

# Simultaneous K-edge subtraction tomography for strontium tracer using parametric X-ray radiation

Yasushi HAYAKAWA

Laboratory for Electron Beam Research and Application (LEBRA),

Institute of Quantum Science,

Nihon University

*Channeling 2016 ( 25–30 Sep. 2016, Sirmione–Desenzano del Garda, Italy )*

# Collaborators

Y. Hayakawa<sup>1</sup>, K. Hayakawa<sup>1</sup>, T. Kaneda<sup>2</sup>,  
K. Nogami<sup>1</sup>, T. Sakae<sup>2</sup>, T. Sakai<sup>1</sup>, I. Sato<sup>3</sup>,  
Y. Takahashi<sup>4</sup>, T. Tanaka<sup>1</sup>

<sup>1</sup>Laboratory for Electron Beam Research and Application  
(LEBRA), Institute of Quantum Science, Nihon University

<sup>2</sup>Nihon University School of Dentistry at Matsudo

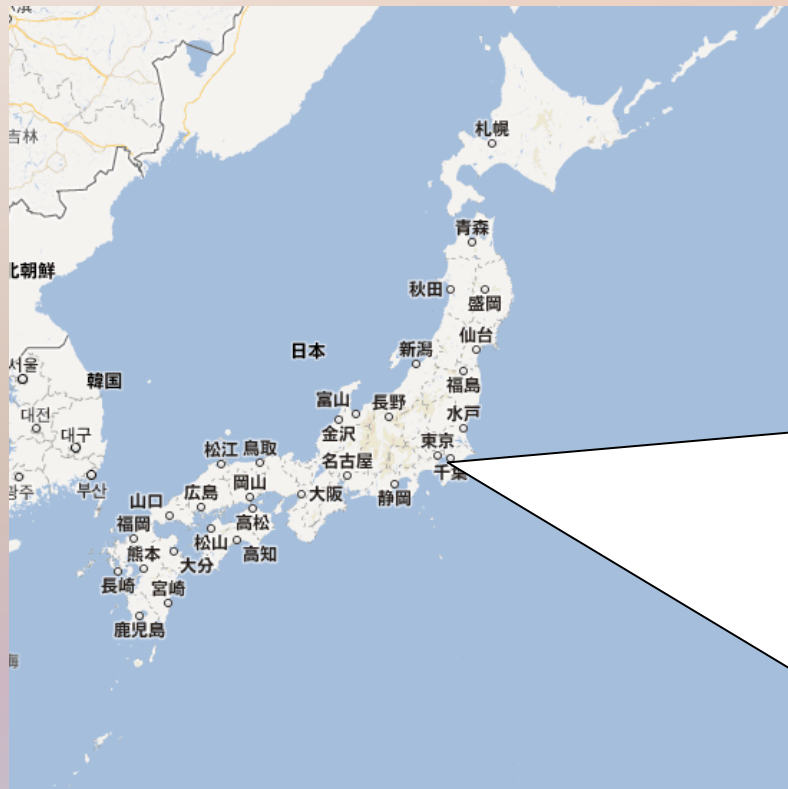
<sup>3</sup>Advanced Research Institute for the Science and  
Humanities, Nihon University

<sup>4</sup>Institute of Materials Structure Science, High Energy  
Accelerator Research Organization (KEK)

# Outlines

- ❑ Status of LEBRA-PXR source
- ❑ Advanced imaging using the PXR source
- ❑ Computed Tomography using PXR
- ❑ Temporal-KES (K-edge subtraction) method
- ❑ Simultaneous-KES method based on PXR
- ❑ Summary

# Nihon University



Funabashi, Chiba

# LEBRA: Laboratory for Electron Beam Research & Application

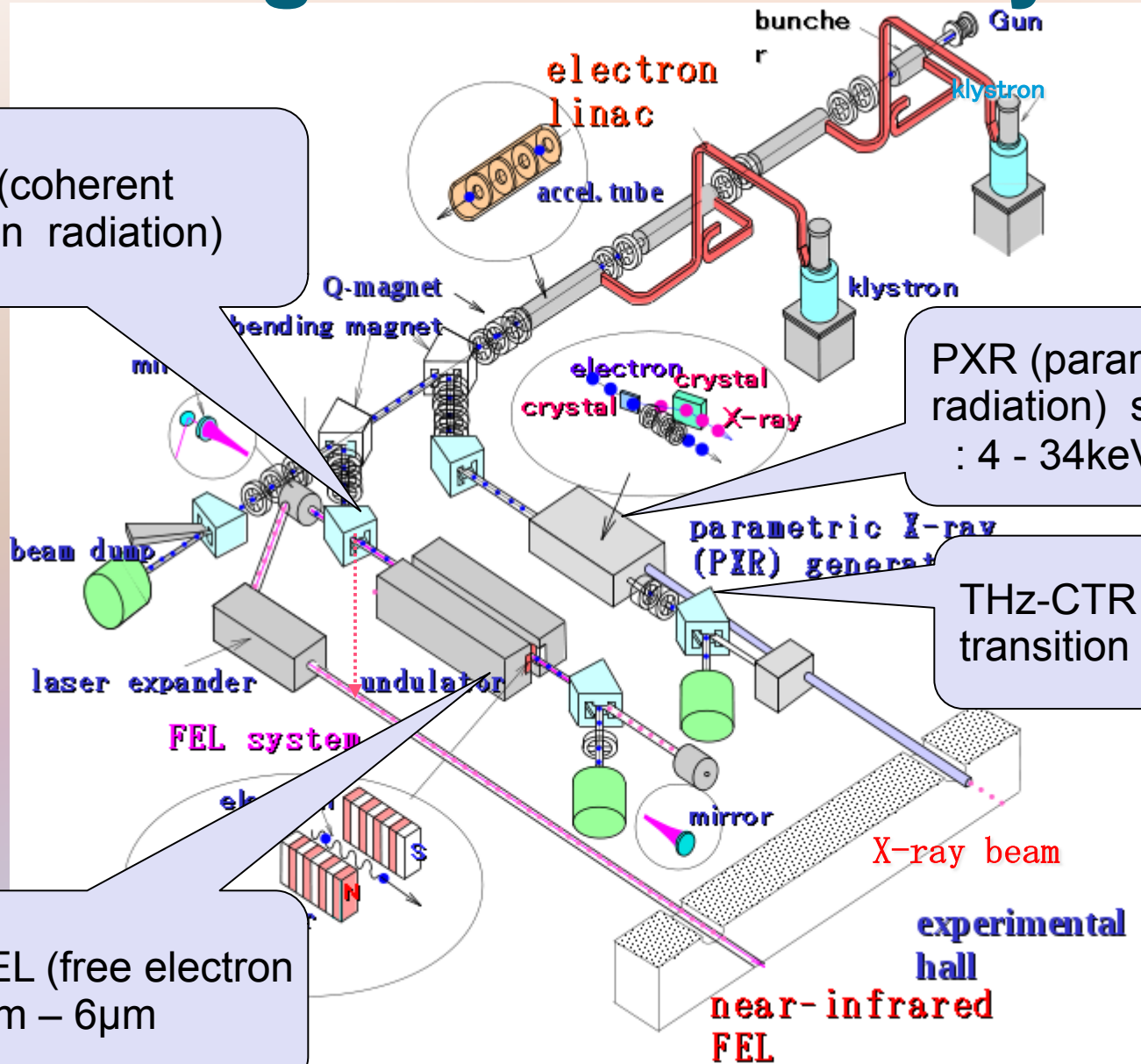


electron energy: 125MeV(max.), 100MeV(typ.)  
average current : 5μA (max.), 1 – 3 μA(typ.)

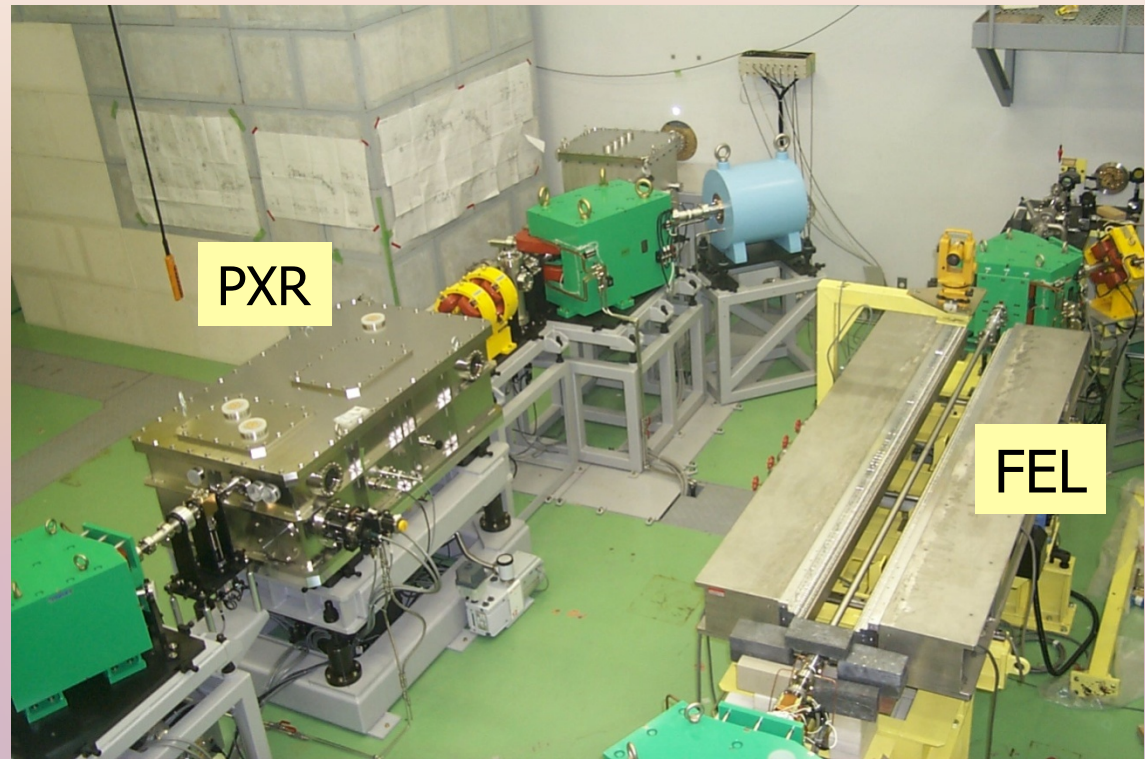
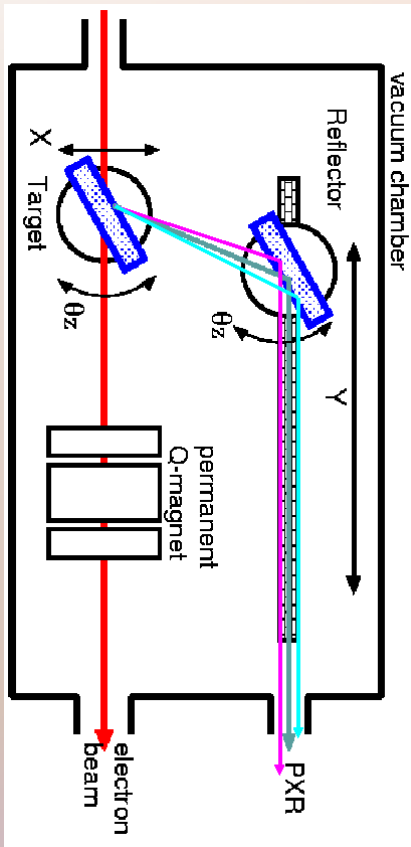
# Specification of LEBRA Linac

electron energy	50 – 100 MeV
accelerating frequency	2856 MHz
bunch length (rms)	0.5 – 3 ps
bunch charge	~ 40 pC
macropulse duration	4 - 20 $\mu$ s
macropulse beam current	~ 130 mA
macropulse repetition rate	2 – 5 pps
average beam current	< 5 <span style="border: 1px solid black; padding: 0 2px;">?</span> A
beam emittance	< $20\pi$ mm mrad

# Tunable light source facility



# LEBRA facility: beamlines (FEL & PXR)



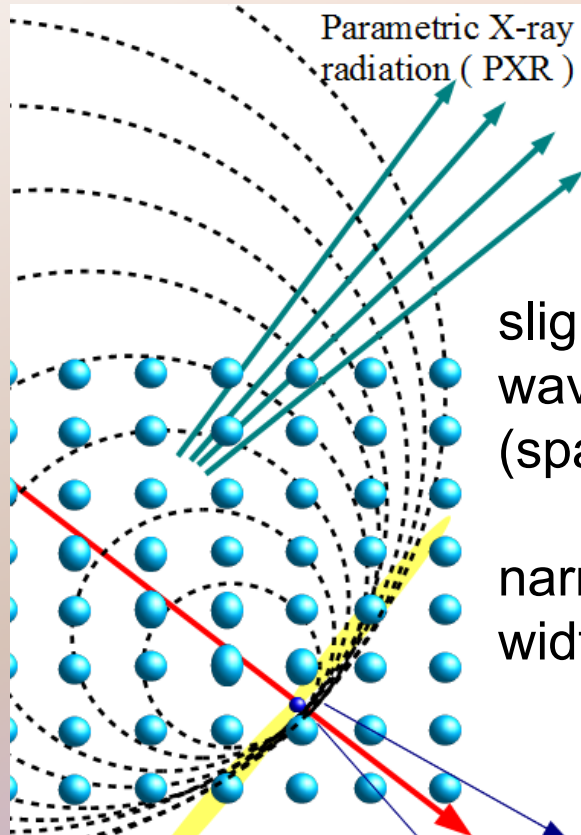
Free electron laser (FEL): 1  $\mu\text{m}$ – 6  $\mu\text{m}$  (near-IR)

Parametric X-ray radiation (PXR): double-crystal system

# Status of LEBRA-PXR source

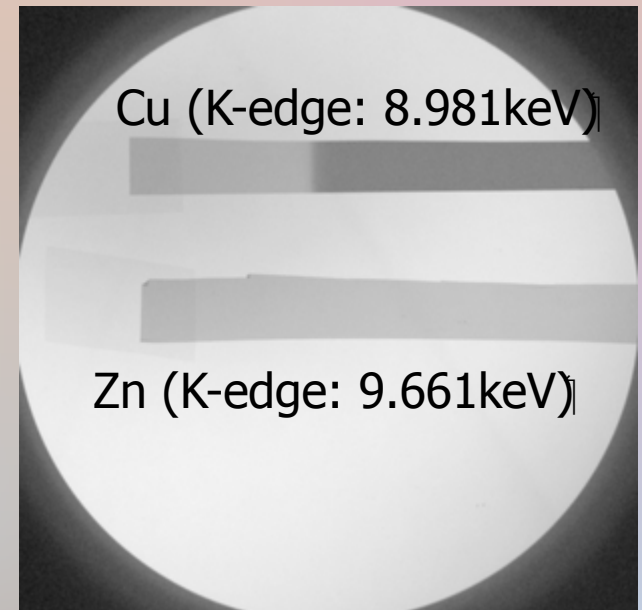
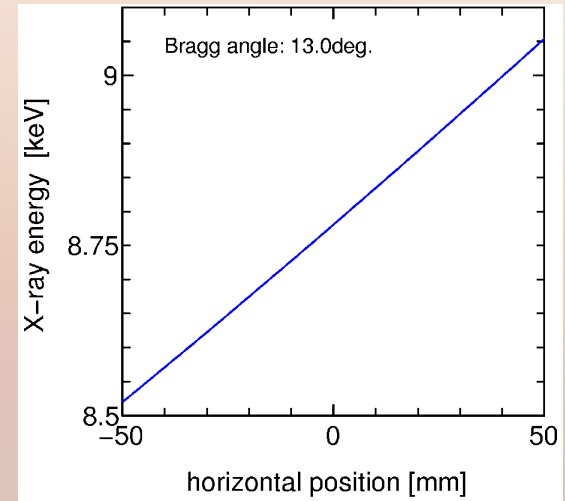
electron energy	100 MeV
X-ray radiator (target)	Si crystal plate
source size (e-beam spot on target)	0.5 – 1 mm in dia.
X-ray energy range	Si(111): 4 – 20 keV Si(220): 6.5 – 34 keV
irradiation port	100 mm in dia.
total photon rate	$\geq 10^7$ /s @17.5keV

# Spatial chirp of PXR beam



slight & continuous  
wavelength-shift  
(spatial chirp)

narrow local spectral  
width (several eV)



Wave front of PXR is different from  
both plane wave and spherical wave.

