Coherent X-Ray (Nanobeams)

& some applications

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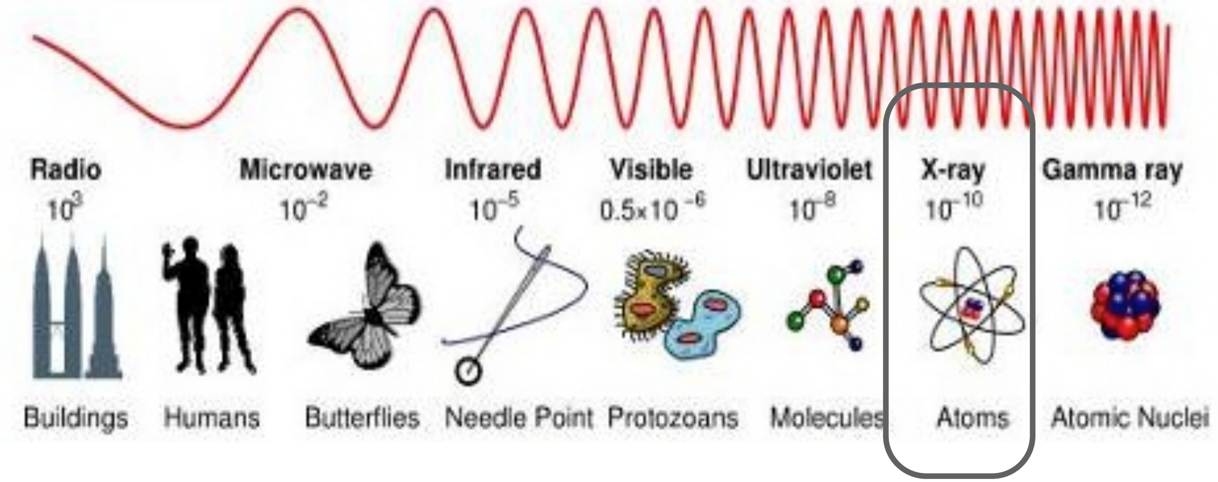
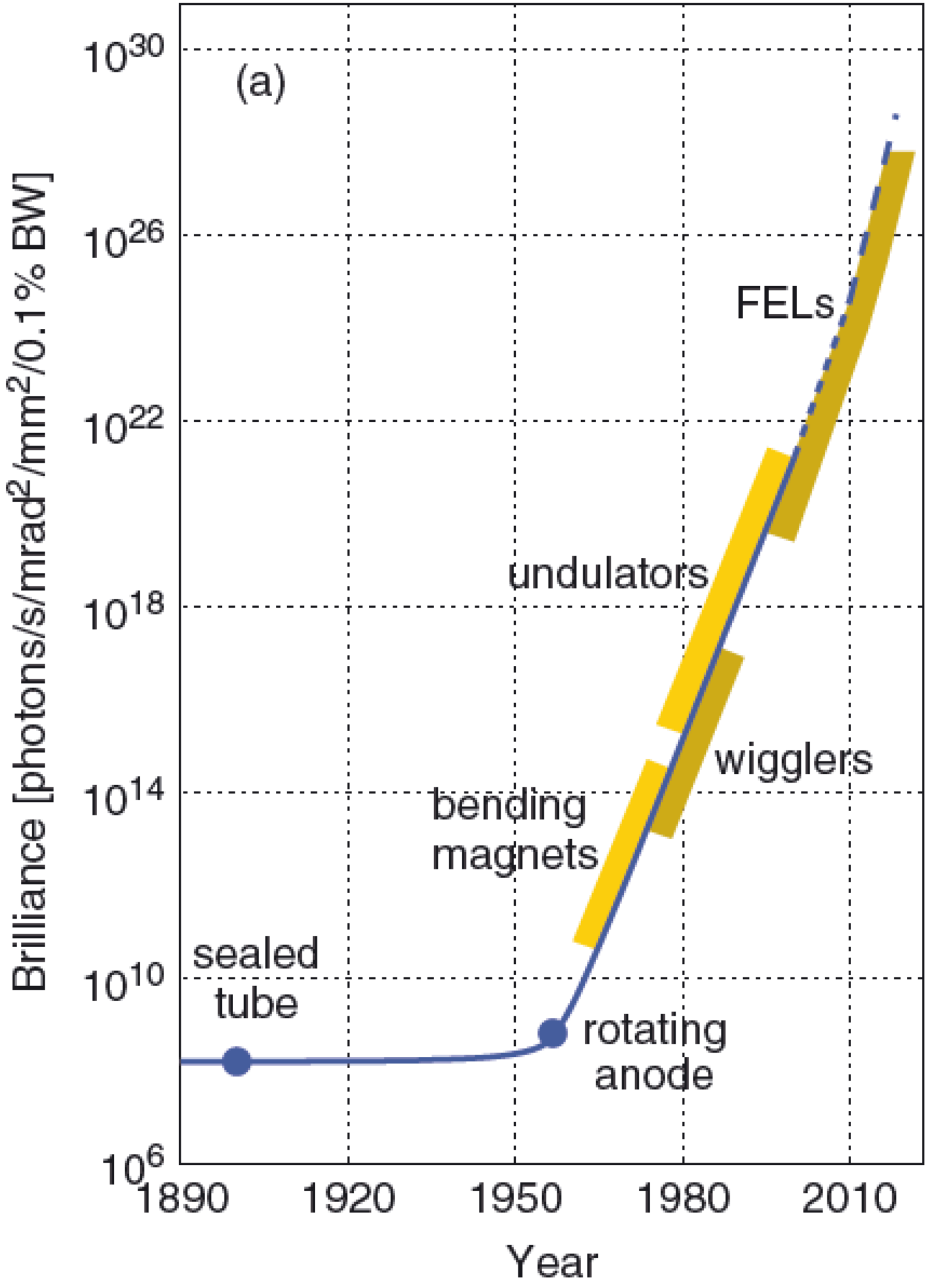
X-rays

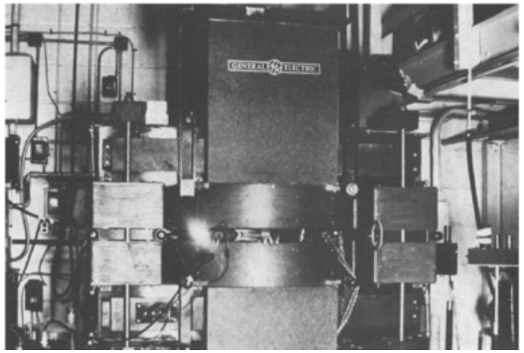


E= 100eV – 100 KeV
λ = *0.01 nm – 10 nm*
ω = **10^{16} Hz – 10^{19} Hz**

- *Energy*	*large penetration depth (inside materials, buried layers), elemental sensitivity*
- *Wavelength*	*interatomic distances, morphology, crystal structure, meso-scale domains*
- *No special environment*	*In-situ & operando : kinetics & dynamics (growth, annealing, erosion, rections indentation, external fields, catalysis, wet, hot, cold …)*

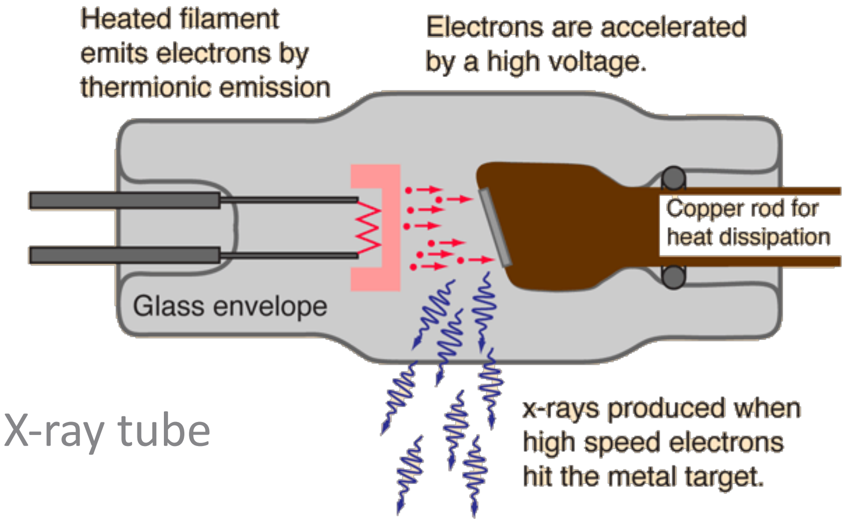
X-Ray Sources





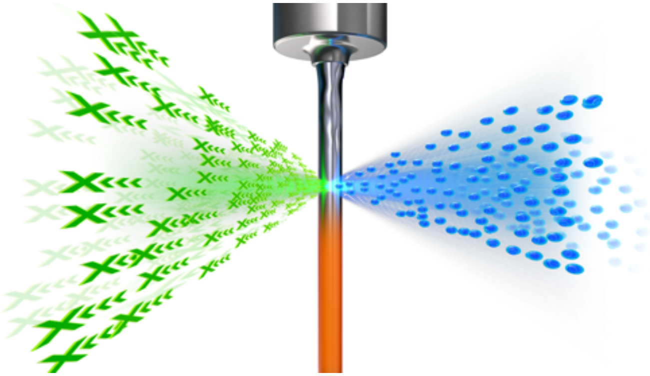
Elder, F.R., Phys Rev **71** (1947)

NOTE: Improvement of Laboratory sources

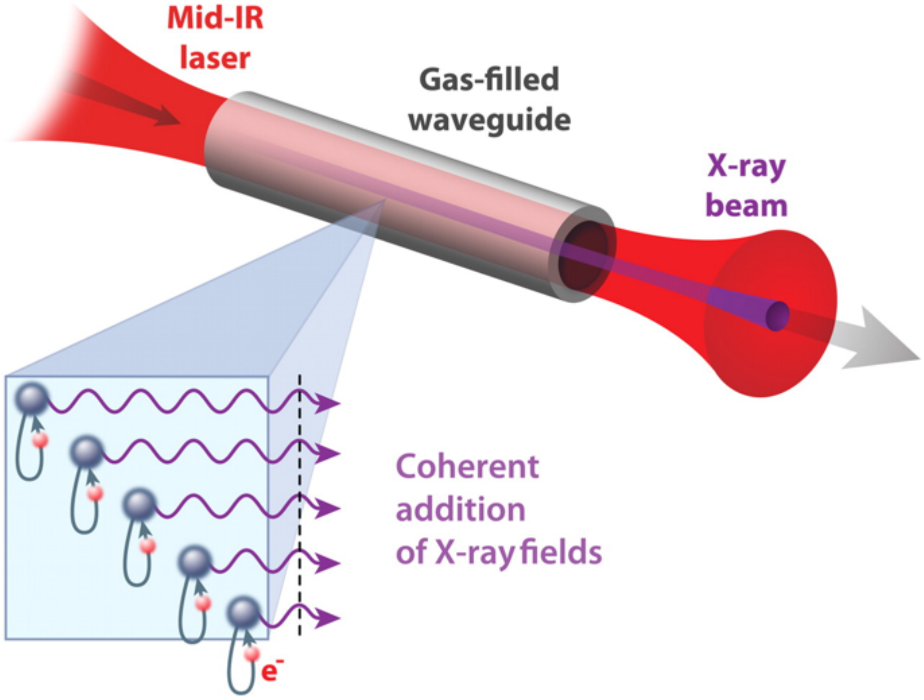


X-ray tube

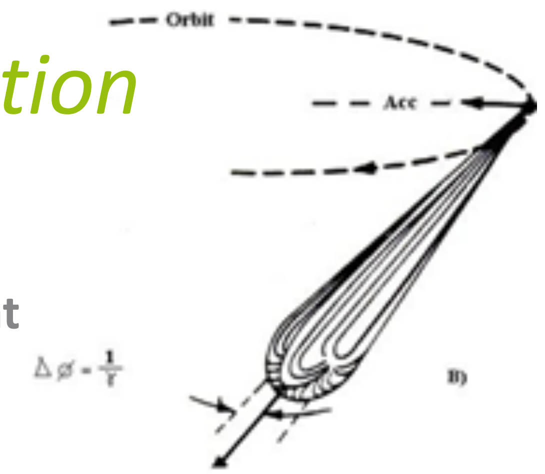
X-ray Metal Jet source, small source and more flux in Tender-Hard regime



X-ray High-Harmonic Generation: more coherence in Soft- regime



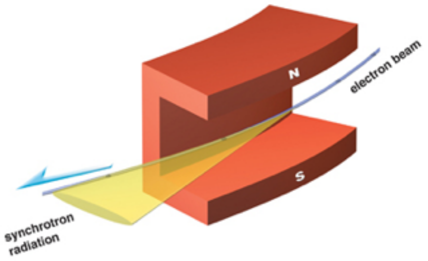
Synchrotron Radiation

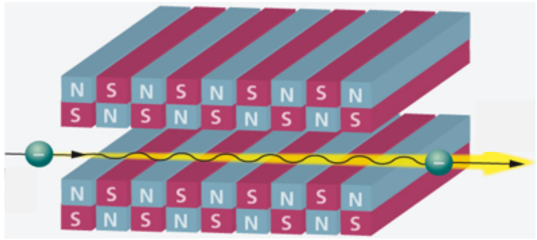


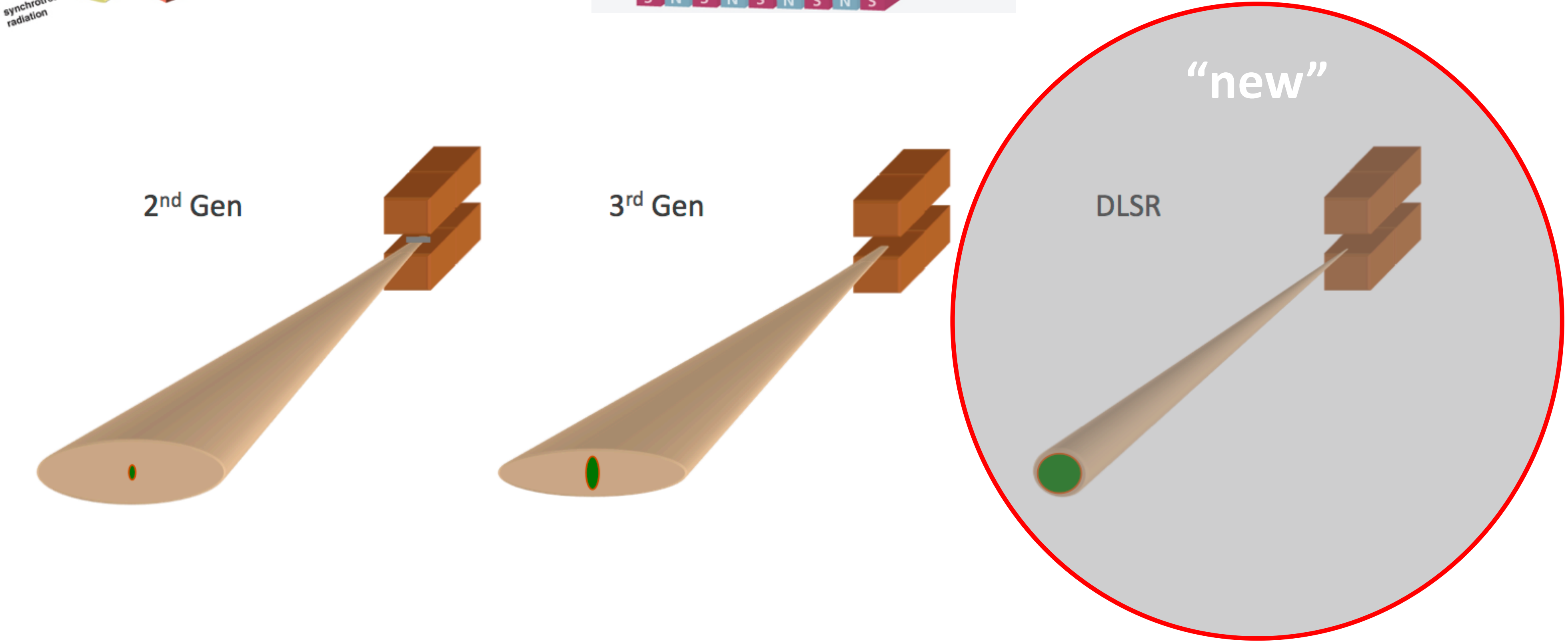
Electromagnetic radiation produced by relativistic electrons that are bent (accelerated) in a magnetic field and thus producing a radiation with an energy ranging from infrared to hard X-rays.

- **Brilliant** - many orders of magnitude brighter than conventional sources.
- **Tunable** – varying Energy of X-ray beam
- **Collimated** – parallel beam, easy to focus, permits studies of complex structures.
- **Polarized** - improves sensitivity
- **Pulsed** - electron bunches produce light pulses (time-resolved experiments)
- **Coherent** - well defined source gives a high degree of coherence

Synchrotron sources





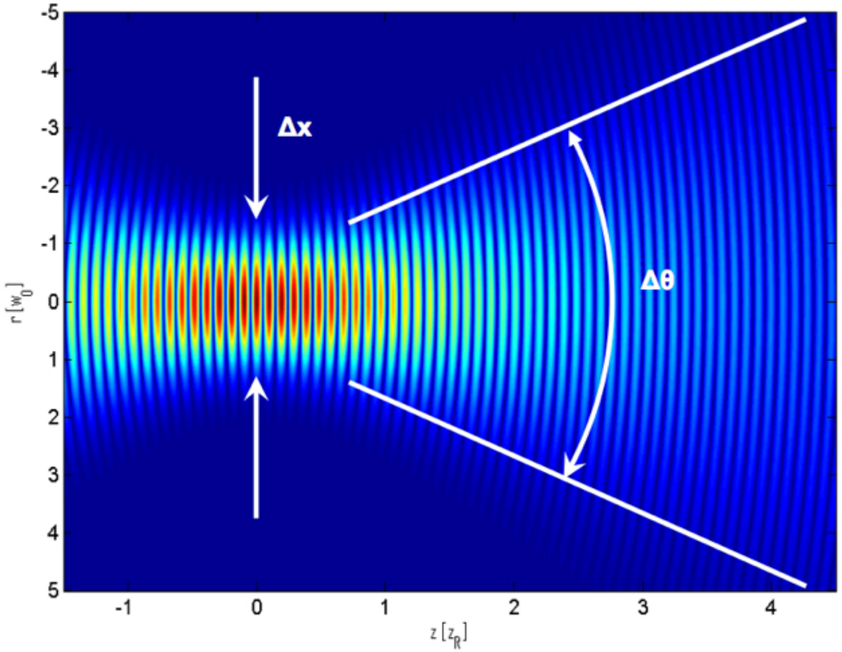


Large emittance, low brilliance,
small coherent fraction and flux

Small emittance, high brilliance,
large coherent fraction and flux

Why are DLSs so Bright ?

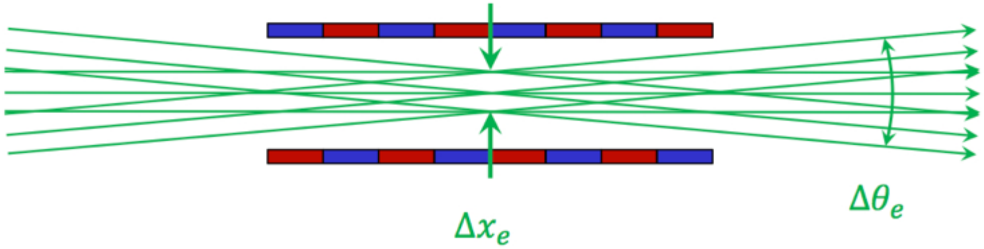
Photon Beam



Photon emittance limit

$$\varepsilon_p = \Delta x_p \Delta\theta_p \geq \frac{\lambda}{4\pi}$$

Electron Beam (in undulator)



electron emittance

$$\varepsilon_e = \Delta x_e \Delta\theta_e$$

In a DLSR light source (e.g. synchrotron)

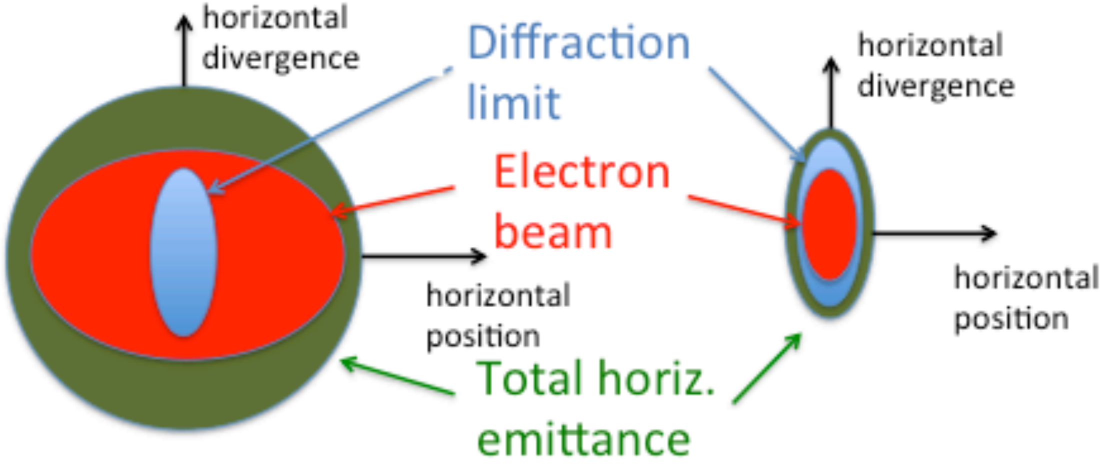
$$\varepsilon_e \leq \varepsilon_p$$

Small & collimated e-beam

Courtesy of Dan Mannix, U. Aarhus, ESS & CNRS

Transverse Coherence of Diffraction Limited Storage Ring Sources

- **Fundamental limit of source size and divergence depends only on wavelength**



$$\varepsilon_r = \text{diffraction limited emittance} = \sigma_\gamma \sigma'_\gamma = \frac{\lambda}{4\pi} = \begin{cases} 80 \text{ pm @ 1 keV} \\ 8 \text{ pm @ 10 keV} \end{cases}$$

- **Coherent fraction = ratio of diffraction-limited emittance to total emittance**

$$f_{coh} = \frac{F_{\mathrm{coh,T}}(\lambda)}{F(\lambda)} = \frac{\sigma_\gamma \sigma'_\gamma}{\underbrace{\sigma_{\mathrm{Tx}} \sigma_{\mathrm{Tx'}}}} \frac{\sigma_\gamma \sigma'_\gamma}{\sigma_{\mathrm{Ty}} \sigma_{\mathrm{Ty'}}}$$

Courtesy of Paul Evans, Wisconsin University

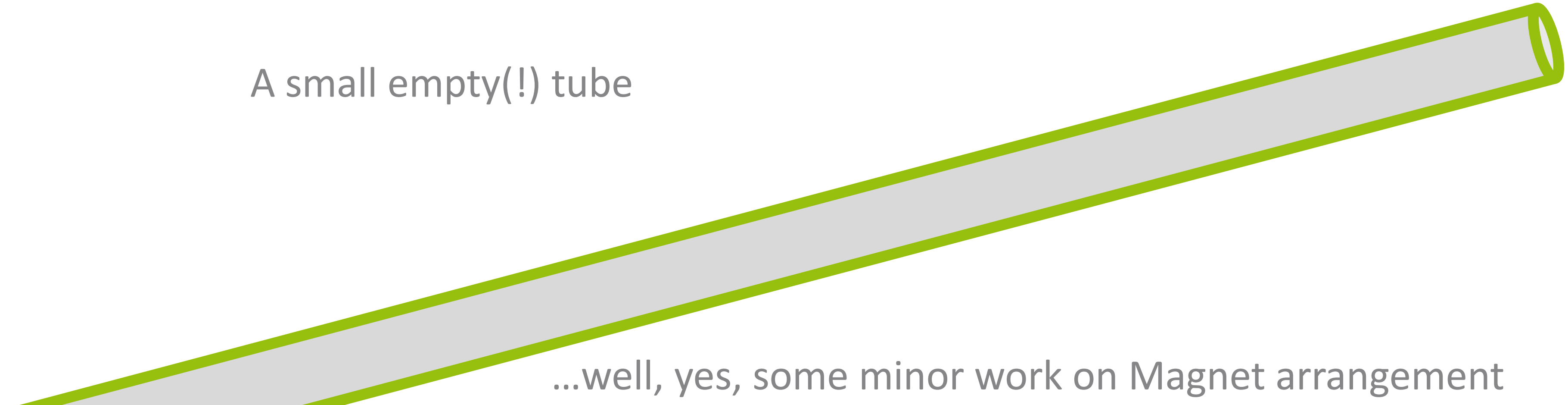
University of Wisconsin-Madison

NSLS-II First Girder:
14-foot, 8-ton structure
holding multiple magnets
June 15, 2010
ε_0= 1 nm rad
Pushing 3rd gen technology

MAX IV 7-BA magnet (2016):
Dipole & multipol. magnets
$\varepsilon_0 < 0.3$ nm rad

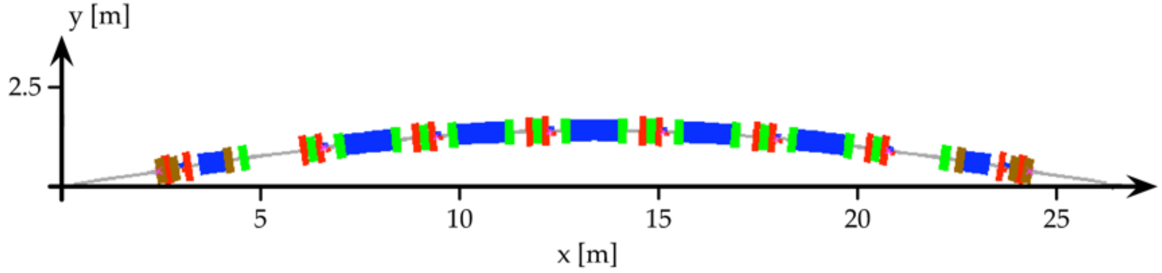
7 x 20 weak (0.5 T) bend-magnets
many 10^2 strong focusing magnets
4th gen, innovative technology

Technological breakthrough?



