RIES Hokkaido University Research Institute for Electronic Science

XDL2011 報告

Workshop 1-
Diffraction Microscopy, Holography and Ptychography using Coherent Beams

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Research Institute for Electronic Science,
Hokkaido University

Outline

- ワークショップ概要
- 回折顕微法
- ホログラフィー
- タイコグラフィー
- Cross-correlation Analysis / Fluctuation Microscopy
- ワークショップの内容

- ワークショップでの私の発表

Workshop 1-

Diffraction Microscopy, Holography and Ptychography using Coherent Beams

Organizers:

Janos Kirz (Lawrence Berkeley National Lab)
Qun Shen (National Synchrotron Light Source II)
Darren Dale (Cornell University)

Purpose:

The purpose of the workshop is to assess the state-of-the-art in the use of coherent hard x-ray beams for high-resolution imaging. Diffraction microscopy, Fourier transform holography and ptychography are making rapid strides, but have yet to realize their full potential due to current limitations of spatial and temporal coherence of hard x-ray beams. We are especially interested in exploring what might be most feasible with a Energy Recovery Linac (ERL) and Ultimate Storage Ring (USR) x-ray sources.

http://erl.chess.cornell.edu/gatherings/2011_Workshops/details1.htm

Description:

Coherence-based imaging experiments are limited mainly by the lack of coherent X-ray sources. Diffraction microscopy and related techniques have started to extend the resolution in X-ray microscopy beyond the limitations of X-ray focusing elements such as zone plates or multilayer Laue Lenses. Calculations indicate that these techniques should be limited only by radiation damage, and/or the diffraction limit. While using short pulse “flash” illumination at X-ray FELs can yield sufficient scattered intensity to image a sample with a single shot, the power in that shot vaporizes the specimen. Hence FELs are not ideal for imaging techniques requiring multiple exposures, for example ptychography and tomography. ERL and USR sources are being conceived to deliver intense, spatially coherent, hard X-ray beams with a quasi-continuous time structure. Sufficient temporal coherence may be provided by the undulator source directly, possibly further filtered using a monochromator. The ideal X-ray energy will depend on the sample, but in general soft X-rays offer higher scattering cross sections and higher coherent flux, but higher energies provide greater penetration through the specimen, and hence the Born approximation (generally assumed in the reconstruction process) is better satisfied. The increased penetration of harder x-rays will also allow greater emphasis on studies of samples in complex environments. The expected high coherent flux may provide the opportunity to study time-sequence series of dynamic phenomena.

For materials where the resolution is not limited by radiation damage, these techniques will provide new capabilities to analyze and visualize porosity, inclusions, defects, and other buried structures. These new imaging techniques can be used in combination with spectroscopy to create a sensitive tool highlighting elemental constituents. Work by Ian Robinson's group has provided a unique new tool to investigate the morphology, strain fields and defects in nanocrystals. This approach makes use of the fine speckles around Bragg peaks, and requires the short wavelength and penetrating power of high-energy X-rays.

For biological samples, such as cells, tissue sections, etc., one can use cryogenic freezing to preserve the morphology of the sample while imaging with a resolution of about 10 nm. High-pressure cryogenic freezing techniques are under development at Cornell to prepare such specimens while minimizing structural perturbations due to the expansion of water on freezing.

Fourier transform holography techniques provide a quick and easy route to a reconstruction at a resolution given by the size of the source of the reference beam. Multiple reference beams, or uniformly redundant arrays have been used to increase the strength of the reference beam. Can this technique be extended to 3D?

Ptychography has been shown to provide a robust "engine" for 2D image reconstruction without the need for finite support. The first 3D reconstruction using this technique was recently demonstrated; the potential impact and limitations of extending this technique to 3D should be explored.

The premise of the workshop is that it is technologically feasible to extend the resolution in X-ray microscopy well beyond the limitations in X-ray optics, and optics-based instruments. The workshop will examine this premise from the standpoint of what might be needed to bring these techniques to routine use.

The workshop will bring together both practitioners of the various coherence-based imaging techniques and scientists from various fields using alternative techniques to analyze samples, to assess the scientific needs, to discuss the technical challenges, distill the best approaches, and to establish the requirements for a successful program.

Some of the technical challenges that need to be addressed could include:

. Suppression of harmonics form the source
. Dynamic range in the detector
. Precision positioning, alignment and rotation of the sample
. Efficient data handling, assembly and reconstruction algorithms
. Pre-and post-exposure assessment of sample quality, suitability, and radiation damage
. Understanding the effects of background from Compton scattering and other sources
. Effects due to the missing data where either a beam-stop is used, or where the substrate interferes with complete 360 degree tomographic data collection
. In the case of diffraction microscopy, determining the finite support, and minimizing the effect of the specimen mount

[Invitation Letter より]

The specific purpose of this workshop is to assess and discuss the new scientific opportunities based on coherent imaging that will be opened up by ERLs and USRs. **The intent of the workshop is not to focus on experiments that have already been done, but rather to brainstorm and discuss experiments that you would like to do, but are impractical with existing sources.** The talks are intended to be short, future-oriented, and each followed by ample discussion time. The goal is to present innovative ideas, new approaches, and out-of-the-box thinking. The workshop will include a poster session where latest results may be presented and augmented by handouts focusing especially on the new science that might be expected from ERLs or USRs.

プログラム（1日目）

イントロダクション

Don Bilderback (Cornell University)

"Energy Recovery Linac (ERL) and Ultimate Storage Ring (USR) Properties "

Sol Gruner (Cornell University)

"X-ray Detectors: State-of-the-art & Future Possibilities "

Qun Shen (National Synchrotron Light Source II)

"New Opportunities with Hard X-ray Diffraction Limited Sources “

回折顕微法、ホログラフィー

Garth Williams (Linac Coherent Light Source)

"Coherent Imaging Without a Laser: getting the most bang for your electrons"

Jim Fienup (University of Rochester)

"X-ray Coherent Diffractive Imaging with an Extended Reference"

Stefano Marchesini (Lawrence Berkeley National Laboratory) ALS COSMIC

"High-efficiency Fourier Holography with Uniformly Redundant Arrays “

プログラム（1日目）続き

生物学応用

Chris Jacobsen (Northwestern University) APS 2-ID

"Imaging With Coherent Beams: let's not do it in a vacuum"

Chae Un Kim (Cornell University)

"Cryopreservation of Structural Integrity under High Pressure "

David Shapiro (National Synchrotron Light Source II) NSLS II

"High-resolution Imaging of Biological Specimens "

Yoshinori Nishino (Hokkaido University) -

"Imaging Cellular Organelles "

John Miao (University of California, Los Angeles) -

"Three-Dimensional Coherent Diffraction Imaging of Materials and Cells

プログラム（2日目）

材料科学応用、タイコグラフィー

Ivan Vartaniants (Deutsches Elektronen-Synchrotron)
"Coherent Diffractive Imaging and Determining Structural Properties from Cross-correlation Analysis "

Ross Harder (Advanced Photon Source) APS ID-34-C
"Probing Strain and Defects in Single Crystals with Coherent X-ray Diffraction"

Pierre Thibault (Technische Universität München) SLS cSAXS
"Ptychography in 2D and 3D"

Harald Ade (North Carolina State University)
"Spectromicroscopy, Resonant Scattering, Possible Extensions to Ptychographic Imaging"

Oleg Shpyrko (University of California, San Diego)
"Magnetic Domains and Dynamics"

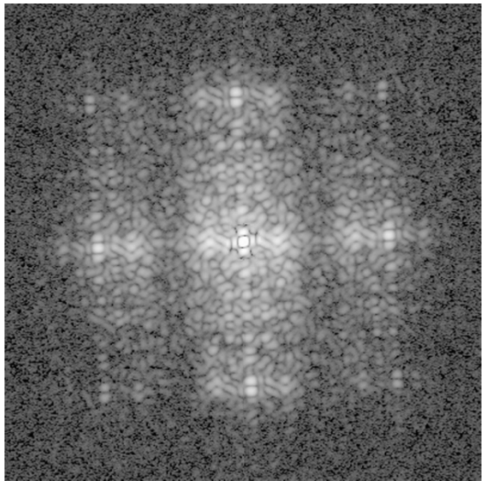
Ian McNulty (Advanced Photon Source)
"Resonant Coherent X-ray Imaging"

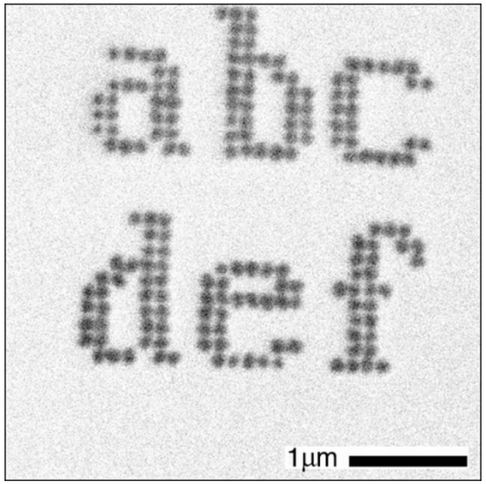
Breakout sessions and summary writing, roundup workshop

X-ray Diffraction Microscopy

"Extending the methodology of x-ray crystallography to allow imaging of micrometer-sized non-crystalline specimens"

Gold dots (~100 nm diameter, 80 nm thick)
on silicon nitride membrane





Diffraction Pattern (log scale)
l=17 Å

Reconstructed Image
spatial resolution: 75 nm

J. Miao, P. Charalambous, J. Kirz & D. Sayre, Nature 400, 342 (1999).

歴史的背景

回折像のオーバーサンプリングによる位相回復の可能性を示唆

D. Sayre, Acta Cryst. **5**, 843 (1952).

反復法による位相回復

W. Gerchberg & W. O. Saxton, Optik 35, 237 (1972).
J. R. Fienup, Applied Optics **21**, 2758 (1982).

周期構造を持たない試料に対するX線回折顕微法のアイデア

D.Sayre, in *Direct Methods of Solving Crystal Structures*,
ed. H. Schenk (Plenum Press, 1991) p. 353.

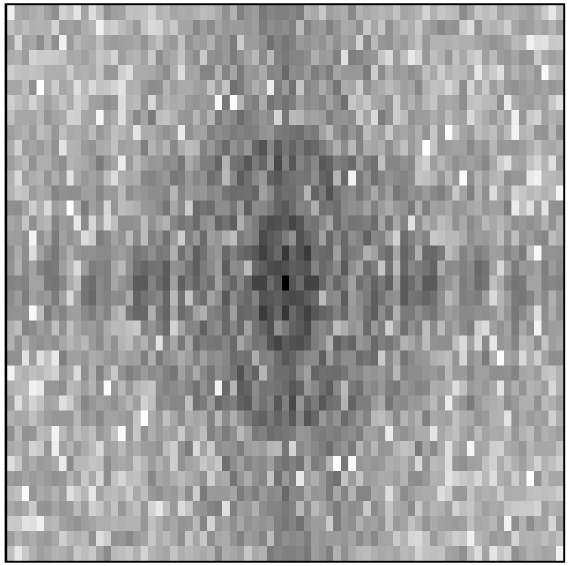
初めての実験

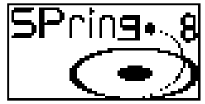
J. Miao, P. Charalambous, J. Kirz & D. Sayre, Nature **400**, 342 (1999).

Oversamping

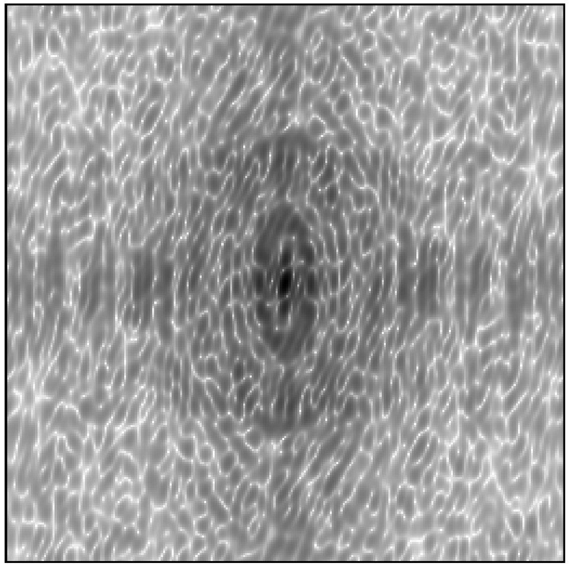
Diffraction Pattern (log scale)

