July 13, 2011, @ Tsukuba

Report of XDL Workshop 6

- Frontier Science with X-ray Correlation Spectroscopies using Continuous Sources -

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The University of Tokyo

Purpose of WS 6

XPCS with Future Continuous Light Sources ?

Energy Recovery Linac, Ultimate Storage Ring

Broadening awareness of XPCS

⟶ wider community

What's XPCS

X-ray Photon Correlation Spectroscopy

Time-resolved Measurement of
Coherent Scattering

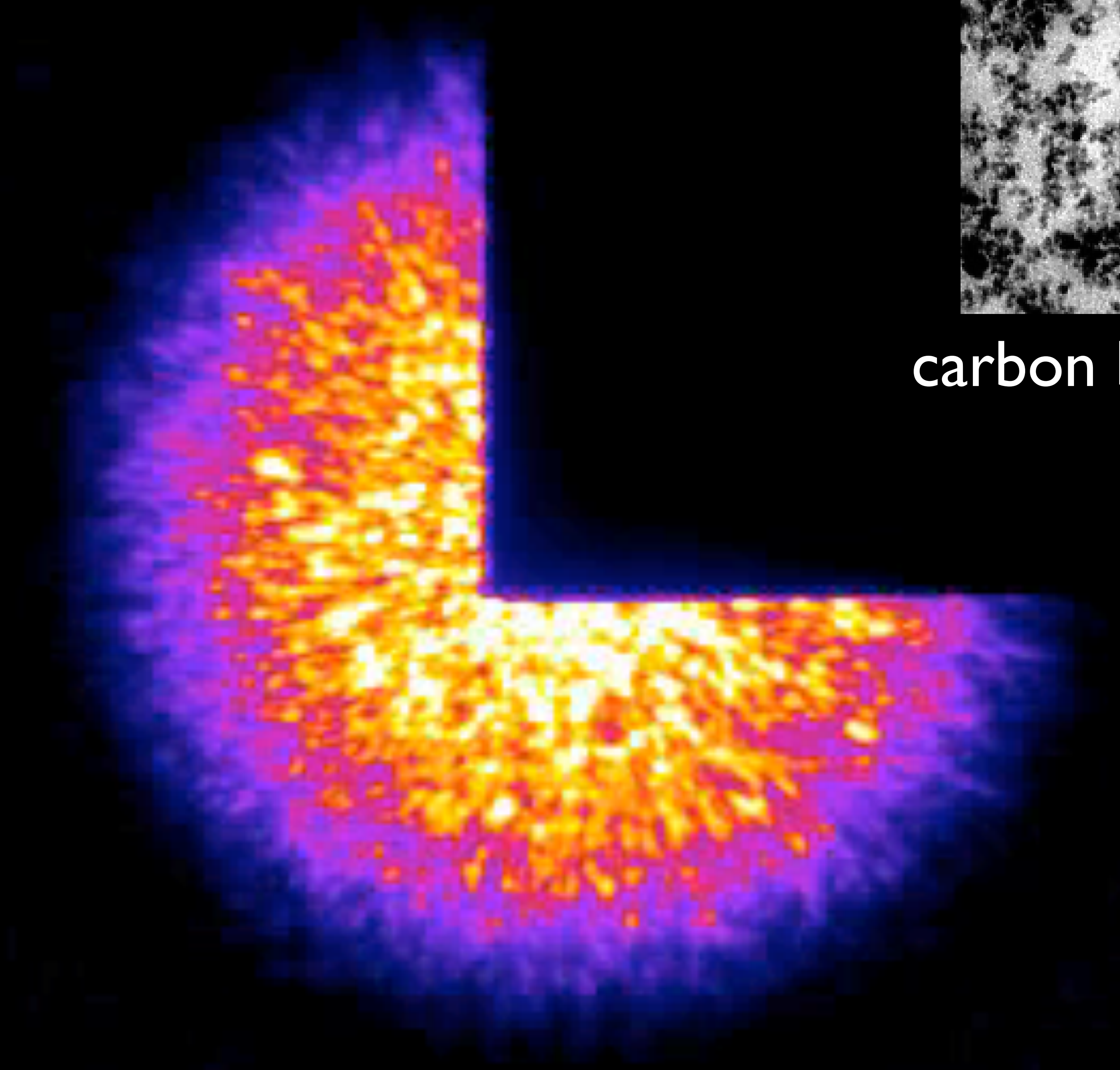
Coherent Diffraction Imaging (CDI)

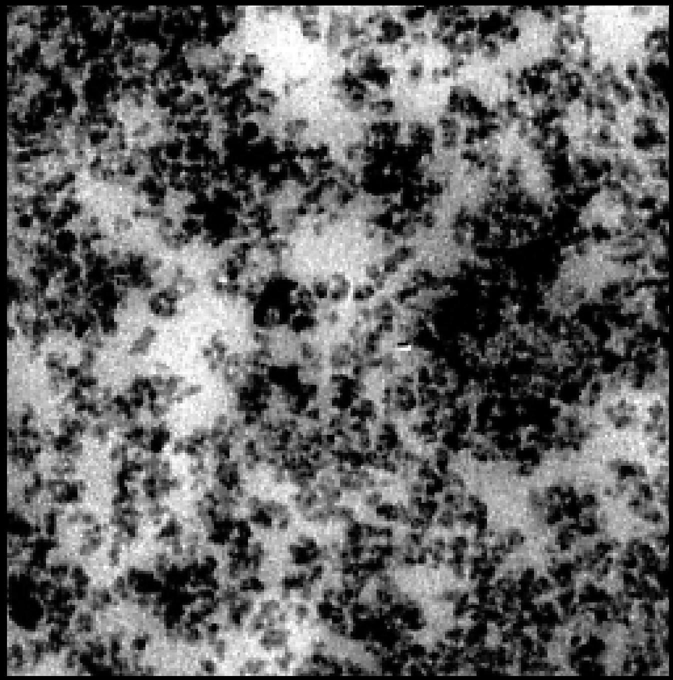
Speckle image: $I(q)$ $\xrightarrow{\text{Fourier Trans.}}$ $\rho(r)$ with Phase info.

XPCS

Speckle images: $I(q, t)$ $\xrightarrow{\text{Time Correlation}}$ Time Corr. of $\rho(r)$

コヒーレントなX線 → スペックル散乱像



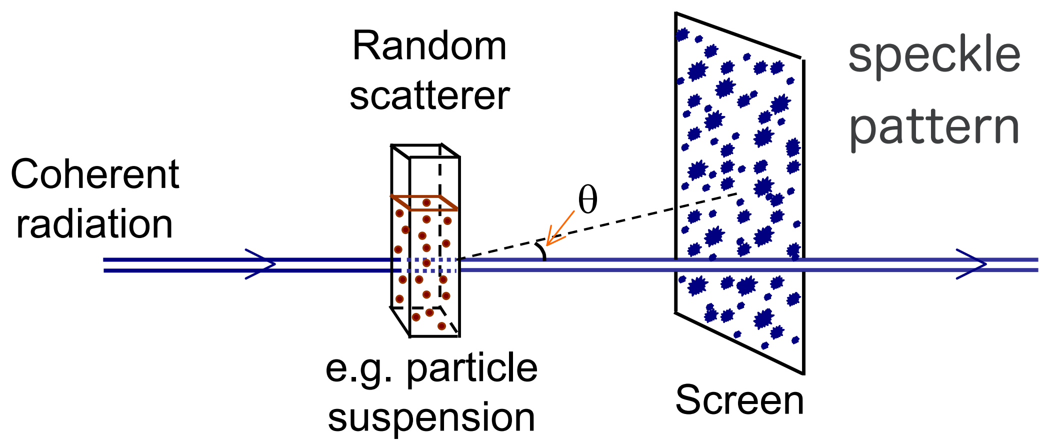


carbon black in rubber

スペックルの揺らぎ ⟶ ゴム中でのナノ粒子の揺らぎ

What is XPCS ?

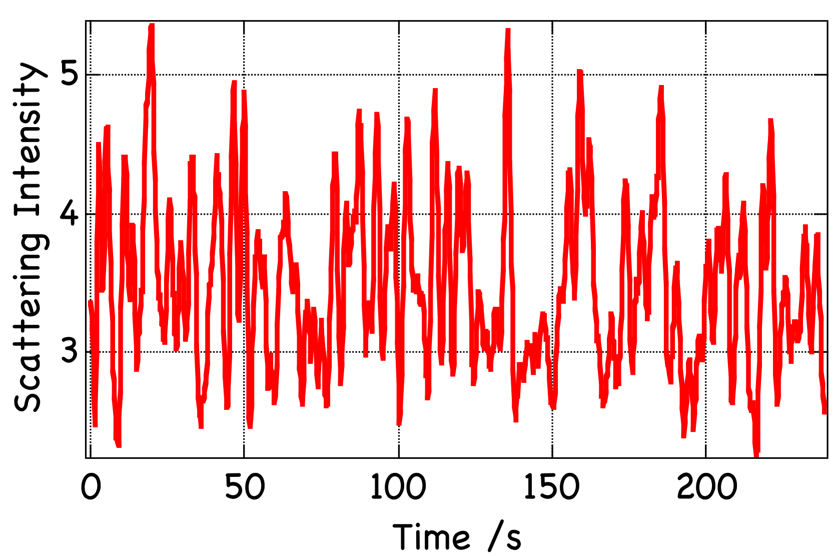
散乱X線強度の揺らぎ → 内部構造の揺らぎ



強度の自己相関関数

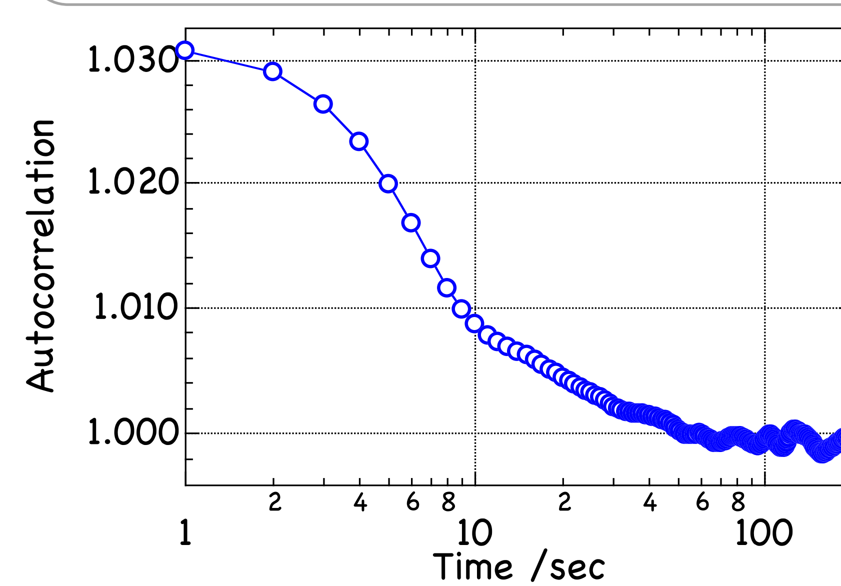
$$g^{(2)}(\boldsymbol{q}, \tau) = \frac{\langle I(\boldsymbol{q}, 0) I^*(\boldsymbol{q}, \tau) \rangle_E}{\langle I(\boldsymbol{q}) \rangle_E^2}$$

Fluctuation of intensity



Autocorrelation →

relaxation time in system



$$G(\boldsymbol{r}, t)$$

van Hove Correlation fn.

Fourier trans. (space)

Fourier trans. (time)

$$f(\boldsymbol{q}, t)$$

Intermediate Correlation fn.

$$S(\boldsymbol{q}, \omega)$$

Dynamic Stricture Factor

XPCS に必要とされる実験条件

- **（部分）コヒーレントなＸ線**
  - ➡ 大強度なＸ線からコヒーレントな部分をピンホールで抽出して用いる。
    空間コヒーレント長 ＞ ビームサイズ
- **高空間分解能・高時間分解能なＸ線検出器**
  - スペックルサイズよりも高い空間分解能
  - 系のダイナミクスよりも速い時間分解能
- **適切な厚みの試料**
  - ➡ 試料への制約条件を明確にしてその範囲内で実験する

Small-Angle X-ray Scattering Using Coherent Undulator Radiation at the ESRF

D. L. Abernathy,[a]* G. Grübel,[a] S. Brauer,[b] I. McNulty,[b] G. B. Stephenson,[b] S. G. J. Mochrie,[c] A. R. Sandy,[c] N. Mulders[d] and M. Sutton[e]

[a]*European Synchrotron Radiation Facility, BP 220, 38043 Grenoble CEDEX, France,* [b]*Argonne National Laboratory, Argonne, IL 60439, USA,* [c]*Massachusetts Institute of Technology, Cambridge, MA 02139, USA,* [d]*University of Delaware, Newark, DE 19716, USA, and* [e]*McGill University, Montreal H3A 2T8, Canada. E-mail: aber@esrf.fr*

(*Received 12 September 1997; accepted 7 November 1997*)

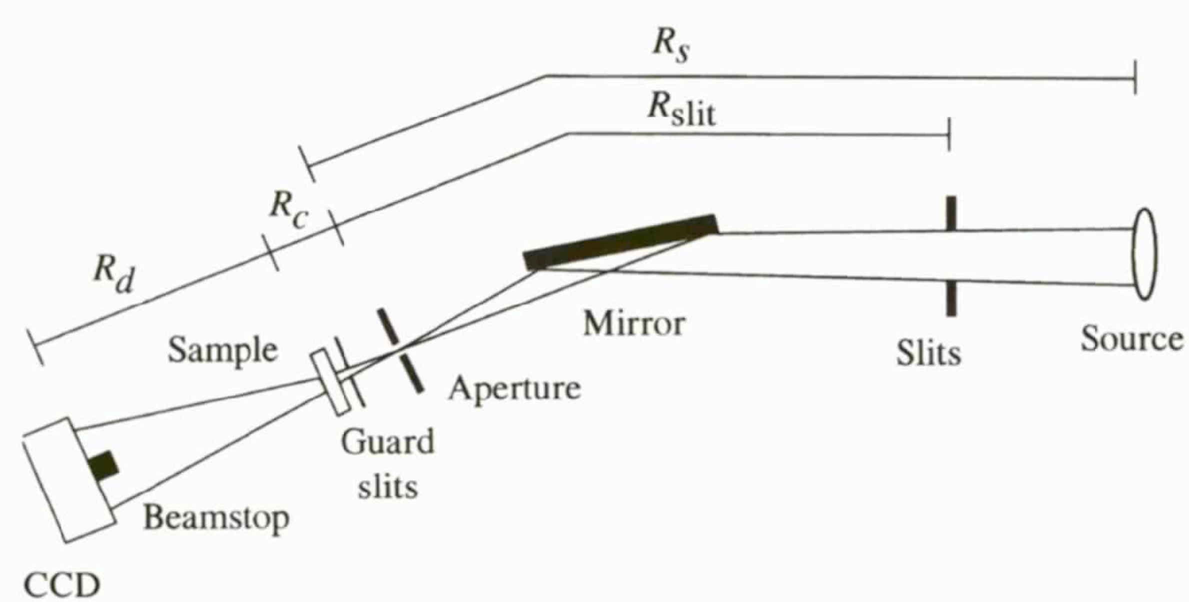


Figure 2
Schematic diagram of the beamline configuration showing major optical components and definitions of distances used in evaluating the static speckle pattern. Lines are guides to the eye indicating possible photon paths from the source to the detector.