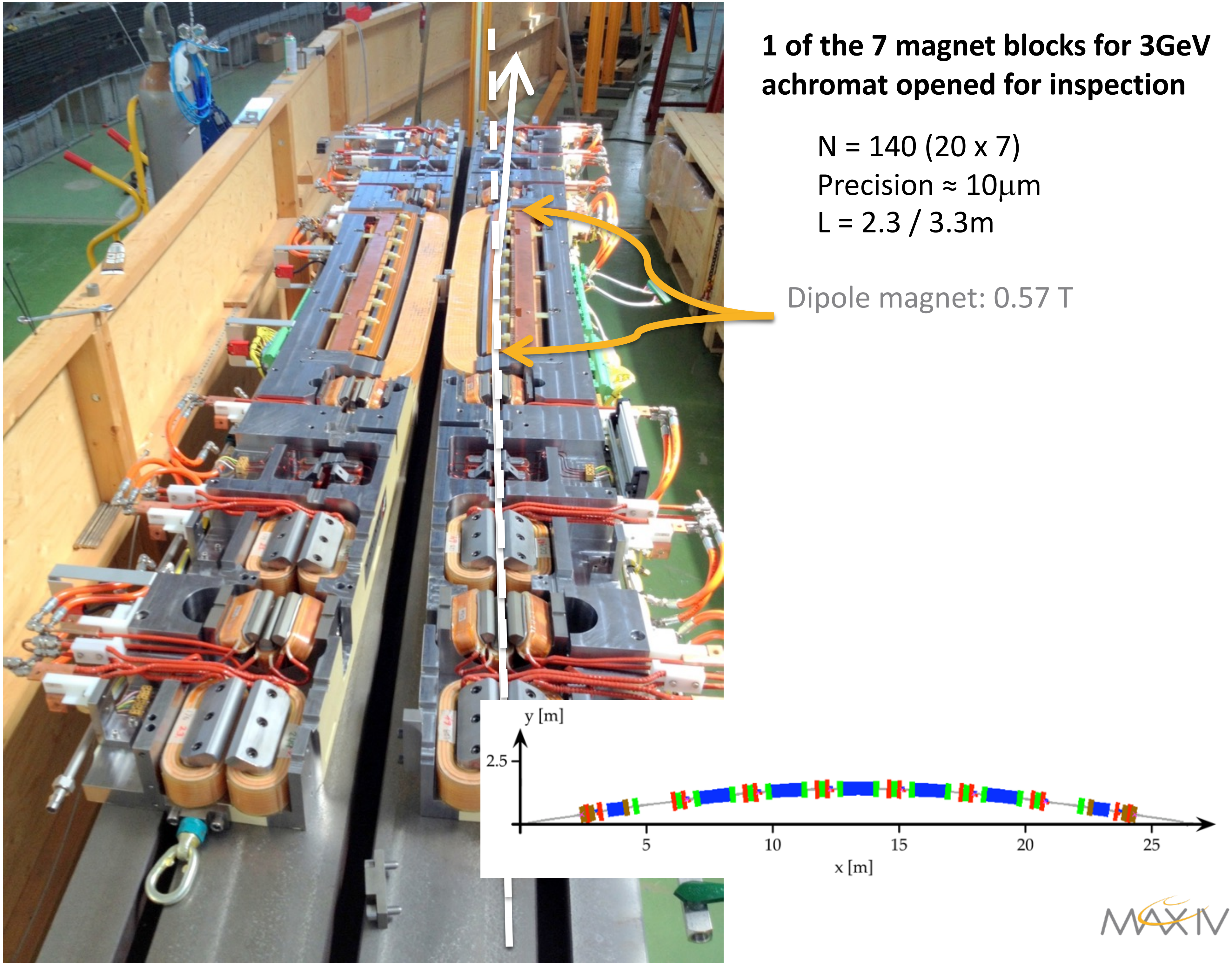
A small empty(!) tube

…well, yes, some minor work on Magnet arrangement

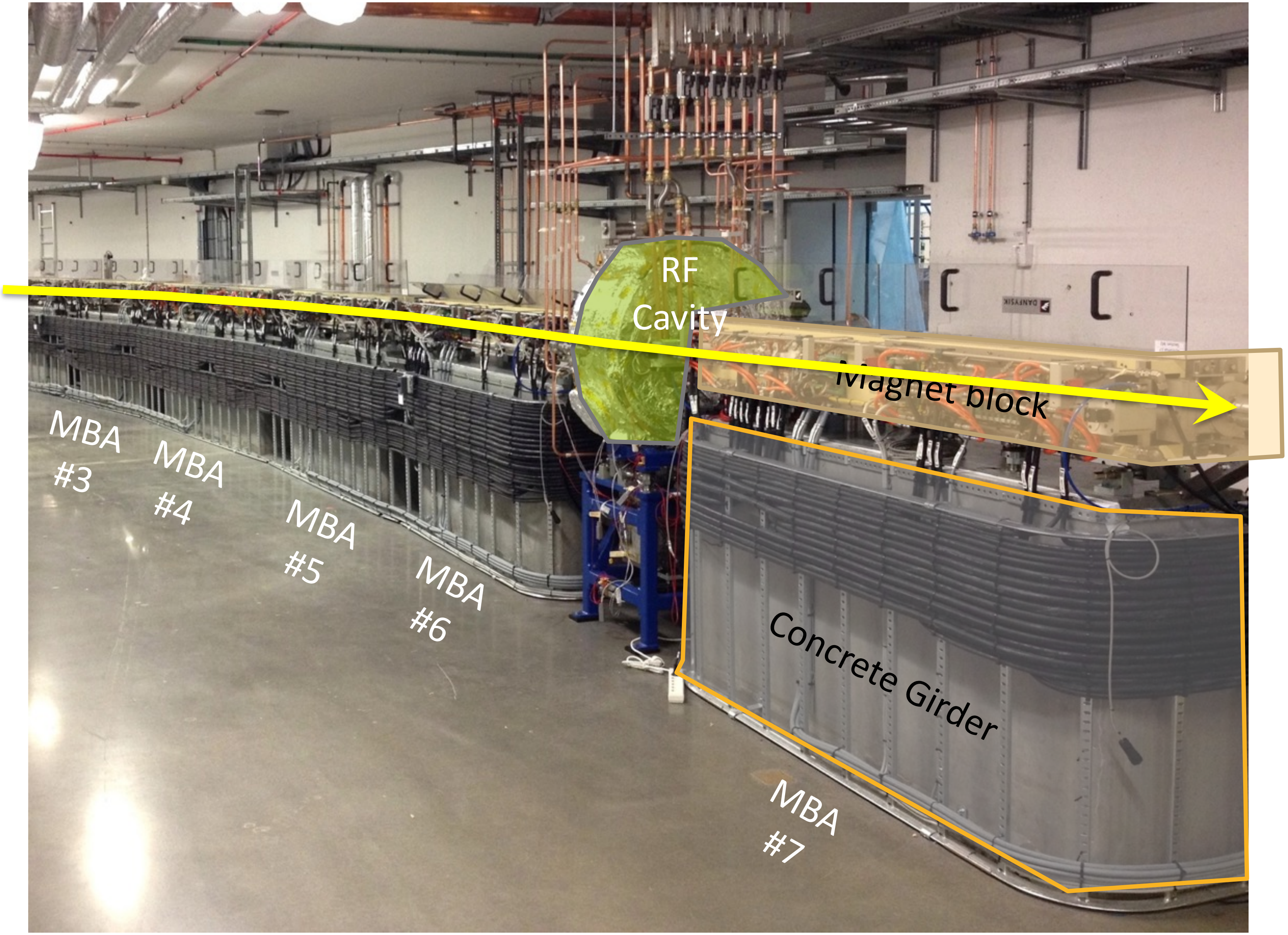


1 of the 7 magnet blocks for 3GeV achromat opened for inspection

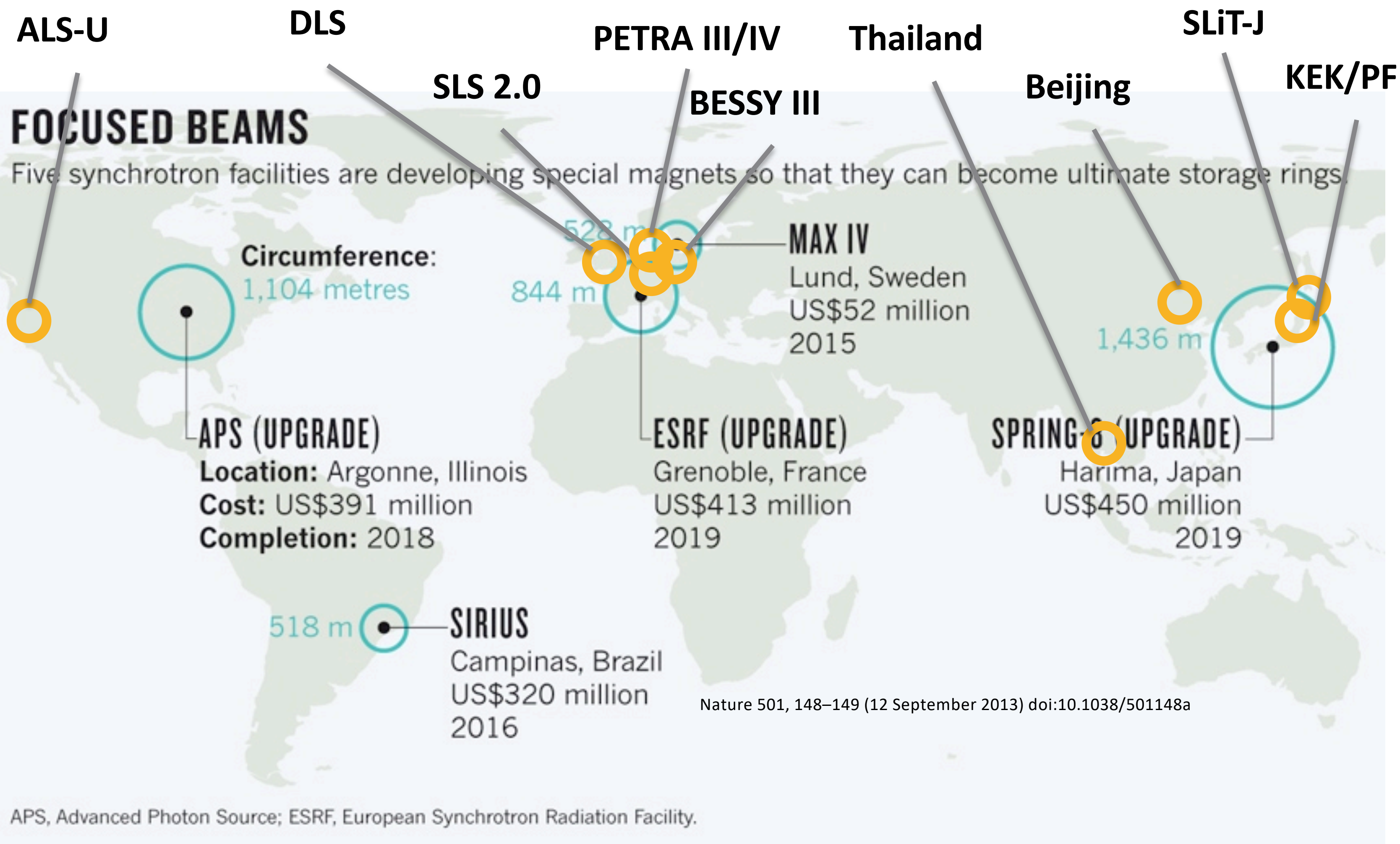
N = 140 (20 x 7)
Precision ≈ 10µm
L = 2.3 / 3.3m


Dipole magnet: 0.57 T

MAX IV 3 GeV Ring

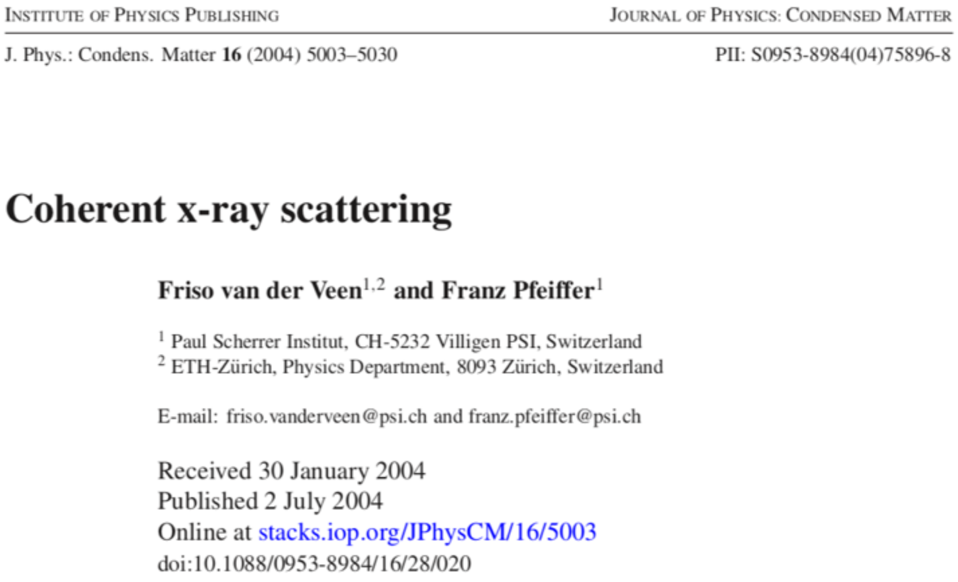


International Scene: DLSR’s all over the world



Nature 501, 148–149 (12 September 2013) doi:10.1038/501148a

Coherence: what ?



INSTITUTE OF PHYSICS PUBLISHING — JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **16** (2004) 5003–5030 — PII: S0953-8984(04)75896-8

Coherent x-ray scattering

Friso van der Veen[1,2] **and Franz Pfeiffer**[1]

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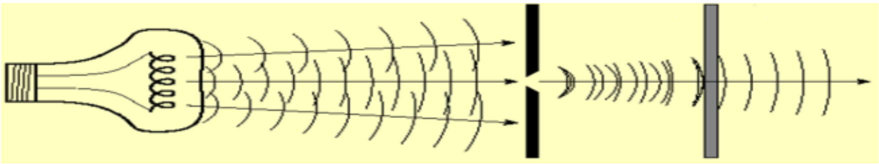
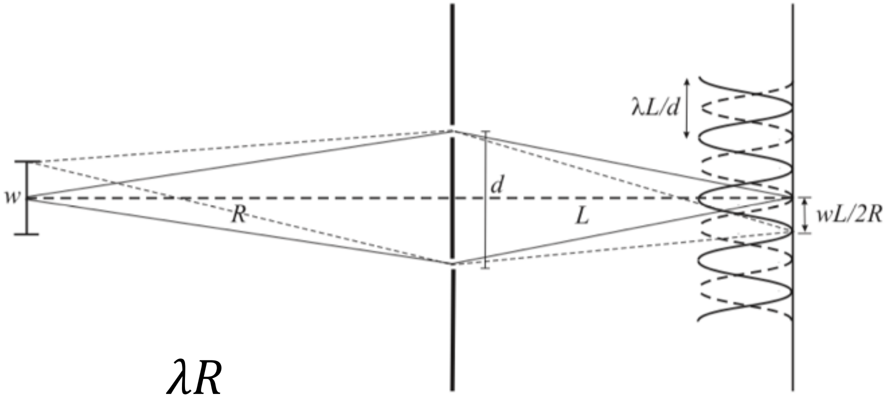
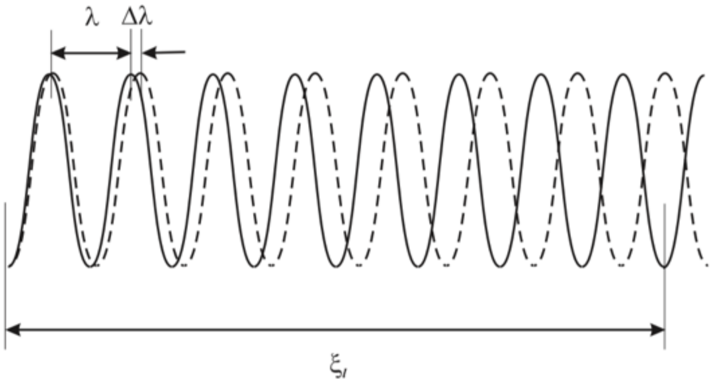
doi:10.1088/0953-8984/16/28/020





$$\xi_T = \frac{\lambda R}{w}$$

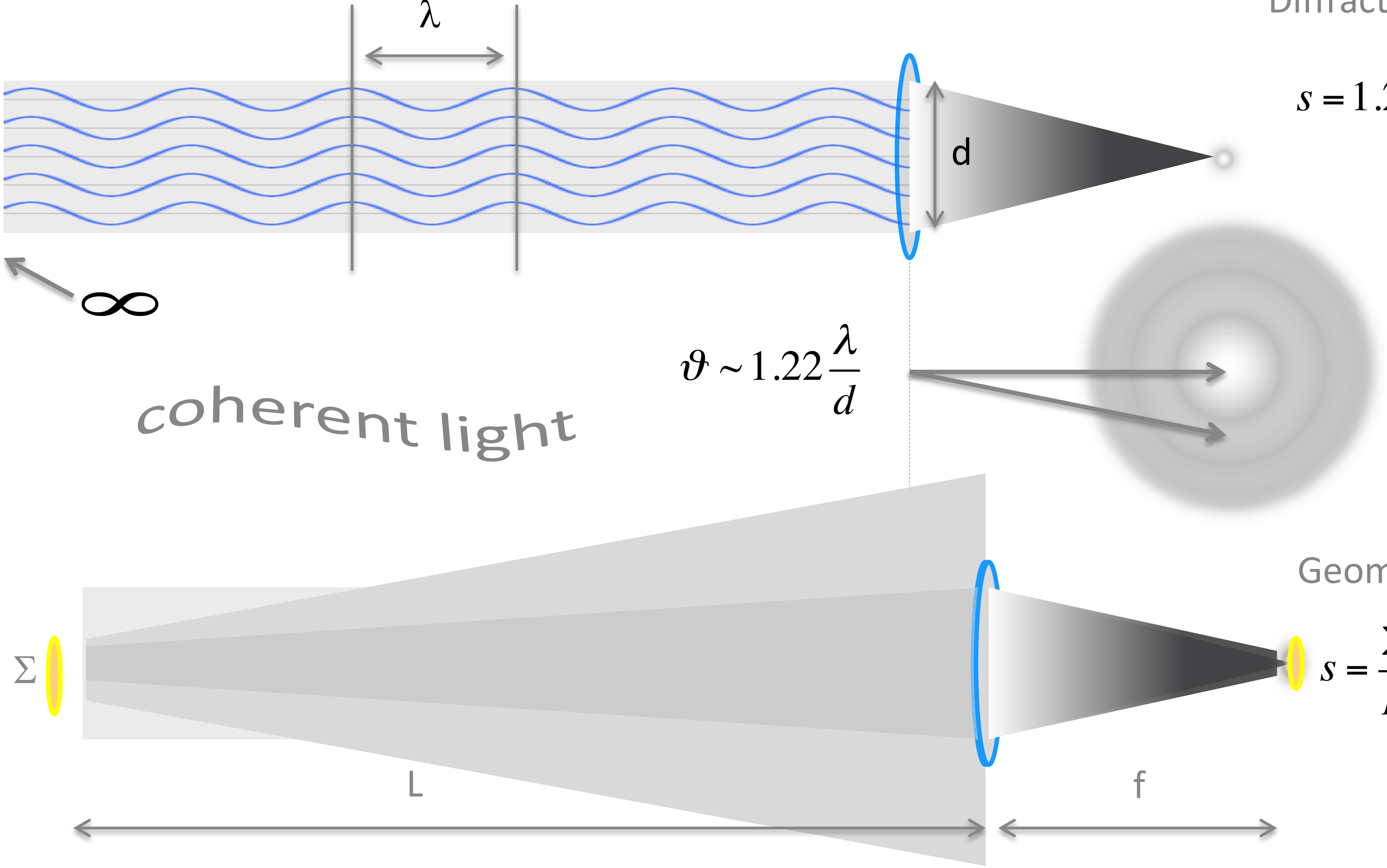


$$\xi_L = N\lambda = (N-1)\cdot(\lambda + \Delta\lambda) \approx \frac{1}{2}\frac{\lambda^2}{\Delta\lambda}$$

Coherence: what for?

