High performance GISAXS

Alexander Hexemer

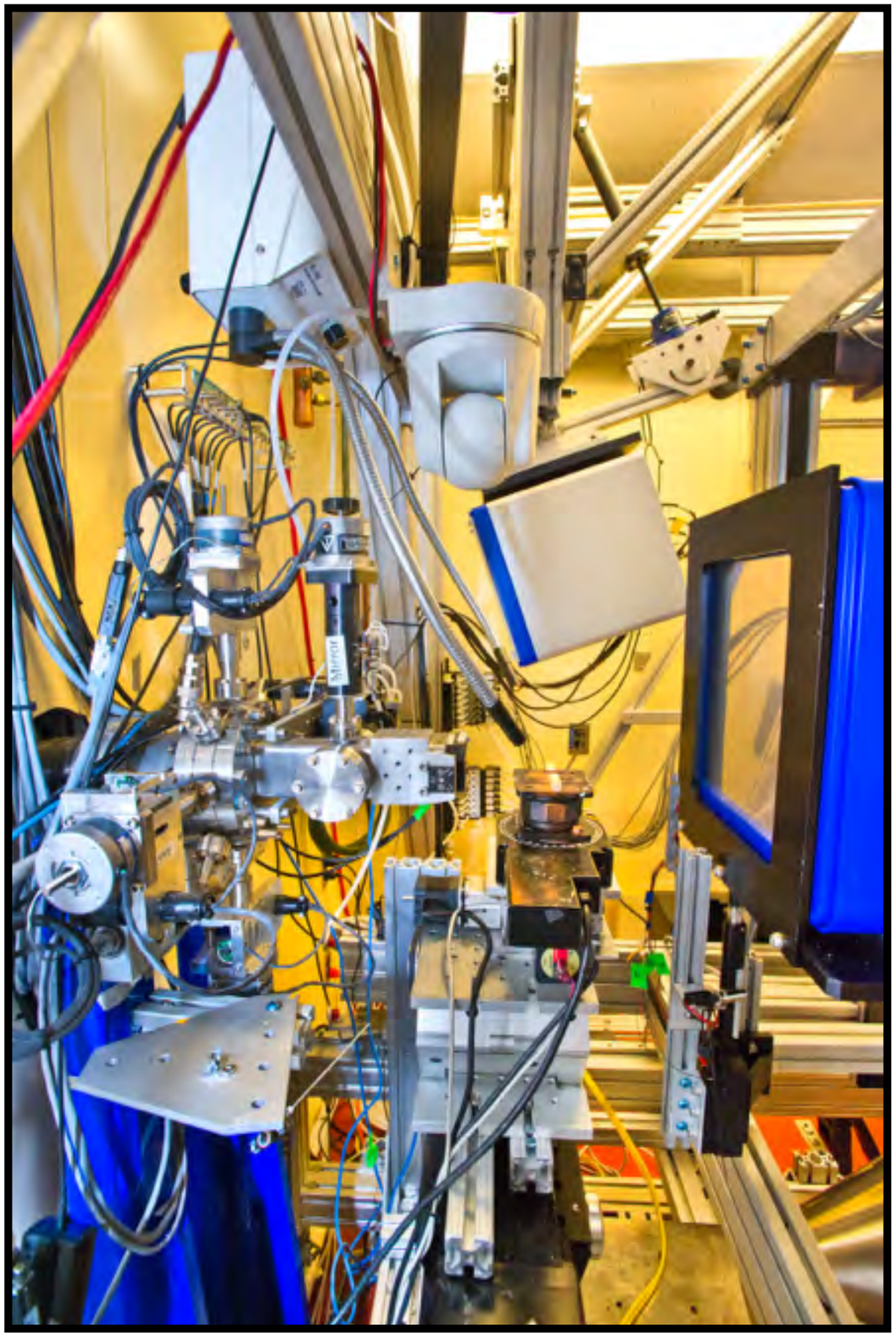
