# Ultrafast Materials Physics

Ian Robinson
Jesse Clark
Ross Harder
Gang Xiong
Marcus Newton
Felix Hofmann

Brookhaven National Laboratory
London Centre for Nanotechnology

Physics Colloquium,
Arizona State Universtity
Tempe, April 2018

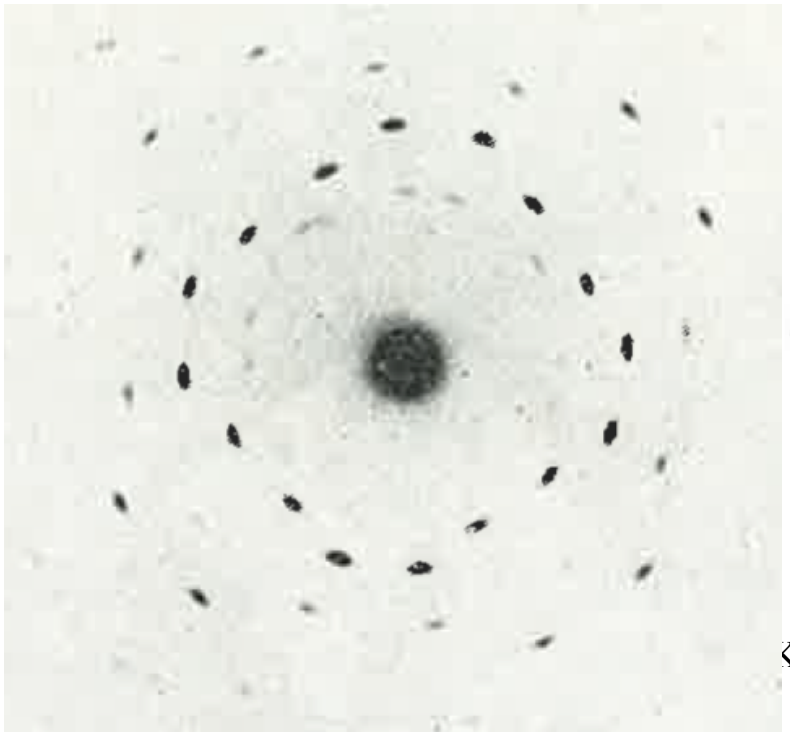
# Outline

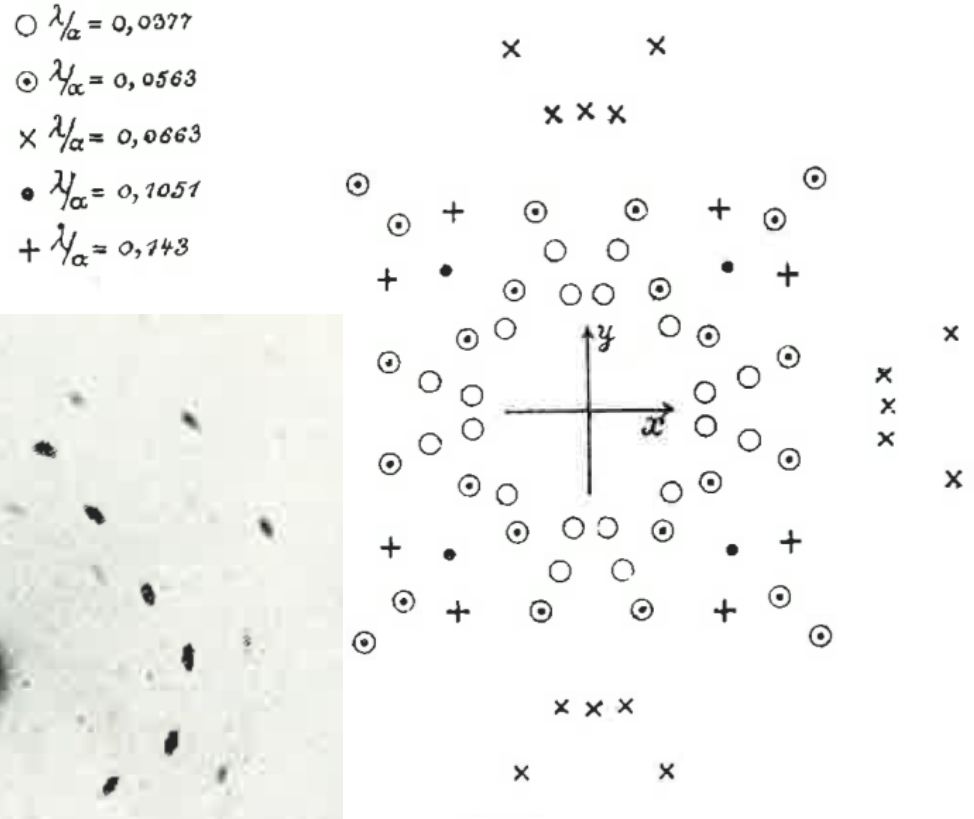
- Bragg Coherent Diffraction Imaging
- Nanocrystal structures
- Crystal strain as complex density
- Nanoscale alloying
- Ultrafast snapshots of moving matter
- Strain Tensor imaging

# Max von Laue, 1879-1960

## Proceedings Bavarian Academy of Sciences, 1912

## Zinc Blende, ZnS, Nobel prize 1914





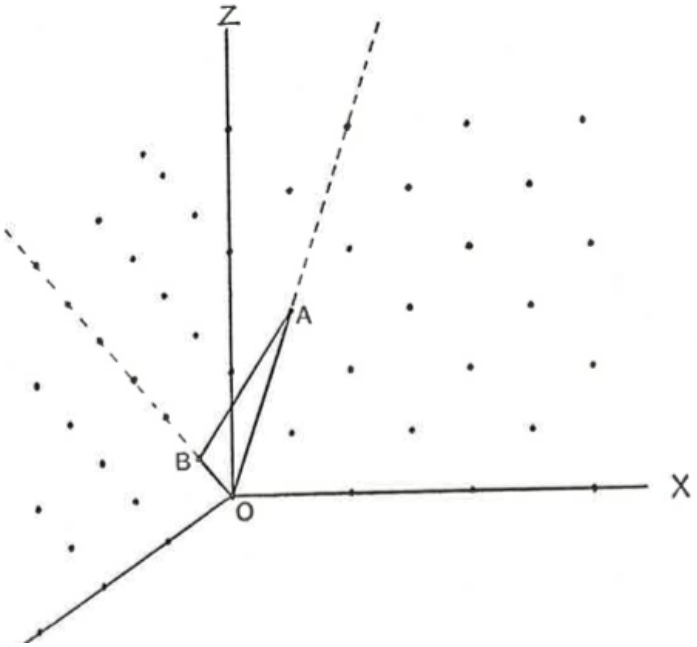

# W. L. Bragg 1890-1971

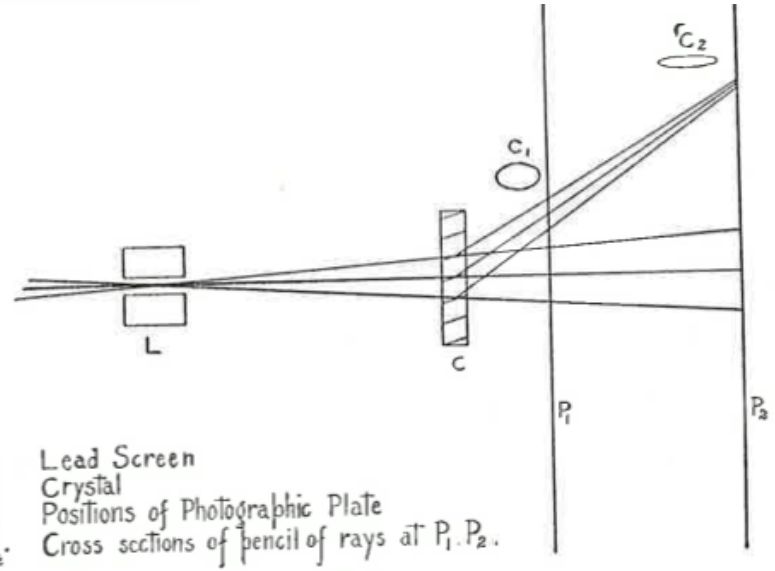
THE DIFFRACTION OF SHORT ELECTROMAGNETIC WAVES BY A CRYSTAL

*by*

*W. L. Bragg, B. A., Trinity College*

(communicated by Prof. Sir J. J. Thomson)
(*Read 11 November 1912*)



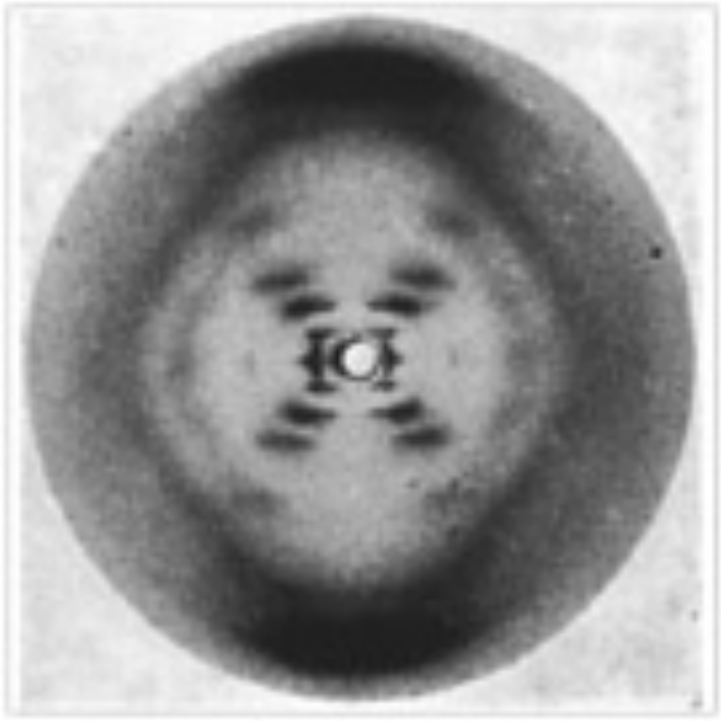




L Lead Screen
C Crystal
P1 P2 Positions of Photographic Plate
C1 C2 Cross sections of pencil of rays at P1 P2.

Fig. 2.

# Diffraction as Fourier Transform

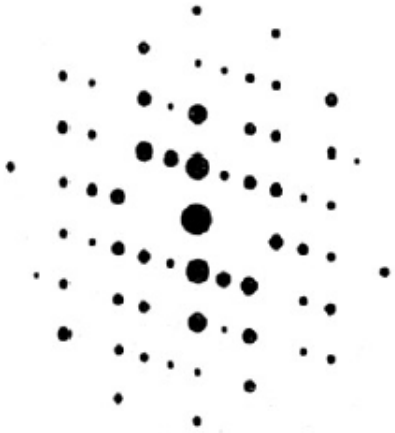
## Rosalind Franklin's DNA "photo 51" 1952

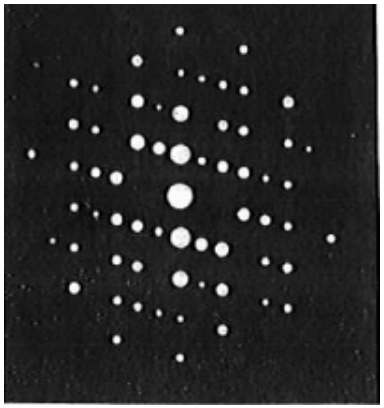



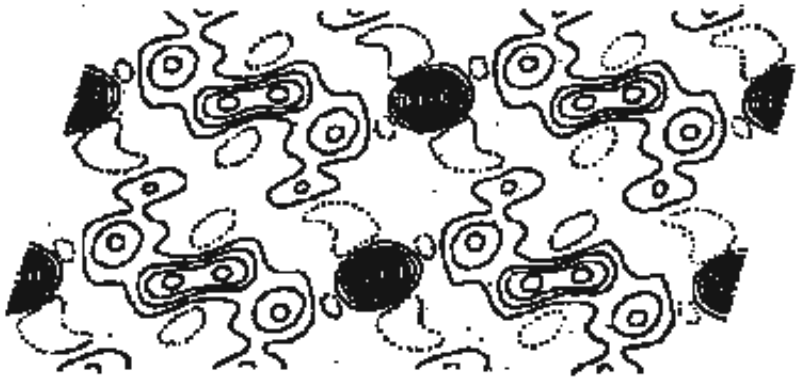
# Rosalind Franklin's DNA "photo 51"

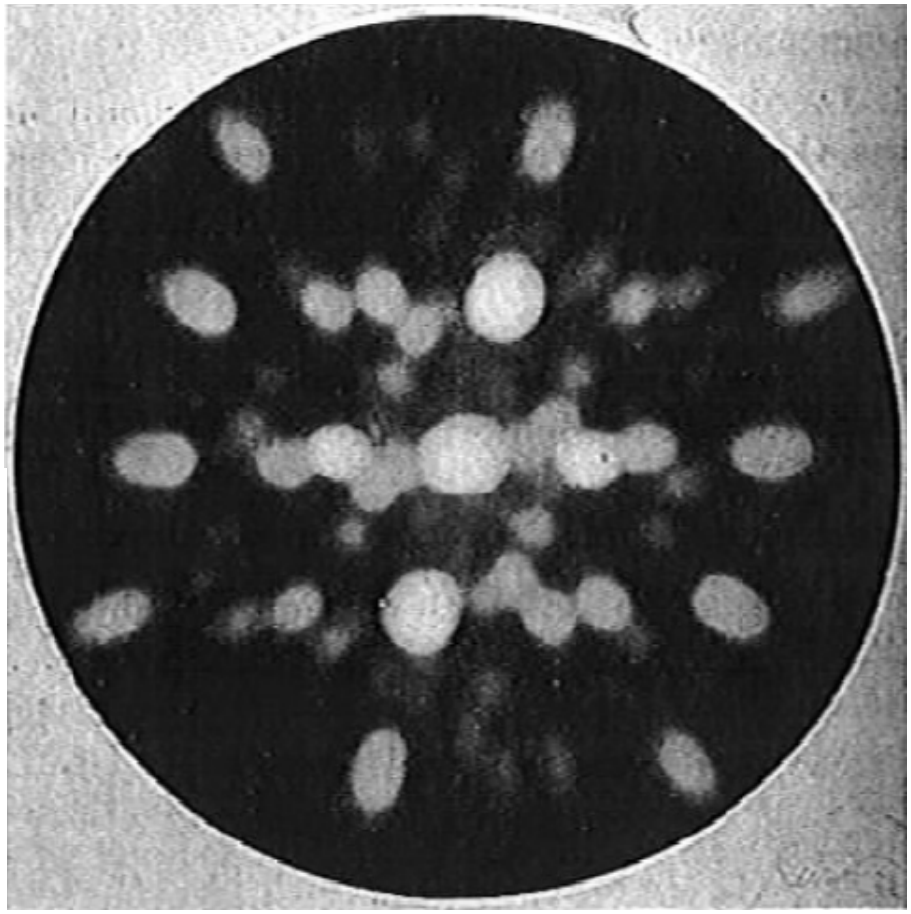
# Bragg's "X-ray Microscope"

W.L.Bragg Nature, 143, 678 (1939): diopside, $CaMg(SiO_3)_2$









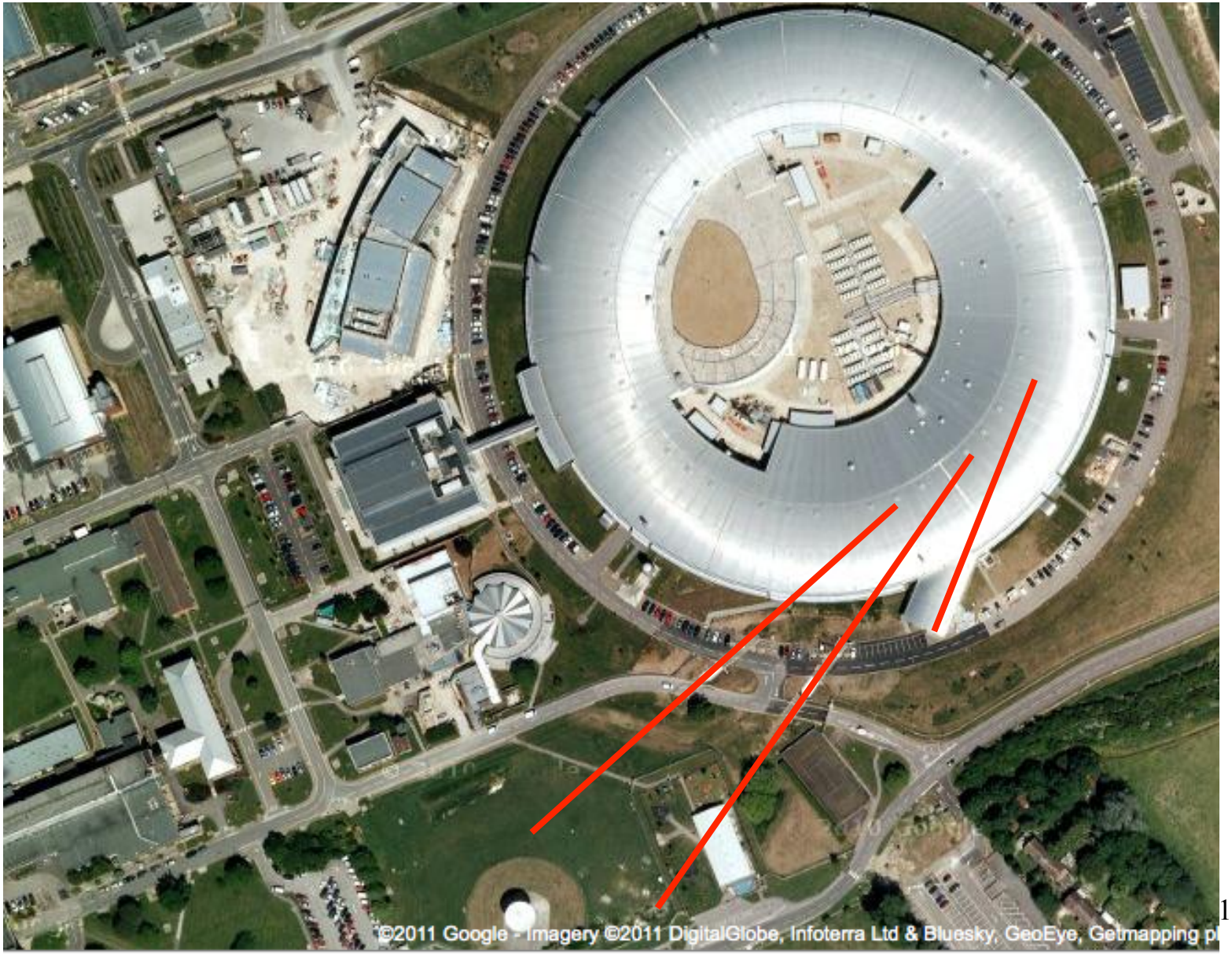
# Arizona X-ray Pioneers

John Cowley
1923-2004

Steve Wilkins
1946-2013

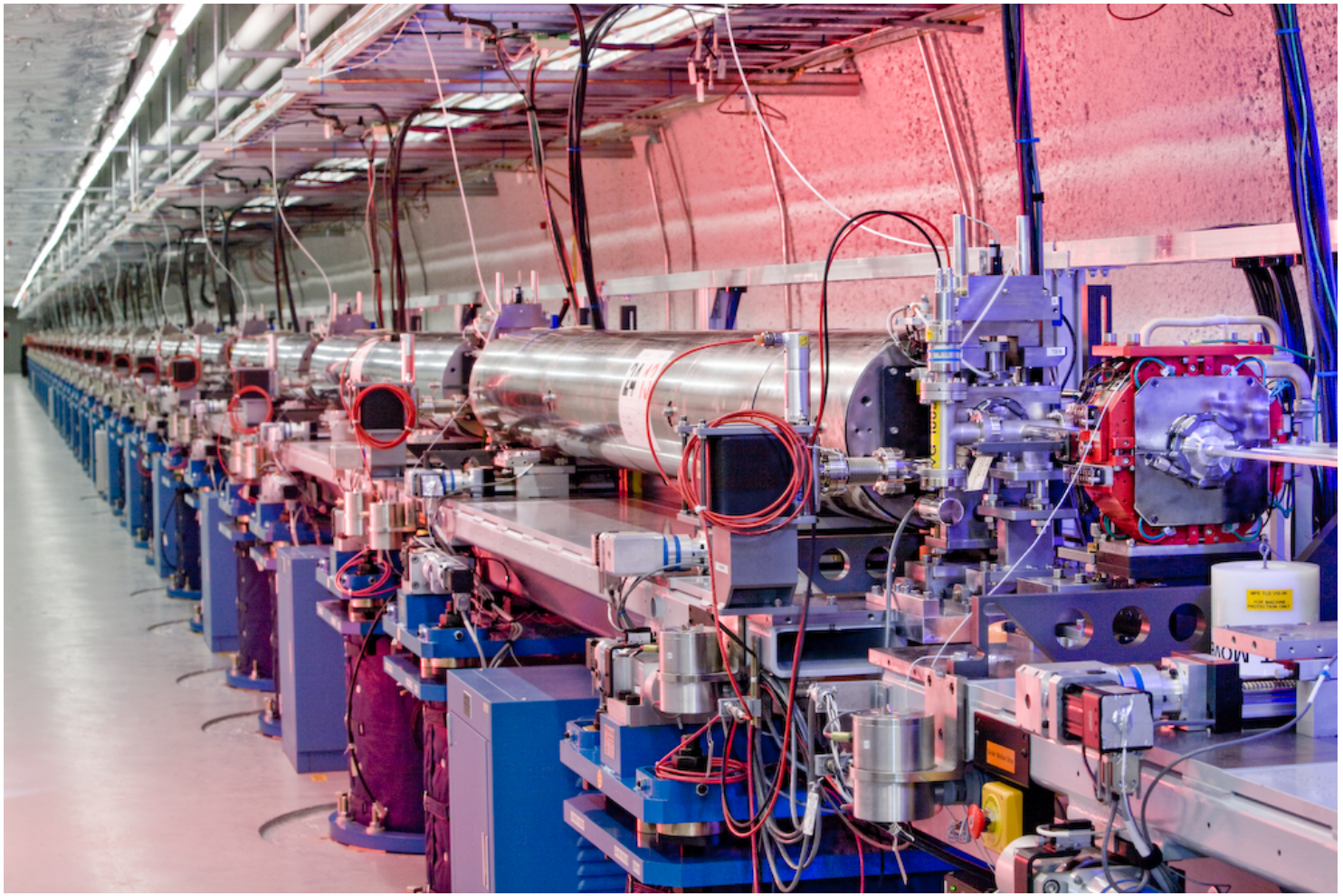
# Advanced Photon Source (ANL)