- **Scanning transmission x-ray microscopy (STXM) & Scanning Diffraction: NanoBEAMS**
  Coherent illumination required for diffraction-limited resolution but images are NOT coherent!
  About 15 instruments world wide
- **X-ray holography**
  Many interesting variants and demonstrations since 1972 but only the ESRF scheme has been used in scientific investigations
- **Coherent x-ray diffraction imaging**
  Five synchrotron labs now including ESRF and growing
- **Phase-contrast imaging**
  Phase contrast always involves some degree of coherence we will discuss how much later
- **X-ray photon correlation spectroscopy**
  Two dedicated beam lines now - expected to double or triple in the next few years
- **New and specialized**
  Ptychography, magnetism…

Nanobeams: how small



Diffraction limited

$$s = 1.22 \cdot \frac{\lambda}{d} f$$

$$\vartheta \sim 1.22 \frac{\lambda}{d}$$

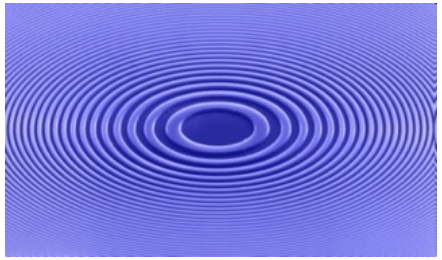
coherent light

Geometry limited

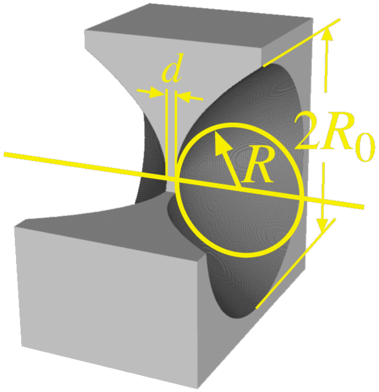
$$s = \frac{\Sigma}{L} \cdot f$$

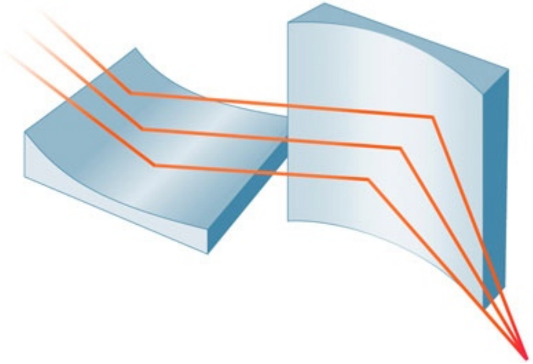
Nanobeams: how

Diffraction
Fresnel zone plates



Refraction
Lenses (convex)

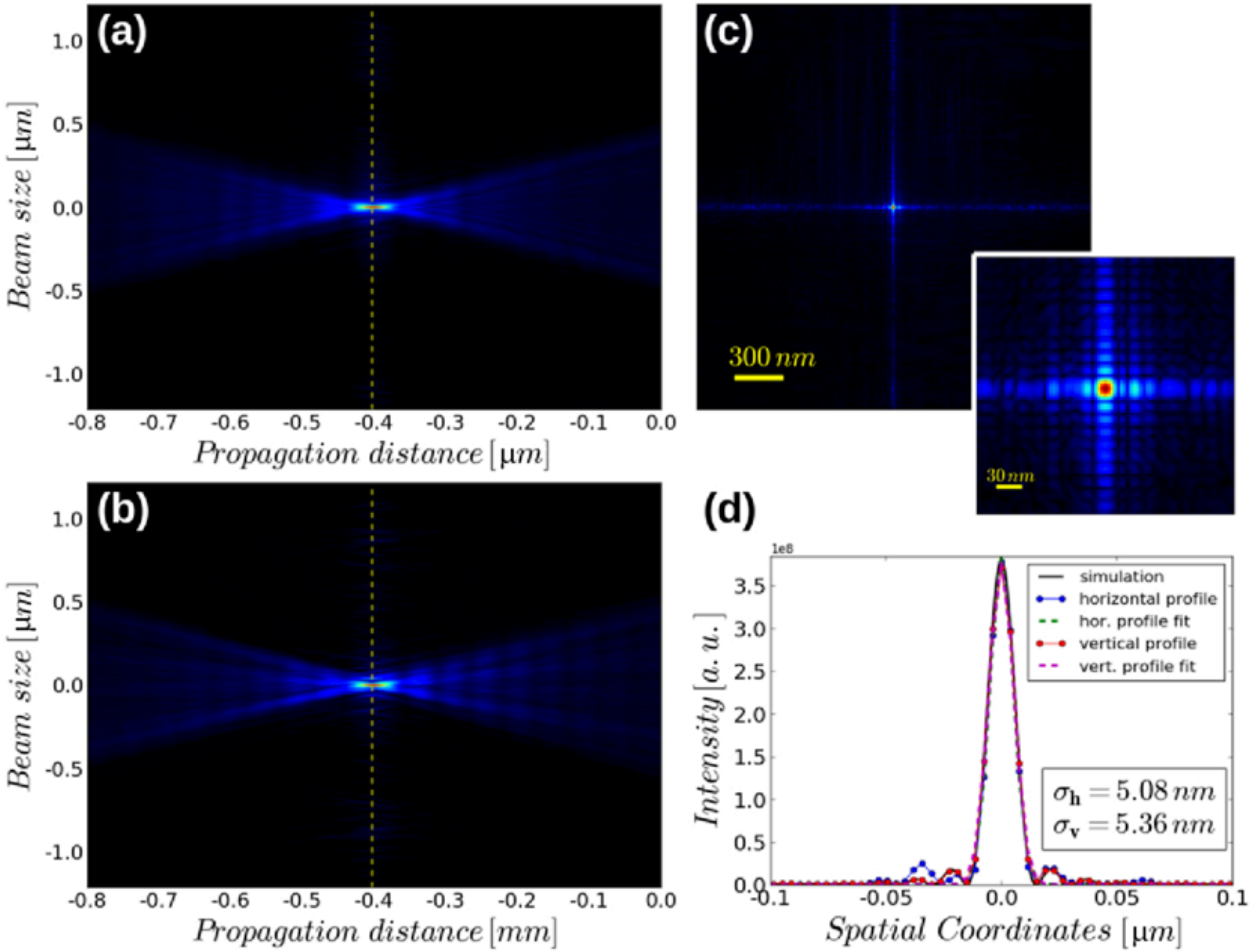




Reflection
Mirrors

Nanofocusing with multilayer-coated KB mirrors

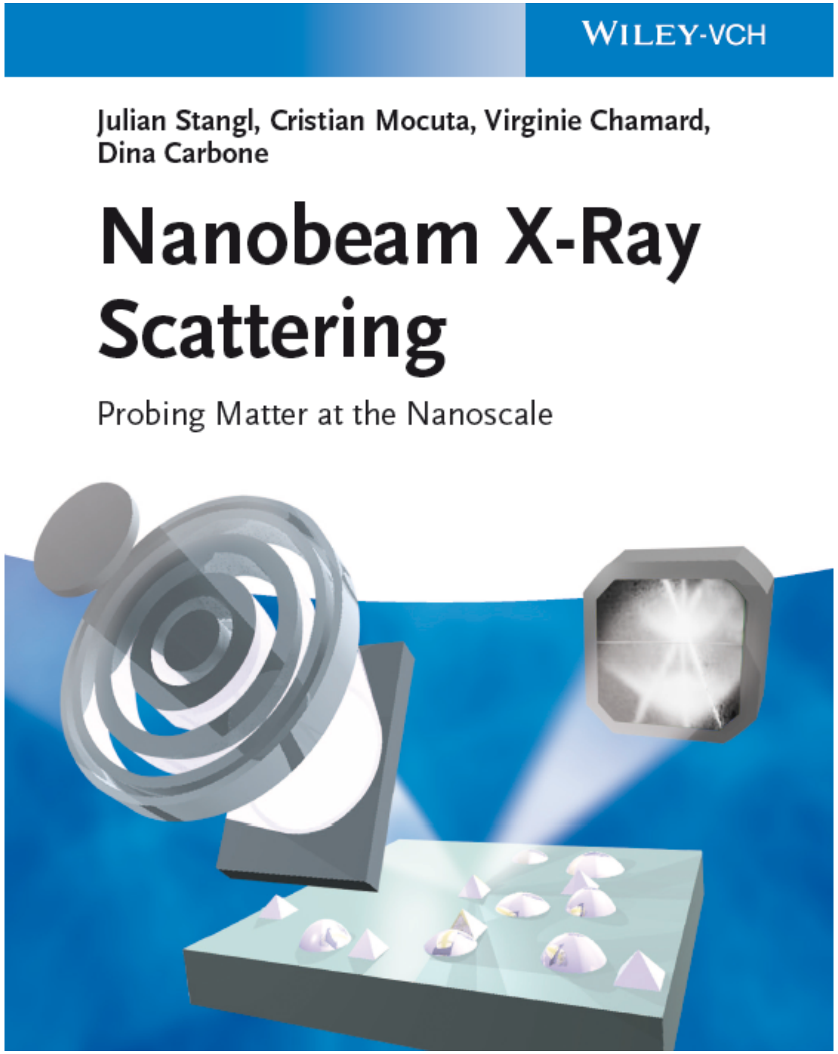
- da Silva, Pacureanu, Yang, Bohic, Morawe, Barrett, and Cloetens, *Optica* **4**, 492 (2017)
- FWHM of 12.0 nm (H) and 12.6 nm (V) at 17 keV



Courtesy of Chris Jacobsen, Argonne National Lab & Northwestern University

Nanobeams: X-ray signals @ nm scale!

Reducing the lateral size of the beam (without loosing photons!) allows to measure ***single*** objects or ***maps***

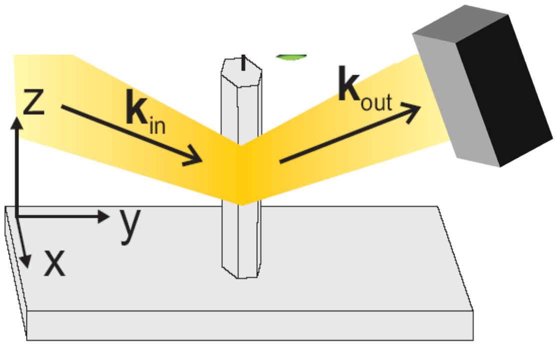

WILEY-VCH

Julian Stangl, Cristian Mocuta, Virginie Chamard, Dina Carbone

Nanobeam X-Ray Scattering

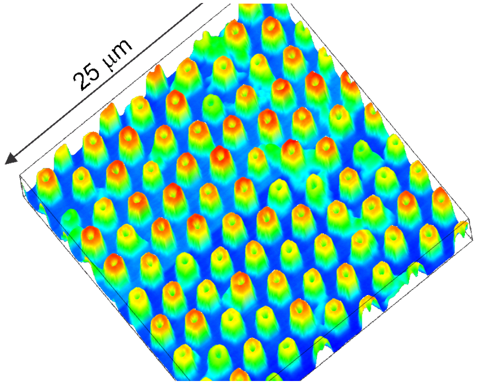
Probing Matter at the Nanoscale

Single object [local probe]



A. Diaz et al *Phys. Rev. B* **79** (2009) 125324

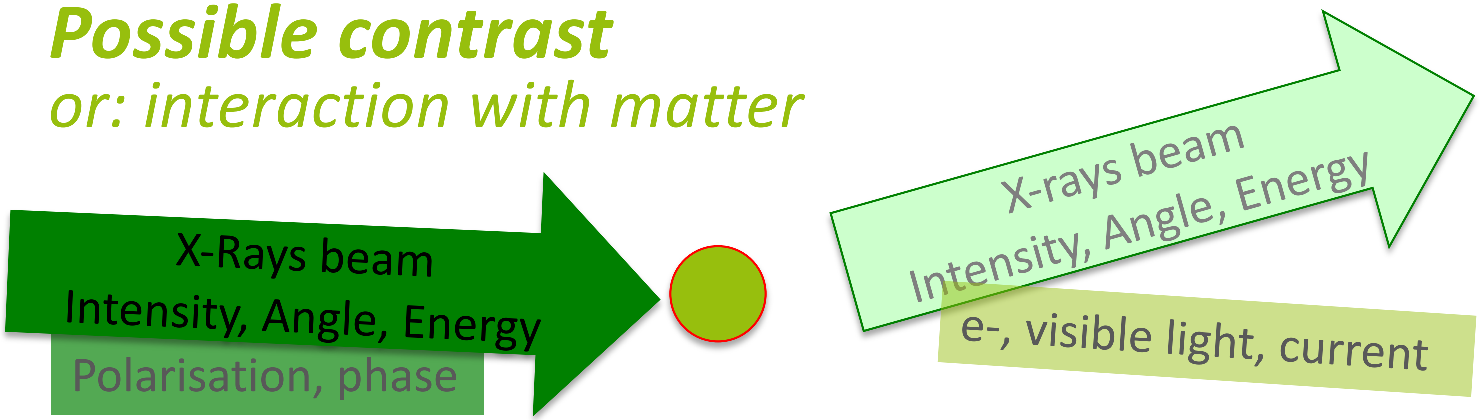
Map [scanning probe]



G. Martinez-Criado *et al Nano Lett.* (2012) **12**, 5829

Possible contrast

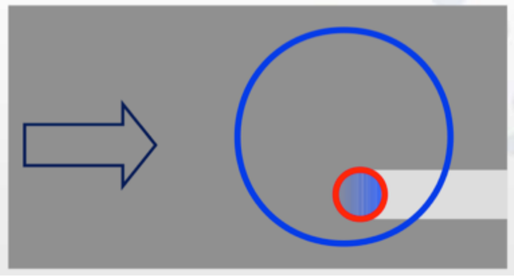
or: interaction with matter

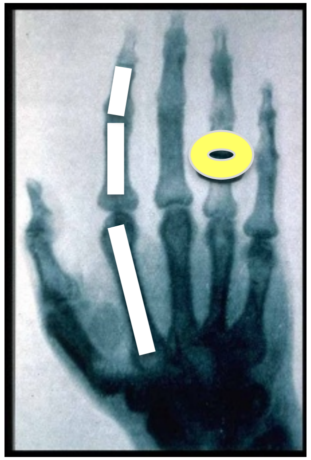

X-Rays beam
Intensity, Angle, Energy
Polarisation, phase

X-rays beam
Intensity, Angle, Energy

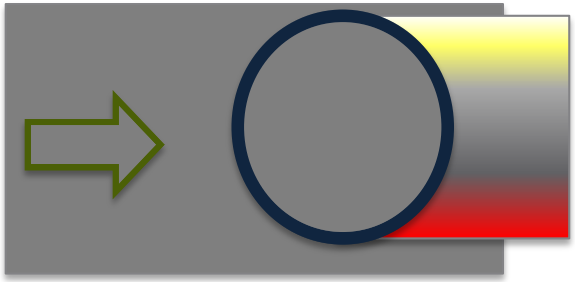
e-, visible light, current

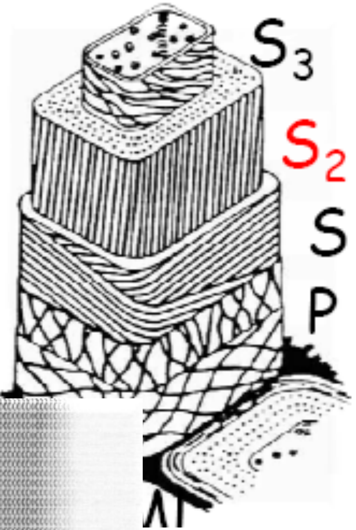
- Absorption I_{out}/I_{in}



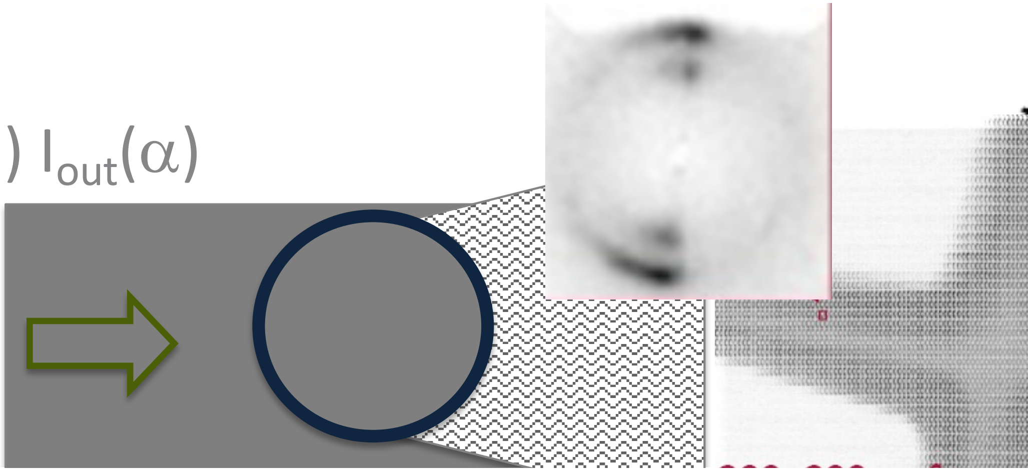


- Spectroscopy $I_{out}(Energy)$

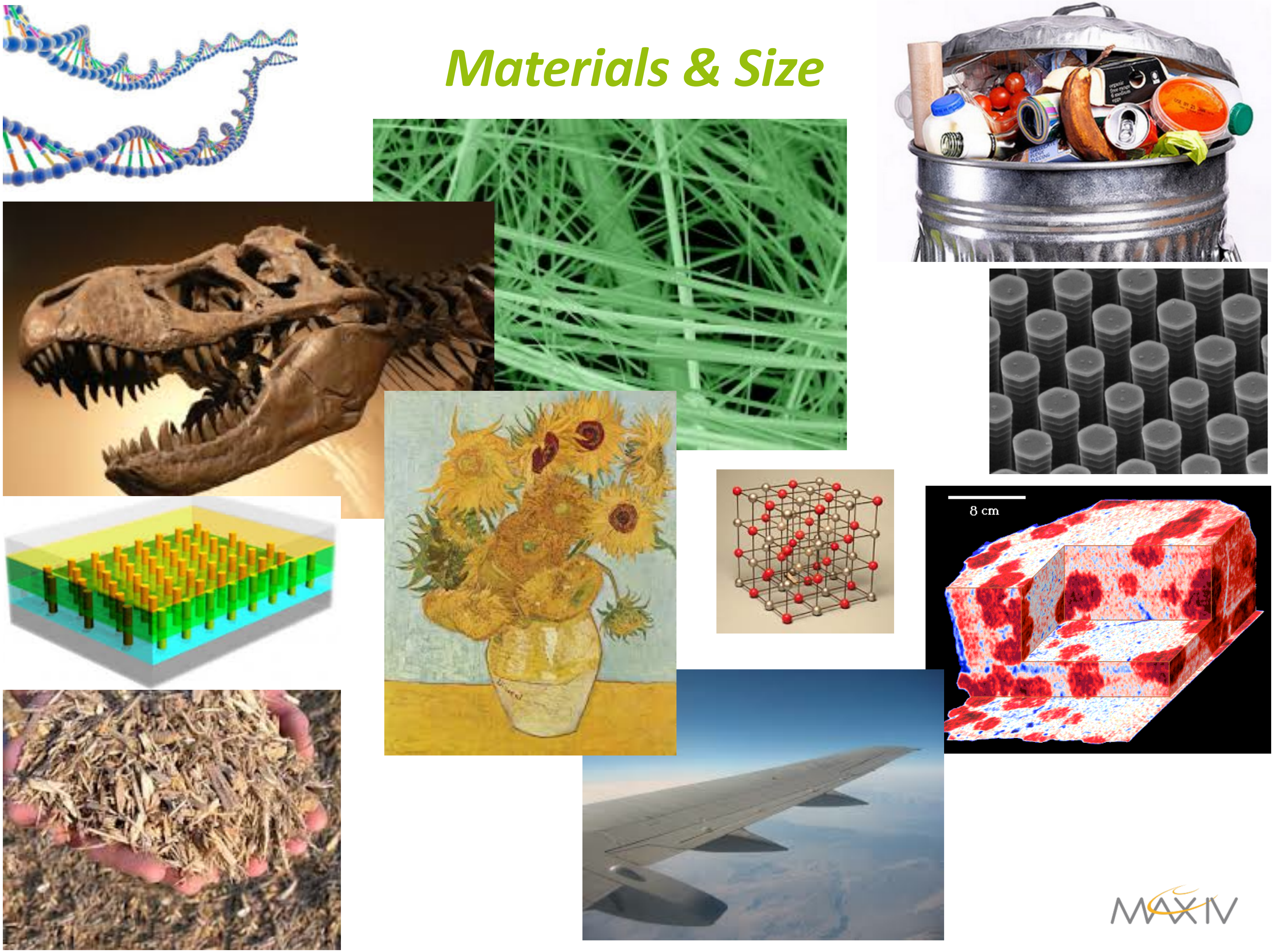




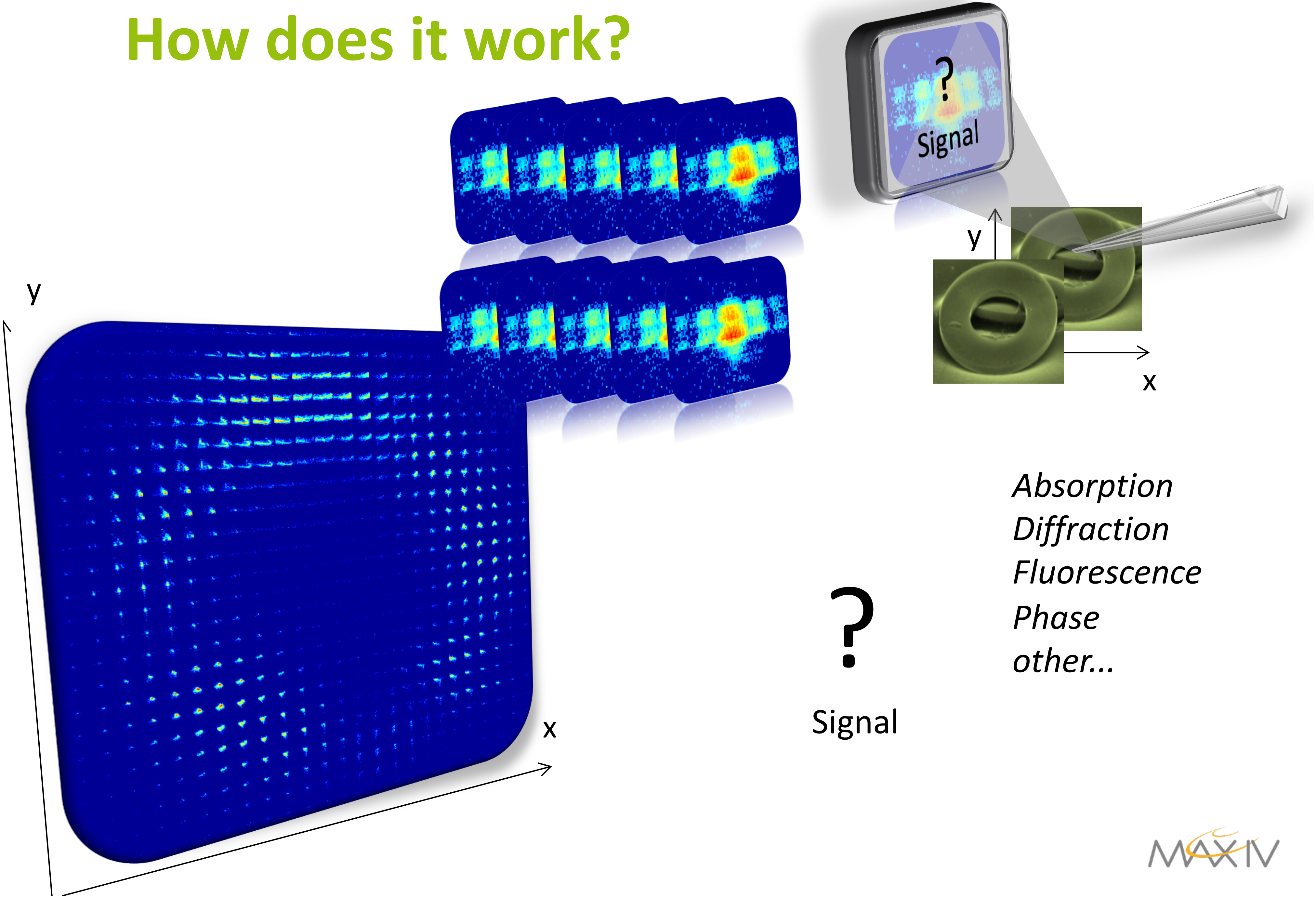
- Scattering (structure) $I_{out}(\alpha)$



Materials & Size



How does it work?



?

