Imaging and dynamics using scattered coherent X-rays

School on X-ray Imaging Techniques at the ESRF

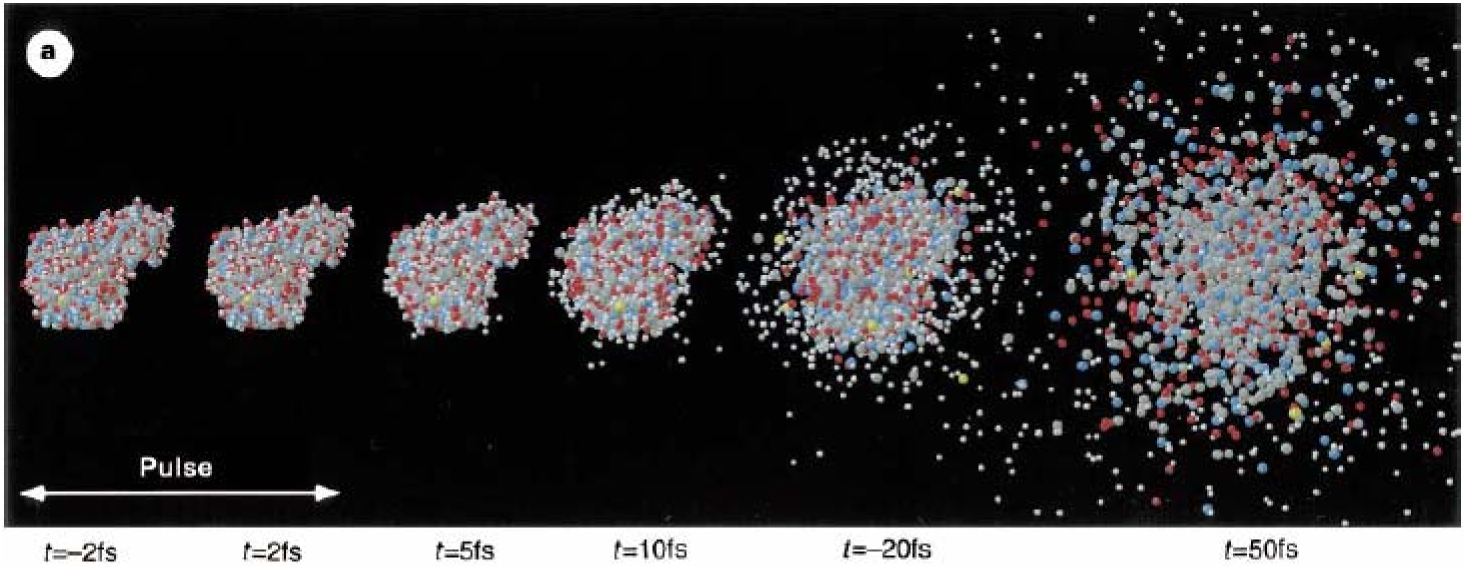
Grenoble

February 5-6, 2007

Anders Madsen
European Synchrotron Radiation Facility

ESRF

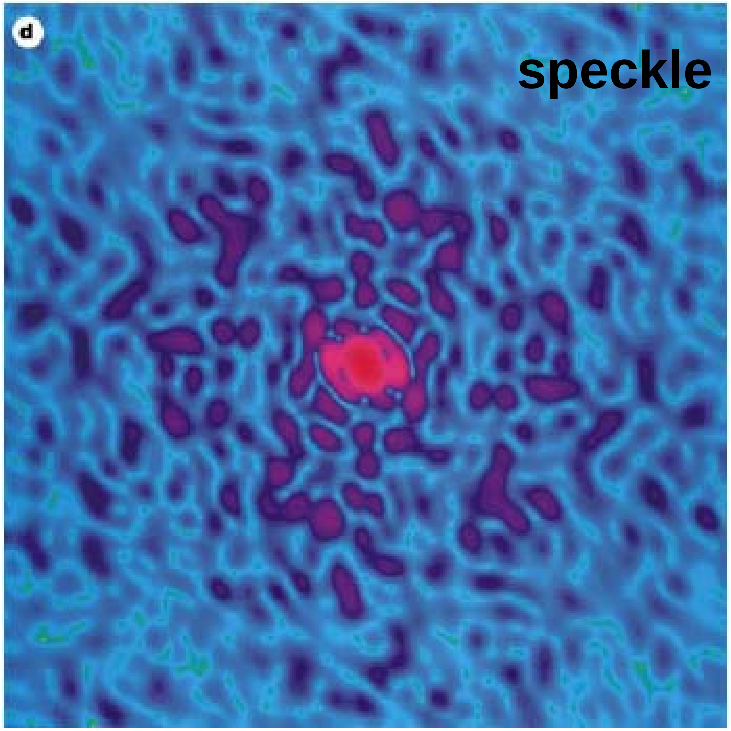
Diffraction imaging of biomolecules with coherent femtosecond X-FEL pulses



Coulomb explosion of T4 lysozyme

R. Neutze *et al.*, Nature **406**, 752 (2000)

Diffraction imaging of biomolecules with coherent femtosecond X-FEL pulses



Simulated coherent scattering image of a single T4 lysozyme molecule.

R. Neutze *et al.*, Nature **406**, 752 (2000)

Outline

- **Coherence**
- **Scattering with Coherent X-rays**
- **Coherent Diffraction Imaging (CDI, XDM)**
- **X-ray Photon Correlation Spectroscopy (XPCS)**
- **Outlook (X-ray laser sources)**

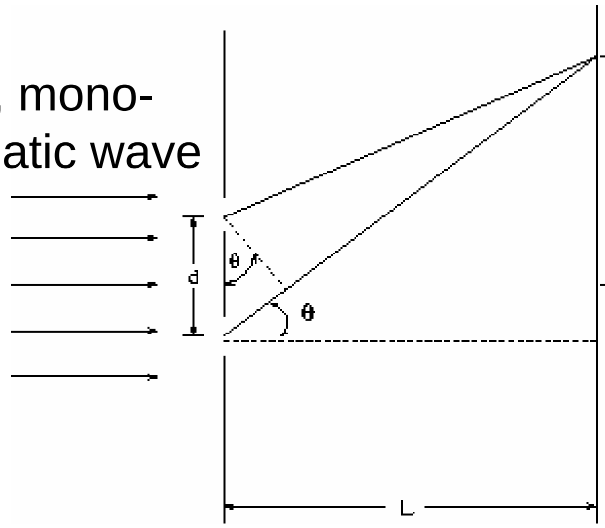
Coherence ?

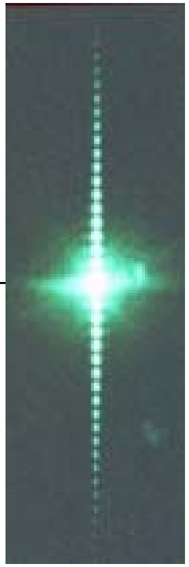
- Quantum mechanics → probability amplitudes
- Optics → interference
- X-ray and neutron scattering

It's all about probability amplitudes and interference !!!

Young's double slit experiment (Thomas Young, 1801)

Plane, mono-chromatic wave





$P=|\Sigma_j \Phi_j|^2$
Φ: probability amplitude
$\Phi_j \sim \exp[-i(\omega t - k l_j)]$
$\omega = ck$, $k = 2\pi/\lambda$, $l_j(L,y)$

$I(y) \rightarrow P(y) \sim \cos^2(\pi y/\Delta)$
$\Delta = \lambda L/d = \lambda/\Theta$
Far field: $\Theta << \lambda/d$

Reasons for losses in visibility/interference/coherence

1) Incoherent superposition of probability amplitudes $P=\Sigma_j|\Phi_j|^2$ (distinguishable alternatives, uncertainty principle)

2) Intensity interference is only observed if event is repeated many times; repetition under non-ideal conditions washes out the visibility

Using a (partially) coherent X-ray source 2) is a major limitation

Non-ideal conditions:

- E_{in}, E_{out}, $\mathbf{k}_{in}$, $\mathbf{k}_{out}$ not well defined in the experiment
- Disorder in the scattering sample
- Limited resolution (temporal and spatial)
- The source is chaotic……..

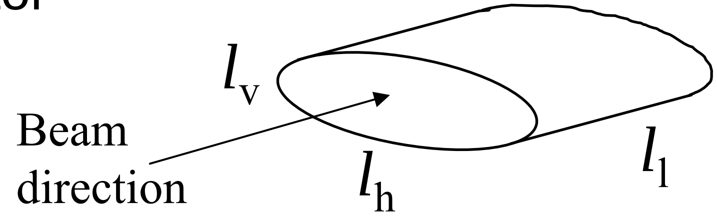
Light sources

- Chaotic sources (spontaneously emitted photons)
  - Light bulb
  - Laboratory X-ray generators
  - Synchrotron and Neutron sources
  - Radioactive nuclei

- One-mode sources (stimulated emission, Glauber light)
  - Unimodal lasers

<u>Decisive parameter</u> : N_C=photons pr. coherence volume

N_C ~10^{-3} for typical ESRF undulator
N_C ~10^{7} for typical optical laser



Coherence volume V_c : $\pi l_h l_v l_l / 4$
horizontal, vertical and longitudinal (temporal) coherence length

Chaotic source (ESRF undulator)

(spontaneous, independent emission in all modes)

Longitudinal (or temporal) coherence

After time $t \gg \tau_0$ the field amplitudes from a chaotic source are no longer correlated due to the wavelength spread

$$\tau_0 \sim 1/\Delta\nu = \lambda^2/(c\Delta\lambda)$$

Longitudinal coherence length $l_l = c\tau_0 \sim \lambda^2/(\Delta\lambda)$ (~1 μm)

Transverse (spatial) coherence

Analogy with Young's double slit experiment :

Transverse coherence length (v,h) : $l_{v,h} \sim \lambda L/d_{v,h}$ (~5-200 μm)

Important source parameters

Brilliance B = photons/sec / [source area × solid angle × bandwidth]

Lateral coherence area $A_t=\pi l_h l_v/4=(\lambda L)^2/(4\pi d_h d_v)$

N_c=photons in V_c ($V_c = A_t \times l_l$)

Coherent solid angle $\Omega_C=A_t/L^2=\lambda^2/(4\pi d_h d_v)= \lambda^2/16A_s$

$$N_c=B \times \tau_0 \times A_s \times \lambda^2/16A_s \times \Delta\nu/\nu = B\,\lambda^3/(16\pi c)$$