J. Synchrotron Rad. (1998), 5, 37-47.

Design and characterization of an undulator beamline optimized for small-angle coherent X-ray scattering at the Advanced Photon Source

A. R. Sandy,[a] L. B. Lurio,[a*] S. G. J. Mochrie,[a] A. Malik,[b] G. B. Stephenson,[b] J. F. Pelletier[c] and M. Sutton[c]

[a]Center for Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139-4307, USA, [b]Materials Science Division, Argonne National Laboratory, Argonne, IL 60439-4832, USA, and [c]Physics Department, McGill University, Montréal, Québec H3A 2T8, Canada. E-mail: larry@slate.imm.aps.anl.gov

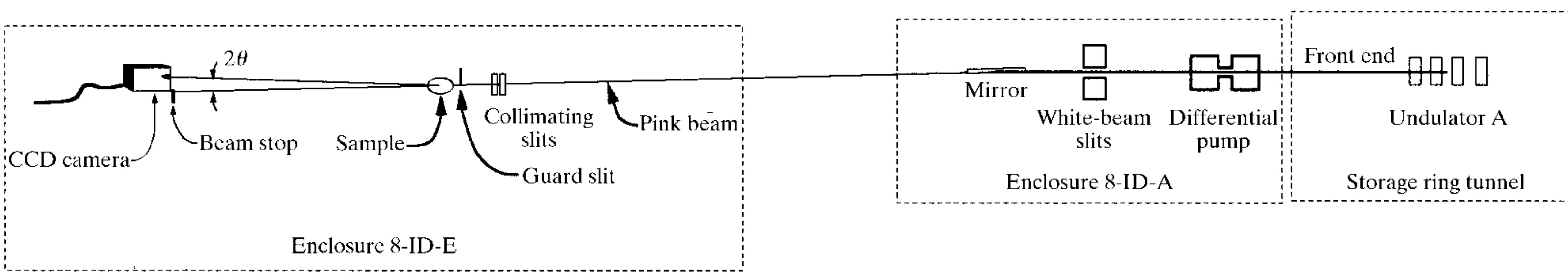
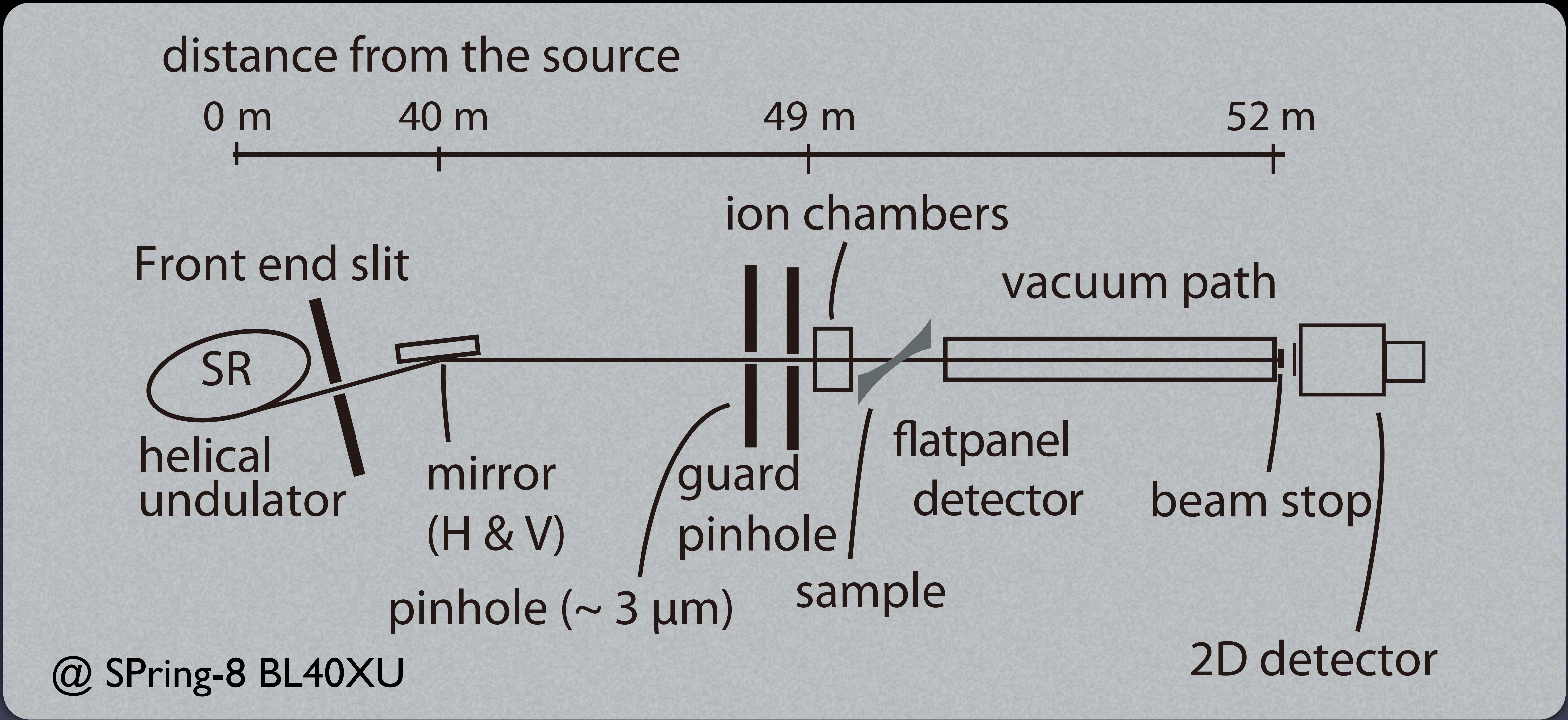


Figure 1
Schematic of the coherent small-angle X-ray scattering beamline and spectrometer.

J. Synchrotron Rad. (1999), 6, 1174–1184.

XPCS Setting @ SPring-8



Y. Shinohara, et al., *J. Synchrotron Rad.*, **17**, 737-742 (2010).
Y. Shinohara, et al., *Macromolecules*, **43**, 9480-9487 (2010).

主要な研究例

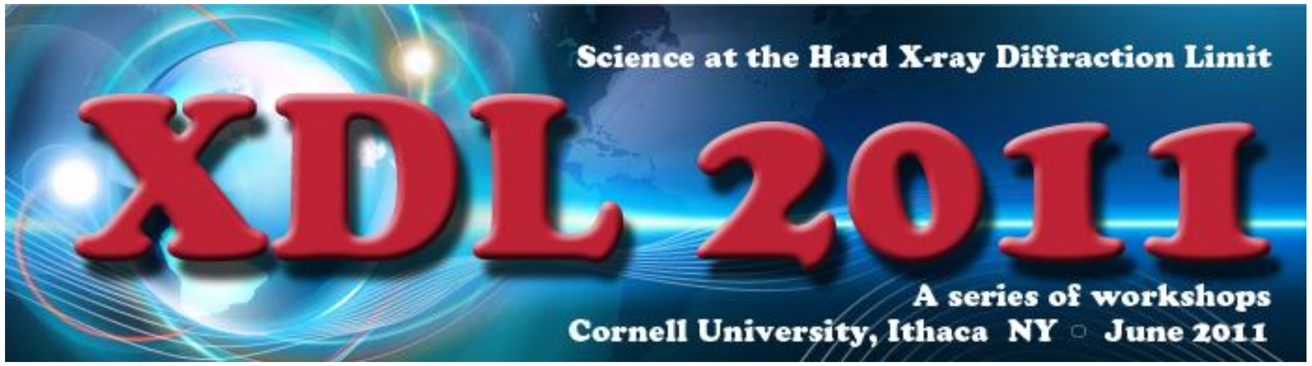
Colloidal Suspension

Surface Dynamics

etc.

Slow Dynamics of Glassy Materials

See G. Grübel, A. Madsen, A. Robert, *Soft-Matter Characterization*, pp. 953-995 Springer-Verlag (2008)



Frontier Science with X-ray Correlation Spectroscopies using Continuous Sources

June 29 & 30, 2011

Robert Purcell Conference Center, Cornell University, Ithaca NY

The purpose:
Future Energy Recovery Linac (ERL) and Ultimate Storage Ring (USR) x-ray sources will be able to deliver coherent hard x-ray beams that are hundreds of time more intense than at most existing storage ring x-ray sources. These intense beams will enable novel ways of probing structural dynamics in matter using correlation spectroscopy (XPCS).

The emphasis is on identifying opportunities and exploring high-impact experiments.

Don Bilderback, Cornell University
"Energy Recovery Linac (ERL) and Ultimate Storage Ring (USR) Properties"

Wes Burghardt, Northwestern University
"XPCS During Shear"

Andrei Fluerasu, National Synchrotron Light Source II
"Dynamics in Soft-matter and Biological Systems: Trends and Opportunities at NSLS-II"

Sol Gruner, Cornell University
"X-ray Detectors: State-of-the-art & Future Possibilities"

Christian Gutt, Deutsches Elektronen-Synchrotron
"X-ray Cross Correlation Analysis (XCCA) and Bond-order in Liquid and Glasses"

Stephen Kevan, University of Oregon
"Probing Magnetic Complexity with Coherent Soft X-ray Beams"

Karl Ludwig, Boston University
"Martensitic Transitions & Opportunities in Non-equilibrium Physics"

Larry Lurio, Northern Illinois University
"Prospects for X-ray Photon Correlation Spectroscopy from Liquid and Soft Matter Surfaces and Interfaces"

Simon Mochrie, Yale University
"Biophysics and Soft Matter"

Michael Pierce, Advanced Photon Source
"XPCS on Surfaces: Challenges and Opportunities"

Maikel Rheinstadter, McMaster University
"Nanobiology: Membranes and Proteins in Motion"

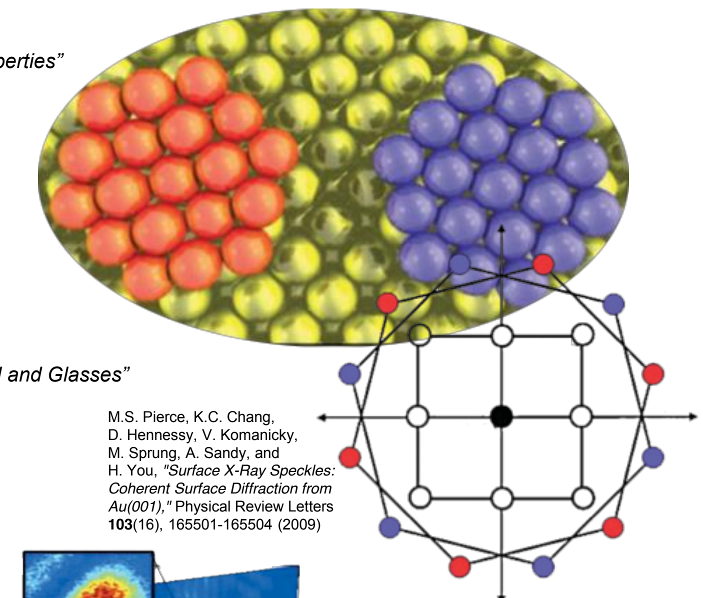
Alec Sandy, Advanced Photon Source
"Scientific Trends and Opportunities from the Perspective of 8-ID"

Bogdan Sepiol, University of Vienna
"Nanoscale Dynamics, Atomic Diffusion"

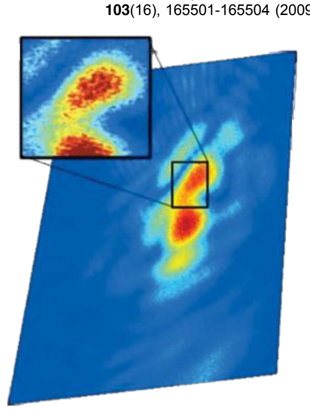
Yuya Shinohara, University of Tokyo
"Hierarchical *Dynamics of Soft Matter and Opportunities at Japanese Future Light Sources"*

Michael Sprung, Deutsches Elektronen-Synchrotron
"Scientific Trends and Opportunities: P10 @ PETRA III"

Mark Sutton, McGill University
"New Opportunities for XPCS"



M.S. Pierce, K.C. Chang, D. Hennessy, V. Komanicky, M. Sprung, A. Sandy, and H. You, *"Surface X-Ray Speckles: Coherent Surface Diffraction from Au(001),"* Physical Review Letters **103**(16), 165501-165504 (2009)



CHESS SSRL DESY

High Energy Accelerator Research Organization (KEK)
Institute of Materials Structure Science (IMSS)
PF Photon Factory

Organizers:
Mark Sutton (McGill University)

XPCS with Future Continuous Light Sources ?

high brilliance ??