# Feature of LEBRA-PXR

- Monochromaticity

energy dispersion (spatial chirp)  $\sim 10\%$

local band width  $\sim 0.1\%$  (several eV)

- Tunability

continuous selection of the center energy

- Large irradiation area

at least 100mm in diameter

cone-beam depending on  $1/\gamma$

- Spatial coherence

phase-contrast imaging is actually possible

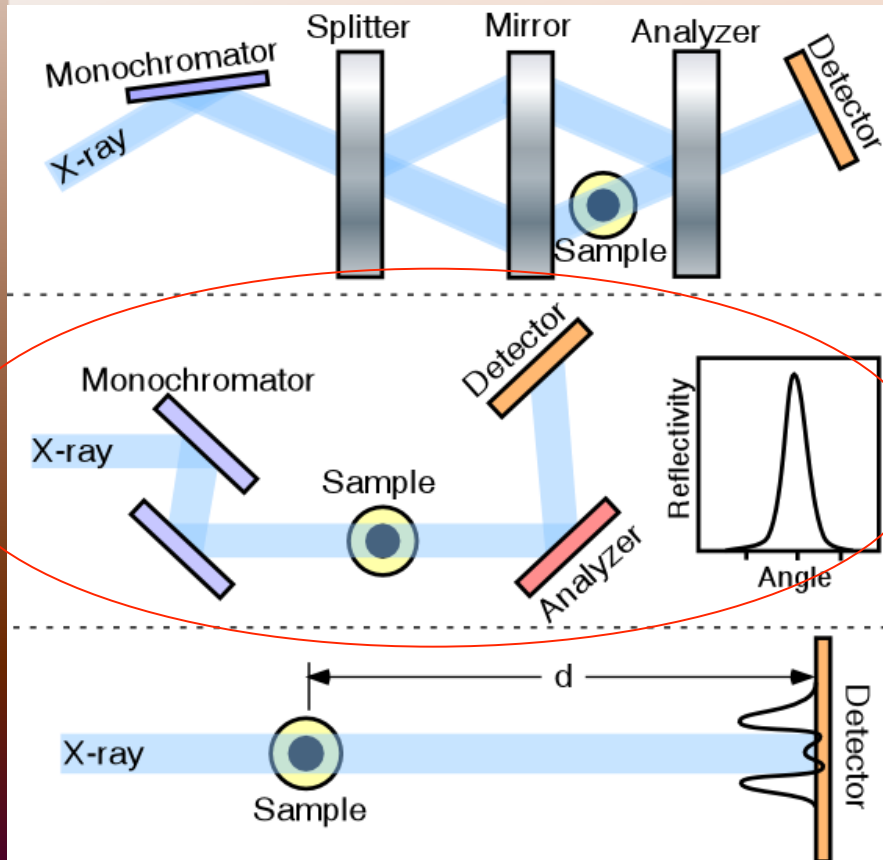
- Stability

X-ray stability depends only on the linac condition

# Application of LEBRA-PXR source

- **Conventional imaging**  
monochromaticity & tunability
- **Diffraction-enhanced imaging**  
refraction (phase-gradient) contrast  
contrast based on small-angle scattering (SAXS)
- **X-ray absorption fine structure (XAFS)**  
energy dispersive type XAFS analysis
- **Computed tomography (CT)**  
monochromaticity & tunability  
propagation-based phase contrast effect

# Phase-contrast X-ray imaging



interferometer-based technique

Si perfect crystal interferometer  
Talbot interferometer

analyzer-based technique

DEI: diffraction-enhanced  
imaging

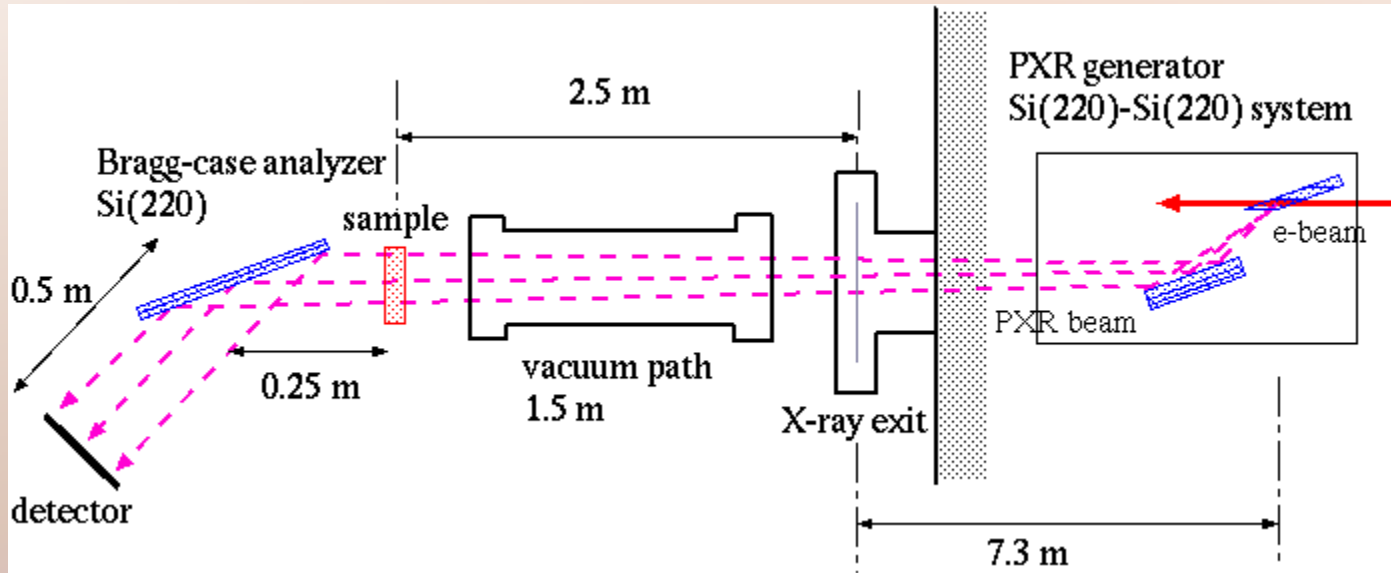
propagation-based technique

R. Fitzgerald: Phys. Today 53 (2000) 23

DEI is possible using PXR.

# Setup of DEI experiments

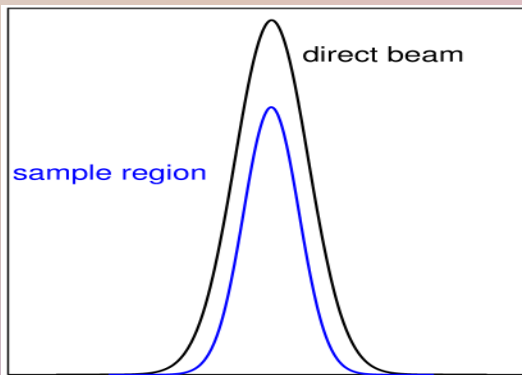
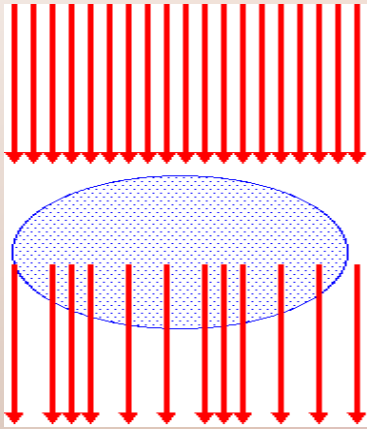
top view



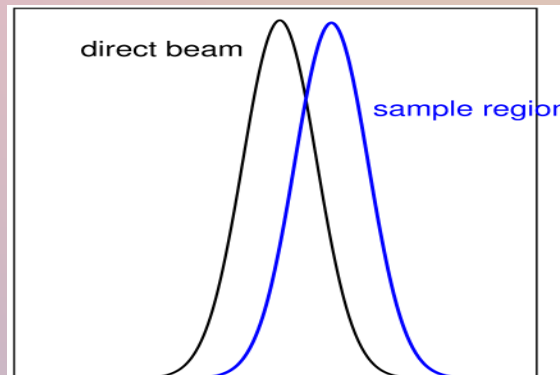
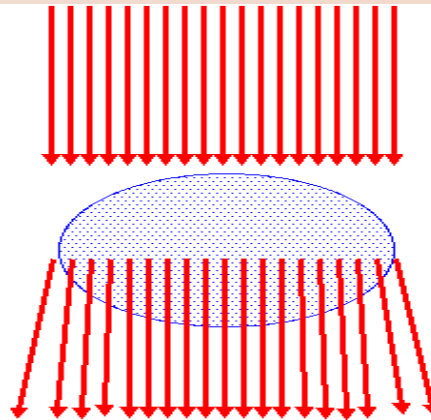
Due to the extension of cone-beam, a wide irradiation field can be obtained without asymmetric analyzer.

The distance between the PXR source and the sample is shorter than 10m.

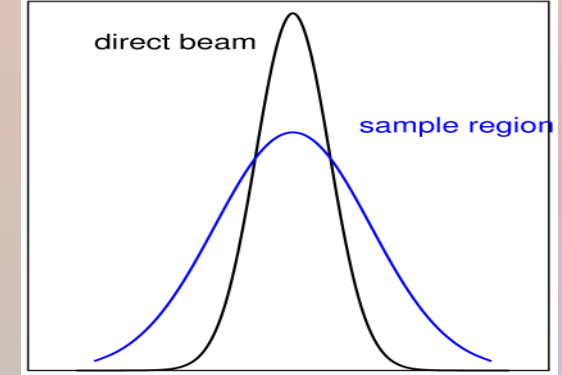
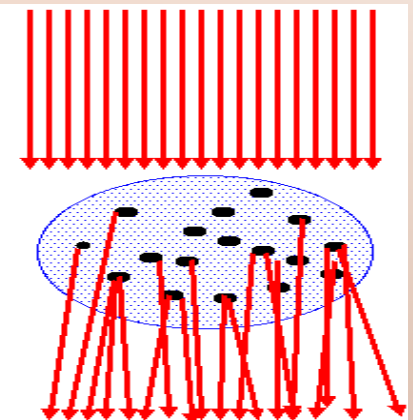
# Interaction between X-rays and material



absorption (amplitude attenuation): reduction of the peak area

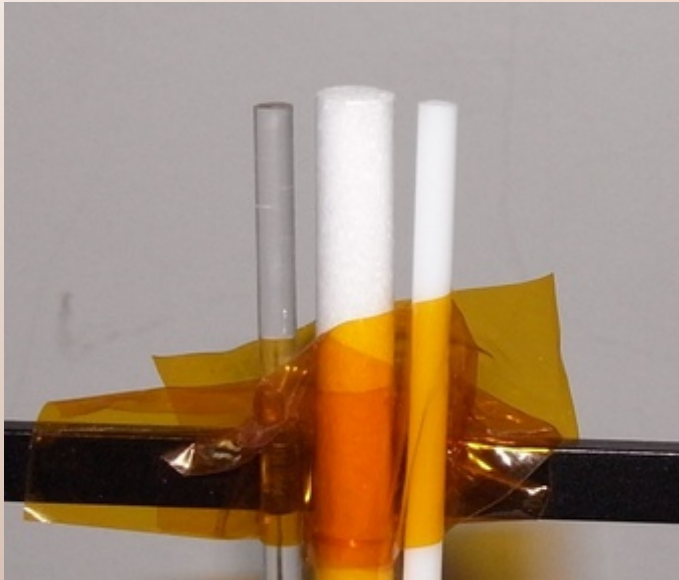


refraction (phase shift): shift of the center position



small-angle scattering (SAXS): reduction of the peak height (or peak broadening)

# Experiment for demonstration



PXR source:

radiator-reflector: Si(220)-Si(220)

electron energy: 100MeV

average beam current:  $3\mu\text{A}$

PXR energy: 25.5keV

photon rate:  $\sim 10^6$  /s /100mm in dia.

Sample:

acrylic rod (3mm in dia.)

density:  $1.17 \text{ g/cm}^3$

styrene-foam rod (6mm in dia.)

density:  $0.16 \text{ g/cm}^3$

polystyrene rod (3mm in dia.)

density:  $0.986 \text{ g/cm}^3$

DEI measurement setup:

analyzer: Si(220)

160mm x 35mm x 5mm

angular step:  $0.4625 \mu\text{rad}$

image sensor: X-ray CCD

(Q.E. @25.5keV  $\sim 10\%$  )

pixel size:  $24\mu\text{m} \times 24\mu\text{m}$

# Image contrast

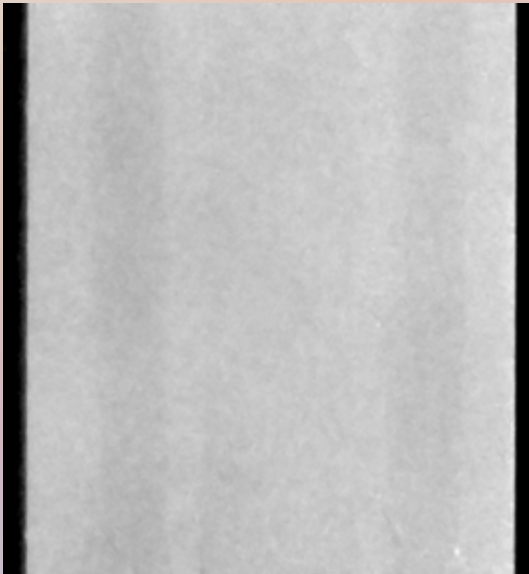
sample:

acrylic rod, styrene-foam rod, polystyrene rod

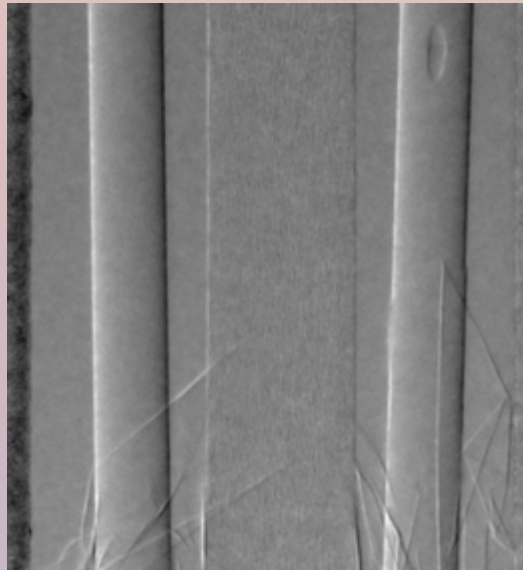
PXR energy: 25.5 keV

Phase contrast is much stronger than absorption contrast.

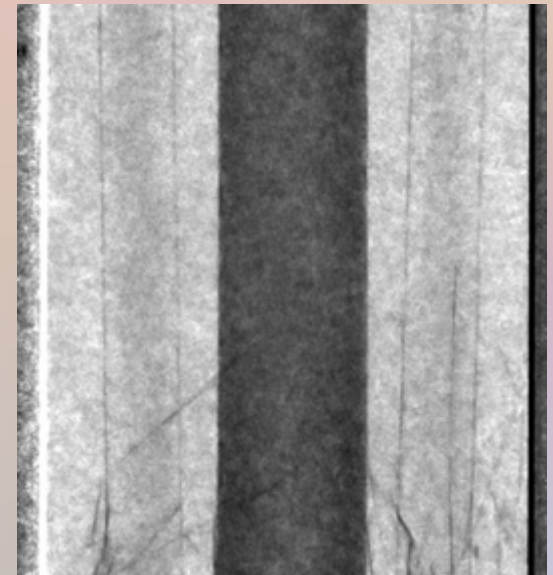
SAXS imaging is sensitive to micro structures of sample material smaller than 1-micron.



absorption

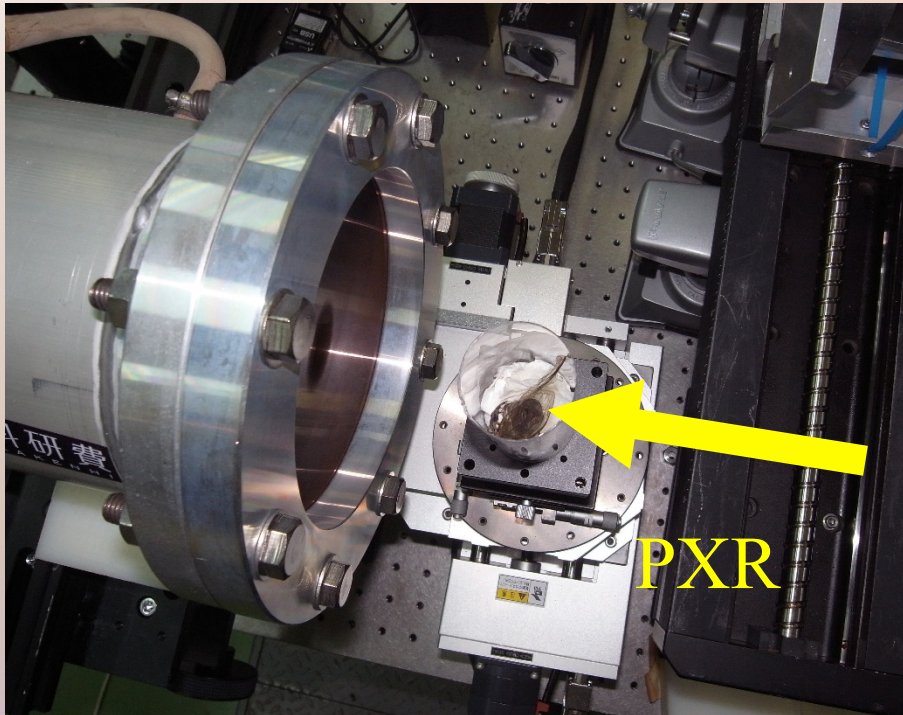


refraction  
(phase gradient map)



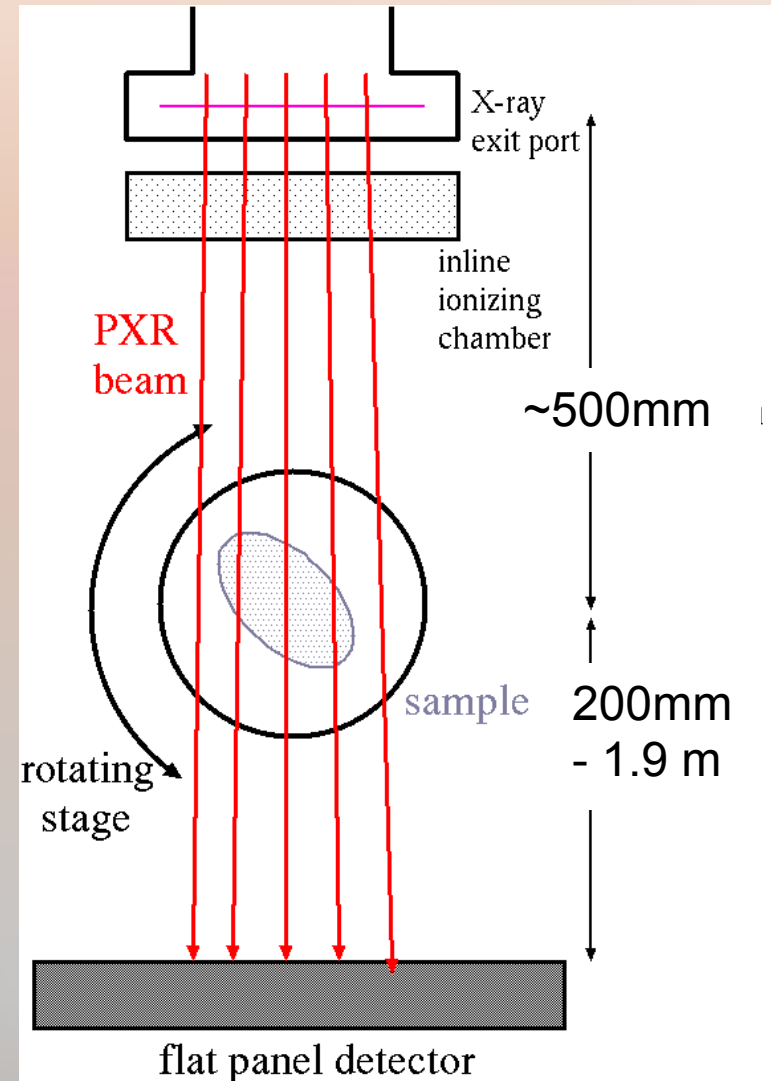
small-angle scattering  
(SAXS contrast)

# Setup for CT experiment using PXR



vacuum  
path

sample &  
rotating stage  
( 0 – 180 deg.)