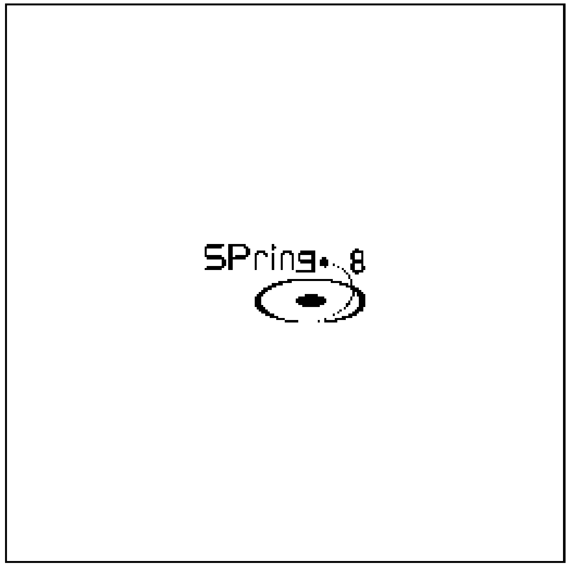
Image





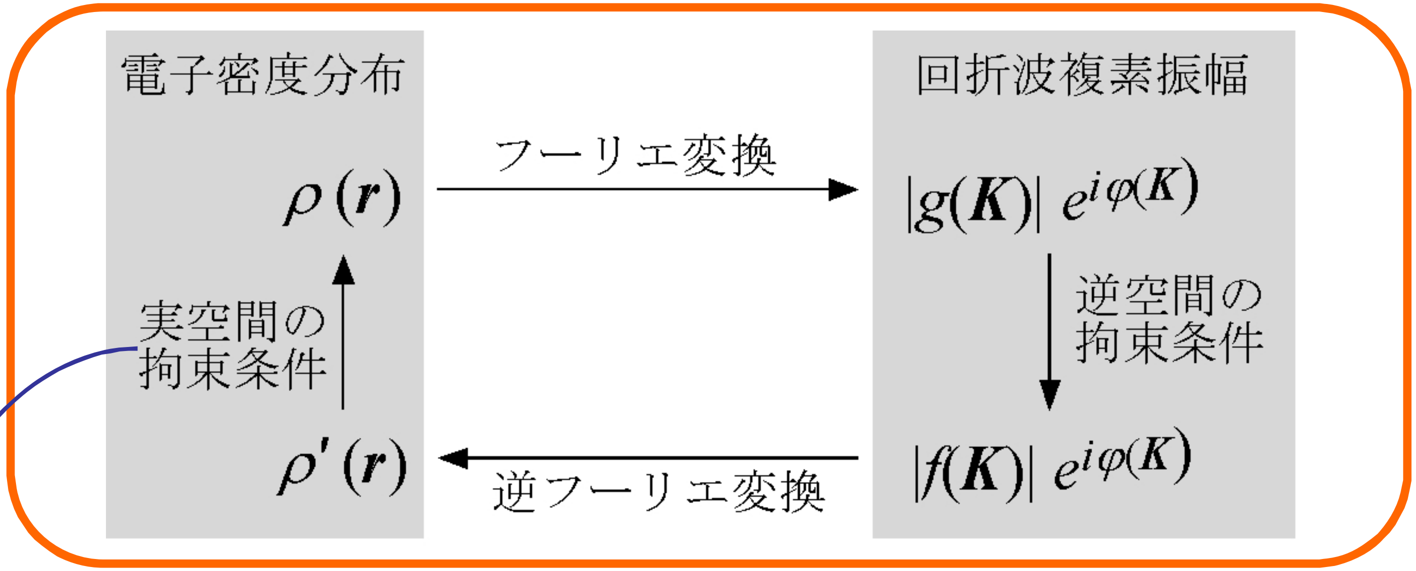
oversampling





no-density region around image

反復法による位相回復



$|f(\boldsymbol{K})|$: 実験で得られた回折強度分布

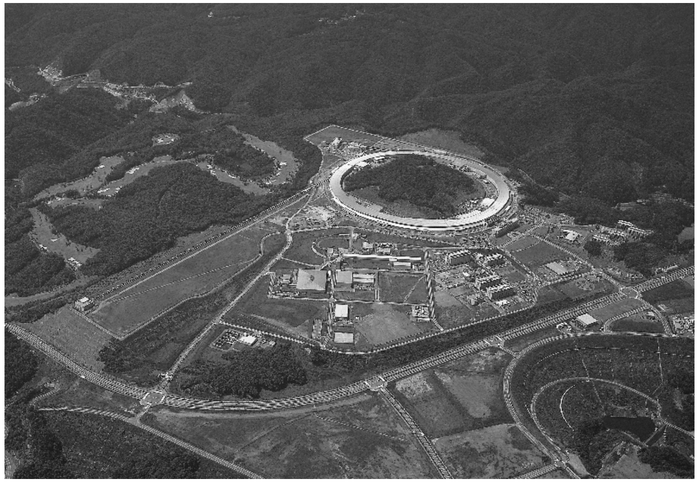
Hybrid Input-Output (HIO) Algorithm

$$\rho_{n+1}(\boldsymbol{r}) = \begin{cases} \rho'_n(\boldsymbol{r}), & (r \in S) \wedge (\rho'_n(\boldsymbol{r}) \geq 0) \\ \rho_n(\boldsymbol{r}) - \beta\rho'_n(\boldsymbol{r}), & \text{otherwise} \end{cases}$$

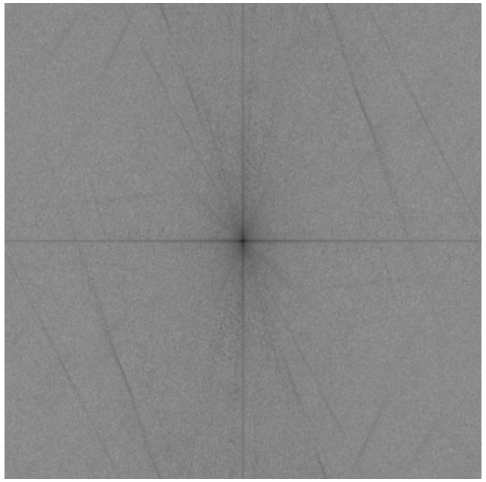
S: Support, β : between 0.5 and 1

R.W. Gerchberg and W.O. Saxton, Optik (Stuttgart) **35**, 237 (1972).
J.R. Fienup, Appl. Opt. **21**, 2758 (1982).

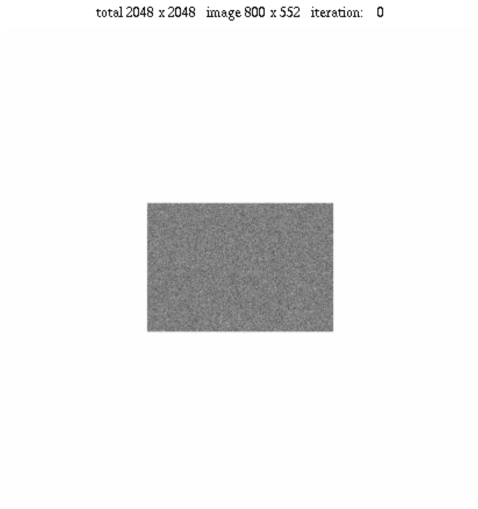
Simulation of the HIO algorithm



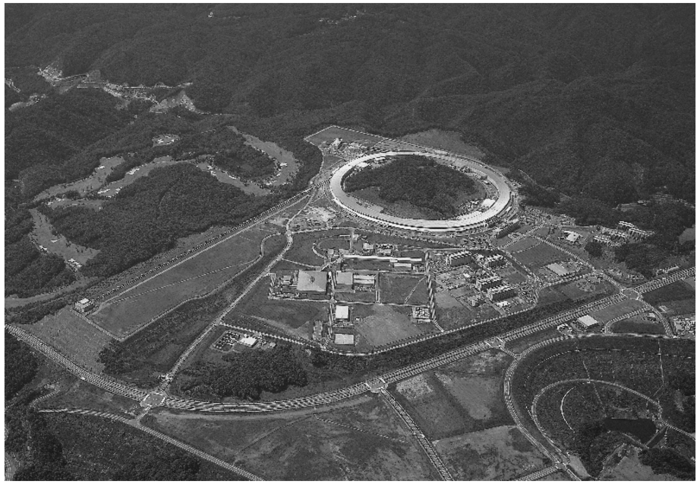
Sample Image



Calculated Diffraction Pattern



Iterative Image Reconstruction

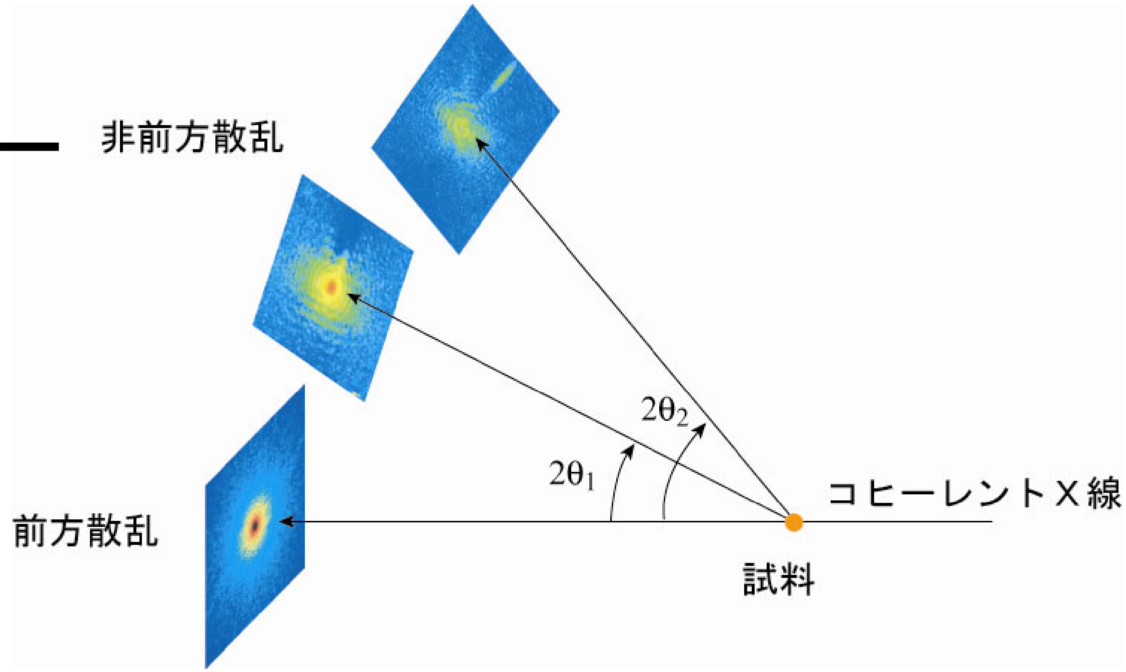


Reconstructed Image after 5000 Iterations

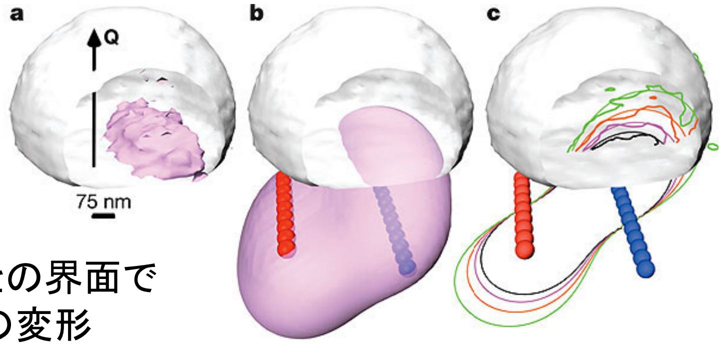
ナノ結晶中のひずみ場分布の可視化

電子密度＋ひずみ場 ←

電子密度 ←



鉛のナノ結晶中のひずみ場の可視化



SiO_2基板との界面で
最大0.5Åの変形

波長1.38Å

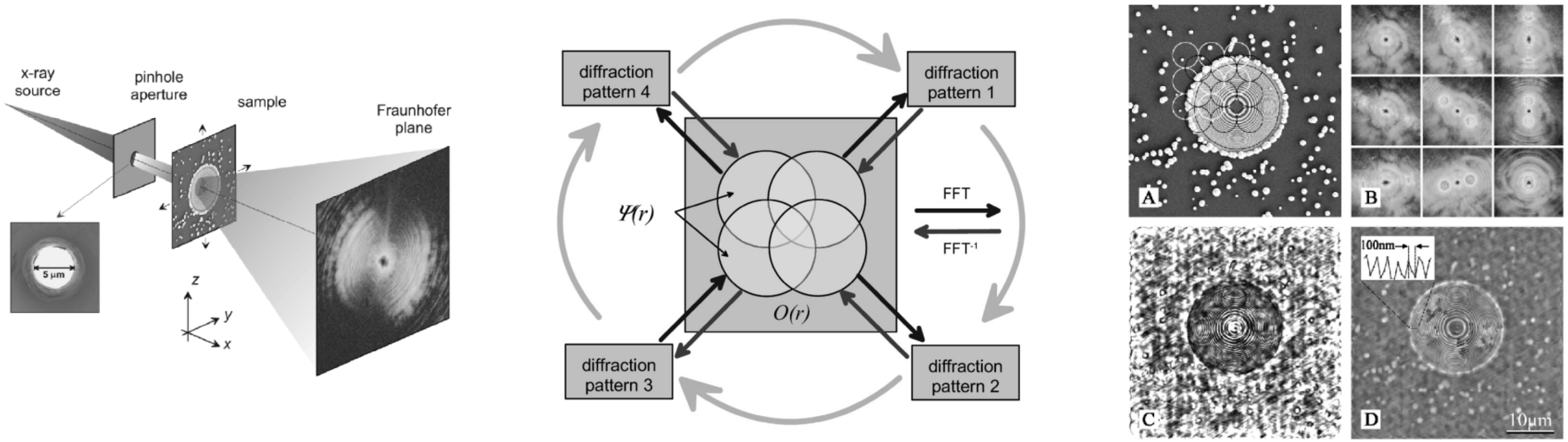
空間分解能: 45 nm

34-ID-C, APS

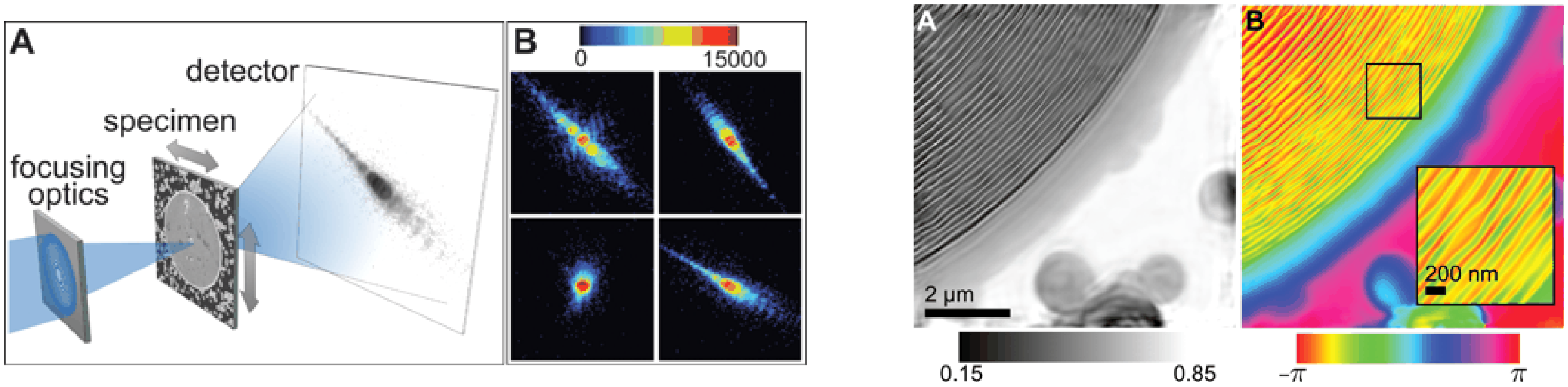
M.A. Pfeifer *et al.*, Nature **442**, 63 (2006)

