atomic) formfactor — position of scatterers

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

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 from an atom:

scattering amplitude of an atom ≡ atomic form factor $f_0(Q)$ [in units of $r_0$]

$\rho(r)$: electronic number density ≡ charge density

$$f_0(\mathbf{Q}) = \int \rho(\mathbf{r}) \exp(i\mathbf{Q}\mathbf{r})\, d\mathbf{r}$$

$$= \begin{cases} Z & Q \rightarrow 0 \\ 0 & Q \rightarrow \infty \end{cases}$$

| | $a_1$ | $b_1$ | $a_2$ | $b_2$ | $a_3$ | $b_3$ | $a_4$ | $b_4$ | $c$ |
|---|---|---|---|---|---|---|---|---|---|
| C | 2.3100 | 20.8439 | 1.0200 | 10.2075 | 1.5886 | 0.5687 | 0.8650 | 51.6512 | 0.2156 |
| O | 3.0485 | 13.2771 | 2.2868 | 5.7011 | 1.5463 | 0.3239 | 0.8670 | 32.9089 | 0.2508 |
| F | 3.5392 | 10.2825 | 2.6412 | 4.2944 | 1.5170 | 0.2615 | 1.0243 | 26.1476 | 0.2776 |
| Si | 6.2915 | 2.4386 | 3.0353 | 32.333 | 1.9891 | 0.6785 | 1.5410 | 81.6937 | 1.1407 |
| Cu | 13.338 | 3.5828 | 7.1676 | 0.2470 | 5.6158 | 11.3966 | 1.6735 | 64.820 | 1.5910 |
| Ge | 16.0816 | 2.8509 | 6.3747 | 0.2516 | 3.7068 | 11.4468 | 3.683 | 54.7625 | 2.1313 |
| Mo | 3.7025 | 0.2772 | 17.236 | 1.0958 | 12.8876 | 11.004 | 3.7429 | 61.6584 | 4.3875 |

table 4.1: J. Als-Nielsen & D. McMorrow

note: atomic form factor is FT of electronic charge distribution

$f_0(Q/4\pi)$ tabulated:

$$f_0(Q/4\pi) = \sum_{j=1}^{4} a_j \exp -b_j(Q/4\pi)^2 + c$$

note:

$f = f_0(Q) + f' + f''$

corrections $f'$ and $f''$ arise from the fact that the electrons are bound in the atom

# Scattering from a molecule:

$F^{mol}(Q) = \sum_{rj} f_j(Q)\, \exp(i\mathbf{Q}\mathbf{r}_j)$

——— $CF_4$

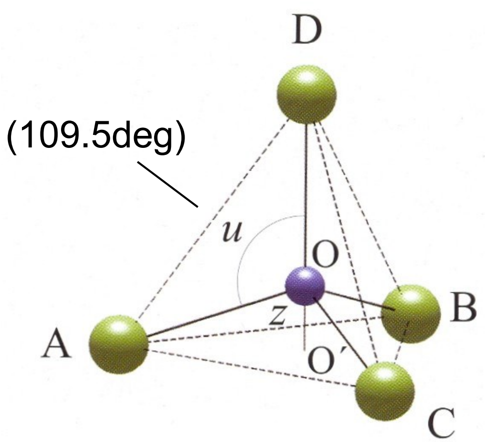
-. - . - $CF_4$ Q not || C-F

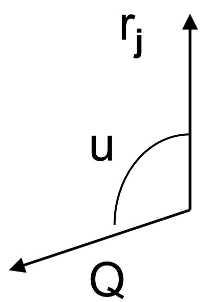
- - - - - molybdenum (also 42 electrons)

<u>example: $CF_4$:</u>

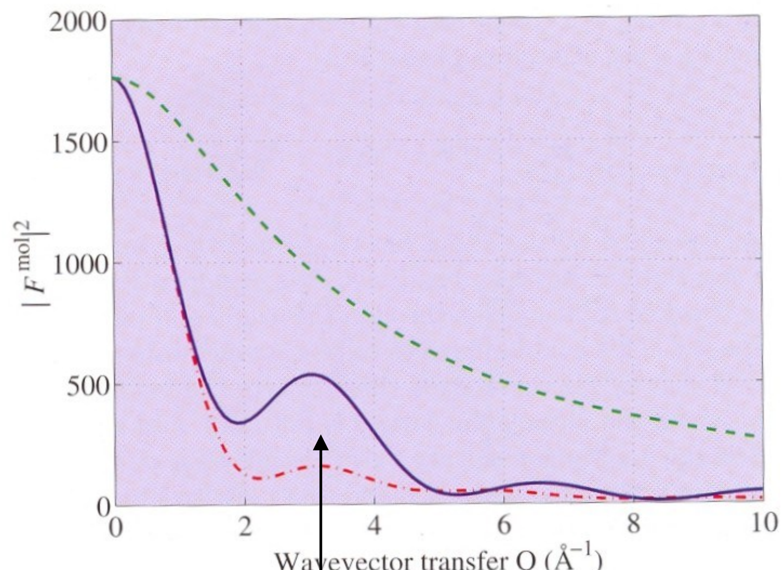
assume OA=OB=OC=OD=1; z=OO'=cos(u) =1/3

(109.5deg)




$\mathbf{Q}\mathbf{r}_j = Qr_j \cos(u) = (1/3)Qr_j$




(F-F bond)

<u>Assume:</u> Q || C-F bond

$F^{mol} = f^C(Q) + f^F(Q)\,[\exp(iQR) + 3\exp(i\mathbf{Q}\mathbf{r}_j)\}$

$= f^C(Q) + f^F(Q)\,[\,3\exp(-/+iQR/3) + \exp(+/-iQR)]$

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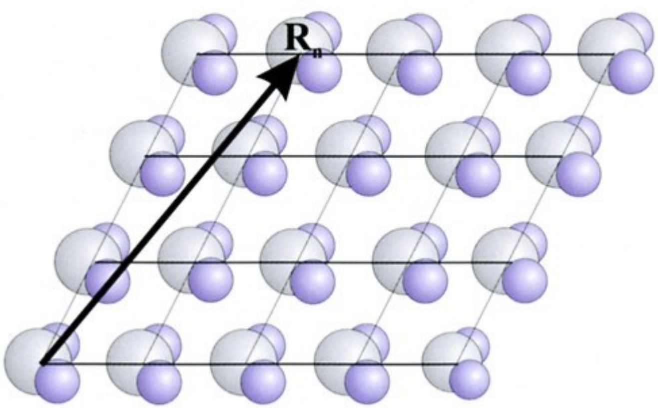
## scattering by a crystal:




$r_{j'} = R_n + r_j$

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

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

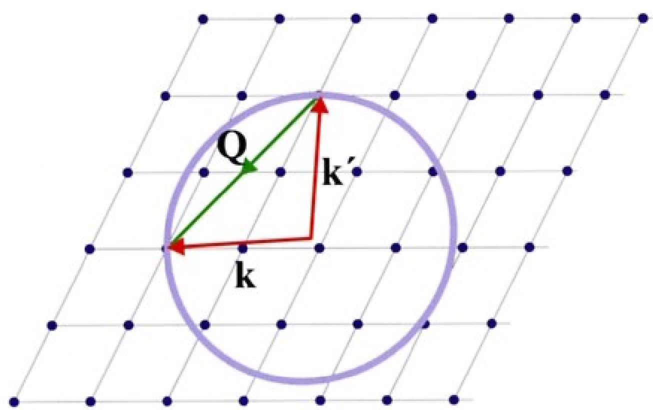
## evaluation of lattice sums:

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

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

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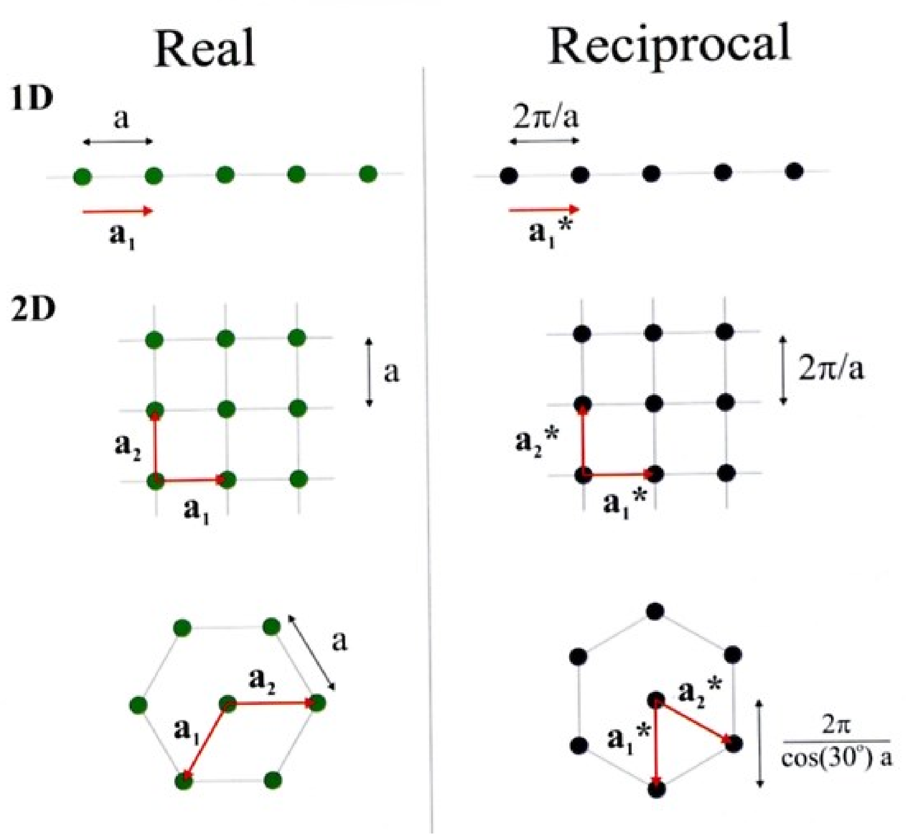
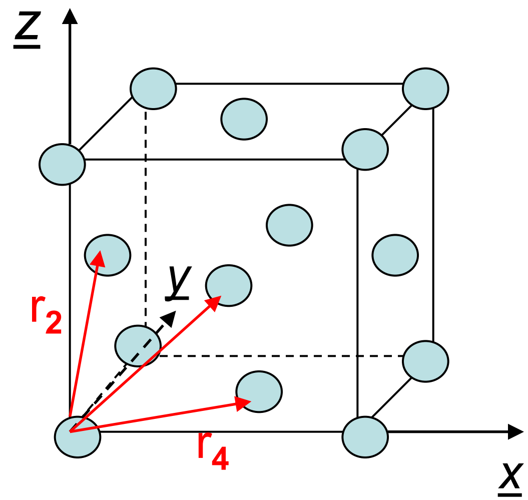
## construction of reciprocal space:

(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




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

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

$r_1 = 0$, $r_2 = \frac{1}{2} a (\underline{y} + \underline{z})$, $r_3 = \frac{1}{2} a (\underline{z} + \underline{x})$, $r_4 = \frac{1}{2} a(\underline{x} + \underline{y})$

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

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

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

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

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

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

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

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

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

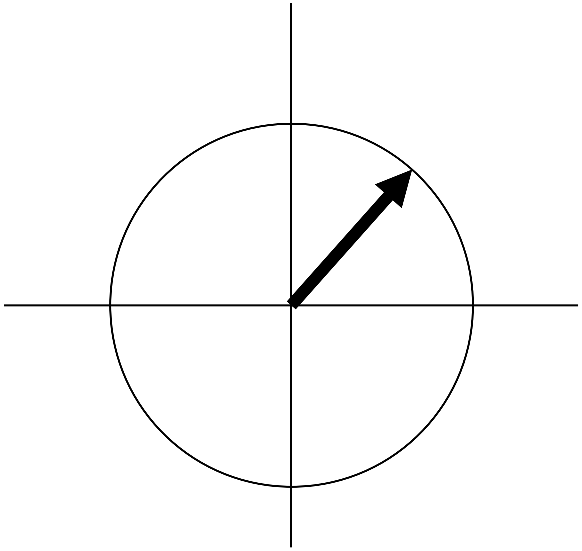
- # The unit cell structure factor for a fcc lattice

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

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

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

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



Reflections:

100 forbidden

111 allowed

200 allowed

■

# Coherence based X-ray techniques:

# Sources of Coherent X-rays

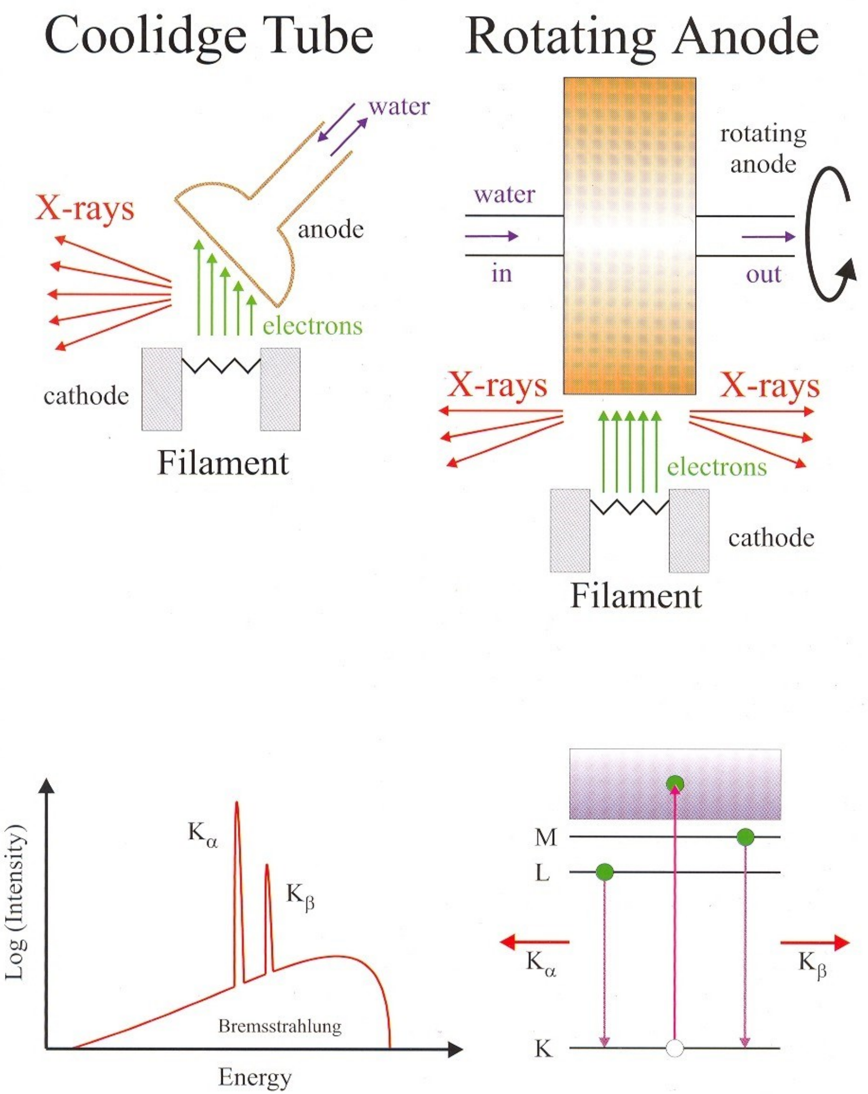
# ▪ Sources of X-Rays

1895 discovered by W.C. Röntgen

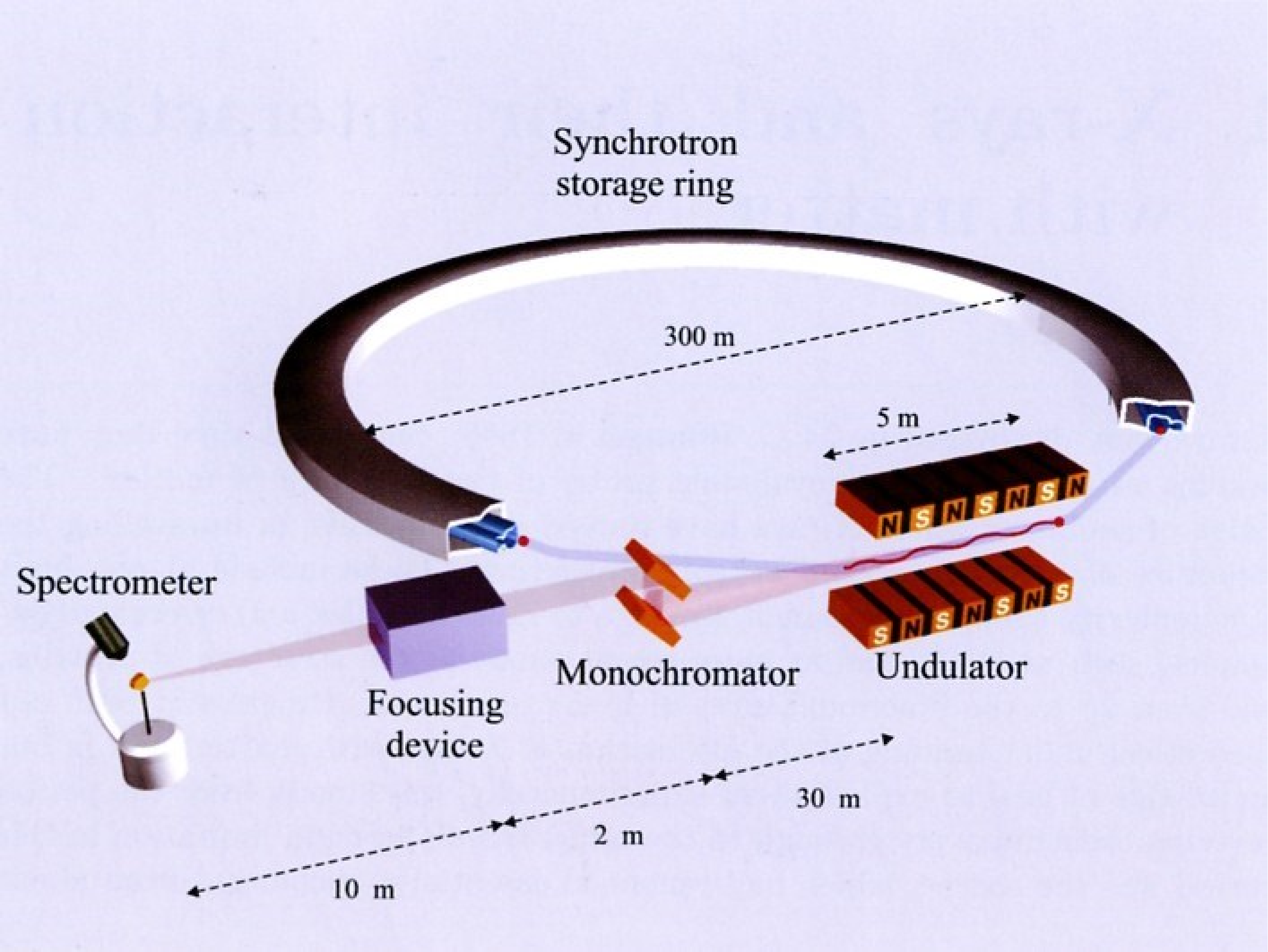
1912 First diffraction experiment (v. Laue)

1912 Coolidge tube (W.D. Coolidge,GE)

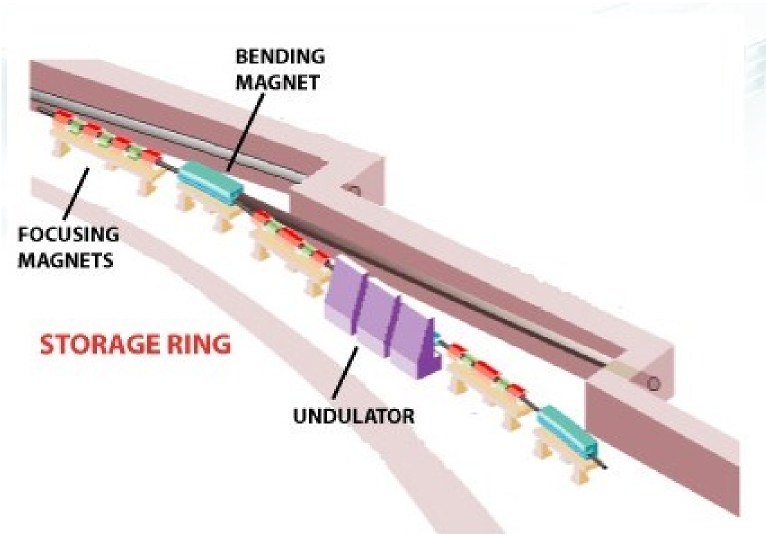
1946 Radiation from electrons in a synchrotron, GE, Physical Review, 71,829(1947)



# A storage ring facility



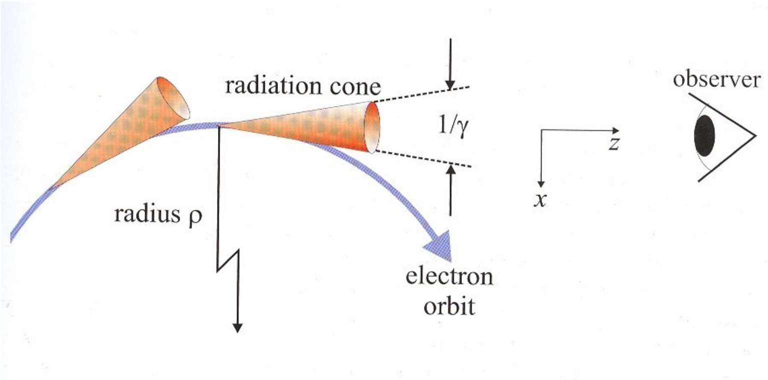
# Synchrotron Radiation Primer




Energy $E_e$ of an electron at speed v:

$$E_e = mc^2/\mathrm{sqrt}\{(1-(v/c)^2\} = \gamma\, mc^2$$

For 5GeV and $mc^2$=0.511 MeV get $\gamma \approx 10^4$




Centrifugal=Lorentz force yields for radius:

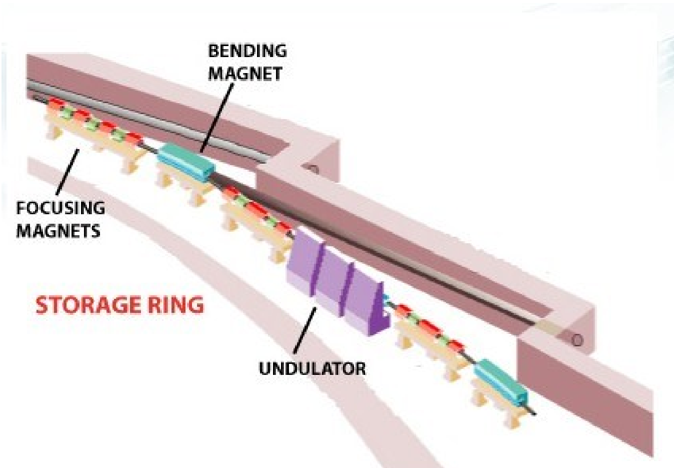
$\rho = \gamma mc /eB = 3.3\ E[GeV]/B[T] \approx 25$ m