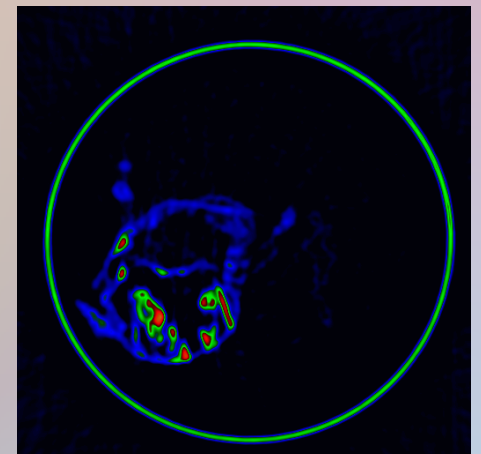
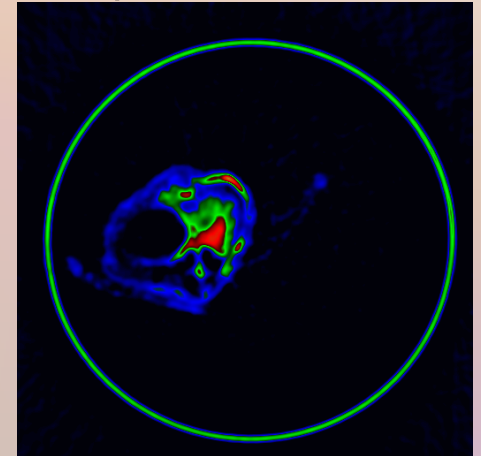
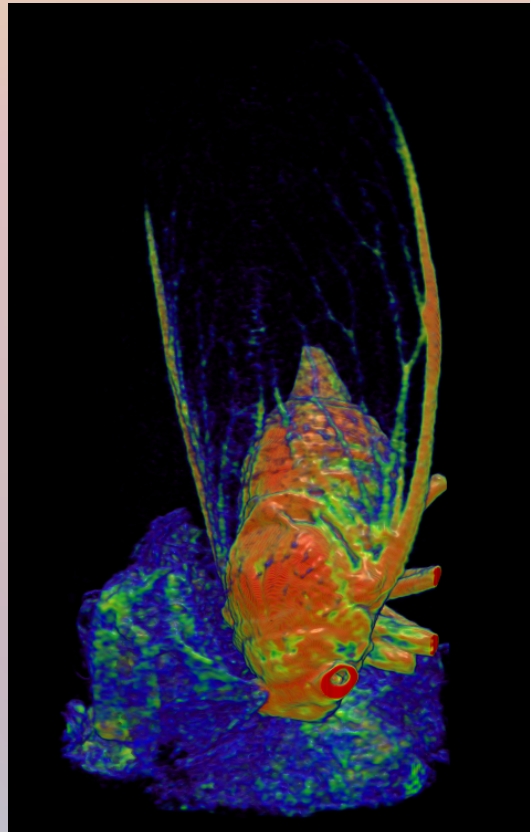
# CT image using PXR

PXR source: Si(220)      PXR energy: 15keV

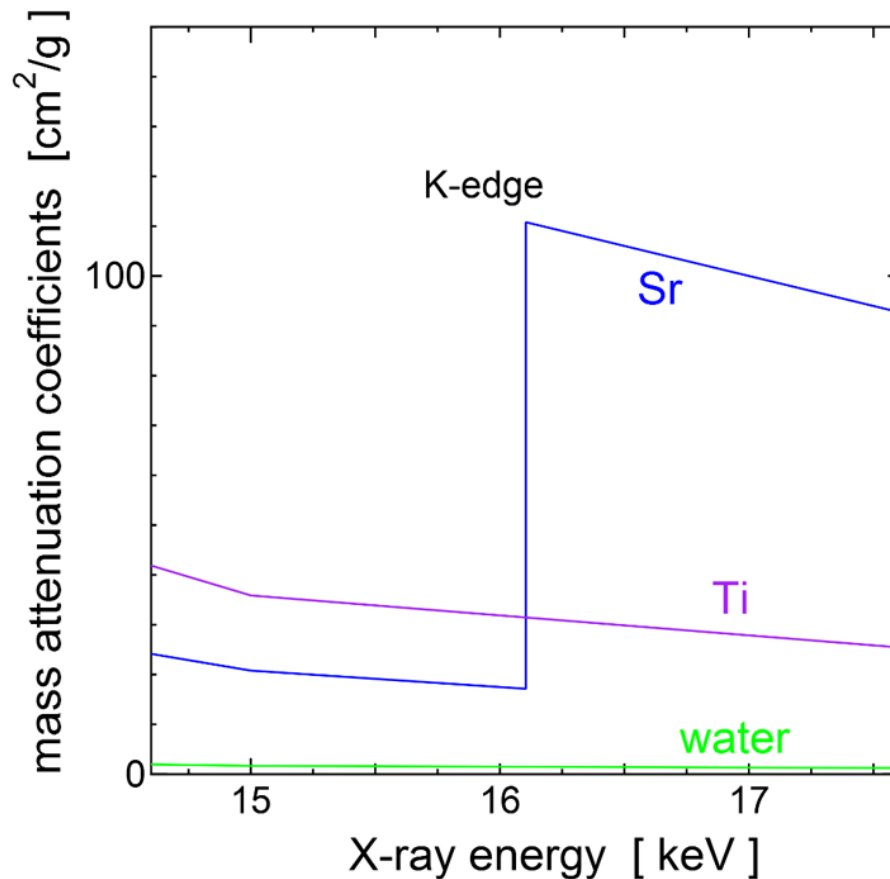
sample: cicada (insect)

measurement time: 20s x 600 ( 3 hours 20min )

FPD: Shad-o-Box 1280HS ( pixel size: 100 $\mu$ m)



# X-ray attenuation around the K-shell edge of Sr



At the K-shell absorption edge of strontium, the X-ray absorption power drastically changes at only the place where strontium exists.

Sr K-edge: 16.105 keV

# Element detection (Sr)



Sample:  
matryoshka doll  
( 3 layers )

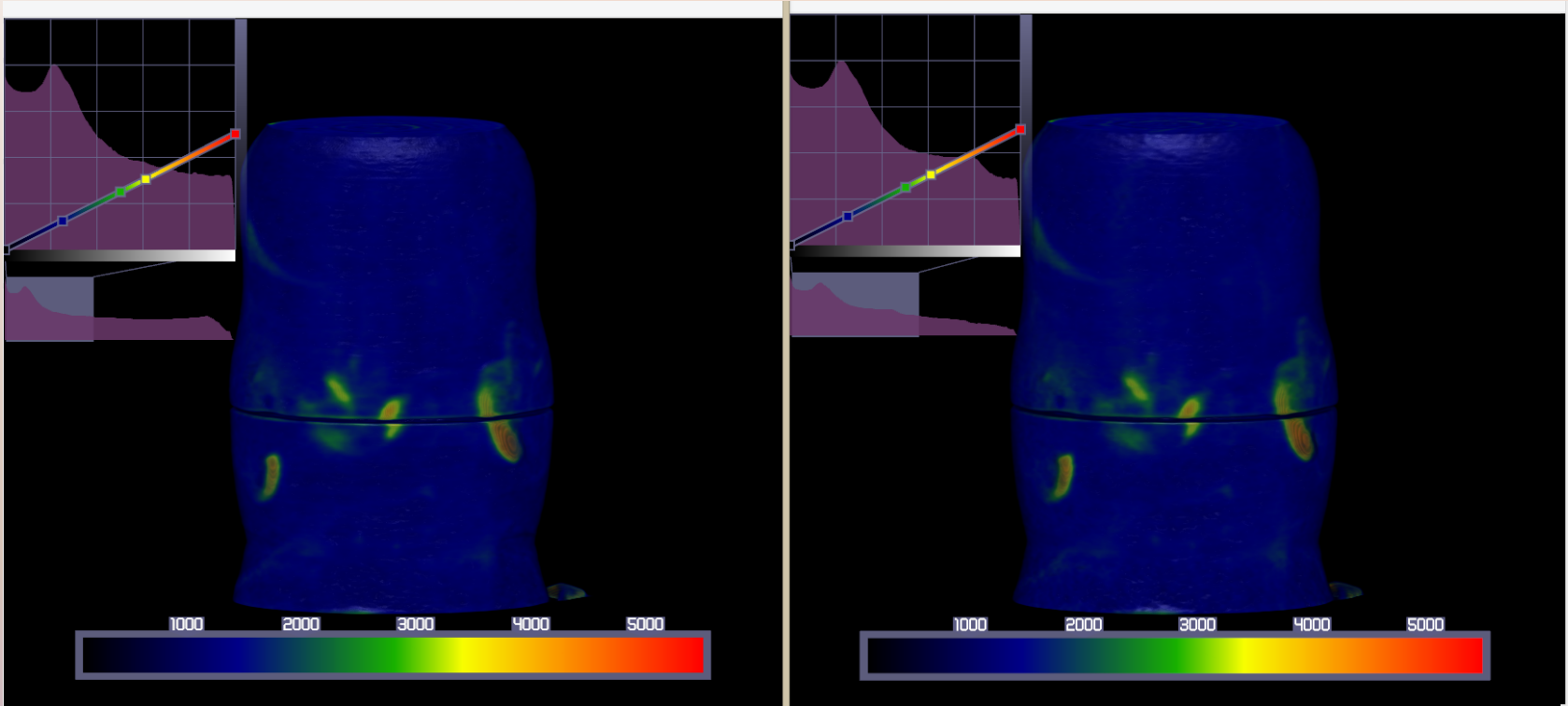
material: wood  
diameter: 34mm  
height: 55mm



polyethylene pellets  
containing  
 $\text{SrTiO}_3$  (STO)  
(white pigment)

density:  $1.0 \text{ g/cm}^3$   
Sr: 4.8 wt %

# Effect of K-shell absorption edge



16.7 keV

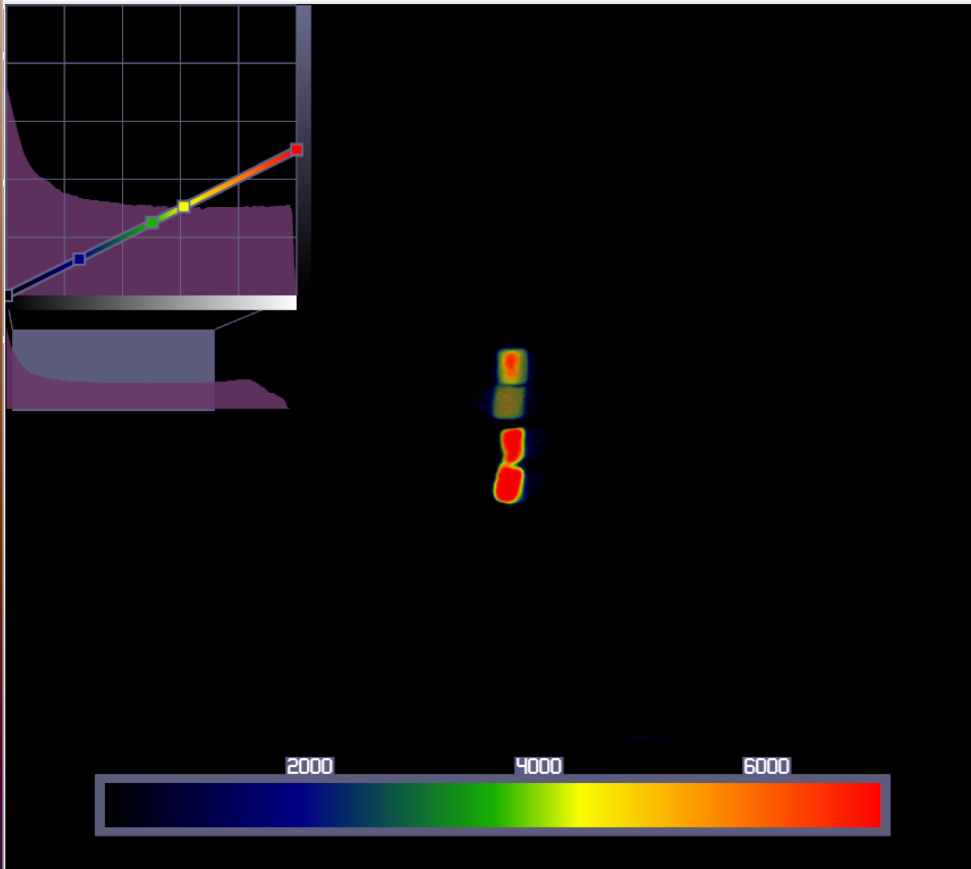
15.5 keV

Sr K-shell absorption edge: 16.1 keV

each measurement time: 1 hr (360 projections)

Both image contrasts are normalized at the wooden region.

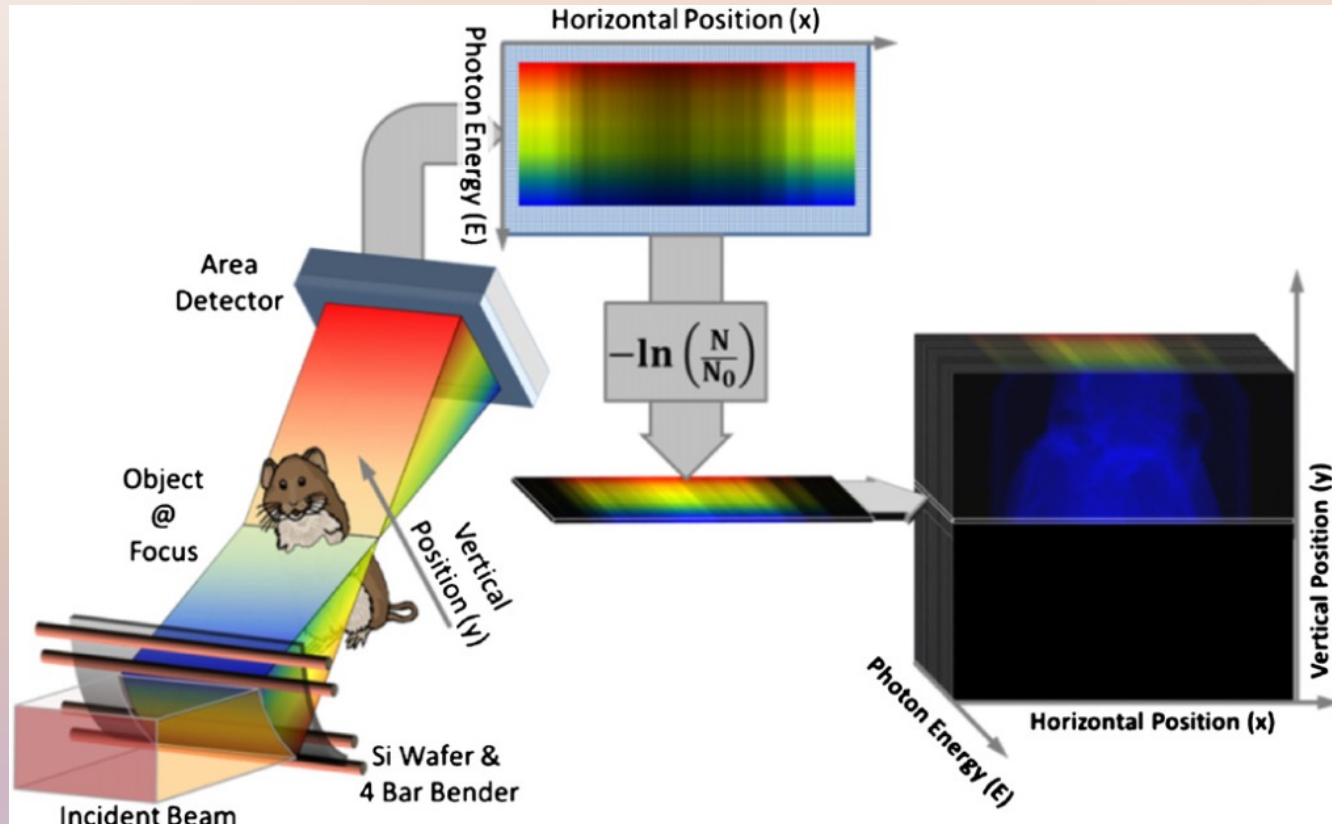
# K-edge subtraction (KES) method



The 3D distribution of Sr element is obtained as difference between the tomographic images of 16.7keV and 15.5keV.

In this case, the datasets of CT were separately acquired. This method, therefore, is referred to as temporal K-edge subtraction (KES).

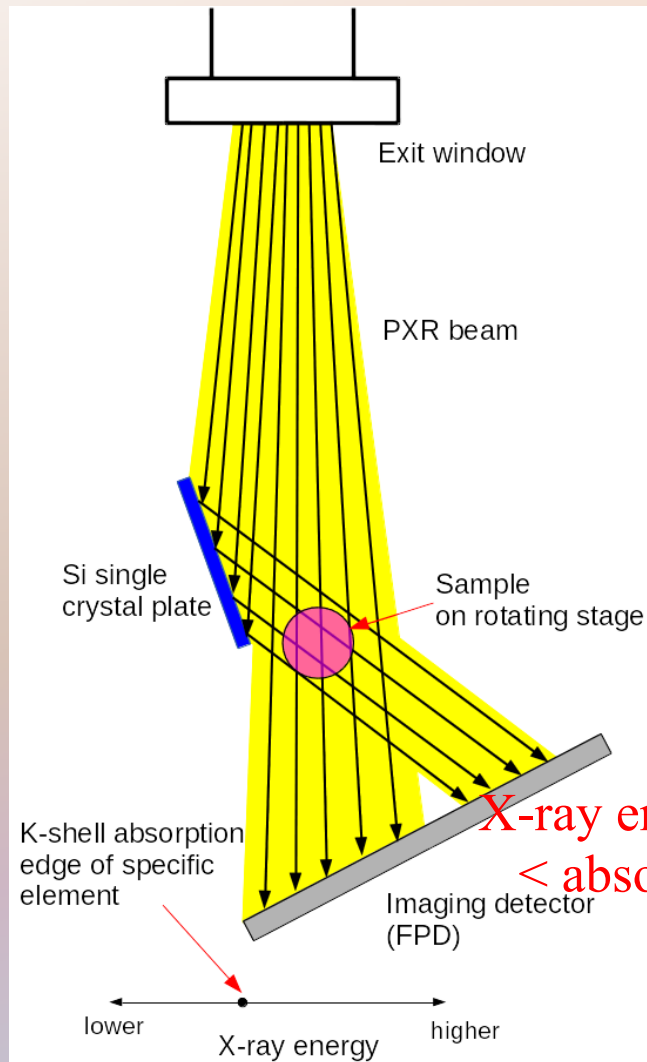
# spectral-KES using SR source



Y. Zhu, et al., Phys. Med. Biol. 59 (2014) 2485–2503.

Simultaneous KES method is developed at Canadian Light Source using bent crystal optics. They call the method “spectral-KES method”.

# Simultaneous-KES CT using PXR



Using the spatial chirp property of PXR, crossing two beams having slightly different energies can be formed using DEI setup.

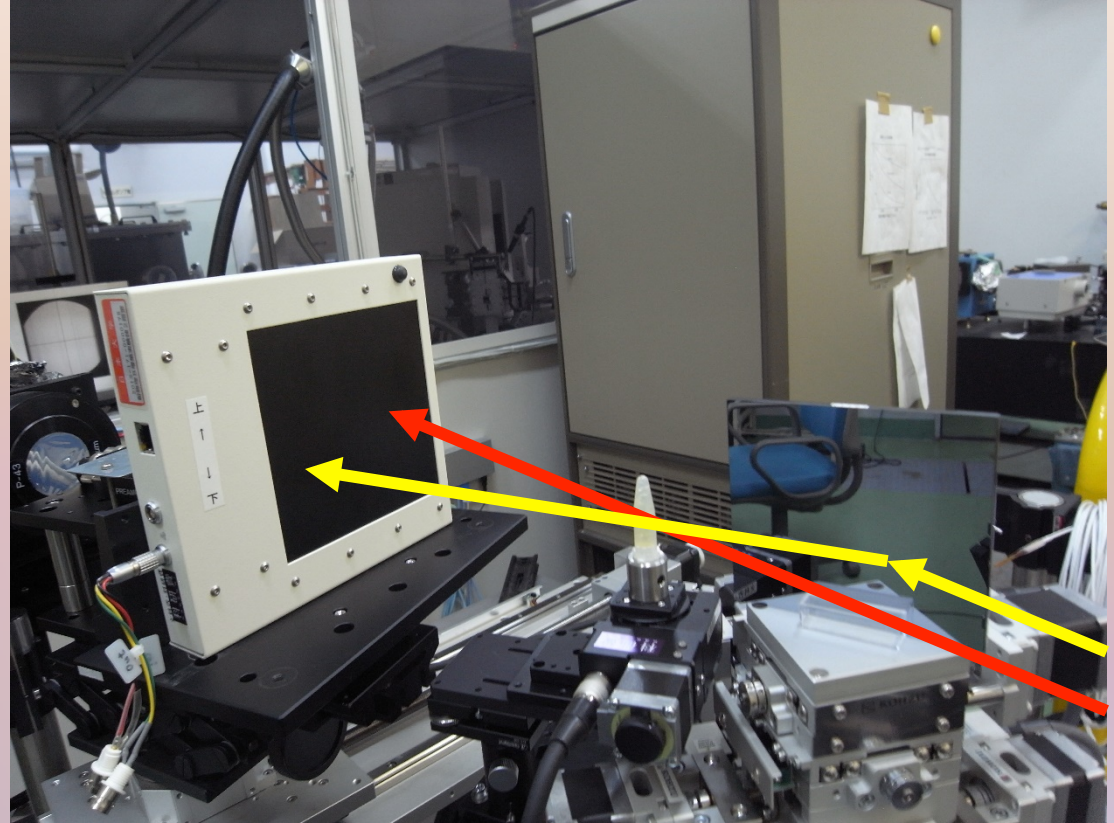
The 2-color beams can be used for simultaneous KES when the center energy of the PXR beam is adjusted to the K-edge energy of a specific element.