ロンドン大学 ユニバーシティー・カレッジ のIan Robinsonらのグループが米国APSで実験を行っている

Ptychography



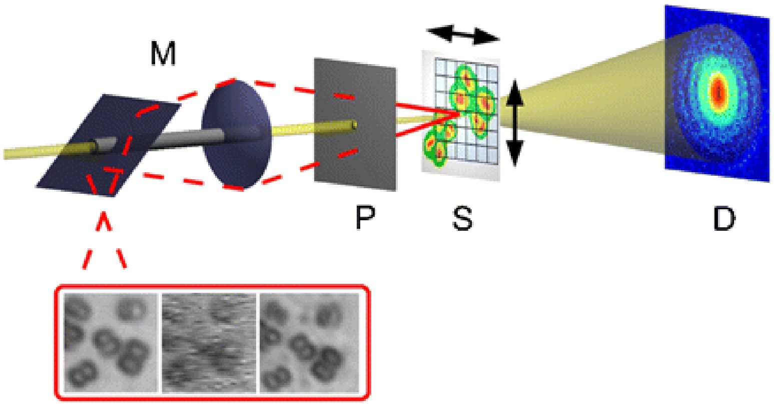
Rodenberg et al., PRL **98,** 034801 (2007)

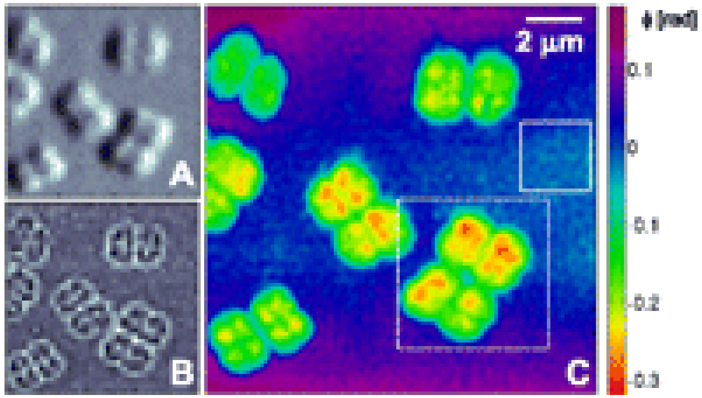


P.Thibault, et al., Science 321, 379 (2008)

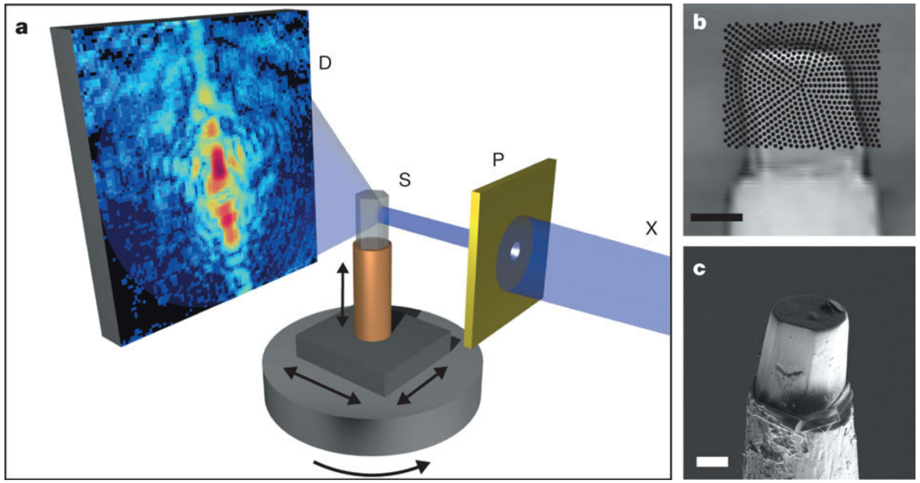
Ptychography

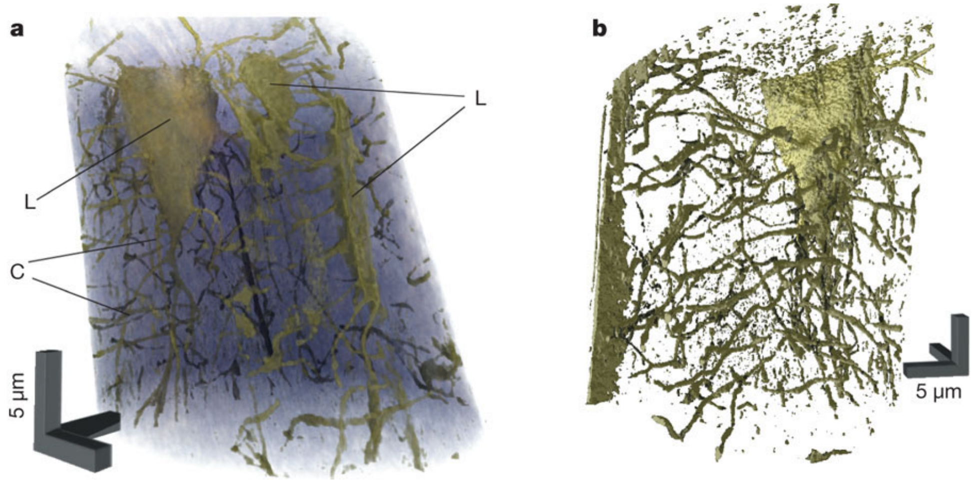
Unstained and unsliced freeze-dried Deinococcus radiodurans





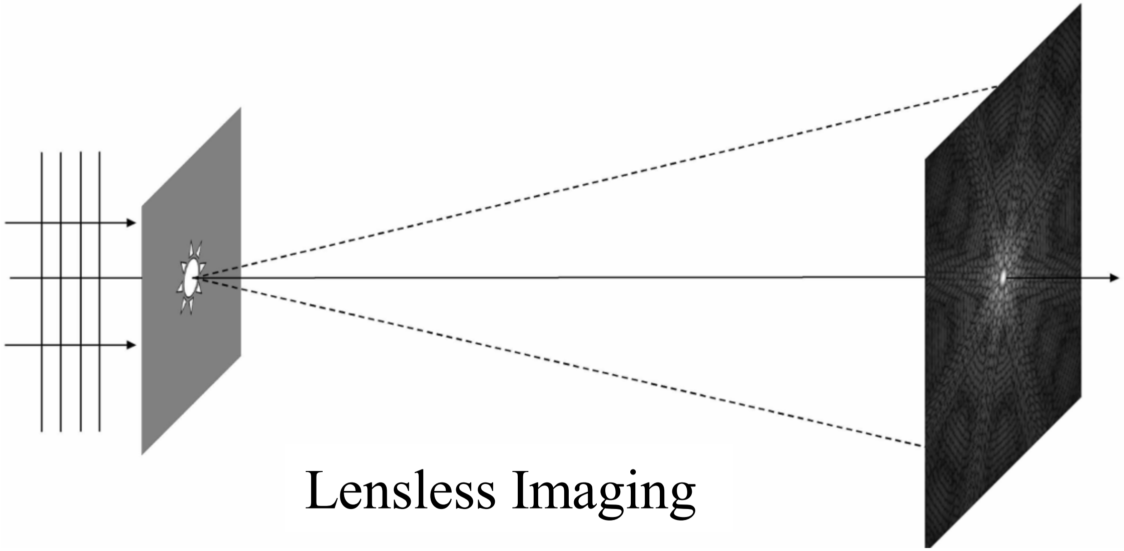
K. Giewekemeyer et al., PNAS **107,** 529 (2010)

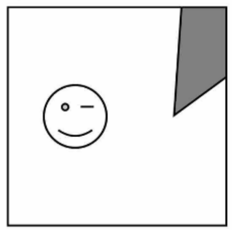




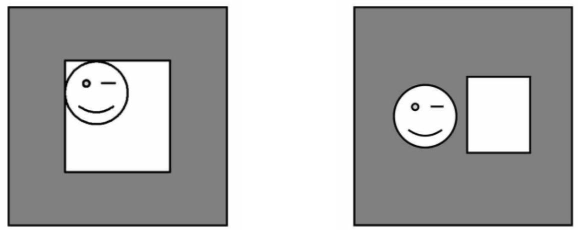
M. Dierolf et al., Nature 467, 436 (2010)

HERALDO (holography with extended reference by autocorrelation linear differential operation)





Obscuring-Tip Reference (as an example)
HERALDO



without Reference
Diffraction Microscopy

Iterative phase-retrieval for reconstruction

Point Reference
Fourier Transform Holography

Rectangular-Hole Reference
Holography by Podorov, Pavlov & Paganin

Non-Iterative Reconstruction

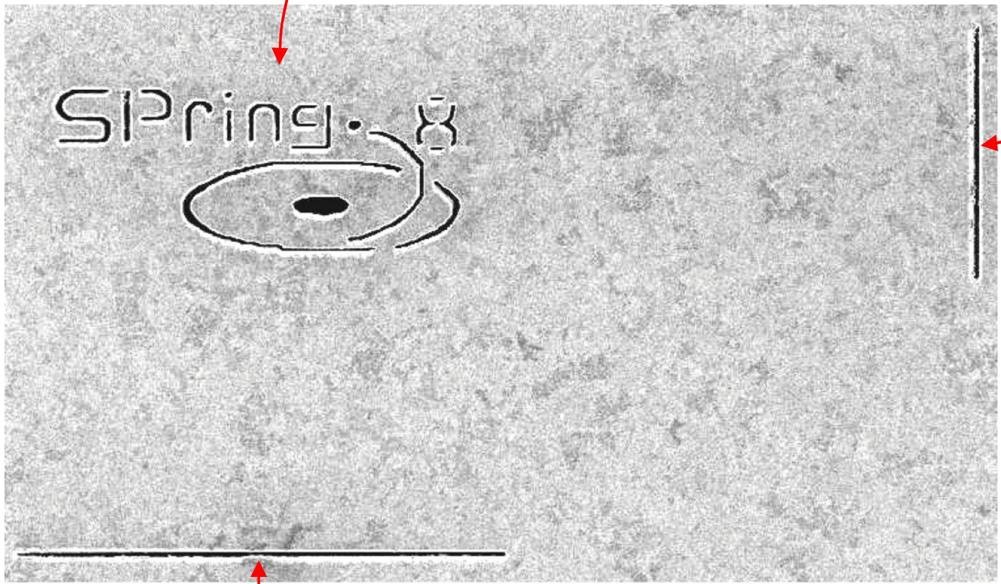
M. Guizar-Sicairos and J. R. Fienup, Optics Express **15**, 17592 (2007).

Femtosecond Snapshot imaging with EUV FEL at SCSS

Holography with Extended Reference by
Autocorrelation Linear Differential Operation (HERALDO)

Sample (SPring-8 logo)

Vertical Reference Slit



Horizontal Reference Slit

50μm

Scanning Ion Microscope (SIM) Image

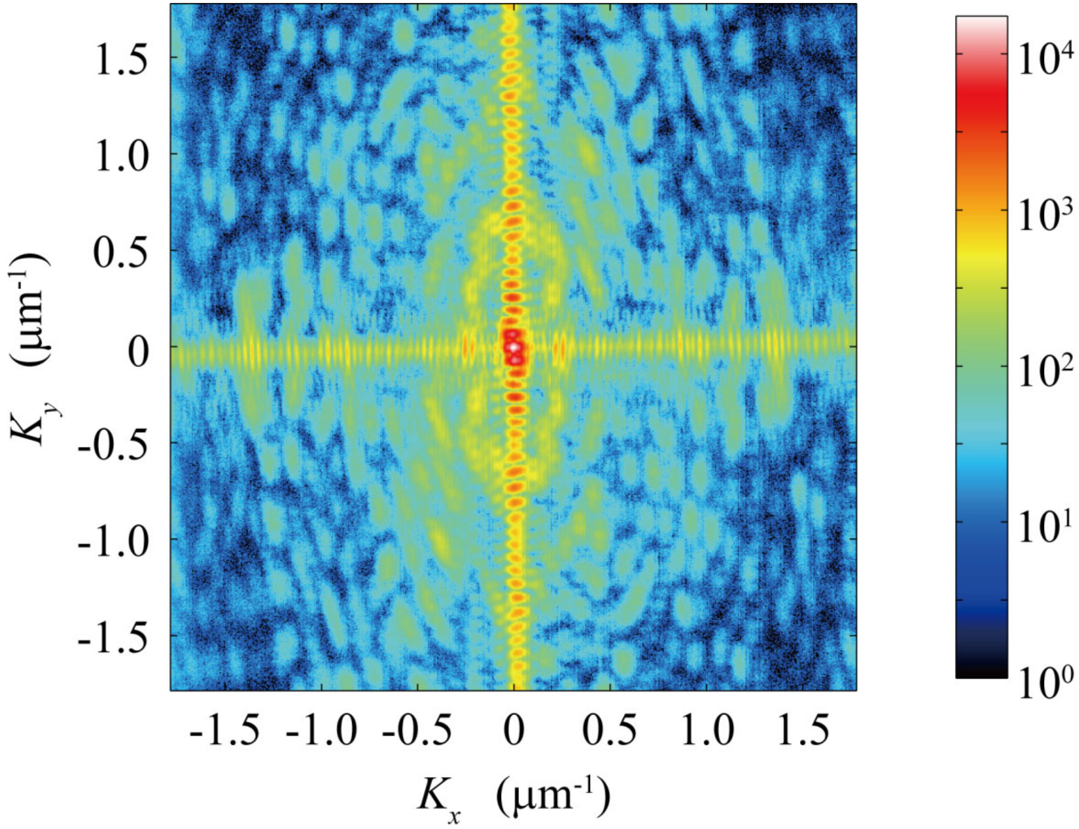
Test pattern milled by FIB
~ 800 nm thick Au deposited
on 100 nm thick Si_3N_4 membrane

Horizontal Slit:
width: ~ 0.9 μm, length: ~ 112 μm
Vertical Slit:
width: ~ 1.2 μm, length: ~ 57 μm