高輝度光源 (ERL & USR)を用いた XPCS の可能性

- High S/N
- Time-resolution
- Coherence fraction --> high intensity

High S/N

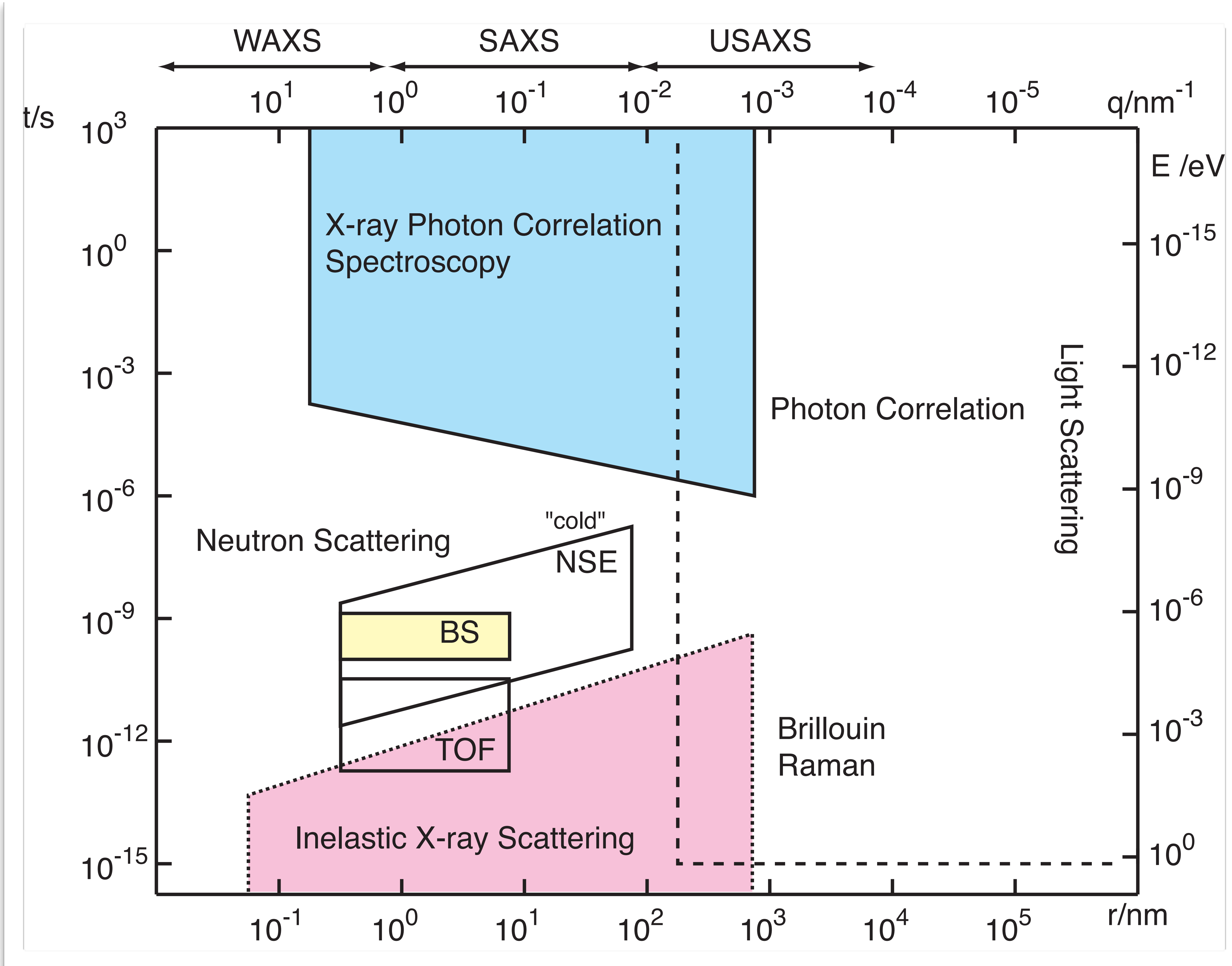
$$\frac{S}{N} = \beta \bar{n} \sqrt{N}$$ from Jakeman (1974)

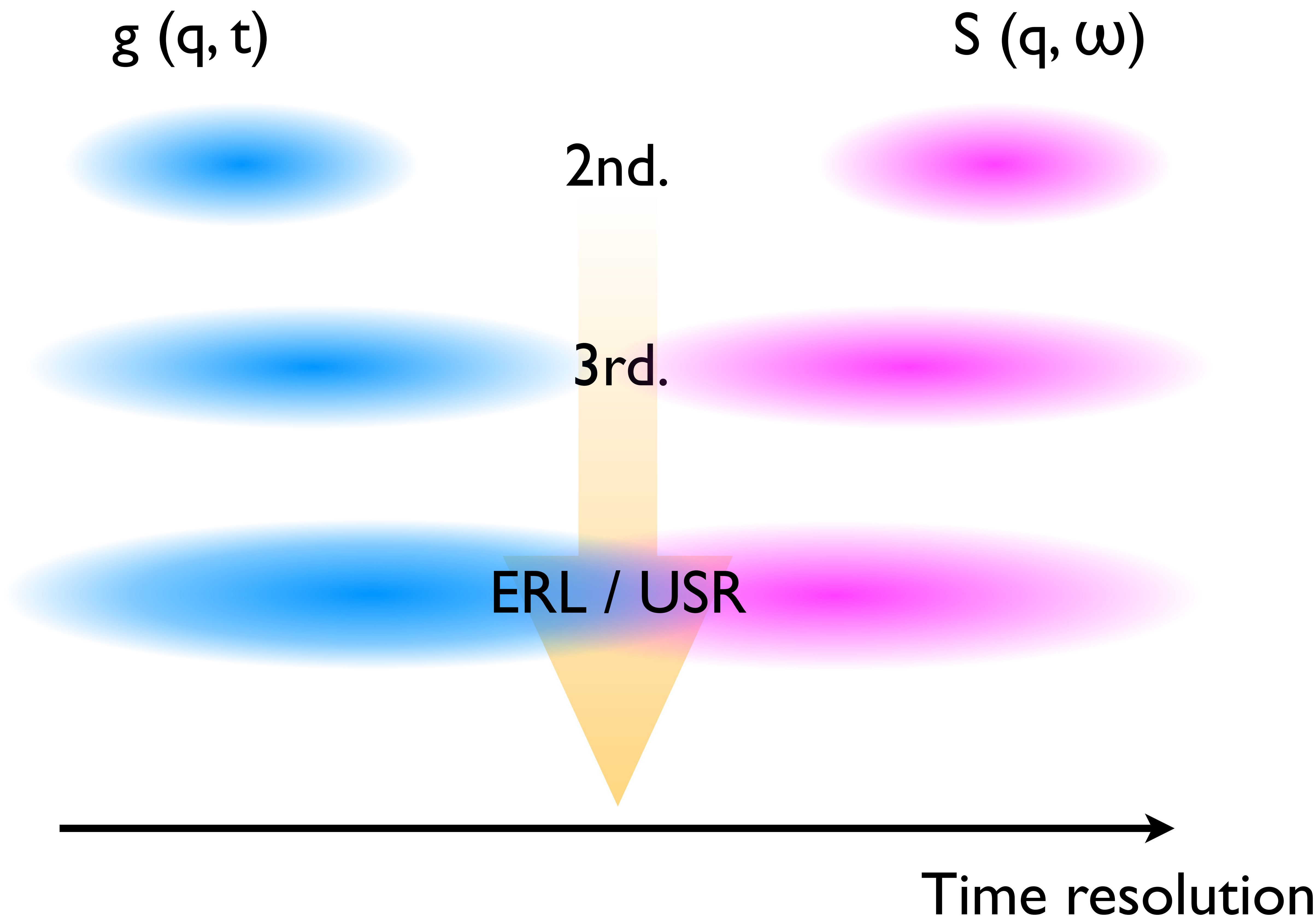
$$= \beta \underline{I_0} \sqrt{\tau_c T N_{\mathrm{sp}}}$$

Count rate (Incident)

M. Sutton, C. R. Physique **9** (2008) 657-667.

Time Resolution




g (q, t)
S (q, ω)
2nd.
3rd.
ERL / USR
Time resolution

Coherence

Beam Size $<$ Transverse Coherent Length

Path difference $<$ Longitudinal Coherent Length

Purpose of WS 6

XPCS with Future Continuous Light Sources ?

Energy Recovery Linac, Ultimate Storage Ring

Broadening awareness of XPCS

⟶ wider community

Workshop Program

- General Description
  - D. Bilderback (Cornell Univ.): ERL & USR
  - S. Gruner (Cornell Univ.): X-ray Detector
  - M. Sutton (McGill Univ.): Review of XPCS

Workshop Program

- Beamlines etc.
  - A. Sandy: 8-ID @ APS
  - M. Sprung: P10 @ PETRA III
  - A. Fluerasu: NSLS II

Workshop Program

- Sciences
  - S. Mochrie (Yale Univ.): Biophysics & soft matter
  - Y. Shinohara (Univ. of Tokyo): Soft Matter
  - W. Burghardt (Northwestern Univ.): Shear
  - L. Lurio (Norther Illinois Univ.): Surfaces & Interfaces
  - M. Pierce (APS): Surfaces

Workshop Program

- Sciences
  - M. Rheinstädter (McMaster Univ.): nanobiology
  - C. Gutt (DESY, Univ. Dortmund): XCAA
  - K. Ludwig (Boston Univ.): non-eq. Physics
  - B. Sepiol (Univ. of Vienna): Atomic diffusion
  - S. Kevan (Univ. of Oregon): Magnetic Systems

Frontier Sciences

- High-angle XPCS
- Heterodyne
- Time-evolution
- X-ray Cross-Correlation Analysis (XCCA)
- Biophysics
- non-equilibrium physics

Shpyrko et al., *Nature*, **447**, 68-71 (2007).

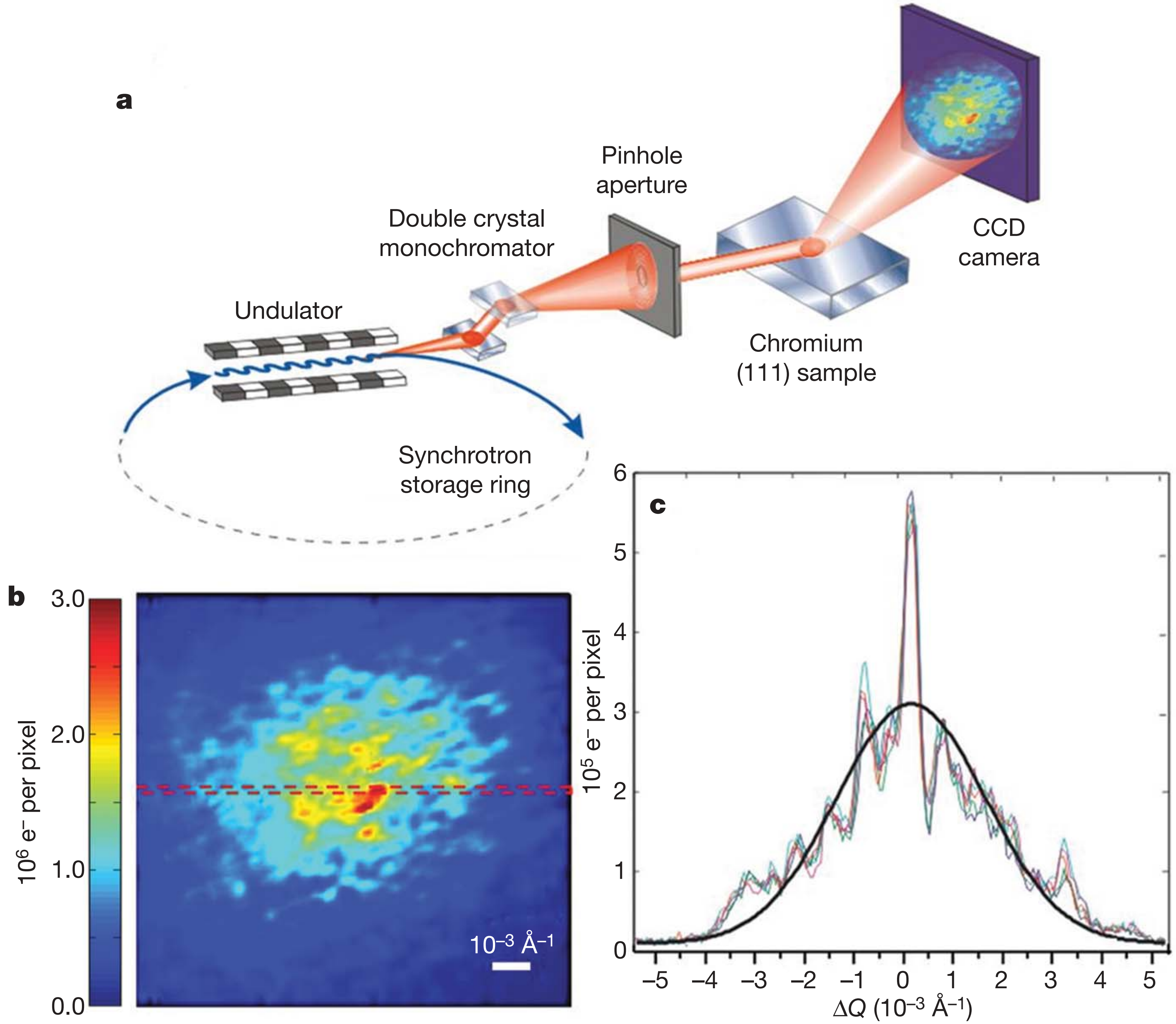
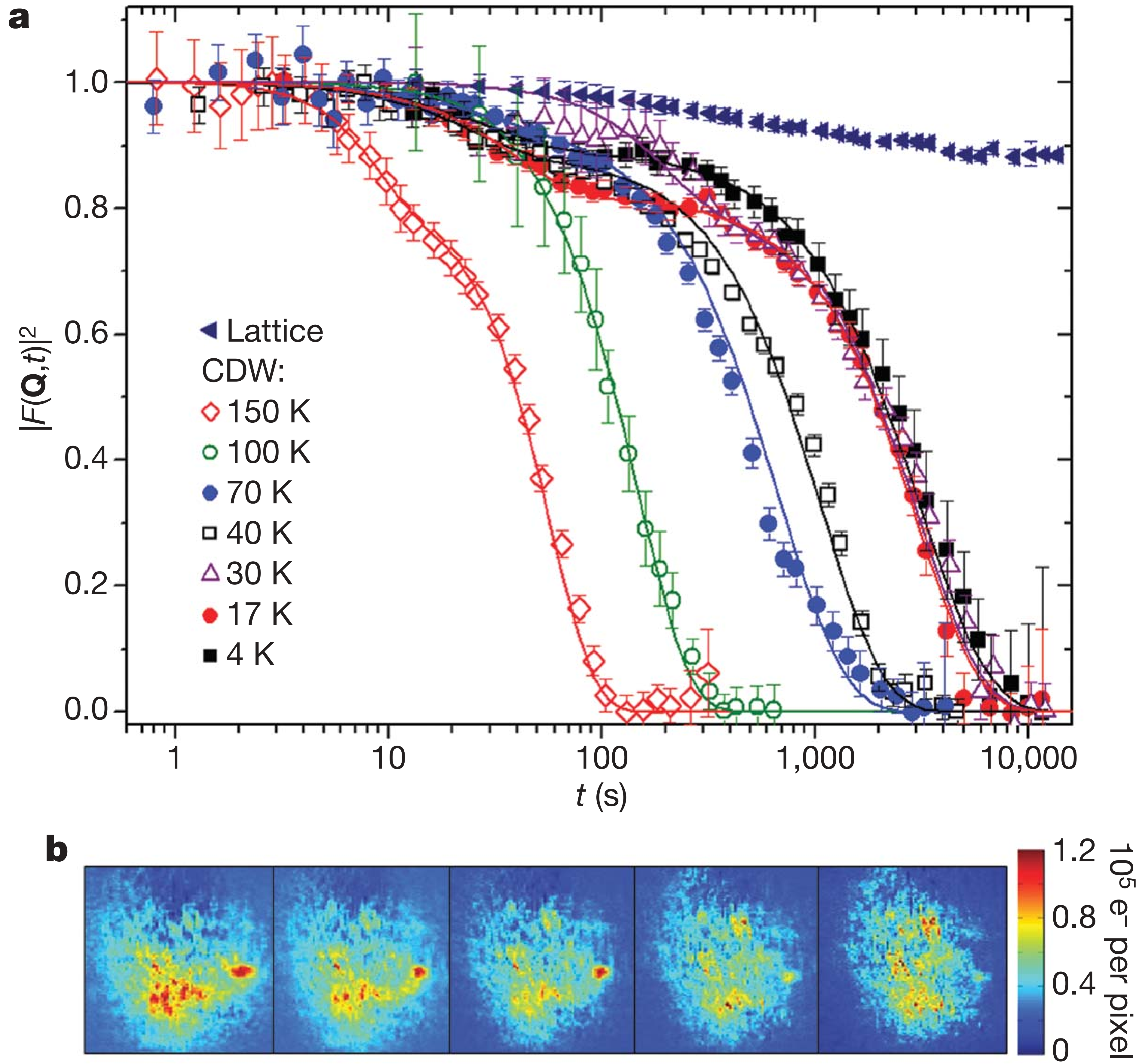


Figure 2 | **X-ray speckle measurements.** **a**, Diagram of the experimental set-up. **b**, CCD image of the X-ray speckle observed for the [200] lattice Bragg reflection. **c**, Intensity distribution for a line scan across the region between the dashed lines in **b**. Five differently coloured and nearly identical lines represent line scans of the portion of speckle pattern shown between the red dashed lines in **b**, taken one hour apart. The black line is a simulated statistically averaged gaussian profile, expected for a completely incoherent beam.