Moreover, KES-CT is possible by rotating the sample.

# Setup for KES-CT experiment

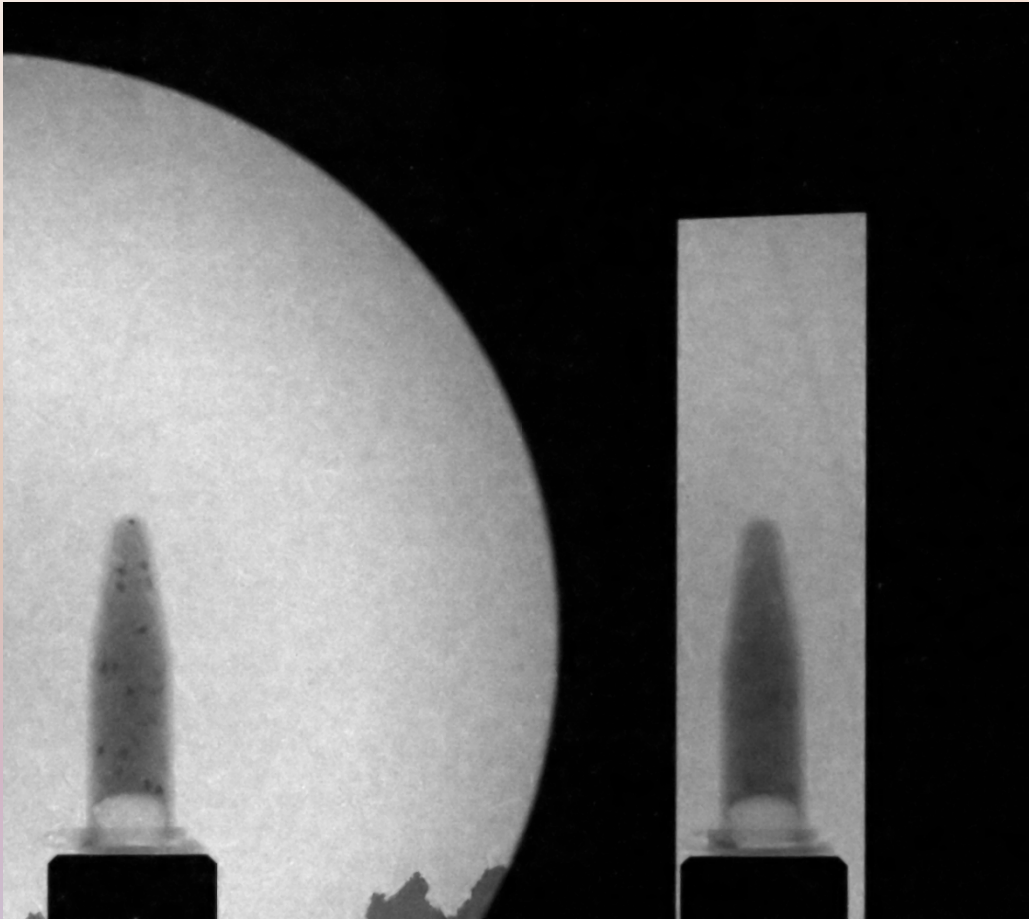


sample:  
epoxy resin  
+  
polyethylene  
fragments colored  
with  $\text{SrTiO}_3$  (STO)



PXR source & DEI analyzer: Si(220)  
PXR energy: 16.105 keV  
FPD: Shad-o-Box 1280HS  
( pixel size: 100 $\mu\text{m}$ )

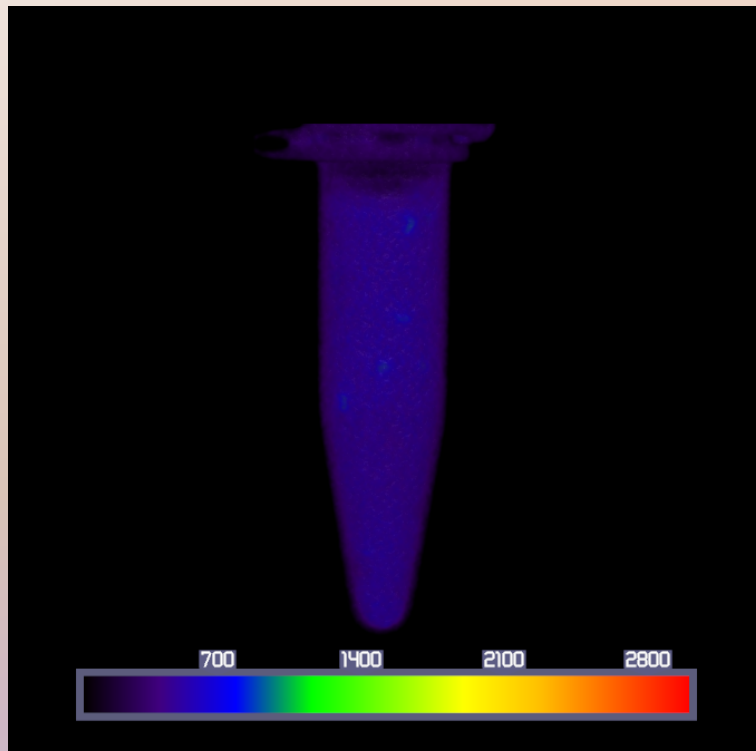
# Simultaneous KES-CT using PXR



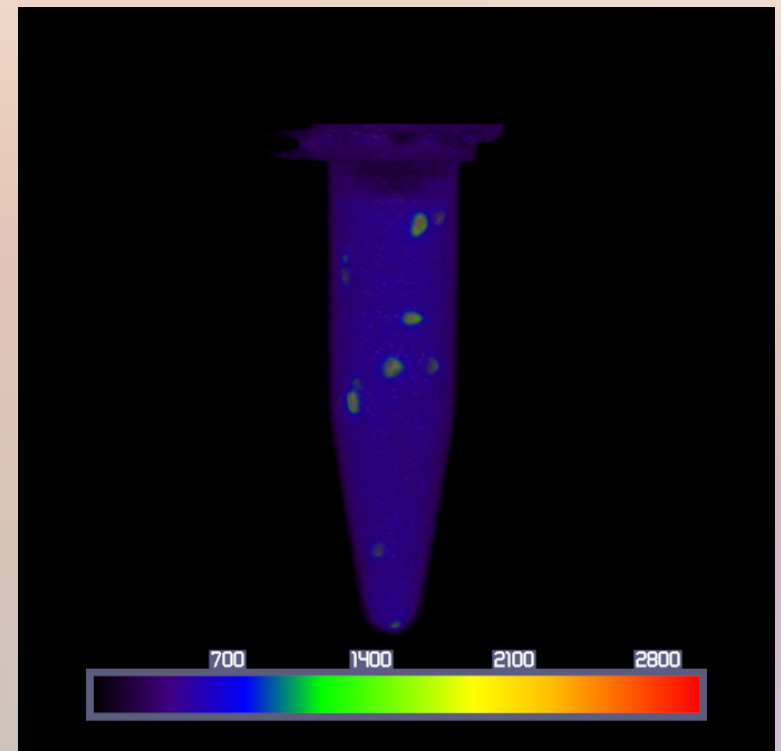
X-ray reflectivity of  
the Si(220)  
analyzer crystal  
~ 75%

projection images: 360 (angular step: 0.5 deg.)  
each exposure time: 20s  
total measurement time: 2 hours

# 3D reconstruction from KES-CT data



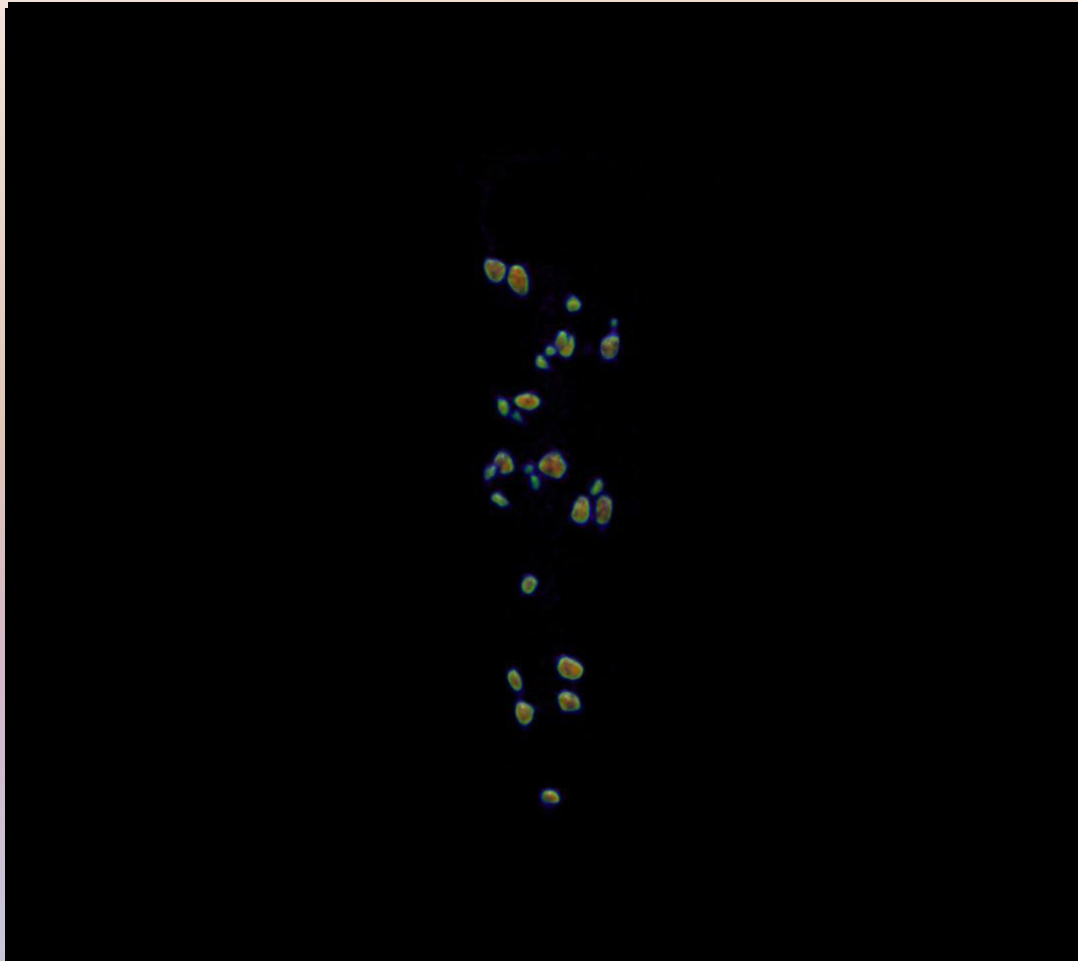
lower than Sr K-edge  
~ 16.0 keV



higher than Sr K-edge  
~ 16.2 keV

Both image contrasts are normalized at the region of epoxy resin.

# Sr distribution obtained by KES-CT



Sr distribution is calculated as subtraction of two CT images.

# Summary

- KES method is useful for the detection of specific element.
- Simultaneous-KES method is one of the current topics in the field of advanced X-ray imaging.
- Combining DEI technique and the energy selectivity of PXR makes it possible to realize simultaneous KES without bent-crystal monochrometer.
- We demonstrated that simultaneous KES-CT using PXR has a capability to detect strontium distribution in a light material sample.
- PXR-KES has a competitive edge in the field of element imaging against other X-ray sources such as SR sources.

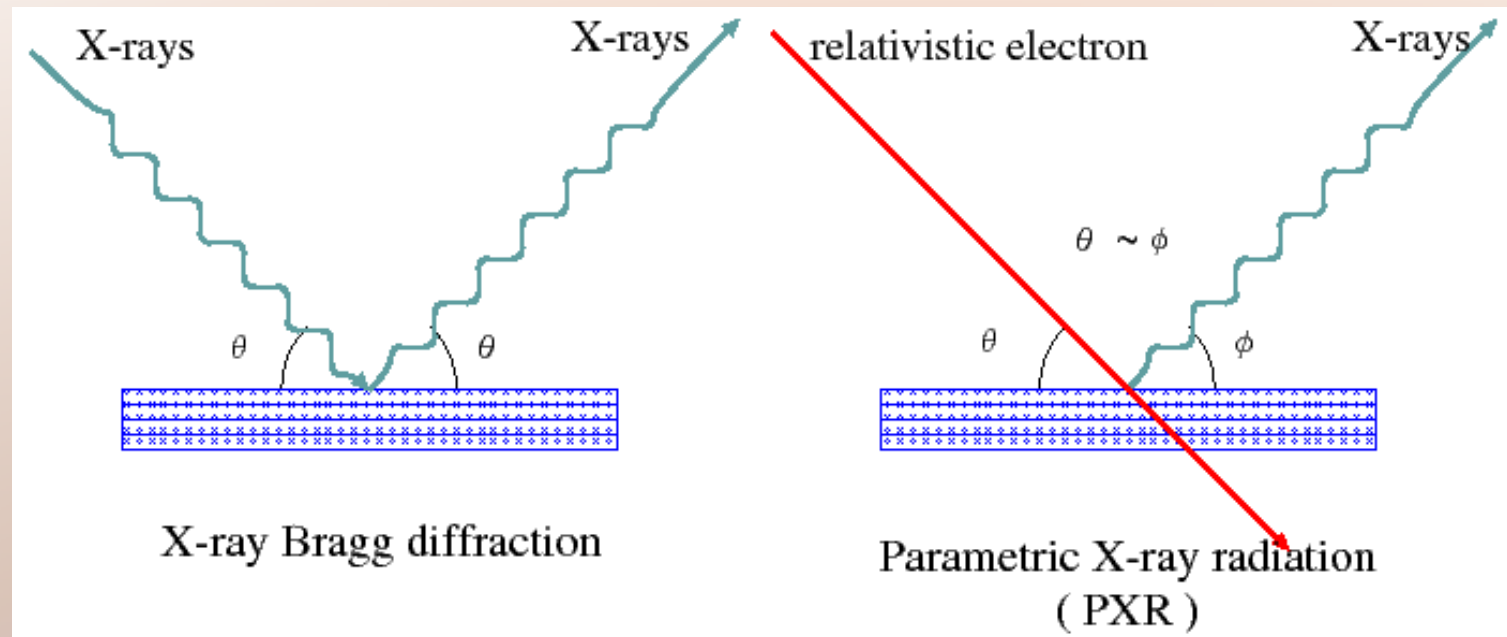
# Acknowledgements

**MEXT.KAKENHI (25286087&16K05008)**

*Thank you for your kind attention !!*

# Appendix

# What is PXR?



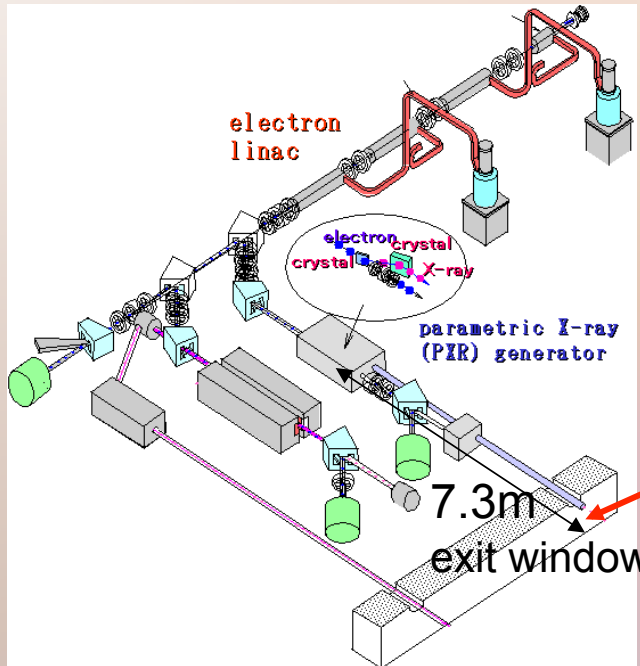
## PXR: Parametric X-ray Radiation

PXR is apparently similar to X-ray Bragg diffraction.

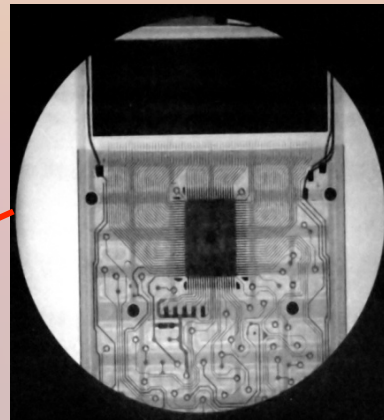
The phenomenon corresponds to Bragg diffraction for virtual photons accompanying the incident electron.

The X-ray energy is tunable depending on the Bragg angle.