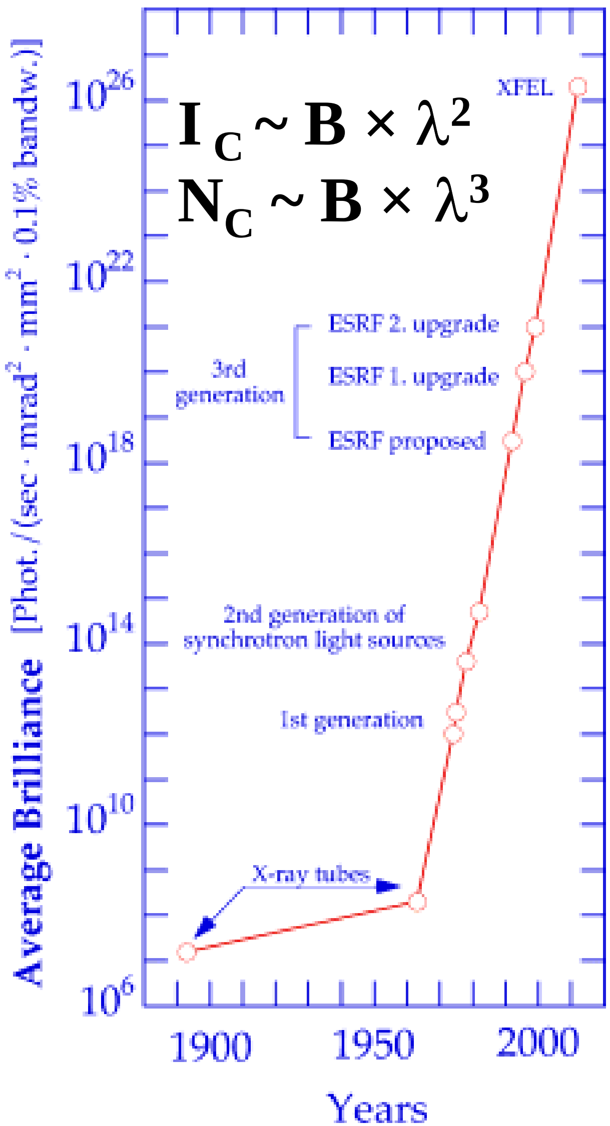
Coherent intensity

$$I_c = (N_c/V_c) \times c \times A_t = B \times (\lambda/4)^2 \times (\Delta\lambda/\lambda) \quad (\sim 10^{10}\ \mathrm{ph/s})$$

Coherent photons scale with λ^3, intensity with λ^2 !

Coherent scattering is Brilliance-hungry !

Coherent X-rays from a partially coherent source



$B=10^{20} \rightarrow \sim 10^{14}$ ph/s/mm^2 (monochromatic)

Low divergence (~10 μrad)

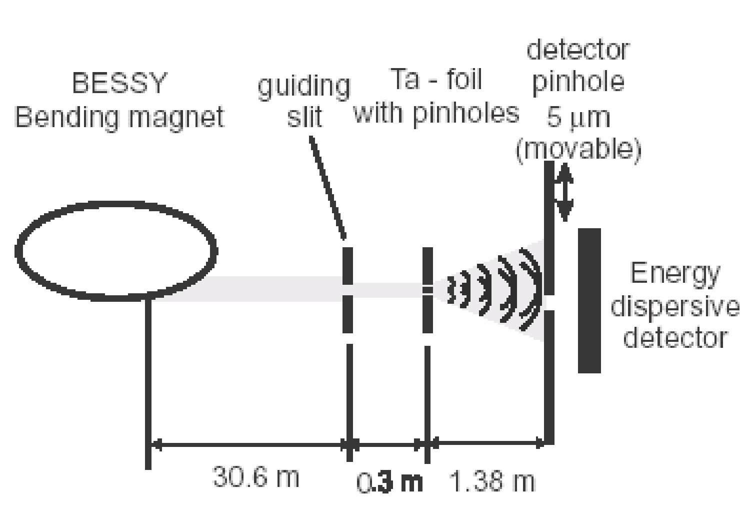
Small source size (~ 25μm)

ESRF beamlines using coherence in experiments:

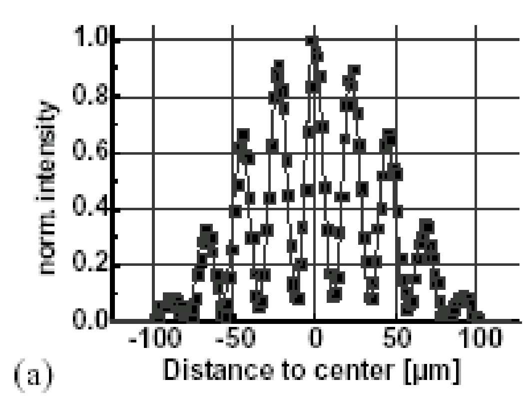
ID01, ID10, ID17, ID19, ID20, ID21, ID22, and more…

Partially coherent light: Coherence lengths

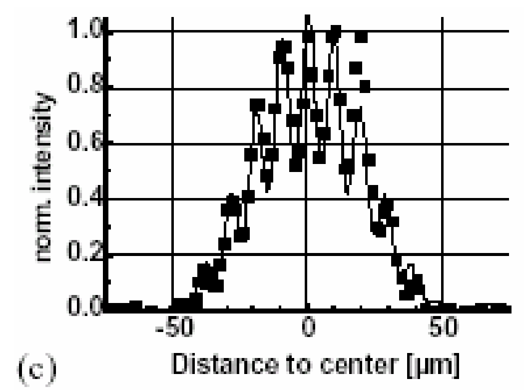
Example: Young's double slit experiment with X-rays



Leitenberger *et al. Physica B* ***336****, 36 (2003)*



λ=2.1Å, d=11μm
Contrast ~ 80%



λ=0.9Å, d=11μm
Contrast ~ 30%

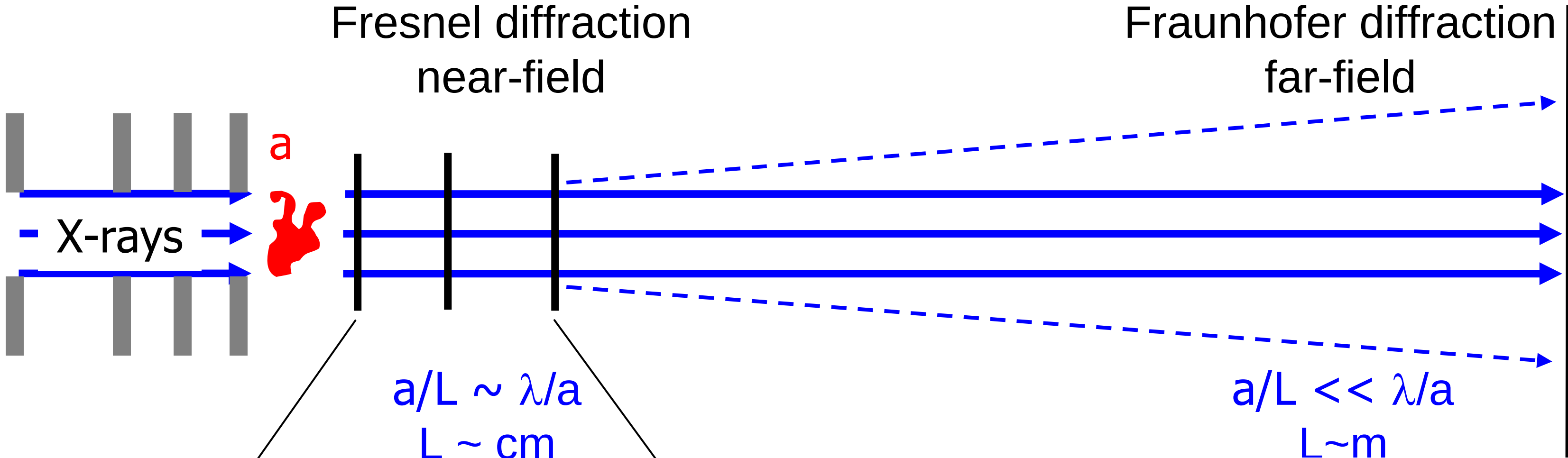
$\Delta y = \lambda L/d$

Transverse coherence length (v,h) : $l_{v,h} \sim \lambda L/d_{v,h}$ (~5-200 μm)

Longitudinal coherence length $l_l = c\tau_0 \sim \lambda^2/\Delta\lambda$ (~1μm)

Contrast $\beta \approx$ (coherence volume)/(scattering volume)

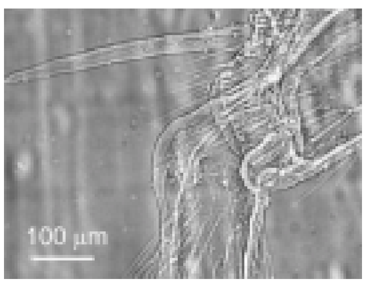
Different reigimes of coherent X-ray imaging/diffraction



Absorption

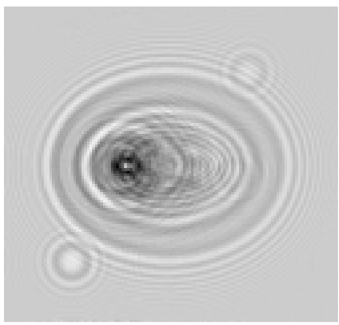
Röntgen (1895)

Phase contrast



Koch *et al.* (1998)

In-line holography



Zhang (2003)