Shpyrko et al., *Nature*, **447**, 68-71 (2007).

Observation of speckle by diffraction with coherent X-rays

M. Sutton*, S. G. J. Mochrie†, T. Greytak†, S. E. Nagler‡, L. E. Berman§, G. A. Held|| & G. B. Stephenson||

* Department of Physics, Centre for the Physics of Materials, McGill University, Montréal, Québec, Canada H3A 2T8
† Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA
‡ Department of Physics, University of Florida, Gainesville, Florida 32611, USA
§ National Synchrotron Light Source, Brookhaven National Laboratory, Upton, New York 11973, USA
|| IBM Research Division, Thomas J. Watson Research Center, Yorktown Heights, New York 10598, USA

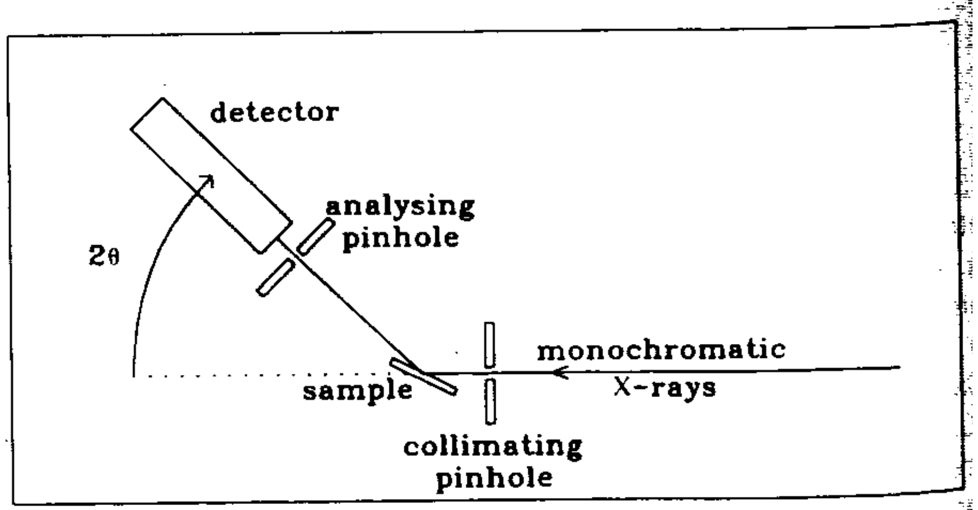


FIG. 1 Schematic representation of the scattering geometry. The angl[e] is shown; α (not shown) is the corresponding angle in the direction no[rmal] to the scattering plane (normal to the page).

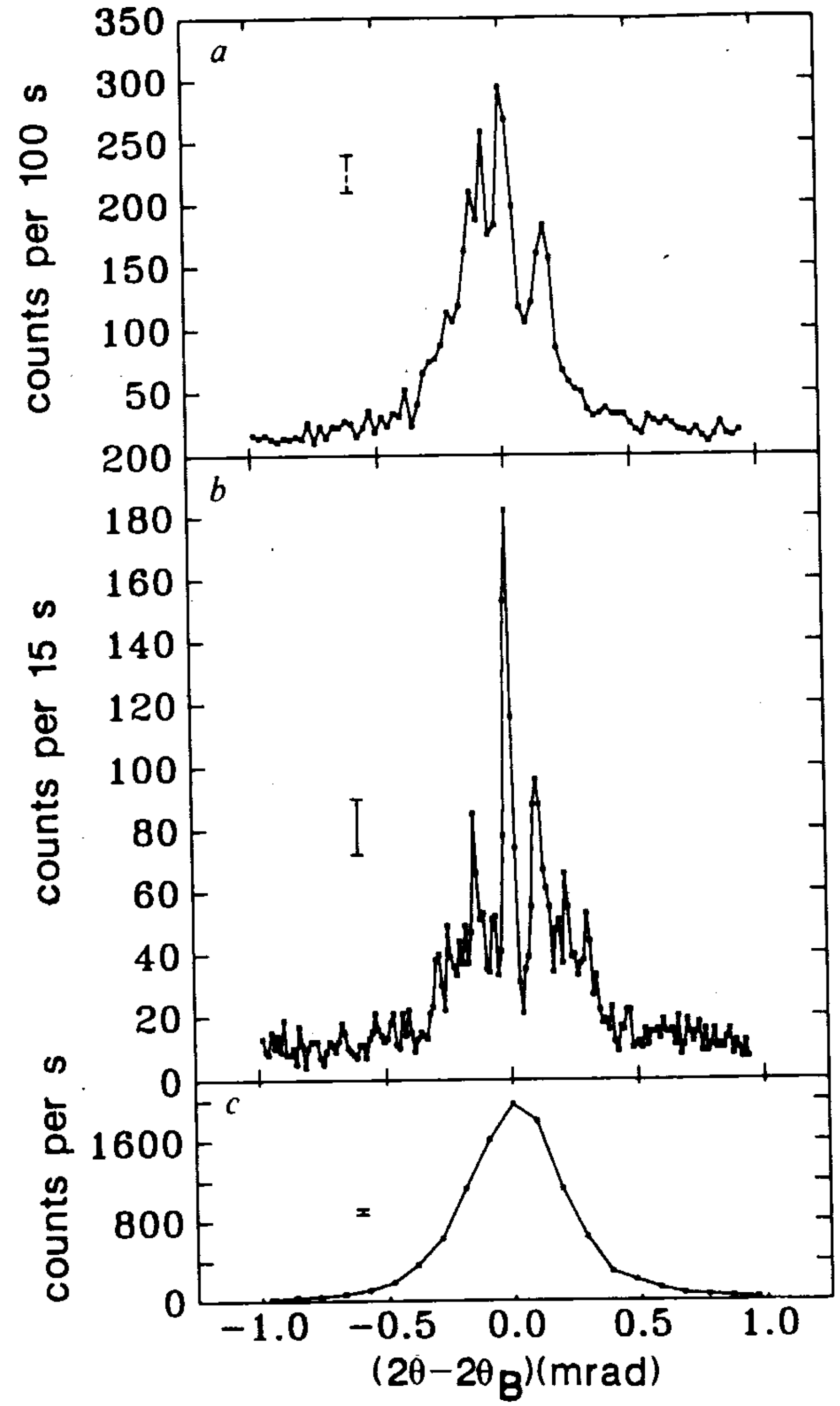


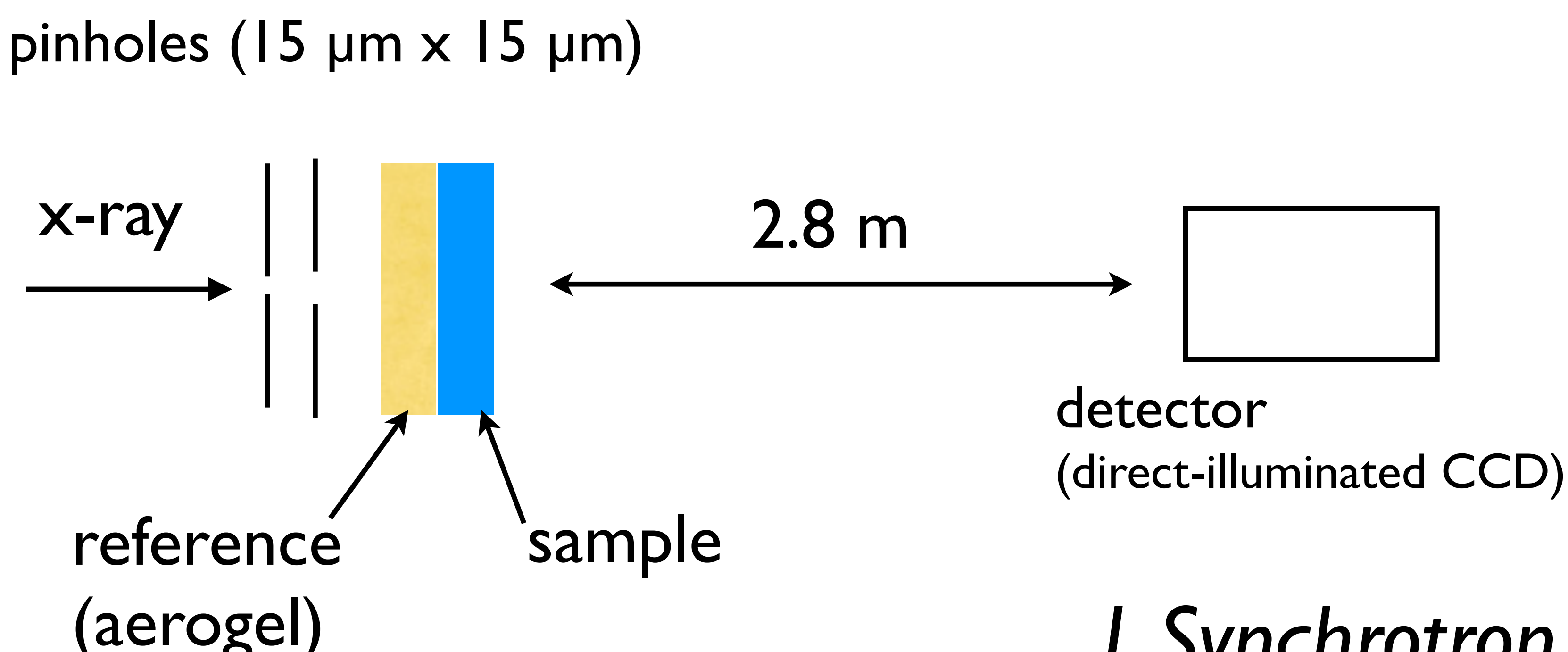
FIG. 4 Speckle patterns measured using *a*, 2.5-μm, *b*, 5 μm and *c*, 50-μm collimating pinholes. The analysing pinholes used were 50, 25 and 100 μm, respectively. Representative error bars are indicated, and the solid lines simply connect the data points. The (001) Bragg angle, $2\theta_B$, is ~23.9°.

Nature **352**, 608- (1991).

X-ray intensity fluctuation spectroscopy by heterodyne detection

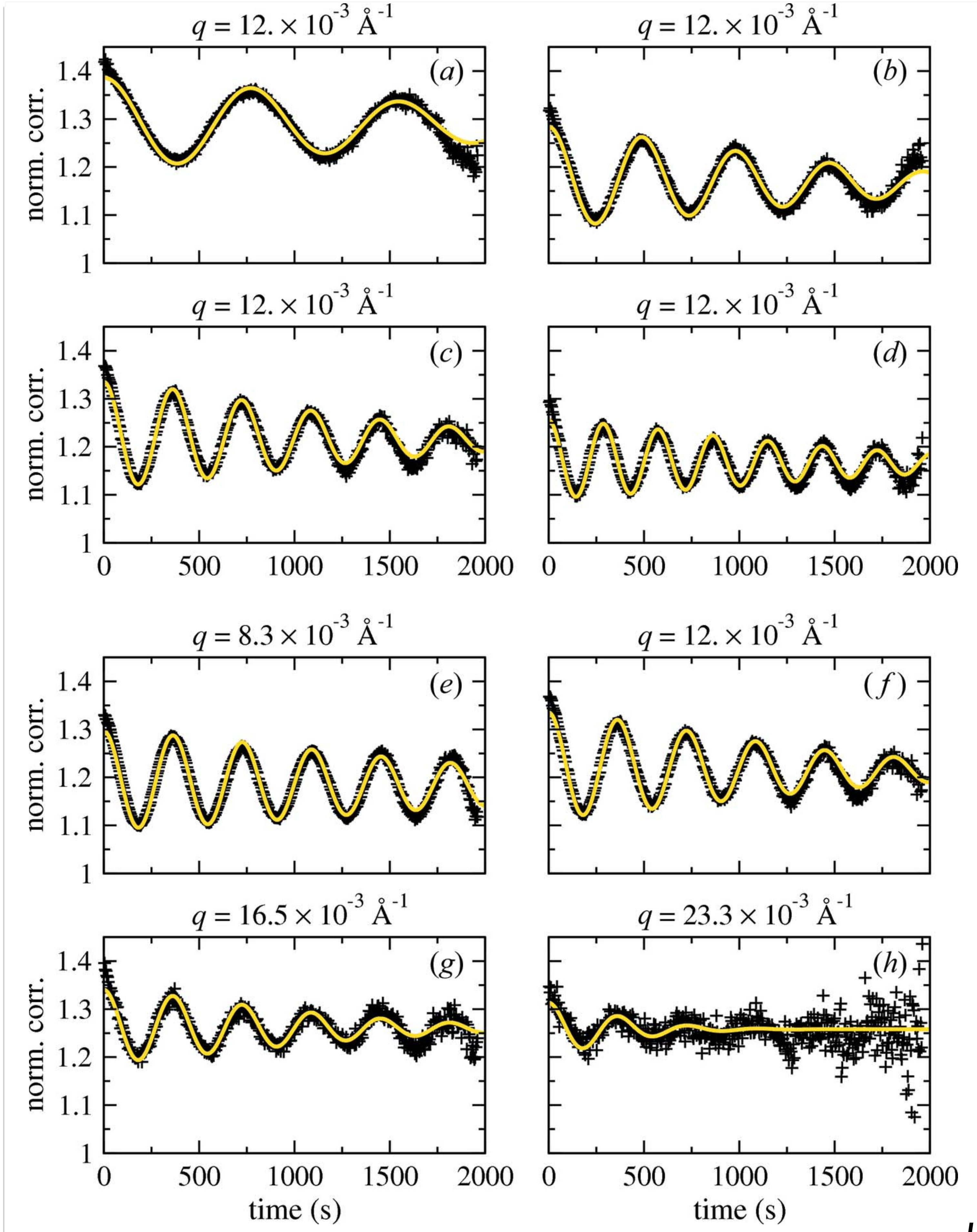
F. Livet,[a]* F. Bley,[a] F. Ehrburger-Dolle,[b] I. Morfin,[b] E. Geissler[b] and M. Sutton[c]