# Diamond Light Source, 2009

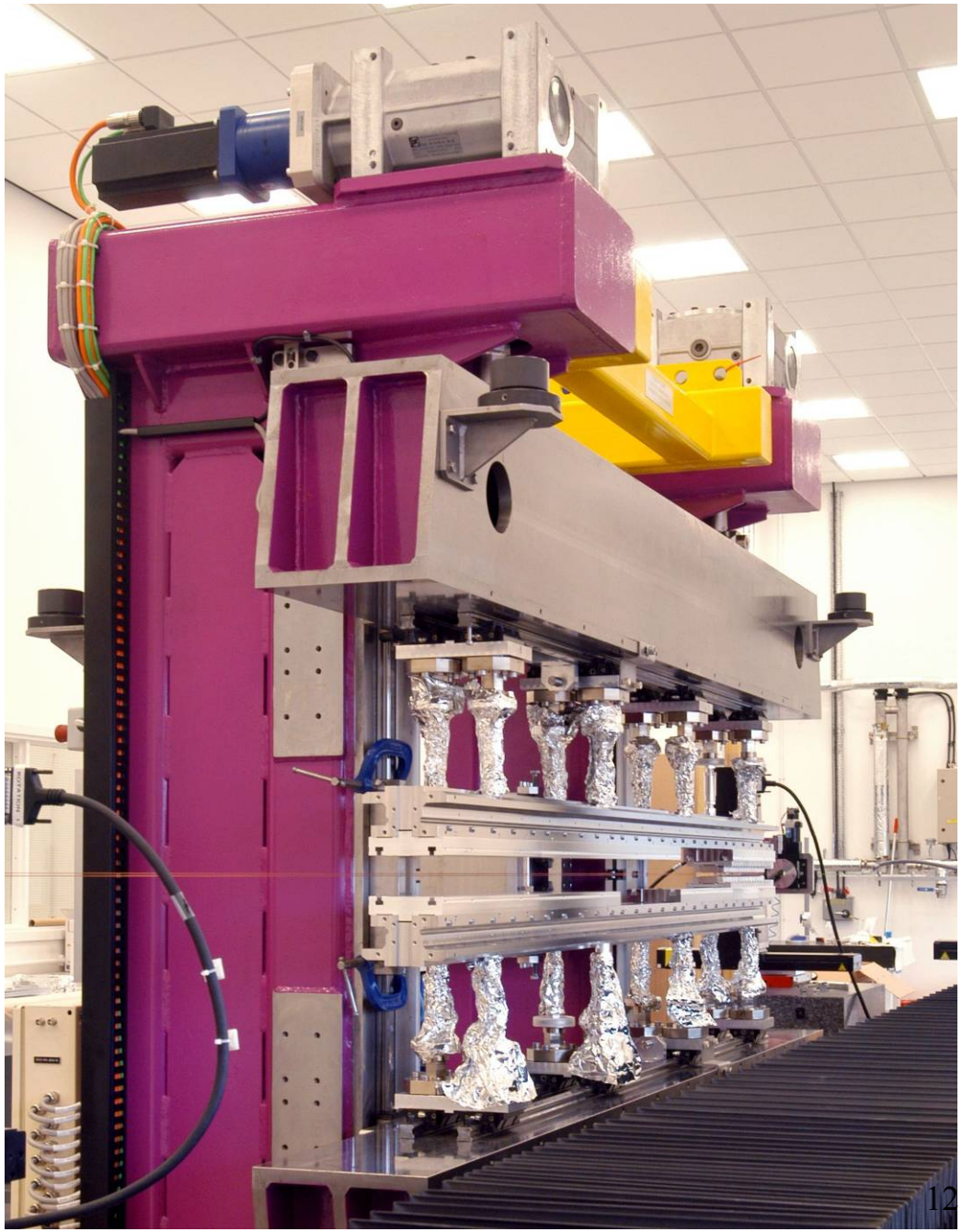


# Linac Coherent Light Source

## X-Ray Free-electron Laser, SLAC, Stanford, USA

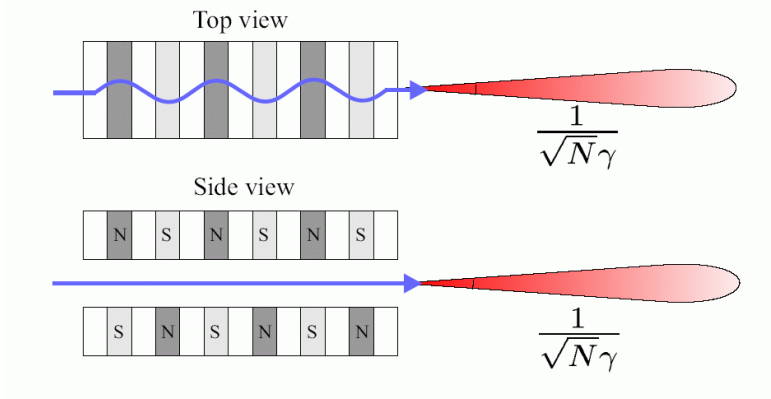


# Diamond in-vacuum X-ray Undulator

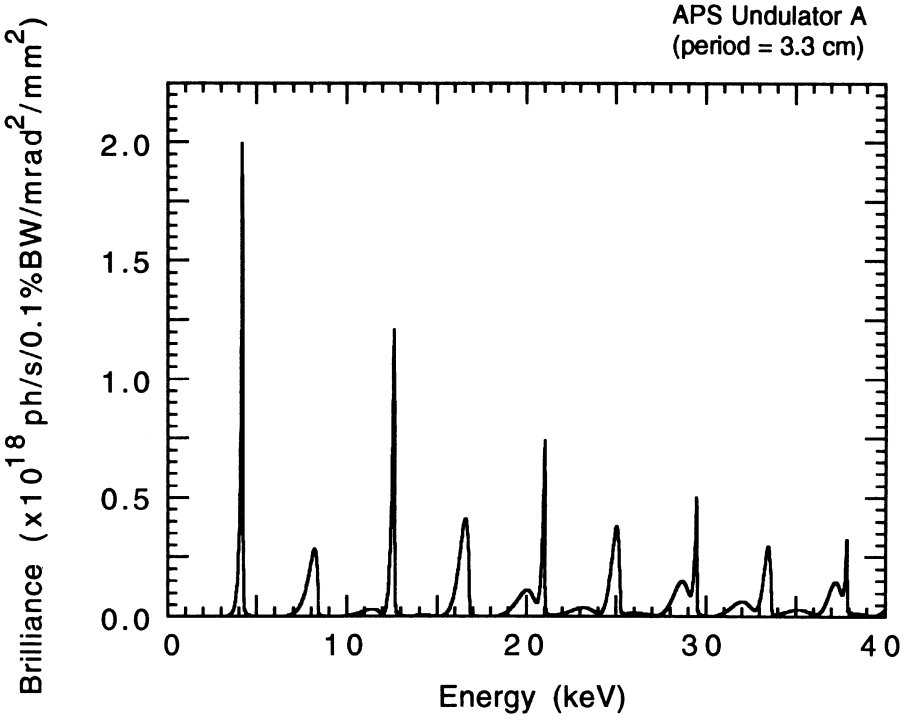


# X-ray Undulator Principle

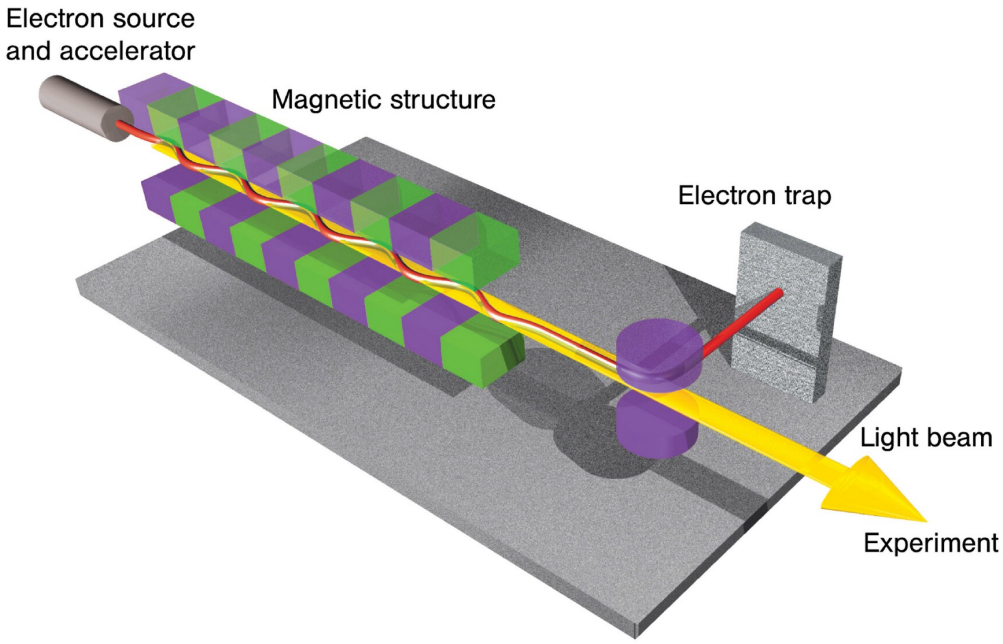
Vitalii Ginzburg, Novosibirsk (1947)

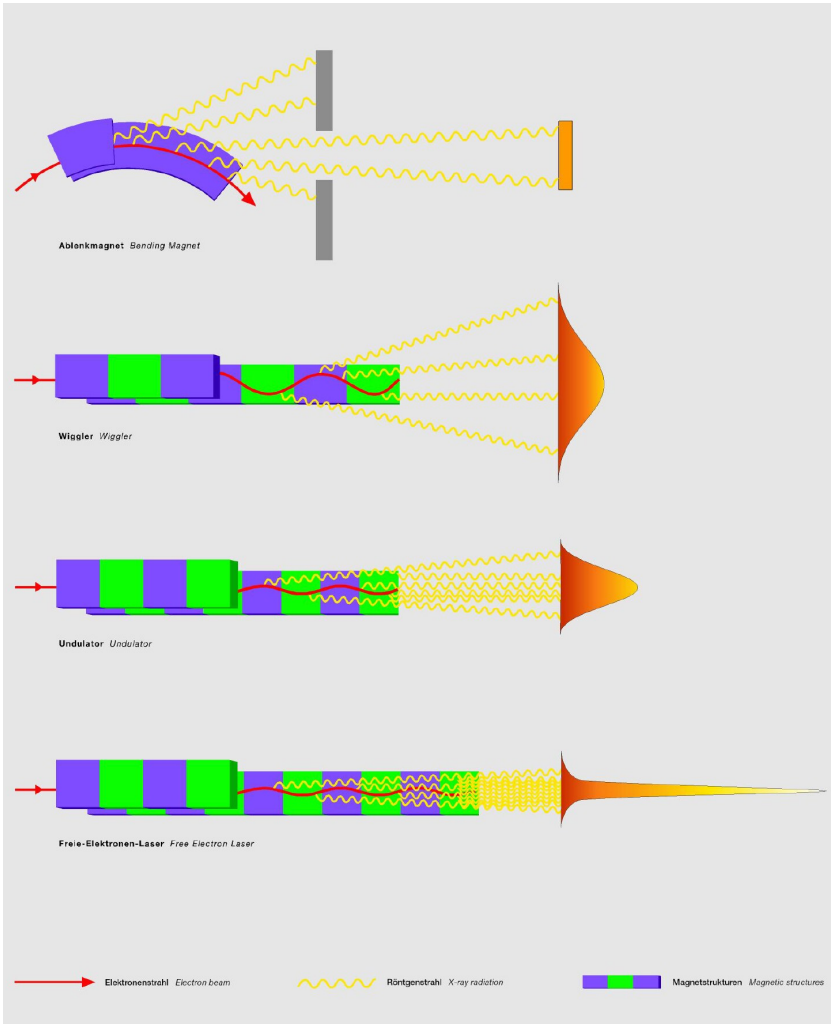



$$\lambda_X = \frac{\lambda_U}{2\gamma^2}\left\{1 + \frac{K^2}{2} + (\gamma\theta)^2\right\}$$

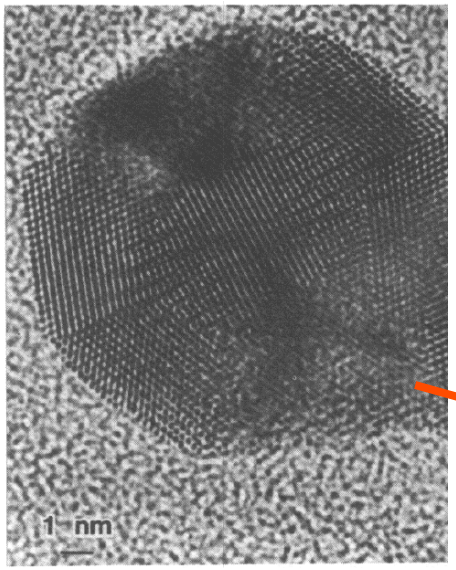


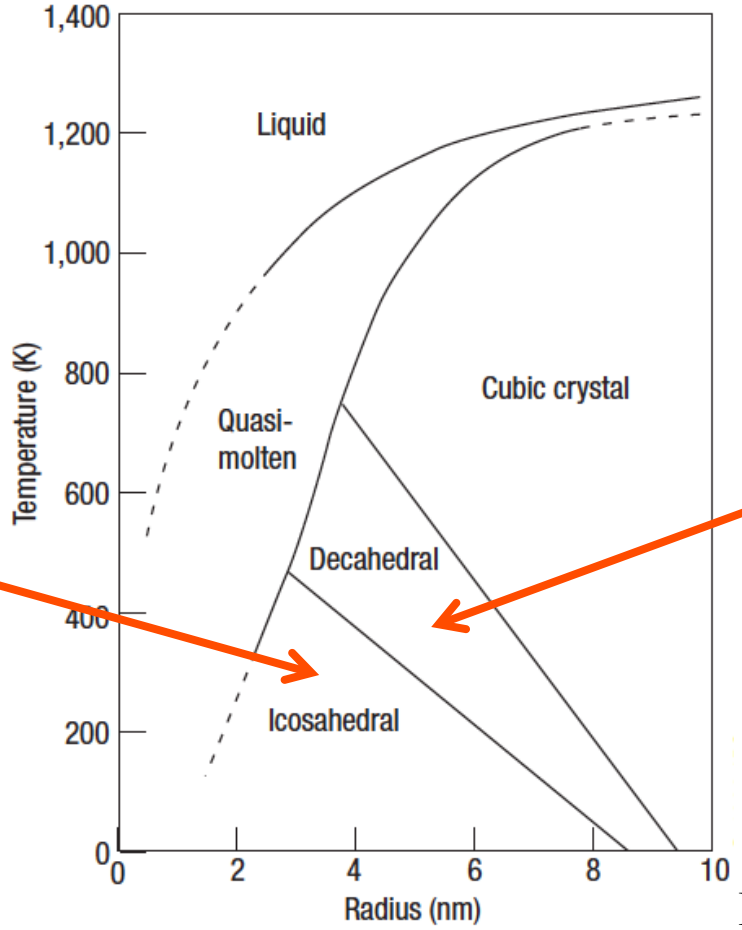
# XFEL principle

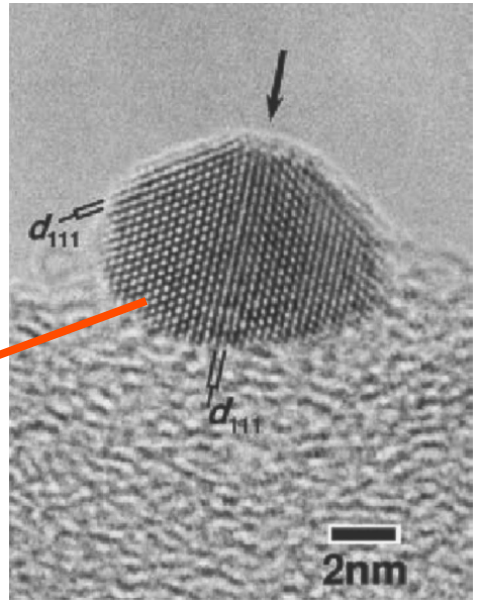


# Structure of Gold vs Size




L. D. Marks, RPP (1994)

Koga and Sugawara (2003)

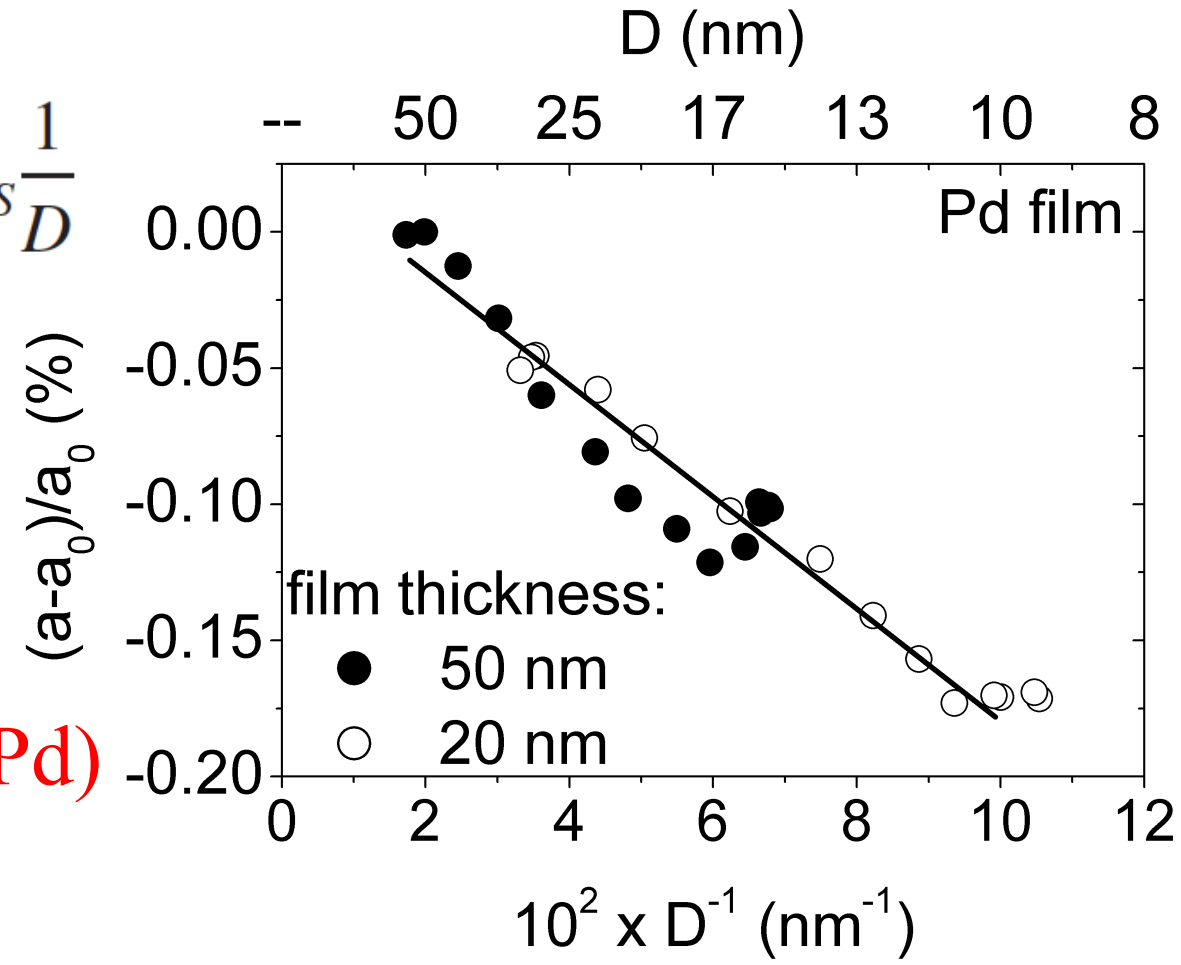
# Contraction of Small Particles

## Gibbs Thomson pressure + Bulk modulus

## Sheng, Welzel & Mittemeijer, APL 97 153109 (2010)

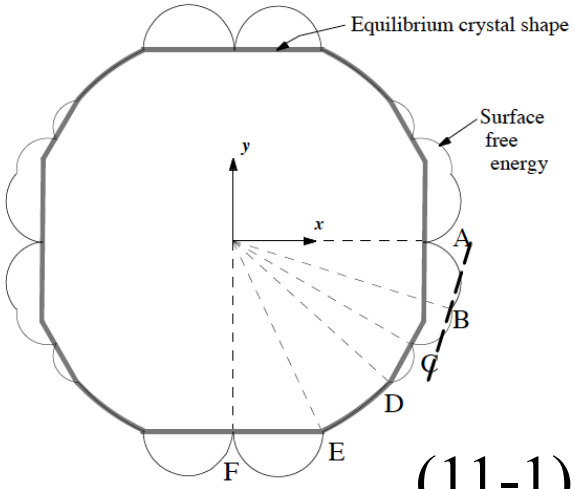
$$\frac{a - a_0}{a_0} = -\frac{4}{3K}\sigma_S\frac{1}{D}$$

$\sigma_S = 2\text{N/m}$ (Pd)