$E_e \approx 6$ GeV, B=0.8 T

Opening angle is of order $1/\gamma \approx 0.1$ mrad


Coherence of light and matter: from basic concepts to modern applications, Vorlesung im GrK 1355, WS 2013 A. Hemmerich & G. Gruebel

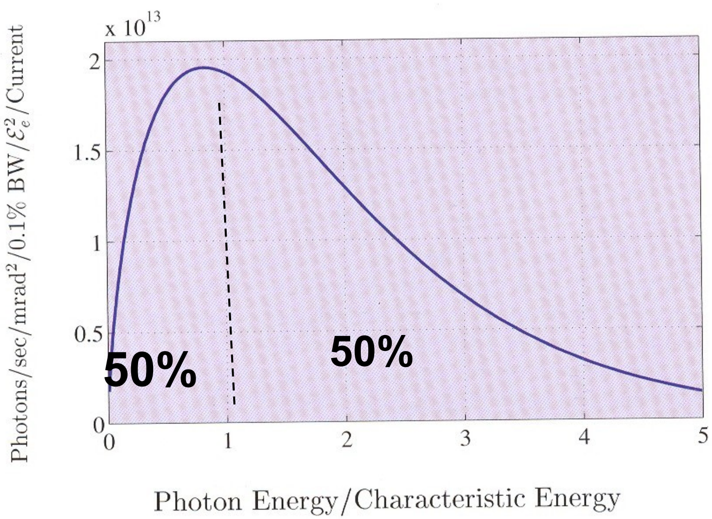
# Bending magnets




<u>Characteristic energy $\hbar\omega_c$ for bend or wiggler:</u>

$\hbar\omega_c$ [keV] = 0.665 $E_e^2$[GeV] B(T) ≈20 keV

Flux ~ $E^2$




<u>Energy loss by synchrotron radiation per turn:</u>

$$\Delta E \text{ [keV]} = 88.5\ E^4\text{[GeV]}/\rho\text{[m]}$$

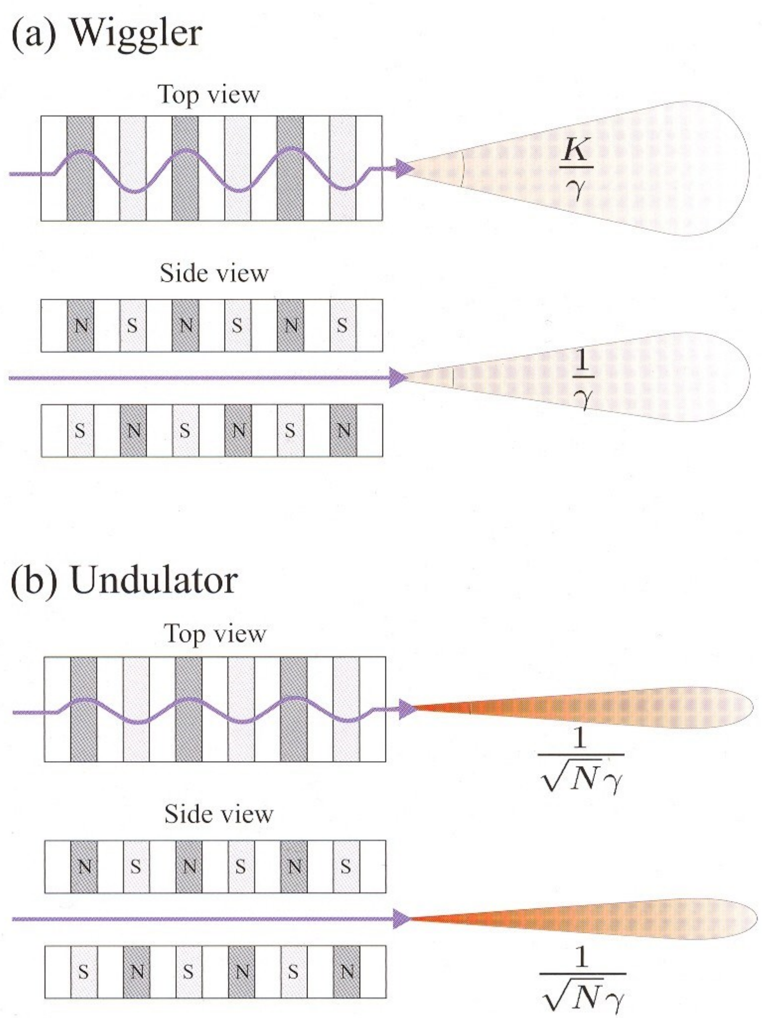
For 1 GeV and ρ=3.33 m: ΔE =26.6 keV/turn

For I=500 mA ≡ 0.5 Cb/s =0.5x6.25x$10^{18}$ $e^-$/s

➔P= 0.5x6.25x$10^{18}$ $e^-$/sx26.6 keV

=8.3125x$10^{22}$x1.6x$10^{-19}$ = 13.3 KJ/s =13.3 KW

# Insertion Devices (wigglers and undulators)




## Wiggler:

$P[kW] = 0.633 E_e^2[GeV]\, B^2[T]\, L[m]\, I[A]$

Flux ~ $E^2 \times N$

N: number poles

## Undulator:

$k = eB / mc\ k_u = 0.934\ \lambda_u\ [cm]\ Bo[T]$

with $\lambda_u$ undulator period

undulator fundamental:

$\lambda_0 = \lambda_u/2\gamma^2 \{(1 + k^2/2 + (\gamma\theta)\}$

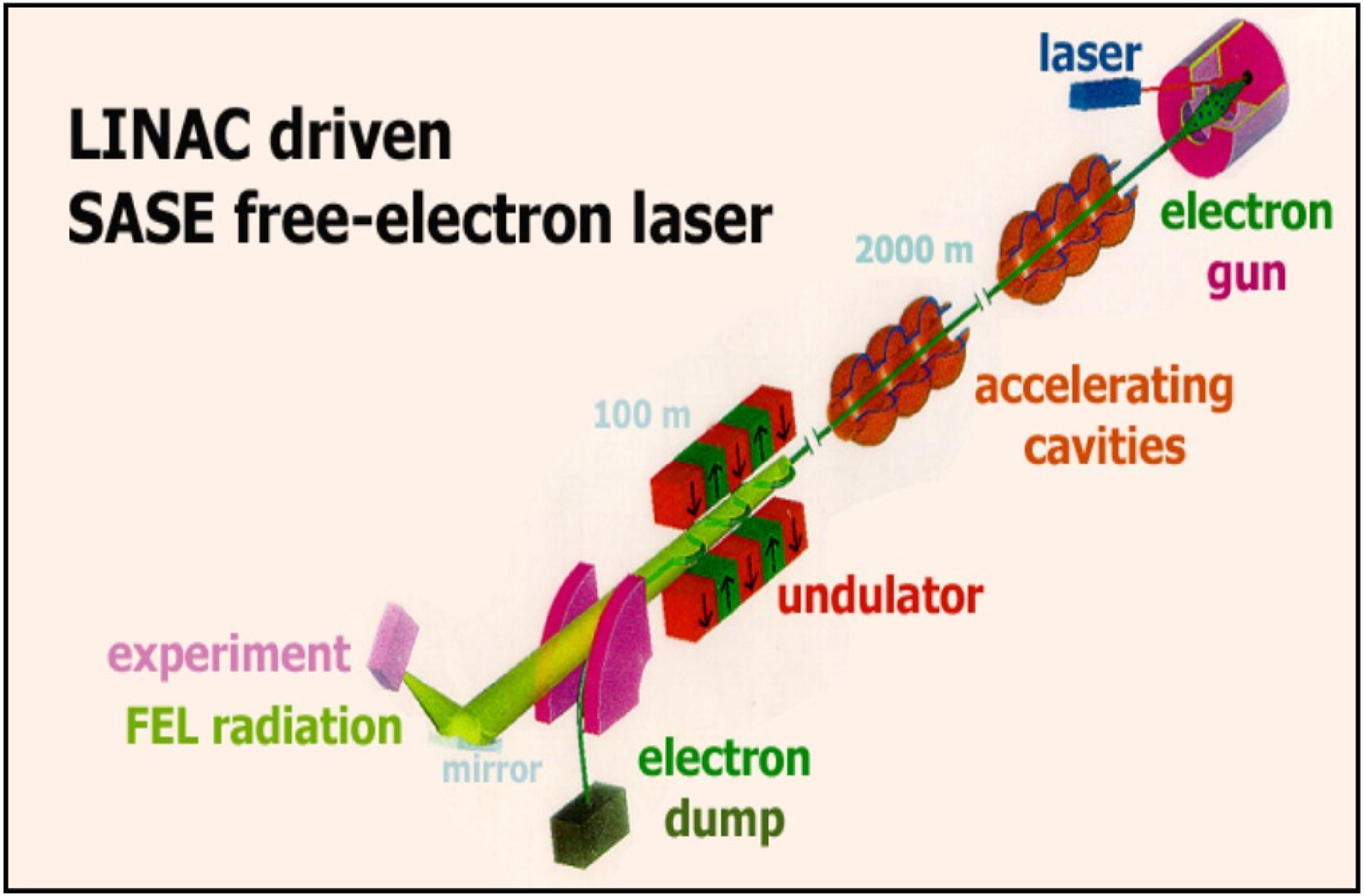
on axis

Flux ~ $E^2 \times N^2$

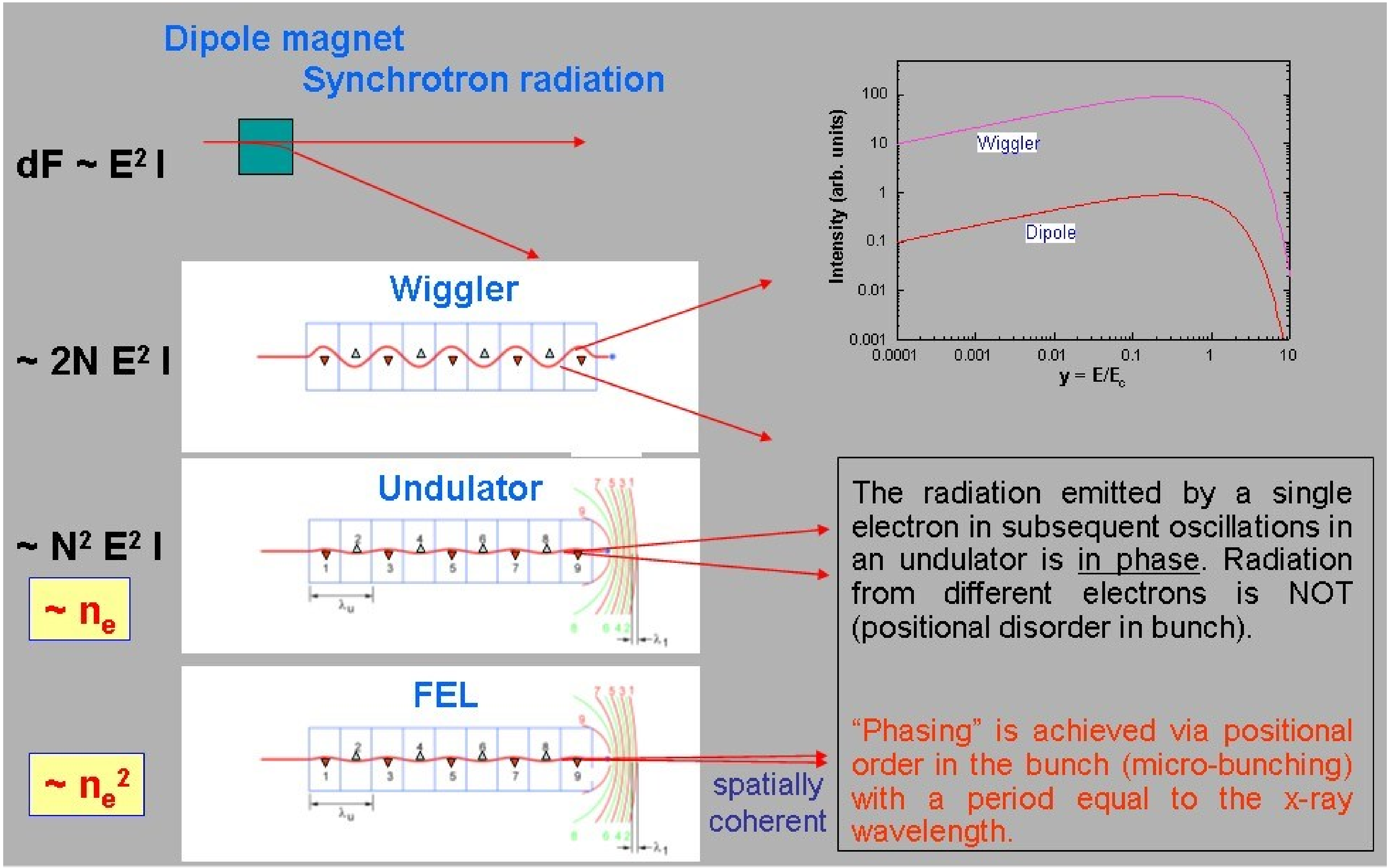
bandwidth:

$\Delta\lambda/\lambda \sim 1/nN$

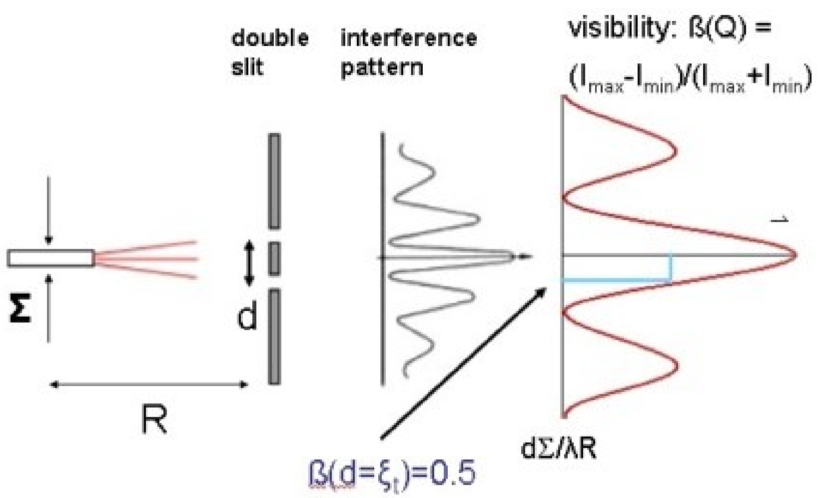
# A Free Electron Laser (FEL)



# ▪ Synchrotron and FEL sources



# Coherence parameters of an undulator source




Coherent Flux:

$F_c = (\lambda/2)^2 \bullet B$

$= 3.5 \bullet 10^{10}$ ph/s

$B = 10^{20}$ ph/s/mm²/mrad²/0.1%bw

$\Delta\lambda/\lambda = 10^{-4}$; $\lambda = 1$ Å

Temporal Coherence:

londitudinal coherence length

$\xi_l = \lambda(\lambda/\Delta\lambda) = 1$ µm

$\Delta\lambda/\lambda = 10^{-4}$; $\lambda = 1$ Å

Transverse coherence length:

$\xi_t^2 \bullet \xi_l$ defines coherence volume

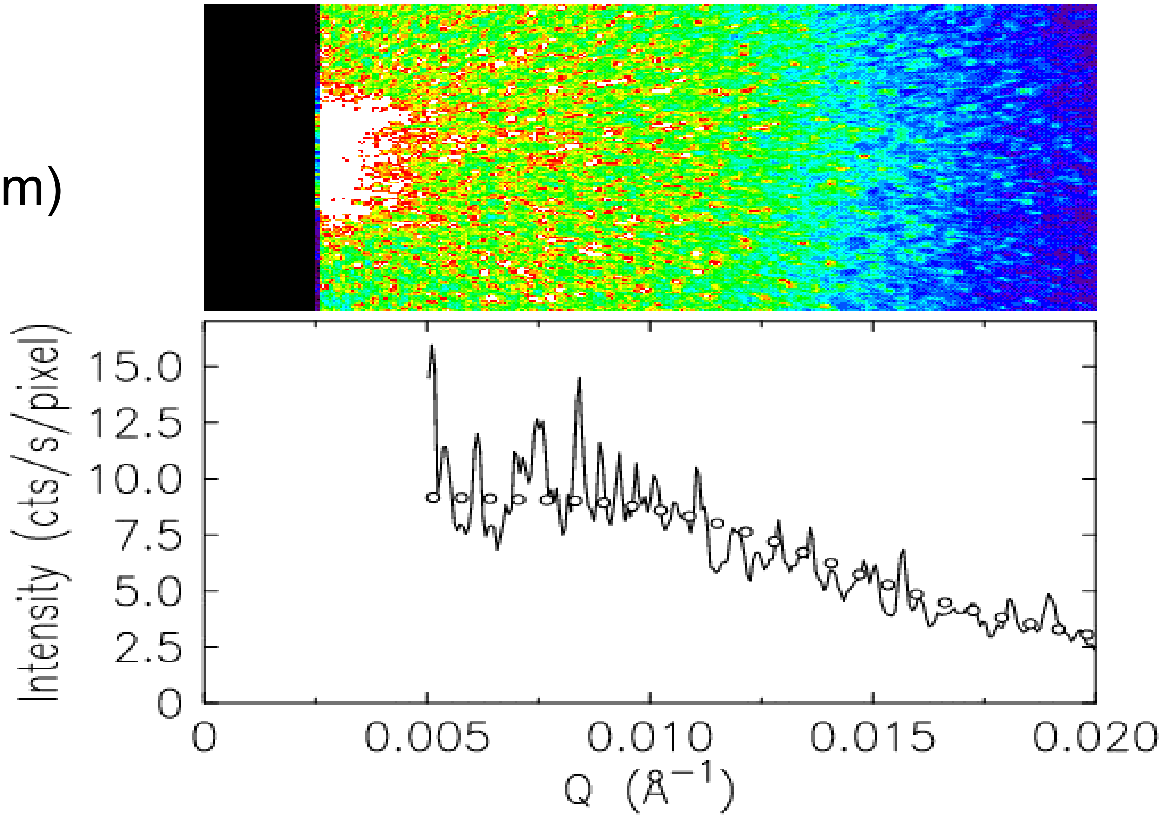
$\xi_t = (\lambda/2)(R/\Sigma)$ = 2.5µm (h), $\Sigma_x$=1mm

= 25µm (h), $\Sigma_z$=0.1mm

(λ=1Å, R=50m)

# Speckle pattern from a porous silica gel

Aerogel
λ=1Å
CCD (22 µm)