[a]LTPCM-ENSEEG, UMR-CNRS 5614, INPG/UJF, BP 75, 38402 St Martin d'Hères, France, [b]LSP, UMR CNRS 5588 UJF, BP 87, 38402 St Martin d'Hères CEDEX, France, and [c]Physics Department, McGill University, Montreal, Québec, Canada H3A 2T8. E-mail: flivet@ltpcm.inpg.fr



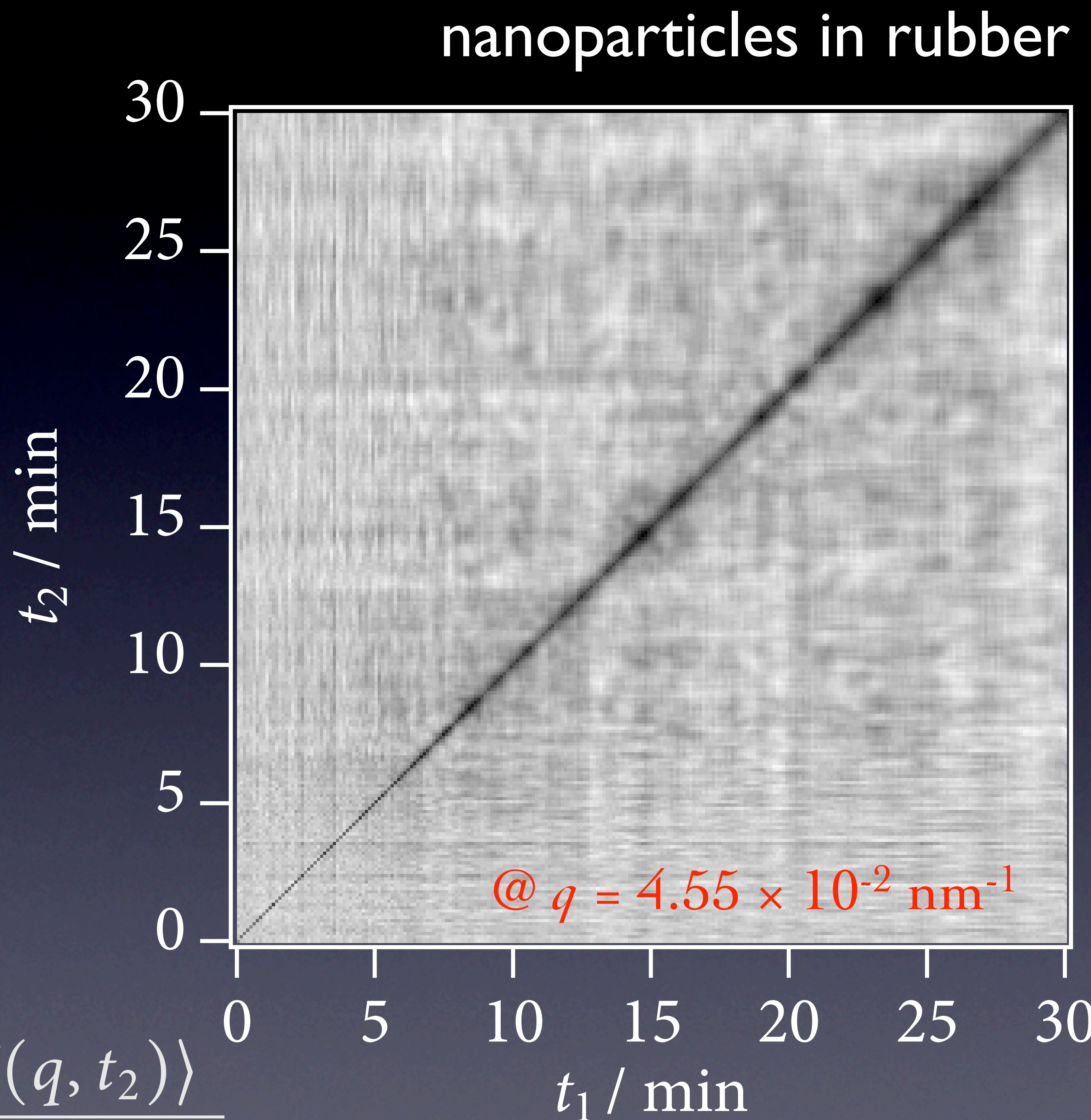
- 10^9 photons/s
- 1.62 Å

J. Synchrotron Rad., **13**, 453-458 (2006).



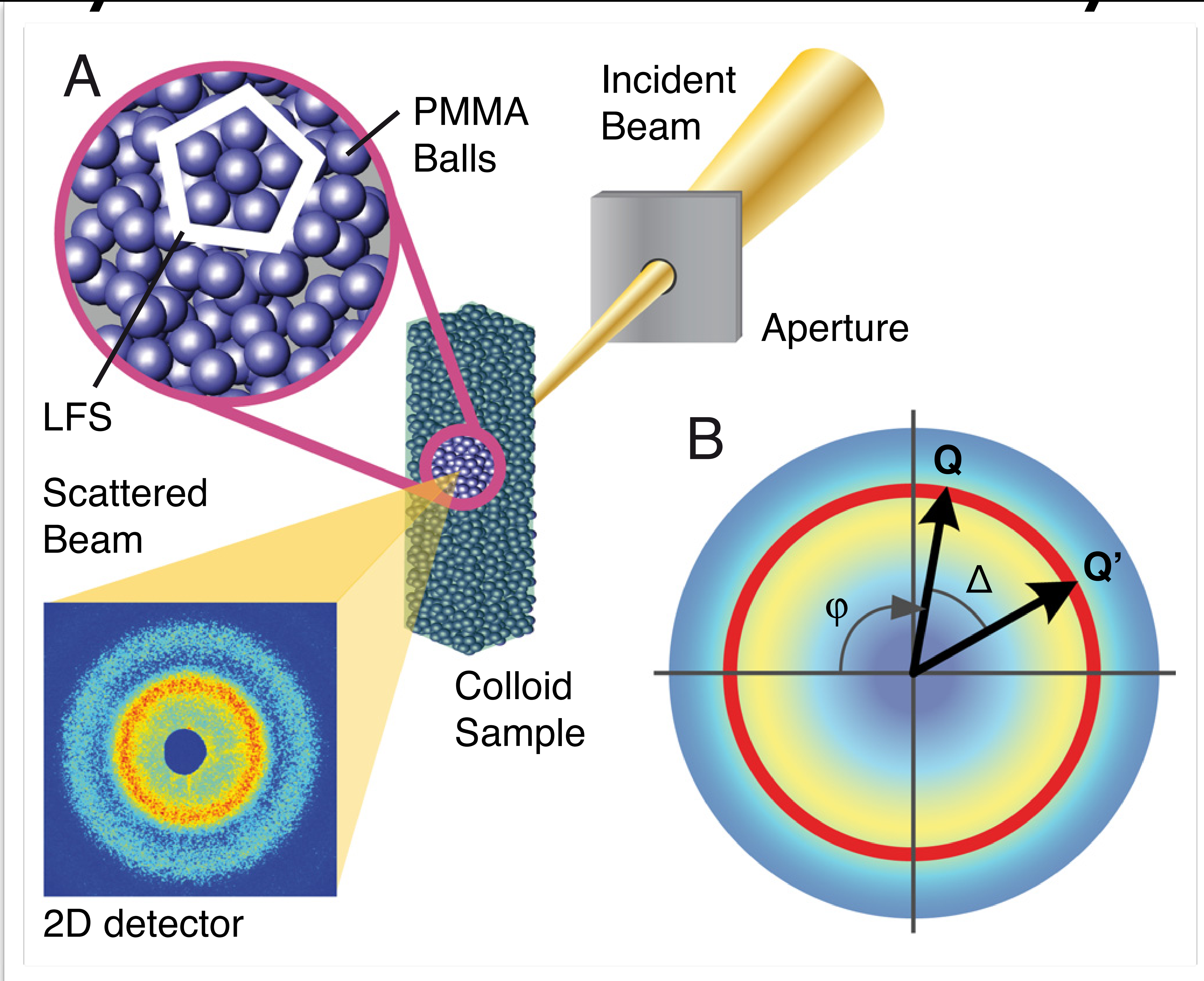
J. Synchrotron Rad., **13**, 453-458 (2006).

Two-time Correlation Fn

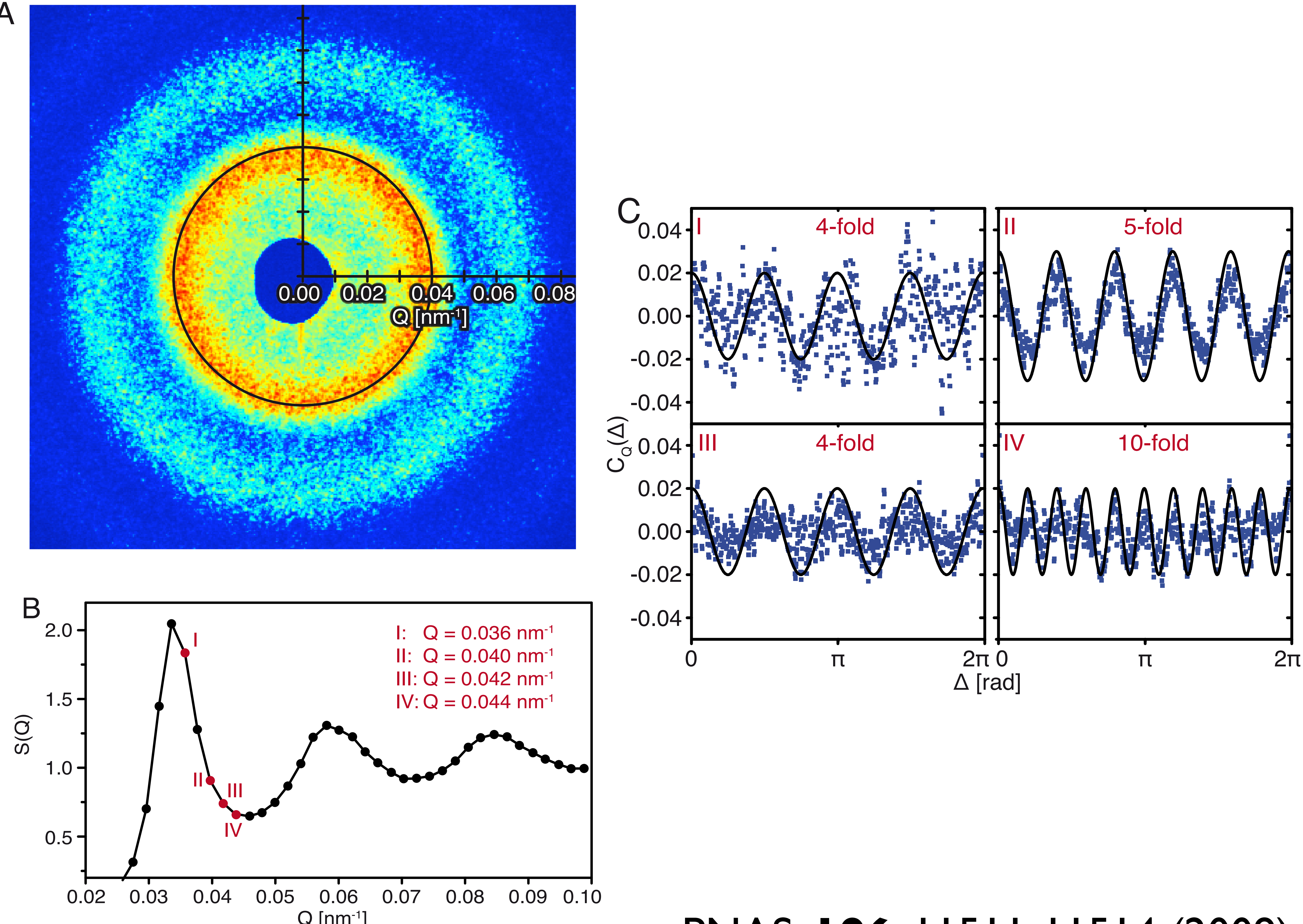


$$C(q, t_1, t_2) = \frac{\langle I(q, t_1) I(q, t_2) \rangle}{\langle I(q, t_1) \rangle \langle I(q, t_2) \rangle}$$

X-ray Cross Correlation Analysis

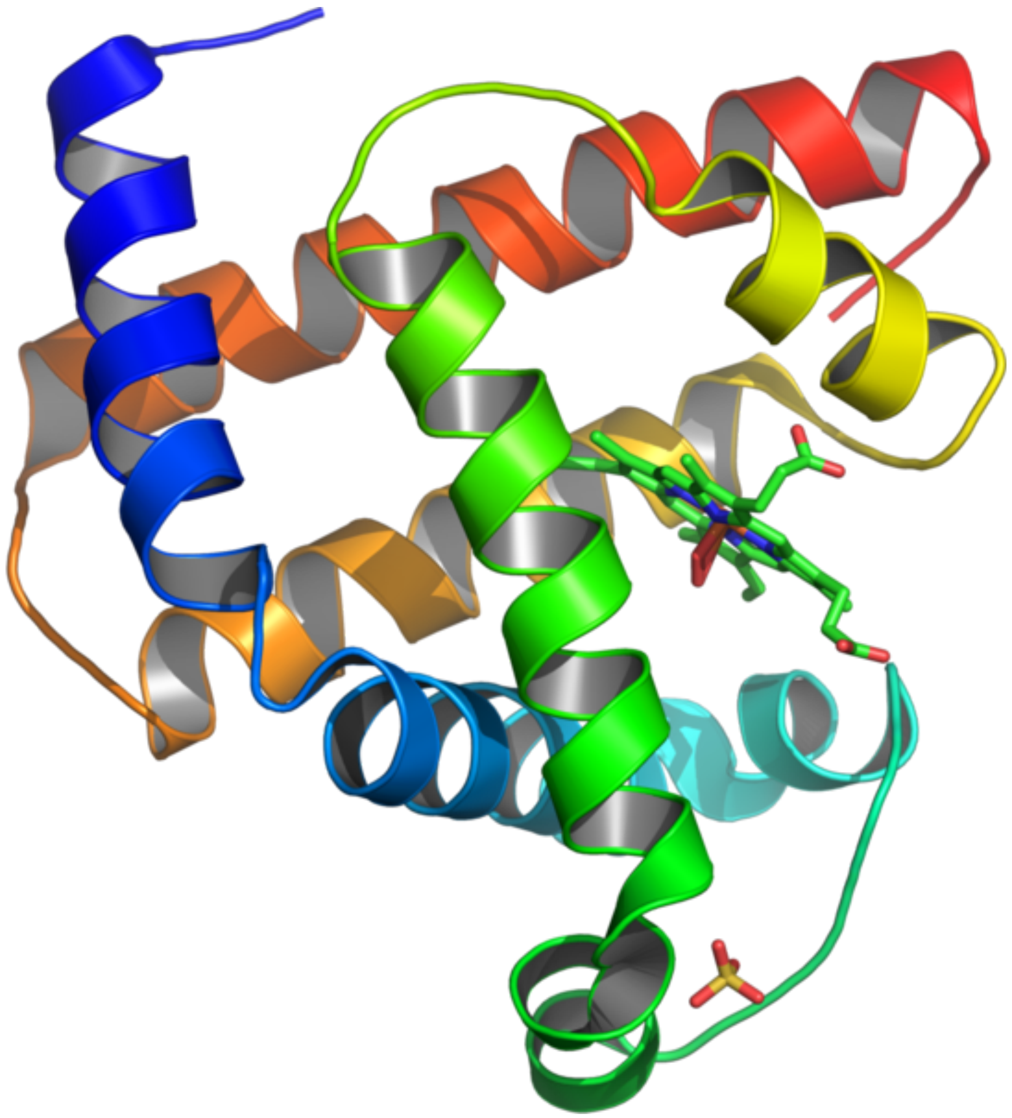


PNAS, **106**, 11511-11514 (2009)



PNAS, **106**, 11511-11514 (2009)

Dynamics in Proteins



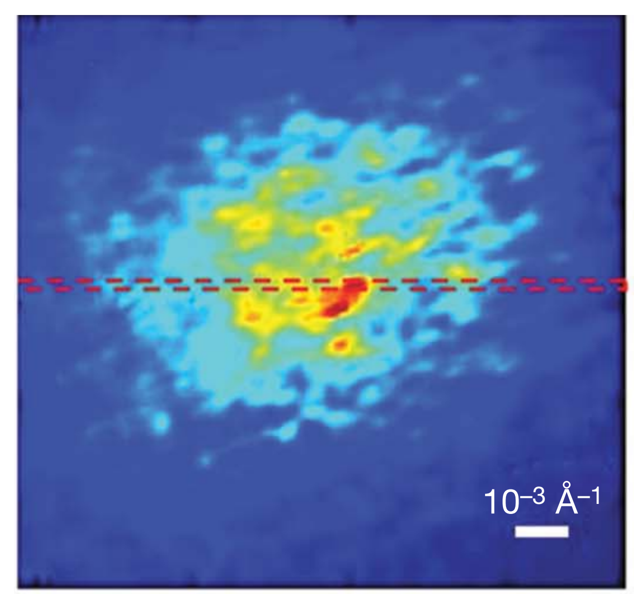
Miyoglobin

from Wikipedia...