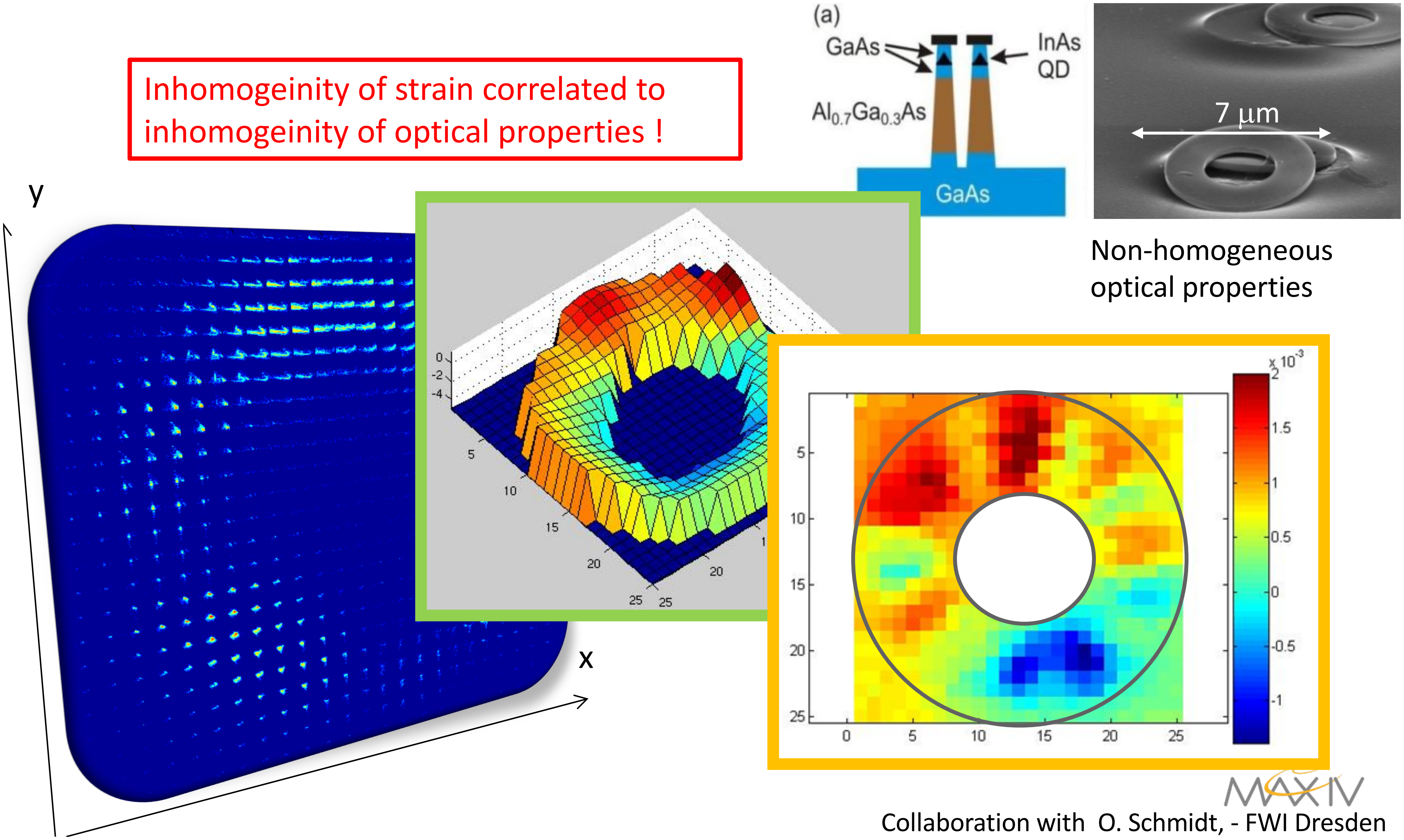
Signal

Absorption
Diffraction
Fluorescence
Phase
other...

2D strain maps: Diffraction

Crystal lattice and strain maps [Res.: beam 500 nm – ID01 ESRF]

Inhomogeinity of strain correlated to inhomogeinity of optical properties !

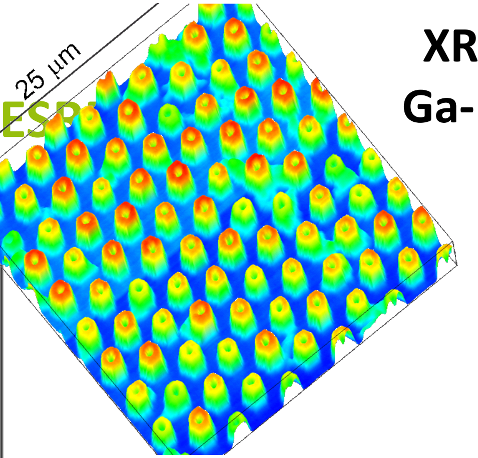


Non-homogeneous optical properties

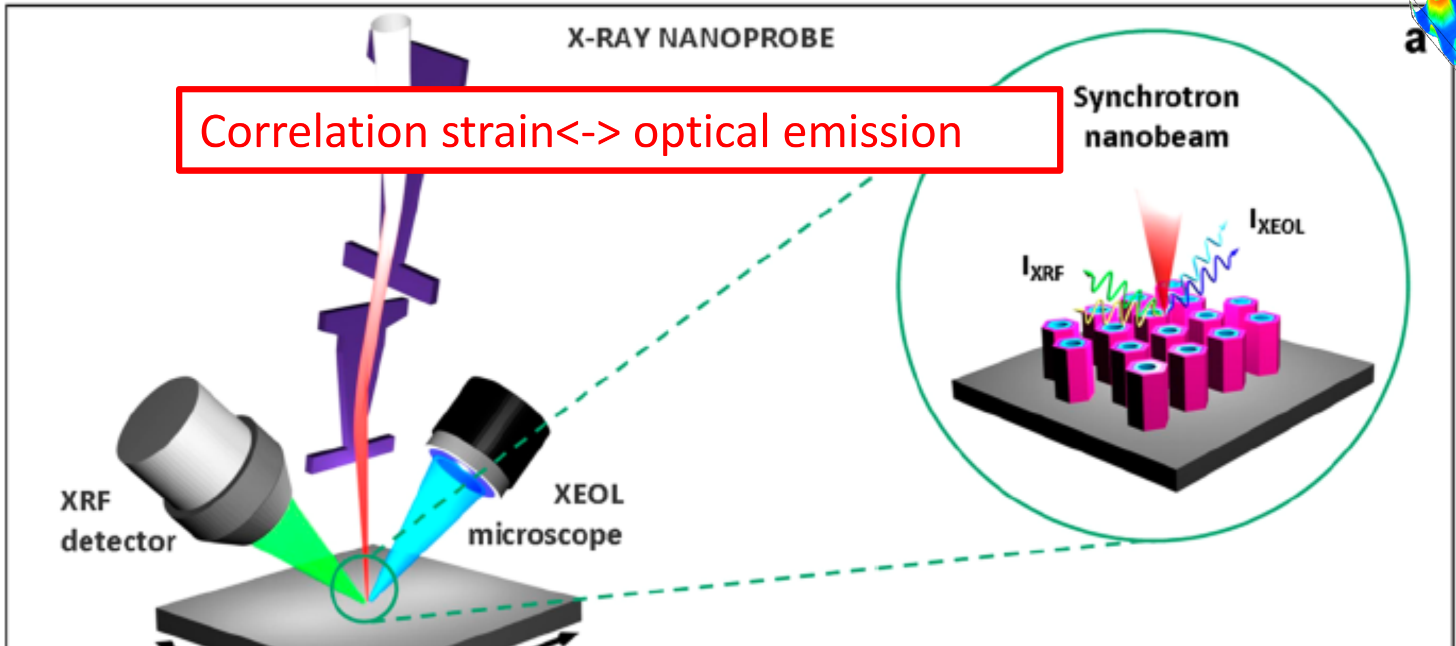
Collaboration with O. Schmidt, - FWI Dresden

2D Fluorescence imaging

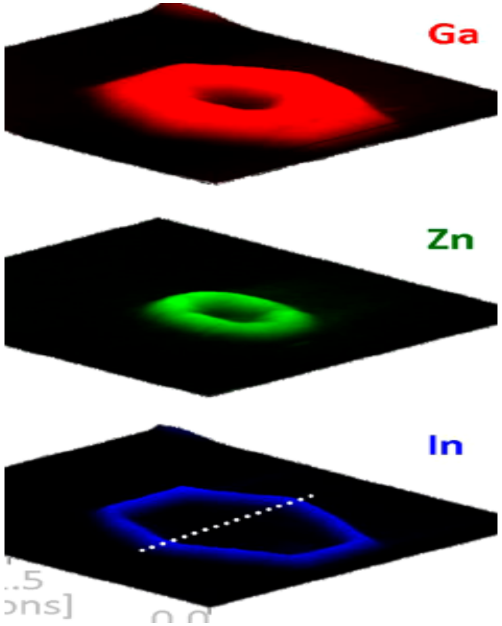
Elemental composition maps [Res: beamsize, 60 nm ID22- ESR

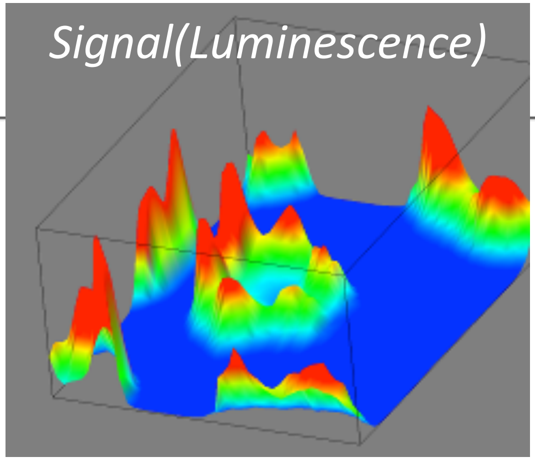


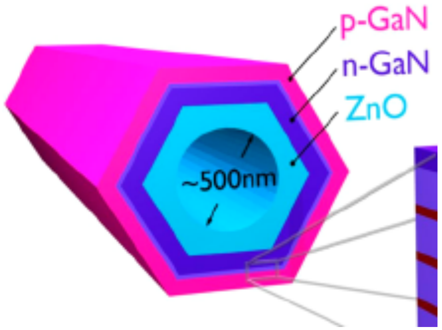
XRF
Ga-K$_{\alpha}$



Correlation strain<-> optical emission




Signal(Luminescence)



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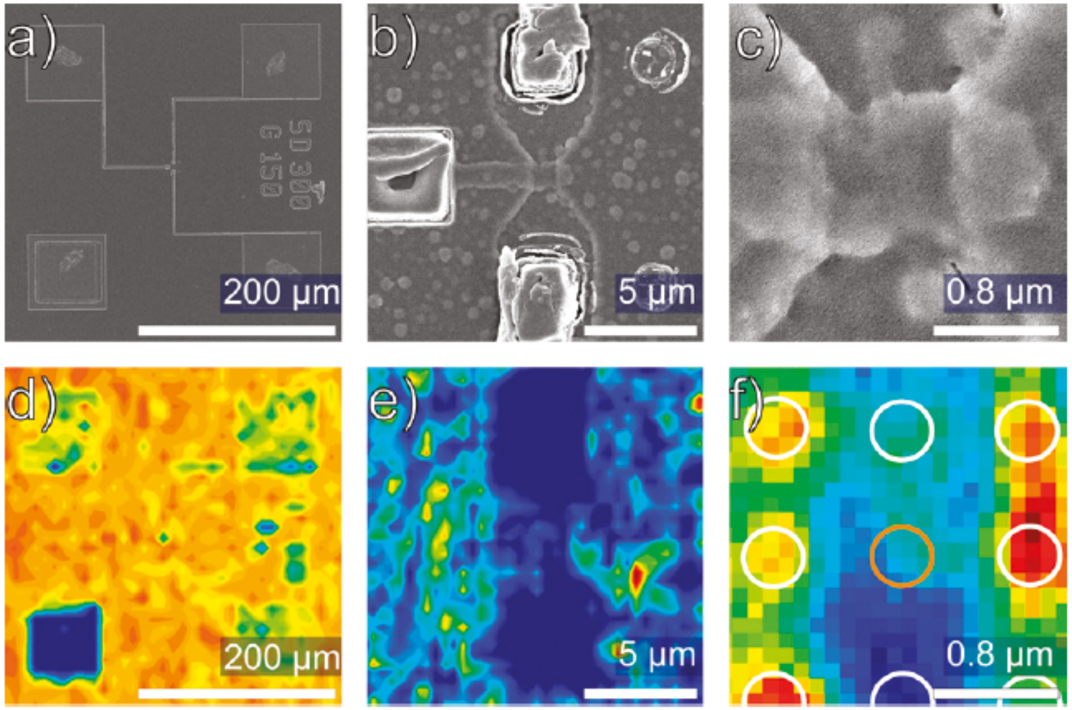
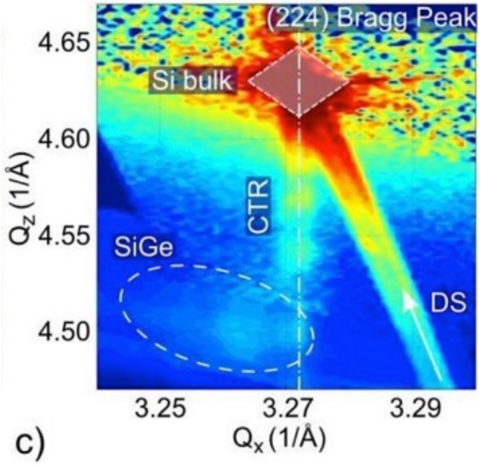
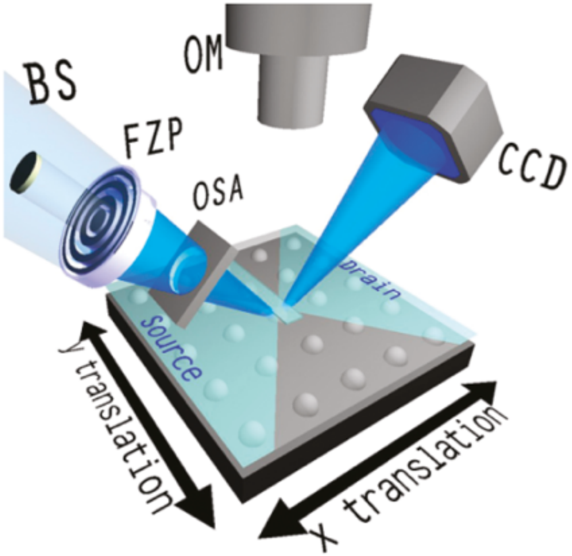
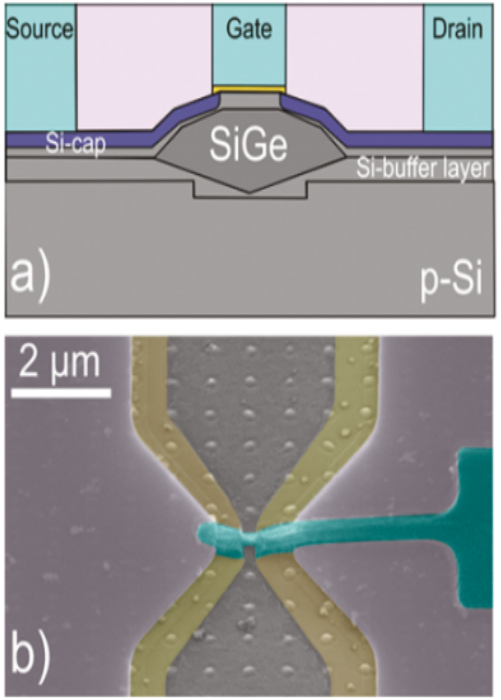
Nanobeams as Local probe

Local atomic structure and strain







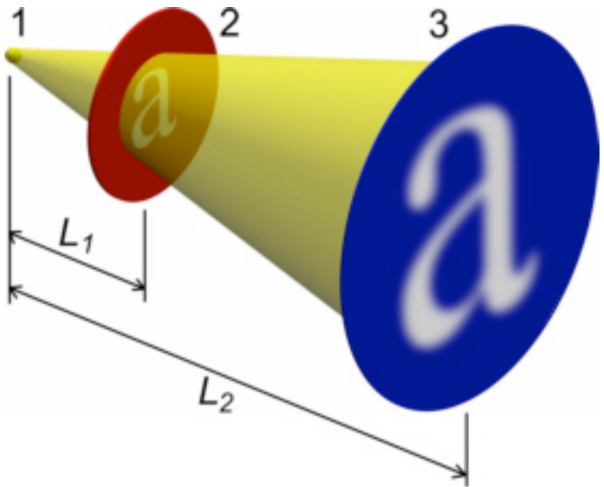


Signal(ϑ)
Data treatment to extract
local *strain & composition*

Structure←→Function
on same structure

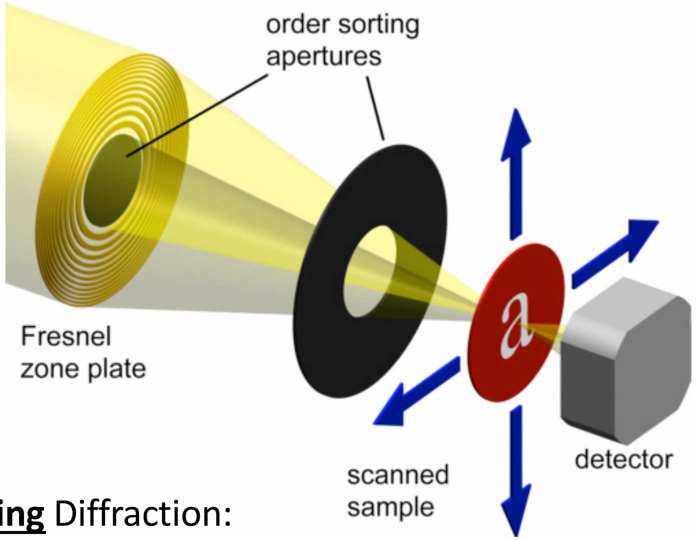
Nano Letters 2011, **11**, 2875

Coherent X-ray imaging techniques

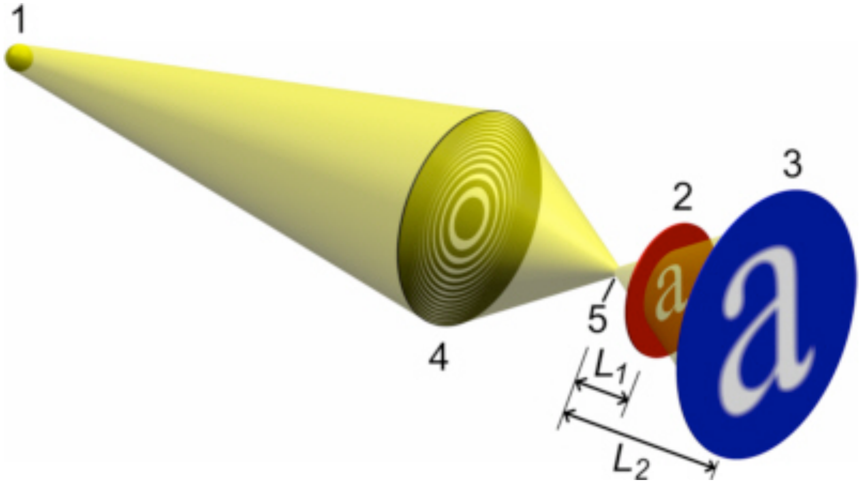


Full-Field Projection: Magnification L_2/L_1

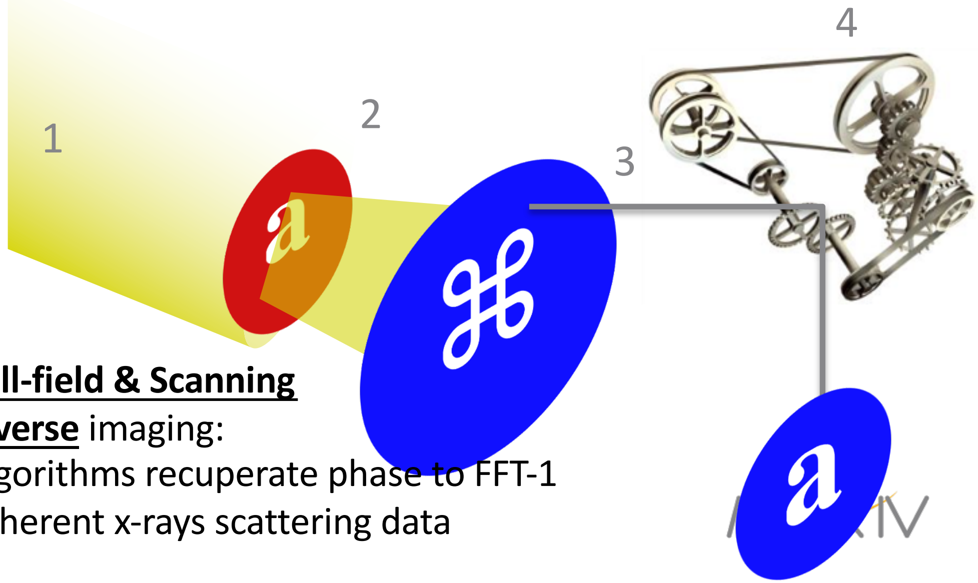
See: www.x-ray-optics.de



Scanning Diffraction:
Lens produces smaller beam on Sample



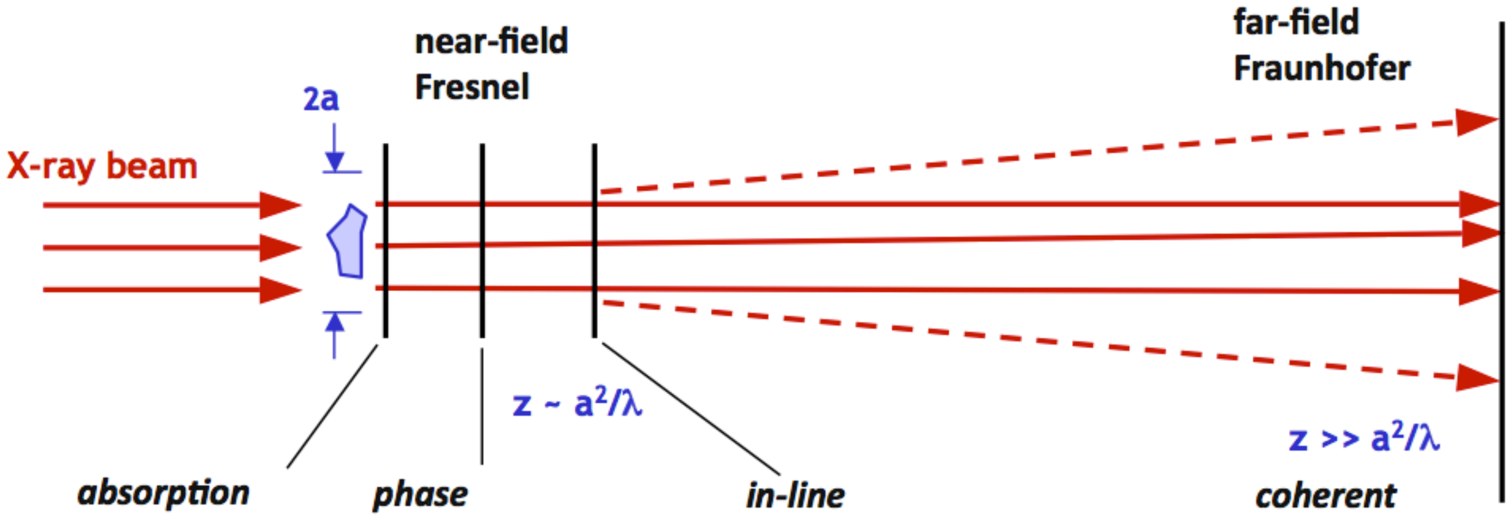
Full-Field Projection: Magnification L_2/L_1
Lens gives demagnified Image of Source
And less blurred image



Full-field & Scanning
Inverse imaging:
Algorithms recuperate phase to FFT-1
coherent x-rays scattering data

From near to Far field

different regimes



absorption radiograph

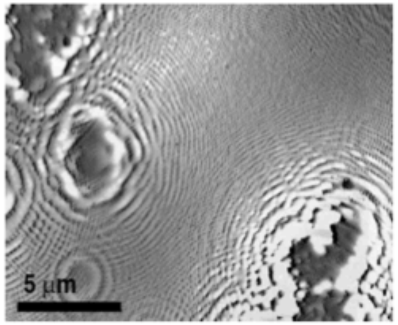
Absorption

phase contrast

Kagoshima (1999)

Refraction

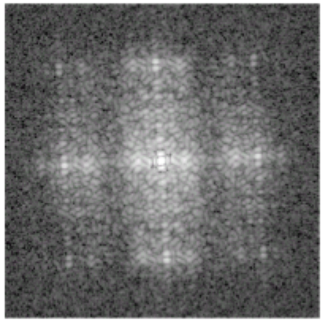
in-line holography



Jacobsen (1990)

Diffraction

coherent diffraction



Miao (1999)

Diffraction

Far Field Coherent diffraction imaging

X-Ray diffraction: distances (size, thickness)
correlation (lattice parameter, structure factor)
density (shape, roughness, intermixing)

X-ray diffraction

Fourier transform of electron distribution

$\mathbf{Q = 2\pi / d}$

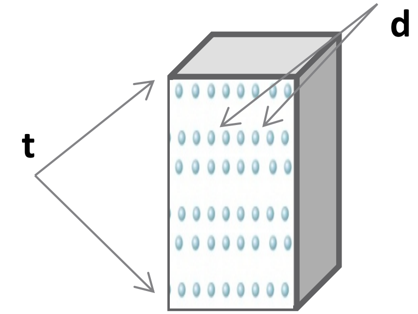
$\mathbf{Q' = 2\pi / t}$

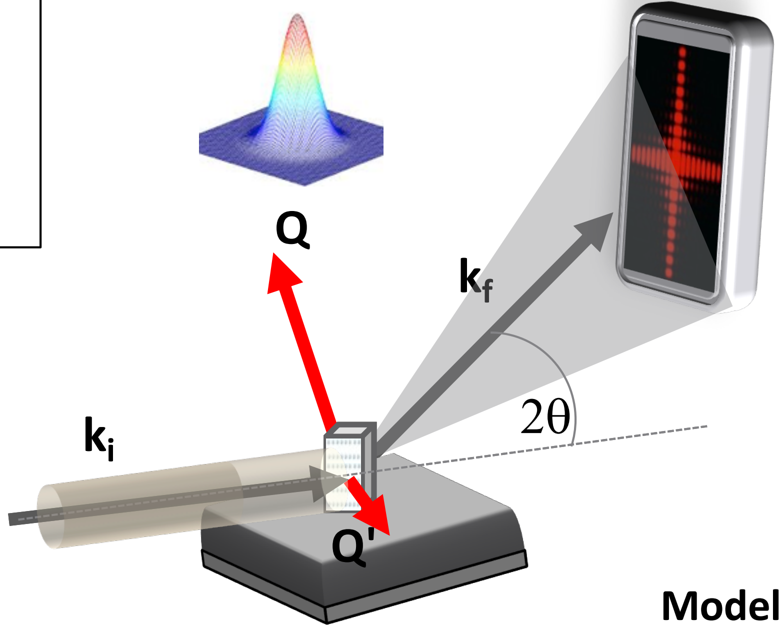
$\mathbf{n\lambda = 2d \sin\theta}$

$\mathbf{Q = (k_f - k_i) \,//\, d, t}$

$$I(\vec{q}) = \left| FT\left\{ \rho(\vec{r}) e^{i\vec{q}\bullet\vec{r}} \right\} \right|^2$$

Partial coherence of x-rays
(chaotic source, polychromaticity)





***Diffracted field**: Information*
e$^-$ density
e$^-$ distribution
=> atomic position: ***strain***
BUT
***Intensity** is measured*

*Can we recuperate **phase**??*

No

Model-dependent analysis

Yes!