# Equilibrium crystal shape

## Wulff construction + Gibbs Thomson (Young-Laplace) pressure




γ = 2N/m

P = 2γ/R = 70MPa

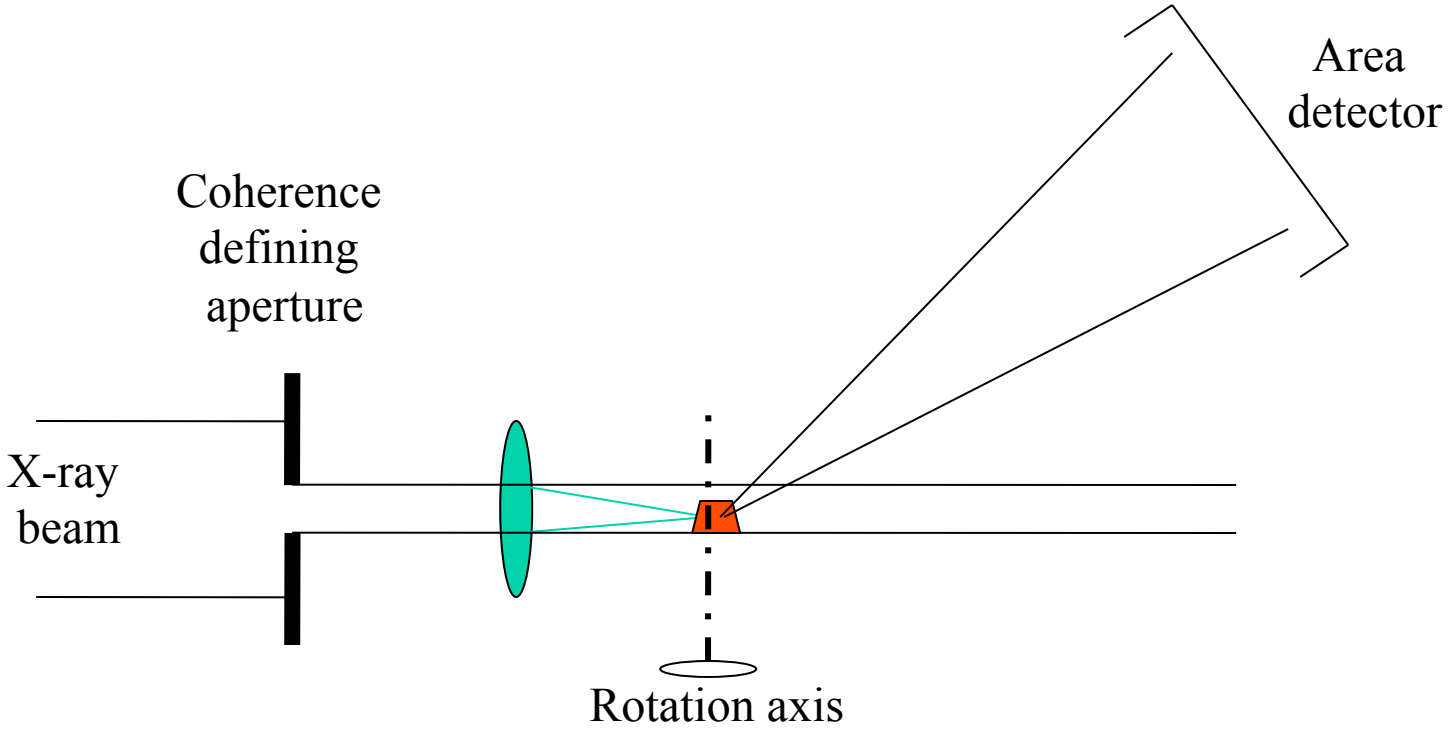
ε = 0.1%, Φ = 0.6 rad

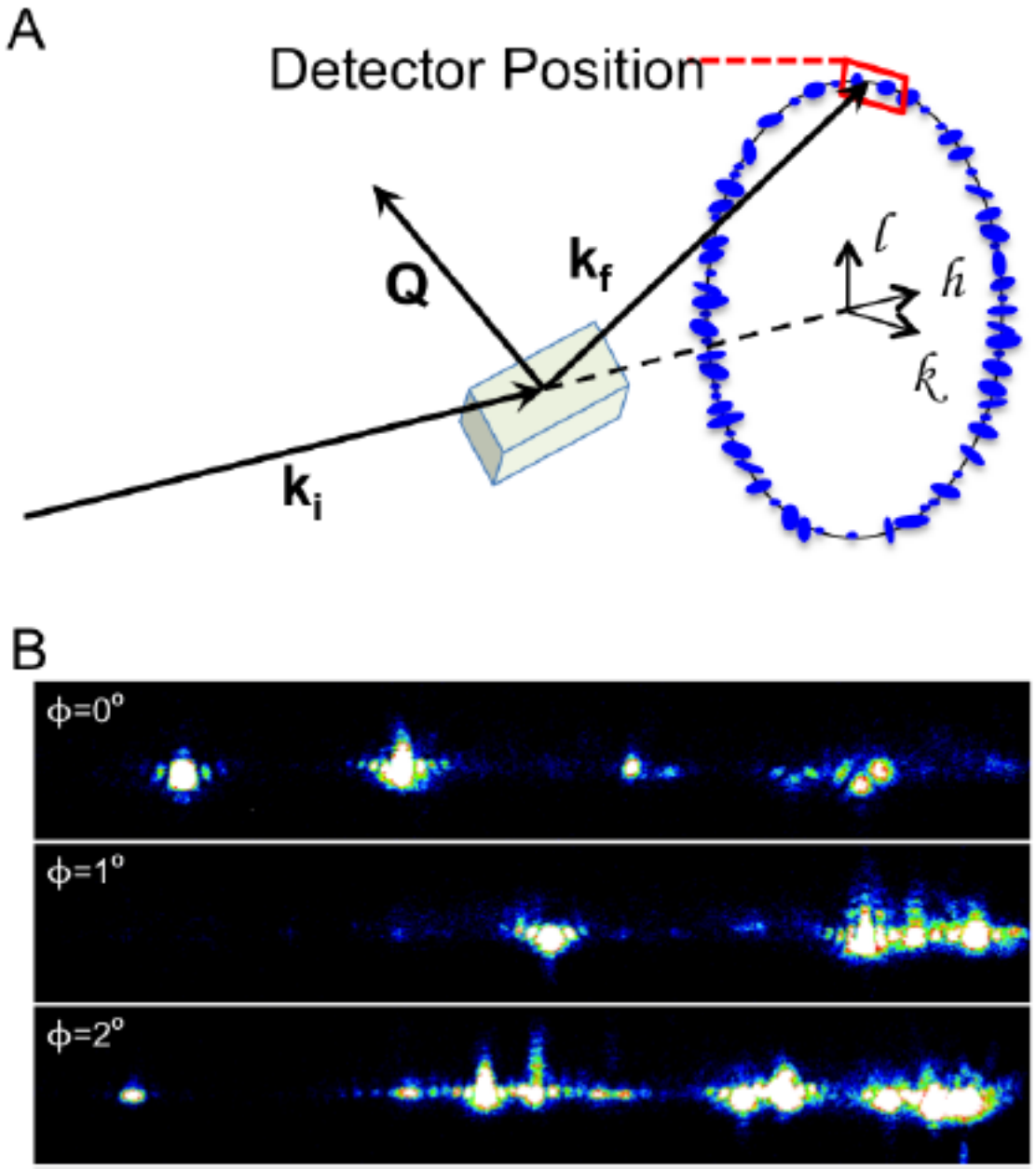
(111)

R = 30nm

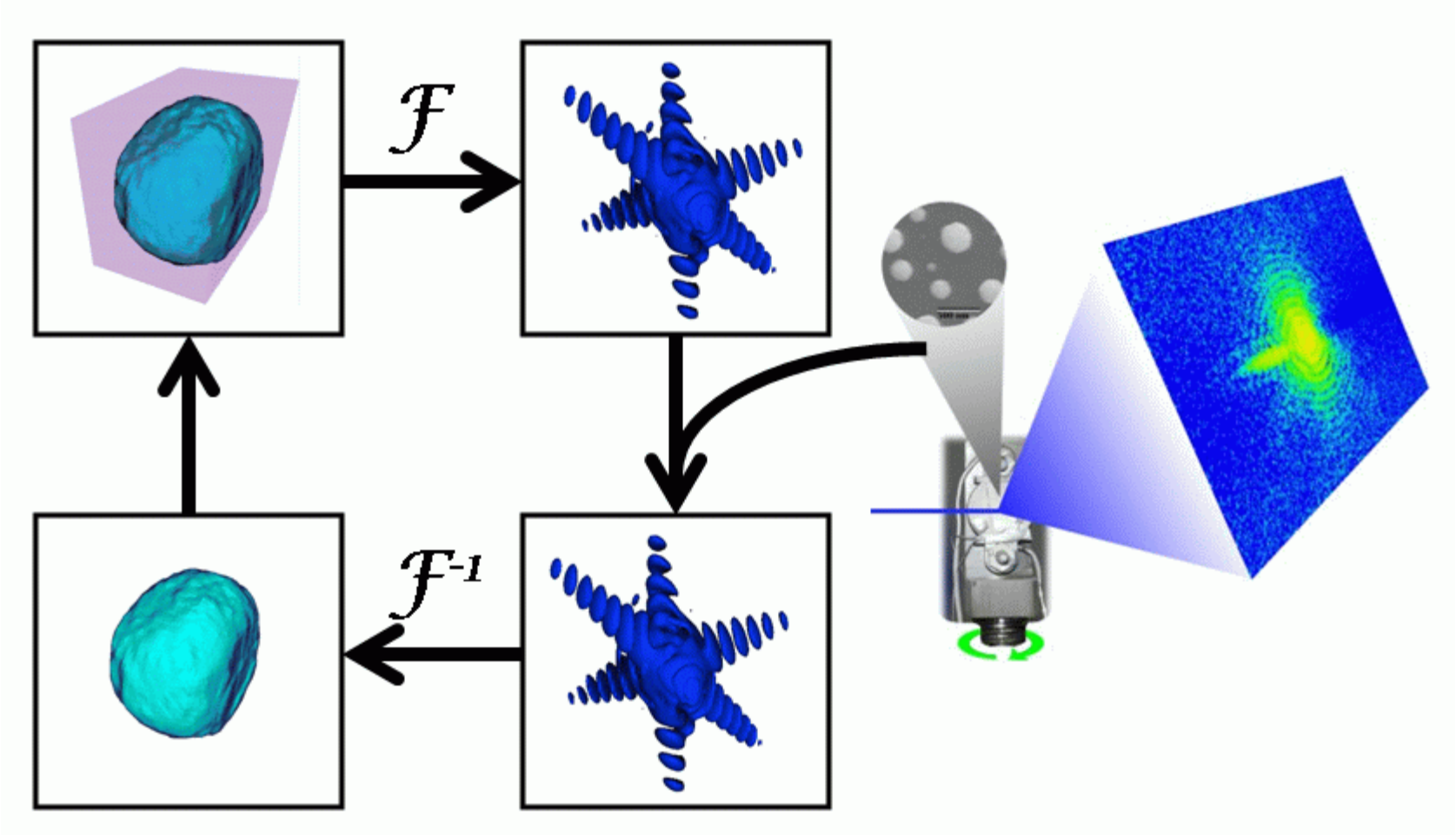
(11-1)