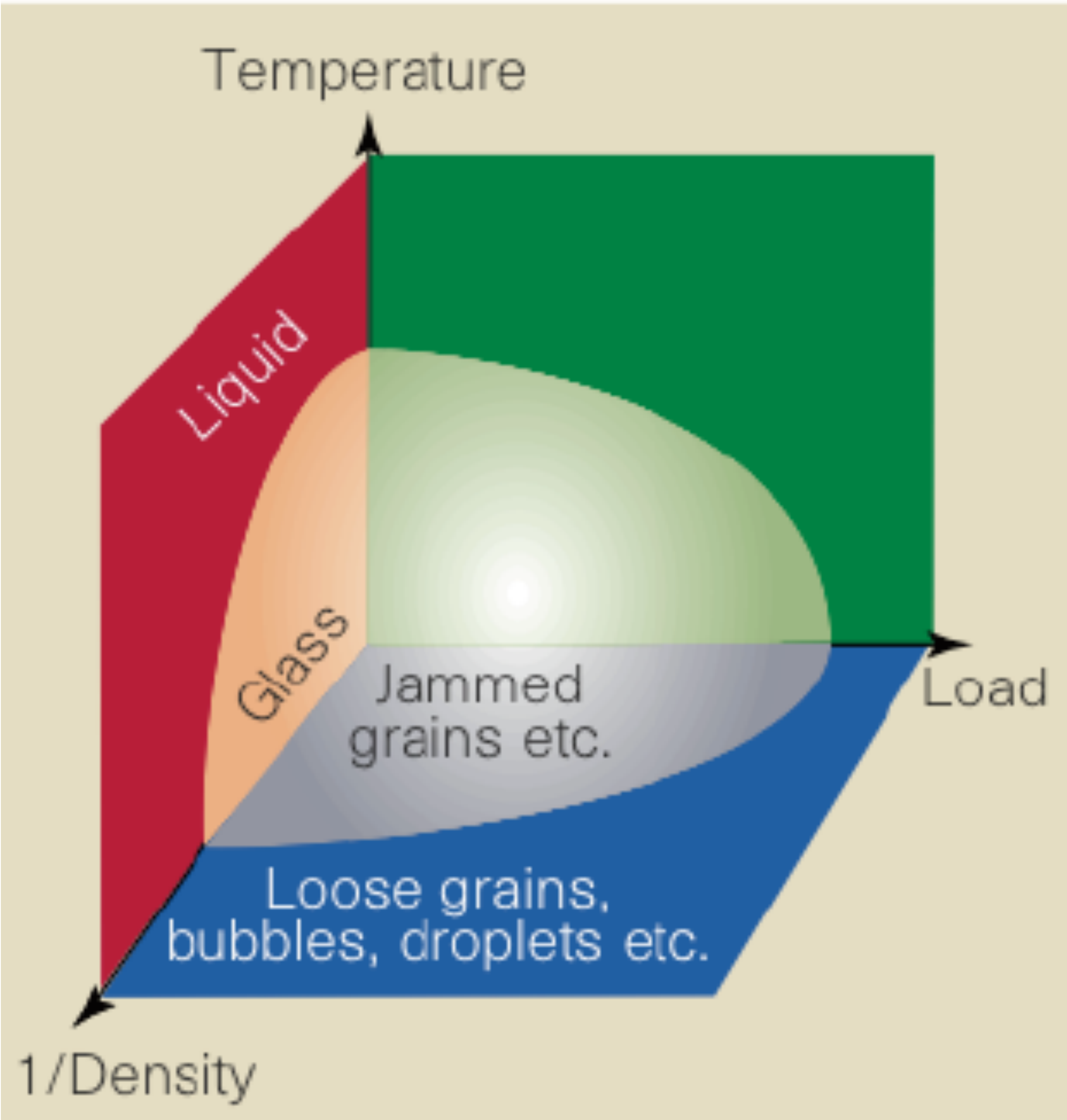
Speckles @ Bragg peaks

↓

Dynamics inside Proteins



Dynamics of Non-equilibrium system

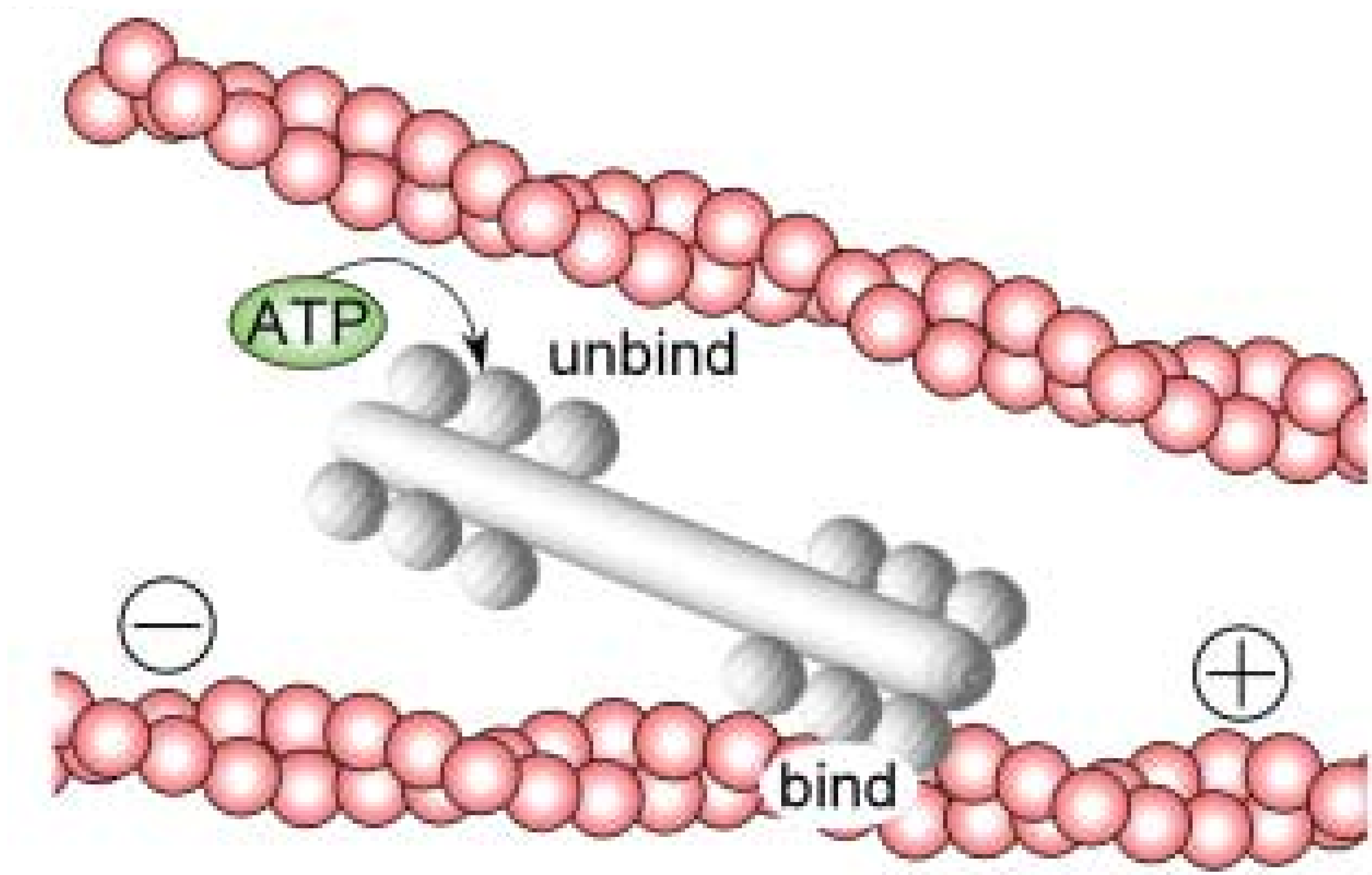


Jamming System

Liu & Nagel, *Nature*, **396**, 21-22 (1998).

- "Fragile Matter" (Cates '98)
  - Hard repulsive particles
- Macroscopic behavior
  - non-elastic constitutive law
- Microscopic behavior
  - "Force Chains"
  - rearrange sensitively to certain deformations

Active Matter



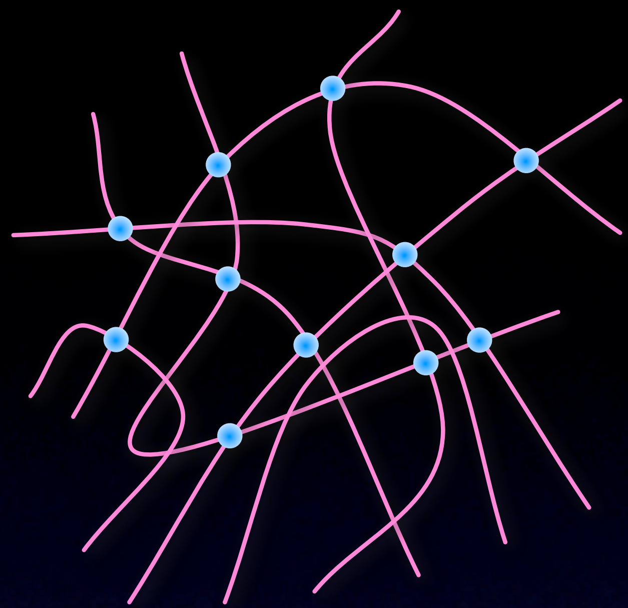
Mizuno et al., *Science*, **315**, 370 (2007).

Hierarchical Dynamics of Soft Matters & Prospects of Japanese Future Light Sources

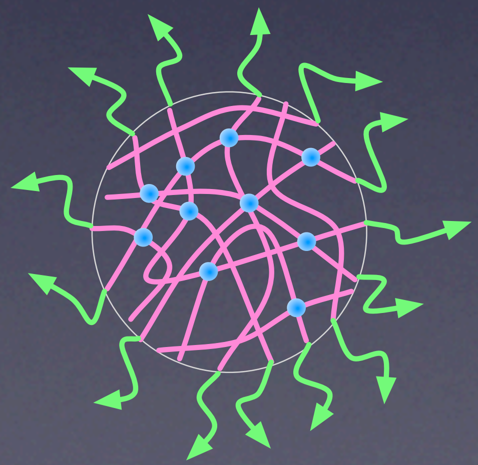
Yuya Shinohara

Department of Advanced Materials Science, Graduate School of Frontier Sciences, The University of Tokyo