Model-free analysis
(!) Coherent x-ray beams

Coherent diffraction imaging

X-ray diffraction

Fourier transform of electron distribution

$$\mathbf{Q = 2\pi / d}$$

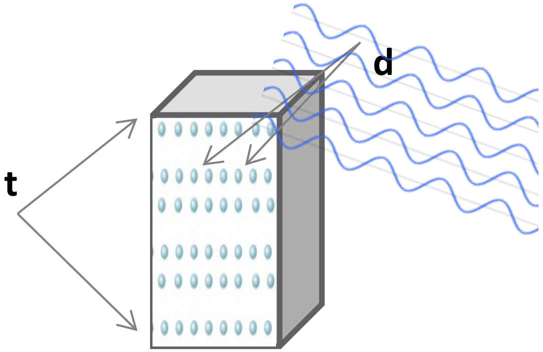
$$\mathbf{Q' = 2\pi / t}$$

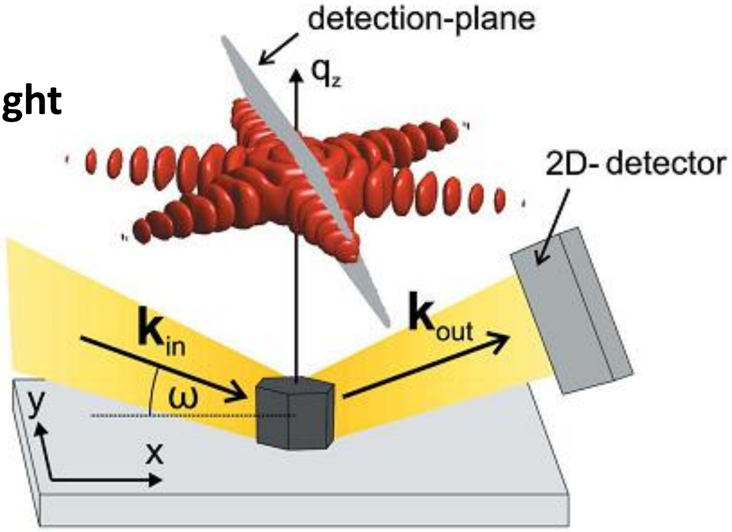
$$\mathbf{n\lambda = 2d \sin\theta}$$

$$\mathbf{Q = (k_f - k_i) // d\ t}$$

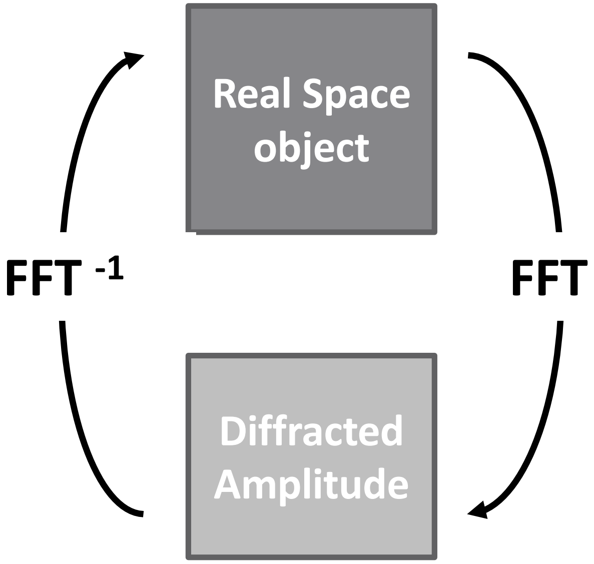
$$I(\vec{q}) = \left| FT\left\{ \rho(\vec{r}) e^{i\vec{q}\bullet\vec{r}} \right\} \right|^2$$

Coherent Light





Phase recovery through mathematical algorithms: ***digital lenses***



Real Space object

FFT

Diffracted Amplitude

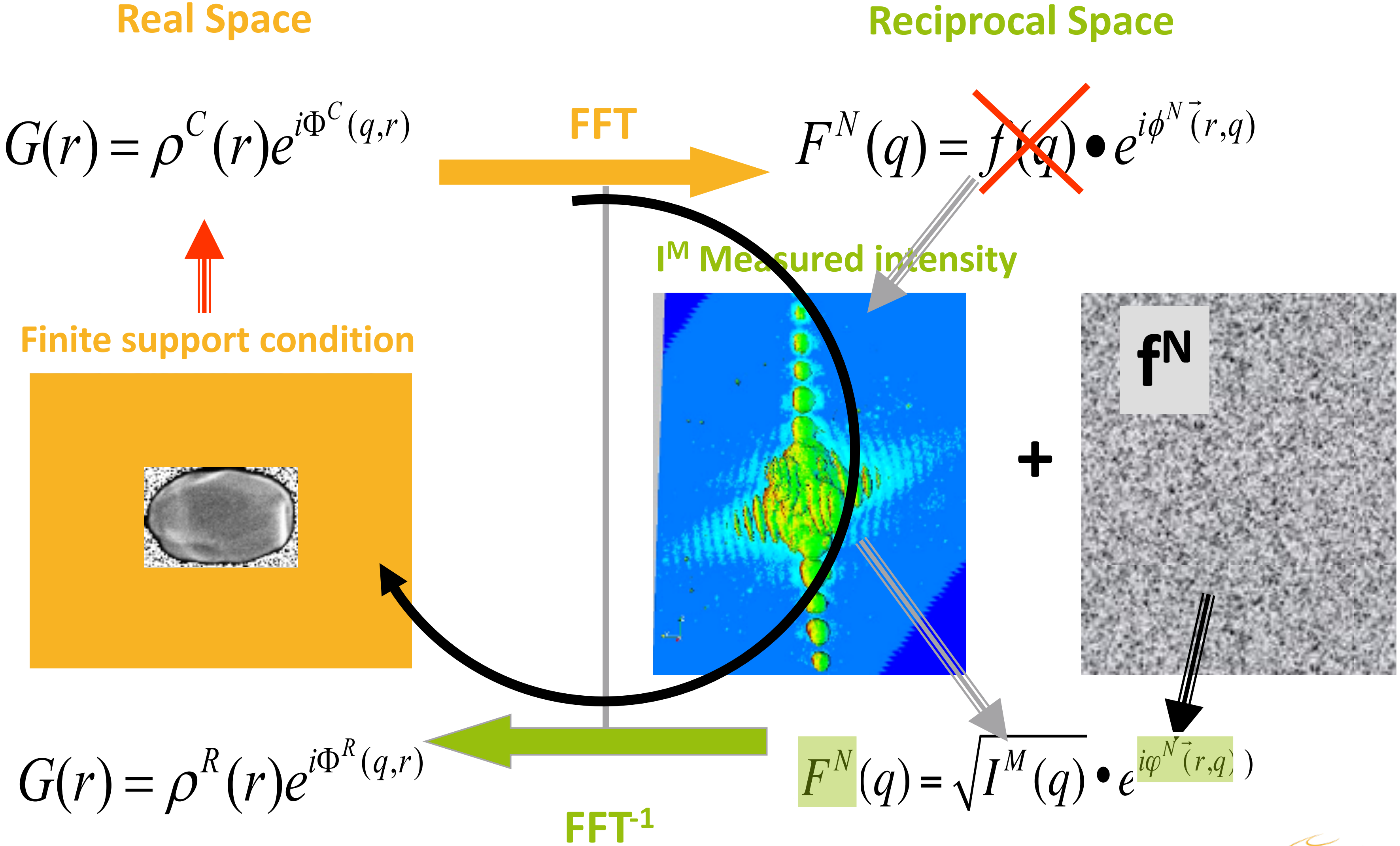
FFT $^{-1}$

Phase Retrieval Algorithm

JR Fienup *Applied Optics 21 2758 (1982)*

Coherent Diffraction Imaging - CDI

$$I(\vec{q}) = \left| FT\left\{\rho(\vec{r})e^{i\Phi(\vec{r})}\right\}\right|^2$$



Real Space

Reciprocal Space

$$G(r) = \rho^C(r)e^{i\Phi^C(q,r)}$$

FFT

$$F^N(q) = \cancel{f(q)} \bullet e^{i\phi^N(\vec{r},q)}$$

I^M Measured intensity

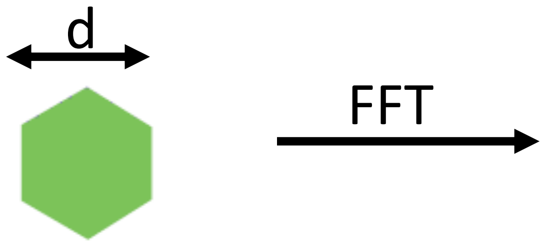
Finite support condition

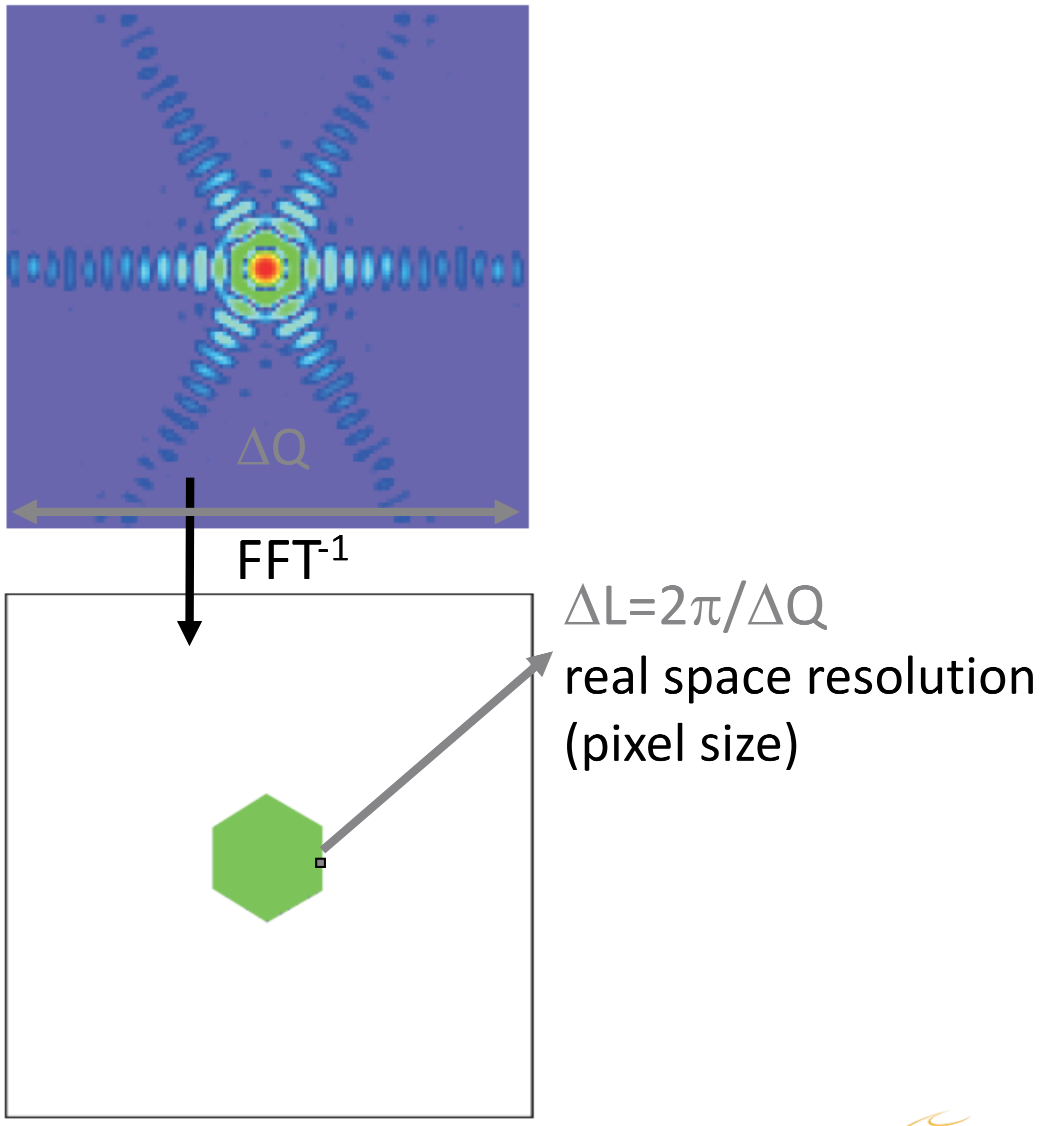
$$G(r) = \rho^R(r)e^{i\Phi^R(q,r)}$$

FFT^{-1}

$$F^N(q) = \sqrt{I^M(q)} \bullet e^{i\varphi^N(\vec{r},q)})$$

A few notes: resolution (!)





2. Measuring *large* ΔQ

increase real space resolution

==>

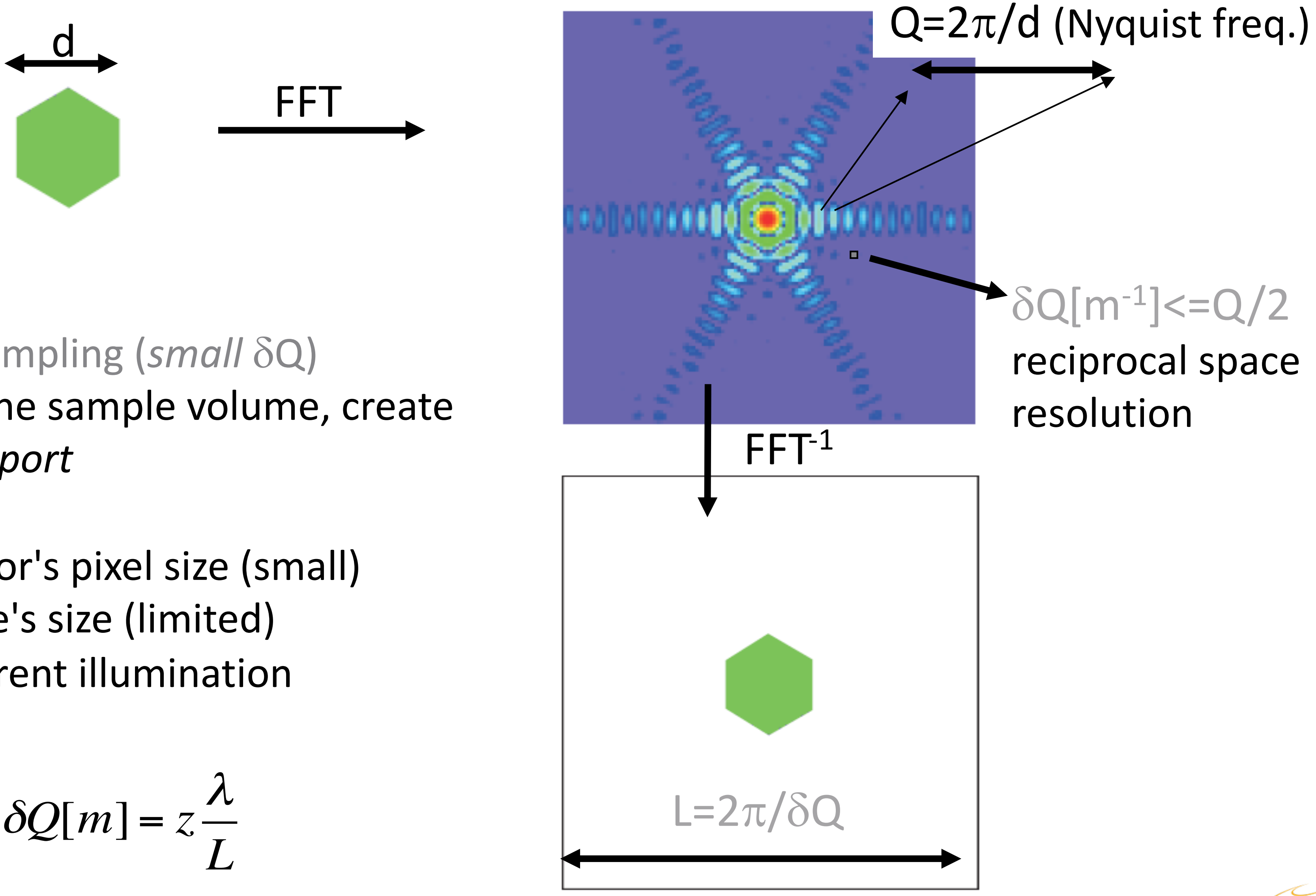
a. Intensity (*focused* beam)

b. Scattering power (large Z)

c. Sample's size (large)

$$\Delta Q[m] = z\frac{\lambda}{\Delta L}$$

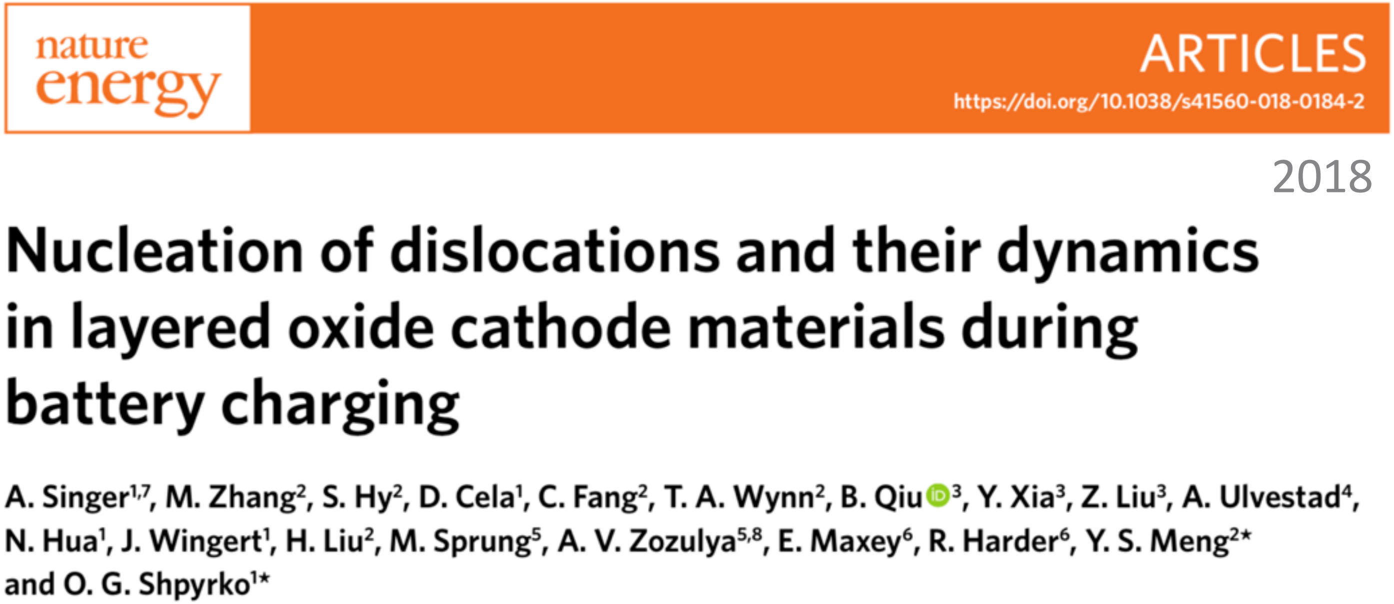
A few notes: over(?)sampling



1. Oversampling (*small* δQ)
confine the sample volume, create *finite support*
==>
a. Detector's pixel size (small)
b. Sample's size (limited)
c. ...coherent illumination

$$\delta Q[m] = z\frac{\lambda}{L}$$

Energy materials: in situ & operando microscopy



nature energy

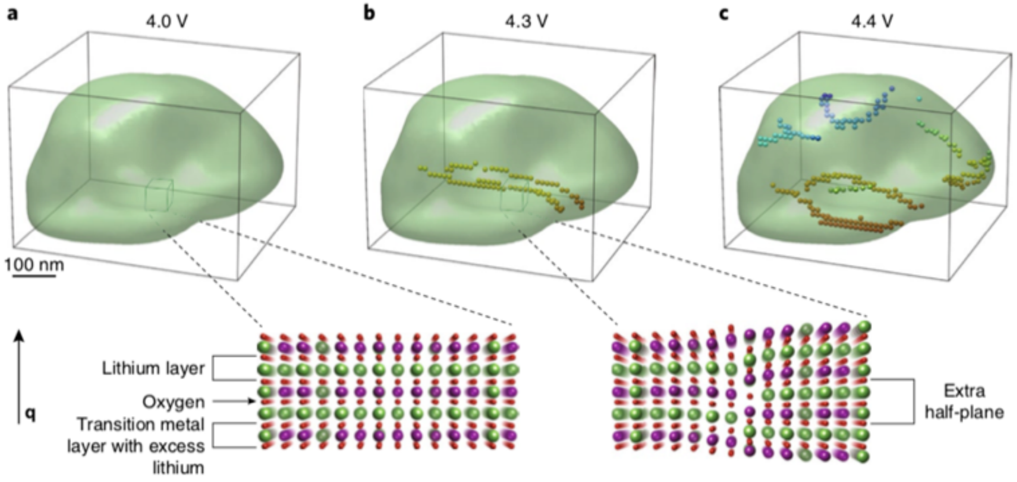
ARTICLES

https://doi.org/10.1038/s41560-018-0184-2

2018

Nucleation of dislocations and their dynamics in layered oxide cathode materials during battery charging

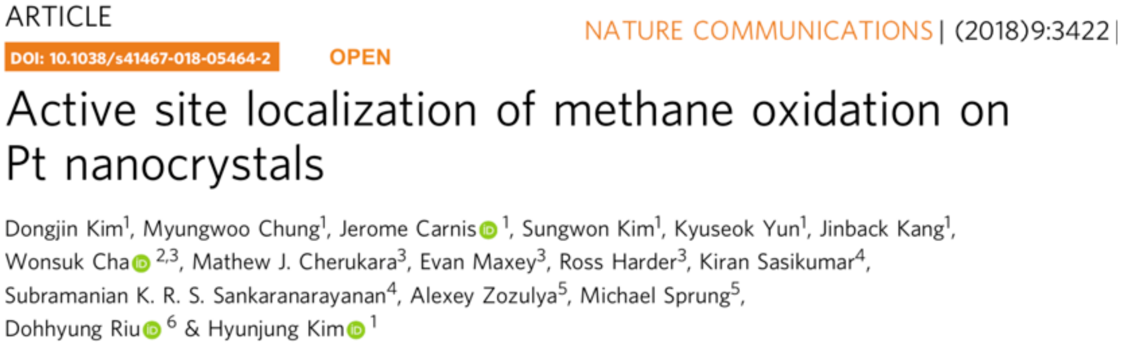
A. Singer[1,7], M. Zhang[2], S. Hy[2], D. Cela[1], C. Fang[2], T. A. Wynn[2], B. Qiu[3], Y. Xia[3], Z. Liu[3], A. Ulvestad[4], N. Hua[1], J. Wingert[1], H. Liu[2], M. Sprung[5], A. V. Zozulya[5,8], E. Maxey[6], R. Harder[6], Y. S. Meng[2]* and O. G. Shpyrko[1]*



In 600nm particles, the formation of stacking faults during operation reduces performances on the long term (fatigue)

? Could the use of smaller particles limit this deterioration?