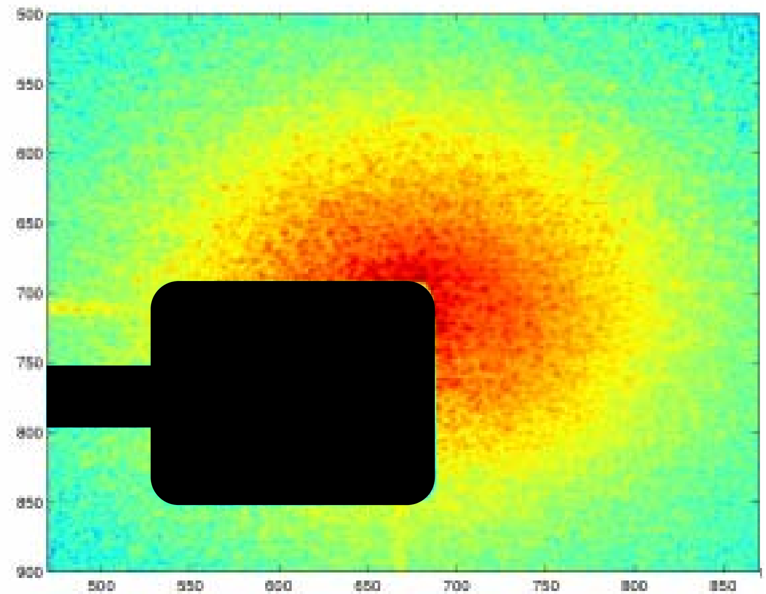


Image reconstruction in the different regimes

- **Absorption regime**
  Easy reconstruction based on attenuation
  3D tomographic reconstruction, inverse Radon transformation
- **Phase contrast regime**
  Edge enhanced contrast
  Transport-of-intensity (TIE) equation
  Holotomographic reconstruction (Talbot effect)
- **In-line holographic regime**
  Holographic reconstuction
  Twin images
- **Diffraction imaging**
  Phase retrieval by iterative procedure (phase problem)
  Real space ←→reciprocal space, application of constraints
  Requires oversampled diffraction pattern

X-ray scattering

X-rays are scattered by electrons

Scattering lengths

Free electron: r_0 (Thomson radius r_0=2.82e-5 Å)

Atom: $f(\mathbf{Q},E)\, r_0$ (atomic form factor $f(\mathbf{Q},E)$)

$$f(\mathbf{Q},E)=f^0(\mathbf{Q})+f'(E)+if''(E)$$

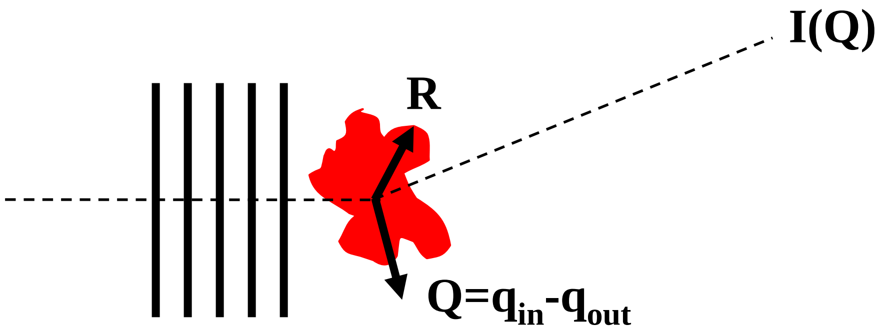
$$f^0(\mathbf{Q})\rightarrow Z \text{ for } Q\rightarrow 0$$

$$f^0(\mathbf{Q})=FT\{\rho(\mathbf{r})\}$$

Connection with refraction: $n = 1-\delta+i\beta = f(\mathbf{Q},E)\rho_a r_0 \lambda^2/2\pi$

More atoms: $F(Q)=\sum f_j(\mathbf{Q},E)\exp(i\mathbf{Q}\cdot\mathbf{r}_j)$

The phase problem



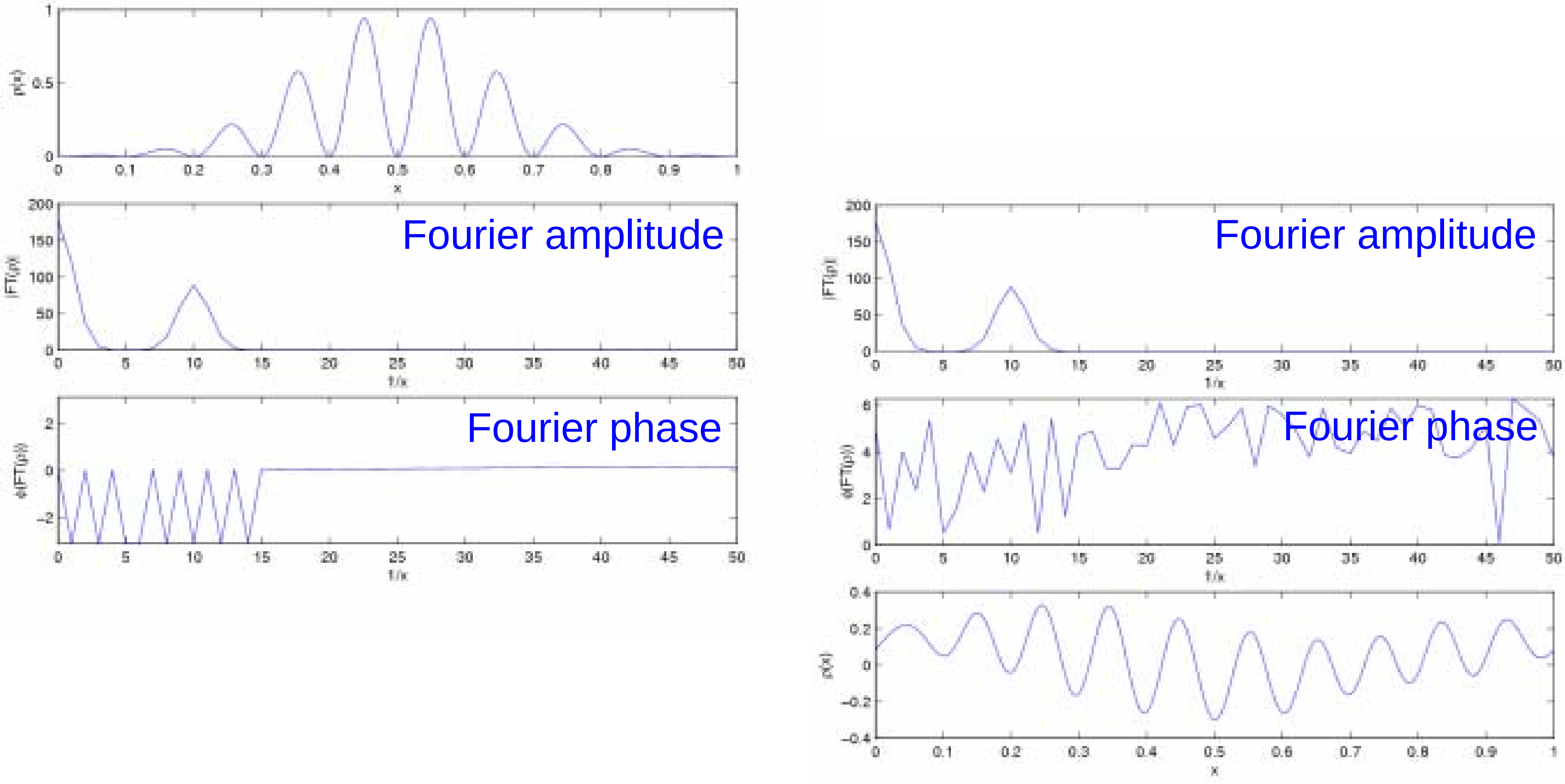
$$E(\mathbf{Q}) \sim \int \rho(\mathbf{R}) \exp[i\mathbf{Q} \cdot \mathbf{R}] d\mathbf{R}$$

$$I(\mathbf{Q}) = E(\mathbf{Q})E^{*}(\mathbf{Q}) = |E(\mathbf{Q})|^2$$

ρ(**R**) Real space ←←← →→→ Reciprocal space E(**Q**)

FT^{-1} FT

Phase matters



Iterative methods in diffraction imaging

Algorithms:

Gerchberg & Saxton (1972)

HIO (Fienup, Miao)

Difference map (Elser)

Shrinkwrap (Marchesini *et al.*)

Curved wavefront approaches (Nugent)

PIE, Faulkner & Rodenburg (2004)

Methodology:

Support, beam vs object (Stadler, Zuo)

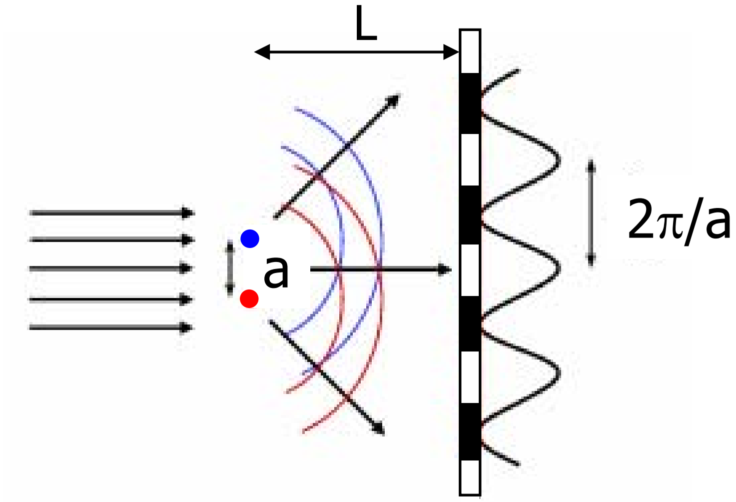
Reference wave (Eisebitt, Nugent)

Beamstops (Chapman, Jacobsen, Zuo)

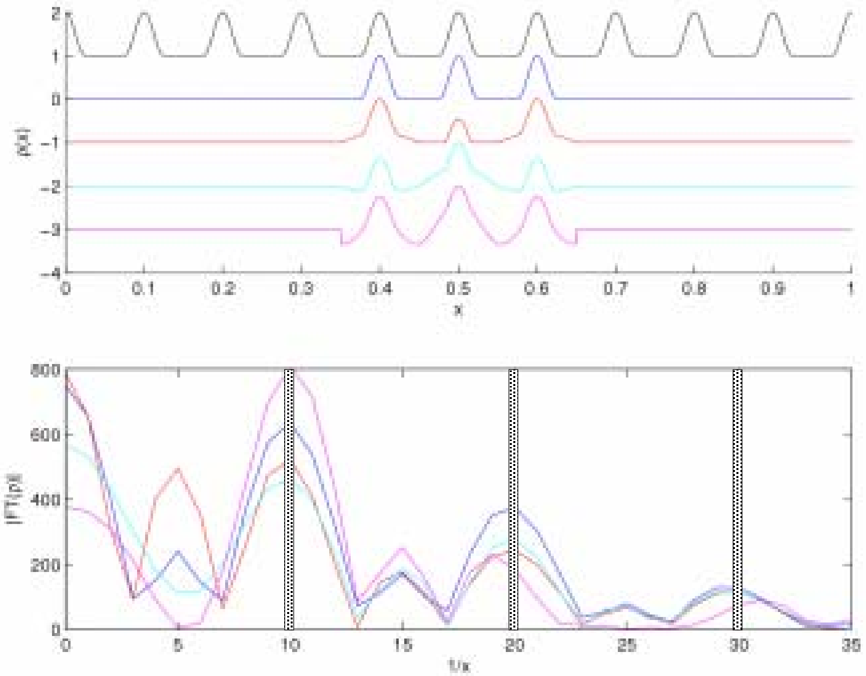
Detectors (Nishino, Chapman)

Radiation Damage (Jacobsen, Chapman)
(electrons or X-rays, imaging or diffraction ?)