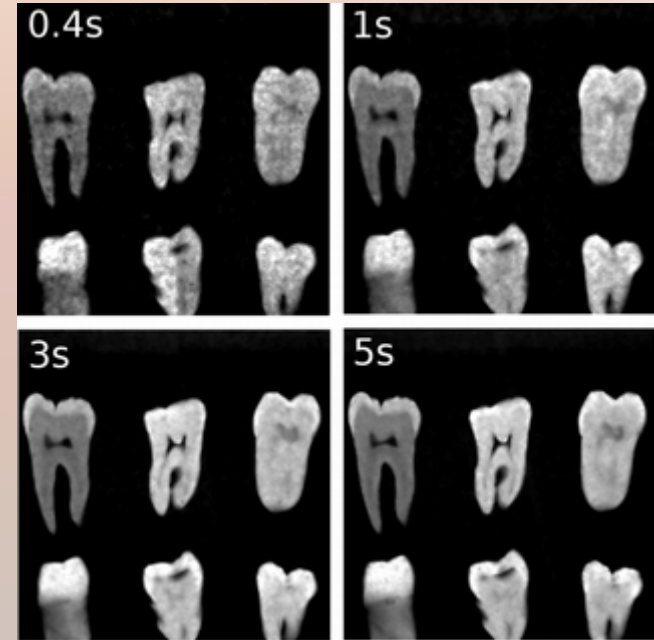
# X-ray imaging (absorption contrast)



diameter: 100mm

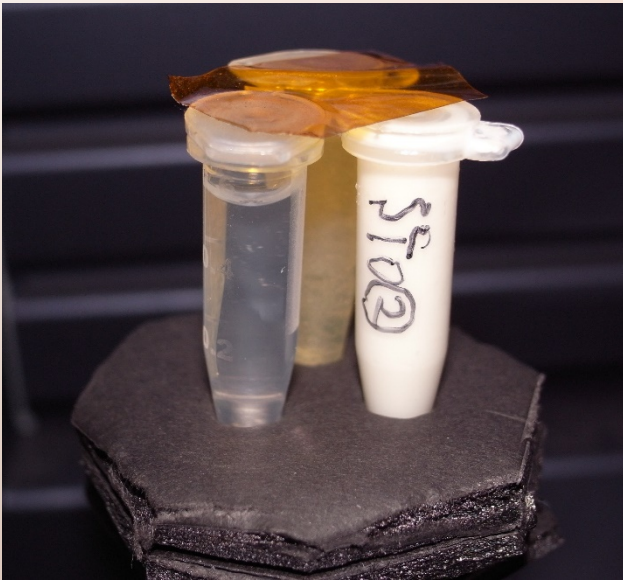


PXR radiator: Si(111) ↑  
PXR energy: 17.5keV (center)  
e-beam: 2.6uA (average)  
sample: calculator  
detector: imaging plate (IP)  
exposure: 10s



PXR radiator: Si(111) ↑  
PXR energy: 17.5keV (center)  
e-beam: 2.6uA (average)  
sample: human tooth  
detector: flat panel detector (FPD)

# Sample 2



Experiment for a material with lower concentration of Sr

ethanol

density:  $0.79 \text{ g/cm}^3$

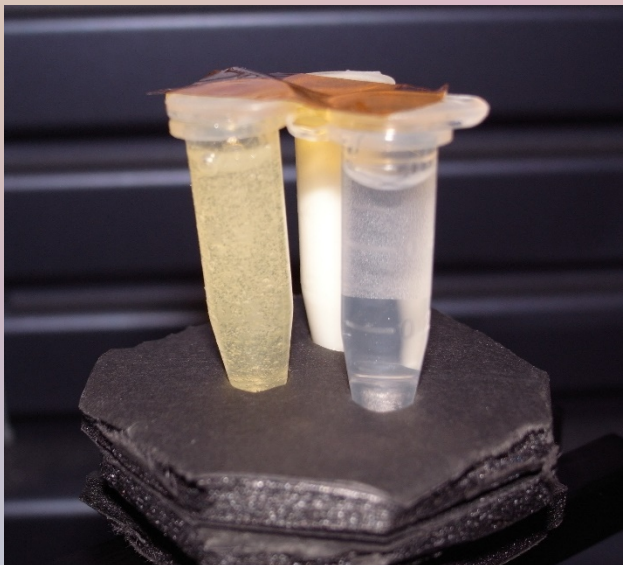
epoxy resin

density:  $\sim 1.2 \text{ g/cm}^3$

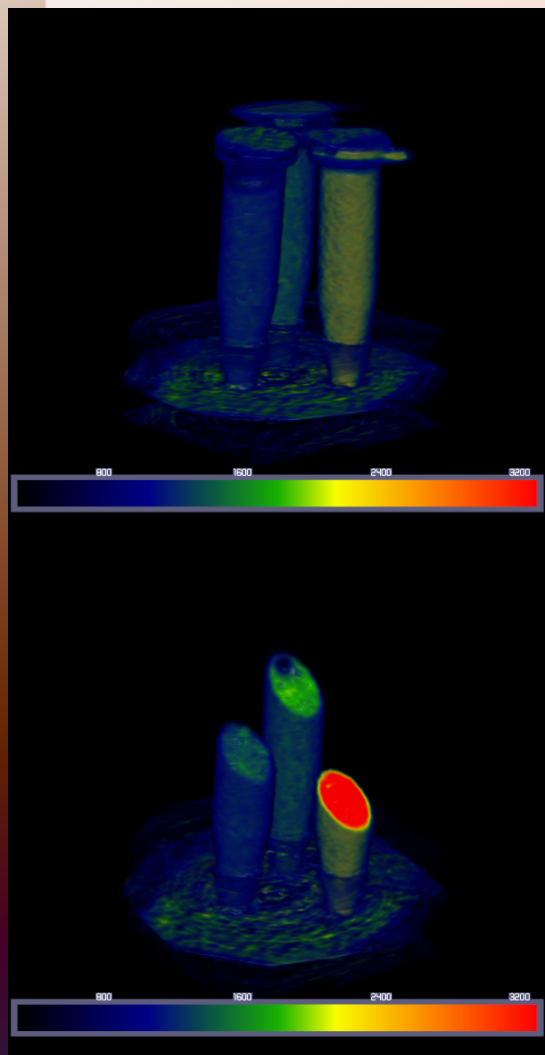
epoxy resin +  $\text{SrTiO}_3$  (STO)

density:  $\sim 1.2 \text{ g/cm}^3$

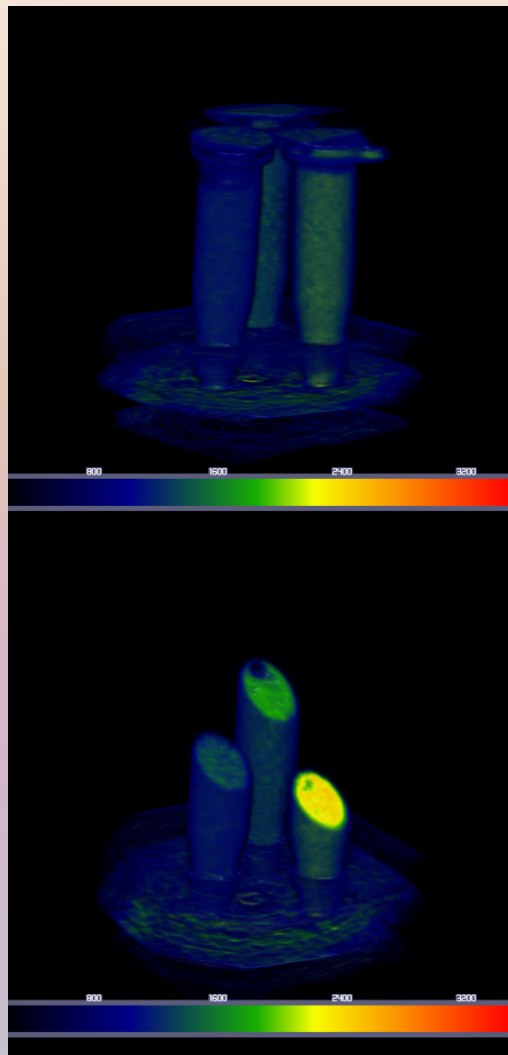
Sr: 0.6 wt %



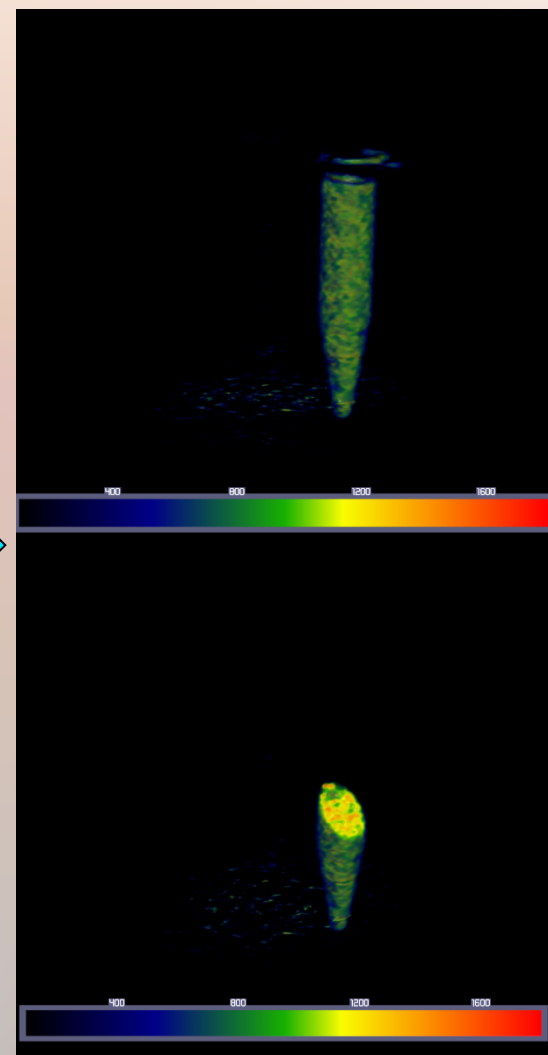
# Comparison of 3D distributions



16.6 keV

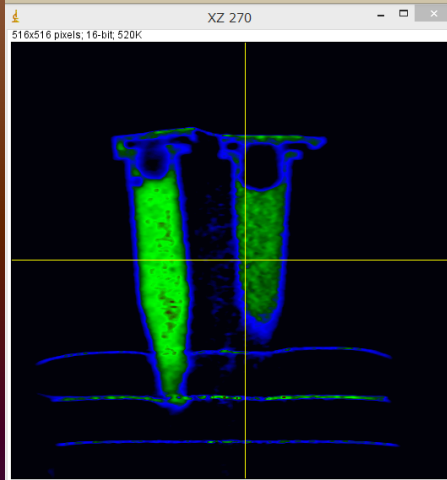
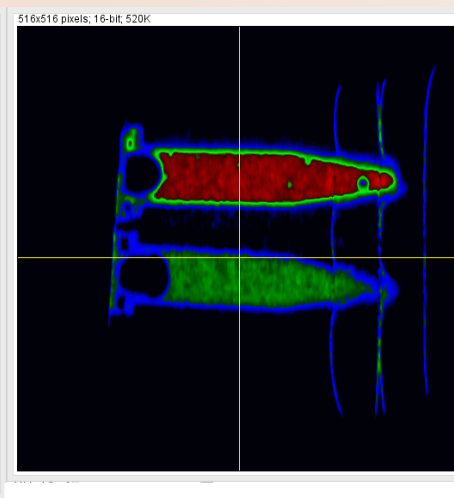
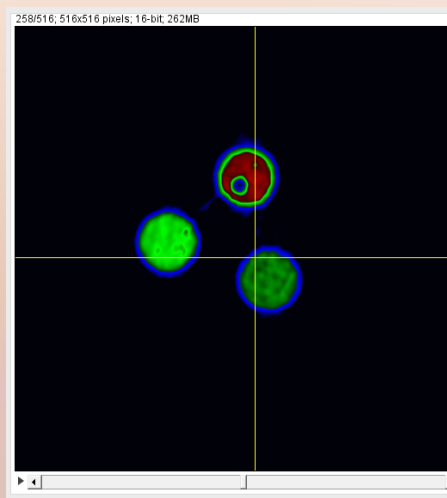
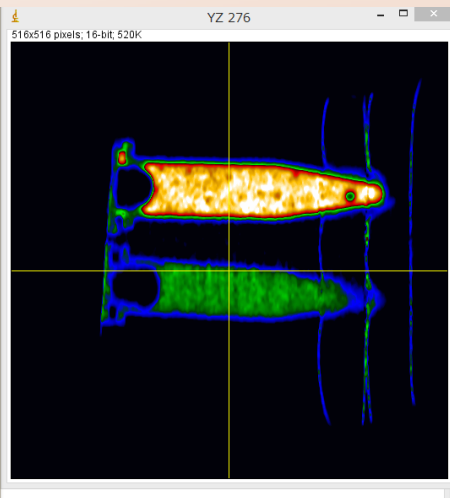
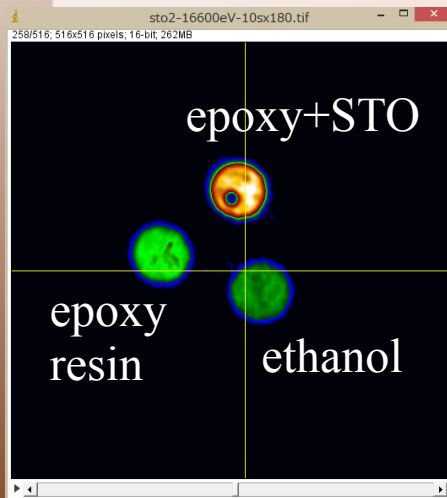


15.6 keV

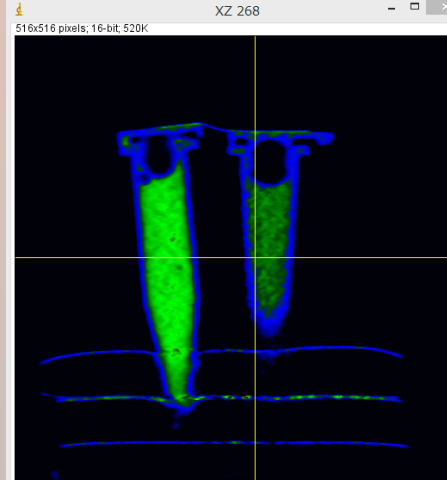


difference

# Result of reconstructions



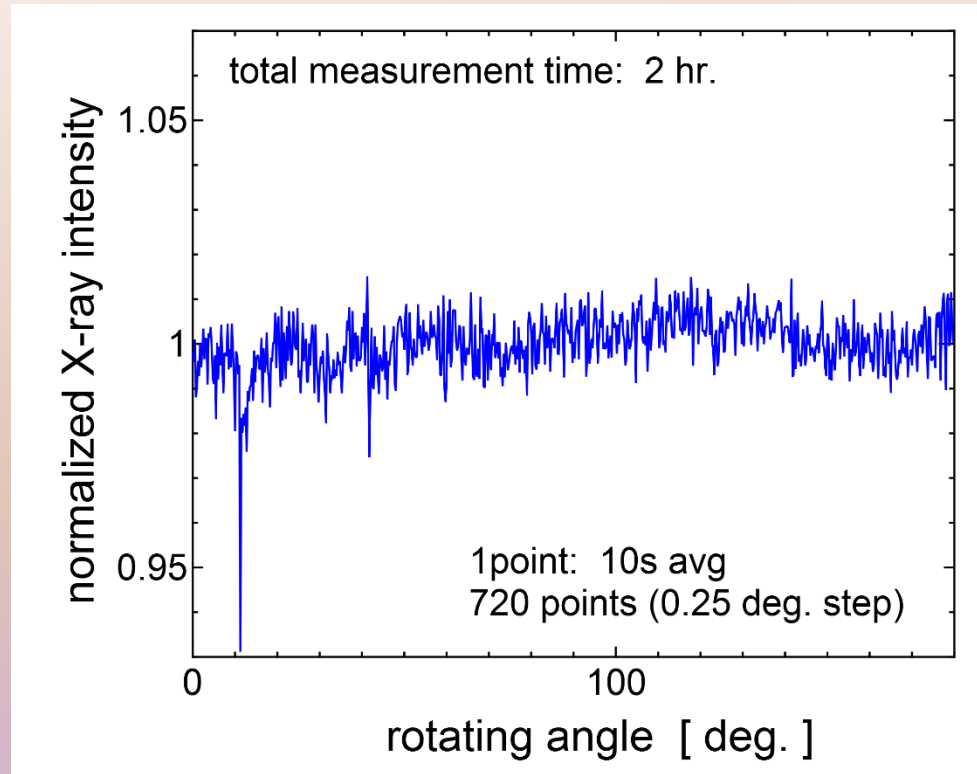
16.6keV



15.6keV

Image contrasts are normalized at the region of ethanol.

# Stability of PXR beam



Long term stability of the PXR intensity was achieved except rare RF faults.

The stability is a great advantage in computed tomography (CT) experiments.

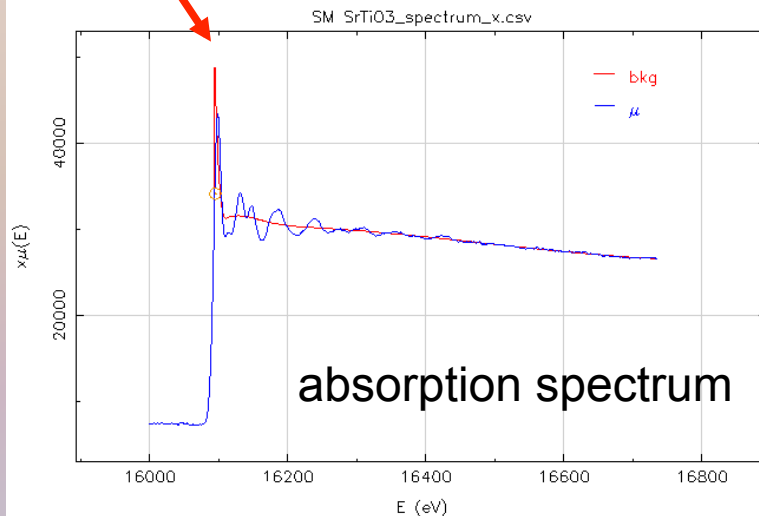
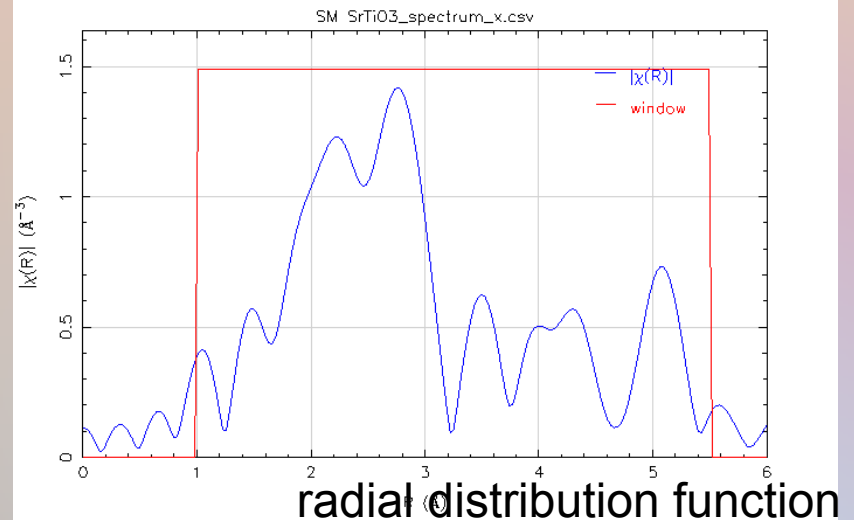
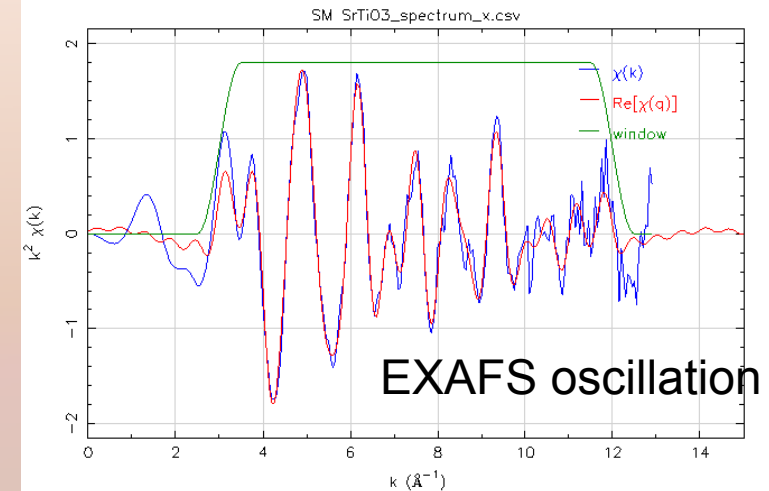
# Typical result of DXAFS experiment



sample  
 $\text{SrTiO}_3$  (white pigments)