# Lensless X-ray Microscope, 2003




A
Detector Position
Q
$k_f$
$k_i$
l
h
k
B
ϕ=0°
ϕ=1°
ϕ=2°

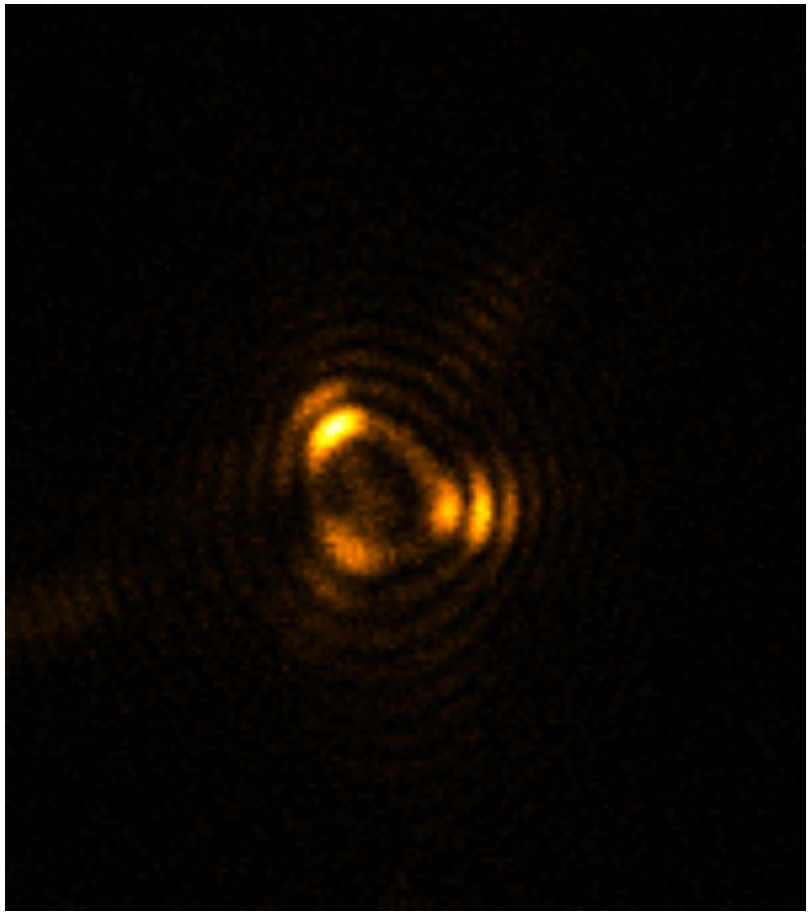
# Generic "Error Reduction" method

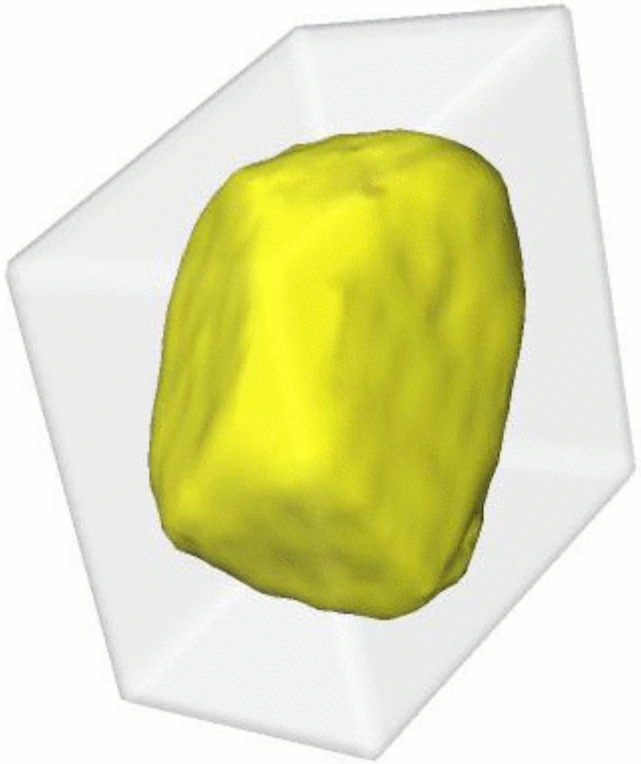



J. R. Fienup Appl. Opt. 21 2758 (1982)
R. W. Gerchberg and W. O. Saxton Optik 35 237 (1972)

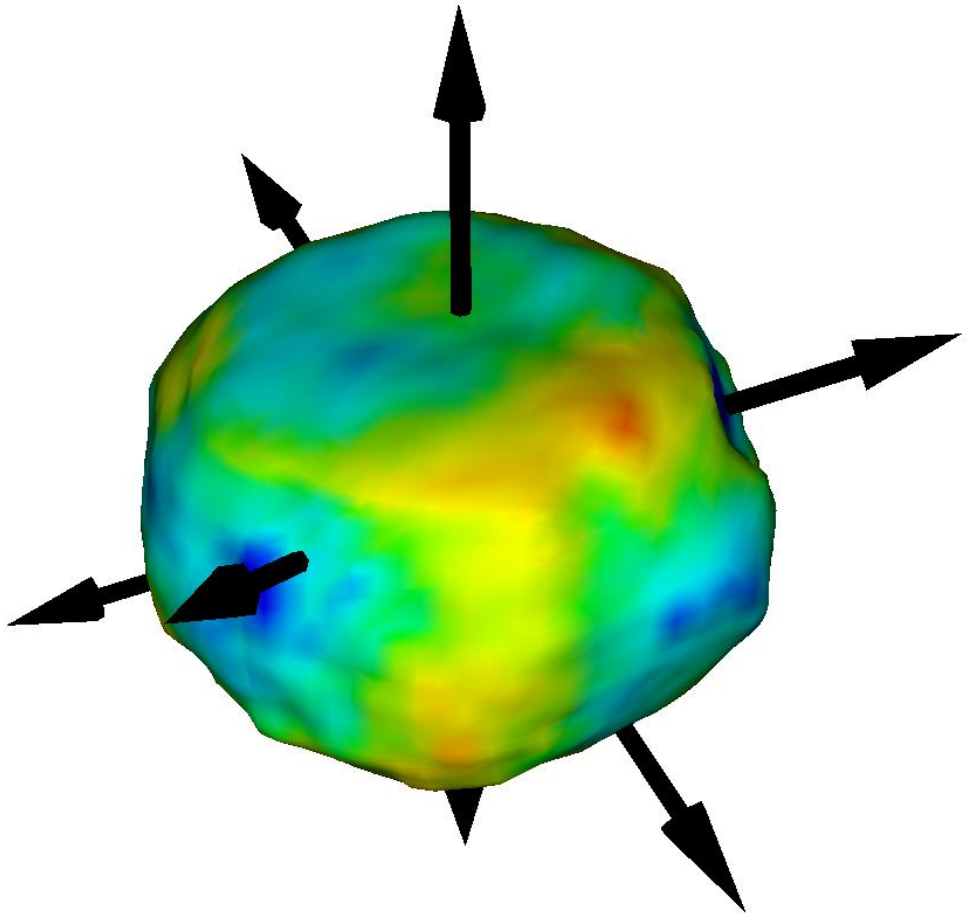
# Gold nanocrystal reconstruction

showing support used for 20 HIO followed by 10 ER



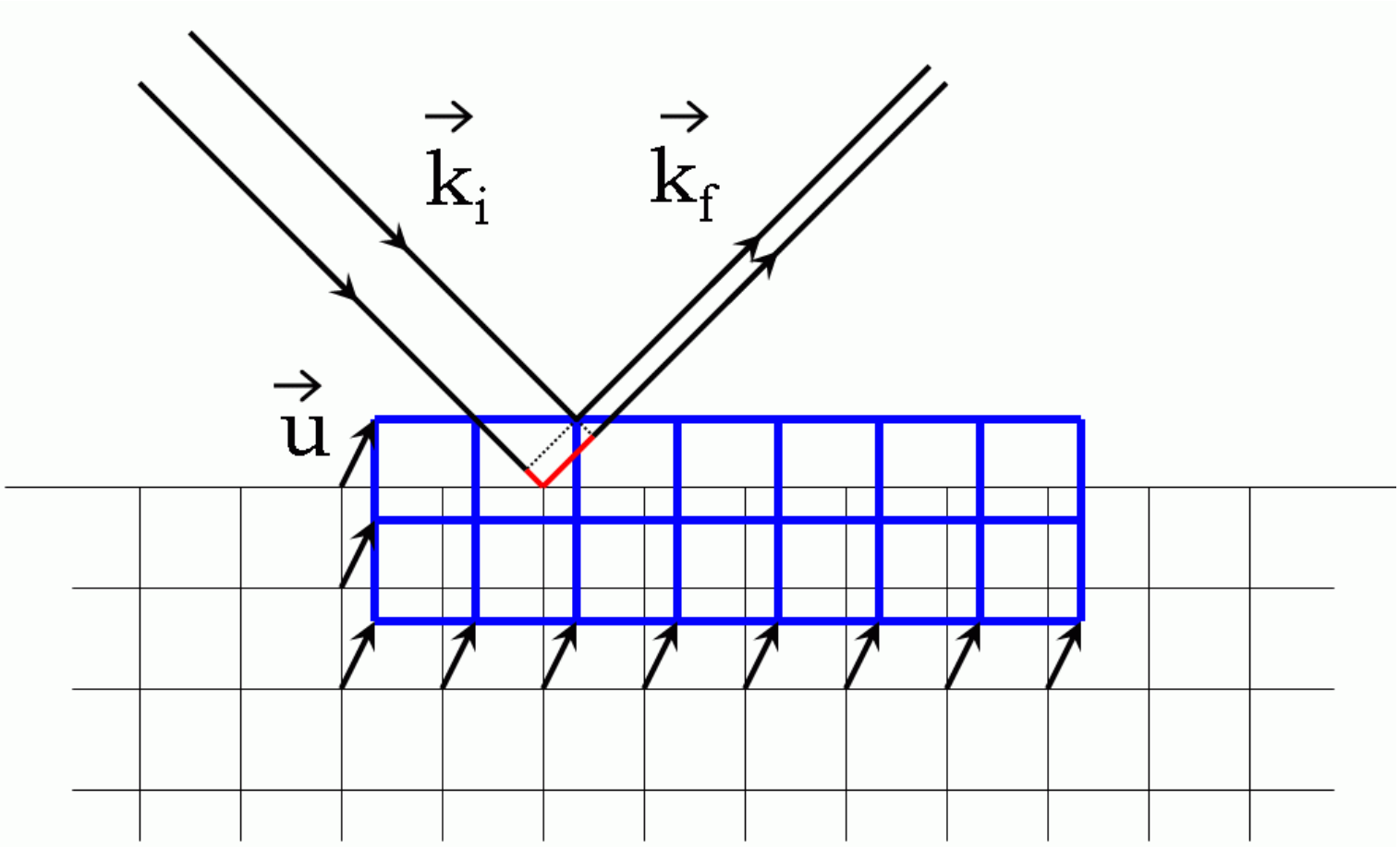


# Phase isosurface of residual strain



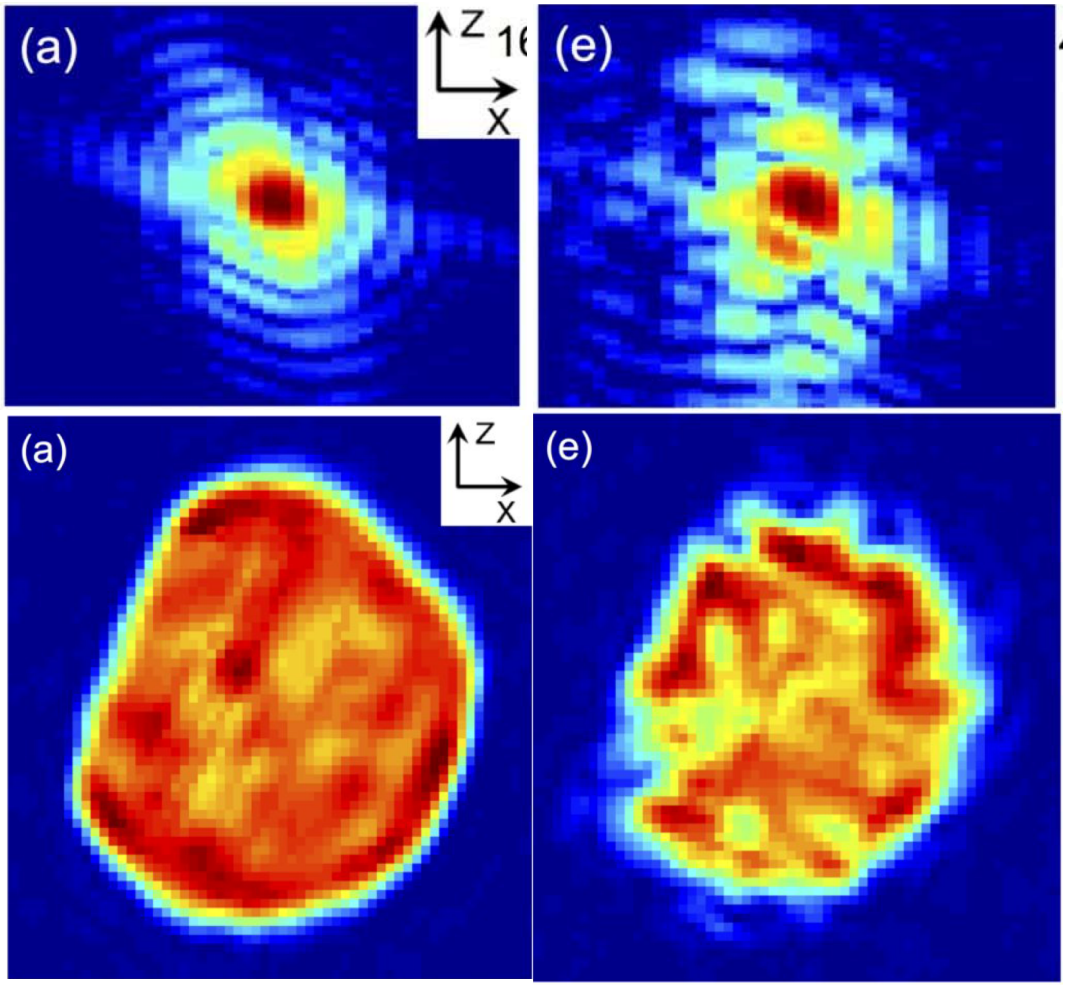
# Sensitivity to strain

$$\Delta\varphi = \mathbf{k}_f \cdot \mathbf{u} - \mathbf{k}_i \cdot \mathbf{u} = \mathbf{Q} \cdot \mathbf{u}$$



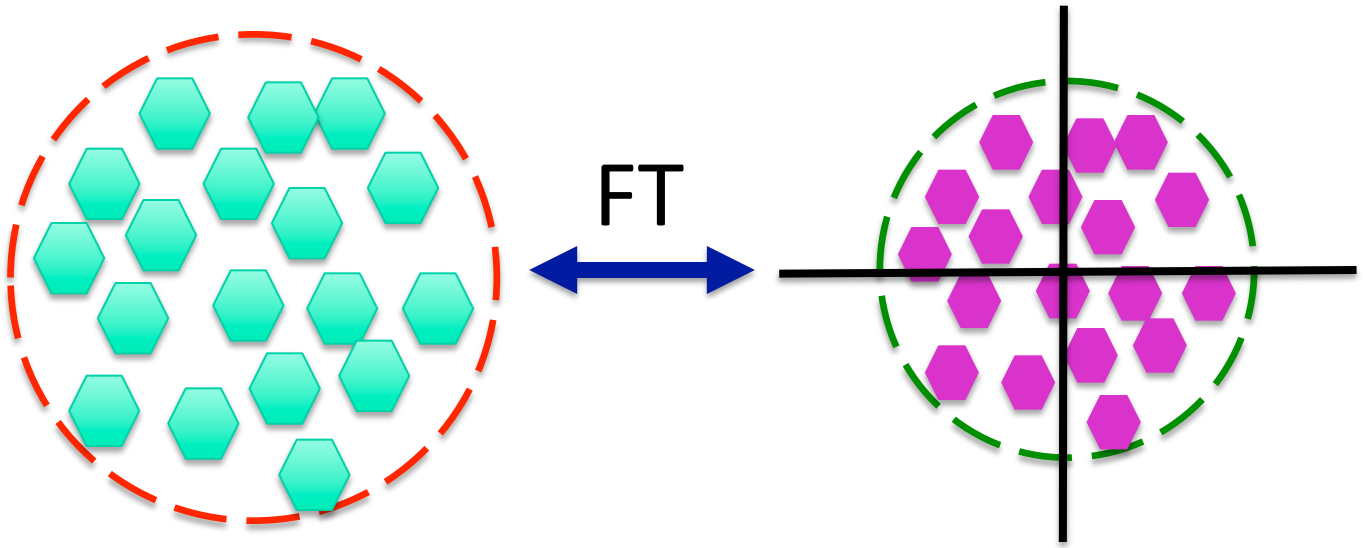
# Copper Diffusion into Gold Nanocrystal

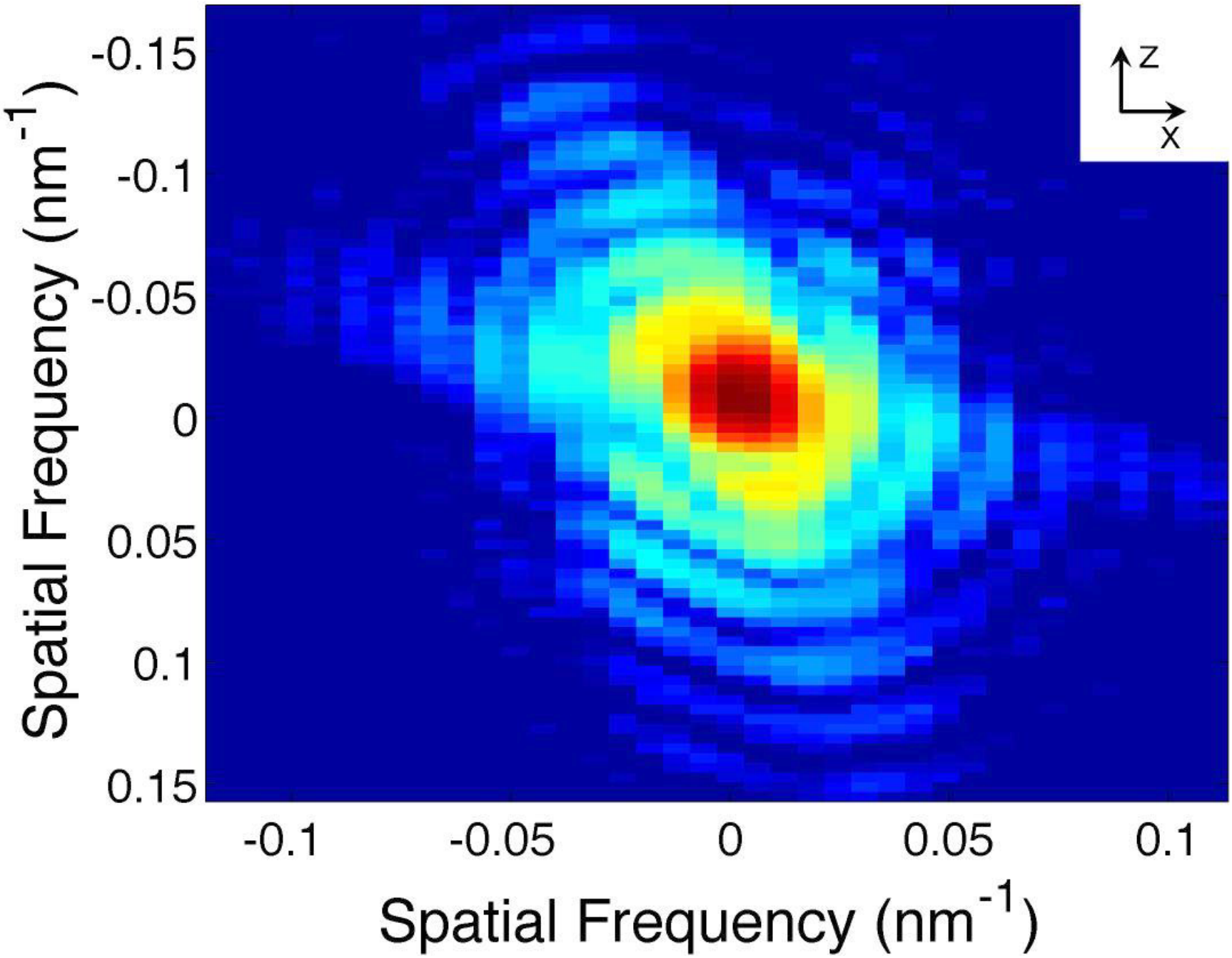
## Gang Xiong, et al, Sci Rep 4 6765 (2013)

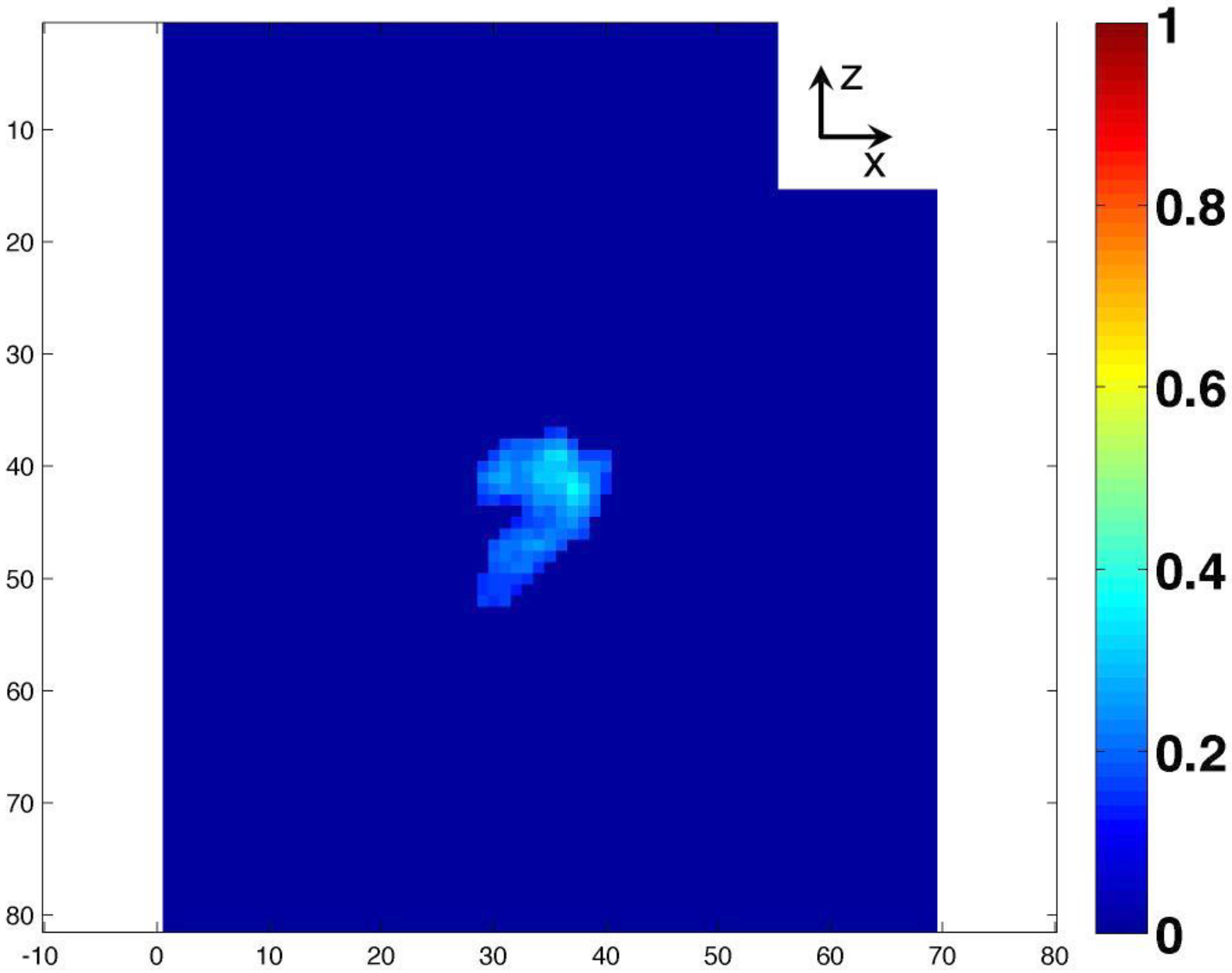


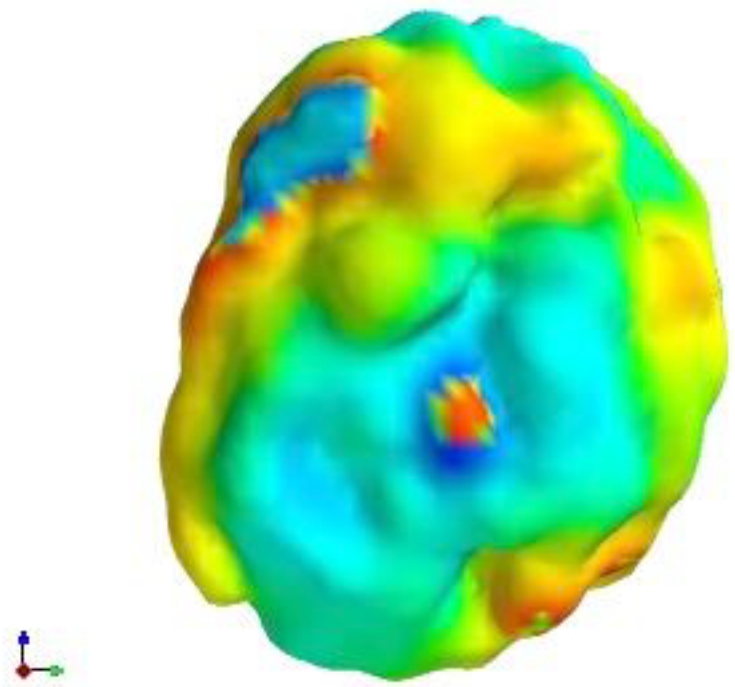
# Domain structures give speckled diffraction

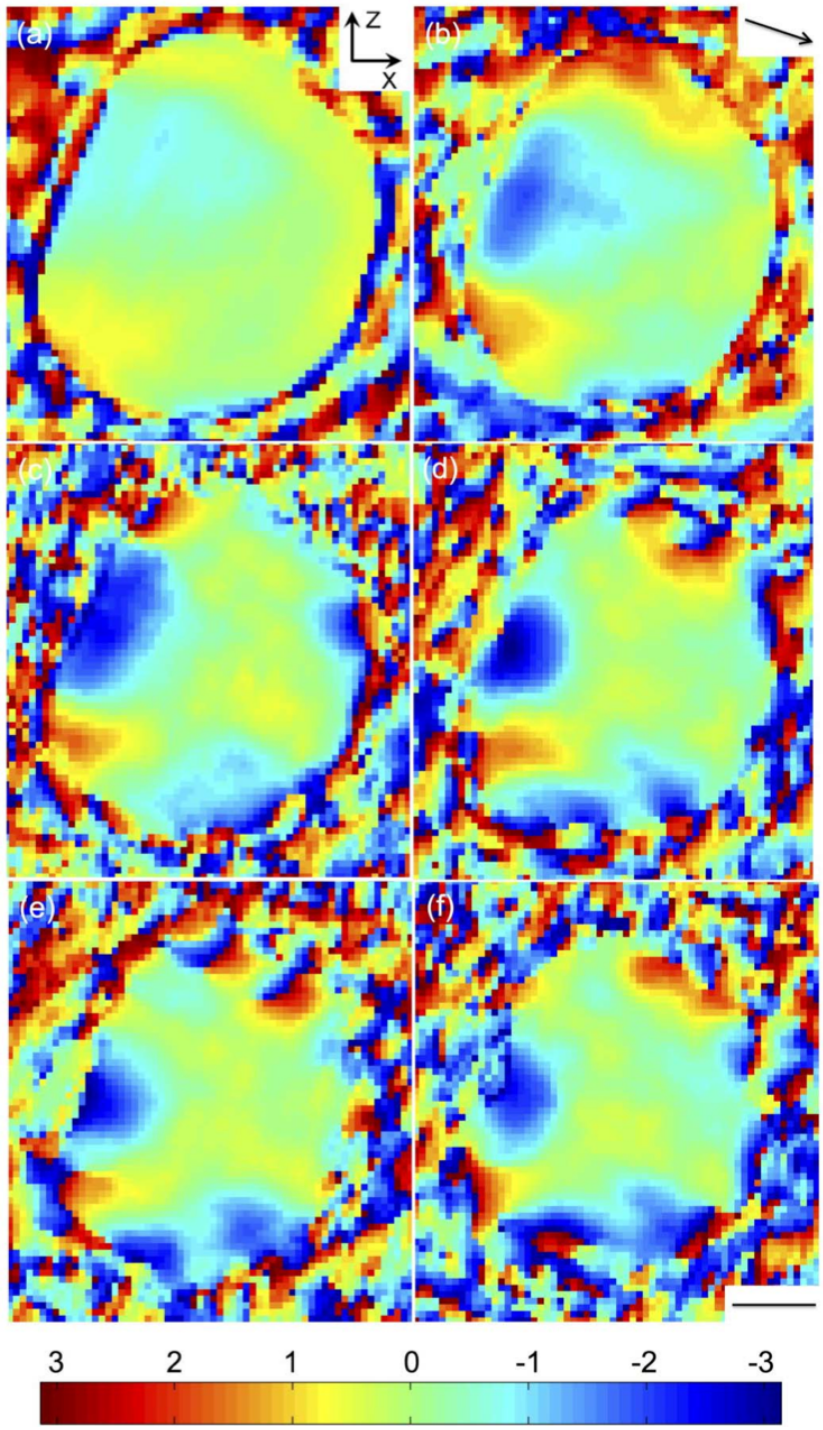
Real Space Reciprocal Space

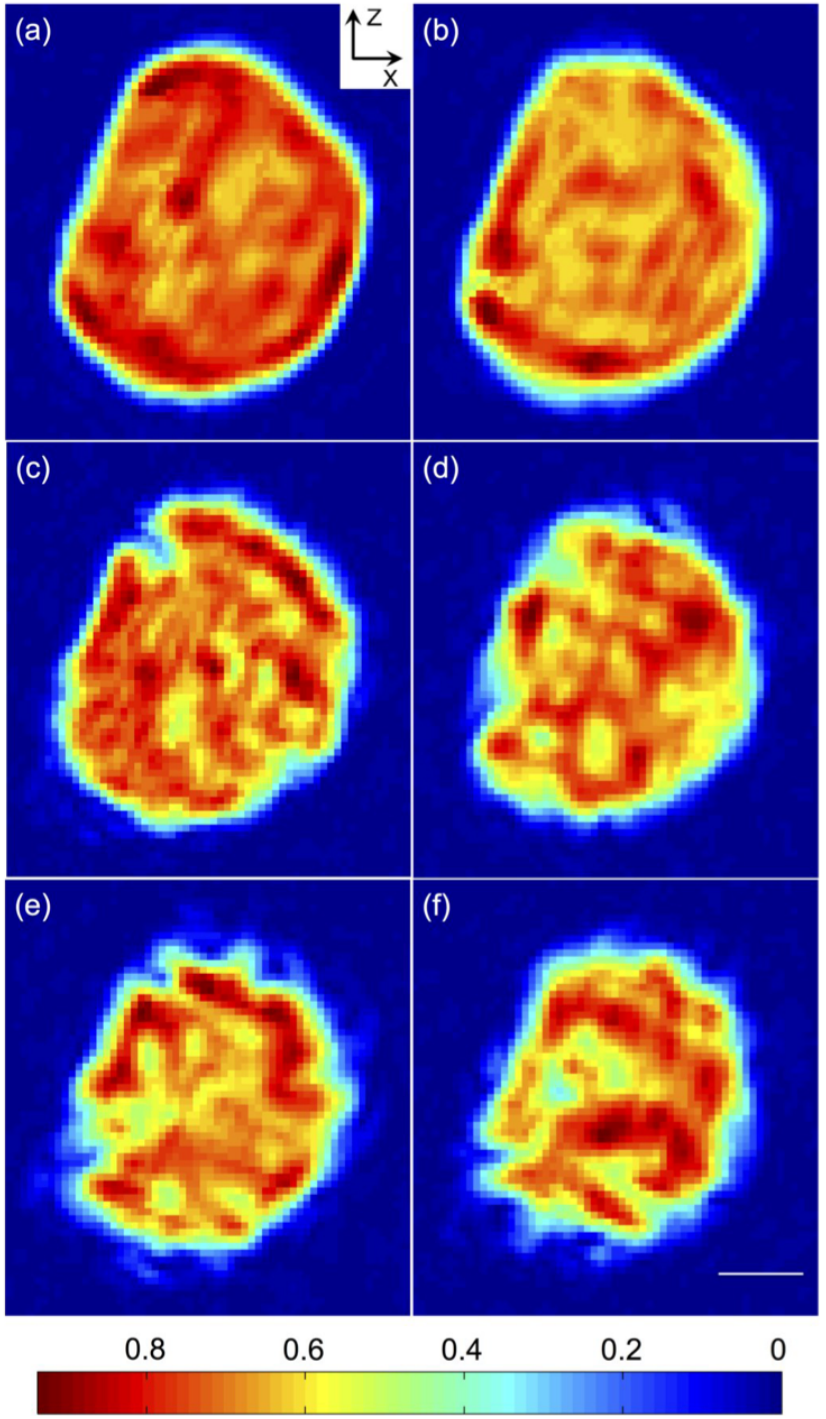



-0.15
-0.1
-0.05
0
0.05
0.1
0.15
Spatial Frequency (nm-1)
-0.1
-0.05
0
0.05
0.1
Spatial Frequency (nm-1)
z
x