Correct sampling



Finite size effects



Sampling at frequency $2\pi/a$ is
NOT ENOUGH to resolve fringes

Minimum oversampling is 2
(regardless of dimension)

$\Delta Q<2\pi/a * (1/2)$ (1D case
$\Delta Q<2\pi/a * (1/2)^{1/2}$ (2D case
$\Delta Q<2\pi/a * (1/2)^{1/3}$ (3D case)

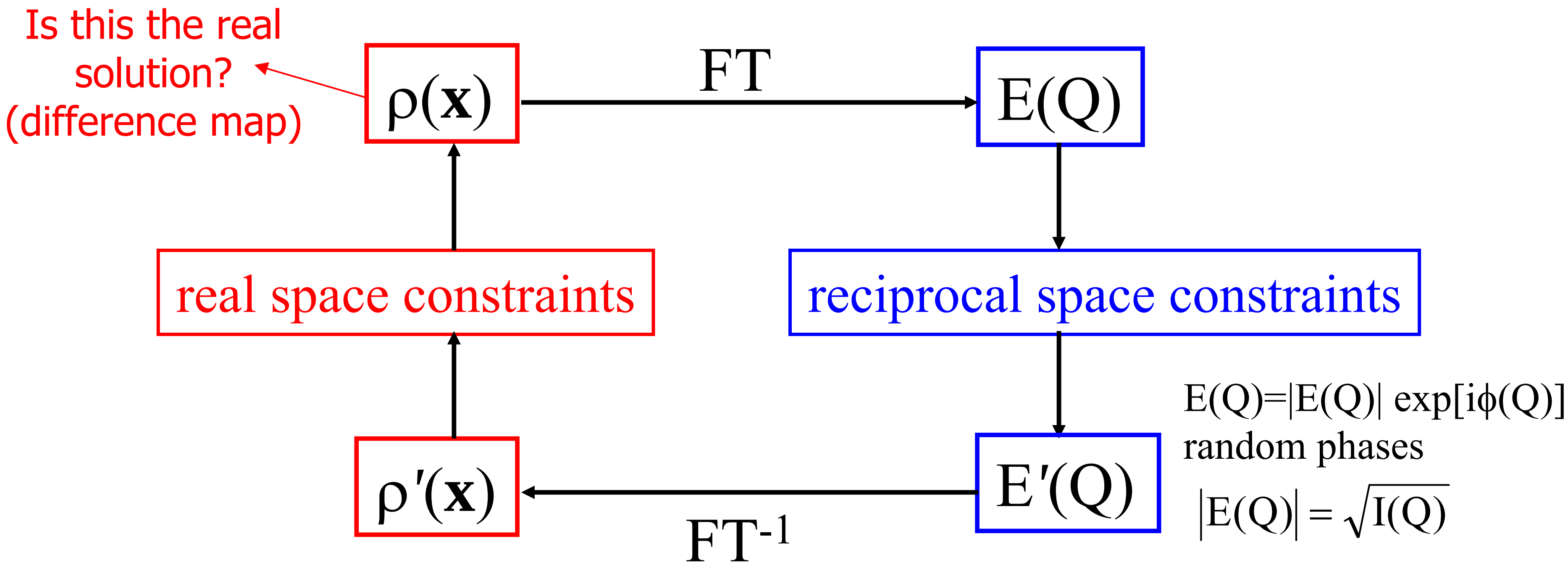
Pixel size $X < \lambda L/2a$ (1D case)

ESRF

Iterative phase retrieval algorithm

J.R. Fienup, Appl. Opt. **21**, 2758 (1982)

R. W. Gerchberg and W. O. Saxton, Optik **35,** 237 (1972)



Real space constraints

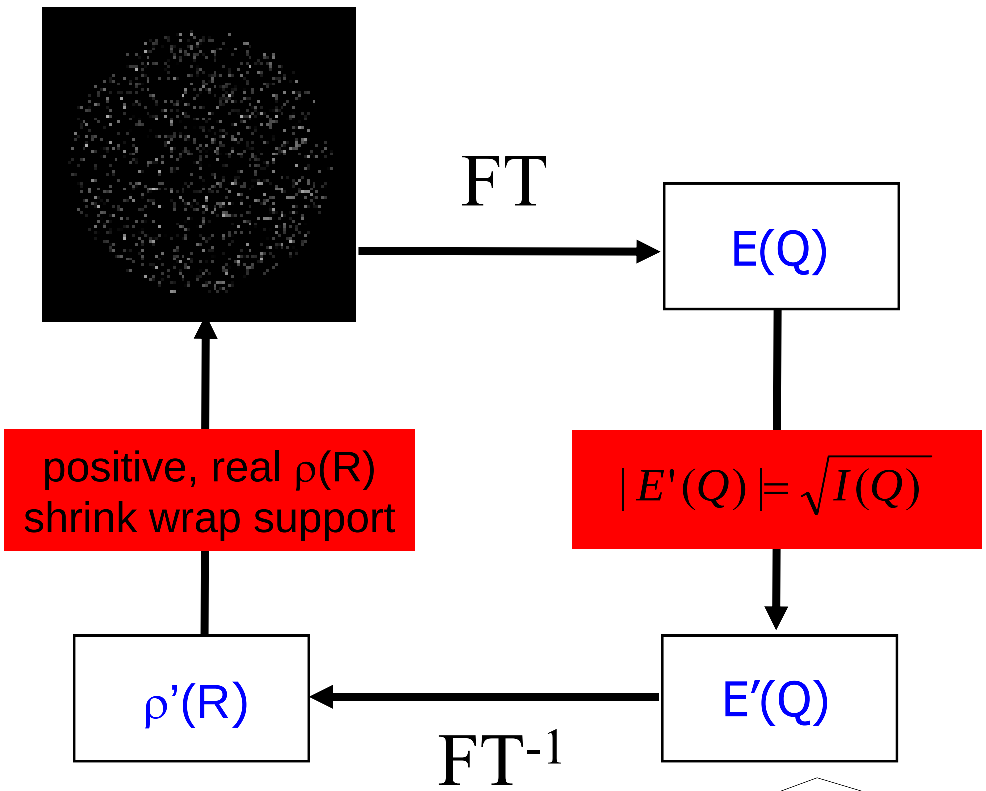
- Finite support (acf, shrink wrap)
- Real ρ (centro-symmetric I(Q)) ?
- positive ρ (sample thickness) ?