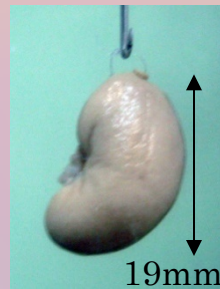
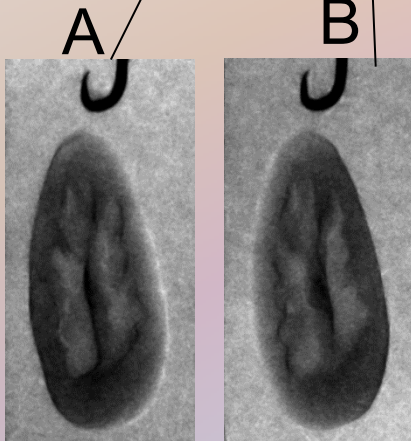
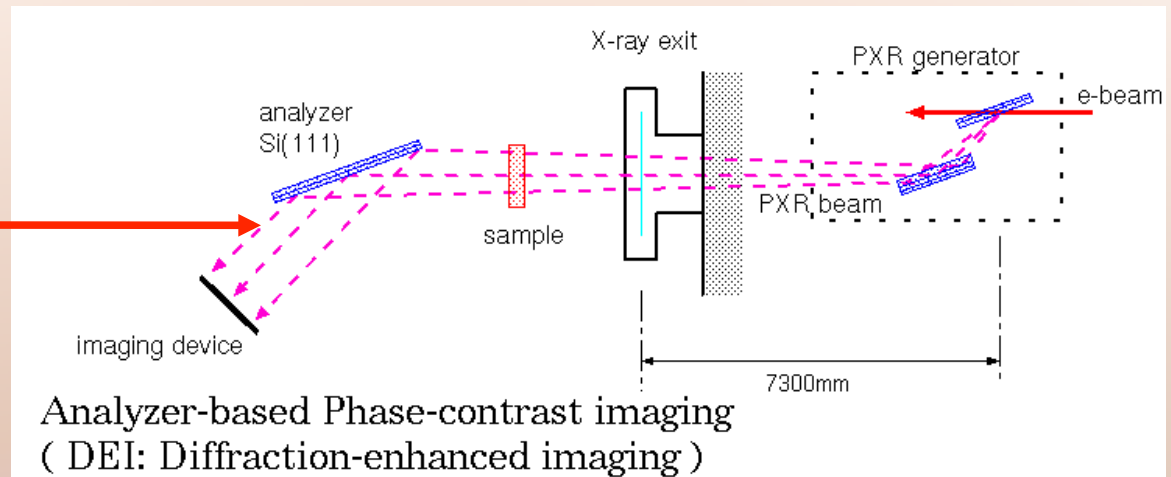
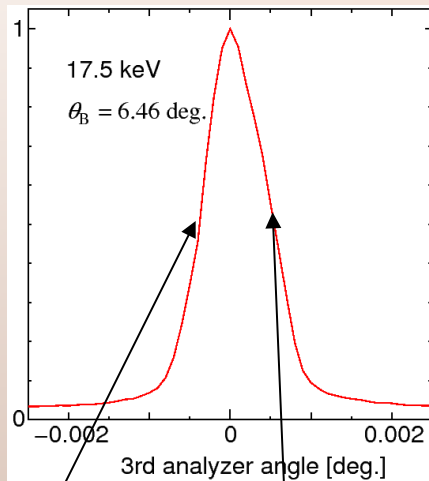
measurement time  
30min

detector: Imaging plate

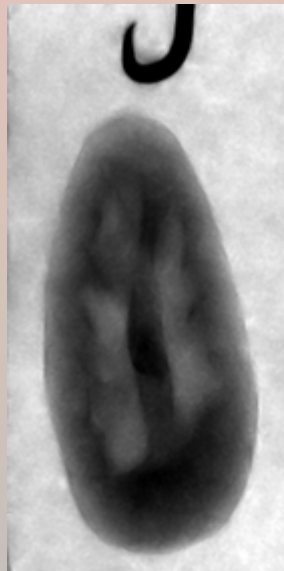


Time- resolved XAFS measurement can be expected using the linac-based source.

# Diffraction Enhanced Imaging (DEI)



kidney of  
mouse

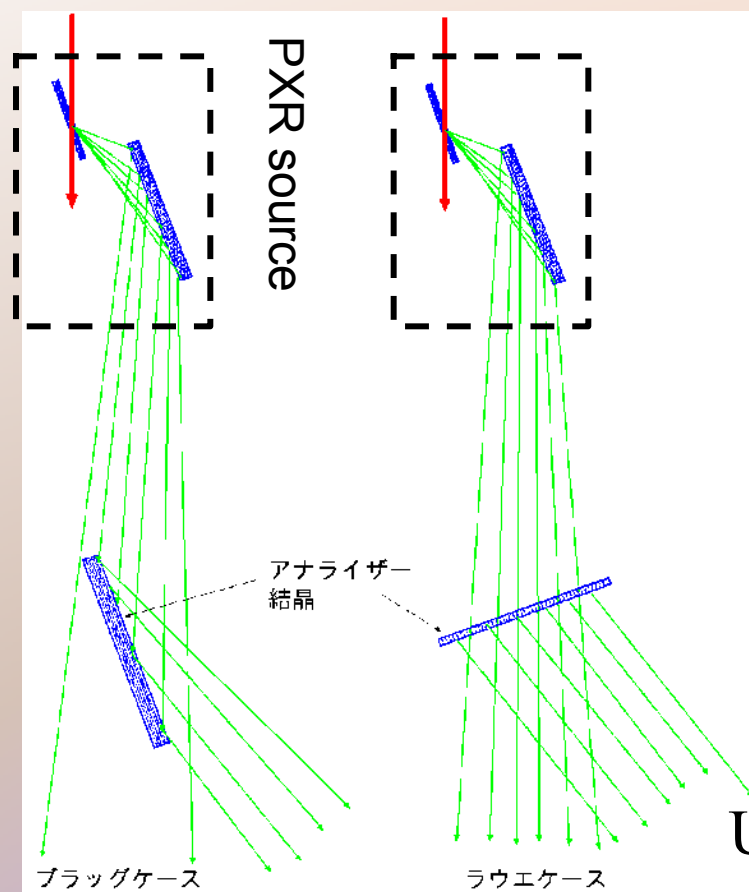


A+B:  
absorption contrast



A-B:  
phase contrast

# (+, -, +) arrangement



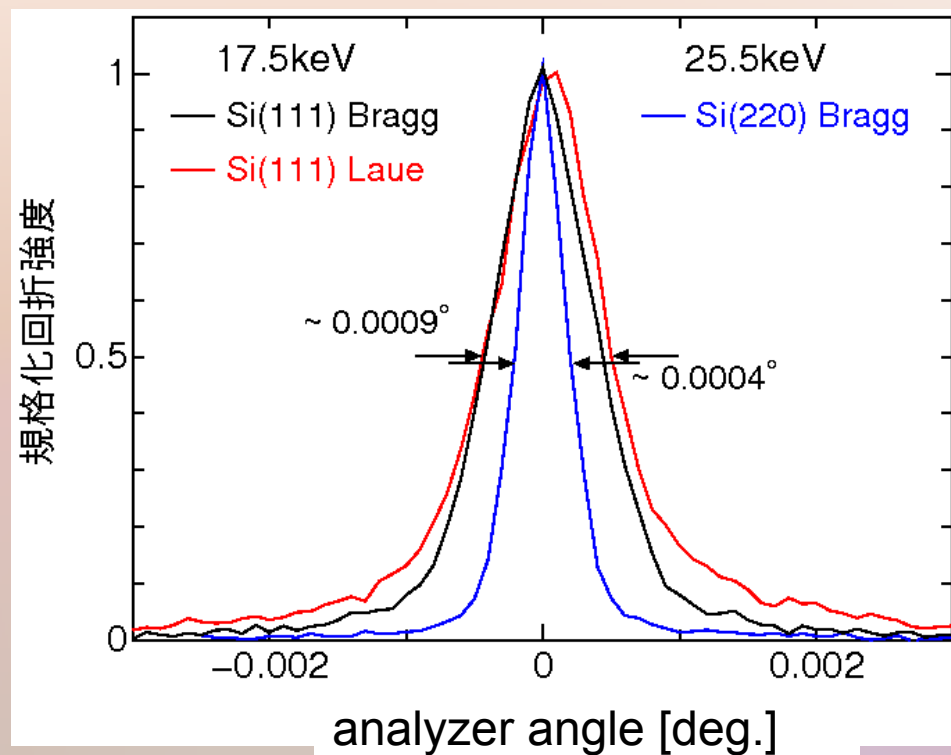
Bragg case

Laue case

Bragg angle:

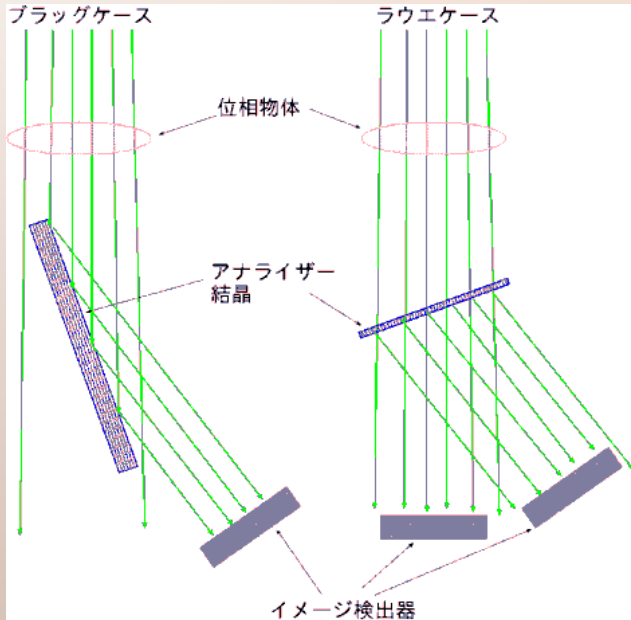
larger for longer wavelengths

smaller for shorter wavelengths (pseudo-plane wave)



Using a 3<sup>rd</sup> analyzer crystal in the (+, -, +) arrangement, the whole of a PXR beam can be diffracted with a narrow angular width despite the cone-beam.

# DEI in a Laue case



The irradiation field of DEI is limited by the size of the analyzer crystal.

Using an analyzer in a Laue case, the active area of 50mm x 50mm is actually available.

PXR radiator:  
Si(220)  
PXR energy:  
17.5keV

sample:  
pig eyeball



# SAXS contrast image

experimental condition:

PXR radiator: Si(220)

PXR energy: 23keV

sample: silica standard particles ( $\phi 1\mu\text{m}$ ,  $\phi 0.2\mu\text{m}$ )

X-ray camera: I.I. CCD ( $\text{QE} \lesssim 10\%$ )

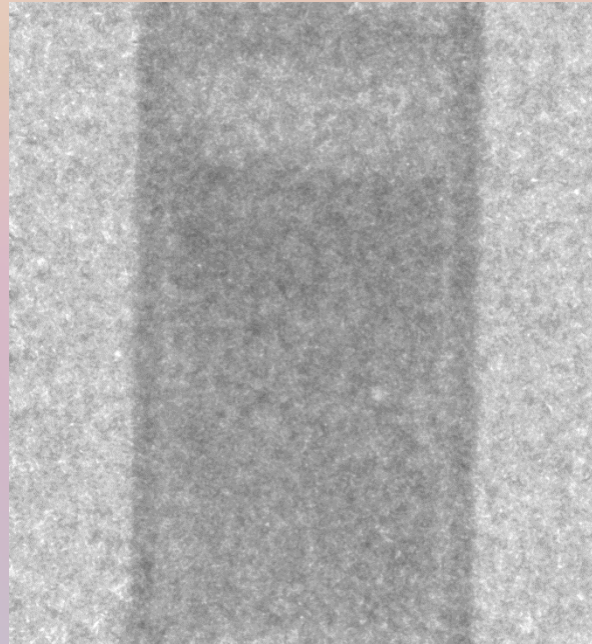


silica powder  
upper layer:

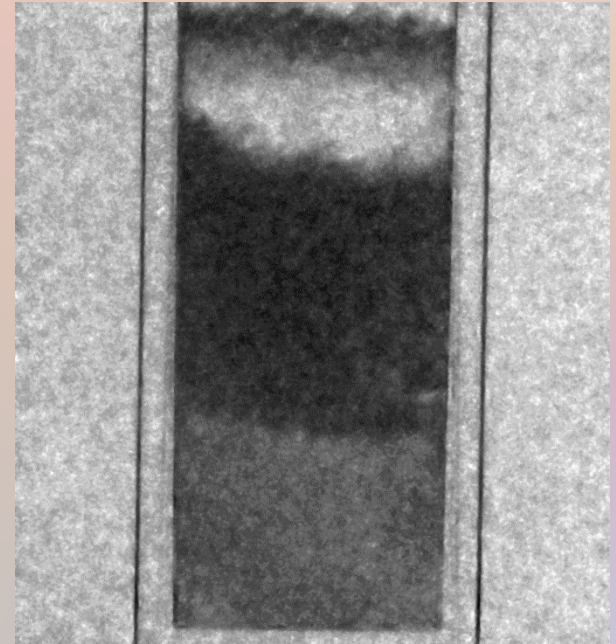
0.2 $\mu\text{m}$  in dia.

lower layer:

1.0 $\mu\text{m}$  in dia.



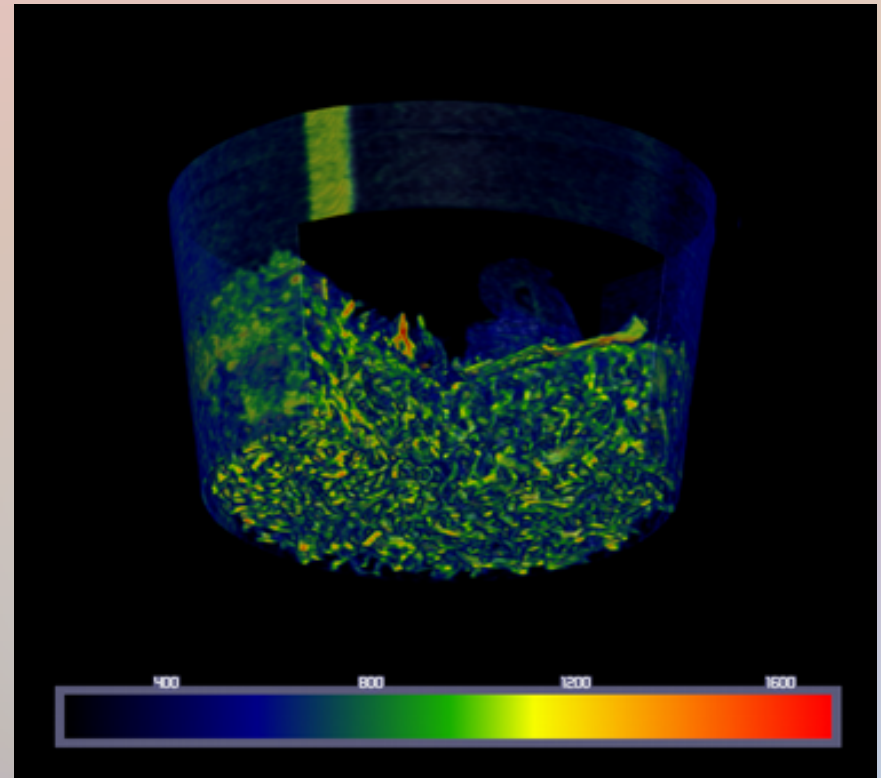
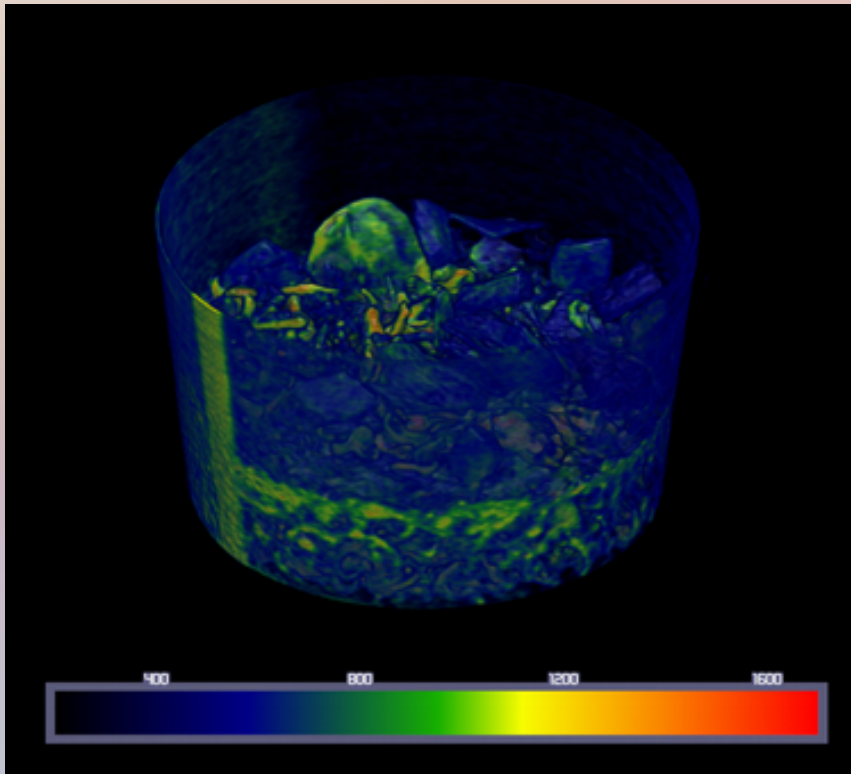
absorption contrast  
image