Y. Nishino *et al*., Appl. Phys. Express, 3, 102701 (2010).

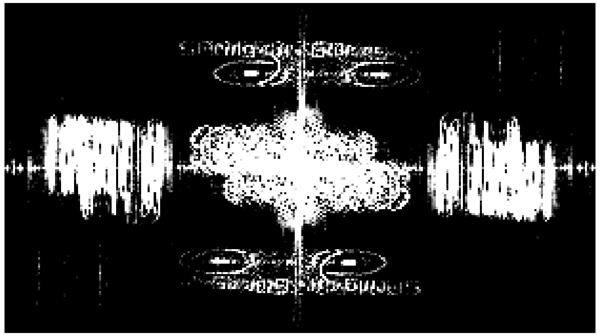
Femtosecond Snapshot imaging with EUV FEL at SCSS

Single-Shot Hologram (λ = 61 nm)

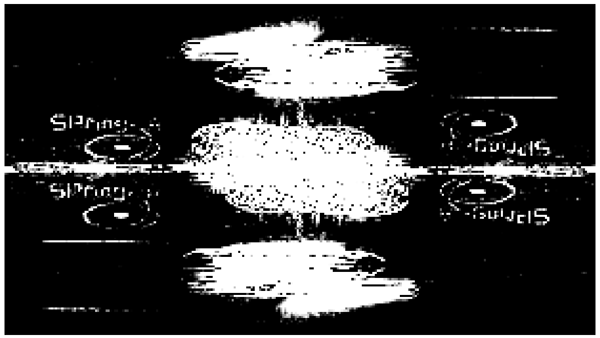


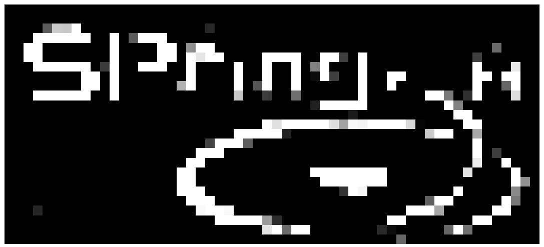
Reconstructed femtosecond snapshot images

Horizontal slit



Vertical slit

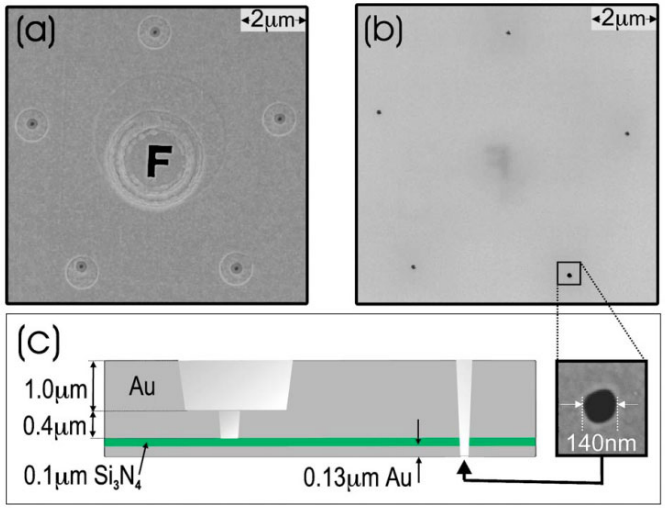




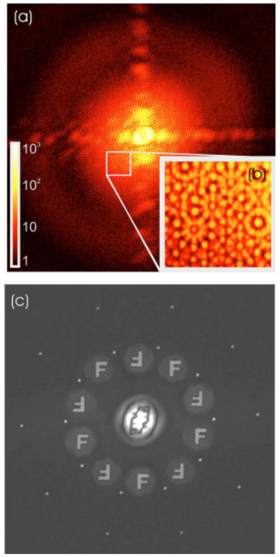
average of 4 copies

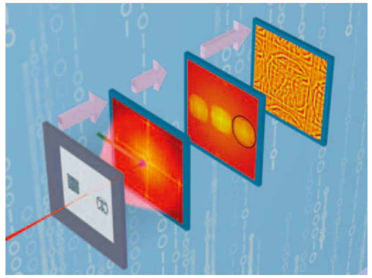
Y. Nishino *et al*., Appl. Phys. Express, 3, 102701 (2010).

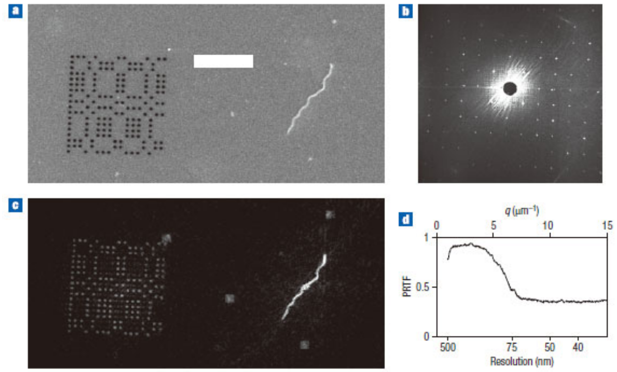
Holograpy



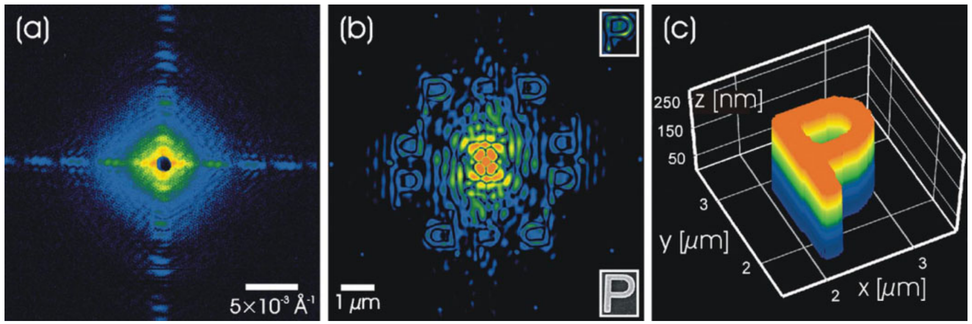
W. F. Schlotter *et al.*,
App. Phys. Lett. **89**, 163112 (2006)

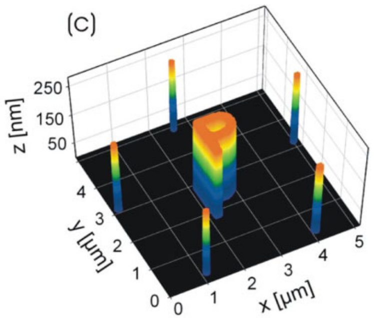






"Massively parallel X-ray holography",
S. Marchesini *et al.*,
Nature Photonics **2**, 560 (2008).

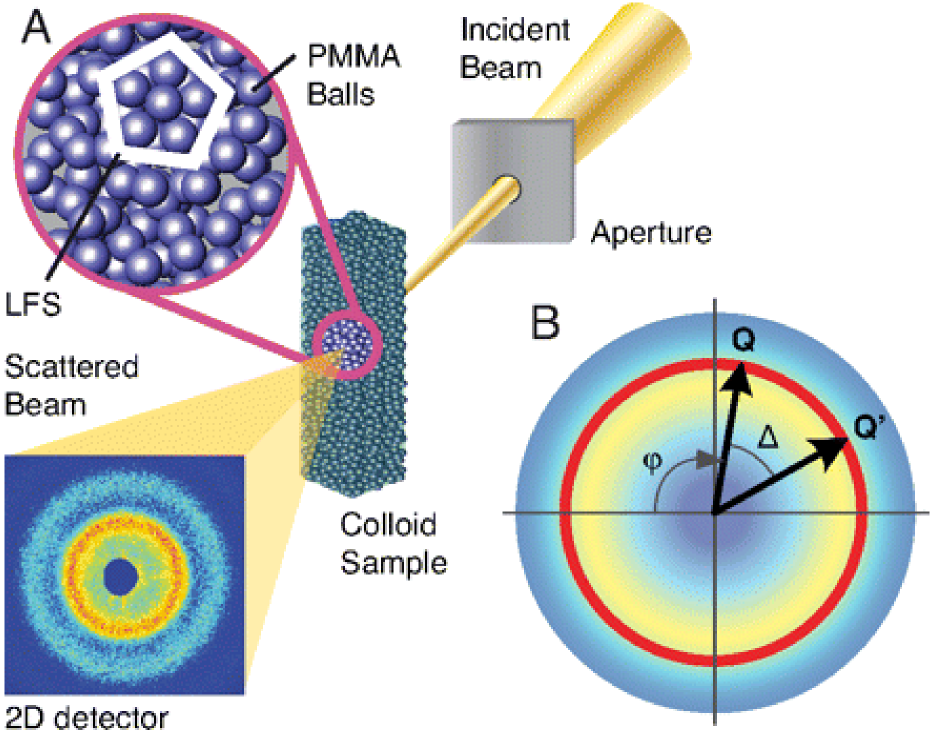


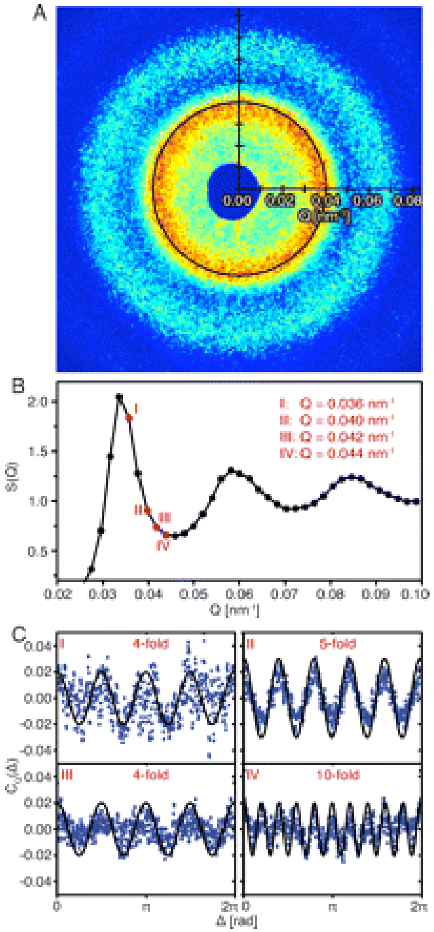


L-M. Stadler *et al.*,
Phys. Rev. Lett. **100**, 245503 (2008).

X-ray cross correlation analysis (XCCA)

hidden local symmetries in disordered matter

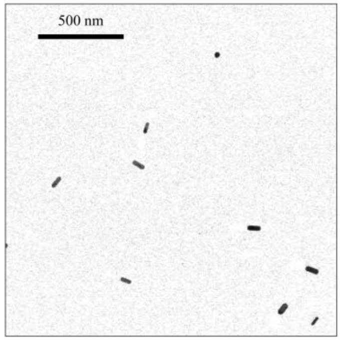




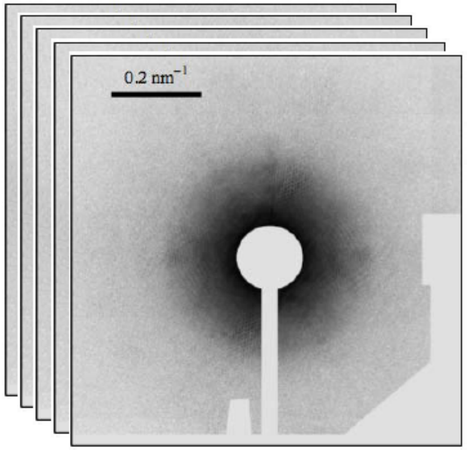
P. Wochner et al., PNAS 106, 11511 (2009)

Fluctuation Microscopy

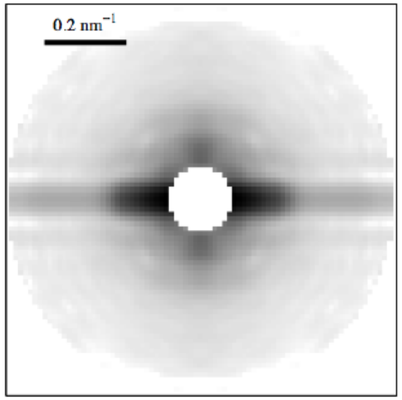
Z. Kam, Macromolecules (1977)



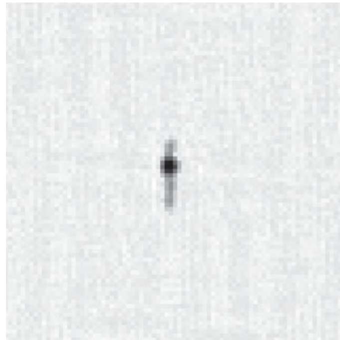
TEM image of randomly oriented 90×25 nm gold nanorods



many snapshot patterns, each with many disordered nanorods



Reconstruction of the *single nanorod* diffraction pattern



Reconstructed Image of gold nanorods

D. K. Saldin *et al*., PRL 106, 115501 (2011)

"The cross-correlation scheme has a disadvantage relative to the single-shot scheme both because of restrictions in the processing and the smaller number of photons scattered per particle."