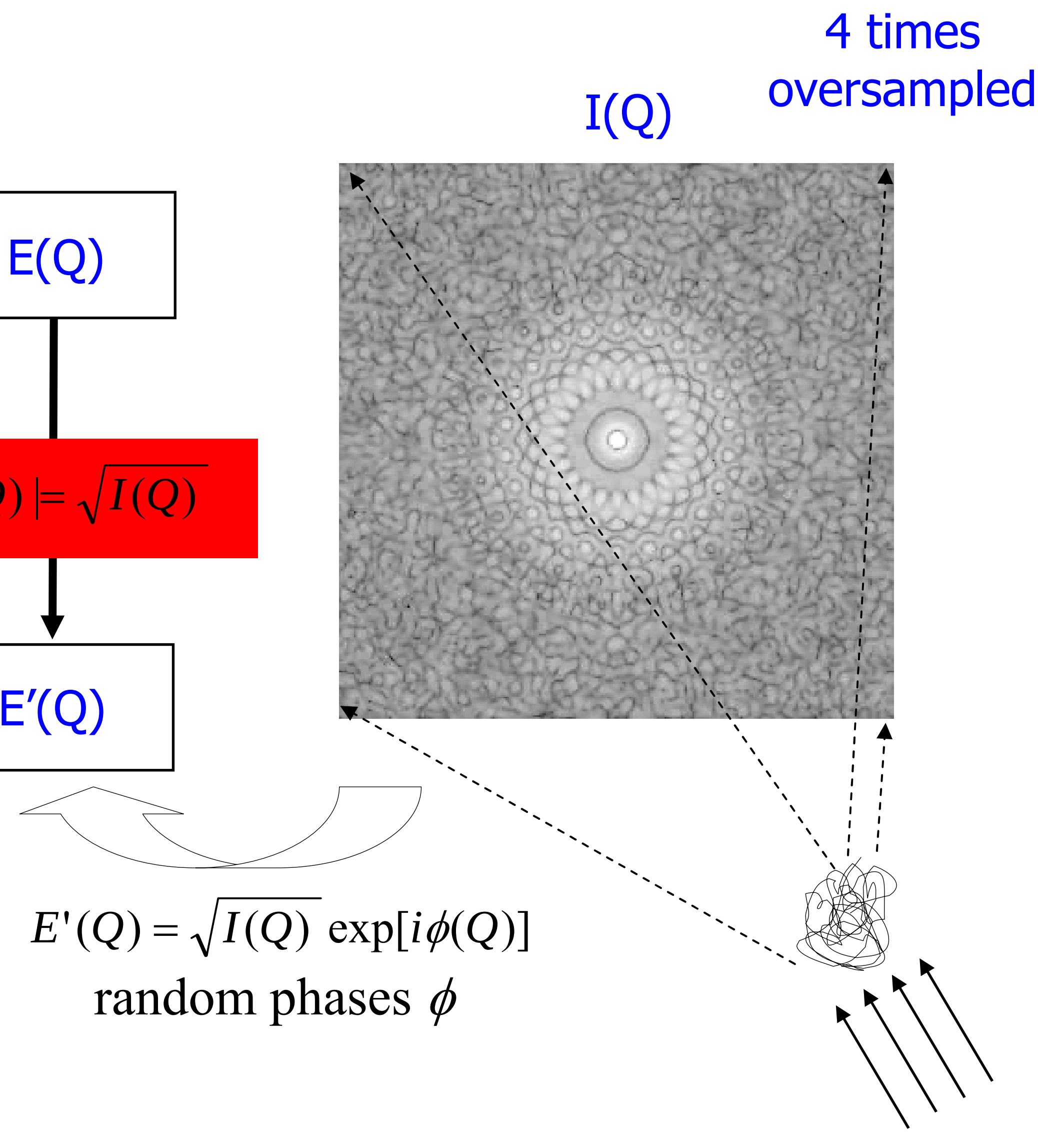
Reciprocal space constraints

$$|E(q)| \rightarrow \sqrt{I(q)}$$

R. Millane *et al*., J. Opt. Soc. Am. **A14**, 568 (1997)

Iterative phase retrieval algorithm

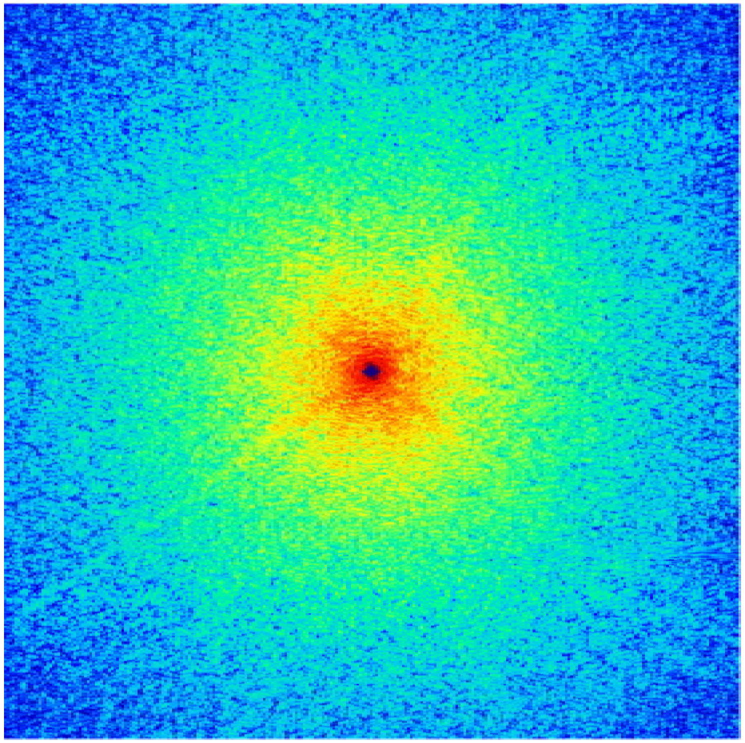


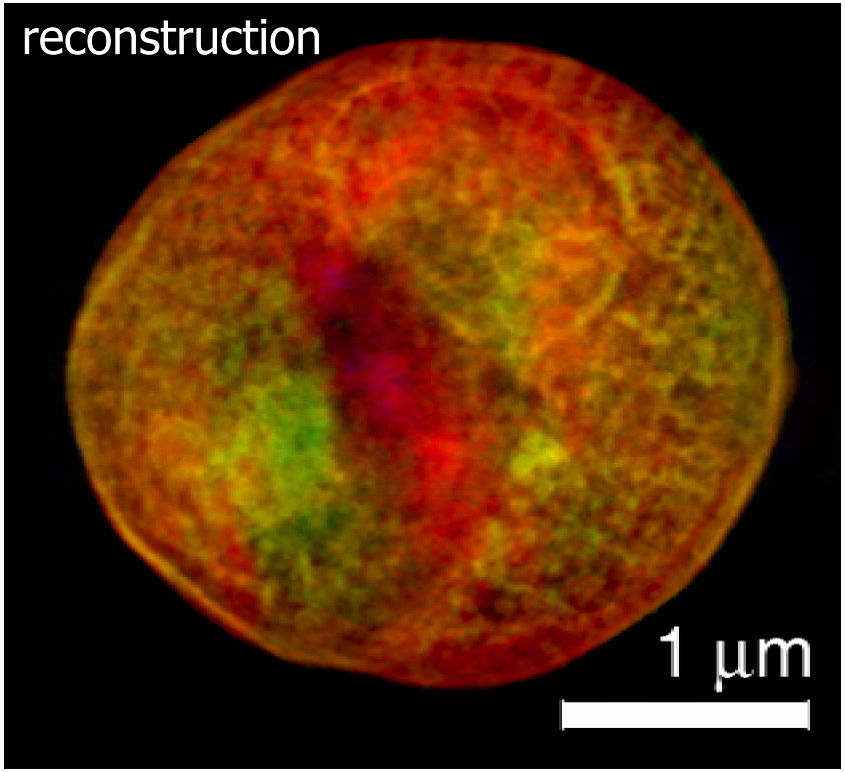


Reconstruction by
E. Lima, ESRF

Coherent scattering from a yeast cell

Speckle pattern, λ=16.5Å (ALS)



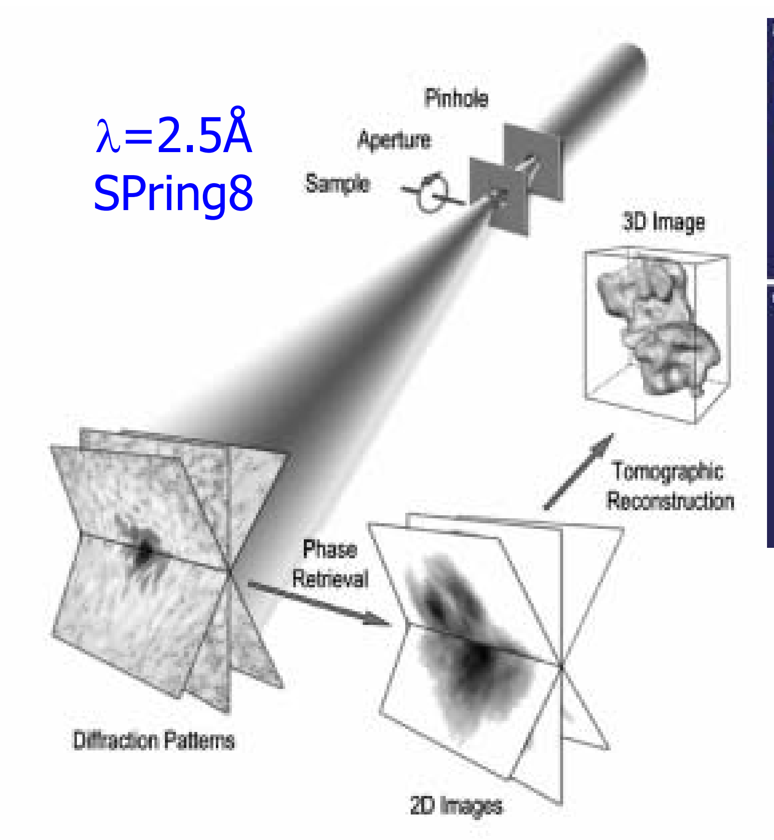


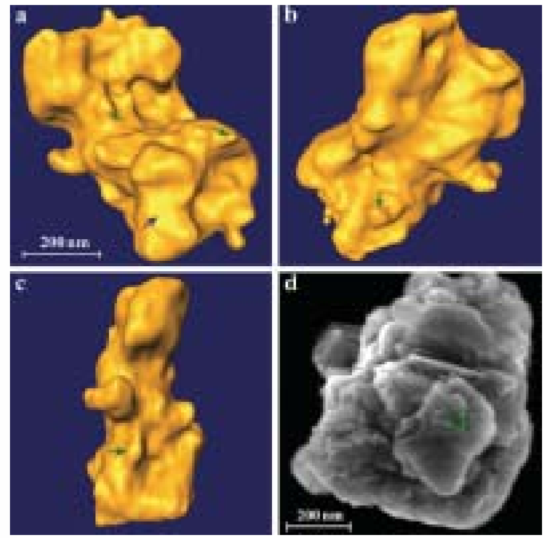
No shrink warp, "hand drawn" support
Averaging iterates (Elser & Thibault, Cornell)
Difference map algorithm
Resolution ~ 30nm

D. Shapiro *et al*, PNAS **102**, 15343 (2005)

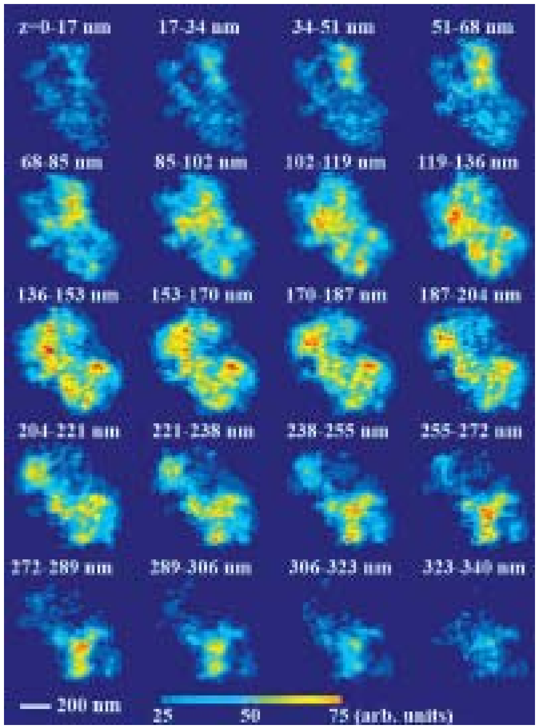
Tomographic reconstruction of core-shell structures