Coherence: strain imaging in-situ



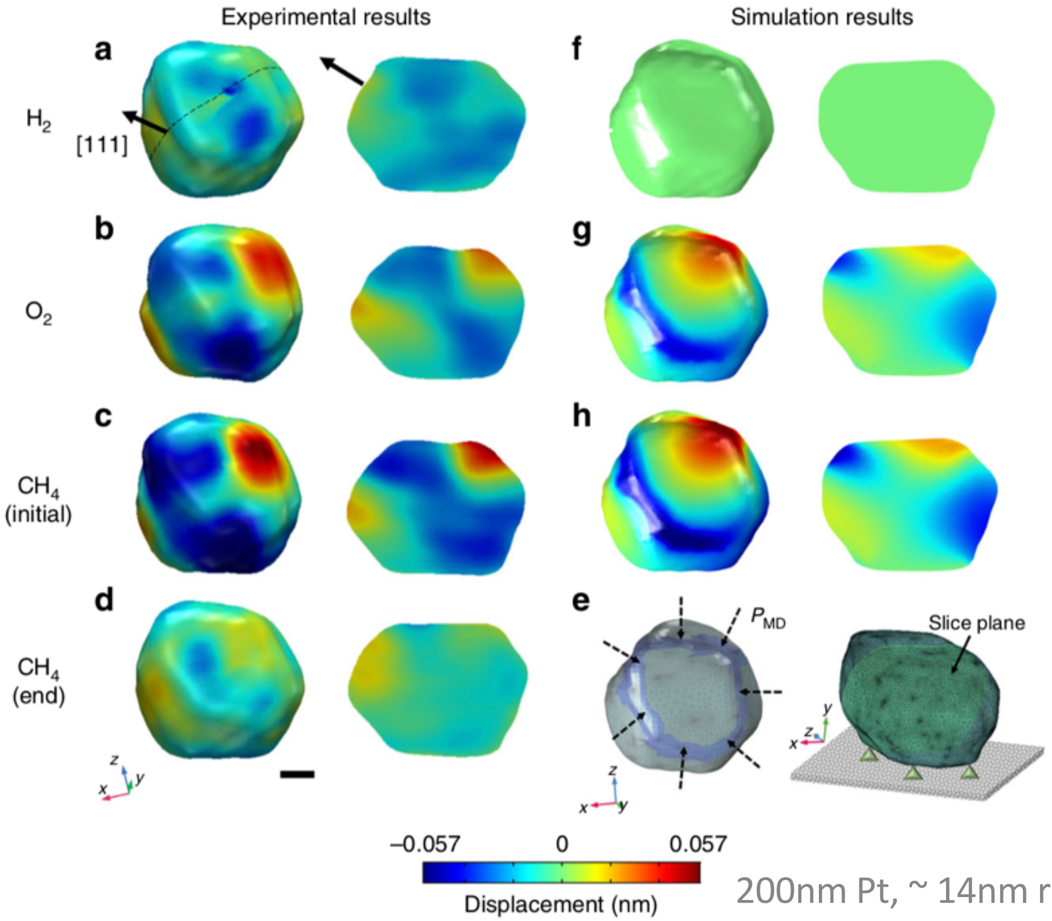
ARTICLE

NATURE COMMUNICATIONS | (2018)9:3422 |

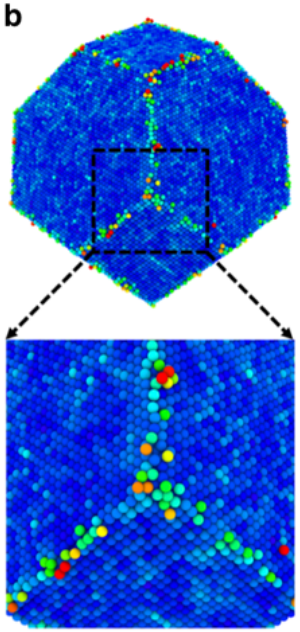
DOI: 10.1038/s41467-018-05464-2 OPEN

Active site localization of methane oxidation on Pt nanocrystals

Dongjin Kim[1], Myungwoo Chung[1], Jerome Carnis[1], Sungwon Kim[1], Kyuseok Yun[1], Jinback Kang[1], Wonsuk Cha[2,3], Mathew J. Cherukara[3], Evan Maxey[3], Ross Harder[3], Kiran Sasikumar[4], Subramanian K. R. S. Sankaranarayanan[4], Alexey Zozulya[5], Michael Sprung[5], Dohhyung Riu[6] & Hyunjung Kim[1]



200nm Pt, ~ 14nm resolution



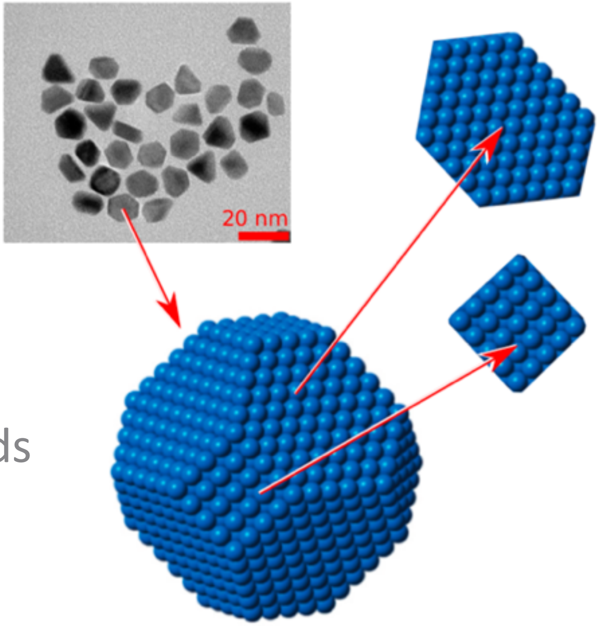
CHALLENGE:

Can we hope to get this resolution?

What about "real life" particle size??

3-30 nm??

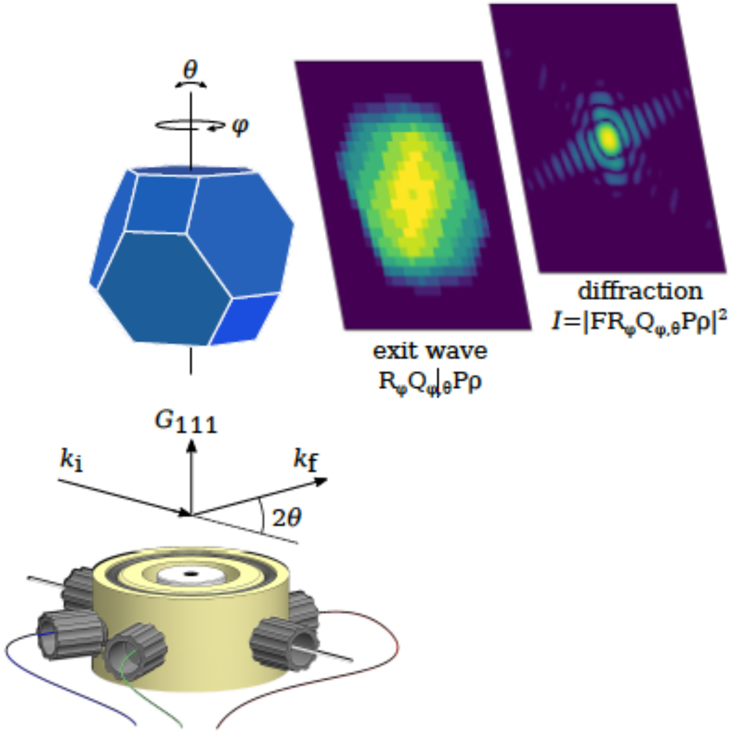
Take the challenge: Pt, Au, Pd 20--60nm @ MAX IV

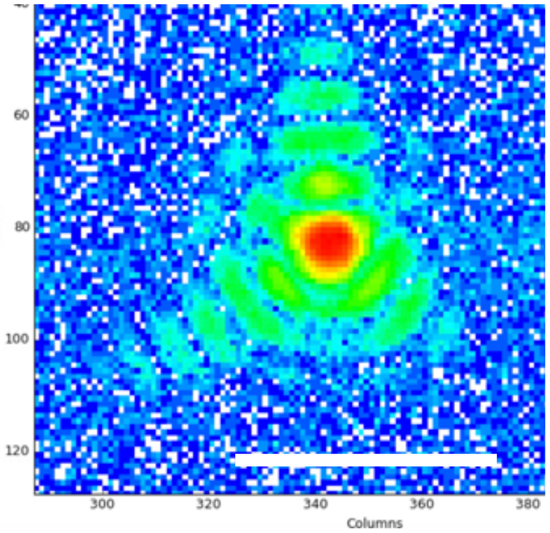


Coherent Bragg imaging of 60 nm Au nanoparticles under electrochemical control at the NanoMAX beamline

A.Bjorling, D. Carbone *et al In Review for JSR*

Particles are unstable in the beam, but good signal is measured even for short counting time (0.1s). Highest q-range corresponds to ~ 8nm



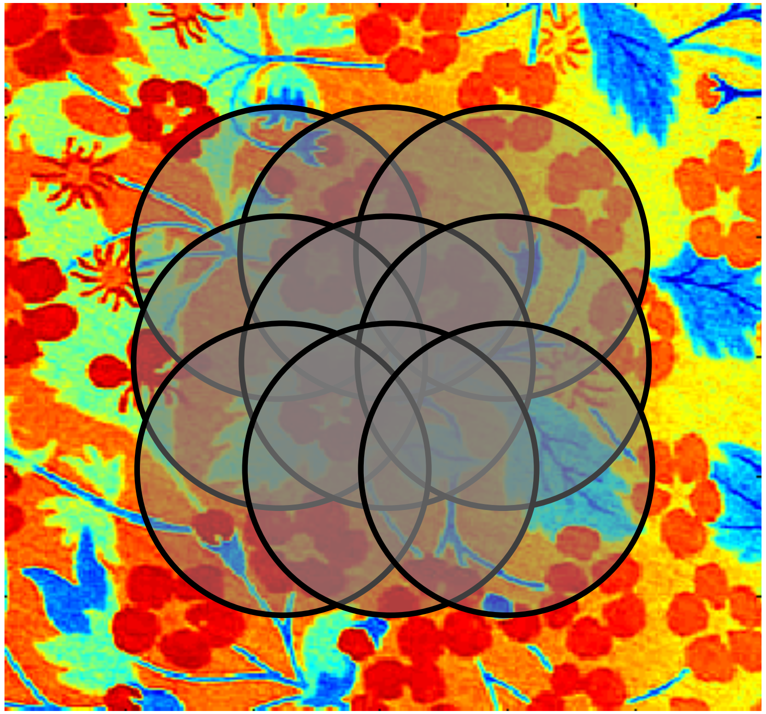


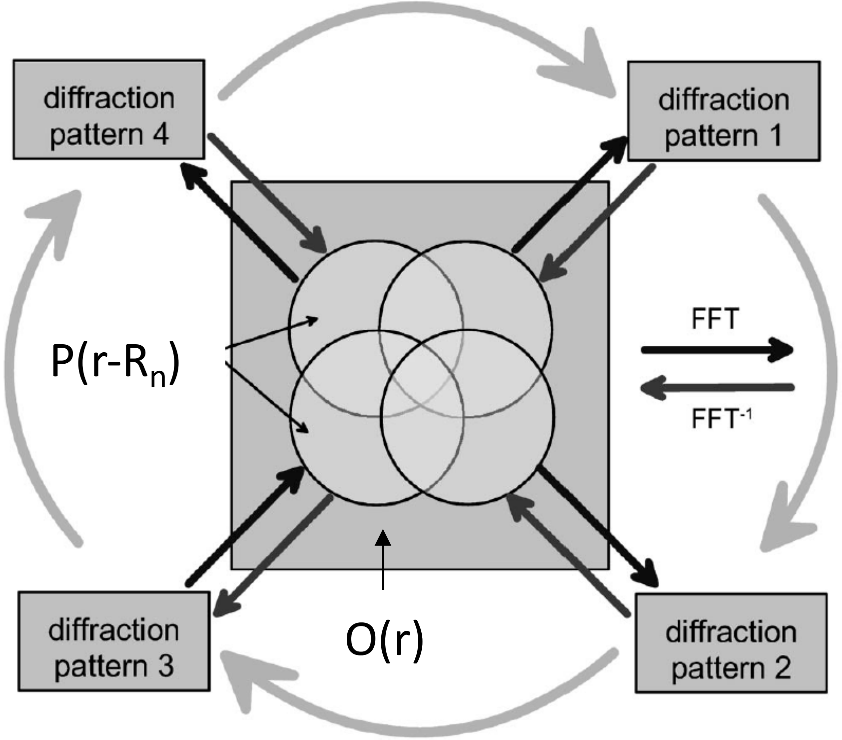
Ptychography – CDI with scanning & overlap

Measurement of several diffraction patterns obtained for different but overlapping illumination areas

$\psi(\mathbf{r}, \mathbf{R}) = O(\mathbf{r} - \mathbf{R}) \times P(\mathbf{r})$

$O(\mathbf{r})$ the *object function,*
$P(\mathbf{r})$ the *illumination function*
$\psi(\mathbf{r})$ the exit wave,
R the displacement of the beam



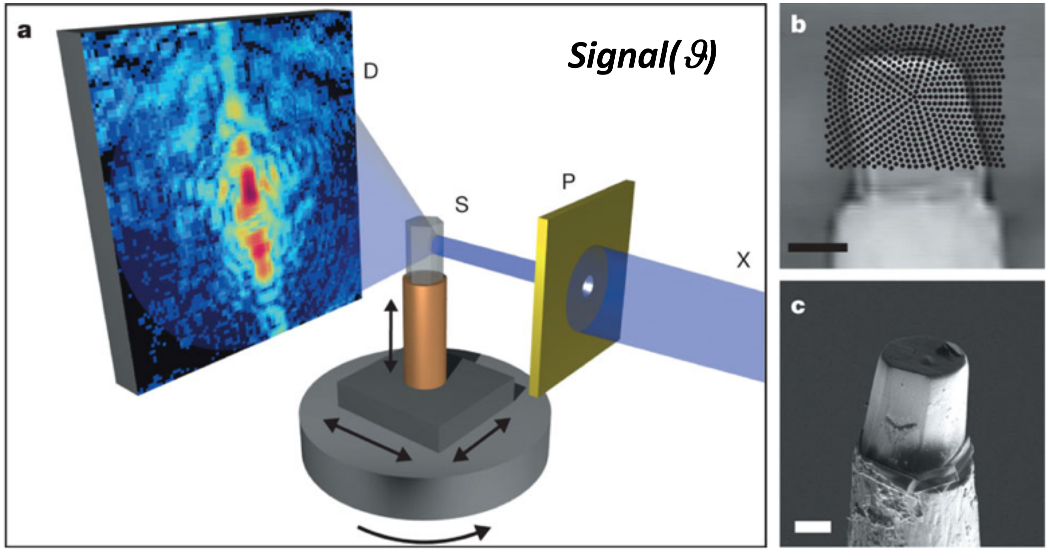


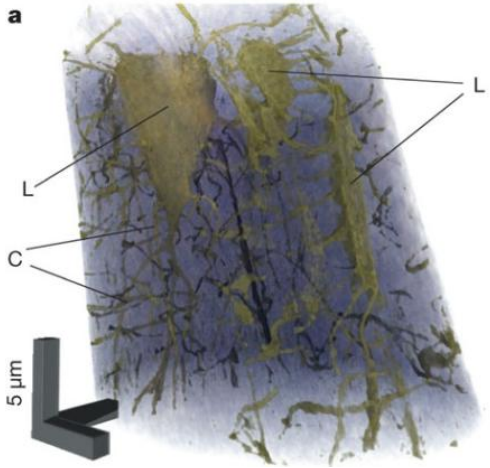
Faulkner et al. PRL ***93*** *(2004); Rodenburg et al. APL* ***85*** *(2004)*

3D with coherent diffraction tomography

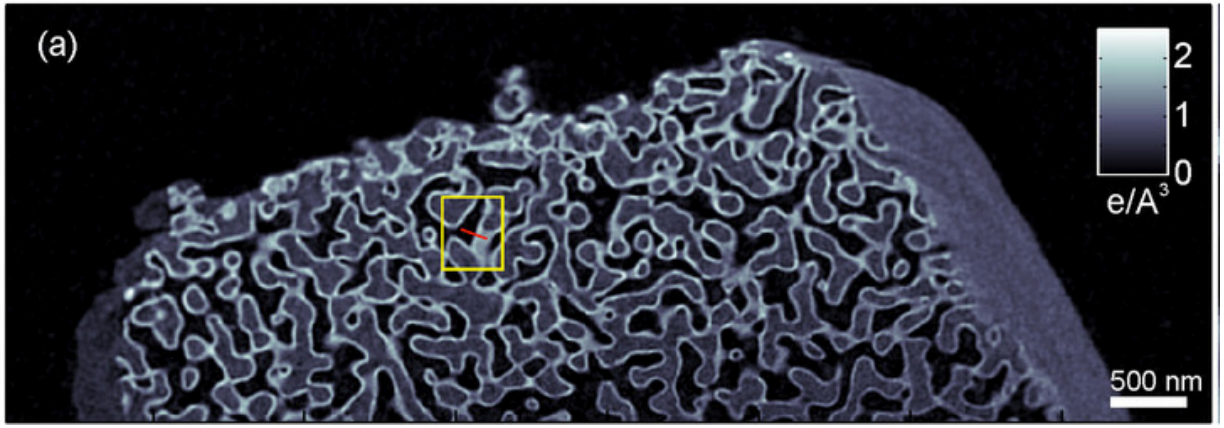
3D quantitative microscopy (morphology biologic and artificial materials)

Bone morphology
M Dierolf *et al. Nature* 2010 **467**, 436-439





2μm beam
& 65nm resolution!!
with phase retrieval
algorithms



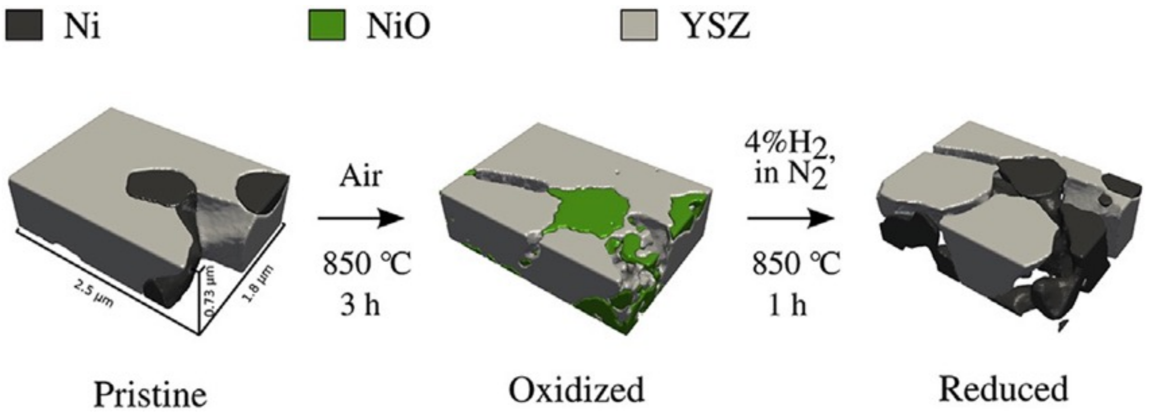
16nm resolution in 3D demonstrated
(strong scattering sample!)