Z
X
10
20
30
40
50
60
70
80
-10
0
10
20
30
40
50
60
70
80
1
0.8
0.6
0.4
0.2
0



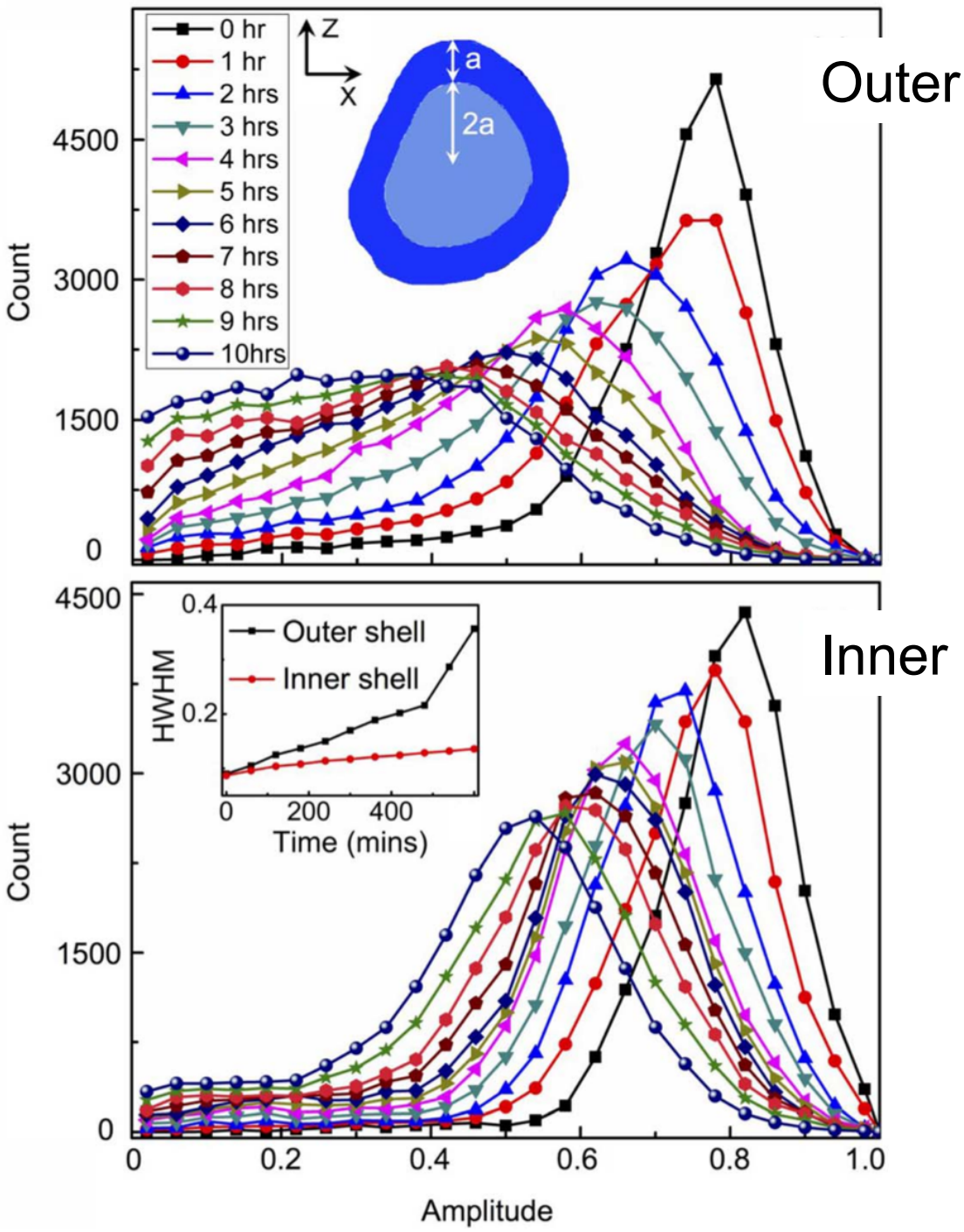

(a)
Z
X
(b)
(c)
(d)
(e)
(f)
3
2
1
0
-1
-2
-3


(a)
Z
X
(b)
(c)
(d)
(e)
(f)
0.8
0.6
0.4
0.2
0

# Atomic Diffusion within Individual Gold Nanocrystal

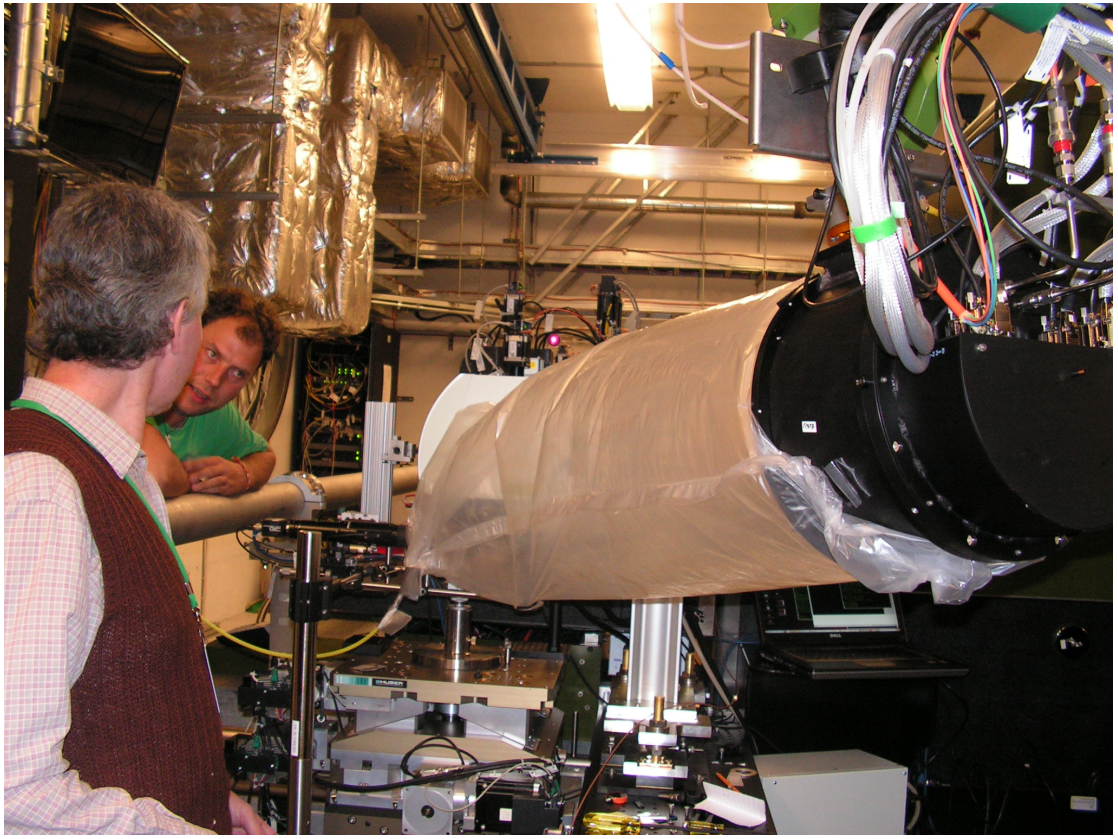
Gang Xiong,
J. N. Clark,
C. Nicklin,
J. Rawle &
I. K. Robinson

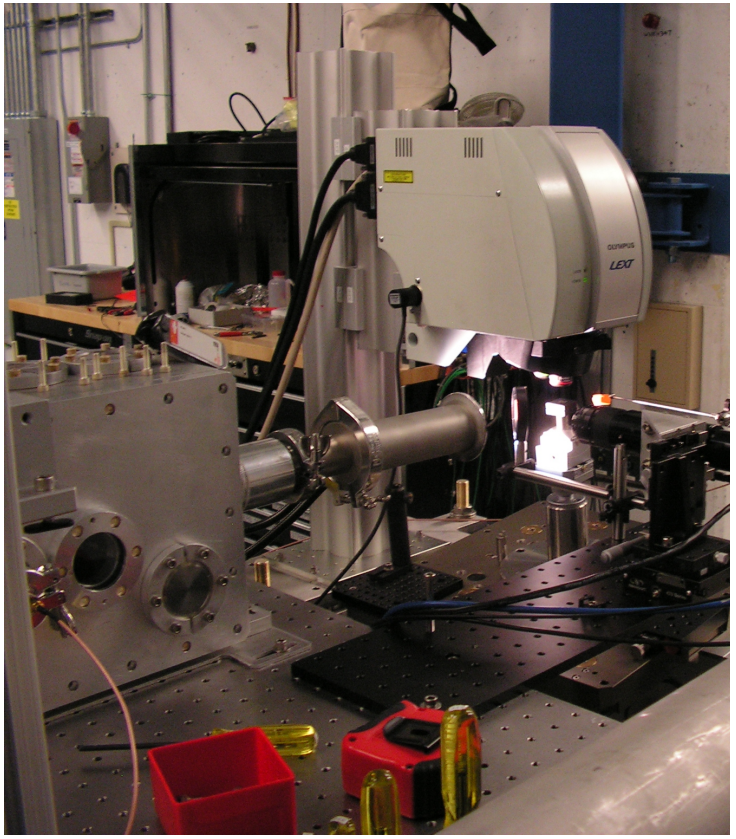
Sci Rep **4** 6765
(2013)



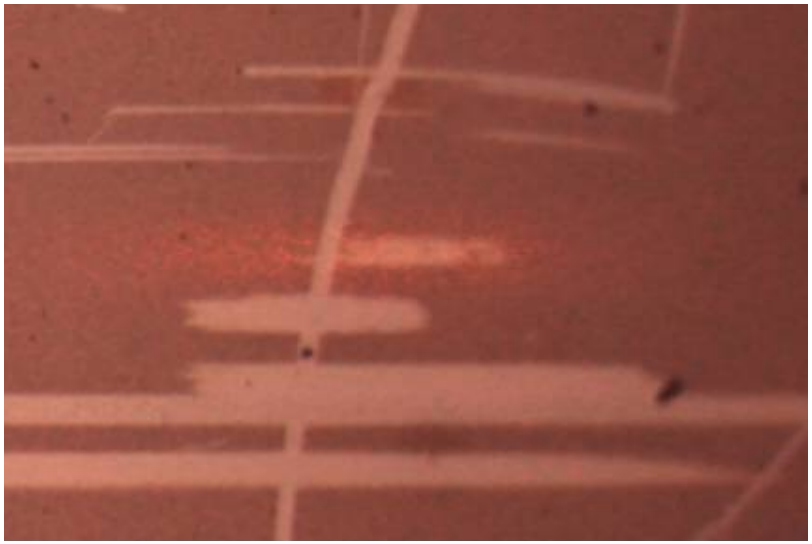
# Pump-probe at LCLS (XPP)

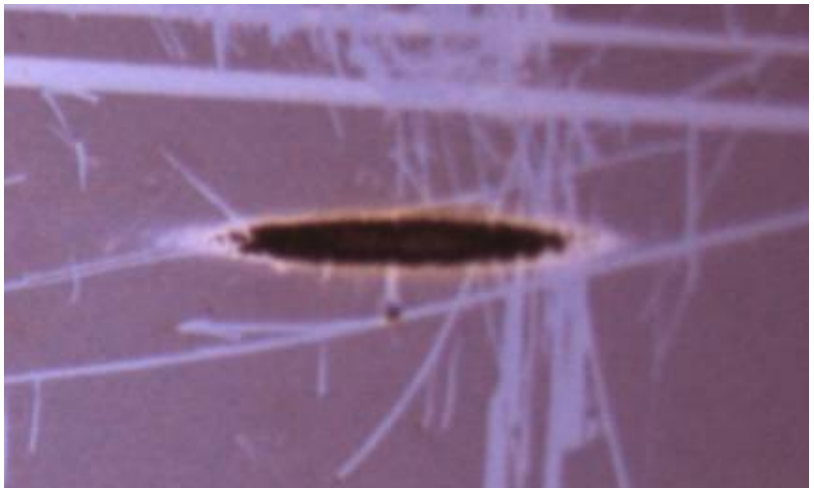
Justin Wark, Loren Beitra, Alexander Korsunsky, Ross Harder, David Fritz ,
Sebastien Boutet, Jesse Clark, Garth Williams, Brian Abbey, Andy Higginbotham,
Diling Zhu, Henrick Lemke, Mattieu Chollet, Marc Messerschmidt

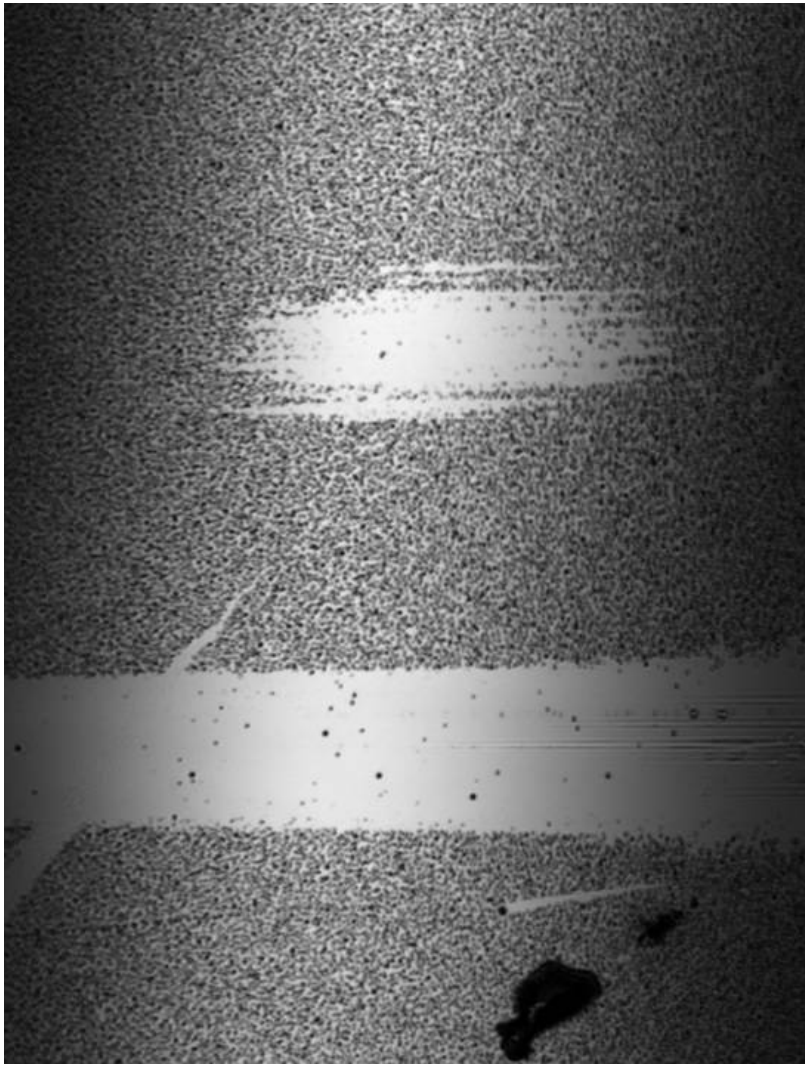




# Visible and Confocal microscopy

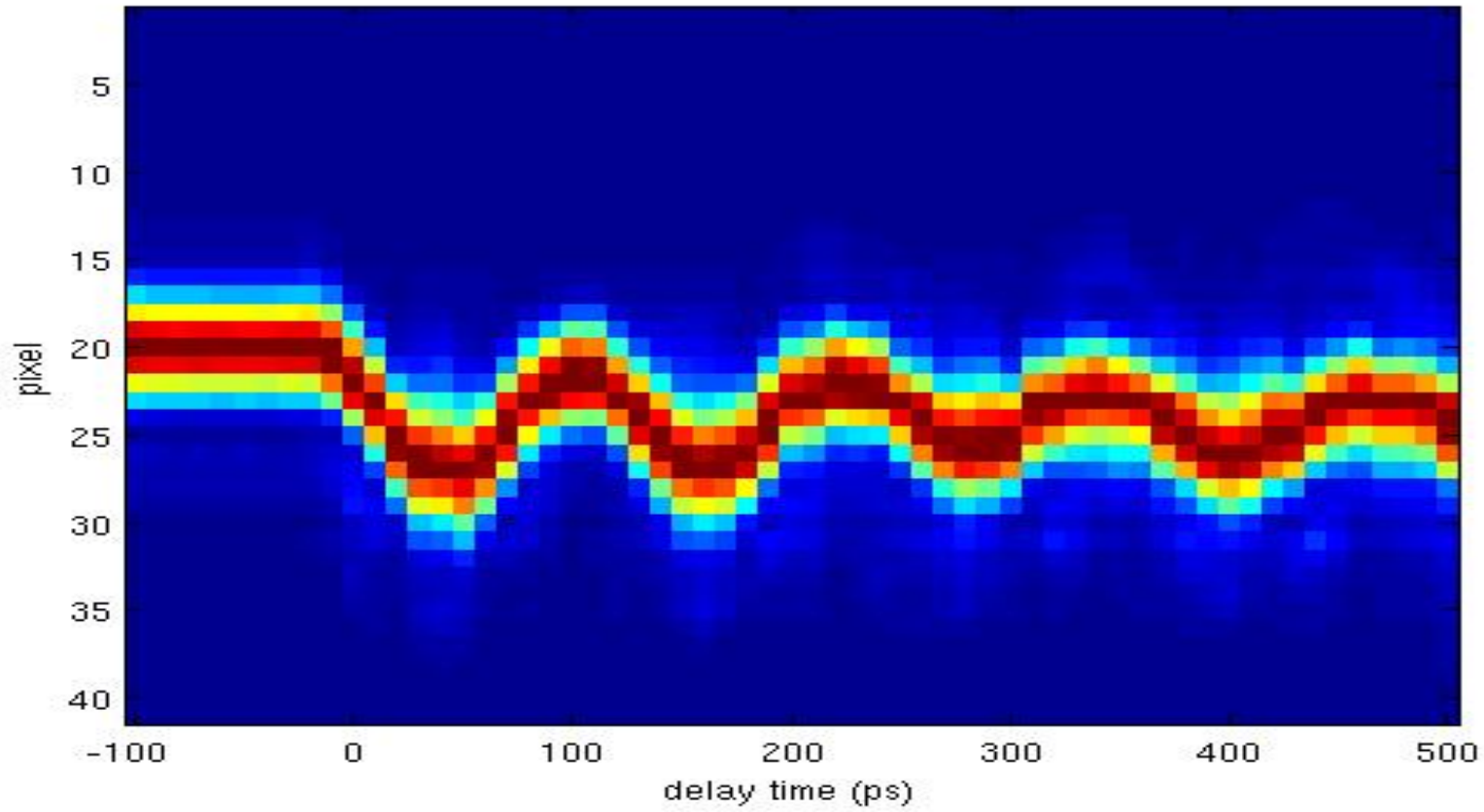






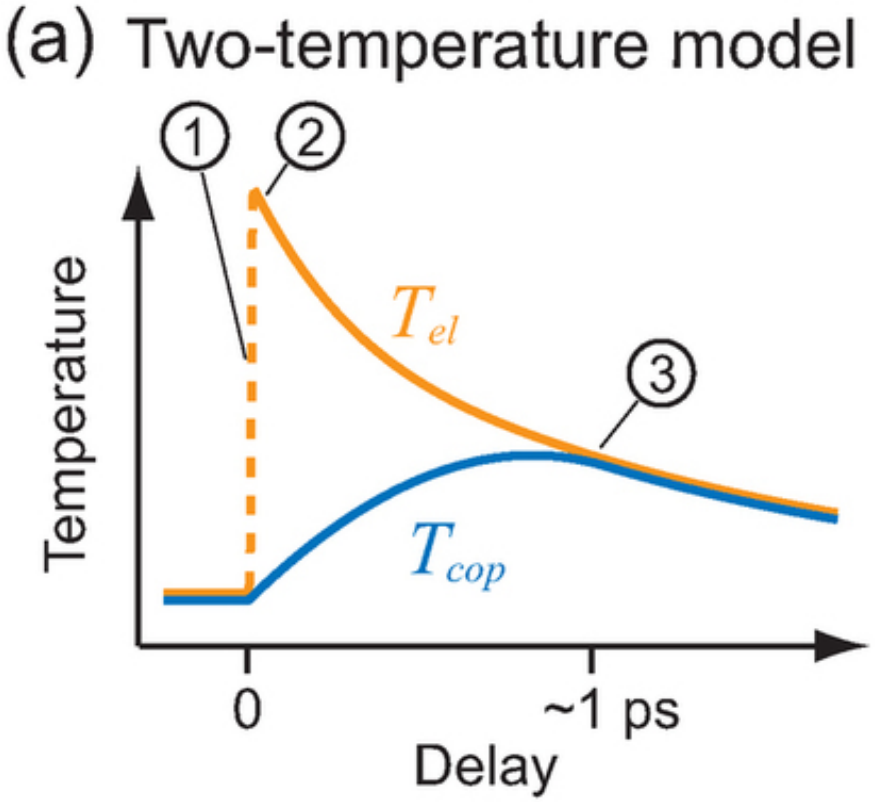
# Pump-probe at LCLS (XPP)