λ=2.5Å
SPring8





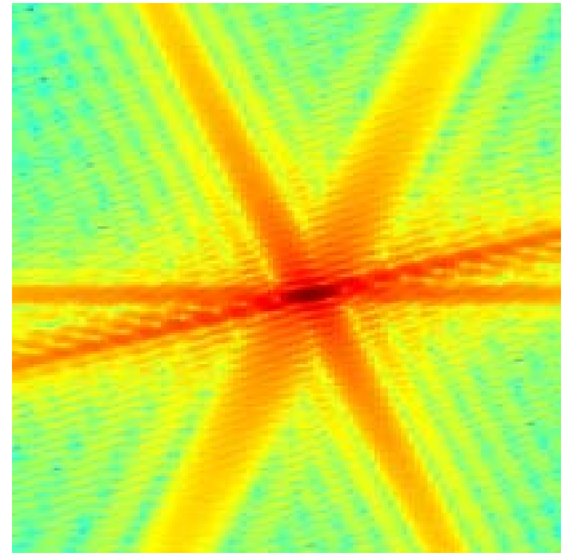
~ 7nm resolution

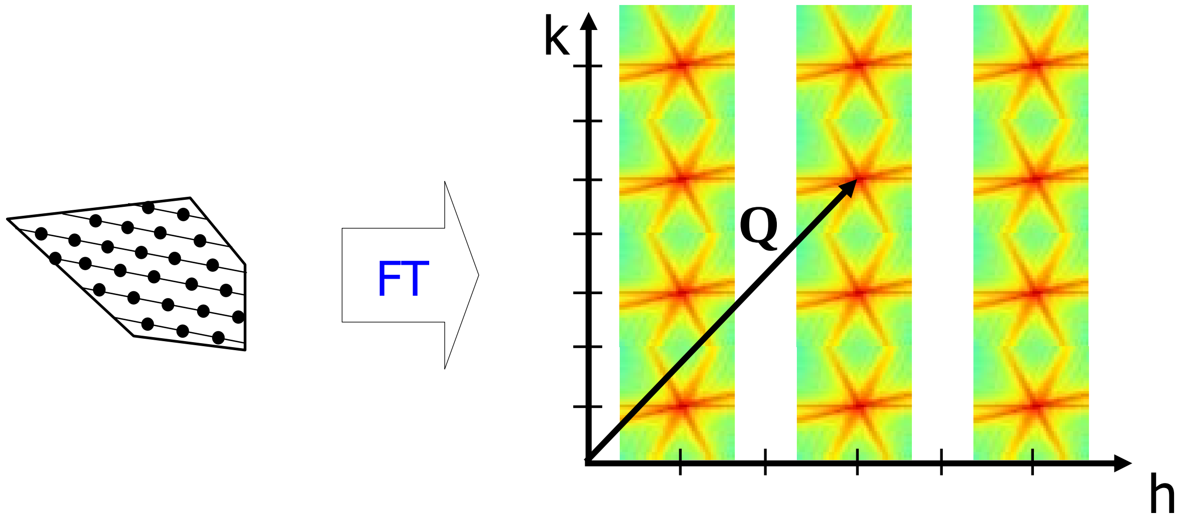


J. Miao *et al*, PRL **97**, 215503 (2006)

GaN (core) – Ga2O3 (shell) structure clearly visible

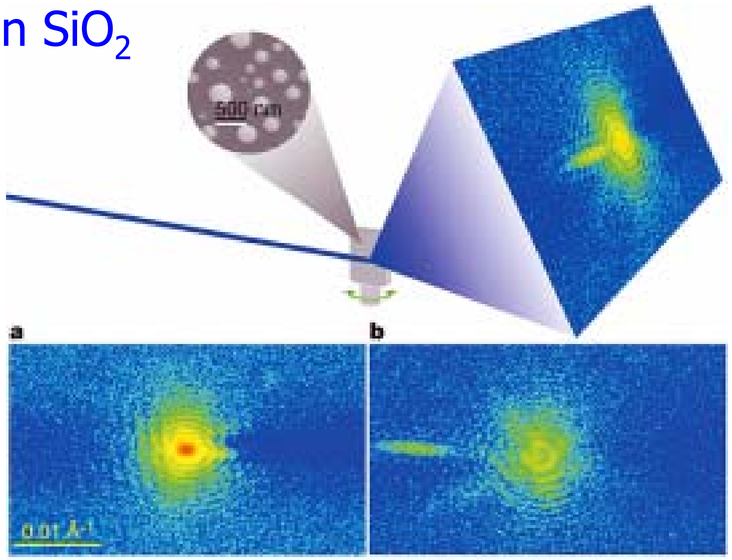
Diffraction imaging of small crystals



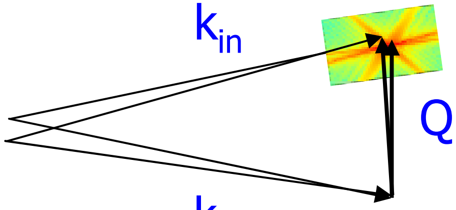


Diffraction imaging of small crystals

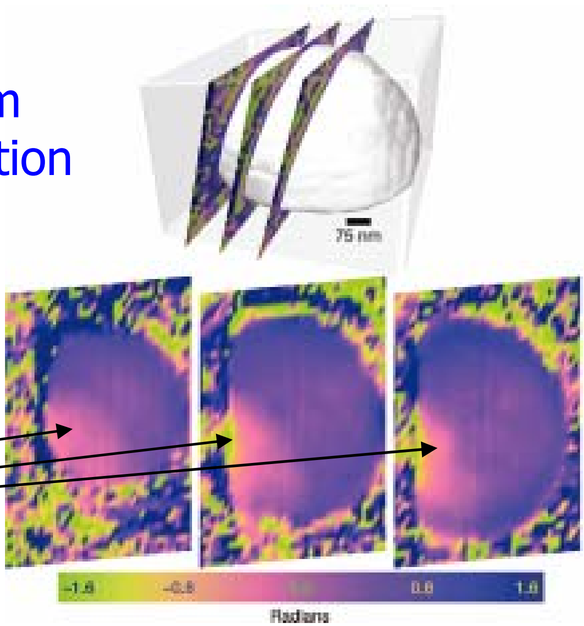
Pb on SiO_2



$Q=k_{in}-k_{out}$



40nm resolution



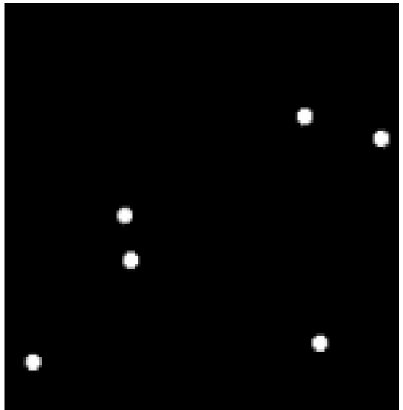
Rocking through the (111) reflection of a Pb nanocrystal

Phase bulge indicates strain inside the nano-crystals arising from the contact forces at the interface

M. A. Pfeifer *et al*, Nature **442**, 63 (2006)

What about dynamics?

real space



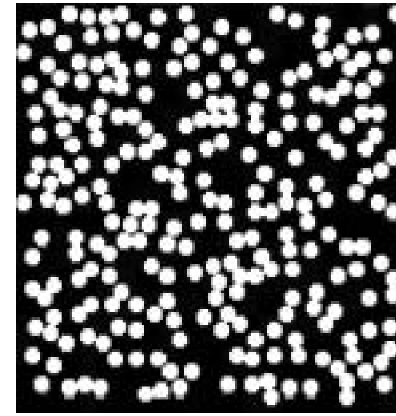
dilute

$<\Delta x^2> = 2D_0 t$

Brownian motion

concen-trated

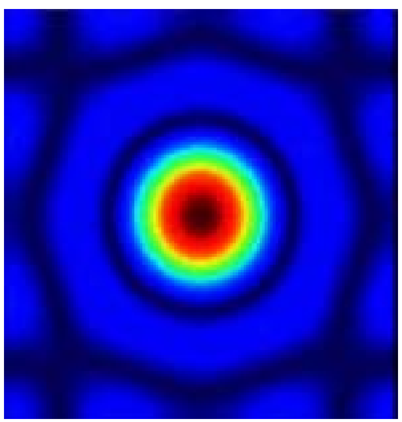
$<\Delta x^2> \neq 2D_0 t$

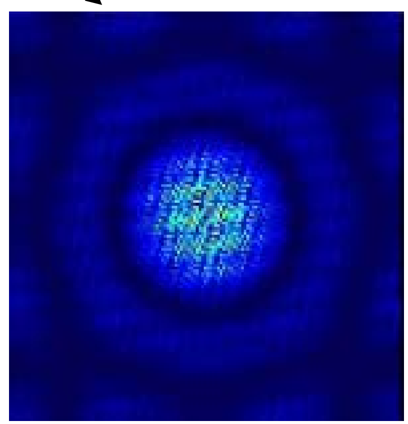


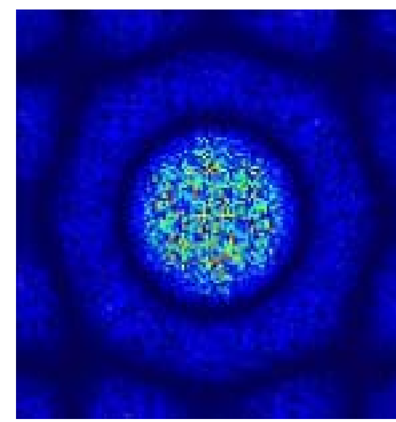
reciprocal space

in-coherent

coherent







$<I(Q)>$

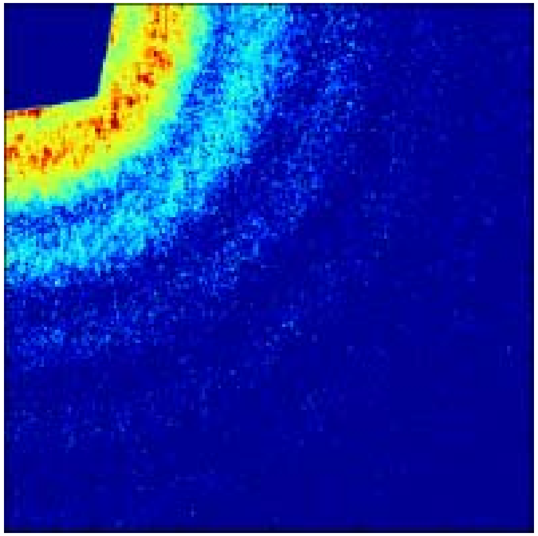
$I(Q,t)$ → dynamic structure factor

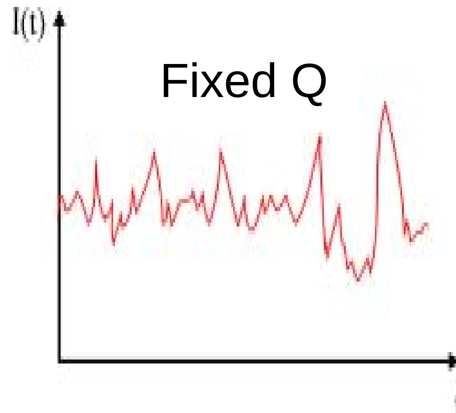
$\Gamma = D_0 Q^2$ (brownian)