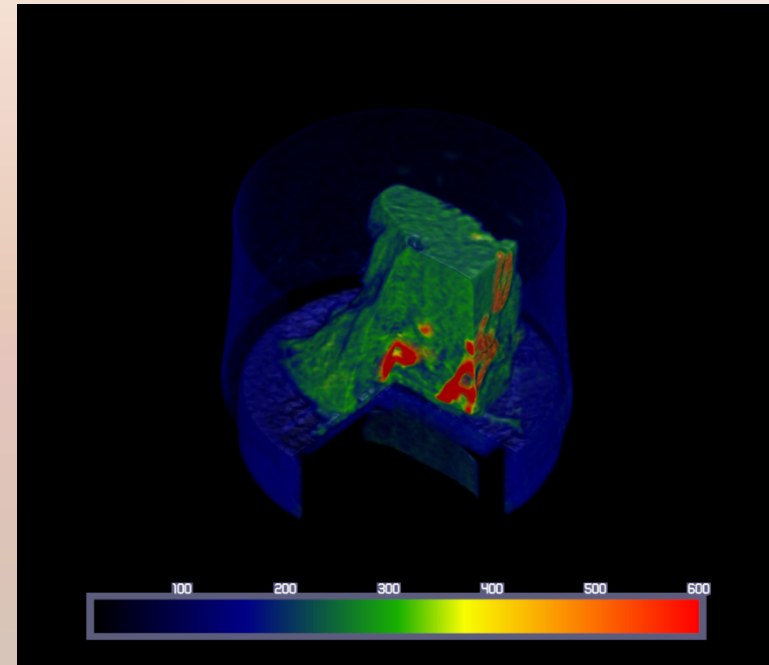
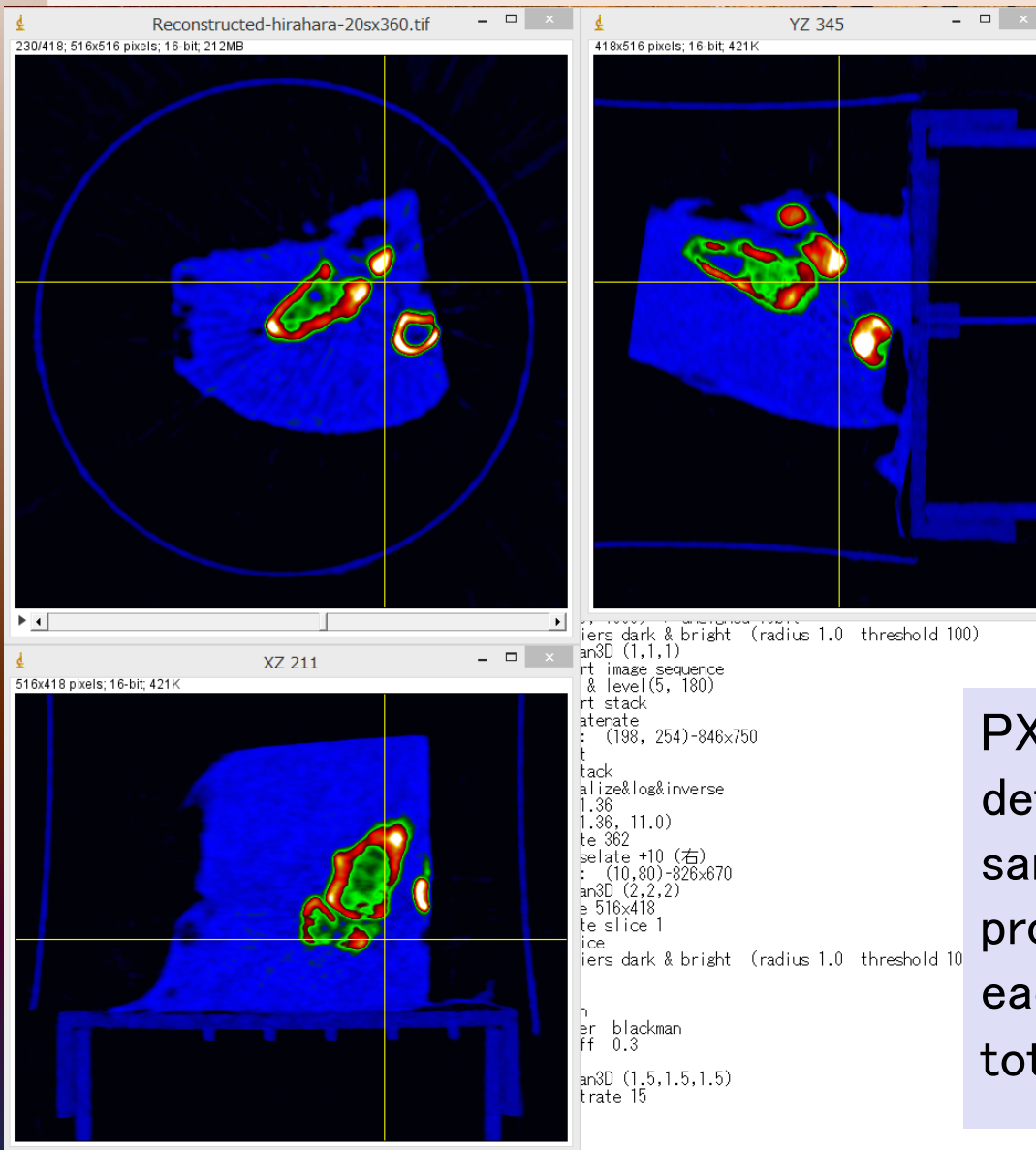
SAXS contrast  
image

# Large sample & Higher X-ray energy

PXR source: Si(220)      PXRenergy: 27keV  
sample: CUP-Noodle ( ~90mm in diameter )  
measurement time: 20s x 360 ( 2 hours )  
FPD: Shad-o-Box 1280 HS ( pixel size: 100 $\mu$ m )



# CT image of biological sample



PXR: 25keV

detector: Shad-o-Box 1280HS

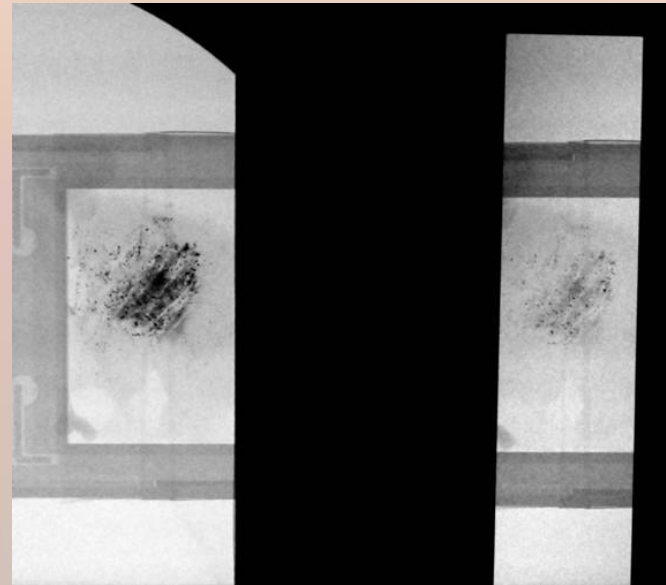
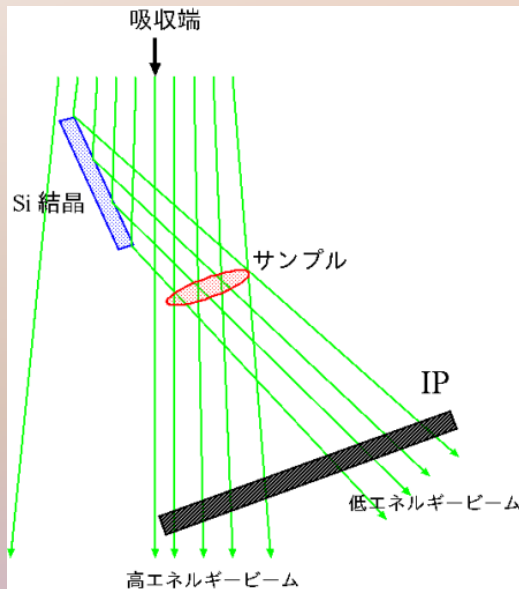
sample: tumor of dog forefoot

projection images: 360

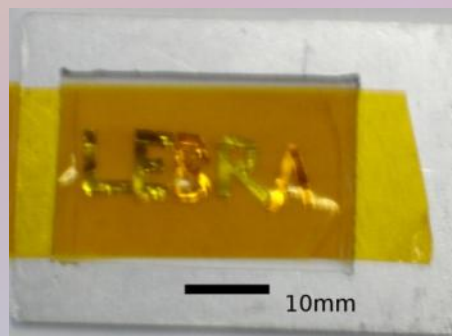
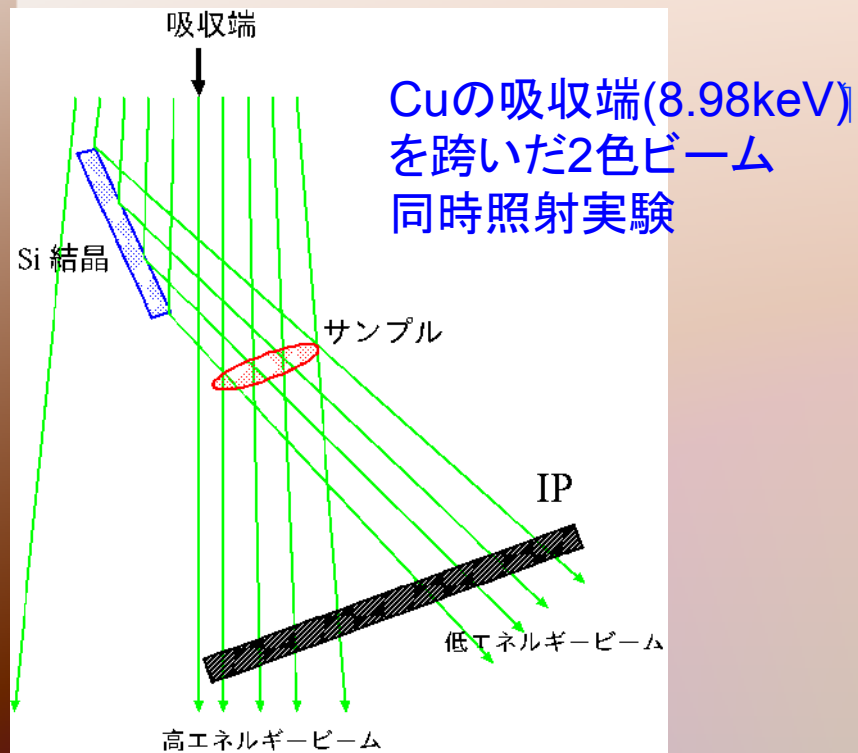
each exposure time: 20s

total measurement time: 2 hours

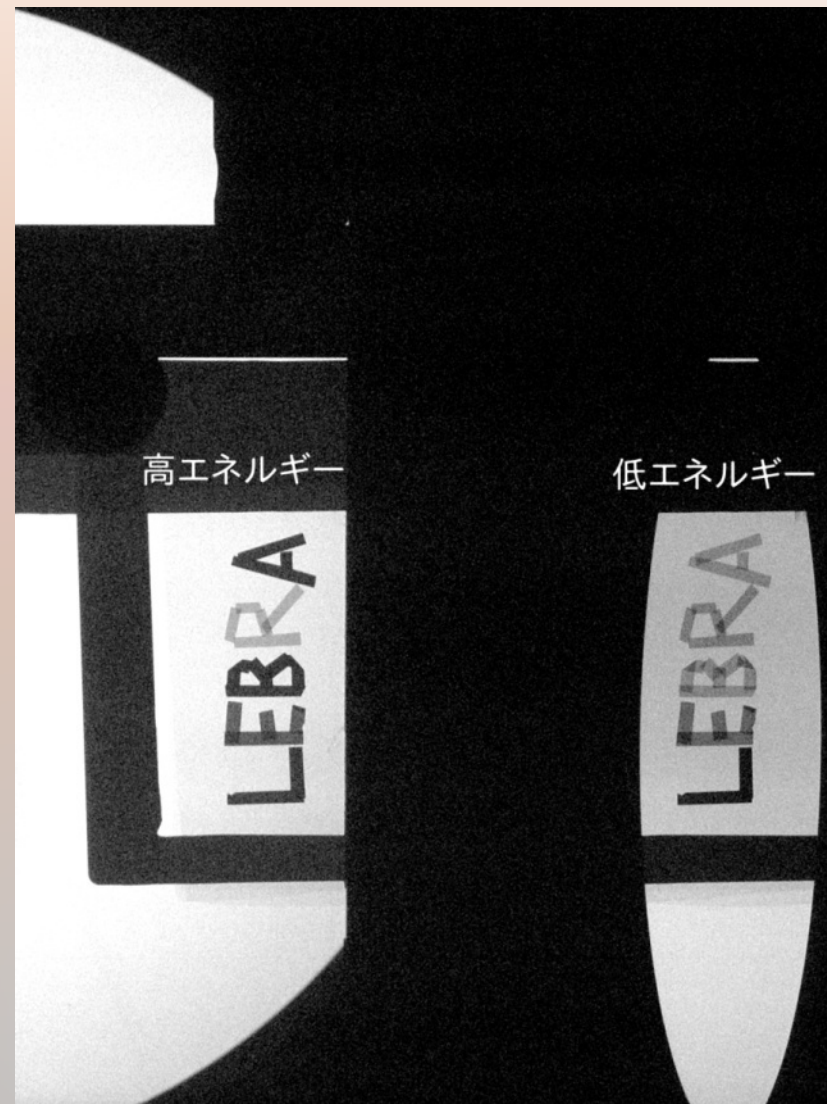
# Arsenic detection using 2-color X-ray beams



# 2色同時撮像による元素イメージング

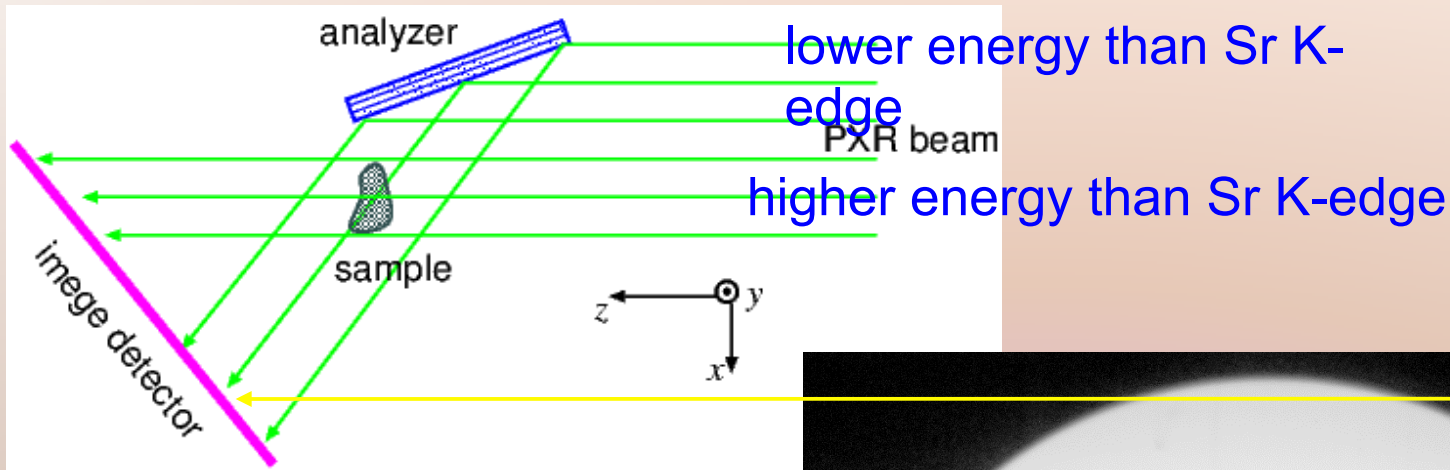


L : Ni (20 $\mu$ m)  
E : Ni-Cu (10 $\mu$ m)  
B, A : Cu (20 $\mu$ m)  
R : Zn (25 $\mu$ m)

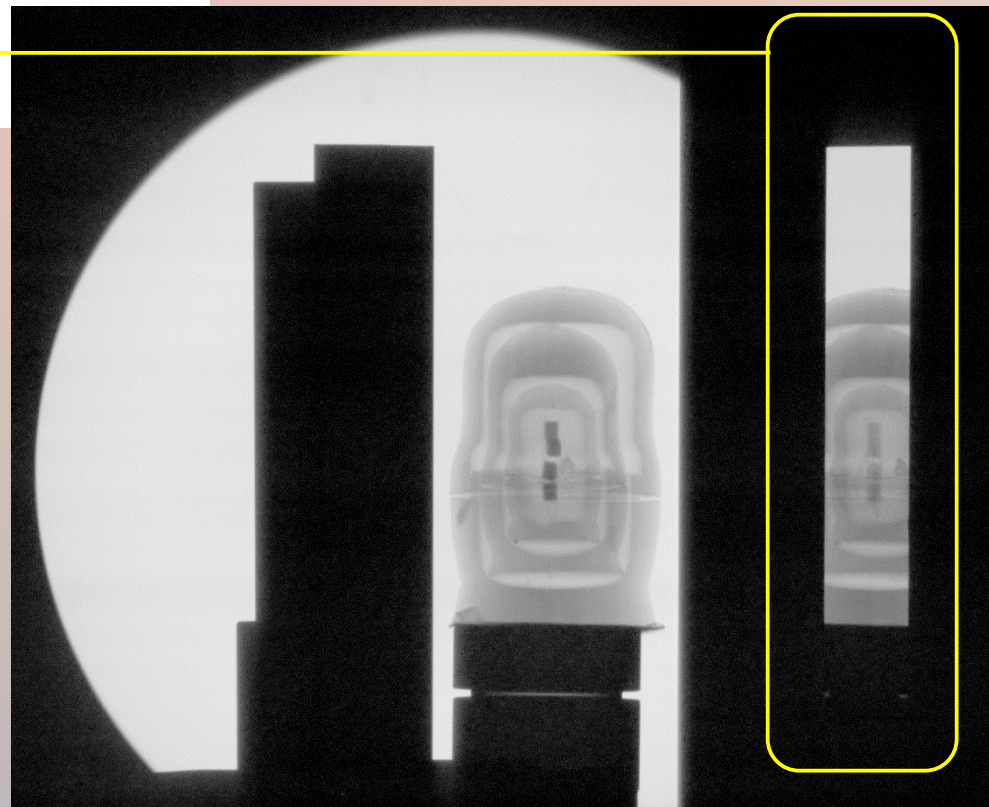


元素(Cu)検出を実証

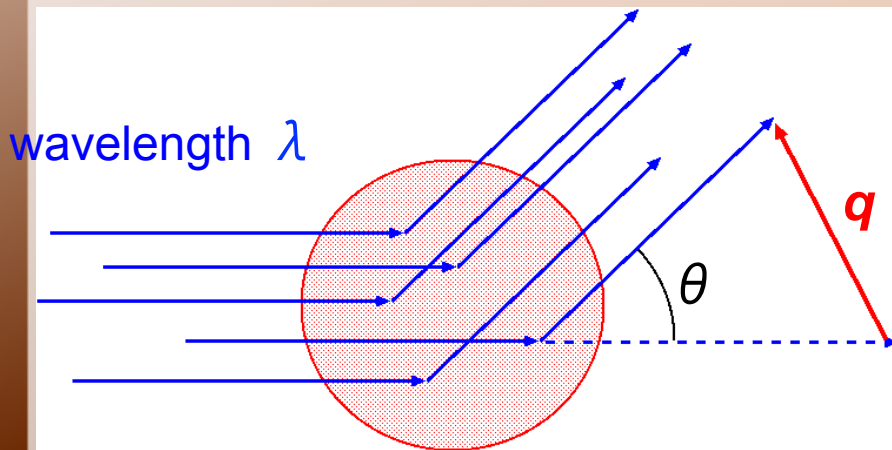
# Stereoscopic X-ray imaging



The technique of analyzer-based phase contrast imaging allows dichromatic crossing beam of X-rays across the absorption edge of specific element. It is useful for the element detection.