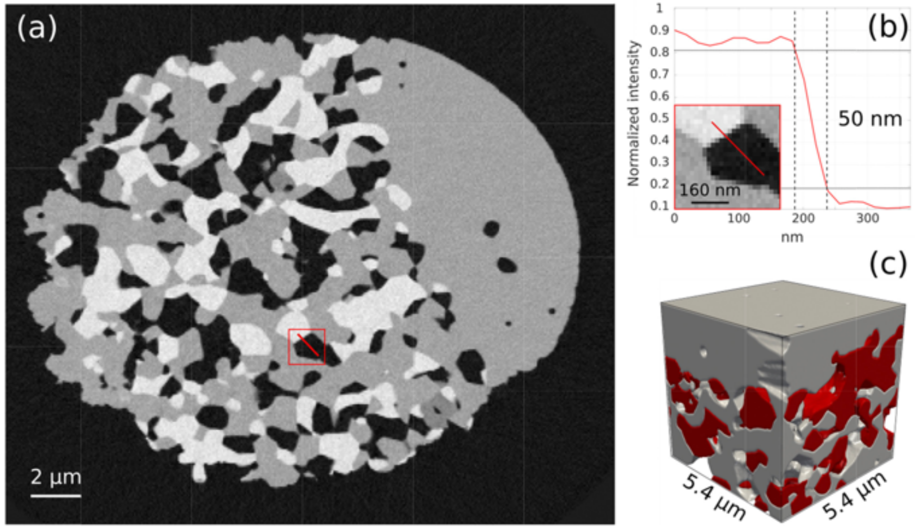
Scientific Reports **4** (2014) Article number:3857

3D with coherent diffraction tomography

3D quantitative microscopy (morphology biologic and artificial materials)

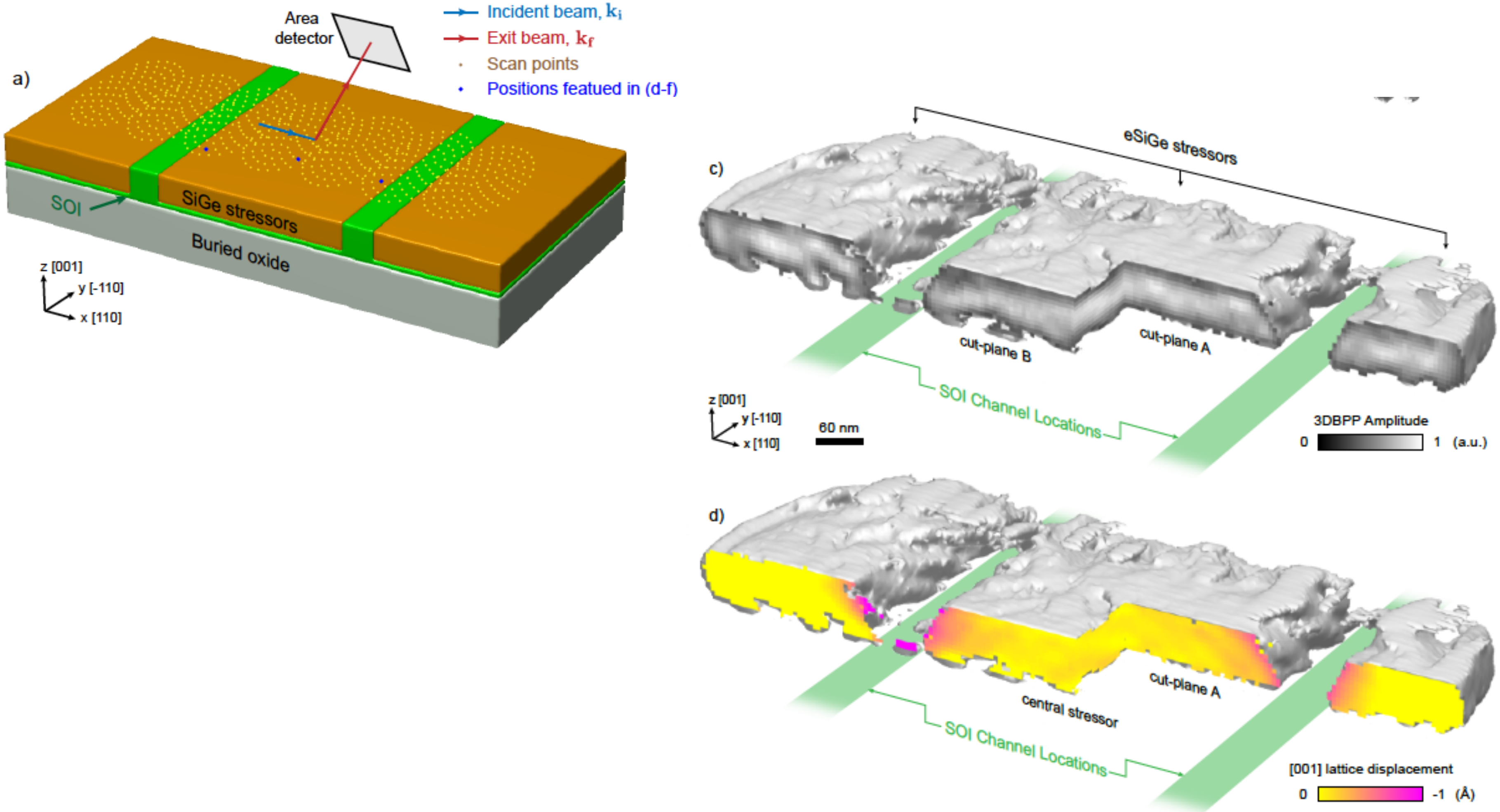
Redox cycle in Solid Oxide Cell
Journal of Power Sources 2017, 520-527





3D with coherent diffraction

3D quantitative microscopy (crystal structure artificial material)



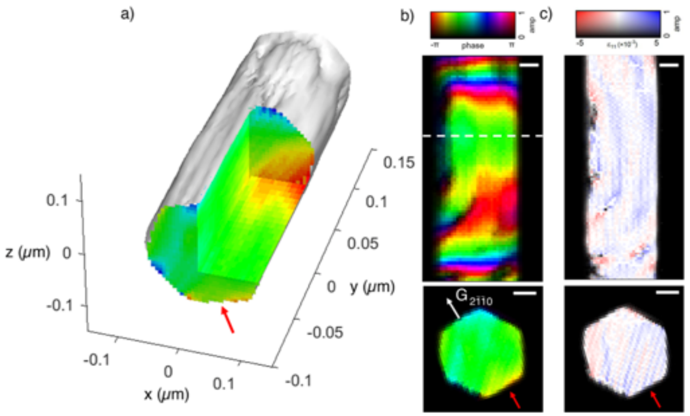
arXiv:1506.01262 [cond-mat.mtrl-sci]

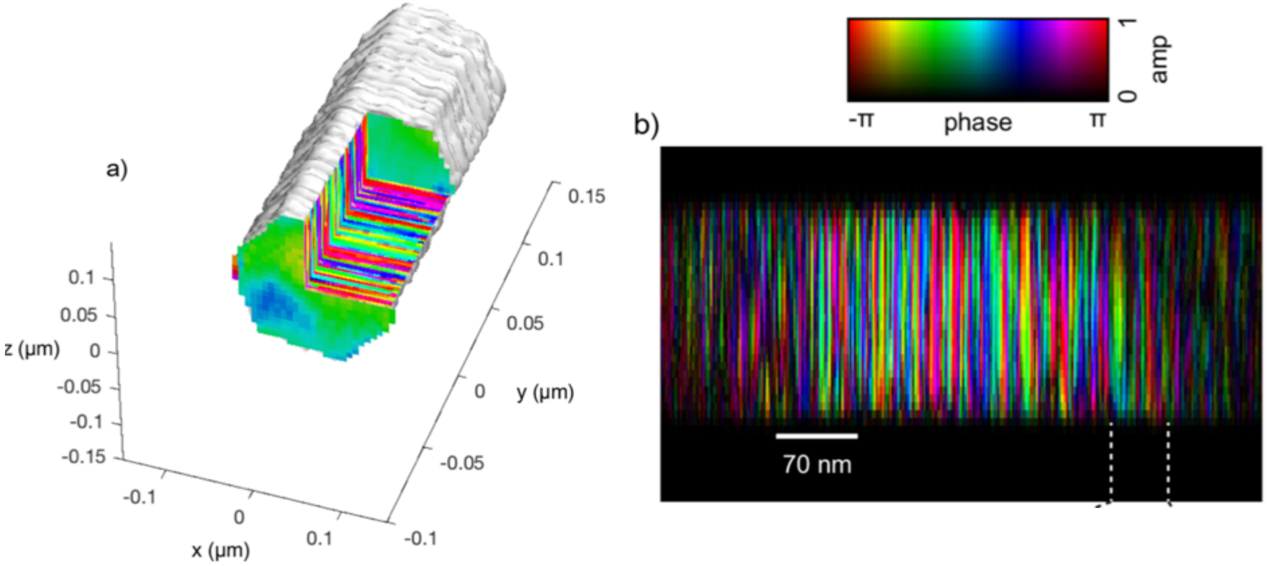
Scanning CDI (Ptycho): pushing the limits @ NSLS II

NANO LETTERS
Letter
Cite This: *Nano Lett.* 2018, 18, 811−819
pubs.acs.org/NanoLett

Measuring Three-Dimensional Strain and Structural Defects in a Single InGaAs Nanowire Using Coherent X-ray Multiangle Bragg Projection Ptychography

Megan O. Hill,[†] Irene Calvo-Almazan,[‡] Marc Allain,[§] Martin V. Holt,[∥] Andrew Ulvestad,[‡] Julian Treu,[⊥] Gregor Koblmüller,[⊥] Chunyi Huang,[†] Xiaojing Huang,[#] Hanfei Yan,[#] Evgeny Nazaretski,[#] Yong S. Chu,[#] G. Brian Stephenson,[‡] Virginie Chamard,[§] Lincoln J. Lauhon,[*,†] and Stephan O. Hruszkewycz[*,‡]





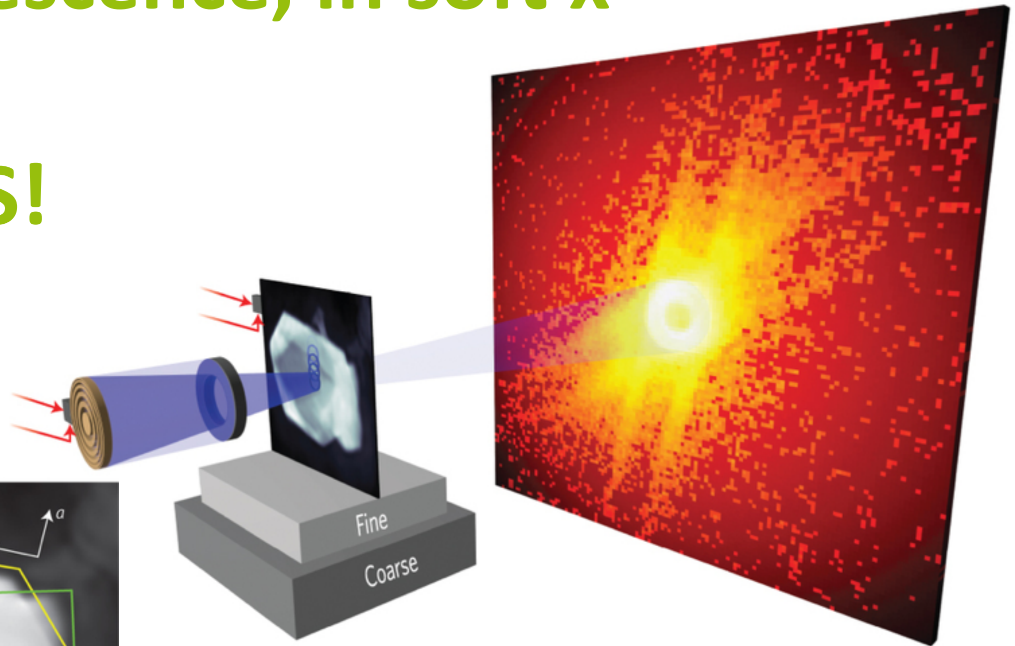
Development of dedicated acquisition schemes and reconstruction algorithms is an essential complement

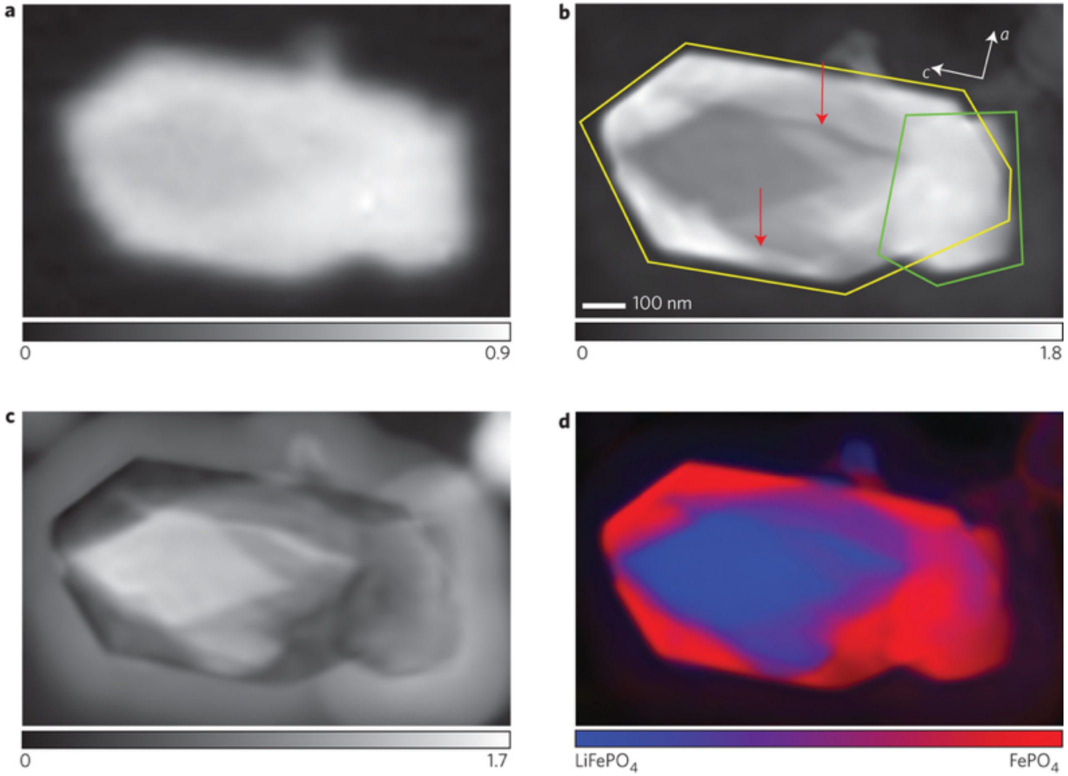
Visualising stacking faults!
Small Beam: 40nm x 80nm

Resulting pixel size: 1.3nm to 6.5nm ~intensity
BUT overall resolution limited by method & stability

Reconstruction reliable within 50nm because of twist

Ptychography and fluorescence, in soft x-ray regime
Record resolution at ALS!





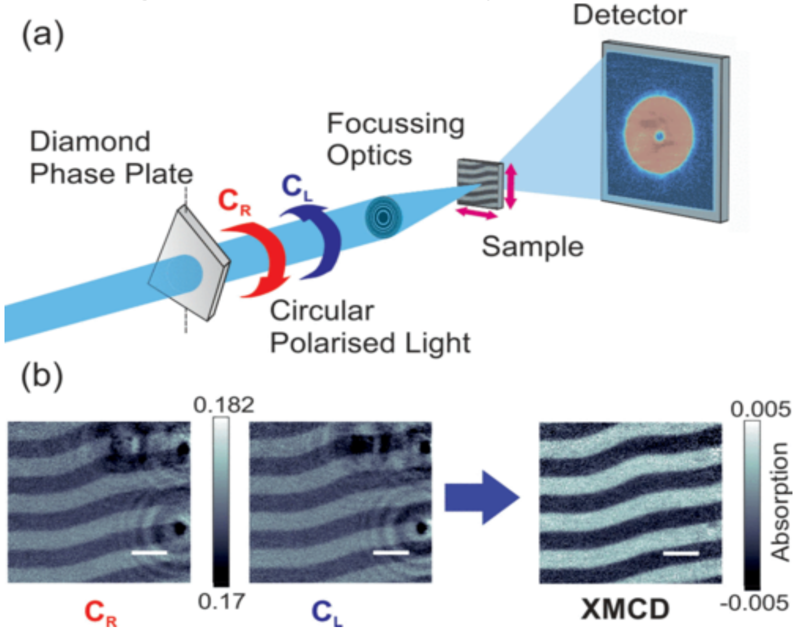
Delithiation in a nanoplate of LiFePO4,

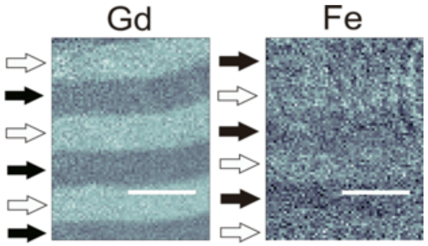
Nature Photonics **8**, 765–769 (2014)

2D & 3D of magnetic materials

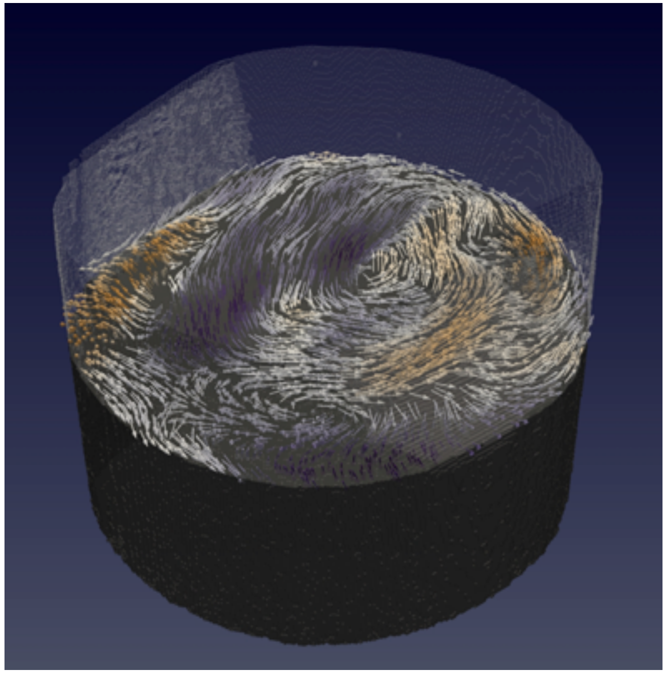
Spectro-microscopy of magnetic GdCo by Ptycho-tomography

Valerio Scagnoli & Claire Donnelly PSI





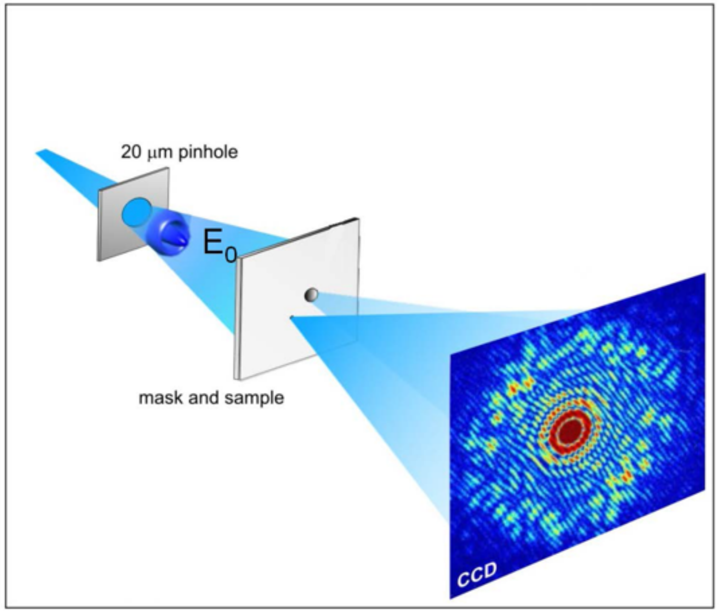
Challenge: Fe K-edge signal



Nature **547**, 290 2017

Holography in soft x-ray regime

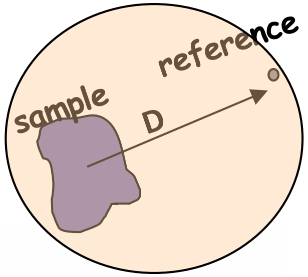
The **phase** is encoded in the diffraction pattern (**amplitude**)
=> No need for inversion algorithms

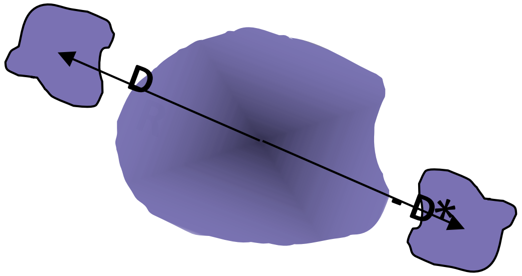


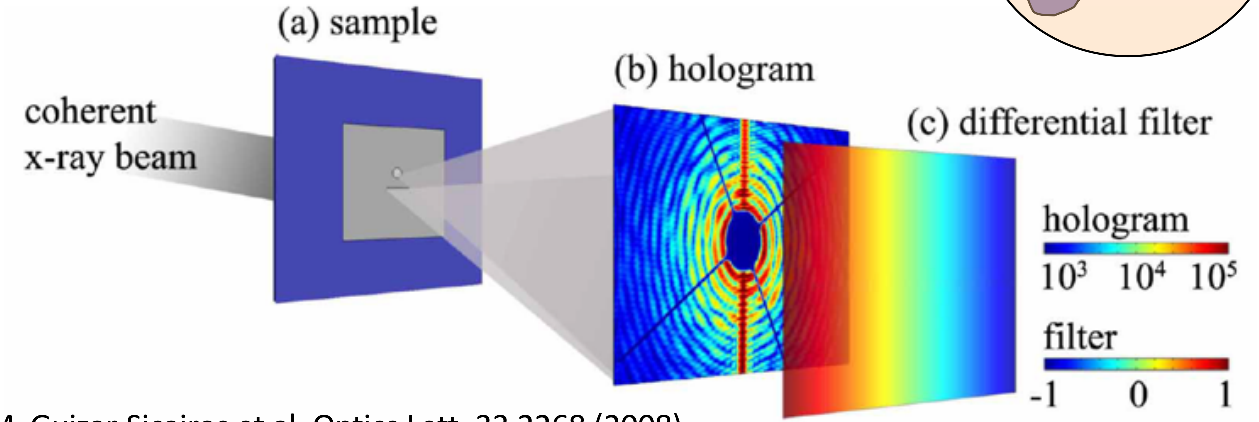
$$
\begin{aligned}
I(\mathbf{q}) &= |\ FT(\rho(\mathbf{r})+\delta(\mathbf{r}\text{-}\mathbf{D}))\ |^2 = |\ \rho(\mathbf{q}) + \exp(i\mathbf{q}\mathbf{D})\ |^2 \\
&= \rho(\mathbf{q})r^*(\mathbf{q}) + 1 + \rho(\mathbf{q})\exp(-i\mathbf{q}\mathbf{D}) + \rho^*(\mathbf{q})\exp(i\mathbf{q}\mathbf{D})
\end{aligned}
$$

From the direct inversion of the **intensity**:

$$
\begin{aligned}
FT^{-1}(I(\mathbf{q})) &= \rho(\mathbf{r}) \otimes \rho(\mathbf{r}) + \delta(0) \\
&\quad + \boldsymbol{\rho(\mathbf{r}\text{-}\mathbf{D}) + \rho^*(\text{-}\mathbf{r}+\mathbf{D})}
\end{aligned}
$$





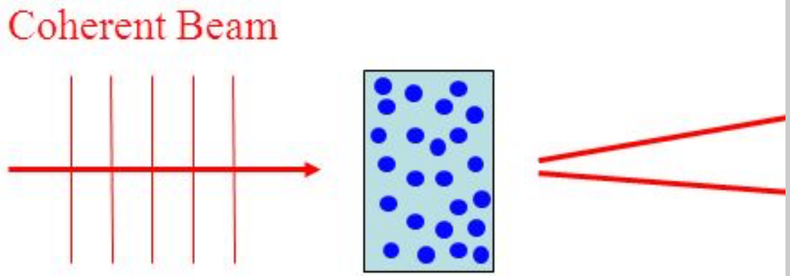


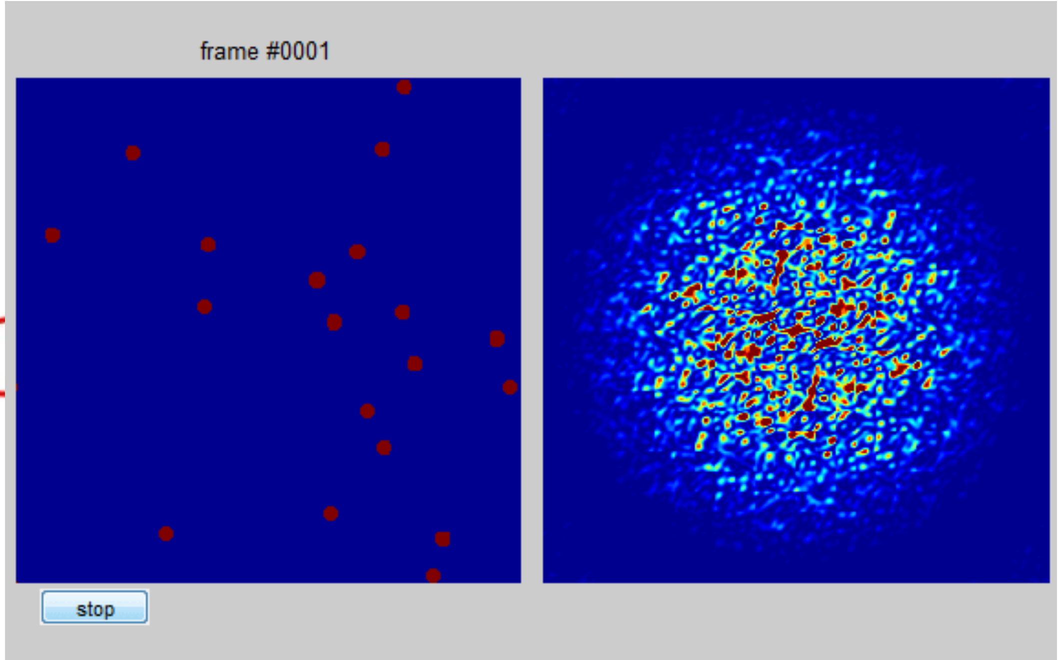
M. Guizar-Sicairos et al. Optics Lett. 33 2268 (2008)
Holography with Extended Reference by Autocorrelation Linear Differential Operator

XPCS

X-ray Photon Correlation Spectroscopy

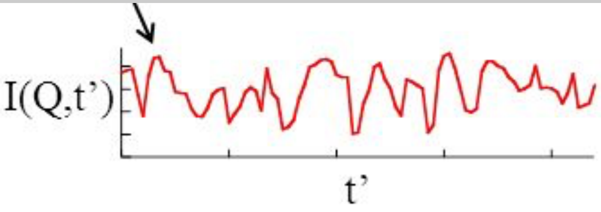
Dynamic light scattering with x-rays





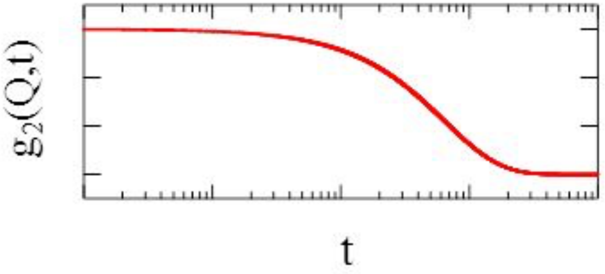
Autocorrelation of intensity…

$$g_2(\vec{Q},t) = \frac{\langle I(\vec{Q},t')I(\vec{Q},t'+t)\rangle}{\langle I(\vec{Q},t')^2\rangle}$$