V. Elser: arXiv:1007.3777v1

プログラム（1日目）

イントロダクション

Don Bilderback (Cornell University)
"Energy Recovery Linac (ERL) and Ultimate Storage Ring (USR) Properties "
Sol Gruner (Cornell University)
"X-ray Detectors: State-of-the-art & Future Possibilities "
Qun Shen (National Synchrotron Light Source II)
"New Opportunities with Hard X-ray Diffraction Limited Sources “

回折顕微法、ホログラフィー

Garth Williams (Linac Coherent Light Source)
"Coherent Imaging Without a Laser: getting the most bang for your electrons"
Jim Fienup (University of Rochester)
"X-ray Coherent Diffractive Imaging with an Extended Reference"
Stefano Marchesini (Lawrence Berkeley National Laboratory) ALS COSMIC
"High-efficiency Fourier Holography with Uniformly Redundant Arrays “

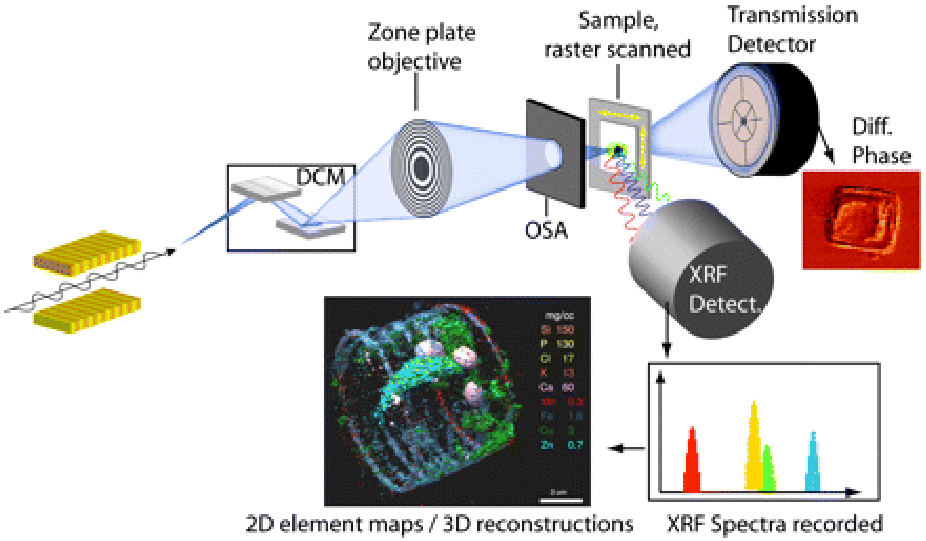
プログラム（1日目）続き

生物学応用

Chris Jacobsen (Northwestern University) APS 2-ID
"Imaging With Coherent Beams: let's not do it in a vacuum"
Chae Un Kim (Cornell University)
"Cryopreservation of Structural Integrity under High Pressure "
David Shapiro (National Synchrotron Light Source II) NSLS II
"High-resolution Imaging of Biological Specimens "
Yoshinori Nishino (Hokkaido University) -
"Imaging Cellular Organelles "
John Miao (University of California, Los Angeles) -
"Three-Dimensional Coherent Diffraction Imaging of Materials and Cells

Scanning X-ray Microscopy

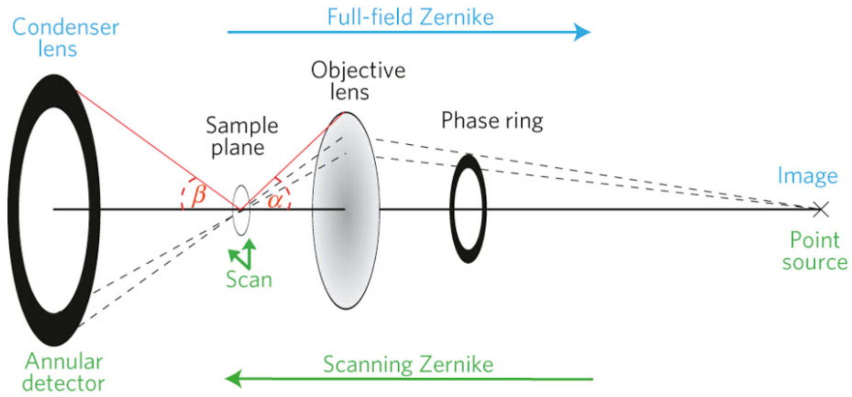
X-ray fluorescence microtomography



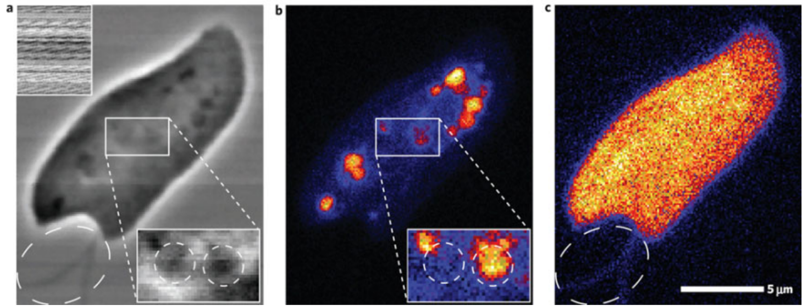
freshwater diatom *Cyclotella meneghiniana*

M. D. de Jonge et al, PNAS 107, 15676 (2010)

Scanning Zernike



Freshwater flagellate *Cryptomonas*



C. Holzner et al., Nature Physics 6, 883 (2010)

プログラム（2日目）

材料科学応用、タイコグラフィー

Ivan Vartaniants (Deutsches Elektronen-Synchrotron)

"Coherent Diffractive Imaging and Determining Structural Properties from Cross-correlation Analysis "

Ross Harder (Advanced Photon Source) APS ID-34-C

"Probing Strain and Defects in Single Crystals with Coherent X-ray Diffraction"

Pierre Thibault (Technische Universität München) SLS cSAXS

"Ptychography in 2D and 3D"

Harald Ade (North Carolina State University)

"Spectromicroscopy, Resonant Scattering, Possible Extensions to Ptychographic Imaging"

Oleg Shpyrko (University of California, San Diego)

"Magnetic Domains and Dynamics"

Ian McNulty (Advanced Photon Source)

"Resonant Coherent X-ray Imaging"

Breakout sessions and summary writing, roundup workshop

Imaging Cellular Organelles

Yoshinori Nishino

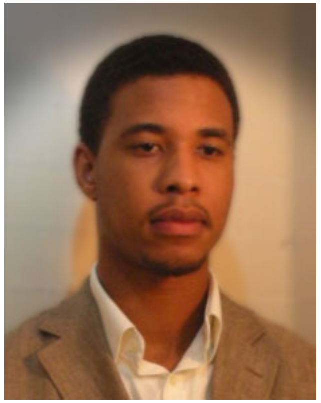
Laboratory of Coherent X-ray Optics,
Research Institute for Electronic Science,
Hokkaido University

Laboratory of Coherent X-ray Optics

- **Laboratory of Coherent X-ray Optics,**
  Research Institute for Electronic Science,
  Hokkaido University

since April, 2010

Professor
Yoshinori Nishino



Assistant Professor
Marcus Newton

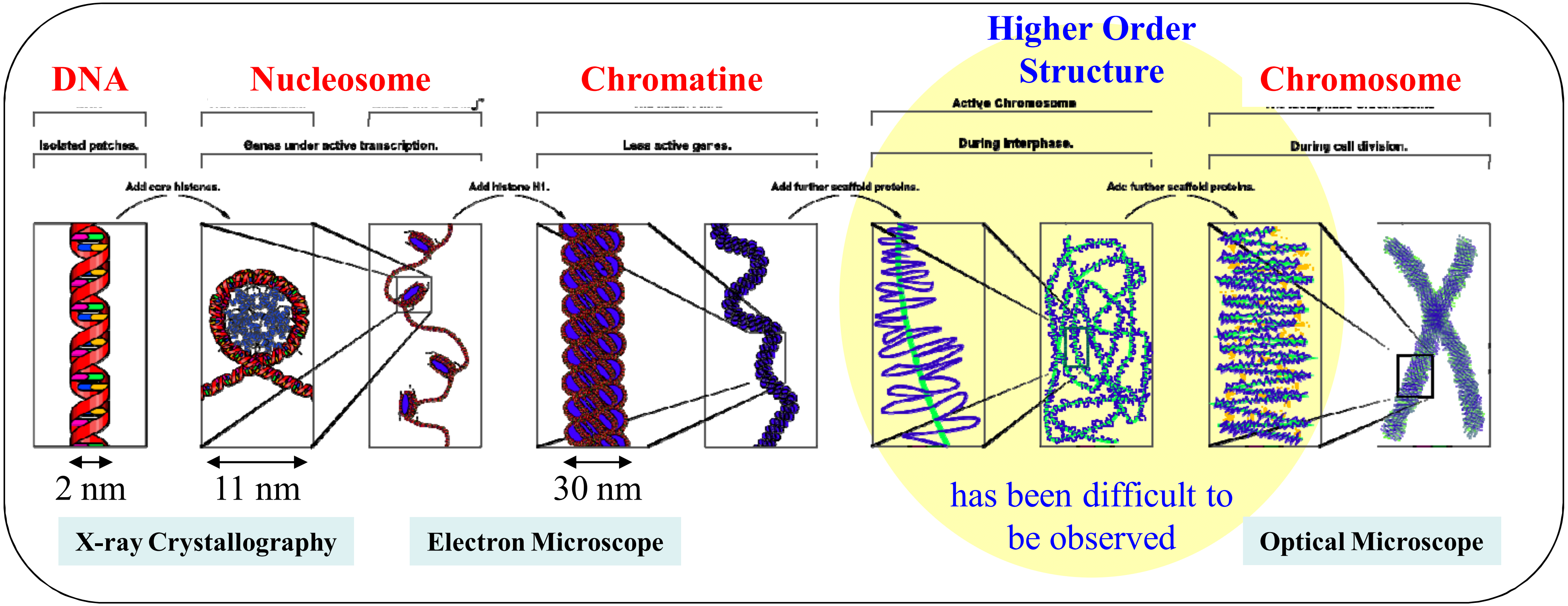
Assistant Professor
Takashi Kimura

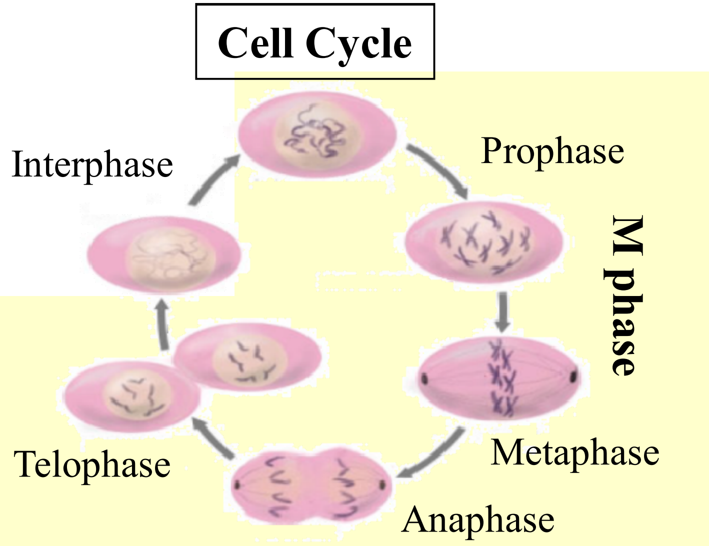
Undergraduate Students	· **Kei Soeta** · **Chie Nagase** · **Arata Mori**
Secretary	· **Kiyo Ssaki**

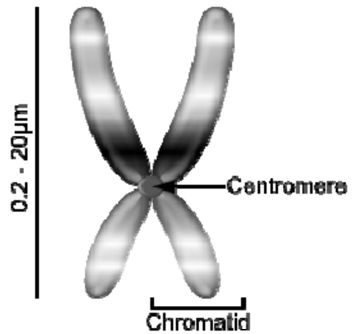
Outline

- Imaging Chromosome
- Discussions
  - Average Structure / Individual Structure
  - Focusing X-rays

Chromosomes





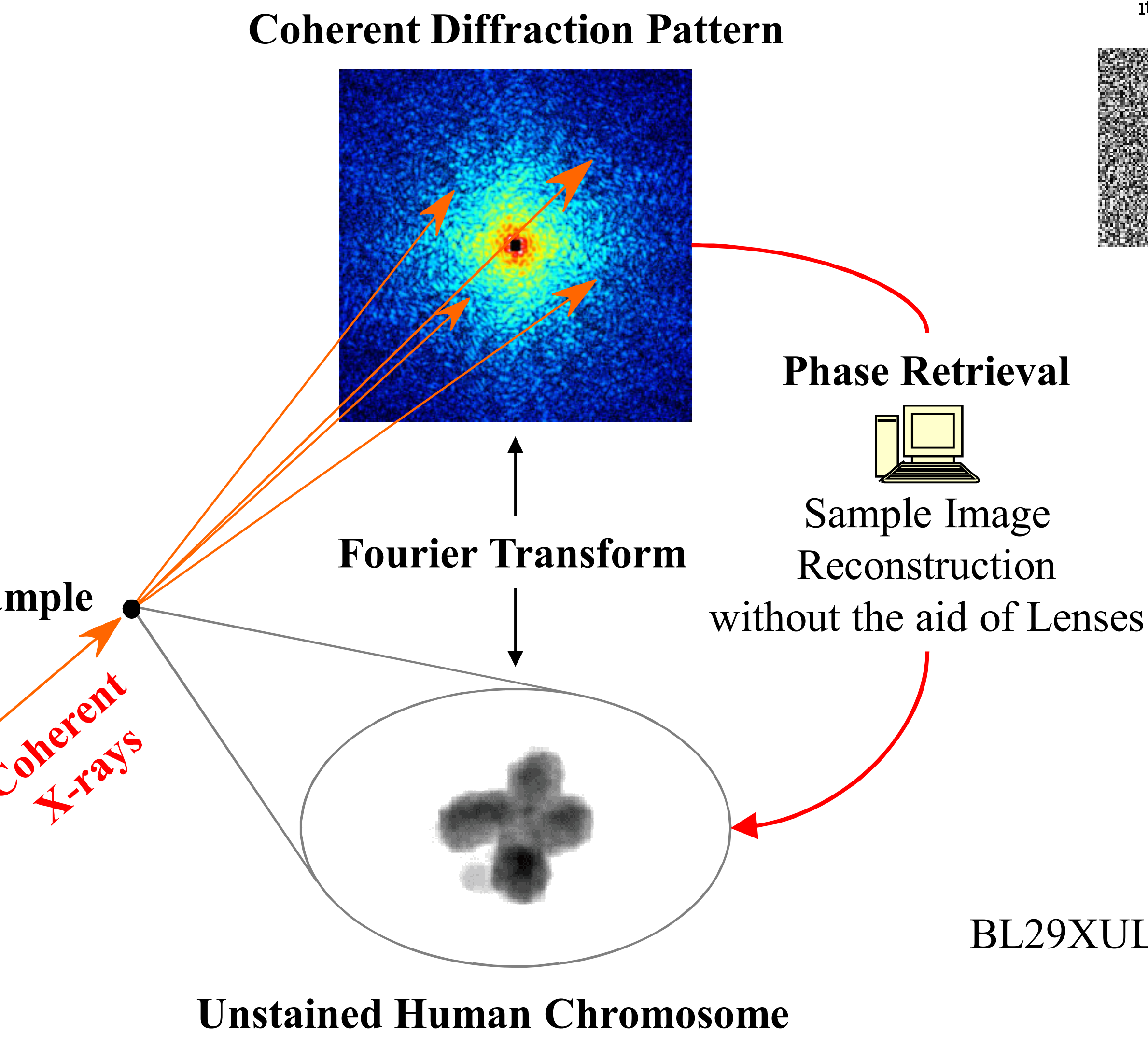


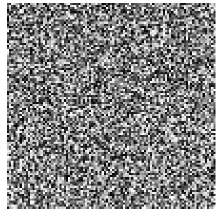
Four Arm Structure:
a pair of sister chromatids attached to each other at the centromere

Coherent X-ray Diffraction

Synchrotron Radiation

SPring-8 (Harima, Japan)





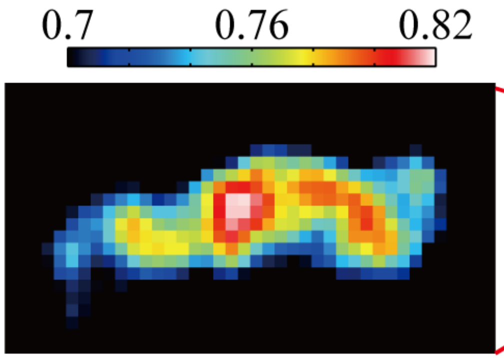
BL29XUL, SPring-8

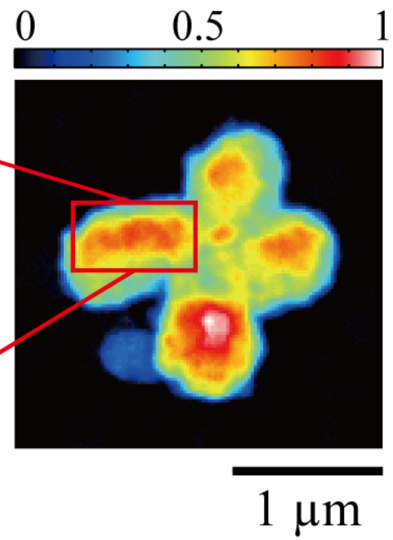
. Nishino, Y. Takahashi, N. Imamoto, T. Ishikawa, and K. Maeshima, Phys. Rev. Lett. **102**, 018101 (2009).