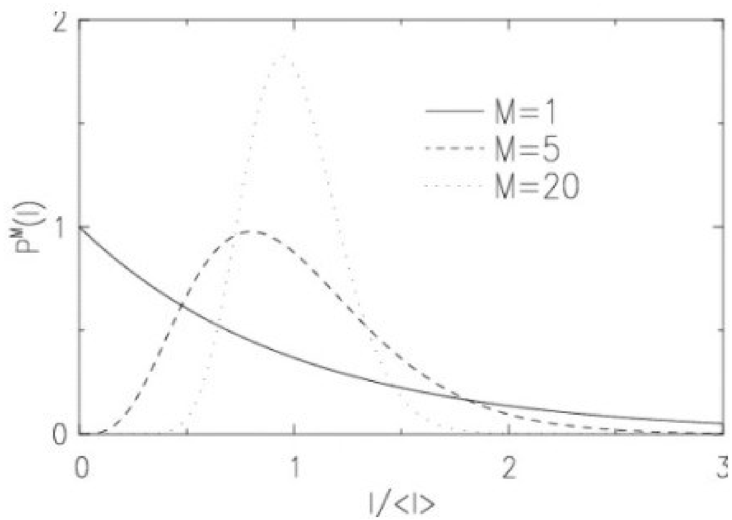



Abernathy, Grübel, et al.J. Synchroton Rad. 5, 37, 1998

## Statistical Analysis of Speckle Pattern (1)

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

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




Mean: <i>

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

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

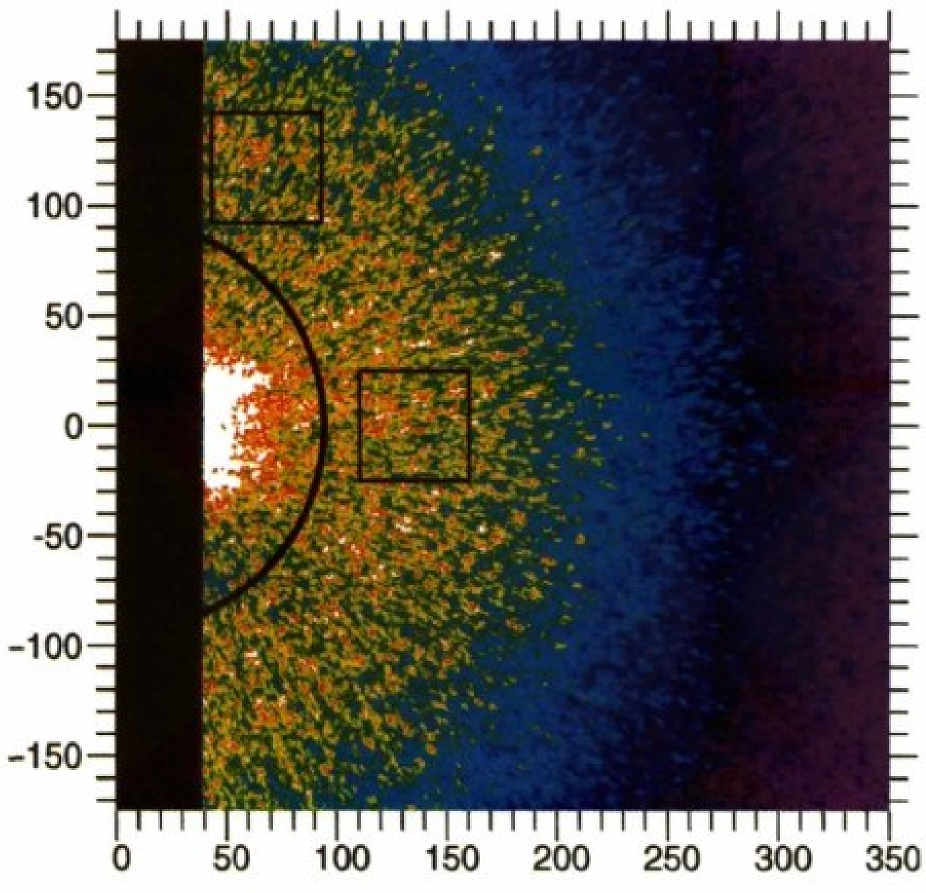
Mean: $\langle I\rangle$

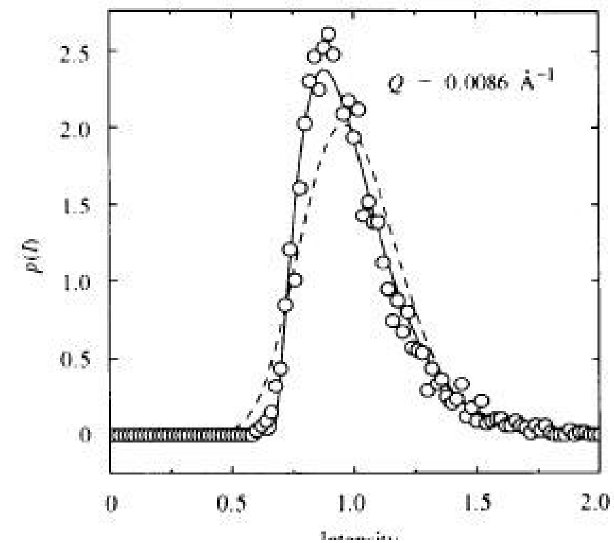
$\sigma = \langle I\rangle/M^{1/2}$

$ß = 1/M$

# Statistical Analysis of Speckle pattern (2)

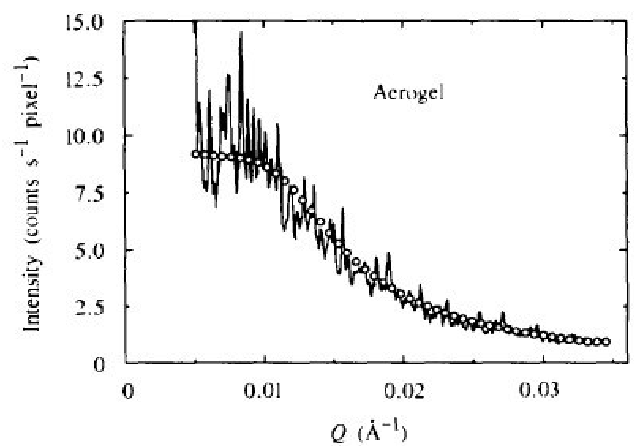
Aerogel, λ=1Å, CCD (22 μm)




M=25 (---)

M=2.7




normalized two-point correlation function:

$$C(\mathbf{r}_1,\mathbf{r}_2) = [ \langle I(\mathbf{r}_1)\bullet I(\mathbf{r}_2)\rangle / \langle I(\mathbf{r}_1)\rangle\bullet\langle I(\mathbf{r}_2)\rangle ] -1$$

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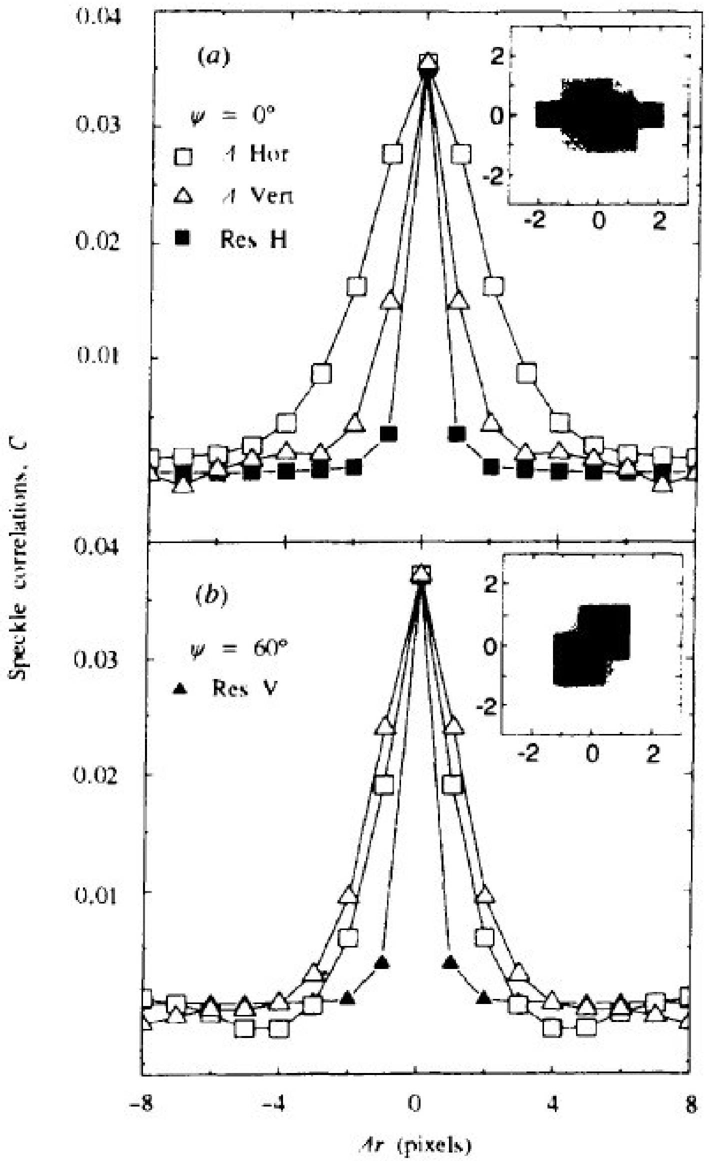
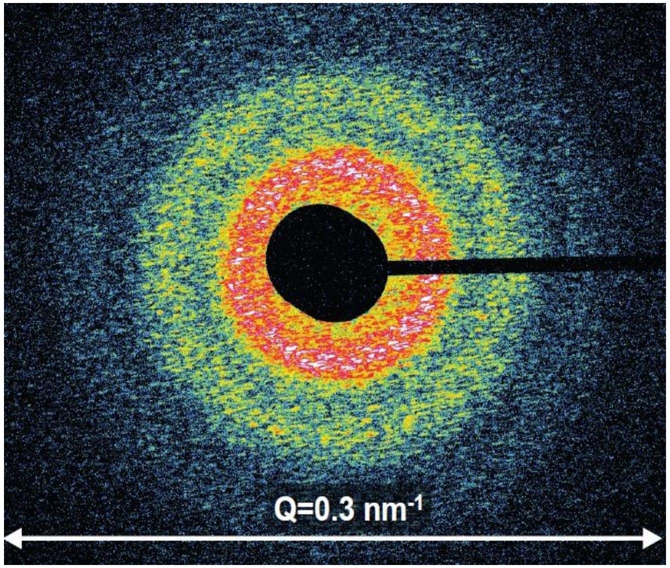
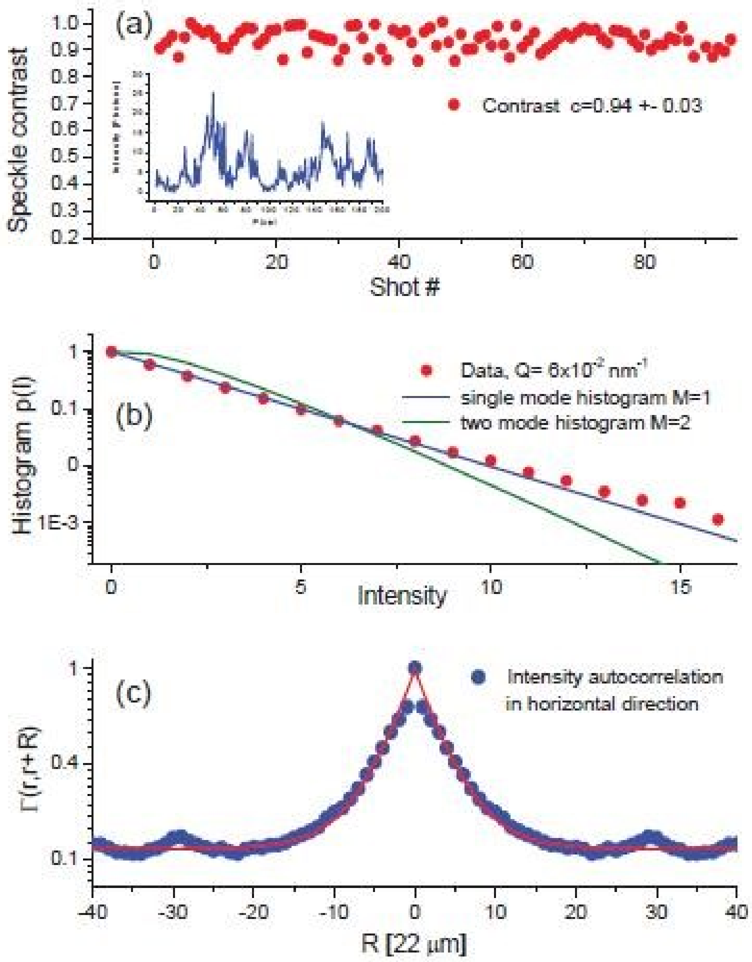