yuya@k.u-tokyo.ac.jp



Soft Matter



Soft Matter

Complexity & Flexibility

Hierarchical Structure

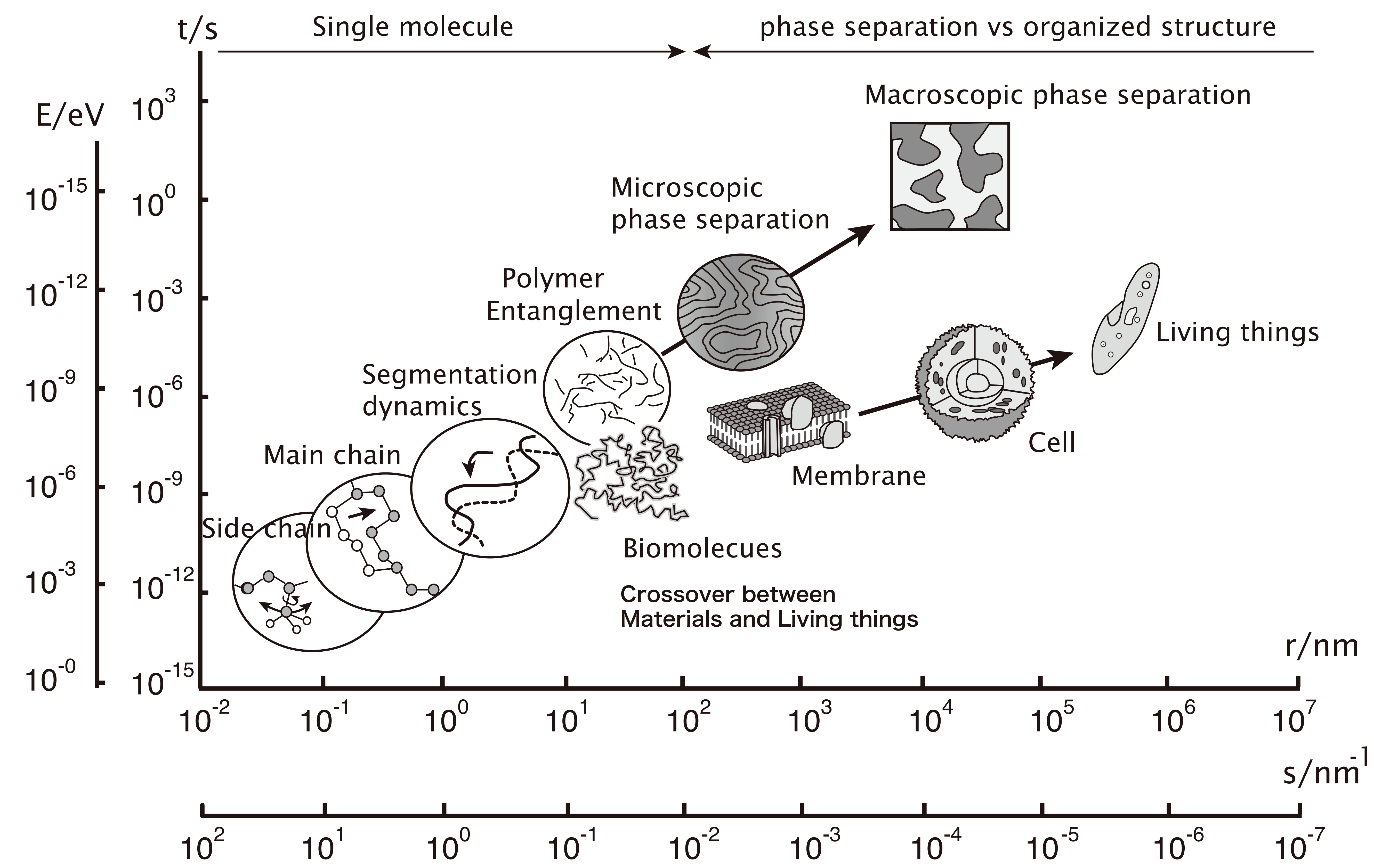
and

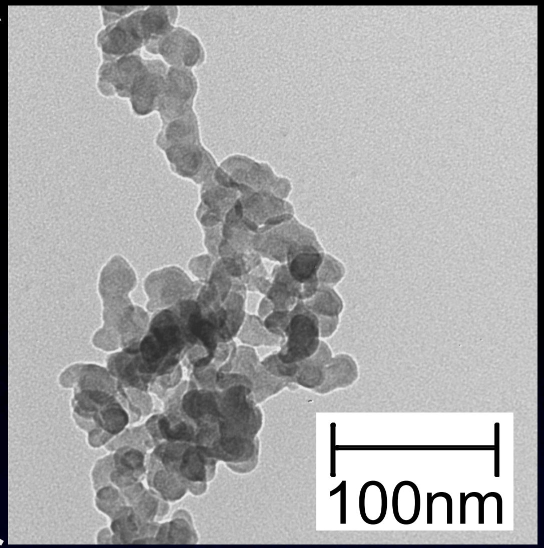
Heterogeneity

←

XPCS using Future Sources

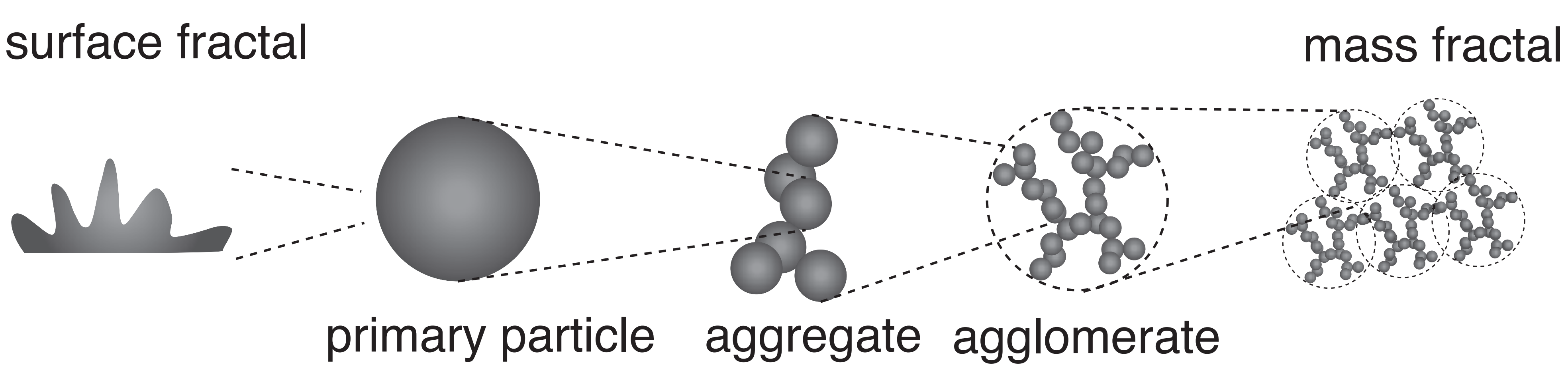
Hierarchical Structure of Soft Matter

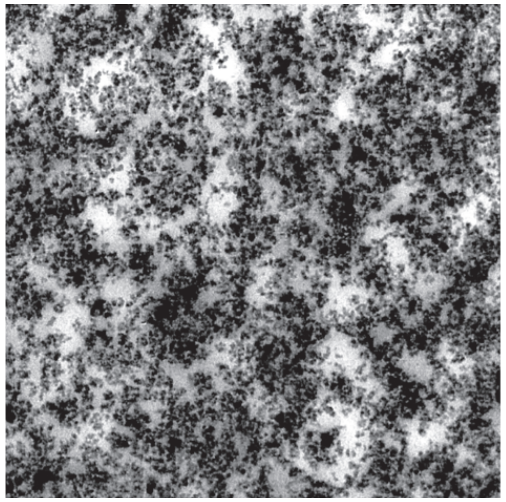




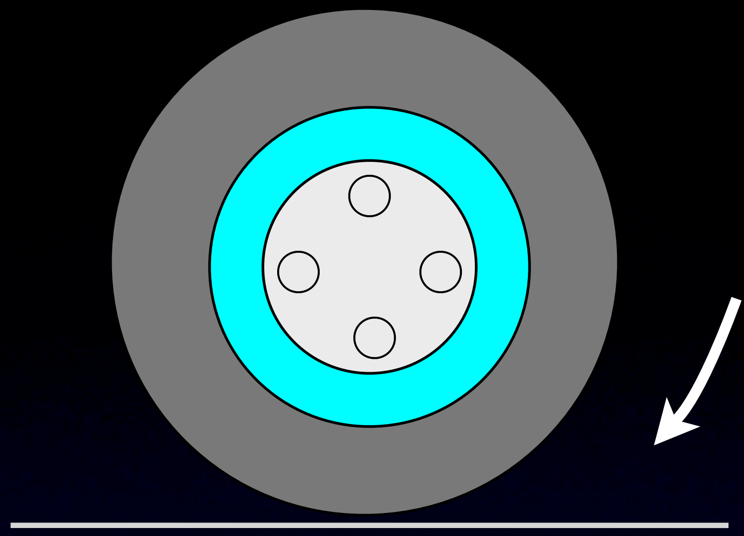
Reinforcement Effect

Time scale ?





TEM Image



Rolling Resistance

(Fuel Efficiency)

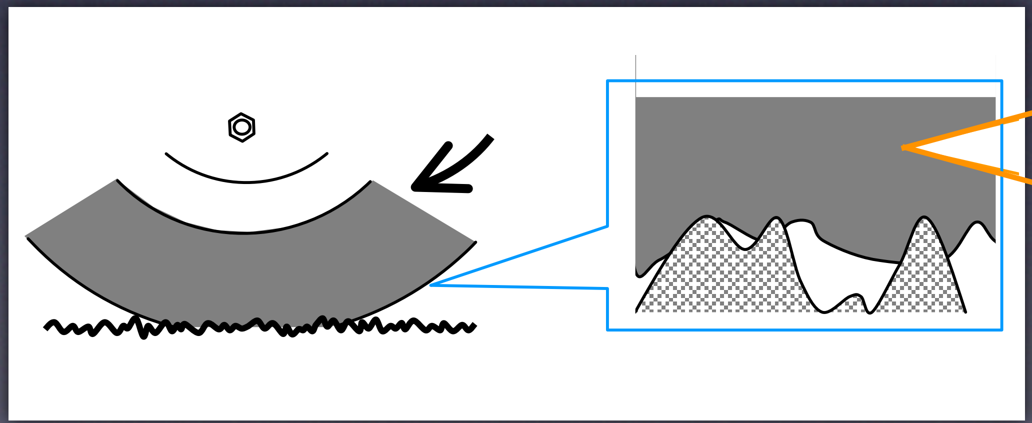
60 km/h → ~ 10 Hz

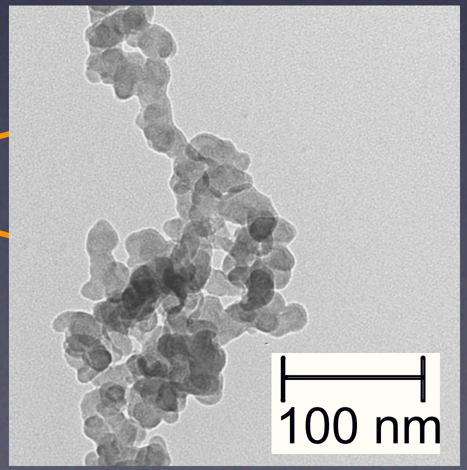
Dynamics of Particles in Different Time Scales ??