$I(Q,t)$

$\Gamma = D(Q) Q^2$

X-ray Photon Correlation Spectroscopy

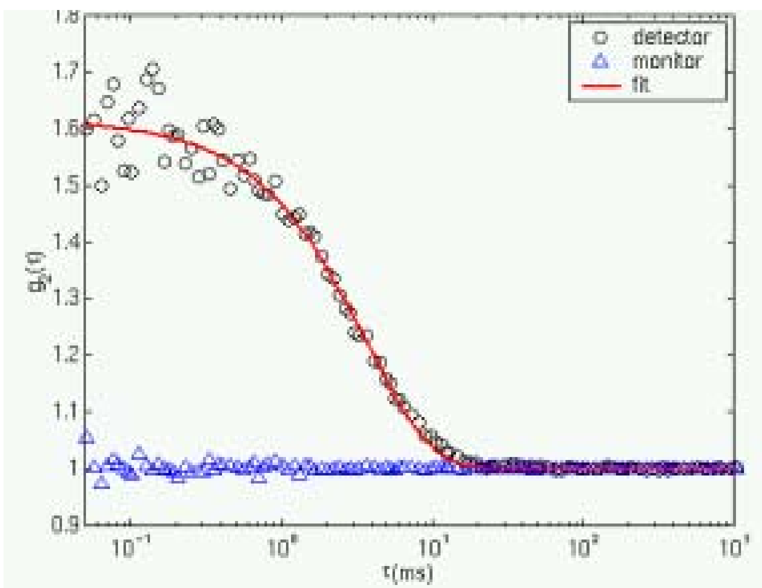




Fixed Q

Temporal auto-correlation function

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

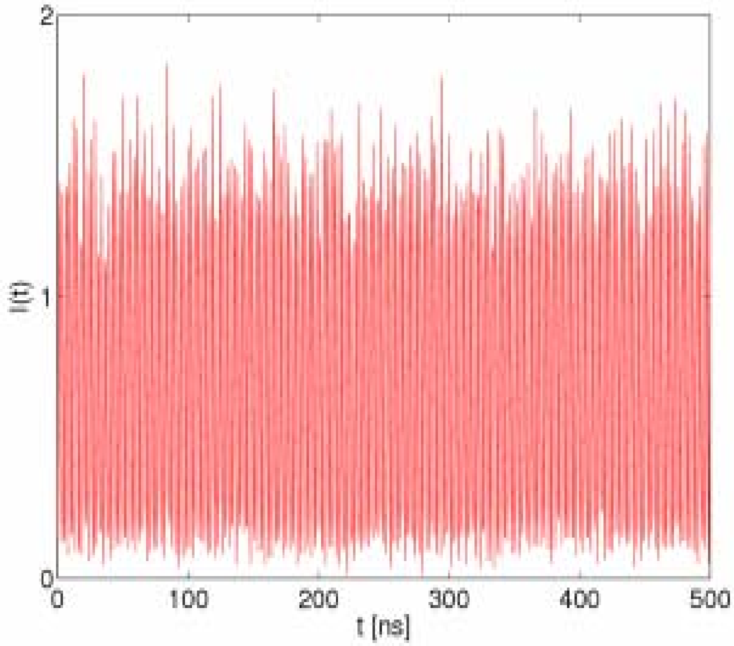


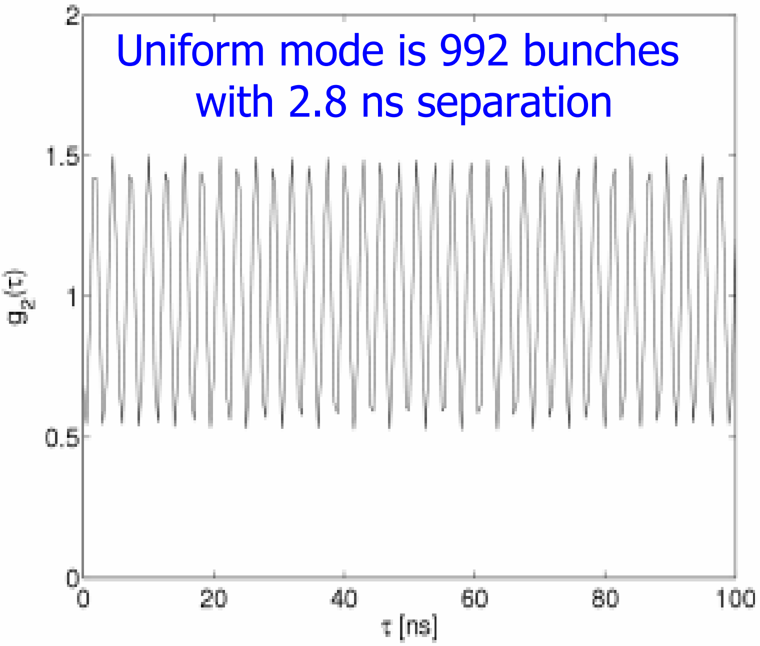
$$g(\tau) \sim \exp(-\Gamma \tau)$$

Relaxation rate Γ vs Q

Intensity fluctuations from the ESRF storage ring

~850 m circumference, 6 GeV, 32 straight sections



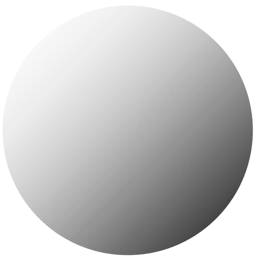


Data taken with APD detector and 2GHz scaler board

XPCS experiment: nanoparticle dynamics in a glass

Samples

Nano-spheres in a glass forming solvent

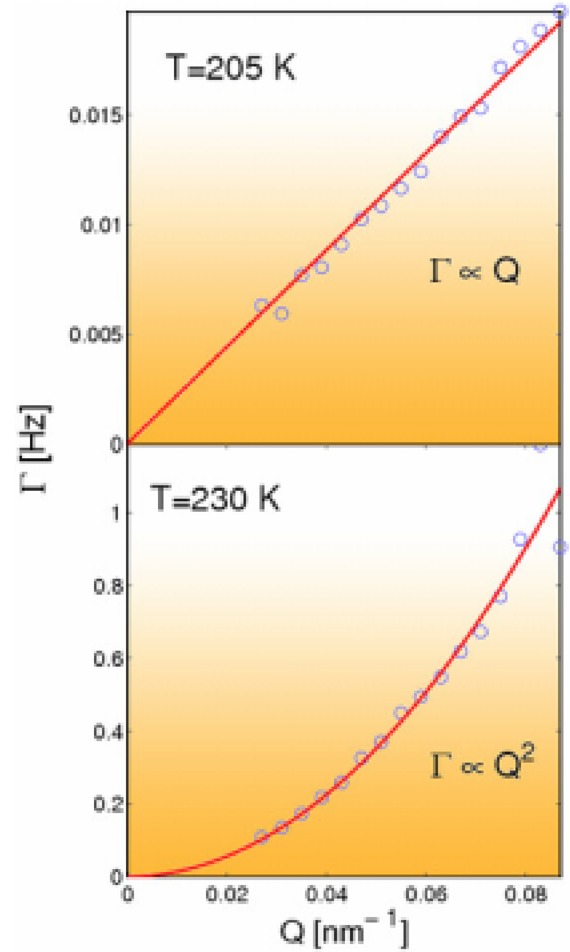


Silica, R=250nm, ϕ~1% (vol.)

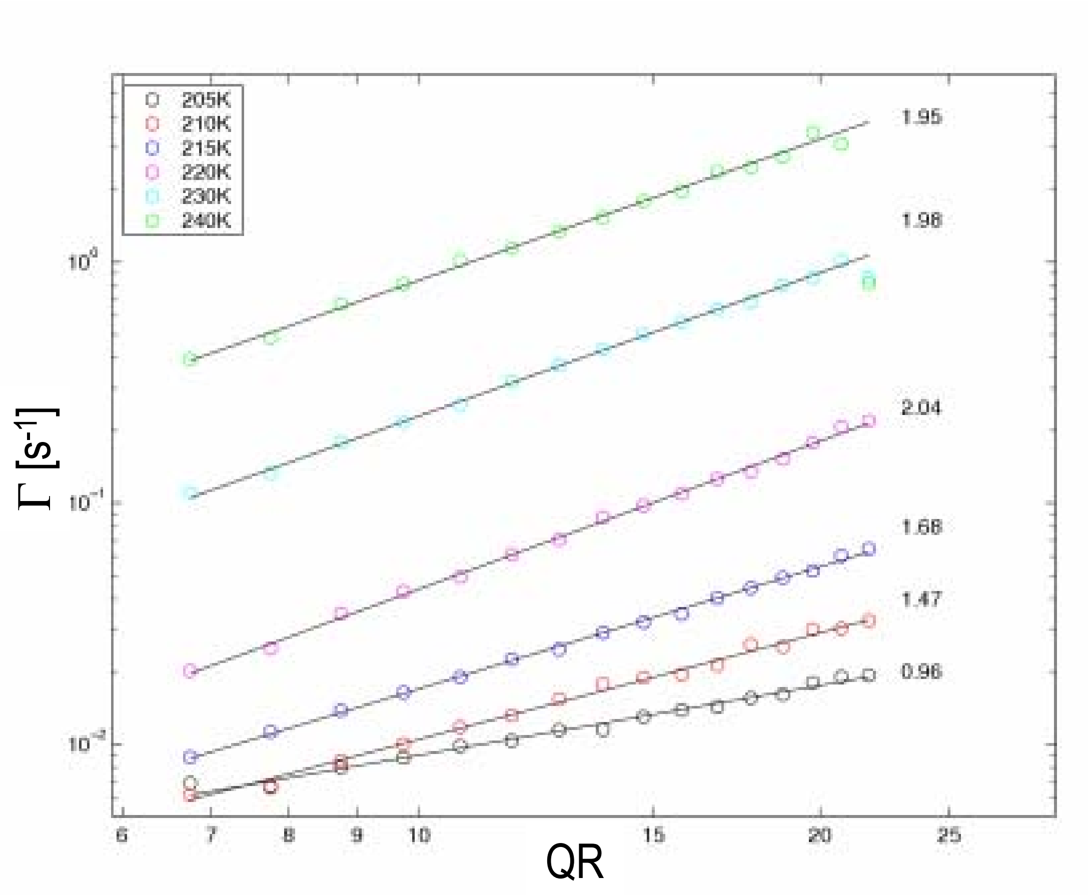
Silica, R=16nm, ϕ~20% (vol.)