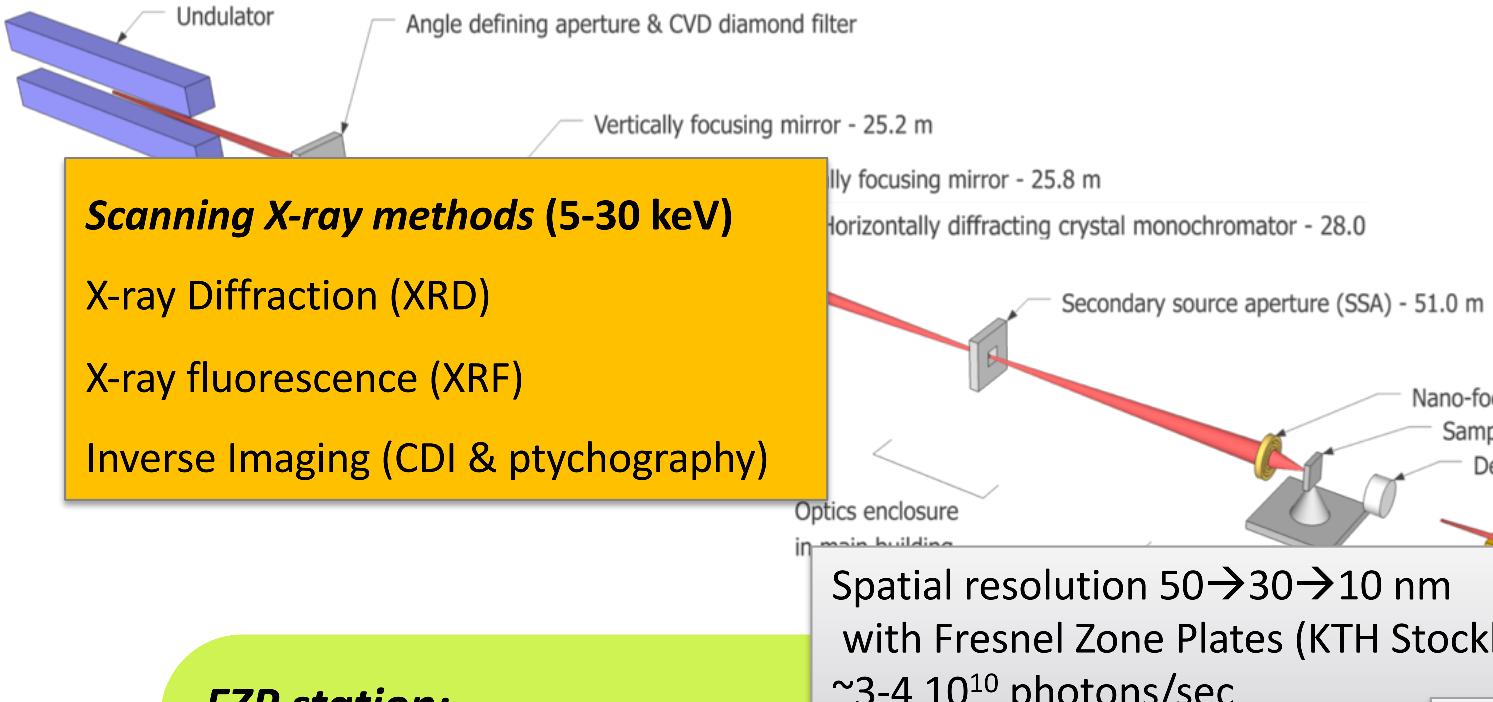


Gives dynamic structure factor:

$$g_2(\vec{Q},t) = 1 + \beta g_1^2(\vec{Q},t) = 1 + \beta[\frac{S(\vec{Q},t)}{S(\vec{Q},0)}]^2$$



NanoMAX overview



***Scanning X-ray methods* (5-30 keV)**

X-ray Diffraction (XRD)

X-ray fluorescence (XRF)

Inverse Imaging (CDI & ptychography)

Spatial resolution 50→30→10 nm with Fresnel Zone Plates (KTH Stockholm)
~3-4 10^{10} photons/sec

Spatial resolution ~100 nm with KB-mirror optics (JTEC)
~2-3 10^{11} photons/sec

FZP station:

Ultimate focus, limited space for sample environment

- In-vacuum
- High stability => little flexibility
- Ad-hoc sample environment
- Cryo-cooling (sample damage)

KB station:

Flexible geometry, large sample environment

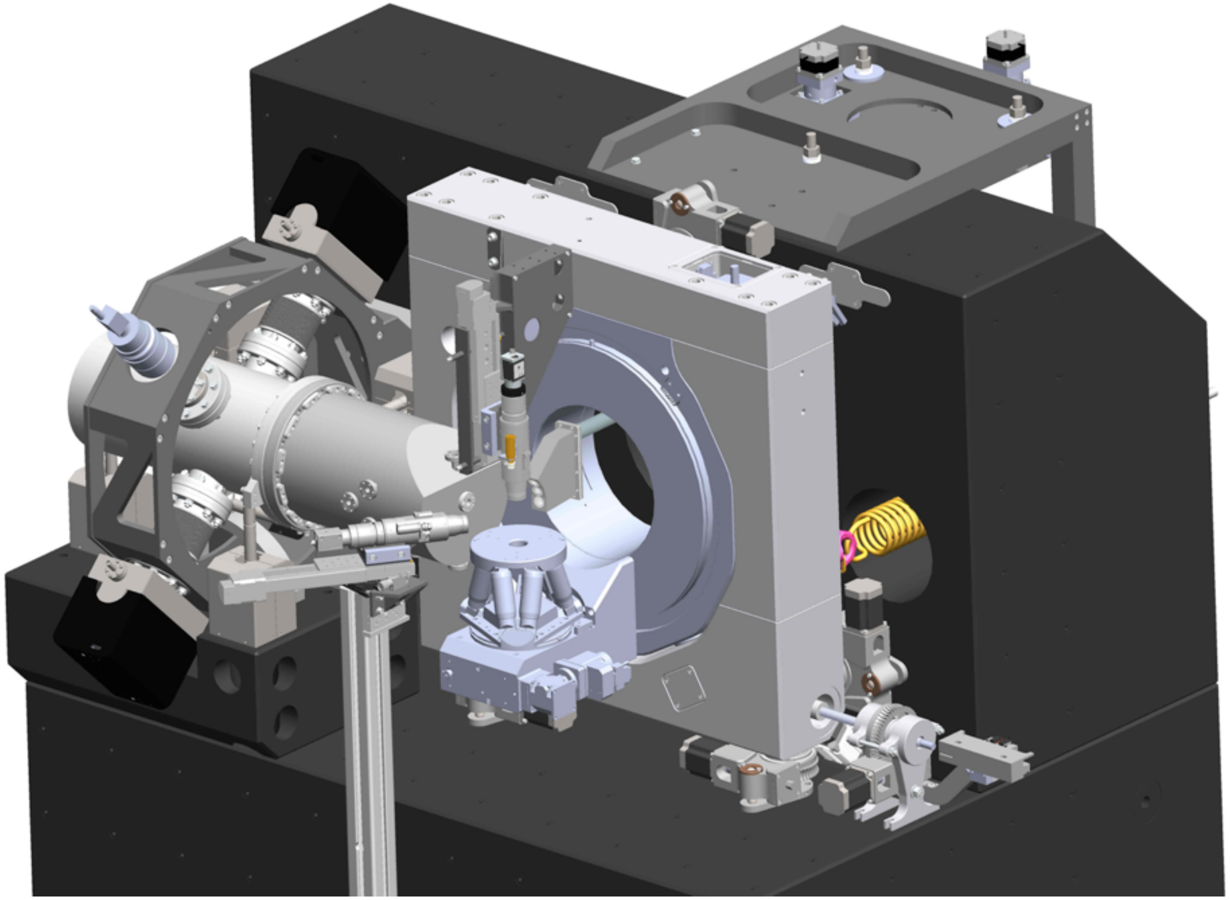
- Multipurpose diffractometer
- Adapted sample environment
- Combined analysis (in-situ): *heating, cooling, external electric / magnetic fields, controlled pressure / gas environment, ...*

NanoMAX:
CXDI EndStation design: flexibility & stability

- **Diffraction & 3D inverse imaging**
- **Spectroscopy**
- **Sample environment (1Kg, 100mm)**

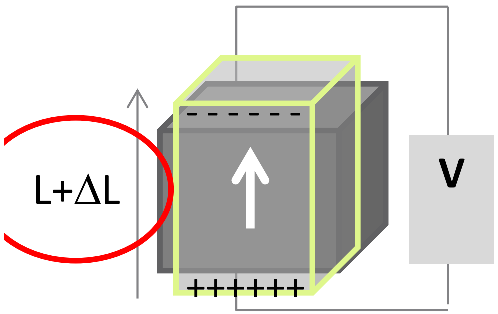
- Data acquisiton & online visualisation
- Program for coherent data analysis (collaboration V. Chamard, P Thibault)

- 2-circles goniometer
- KB mirror optics
- On solid granite block
- Detectors decoupled from sampl
- 5 Kg total load (sample environm
- Huber 2-circles
- Simple design for support
- In-line and top view microscopes



Local probe

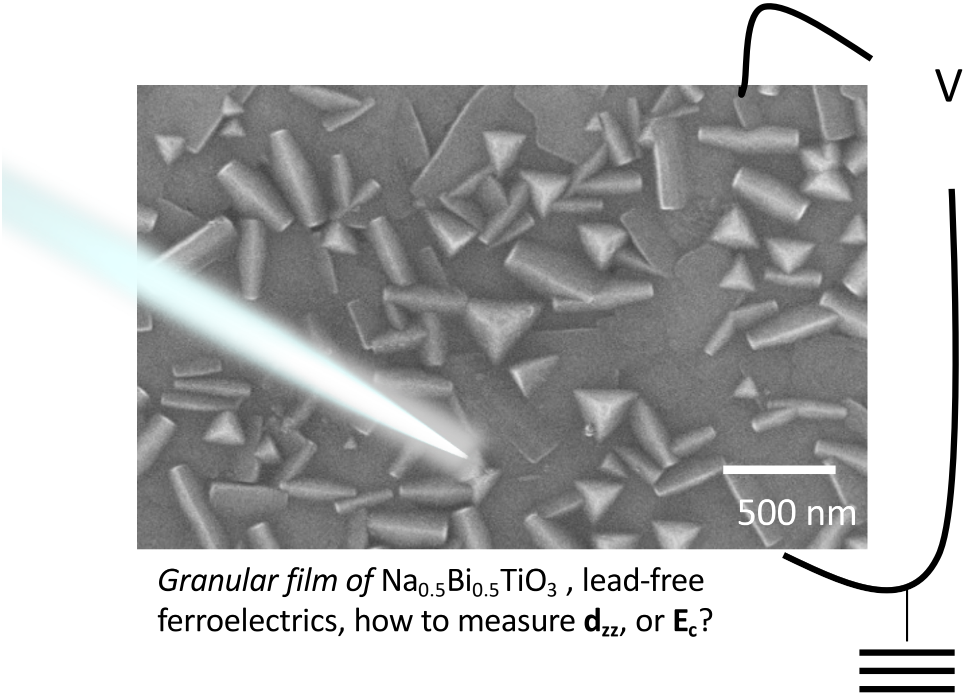
Local response in piezoelectrics



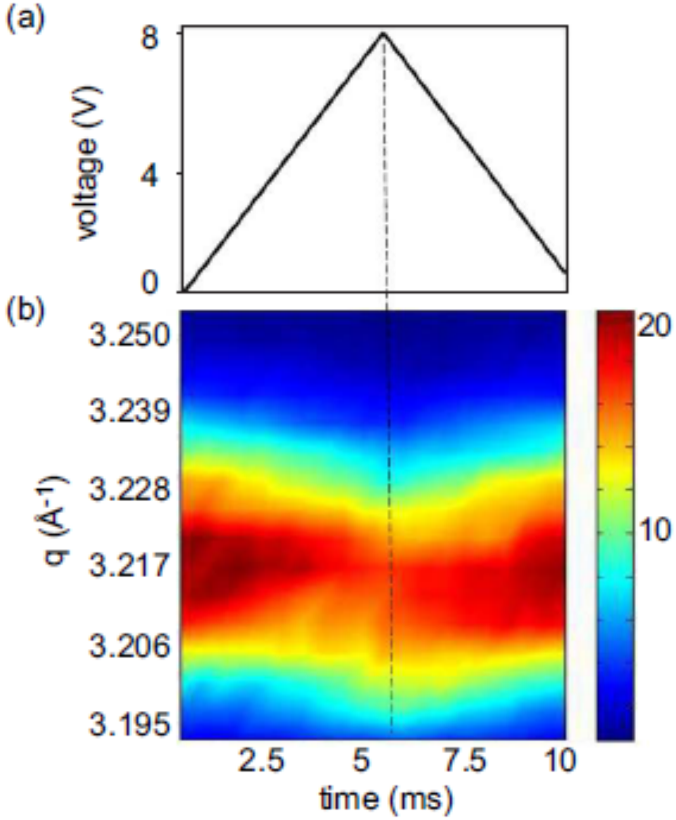
$\varepsilon = \mathbf{d}_{zz}E$

E [V/cm]

$\varepsilon = \Delta L/L$



Granular film of $Na_{0.5}Bi_{0.5}TiO_3$, lead-free ferroelectrics, how to measure $\mathbf{d}_{zz}$, or $\mathbf{E}_c$?



$$d_{zz} = \frac{\Delta L}{L} \cdot \frac{1}{E}$$

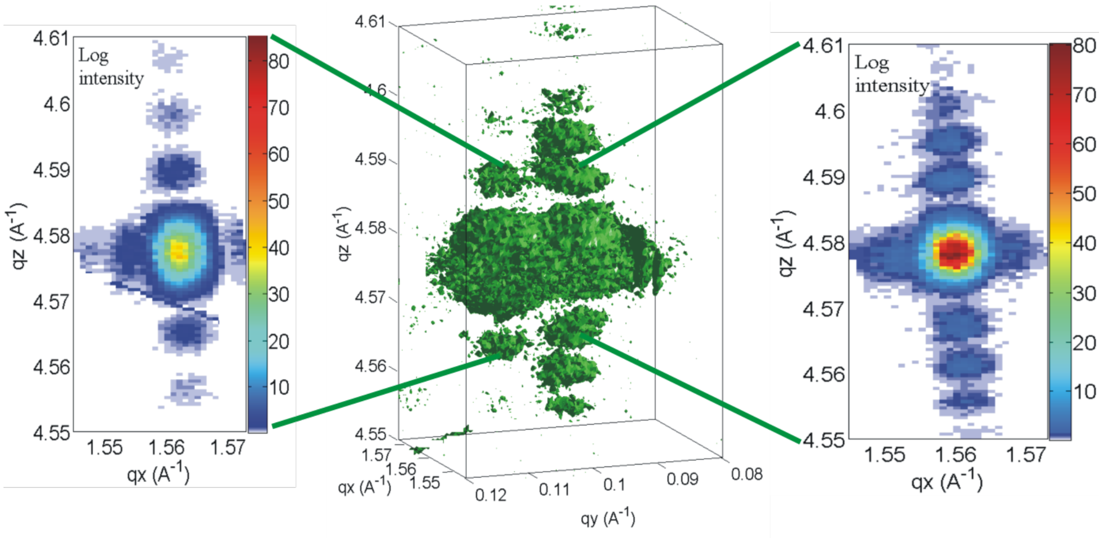
d_{33} (pm/V)
66 ± 2
55 ± 3
88 ± 6
48 ± 2
73 ± 8
92 ± 3

Distribution of properties reflects morphological inhomogeinity

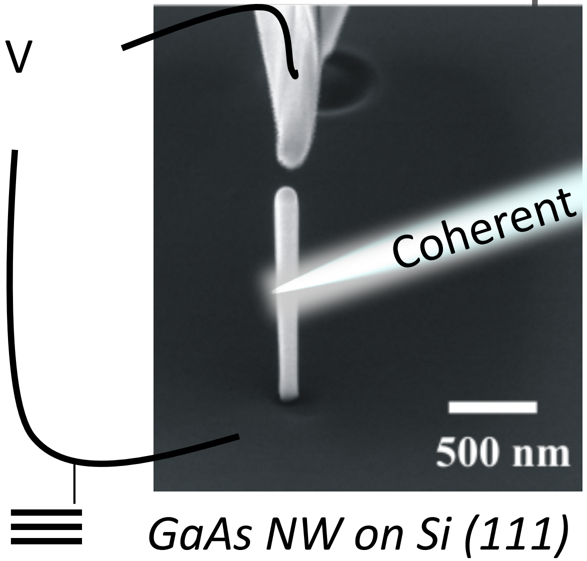
Appl. Phys. Lett. 105 242901 (2014)

Local probe

GaAs NanoWires - transport

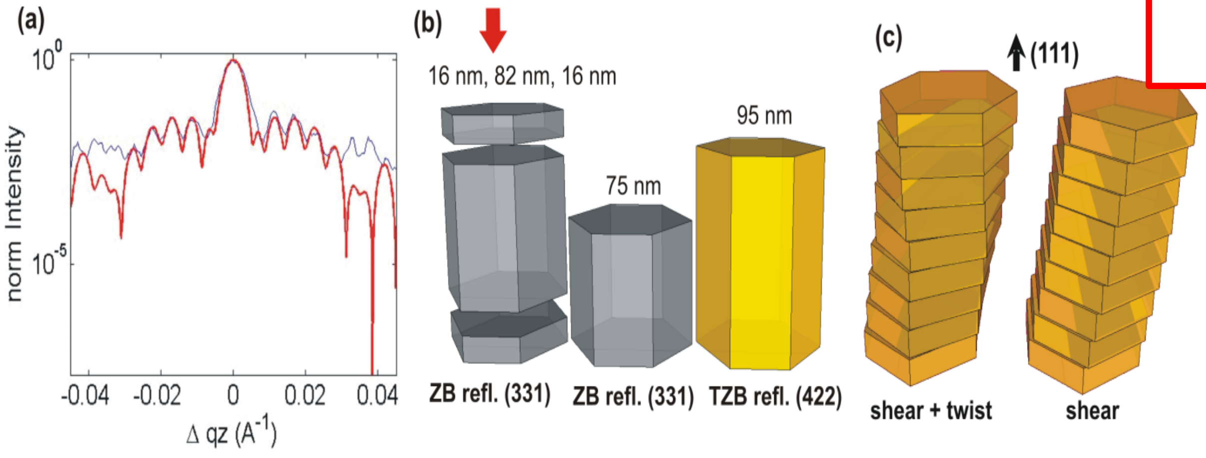


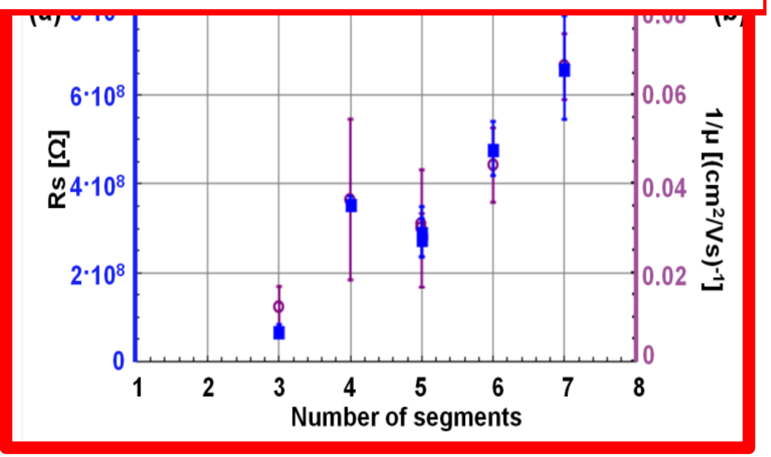
From **coherent diffraction patterns** one can extract the type and the number of different segments in each NW,.



GaAs NW on Si (111)

Distribution of properties reflects structural inhomogeinity





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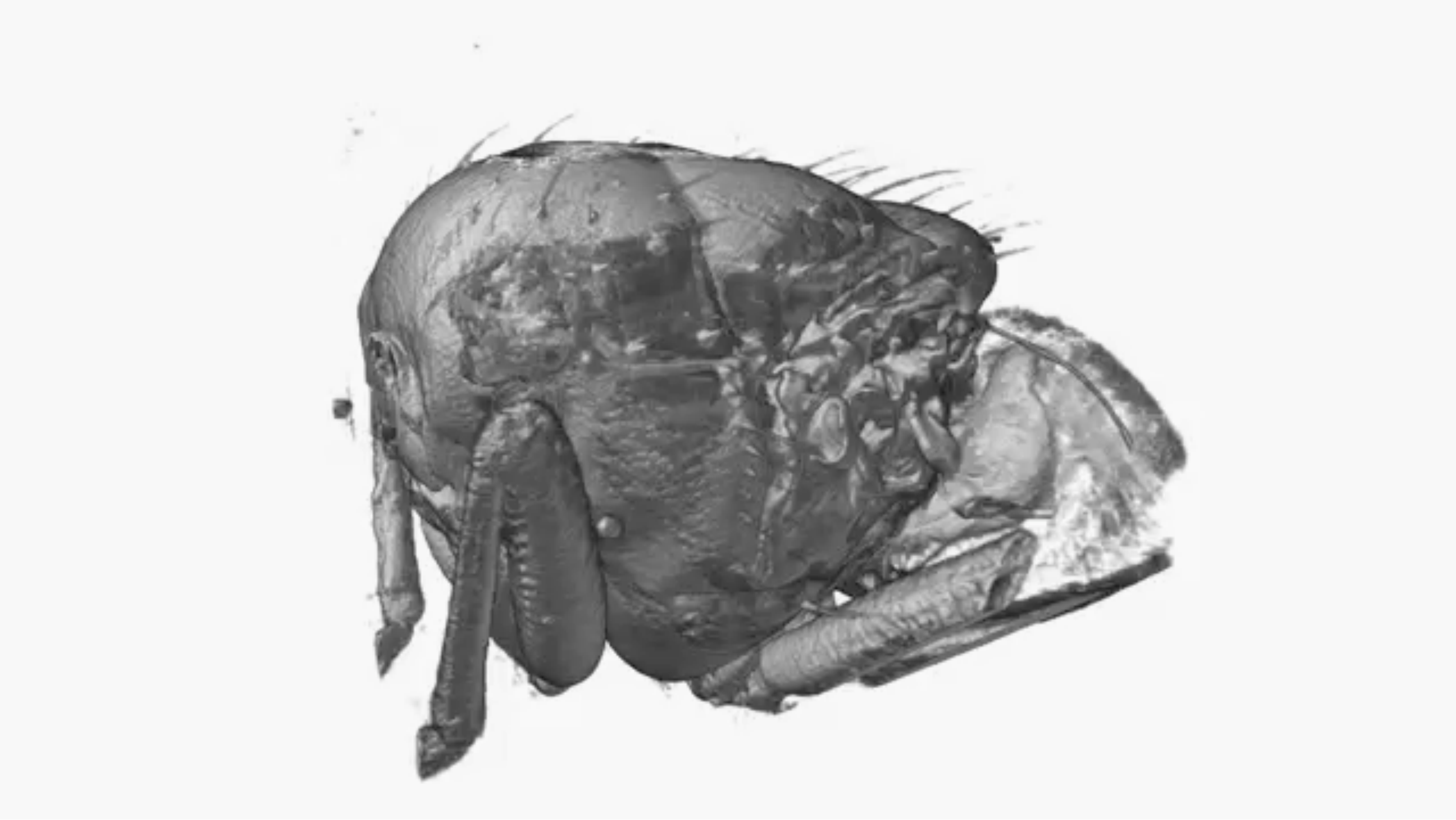
"Live"-science: 4D imaging



Wing oscillation - 150 beats per second.
Study important for biomimicry engineering

Stampanoni et al, PSI -March 2014