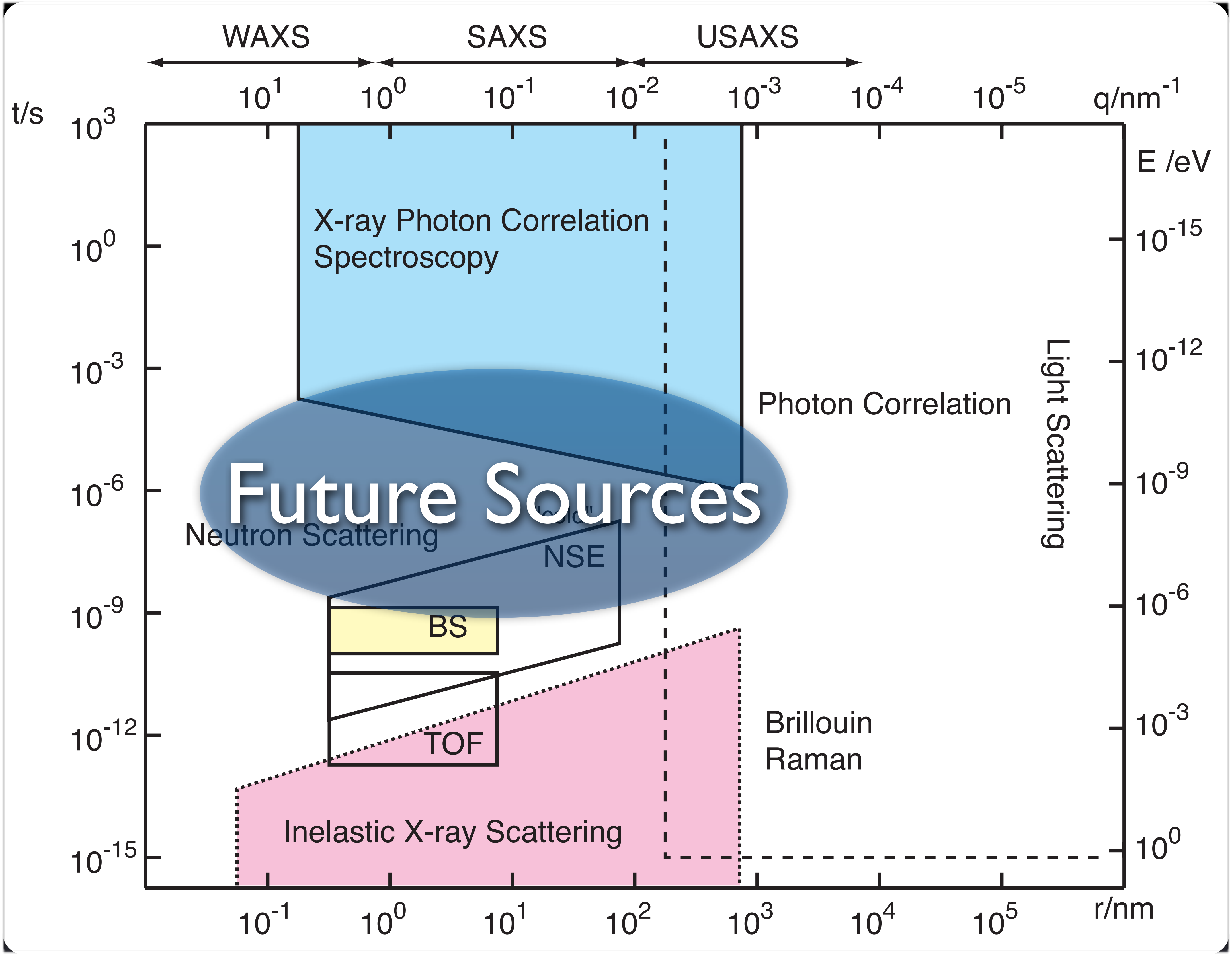
Traction Performance

@ brake, wet road surface

~ 10^4 - 10^6 Hz

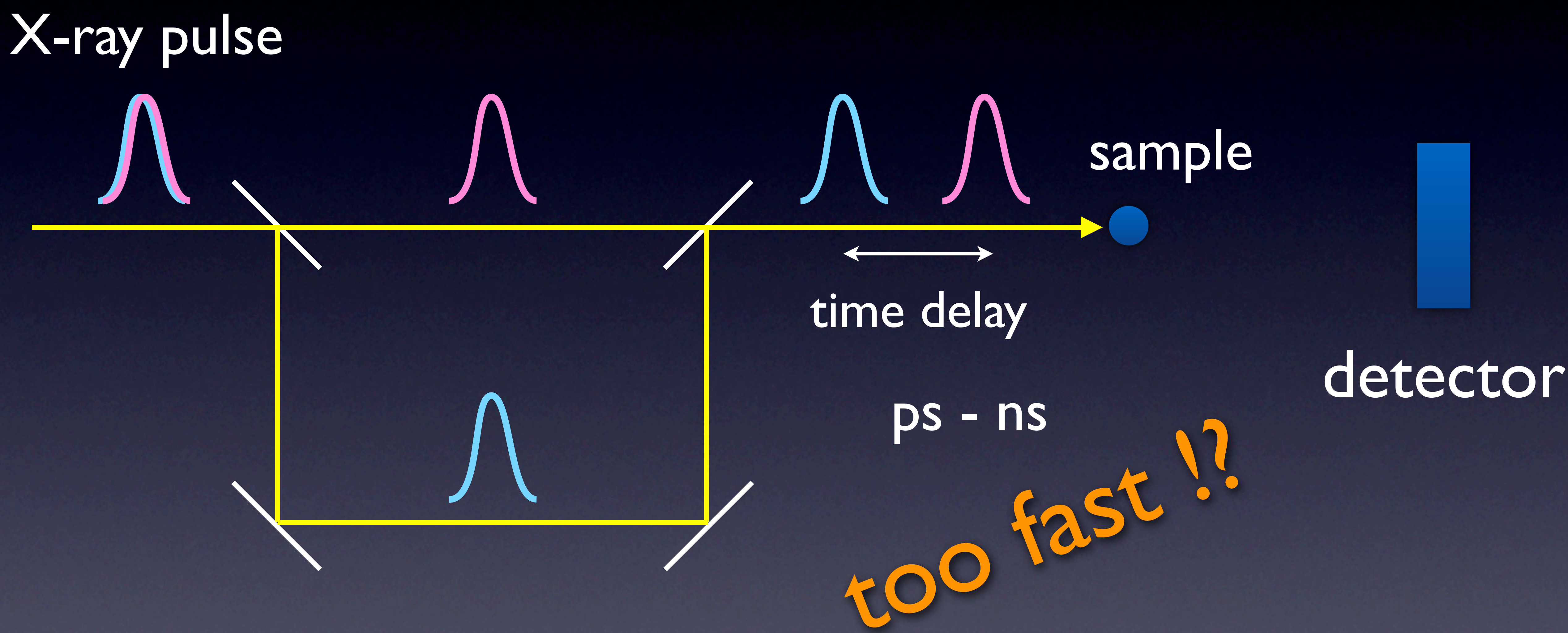




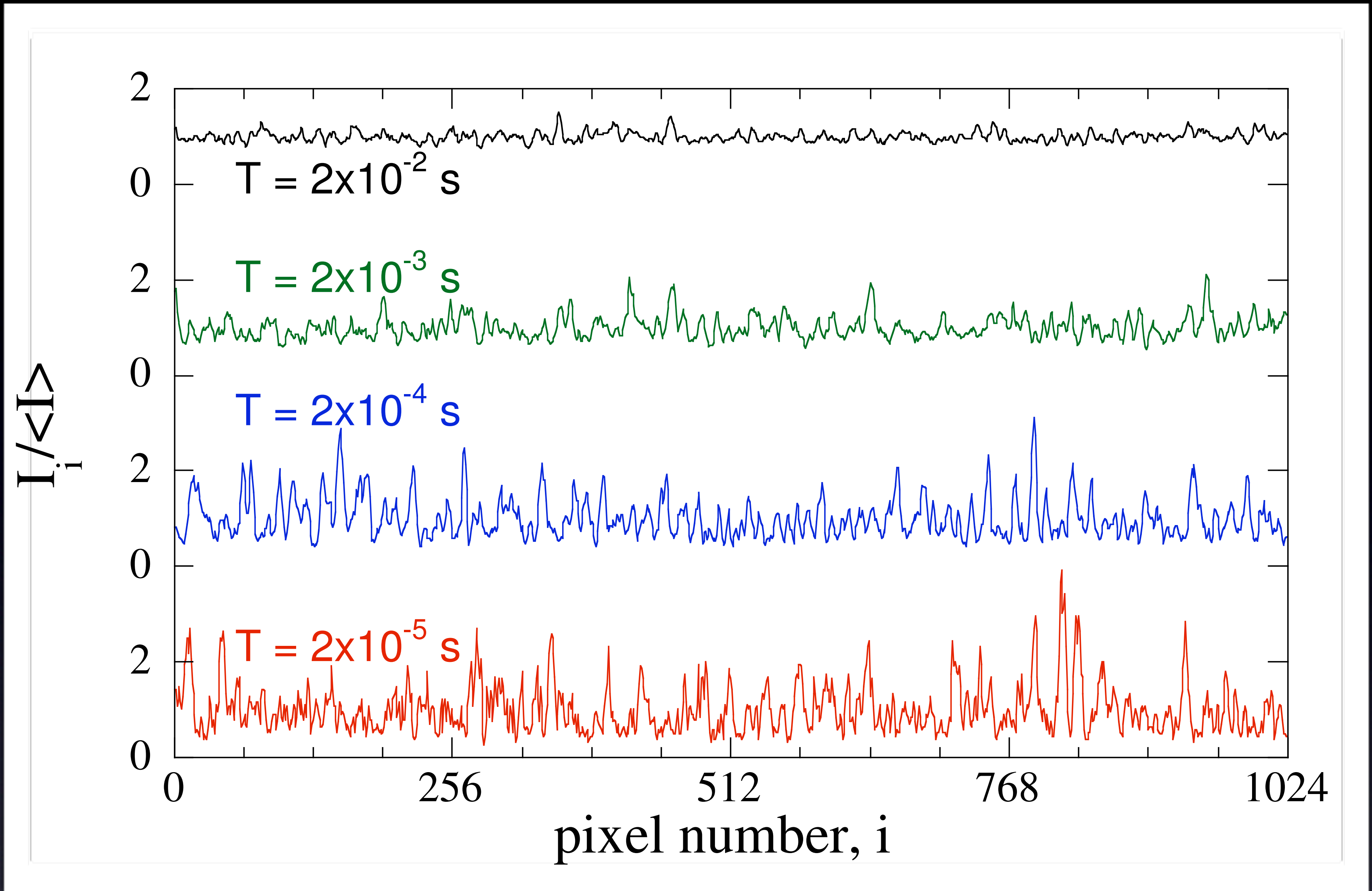


WAXS
SAXS
USAXS
t/s
q/nm^{-1}
10^{1} 10^{0} 10^{-1} 10^{-2} 10^{-3} 10^{-4} 10^{-5}
10^{3} 10^{0} 10^{-3} 10^{-6} 10^{-9} 10^{-12} 10^{-15}
E /eV
10^{-15} 10^{-12} 10^{-9} 10^{-6} 10^{-3} 10^{0}
X-ray Photon Correlation Spectroscopy
Photon Correlation
Light Scattering
Future Sources
Neutron Scattering
NSE
BS
TOF
Brillouin
Raman
Inelastic X-ray Scattering
10^{-1} 10^{0} 10^{1} 10^{2} 10^{3} 10^{4} 10^{5}
r/nm

Speckle Visibility Spectroscopy with FEL



SVS with ERL



D. J. Durian et al.,

Phys. Rev. Lett. **90**, 184302 (2003).
Rev. Sci. Instrum. **76**, 093110 (2005)

Changing Exposure Time --> Change in Visibility of Speckle Patterns

Continuous Source --> XPCS in milli-, micro- & nano-seconds !!

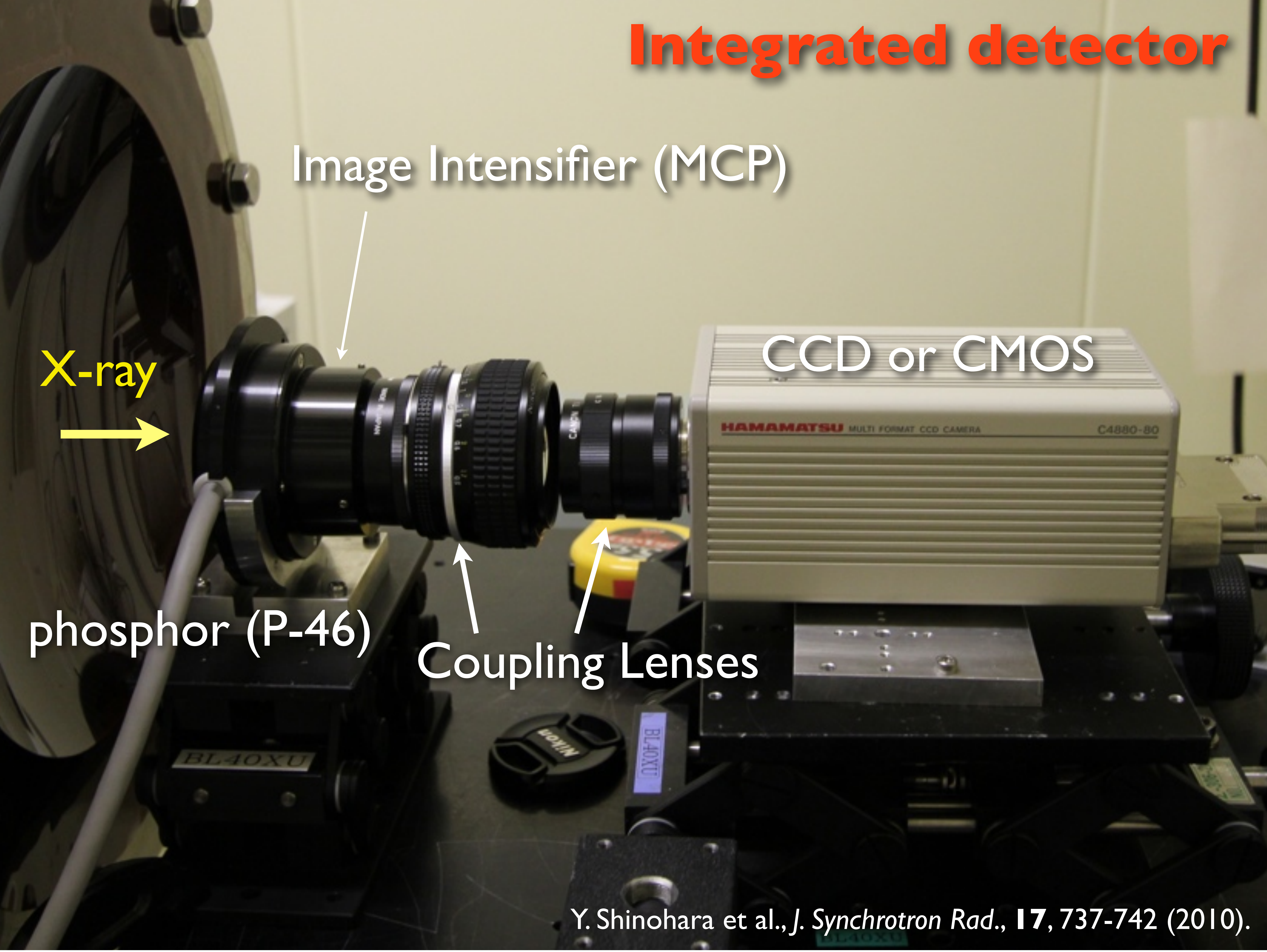
important for Soft Matter Science

XPCS in milli-, micro- & nano-seconds

How ??

Let me introduce..... our detector for XPCS

Integrated detector



Y. Shinohara et al., *J. Synchrotron Rad.*, **17**, 737-742 (2010).

Advantage

Exp. time: easily adjustable (Electronic-shutter)

Image sensor: wide selection

high count rate (integrated detector)

(relatively) low-cost !

high compatibility with ERL (??)

Scattering Study of Soft Matter...

Averaged Structure & Dynamics

SAXS, (conventional) XPCS....

Highly Coherent X-ray

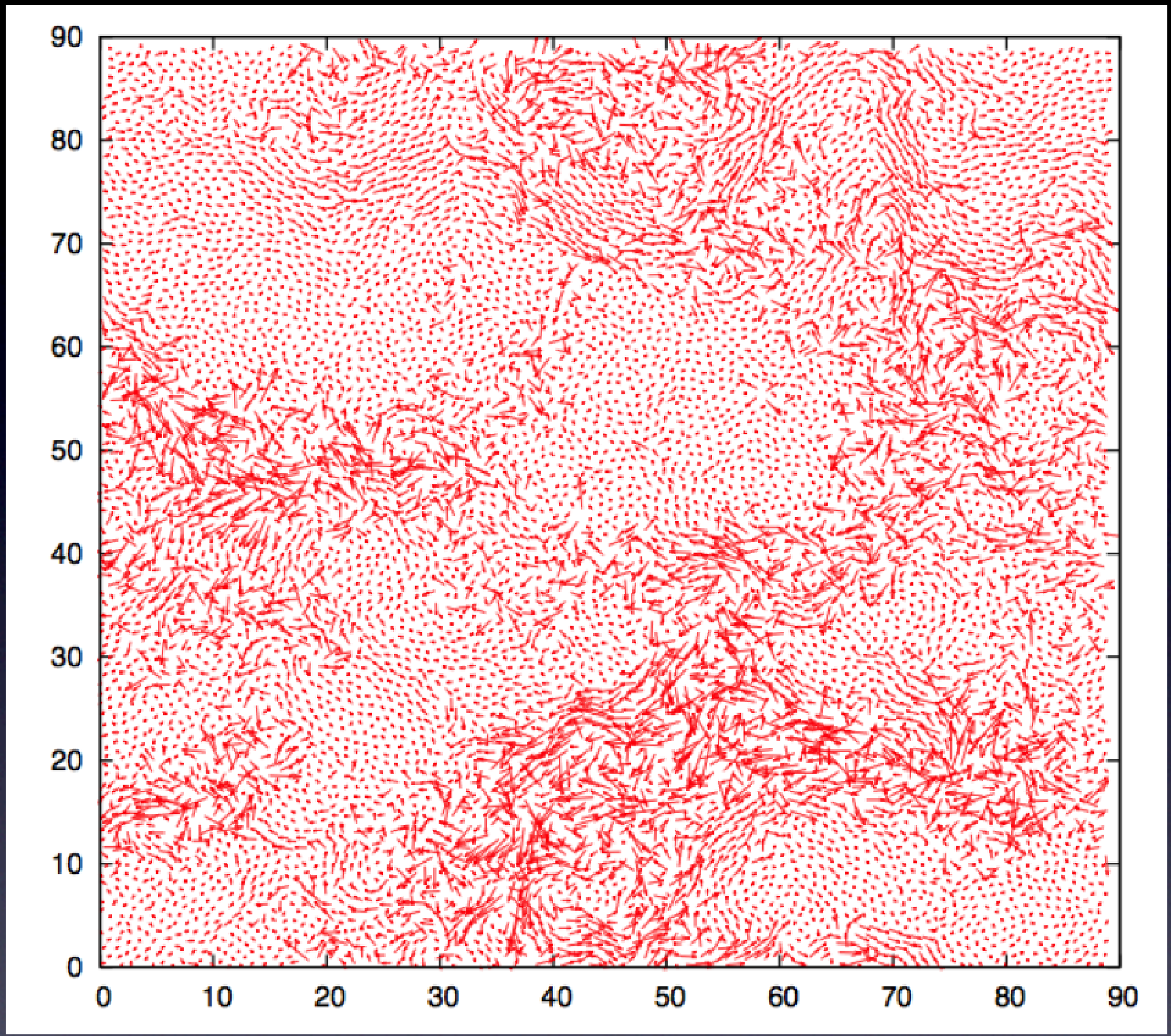
~~Averaged~~ Structure & Dynamics

	Structure	Dynamics
Conventional	SAXS (averaged structure)	Conventional XPCS (averaged dynamics)
Future	Coherent Diffraction Imaging	Future XPCS

Local, heterogeneous, detailed distribution...

Dynamical Heterogeneity

Visualization of Dynamical Heterogeneity



L. Berthier, *Physics*, **4**, 42 (2011)

Highly Brilliant
Continuous source

+

ex.)
Near-Field Scattering

R. Cerbino & A. Vailati, *Curr. Opin. Colloid In.*, **14**, 416 (2009)

Mapping of Dynamics in nm - mm scale !!

Brief Summary

XPCS with Future Light Sources

- Dynamics in wider temporal scale
- Distribution of Dynamics

課題

- 検出器＆ソフトウェア
- 一般社会（産業）への応用