# Small angle X-ray scattering (SAXS)



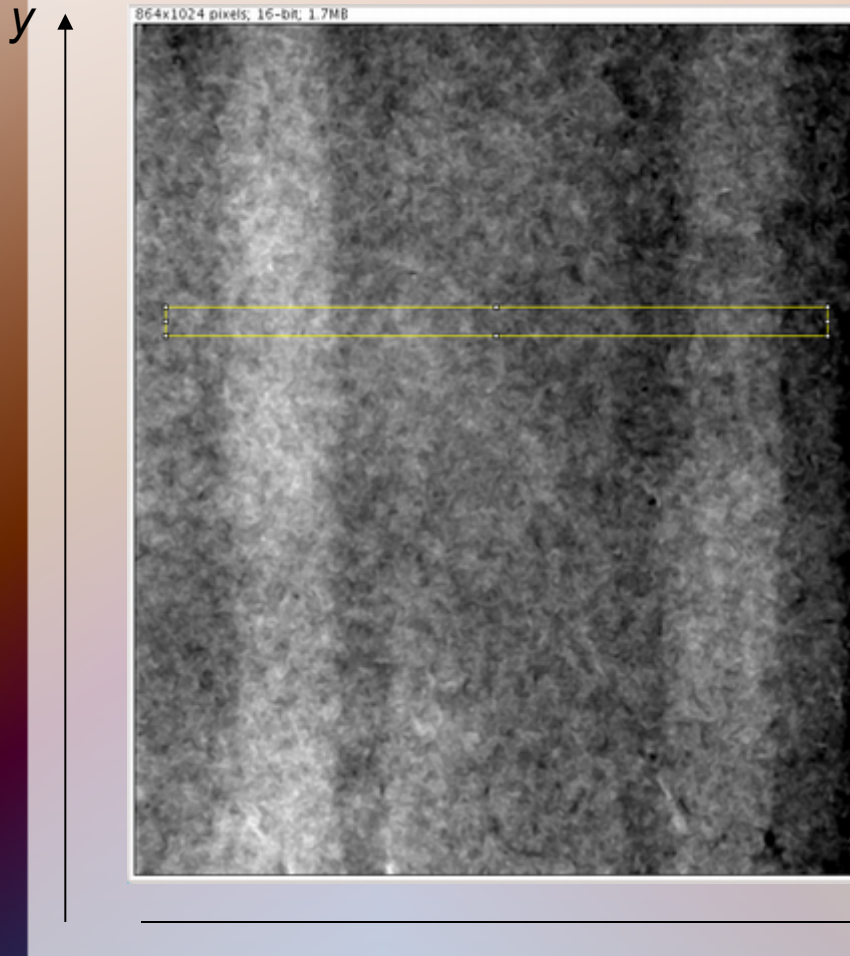
$$q = |\mathbf{q}| \\ = (4\pi / \lambda) \sin(\theta/2)$$

Guinier approximation:

$$I(q) = I_0 \exp\left(-\frac{1}{3} R_g^2 q^2\right)$$

$R_g$  : inertial (gyration) radius

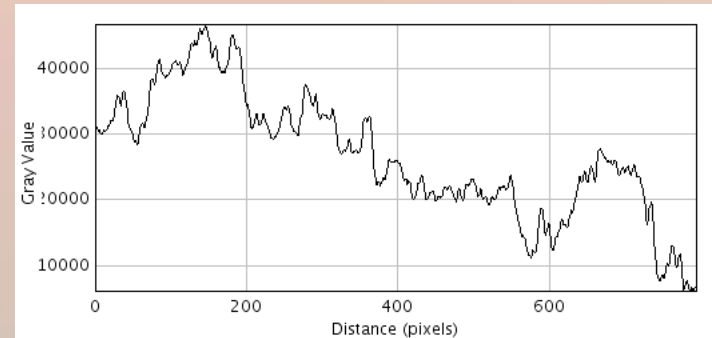
# absorption-contrast image



complex refractive index:

$$n(x,y) = 1 - \delta(x,y) + i \beta(x,y)$$

$\delta, \beta \propto \rho$  : density



→ x

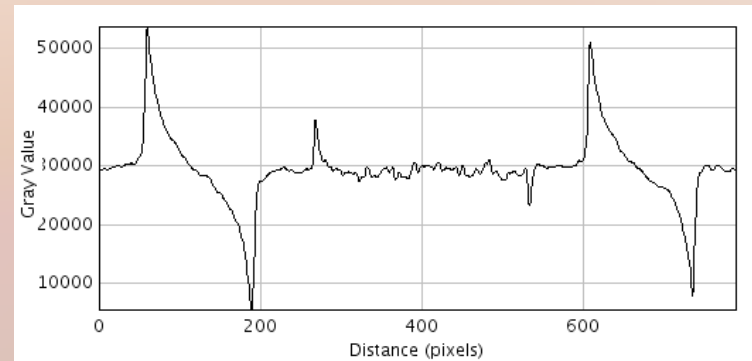
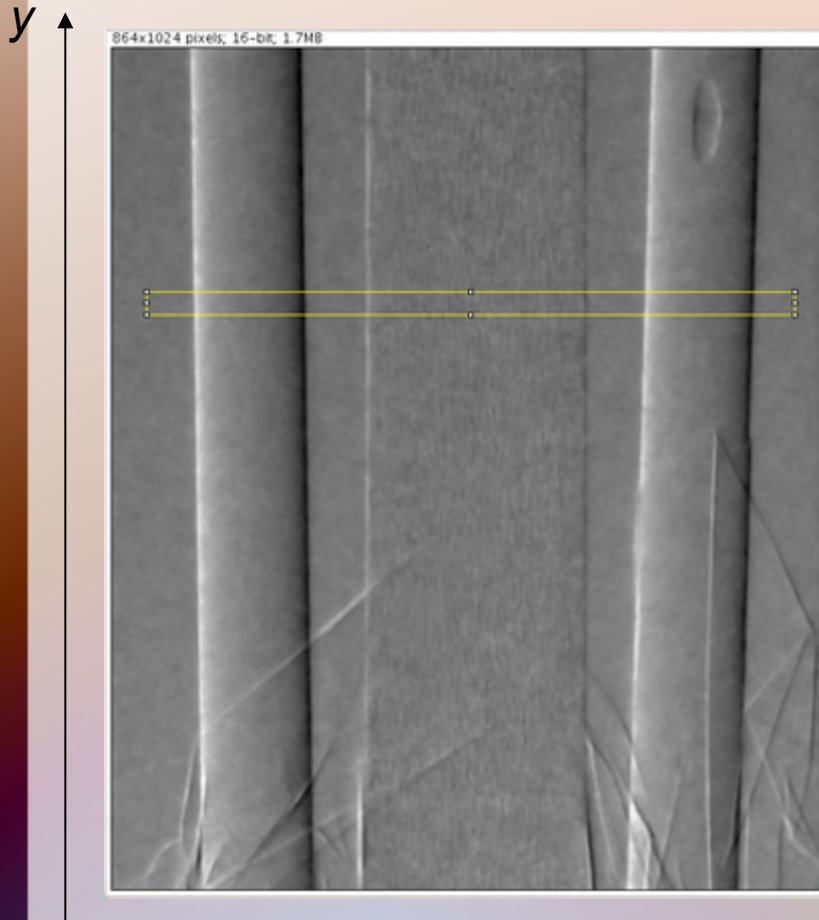
Integral with respect to  $\theta$

$$I_{\text{abs}} = \sum I(x,y, \theta)$$

$$\ln(I_{\text{abs}}(x,y)/I_0) \propto \beta(x,y)$$

$$\propto \rho(x,y)$$

# phase-gradient image

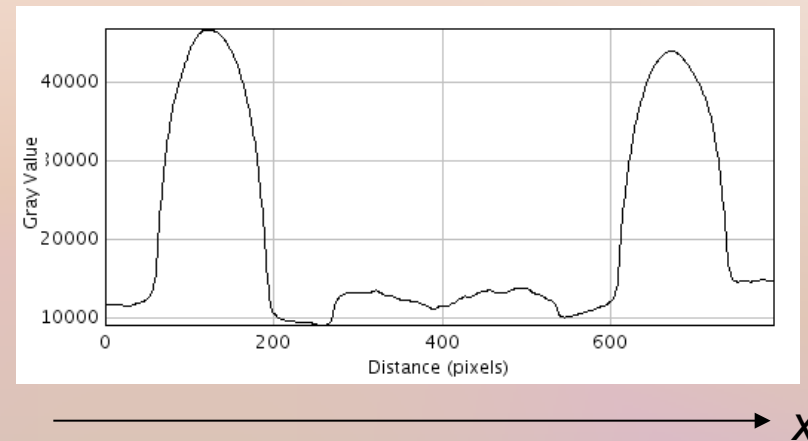
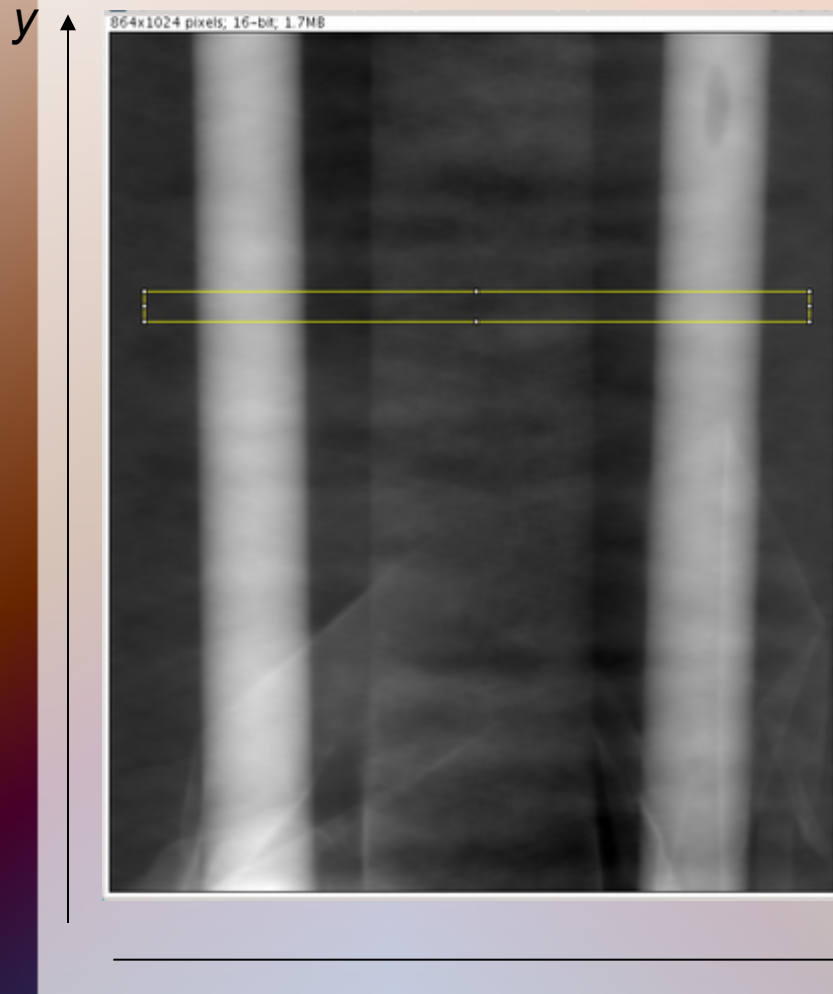


phase-gradient  
(refraction-contrast) map

$$\sum \theta I(x,y, \theta) / \sum I(x,y, \theta)$$

$$x \propto \partial \delta(x,y) / \partial x$$

# phase image

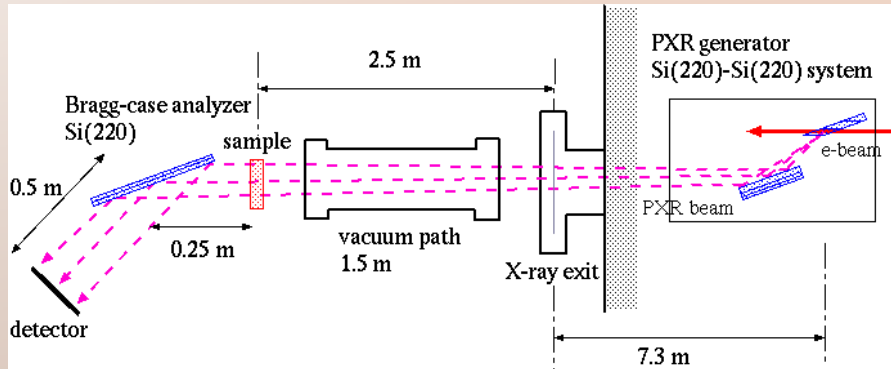


phase map

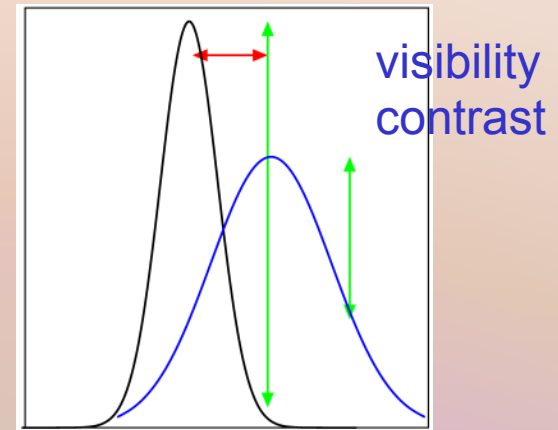
$$\delta(x,y) = \int \partial \delta(x,y) / \partial x \, dx$$

$$\propto \rho(x,y)$$

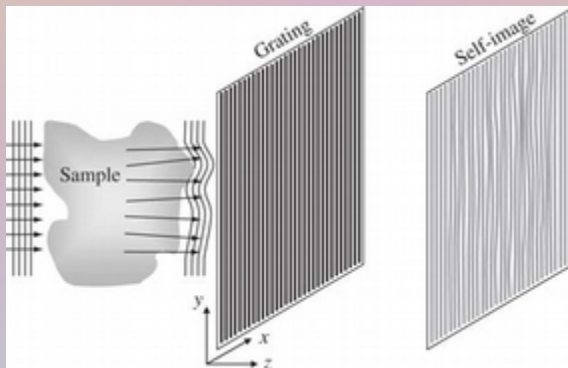
## diffraction-enhanced imaging



phase contrast

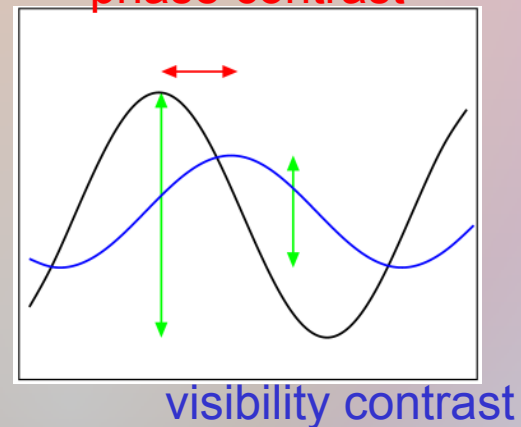


## Talbot interferometer imaging

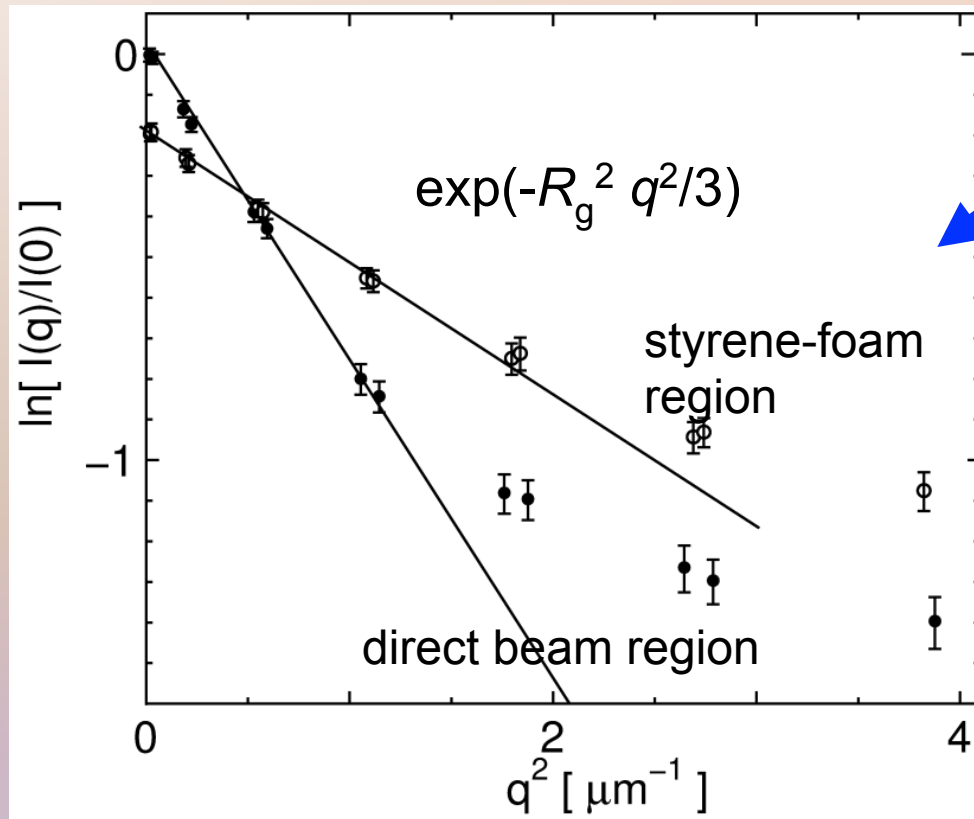


A. Momose et al.: JJAP 42 (2003) L866.

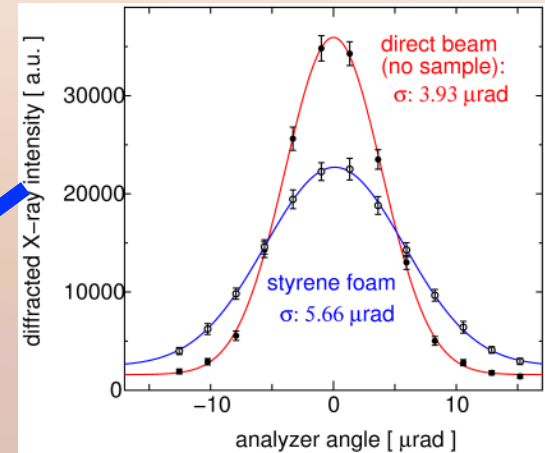
phase contrast



# Guinier plot



$$q = (4\pi / \lambda) \sin(\theta/2)$$



inertial radius  
 $R_g \sim 1 \mu\text{m}$   
 $< \text{pixel size } (24 \mu\text{m})$

For more exact estimation, the sample thickness has to be optimized.