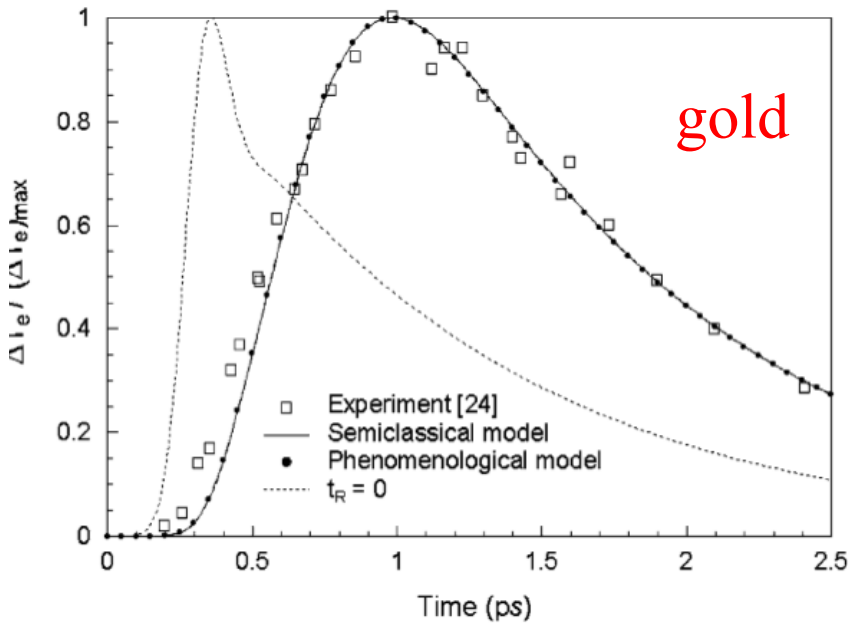
Justin Wark, Loren Beitra, Alexander Korsunsky, Ross Harder, David Fritz ,
Sebastien Boutet, Jesse Clark, Garth Williams, Brian Abbey, Andy Higginbotham,
Diling Zhu, Henrick Lemke, Mattieu Chollet, Marc Messerschmidt



# "Two-temperature" model

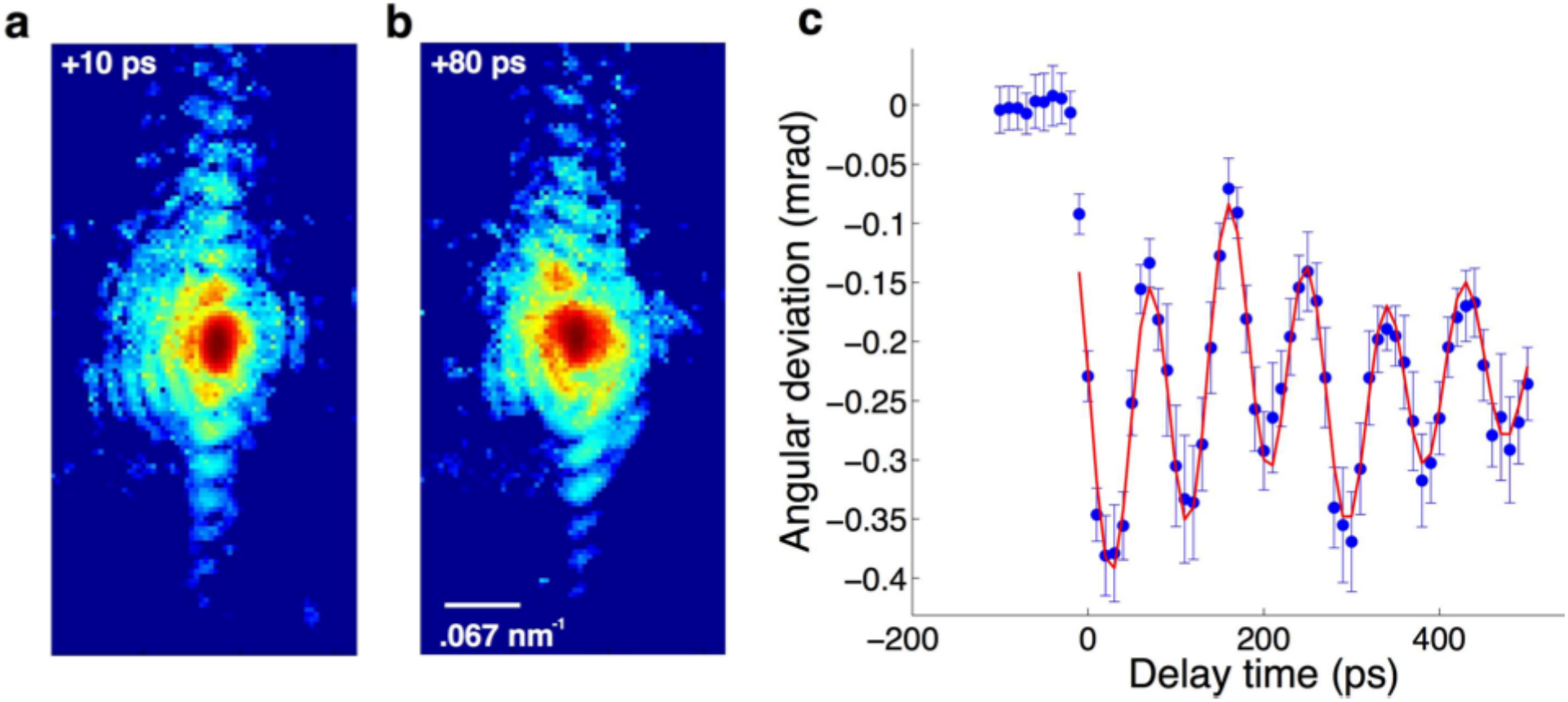
Y. Ishida et al, Nature Scientific Reports 1 64 (2011)
J.K. Chen et al, Int J. Heat Transfer 49 307 (2006)



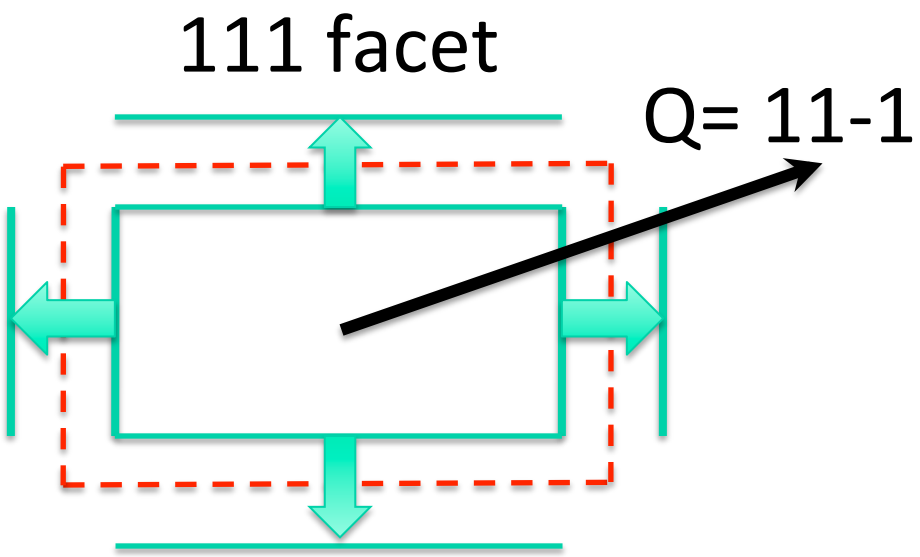

ig. 2. Comparison of the change in electron temperature at
ıe front surface of an 80-nm gold film irradiated by a 2.8 mJ/
$n^2$, 800 nm, 150-fs laser pulse.

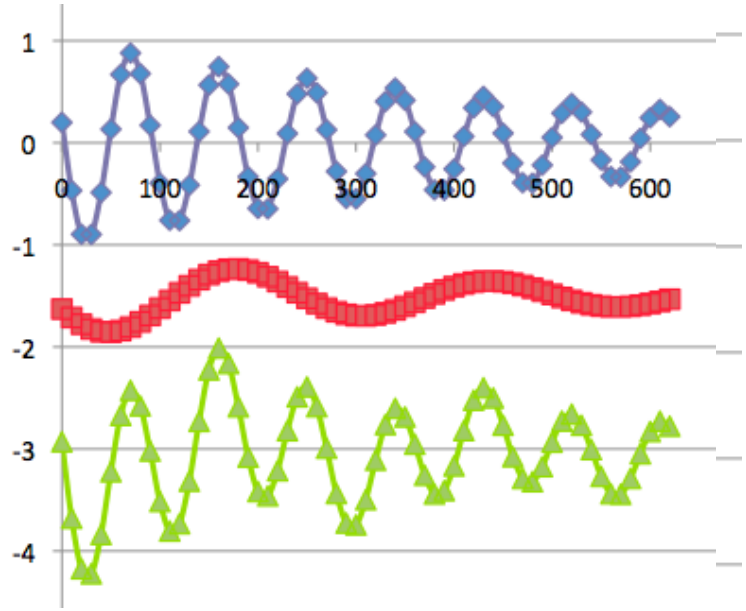
# Time resolved Bragg peak position



# Two Normal Modes of Vibration

$$S(\tau) = \sum_{n=1}^{N} A_n \exp\left[-(\tau/\tau_{d,n})^2\right] \cos(\omega_n \tau + \varphi_{0,n})$$




$T_1$ = 90ps $h_1$ = 145nm $c_s$ = 3240 m/s

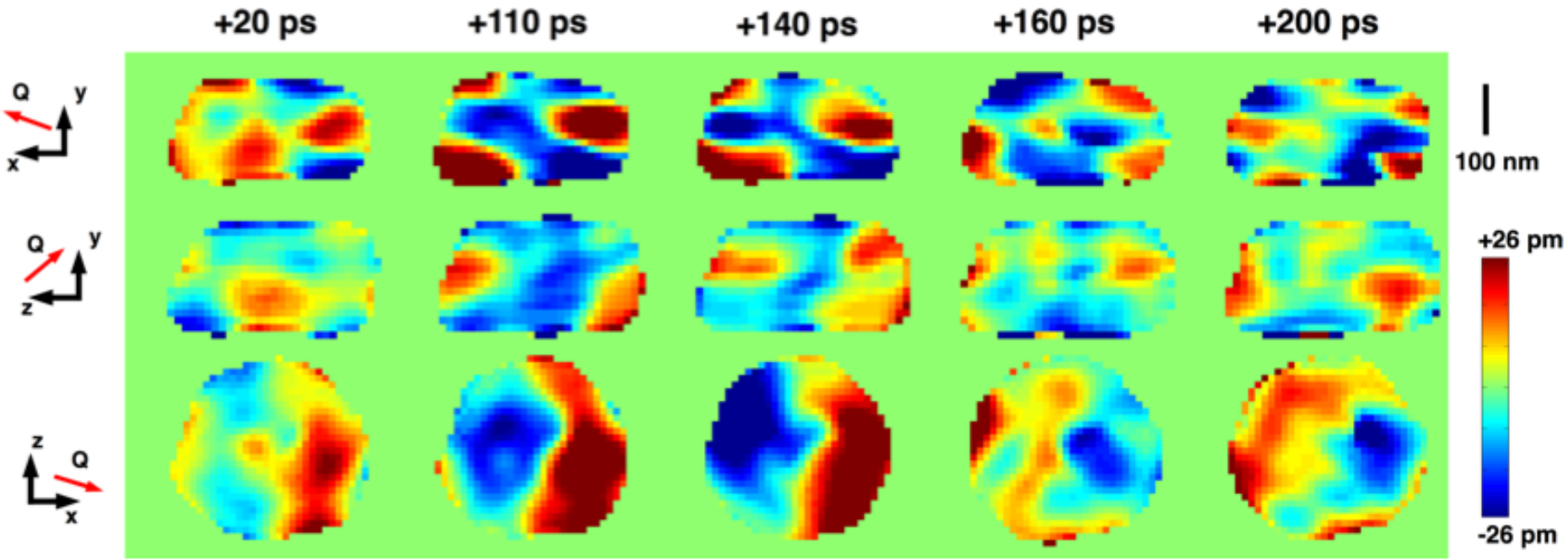
$T_2$ = 259ps $h_2$ = 420nm

# Dynamic imaging of displacements

## CDI inversion of 3D diffraction patterns

## 1000 frames averaged at each point of rocking curve

## Jesse Clark et al, Science 341 56 (2013)

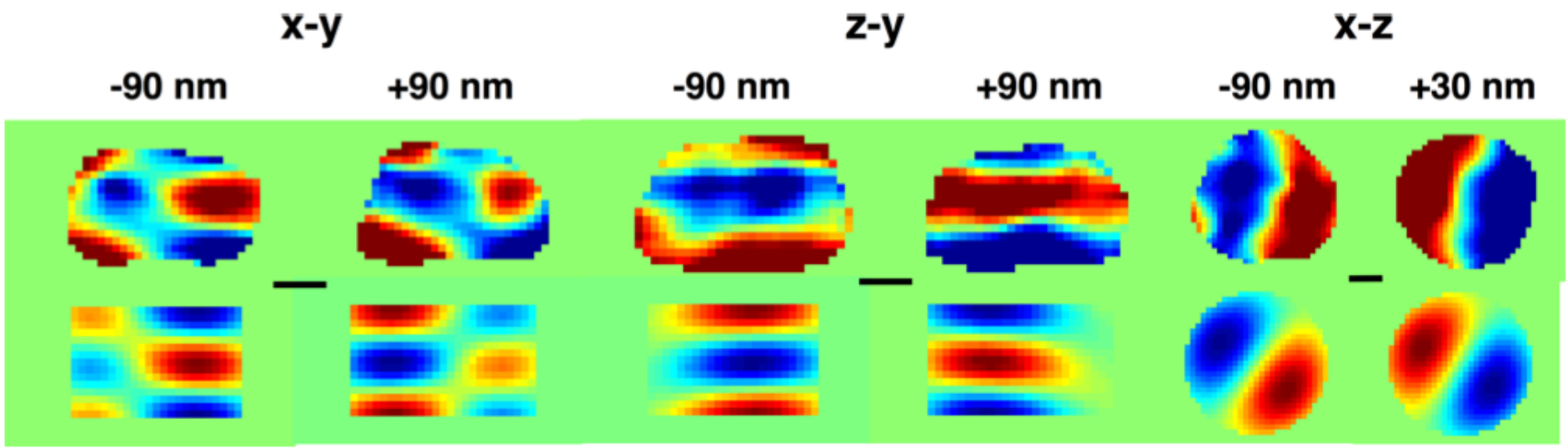


# Dynamic imaging of displacements

## CDI inversion of 3D diffraction patterns

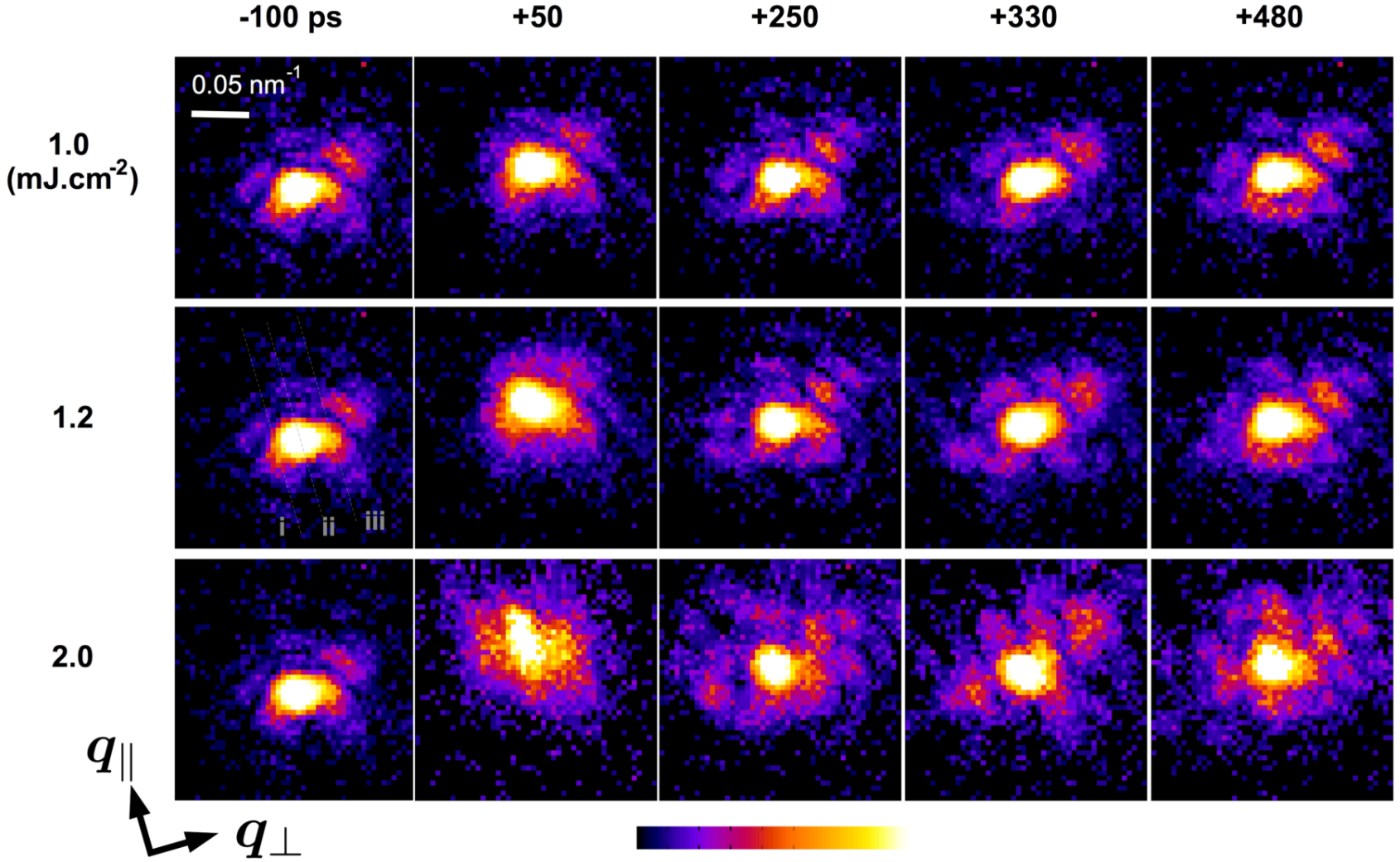
## Comparison with (1,1) normal mode of cylinder