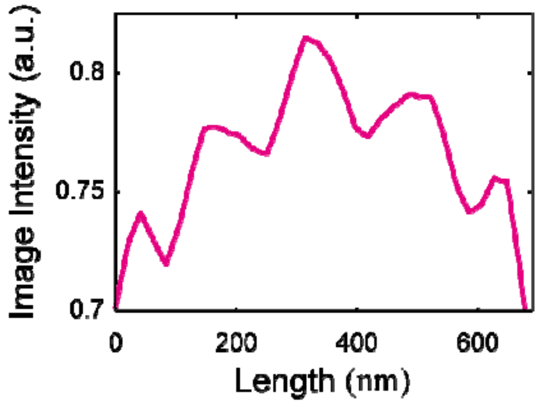
News Articles: nature Nature **457**, 238 (2009). physicstoday Physics Today, **62** (2), 20 (2009).

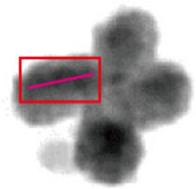
2D Observation of unstained human chromosome

Magnified Image









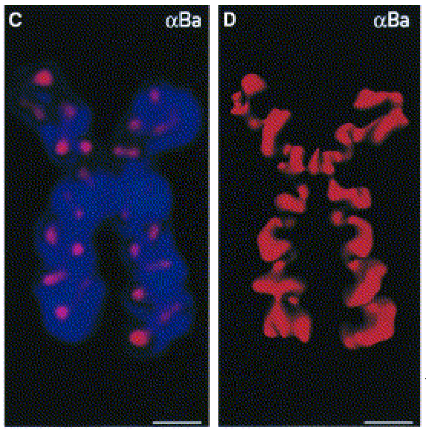
A different color scale is used to enhance the structure.

spatial resolution = 38 nm

First observation of axial structure in unstained chromosome

Y. Nishino *et al*., Phys. Rev. Lett. **102**, 018101 (2009).

Immunofluorescence Microscope Image



bar: 1 µm

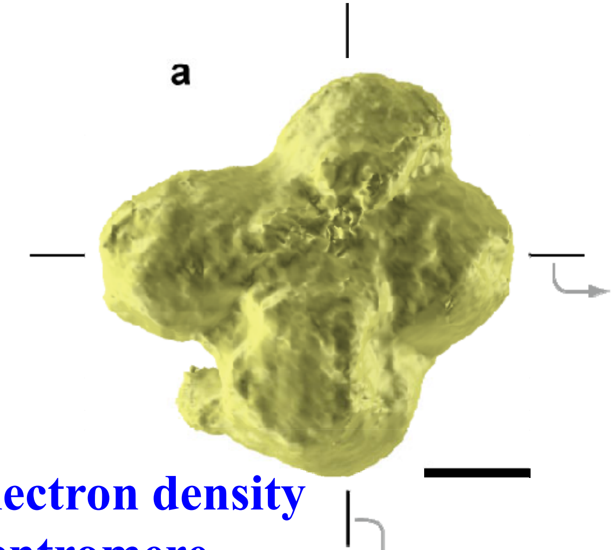
condensin antibody (red)

helically folded axial structure

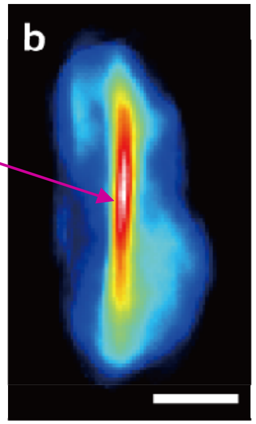
E. Boy de la Tour & U.K. Laemmli, Cell 55, 937 (1988)

K. Maeshima & U. K. Laemmli, Developmental Cell 4, 467 (2003)

3D observation of unstained human chromosome



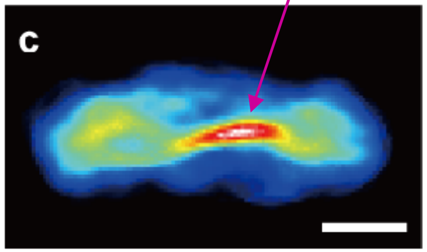
• **highest electron density around centromere**



centromere

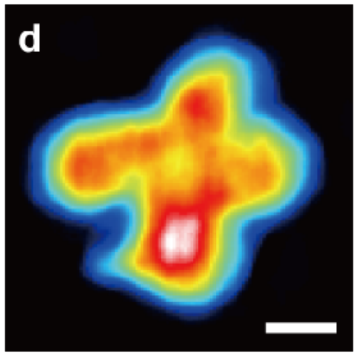
Vertical Slice

• **high density near axis**



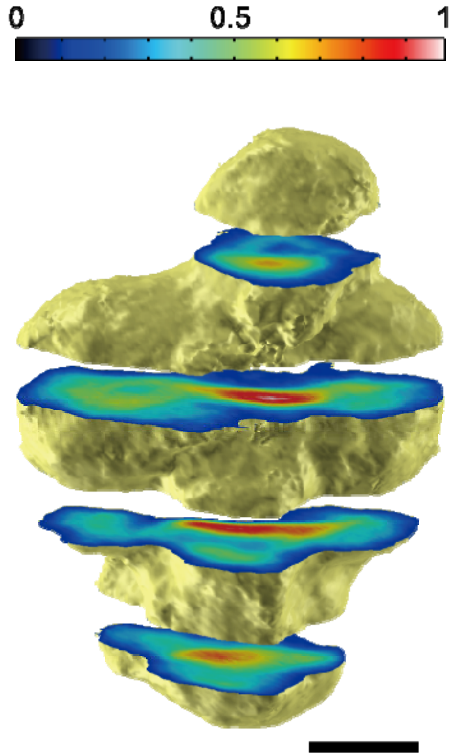
centromere

Horizontal Slice



Generated Projection Image

• **consistent with 2D reconstruction**



scale bars = 500 nm

spatial resolution = 120 nm

• **wavy (helical) structure was not observed**

First observation of a cellular organelle in 3D by using hard X-rays

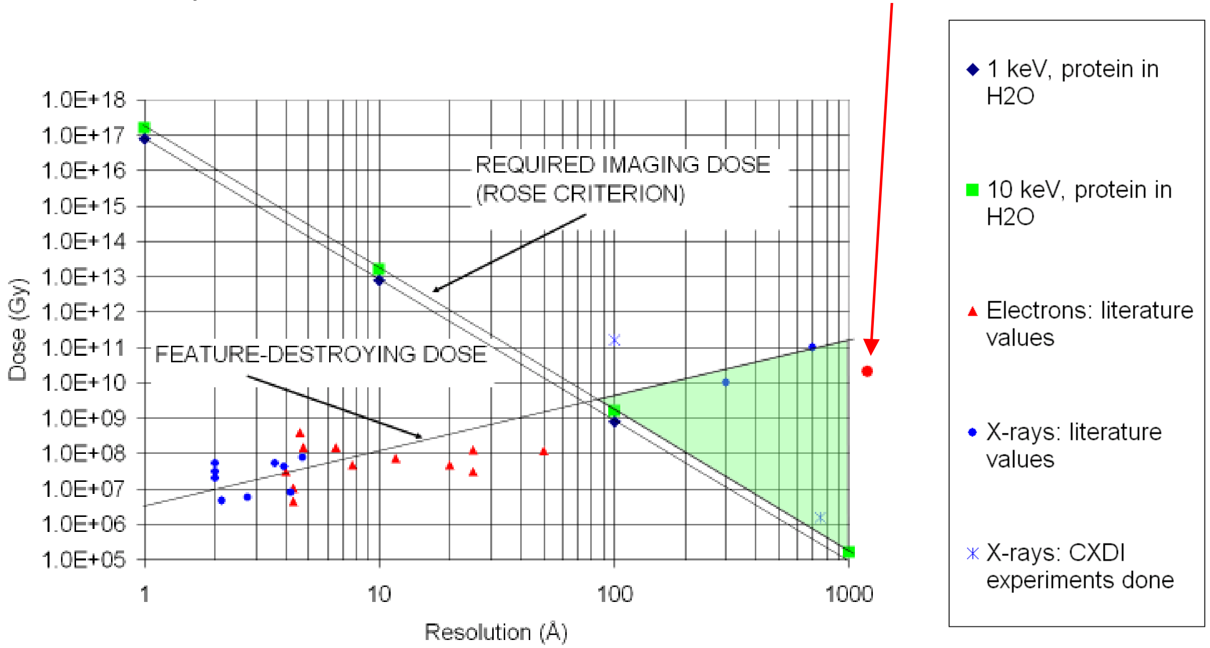
Y. Nishino *et al*., Phys. Rev. Lett. **102**, 018101 (2009).

Spatial Resolution

Estimated Dose

- Single diffraction: 4×10^{8} Gy
- 3D diffraction: 2×10^{10} Gy

our 3D reconstruction
(close to feature-destroying dose line)



M. R. Howells et al., J. Electron Spectrosc. Relat. Phenom. **170**, 4 (2009) .

S. Marchesini et al., Optics Express 11, 2344 (2003).

For higher spatial resolution

- cooling the sample cryogenically
- optimizing the dose
- improving the phase retrieval method

Average Structure / Individual Structure

Classical X-ray Methods

- X-ray Crystallography
- Small-Angle X-ray Scattering (SAXS)

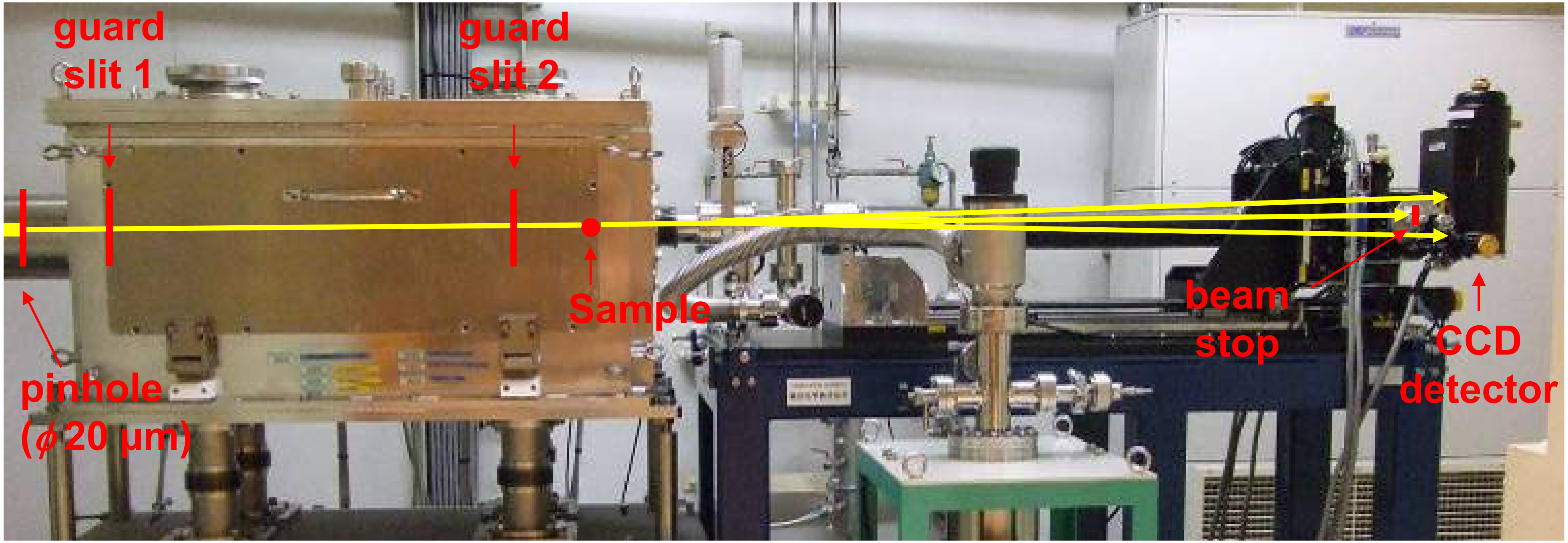
Average Structure

Less Radiation Dose to Each Individual Biological Object
→ Higher Resolution

Solution Scattering of Biomolecules

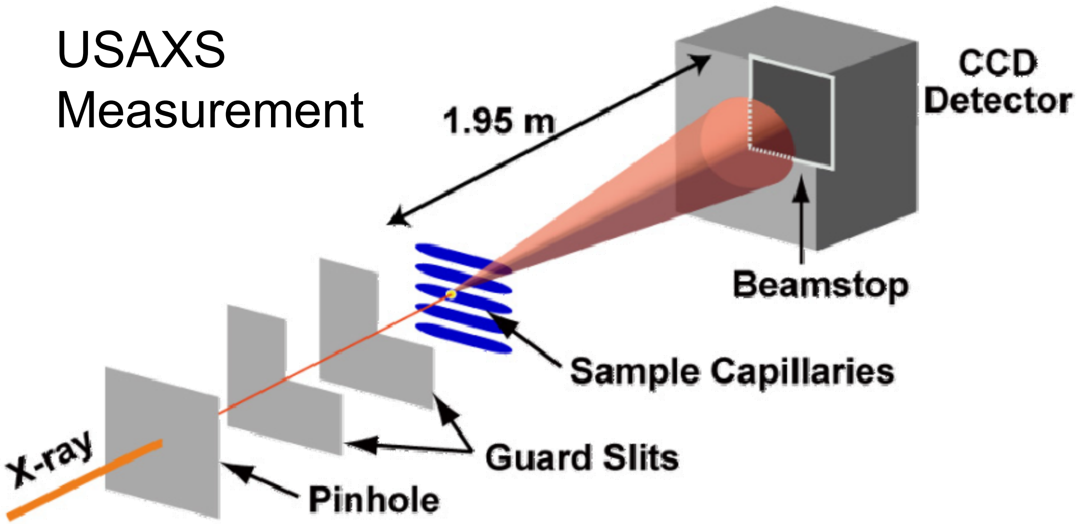
- for bio-molecules difficult crystallize
- nearly physiological conditions