$C(\mathbf{r}_1,\mathbf{r}_2) = [\ \langle I(\mathbf{r}_1)\bullet I(\mathbf{r}_2)\rangle / \langle I(\mathbf{r}_1)\rangle\bullet\langle I(\mathbf{r}_2)\rangle ]\ -1$

width: $\Delta C$; contrast: $ß = C(\mathbf{r},\mathbf{r})$

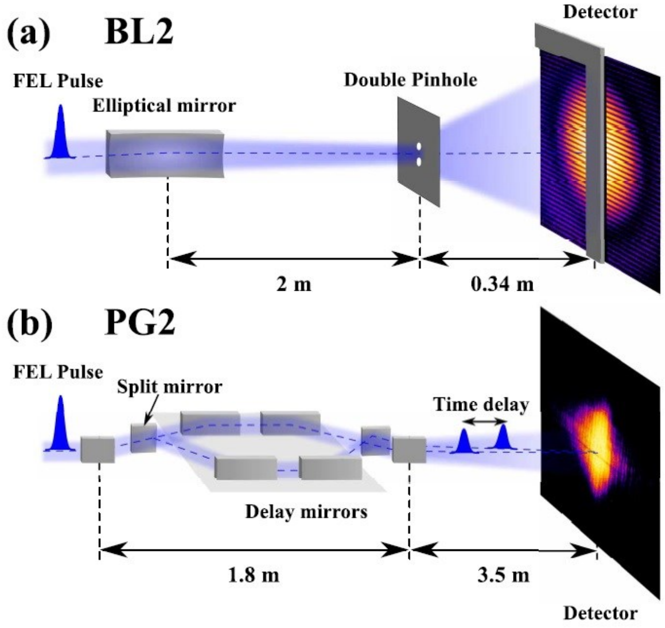
# ▪ The Linac Coherent Light Source (LCLS)

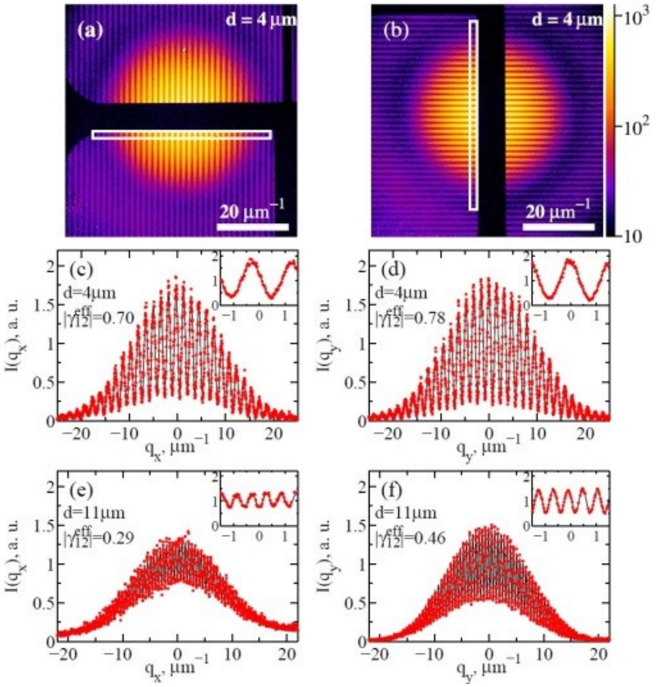


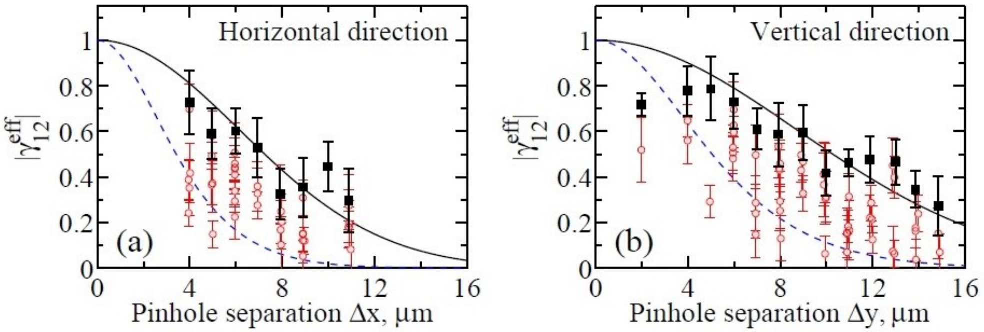




Single pulse hard X-ray speckle pattern captured from nano-particles in a colloidal liquid (photon wave-length $\lambda$= 1.37 Å

# Coherence properties of the FLASH FEL

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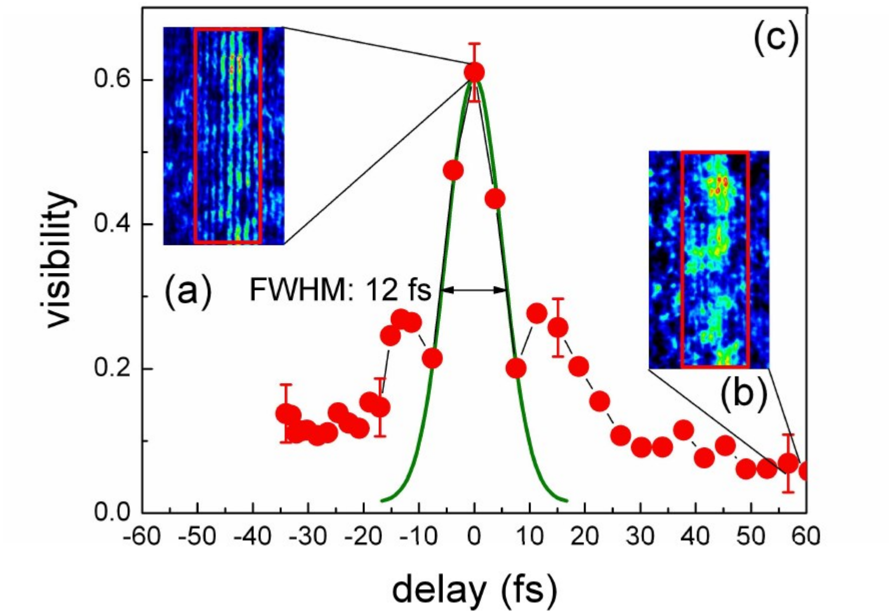
# Spatio-temporal coherence properties of FEL pulses

**Two beam interference**:

fringe spacing = $\lambda$ / sin $\alpha$
coherence length $l_c$= sqrt(2ln2/$\pi$) ($\lambda^2/\Delta\lambda$)
coherence time $\tau_c$= sqrt(2ln2/$\pi$)($\lambda^2/\Delta\lambda$)/c




**FLASH:** **$\lambda$ = 24 nm**

**$\alpha$ = 0.18 – 0.7 mrad**

**$\tau_c$= 7.5 fs**

R. Mitzner, B. Siemer, M. Neeb, T. Noll, F. Siewert
S. Roling, M. Rutkowski, A.A. Sorokin, M. Richter,
P. Juranic, K. Tiedtke, J. Feldhaus, W. Eberhardt,
and H. Zacharias

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Imaging techniques:

Lensless Imaging, Fourier Transform Holography