## Jesse Clark et al, Science 341 56 (2013)



# Dependence on Laser Fluence

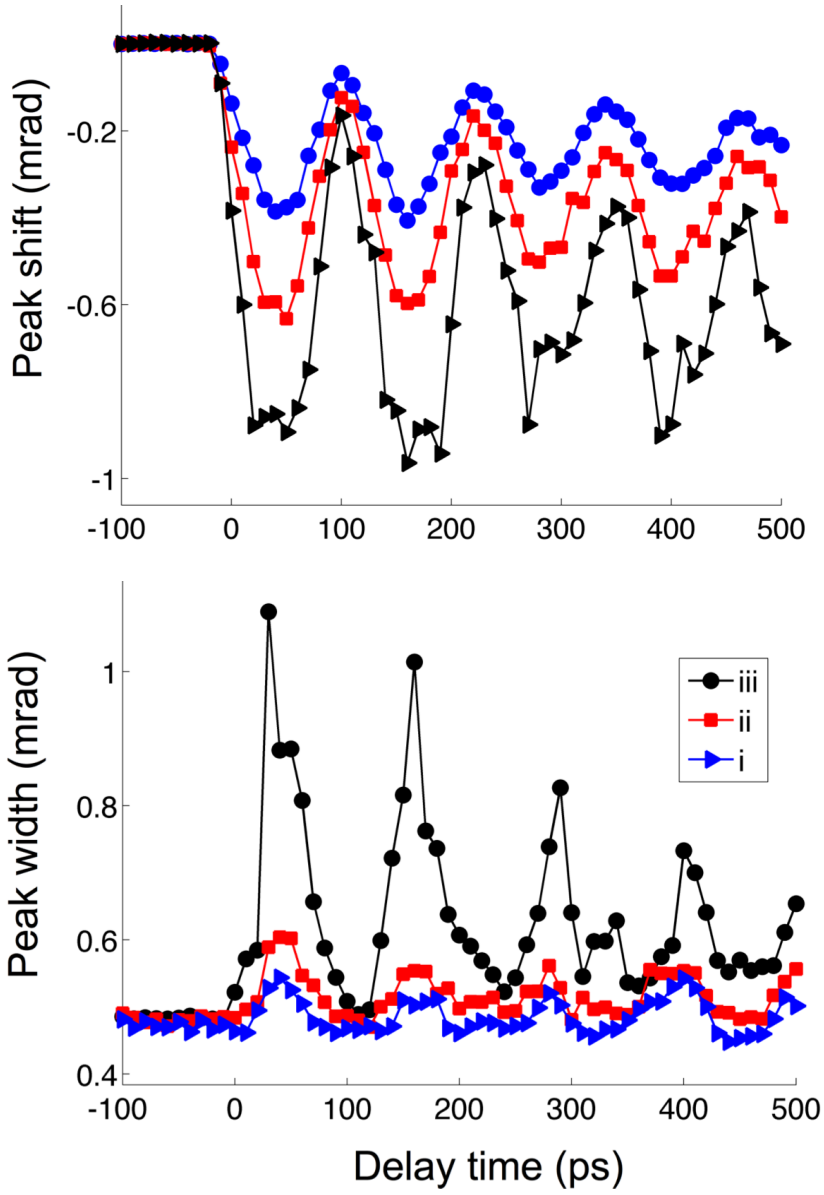
## Jesse Clark et al, to be published



# Dependence on Laser Fluence

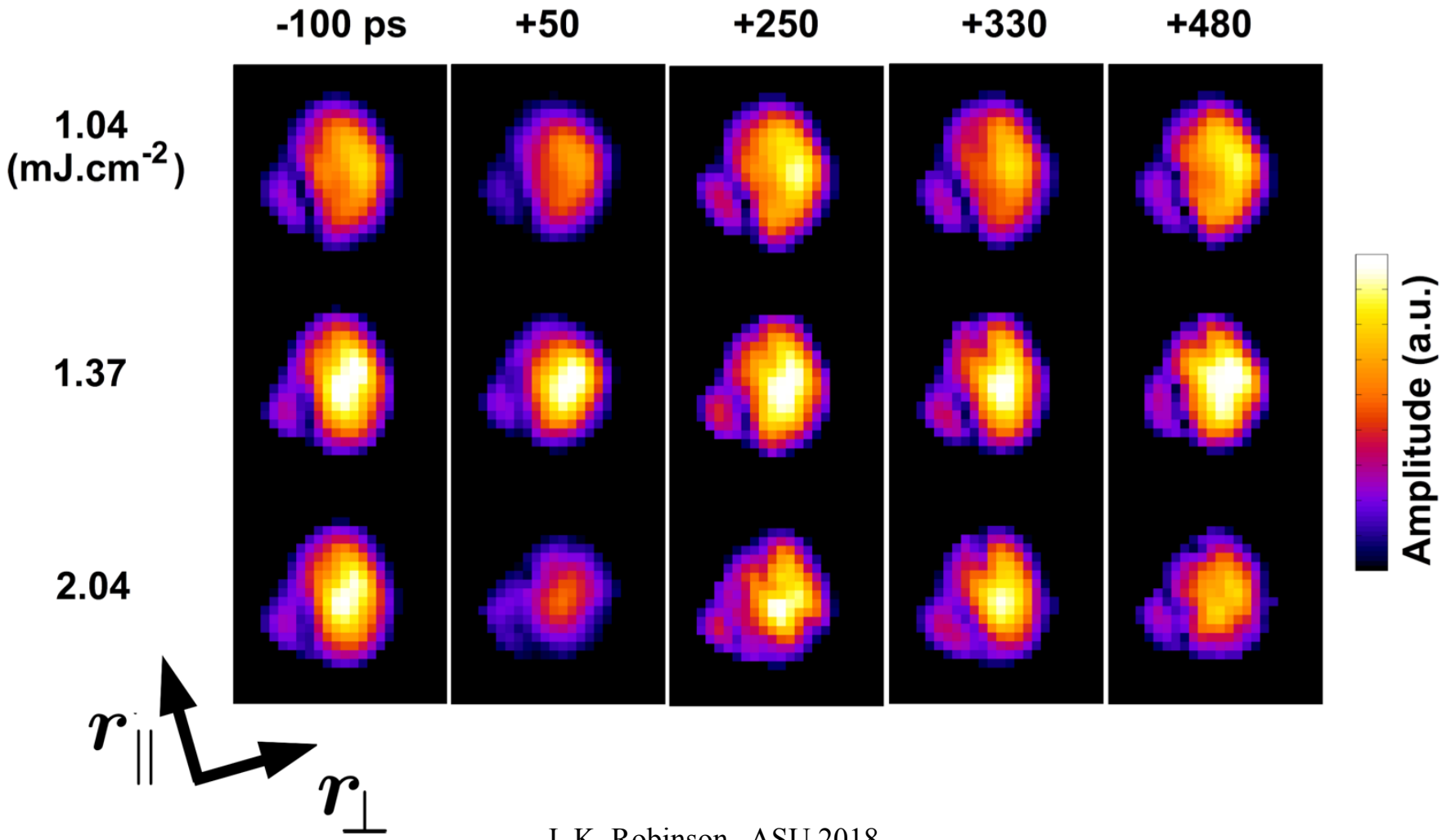
Jesse Clark et al, to be published

1.0 mJ cm$^{-2}$
1.2 mJ cm$^{-2}$
2.0 mJ cm$^{-2}$

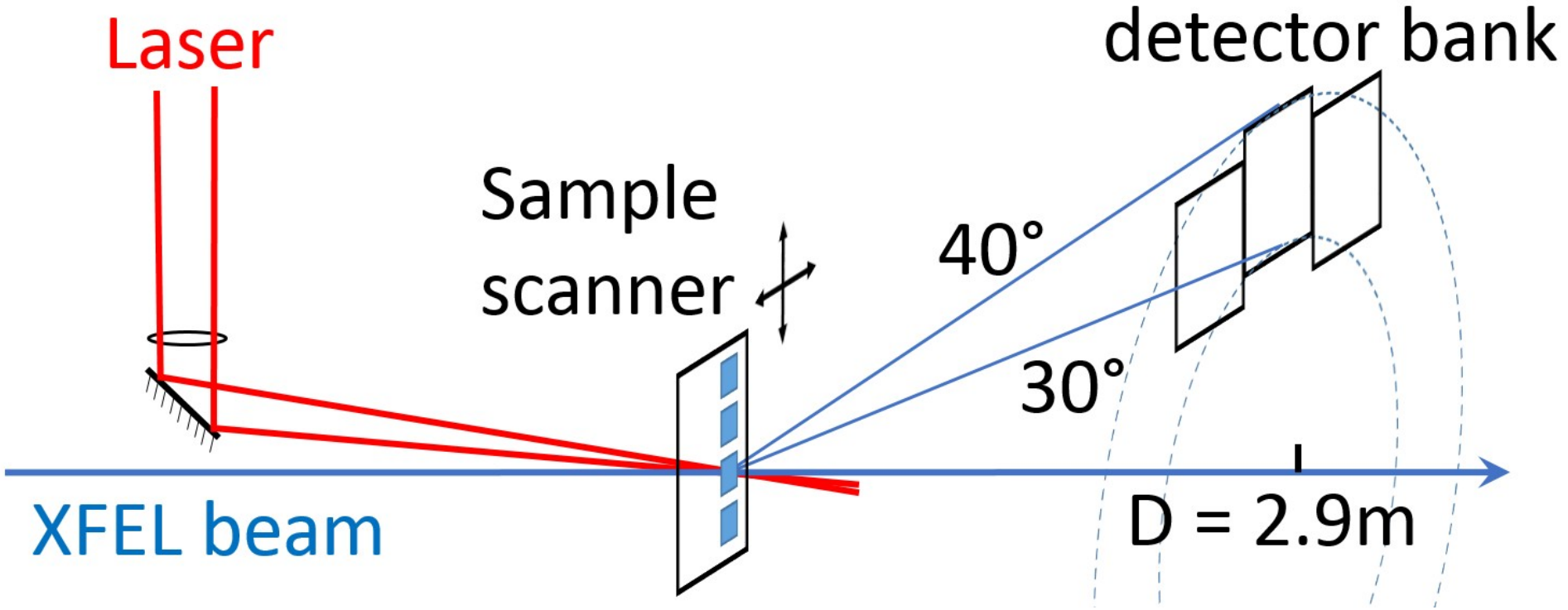


# Dependence on Laser Fluence

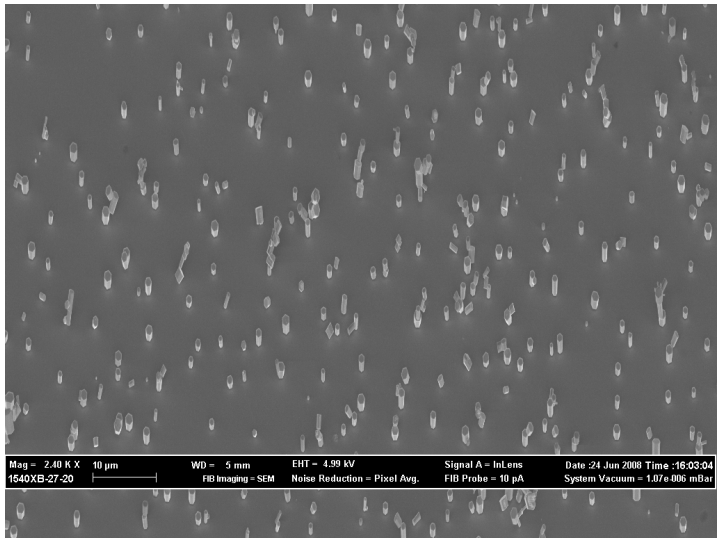
## Jesse Clark et al, to be published

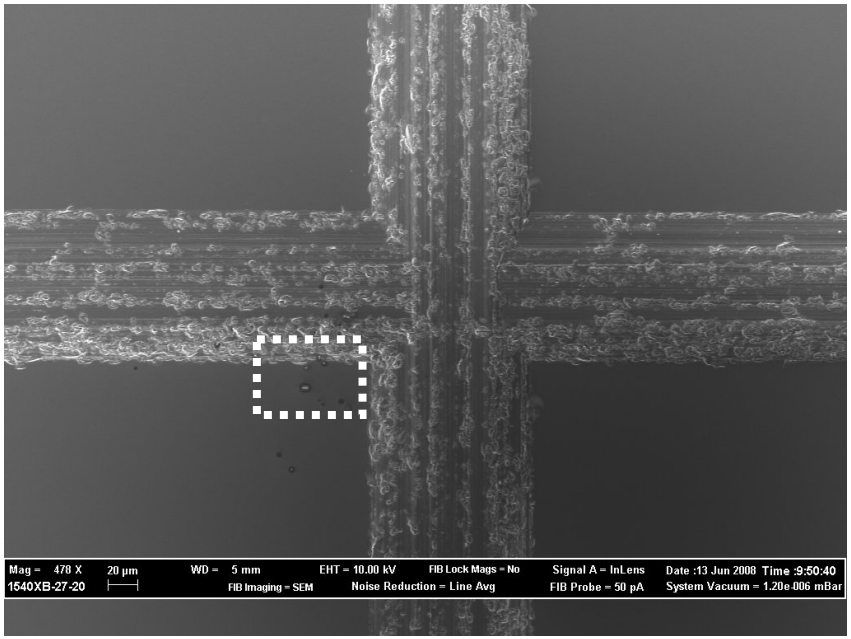


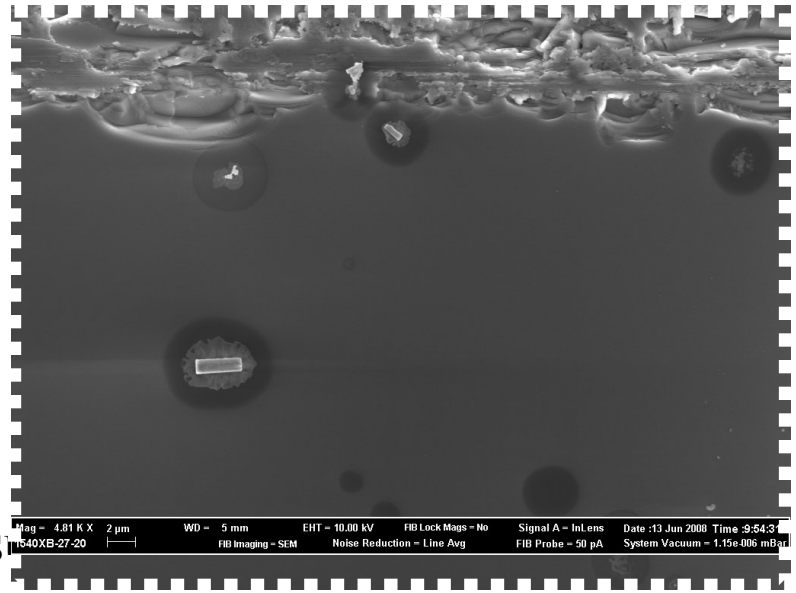
# Ultrafast Materials Physics
# Fixed Geometry Setup



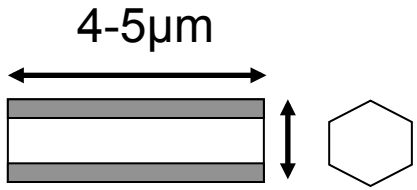
# ZnO Sample Preparation



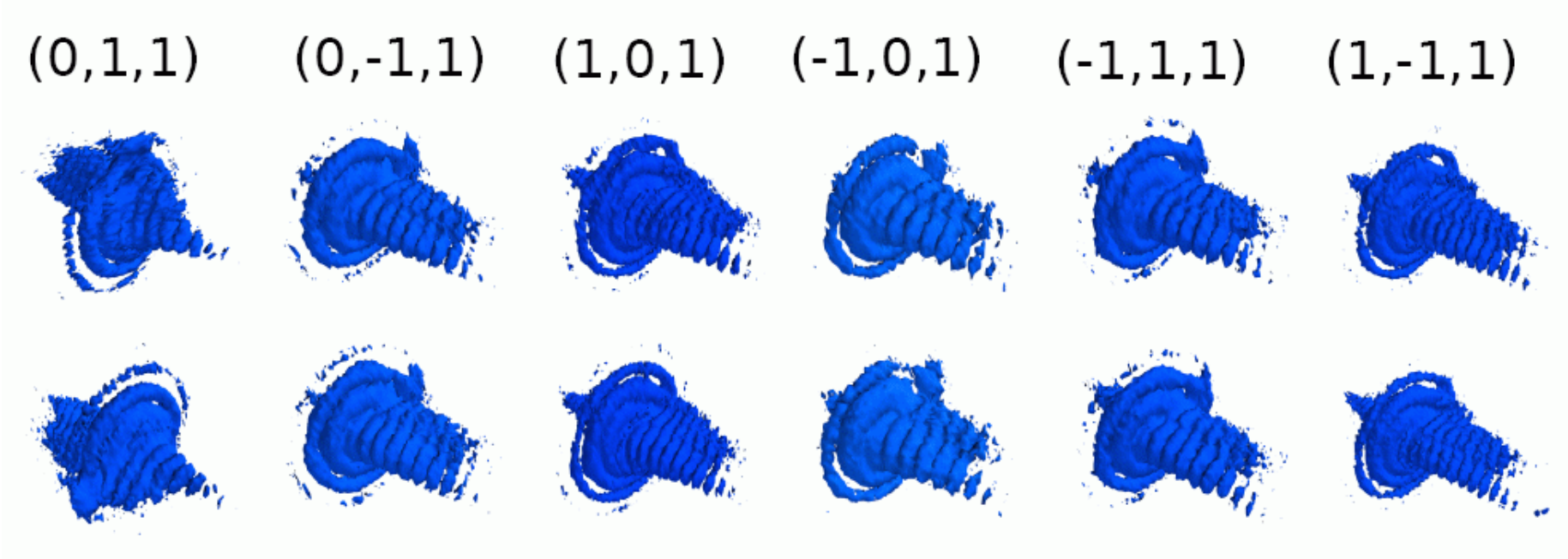




Dimensions:

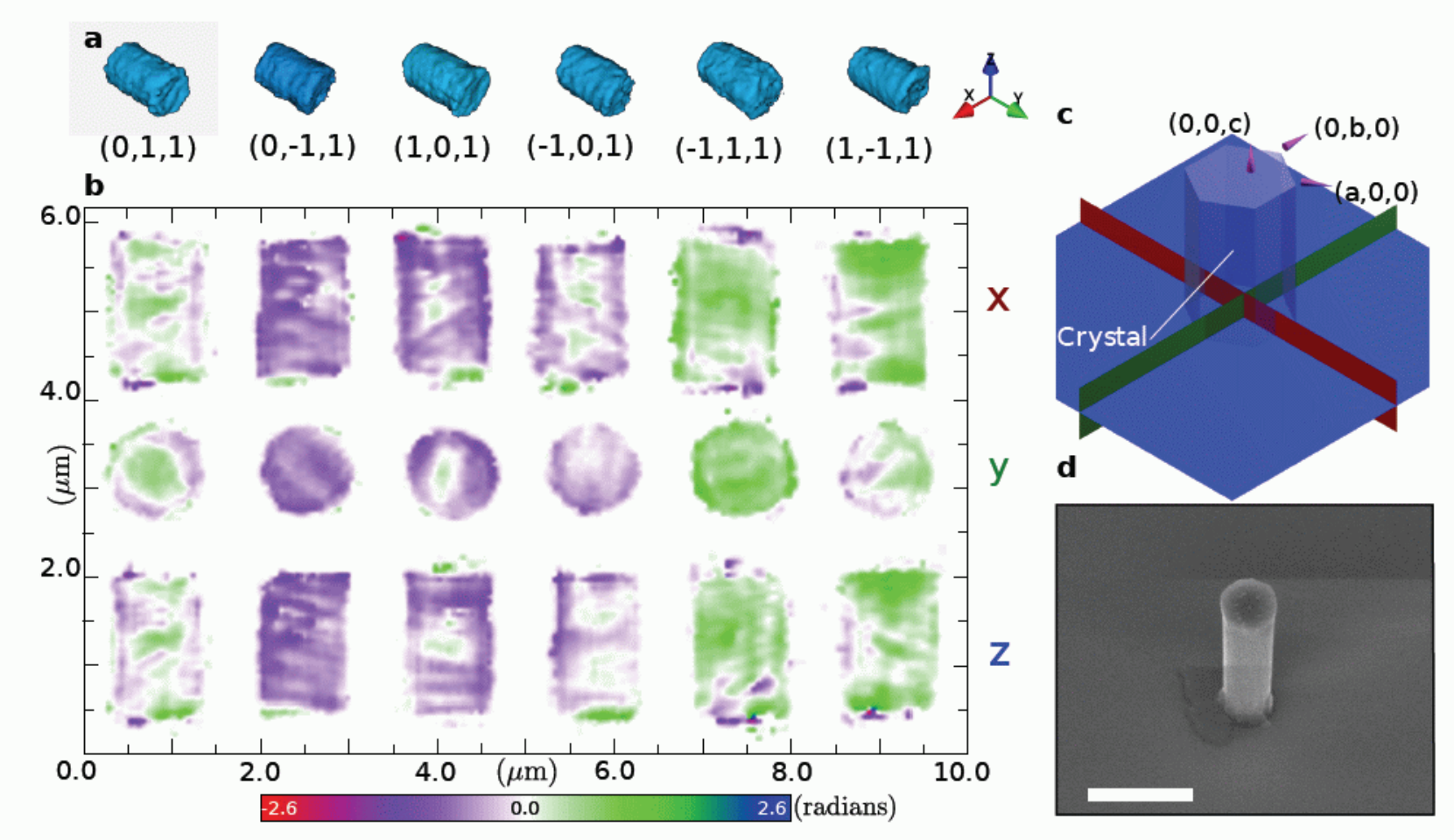



I. K. Robinson, AS

# Extension to 6 Bragg Peaks

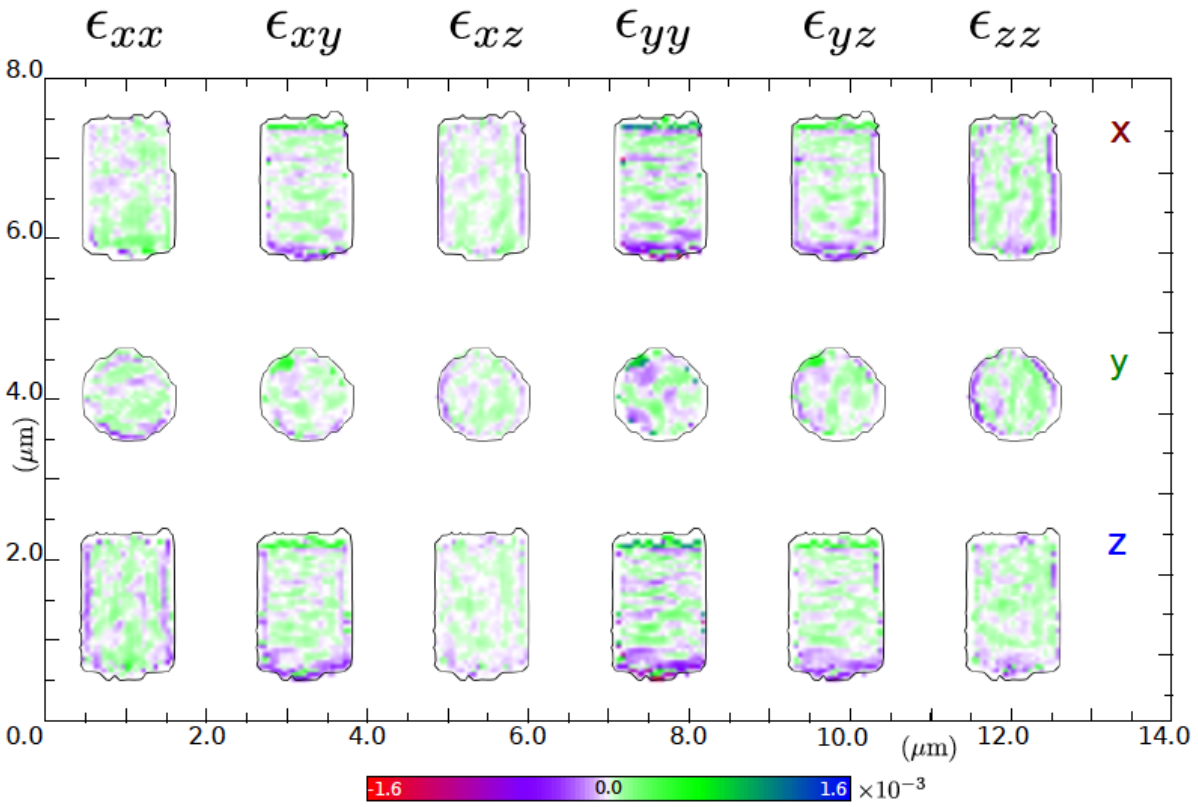


# Extension to 6 Bragg Peaks

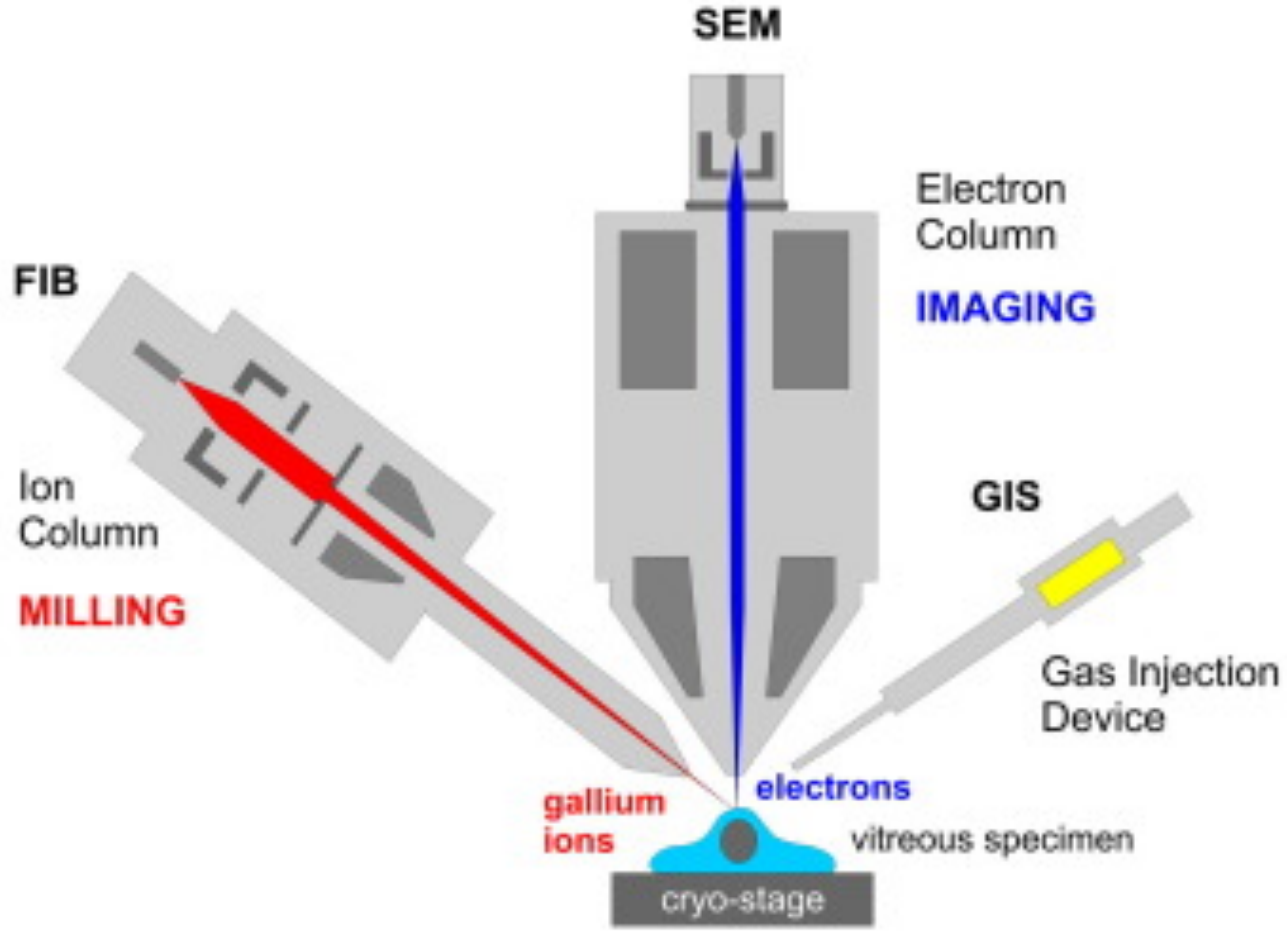


# Full Strain Tensor

$$\epsilon_{ij} = \frac{1}{2}\left(\frac{\partial u_j}{\partial x_i} + \frac{\partial u_i}{\partial x_j}\right) \quad , \quad \tau_{ij} = \left(\frac{\partial u_j}{\partial x_i} - \frac{\partial u_i}{\partial x_j}\right)$$