Diffraction Microscopy Instrument at SPring-8



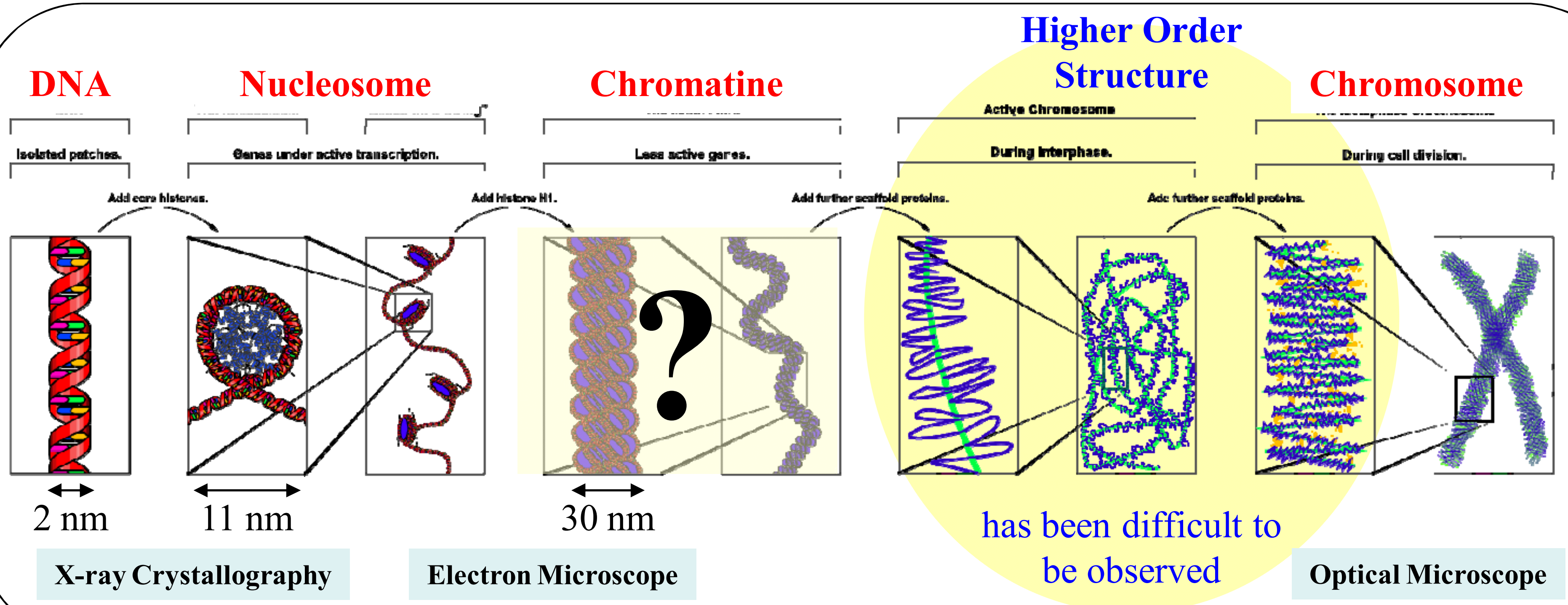
Hard X-ray Beamline BL29XUL

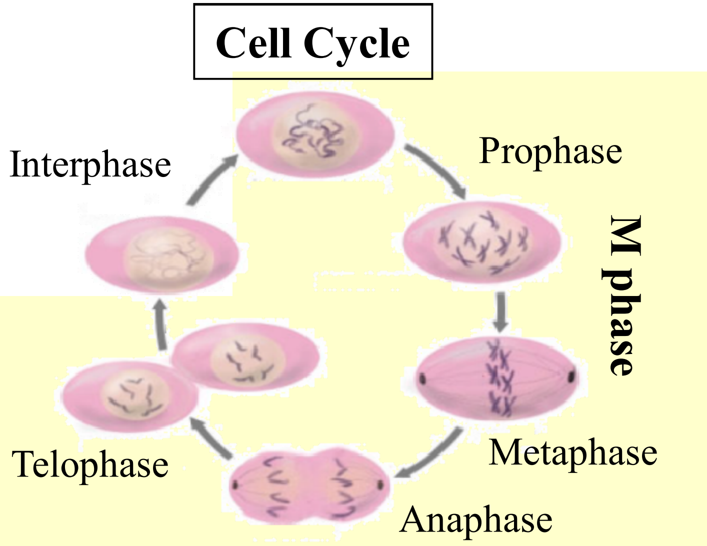
USAXS
Measurement

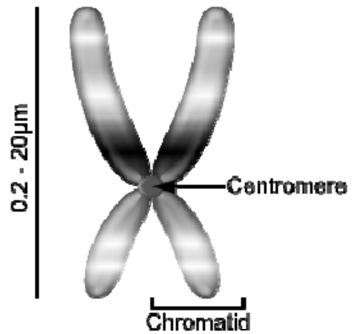


- **CCD Detector:**
  Princeton Instruments PI-LCX 1300
  1300 × 1340 Pixels
  Pixel Size: 20 μm × 20 μm
  Direct Illumination
  Deep Depletion

Chromosomes



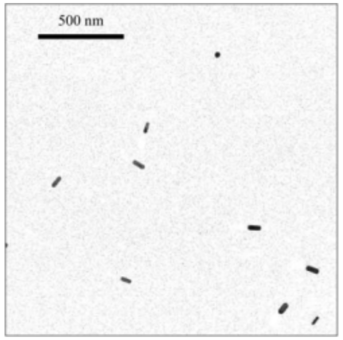




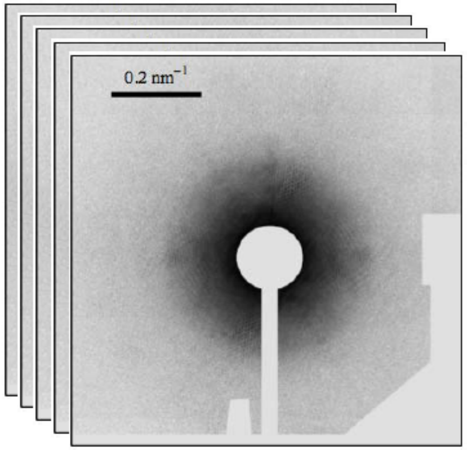
Four Arm Structure:
a pair of sister chromatids attached to each other at the centromere

Fluctuation Microscopy

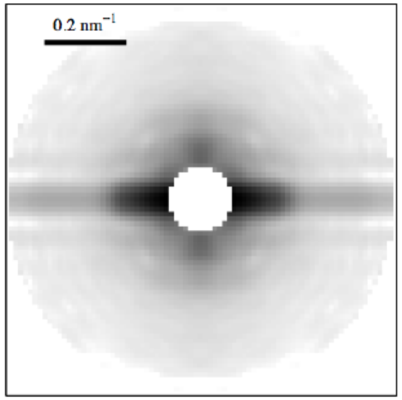
Z. Kam, Macromolecules, 10, 927 (1977)



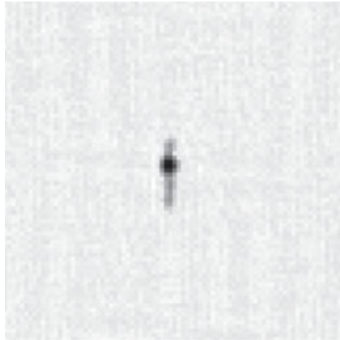
TEM image of randomly oriented 90 × 25 nm gold nanorods



many snapshot patterns, each with many disordered nanorods



Reconstruction of the *single nanorod* diffraction pattern



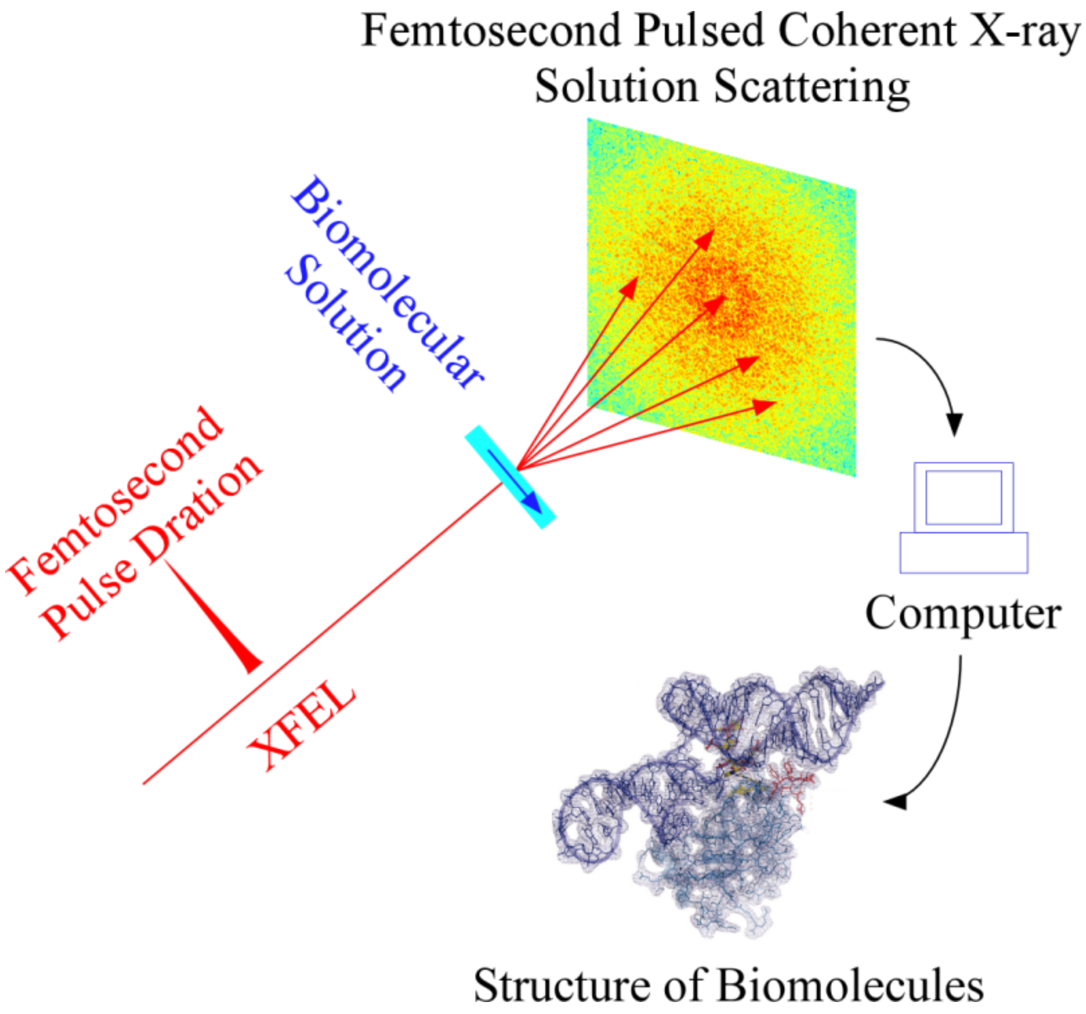
Reconstructed Image of gold nanorods

D. K. Saldin *et al*., PRL 106, 115501 (2011)

“The cross-correlation scheme has a disadvantage relative to the single-shot scheme both because of restrictions in the processing and the smaller number of photons scattered per particle.”