Solvent: Propanediol (T_G ~ 170K)
Liquid at elevated temp. (η ~ 0.1 Pa·s @ 290K)

R=250nm, ϕ~1%
XPCS data



Results, R=250nm, ϕ~1%

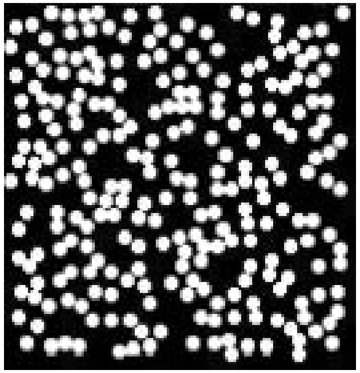


n=2
(Brownian motion)

n→1
(hyperdiffusion?)

Higher concentrations….

Higher concentration → deviations from Brownian motion

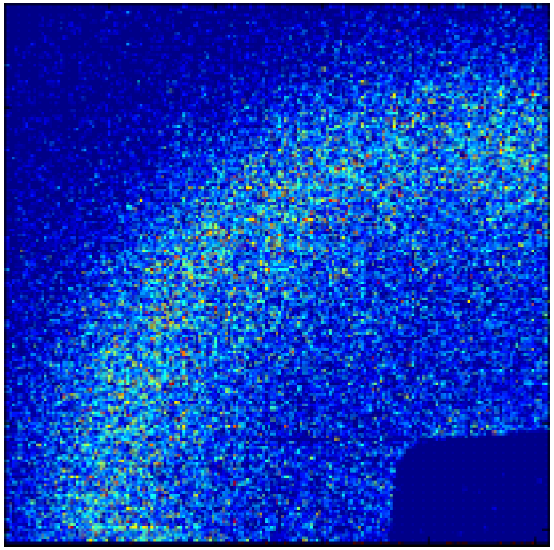


$S(Q=Q_{max}) > 1$ → at $Q=Q_{max}$ the dynamics is slower than simple Brownian motion ("de Gennes narrowing")

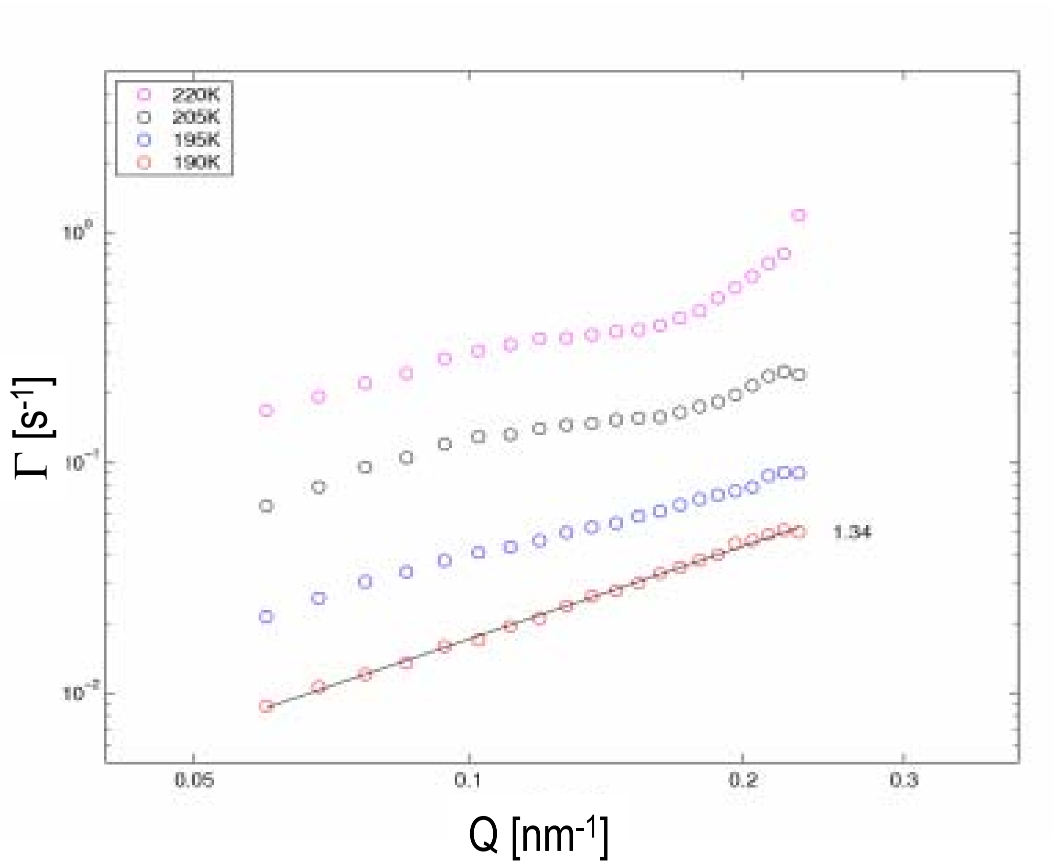
At higher concentrations inter-particle interactions start playing a role

Smaller particles → different lengthscales probed → dynamics is faster

R=16nm, ϕ~20%
ring-of-scattering
at Q_{max}~ 0.2 nm^{-1}



Results, R=16nm, ϕ~20%



"de Gennes narrowing" disappears for $T \rightarrow T_G$ (collective motion)

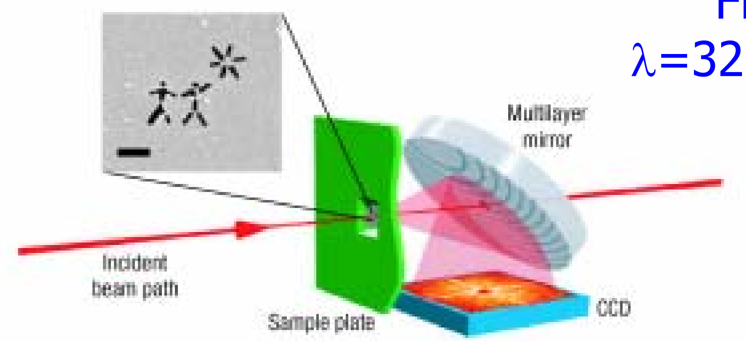
Conclusion:
Intermittent dynamics (n=1) driven by stress relaxations of the solvent become important much before T_G

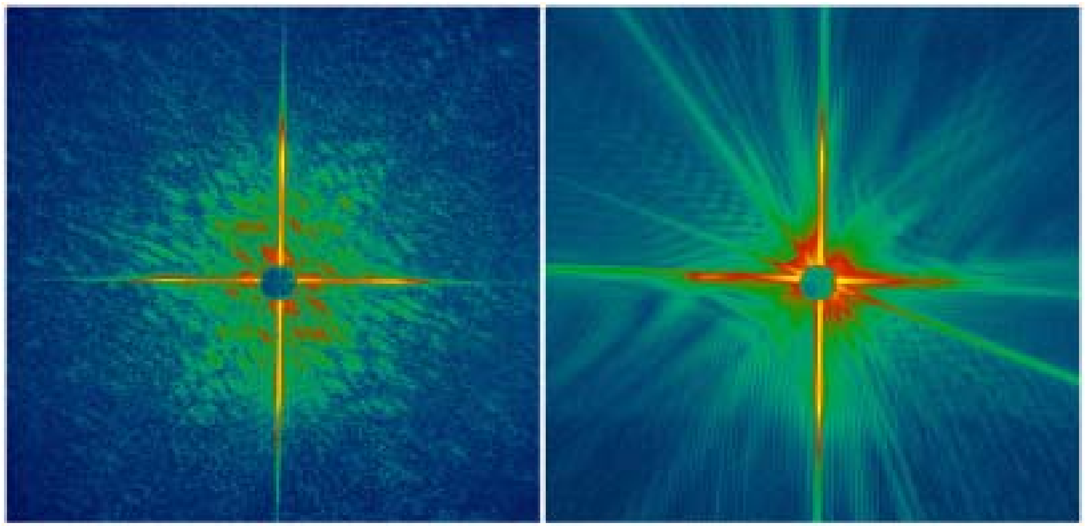
Localized stress relaxations seem to lead to collective dynamics

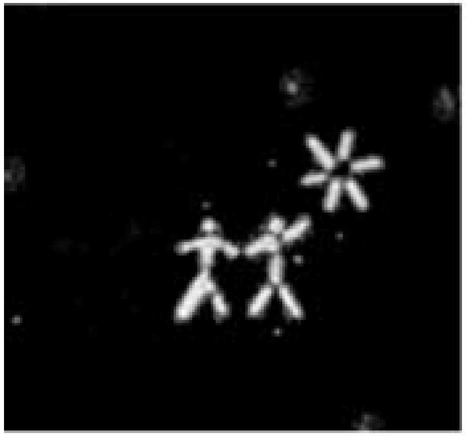
http://www.esrf.fr/news/spotlight/spotlight39/spotlight39xpcs/

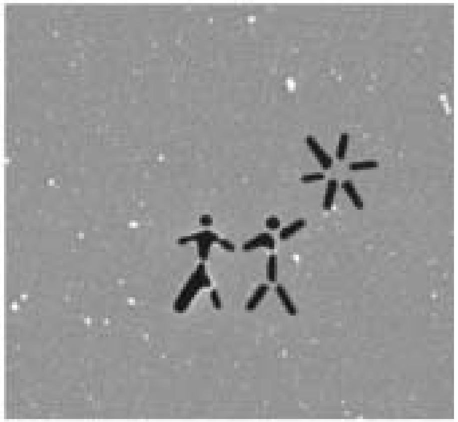
Time resolved reconstruction
(merging Coherent Diffraction Imaging and XPCS)

FLASH VUV-Laser (DESY, Hamburg)
λ=32nm, 25fs pulse duration, 10^{12} ph/pulse









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

Outlook

The future has already started

Pulsed X-ray lasers

- First 25 fs CDI experiment at the FLASH VUV laser
- LCLS (Stanford) & European XFEL coming online (~2009-2013)

Continuous coherent X-ray sources

- Energy recovery linac (Cornell, APS,??)

CDI/XPCS at ESRF

- XPCS activities continue, probably with more beamlines involved
- CDI is taking off (ID01, ID10 + more?)
- Scientific cases for CDI (SAXS/WAXS)
- Coherence is a "hot" topic in the forthcoming "purple book"