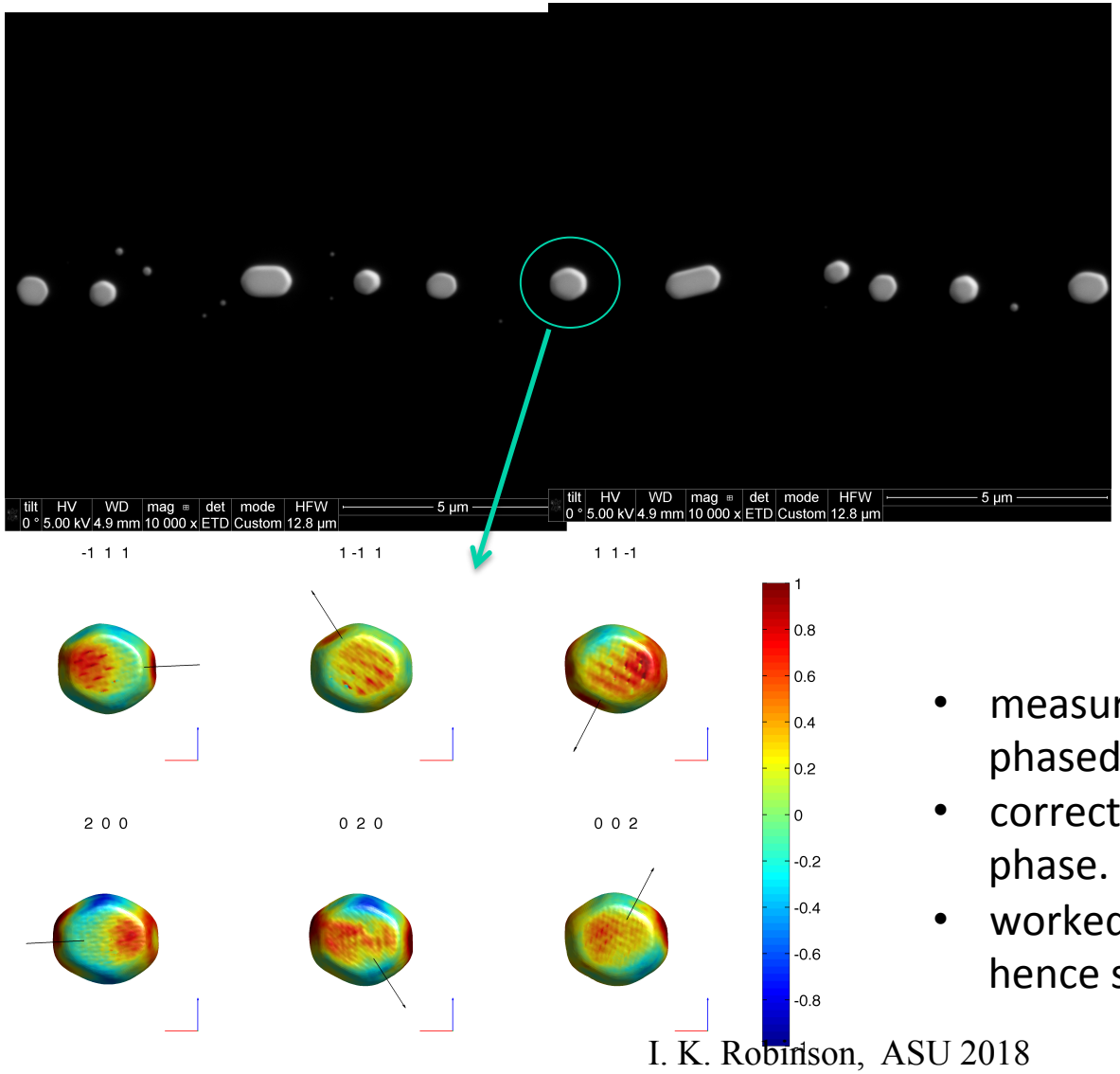


# Focused Ion Beam (FIB)



# Unimplanted Gold Nanocrystals

## Felix Hofmann, Oxford University 34-ID-C

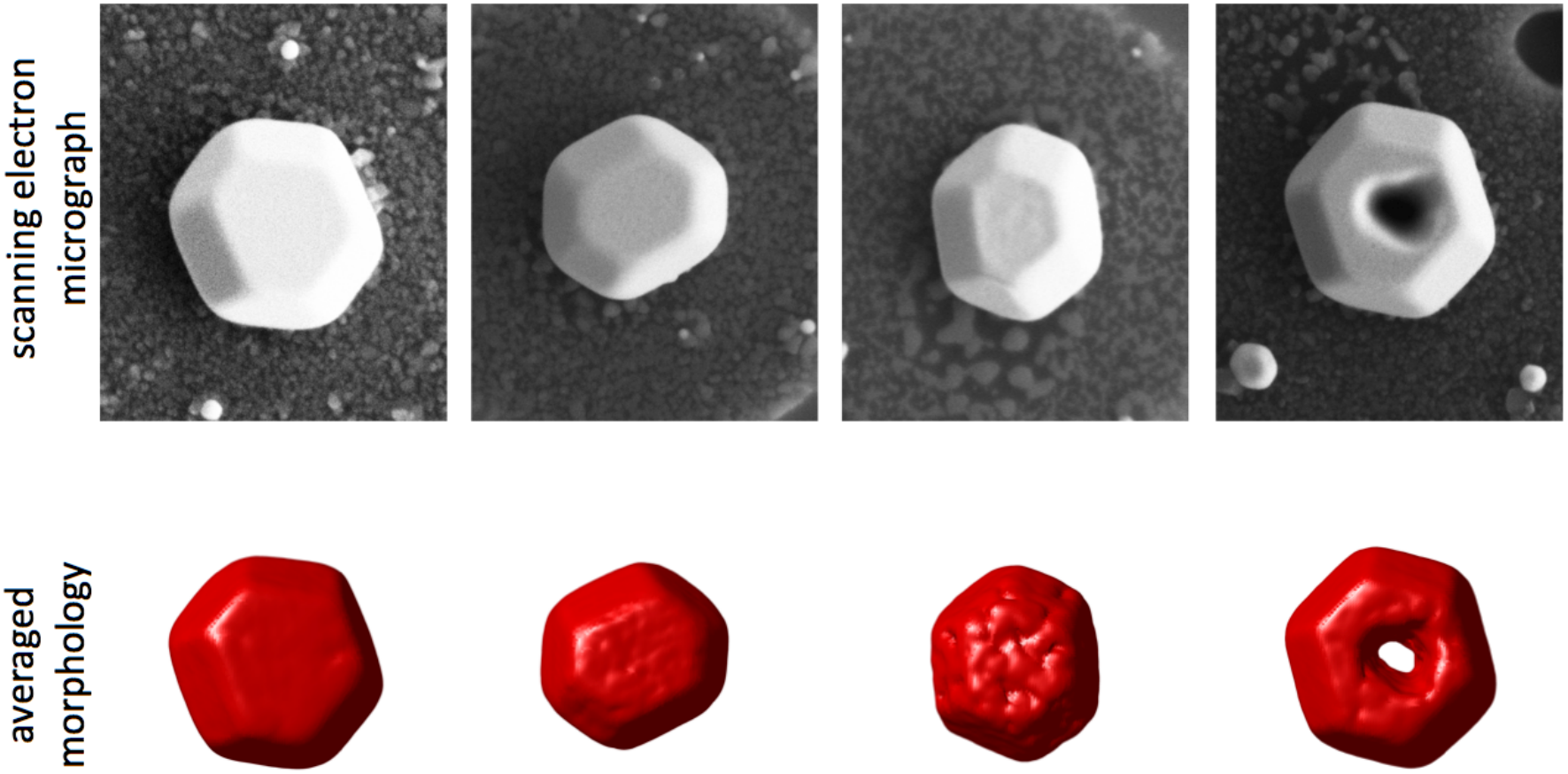



phases from different reflections

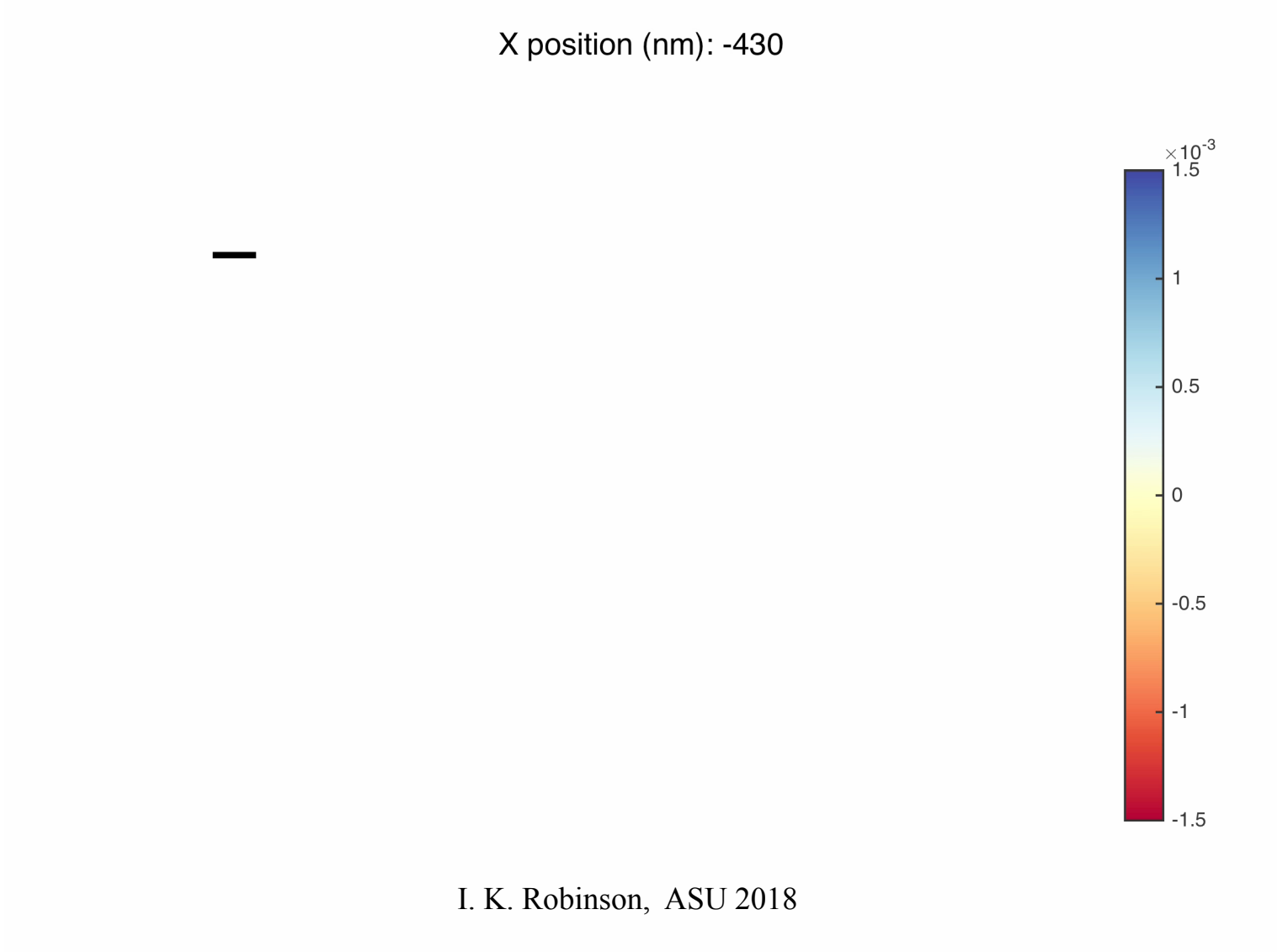
- measured 6 reflections at 9 keV. all phased fine.
- corrected for refraction-induced phase.
- worked out displacement field and hence strains

# Shape comparison: BCDI and SEM

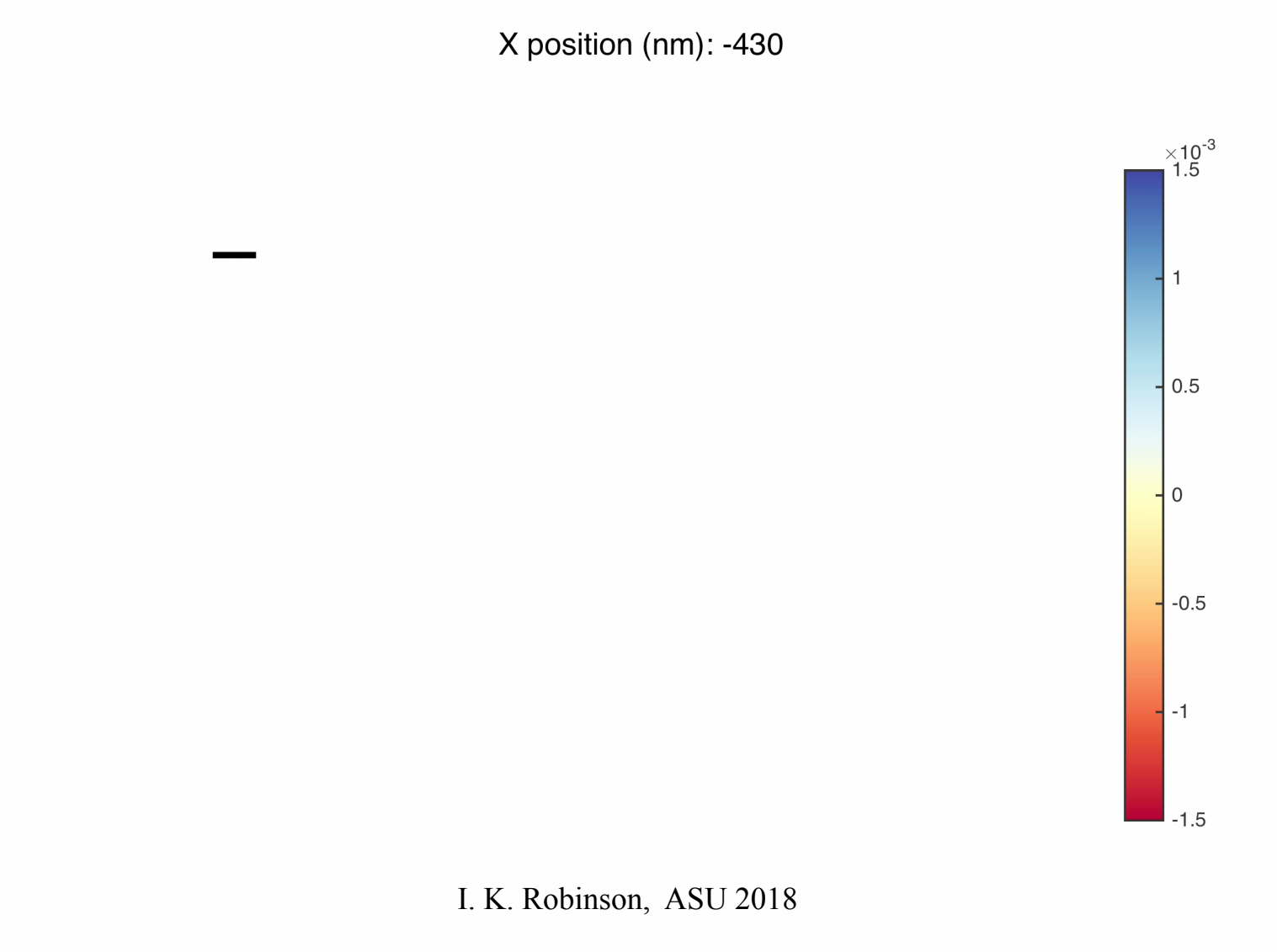
## F. Hofmann et at Nature Scientific Reports 7 45993 (2017)



# Starting crystal (yz plane)

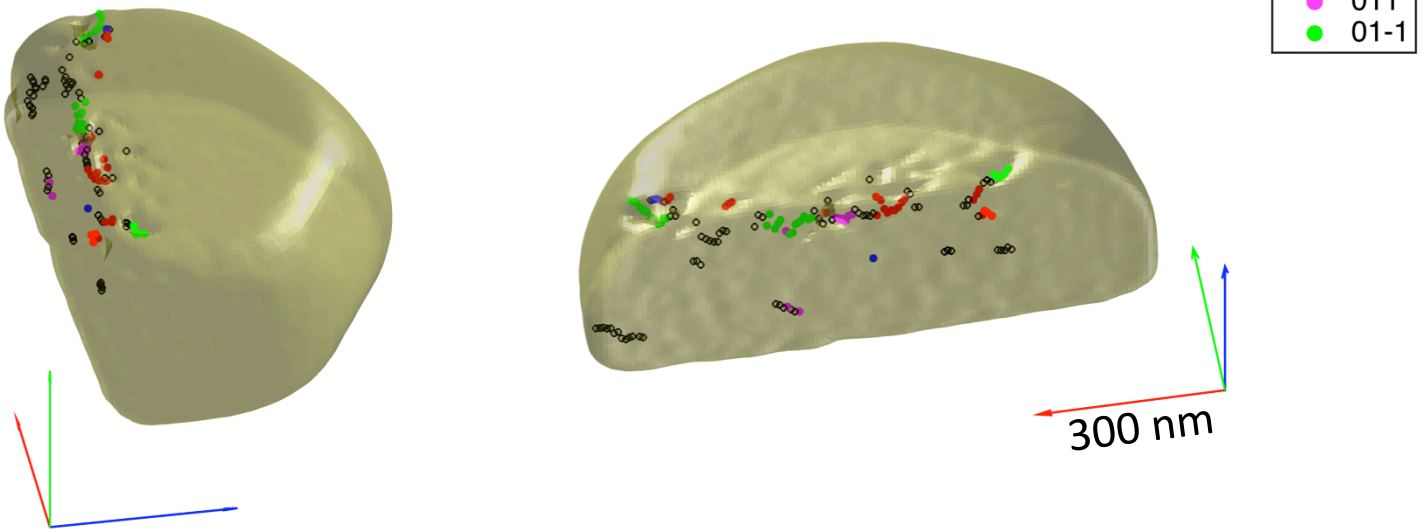


# After irradiation (yz plane)



# Dislocations

F. Hofmann et at Nature Scientific Reports 7 45993 (2017)




Amplitudes and phases of different reflections show jumps and pipes of reduced intensity consistent with dislocations. Dislocations identified in the crystal are shown above. They are colour coded according to their burgers vector direction, determined using Q.b contrast. Dislocations for which the burgers vector direction could not be identified are shown in black.

- dislocations are mostly formed at top edge of crystal where FIB beam has normal incidence component. These can form larger loops.
- some defects can also be seen on the milled face, although these are small.

# Bragg Coherent X-ray Diffraction

- Complex density can image strain
- Strain associated with nano-shape
- Diffusion of Cu in Au nanoparticle
- Ultrafast snapshots of vibrations
- Transient melting of nanoparticles
- Strain tensor imaging