V. Elser: arXiv:1007.3777v1

Pulsed Coherent X-ray Solution Scattering



Conventional SAXS

Averaging

- Ensemble Average
- Time Average

⇩

Concentric Scattering Pattern
(1 Dimensional Data)

Solution Scattering with XFEL

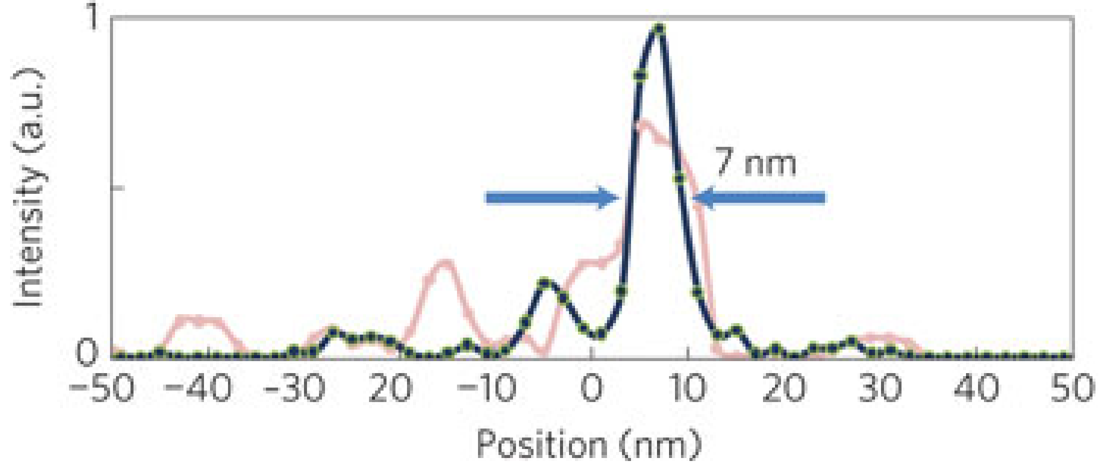
Unaveraged Data Acquisition

- Illuminating a Small Number of Moleculses
- Ultrashort Pulse Duration

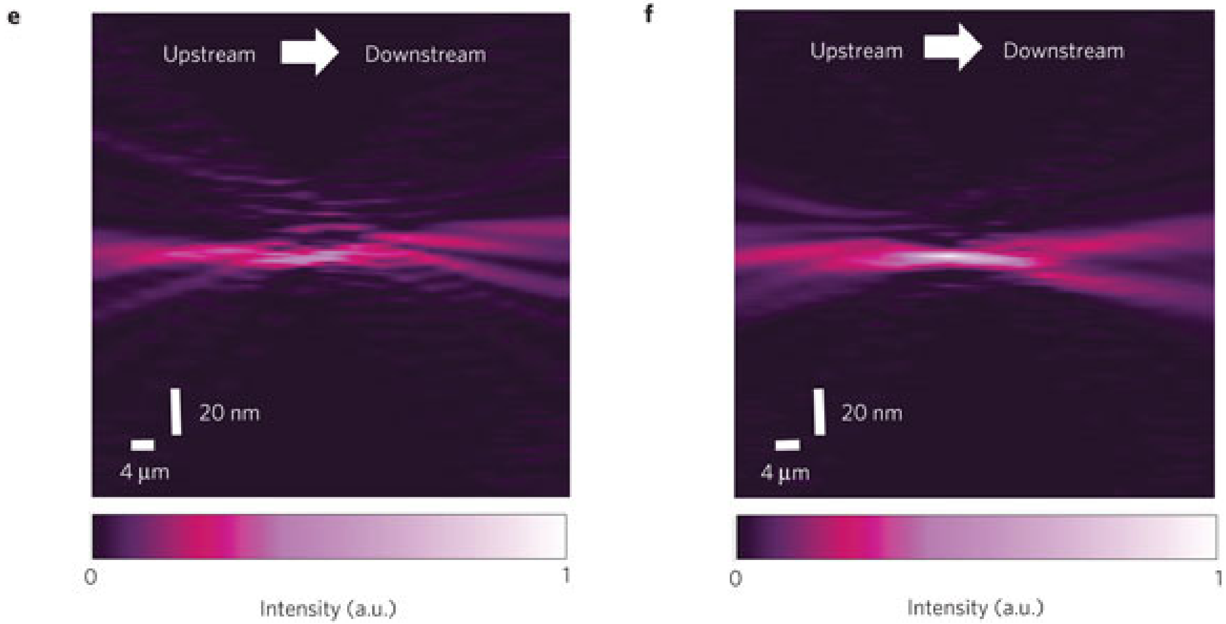
⇩

Speckled Coherent Scattering Pattern
(2-Dimensinal Data)

X-ray Focusing down to world's smallest 7 nm



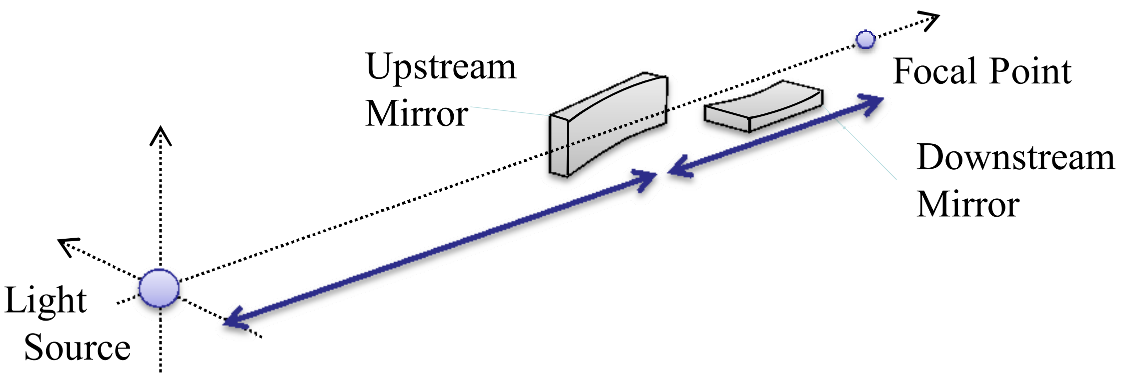
20 keV X-rays



nature physics

H. Mimura *et al*., Nature Physics **6**, 122 (2009).

Beamline Requirements



Geometrical Demagnification
= (Lens-to-Focus)/(Source-to-Lens)

Example:

Source Size: ~10 μm
Focal Size: ~10 nm
Working Distance ~100 mm
↓
Beamline Length ~100 m

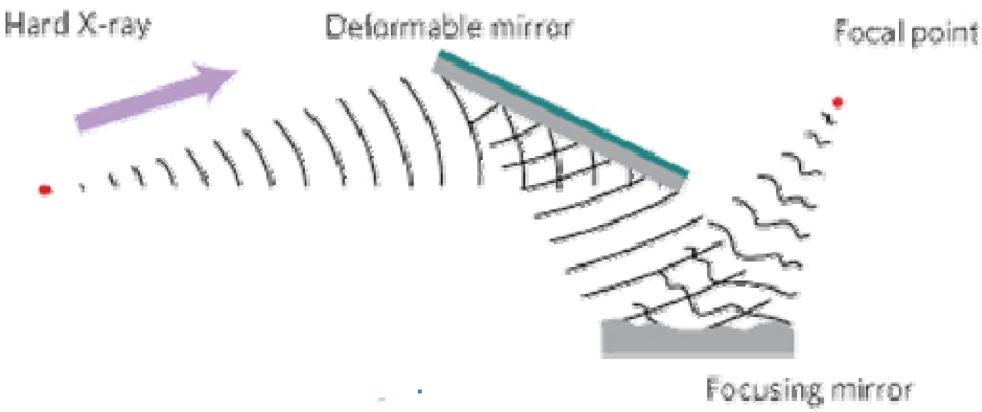
wave optics

$$\text{focal spot size} \sim \frac{\lambda}{\text{N.A.}}$$

ERL or URL:
Coherently Illuminate Focusing Mirrors without Loss

- Diffraction Limited Source both Vertically and Horizontally
- Long Beamline
- Stability

Adaptive Optical System for Hard-X-ray Focusing

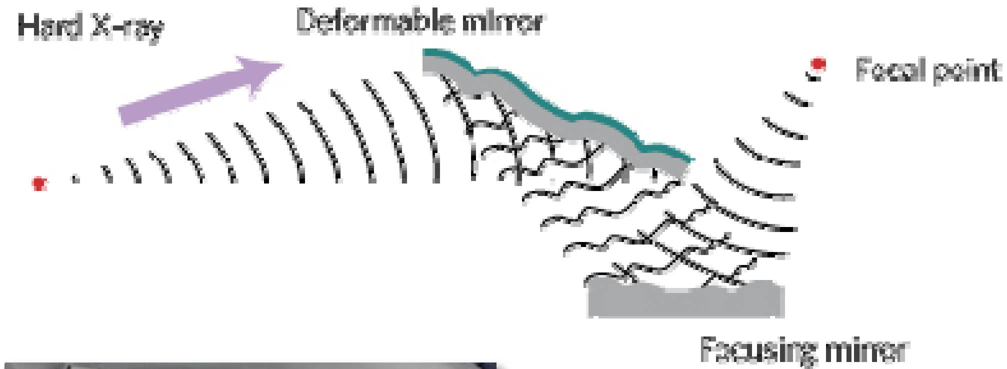


***in situ* determination of the wavefront error of X-ray focusing mirror**

Iterative phase-retrieval method
using the intensity profiles around
the beam waist

H. Yumoto, *et al*. , RSI **77**, 063712 (2006).

20 platinum/carbon bilayers



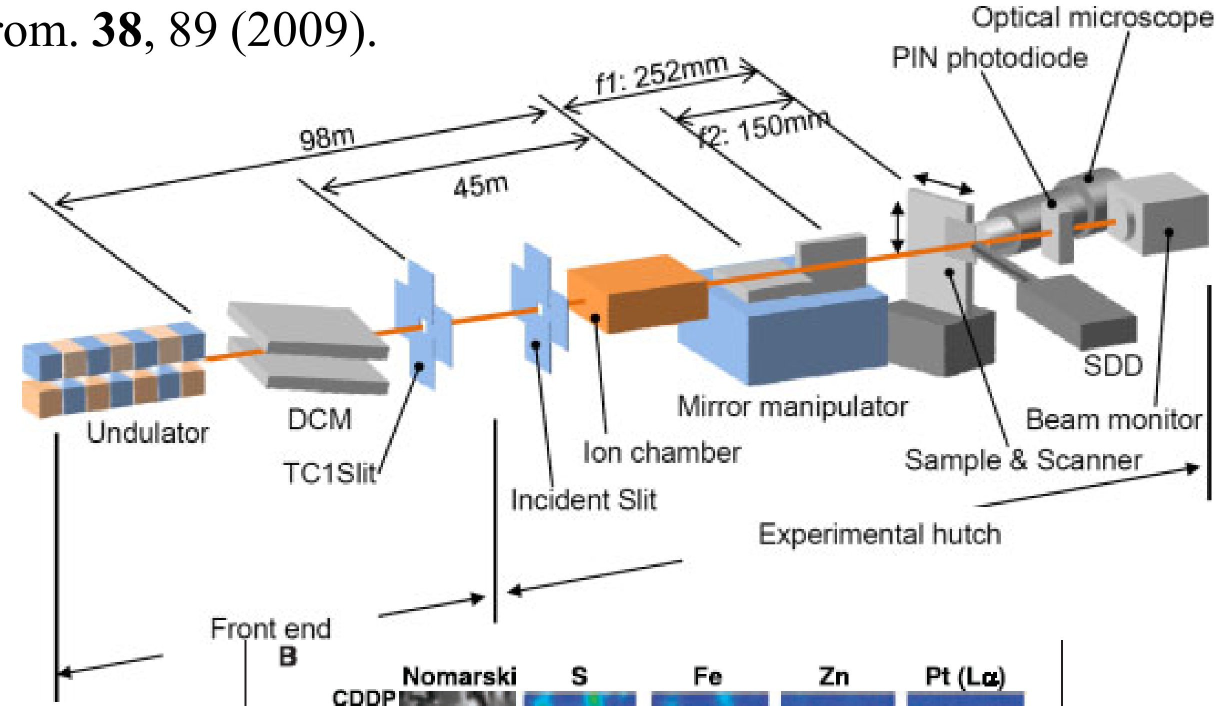
Adaptive optics to compensate figure error of the focusing mirror

T. Kimura *et al*., JJAP **48**, 072503 (2009).

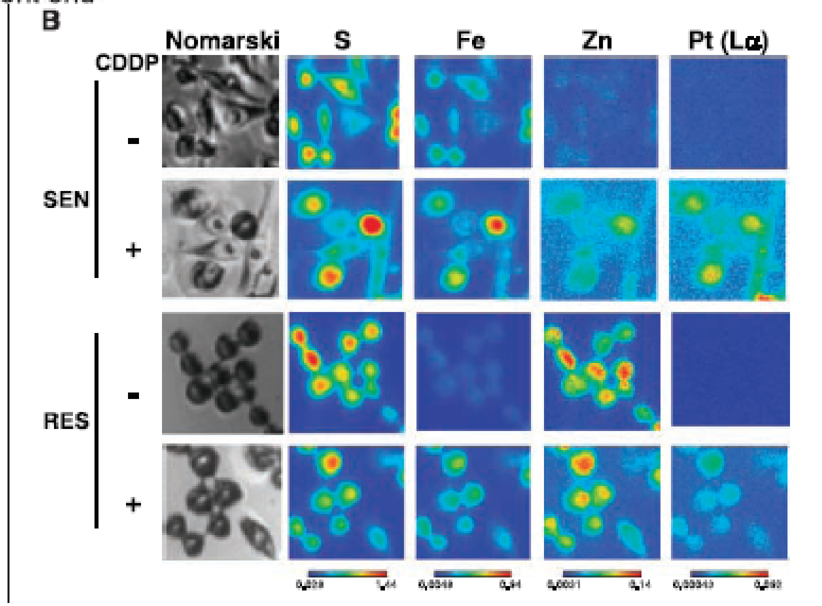
H. Mimura *et al*., Nature Physics **6**, 122 (2009).

Scanning X-ray Fluorescence Microscope

S. Matsuyama *et al.*,
X-Ray Spectrom. **38**, 89 (2009).



SPring-8
BL29XU EH2



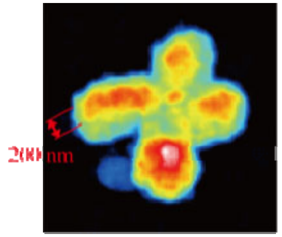
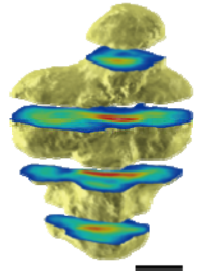
M. Shimura *et al.*,
Cancer Research **65**, 4998 (2005).

Collaborators

- Hokkaido Univ.: T. Kimura, M. Newton
- National Institute of Genetics: K. Maeshima
- Osaka Univ.: Y. Takahashi, S. Matsuyama, K. Yamauchi
- Univ. Tokyo: H. Mimura
- National Center for Global Health and Medicine: M. Shimura
- JASRI: Y. Joti
- Univ. Tokyo: S. Takeuchi
- RIKEN SPring-8: Y. Bessho, T. Ishikawa

Summary

- Imaging Chromosome
  - X-ray diffraction microscopy enable high-contrast imaging for relatively thick samples.
  - 2D & 3D observation of unstained human chromosome

- Average Structure / Individual Structure
  - SAXS
  - cross-correlation

- Focusing X-ray
  - X-ray focusing down to 7 nm
    - long beamline
    - in situ determination of figure error of X-ray focusing mirror
    - Adaptive optics to compensate figure error