Early Career Award Program

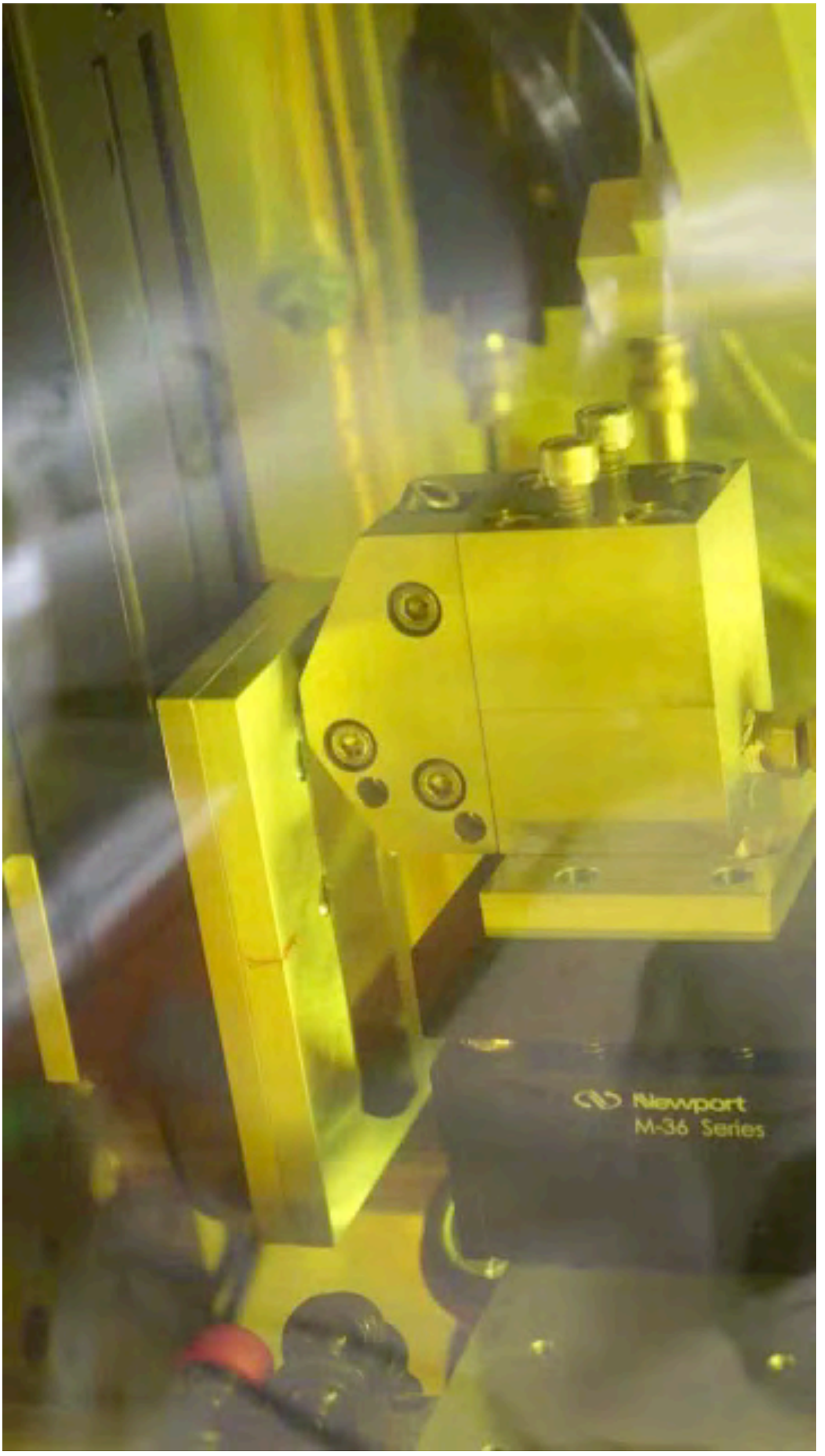
CAMERA

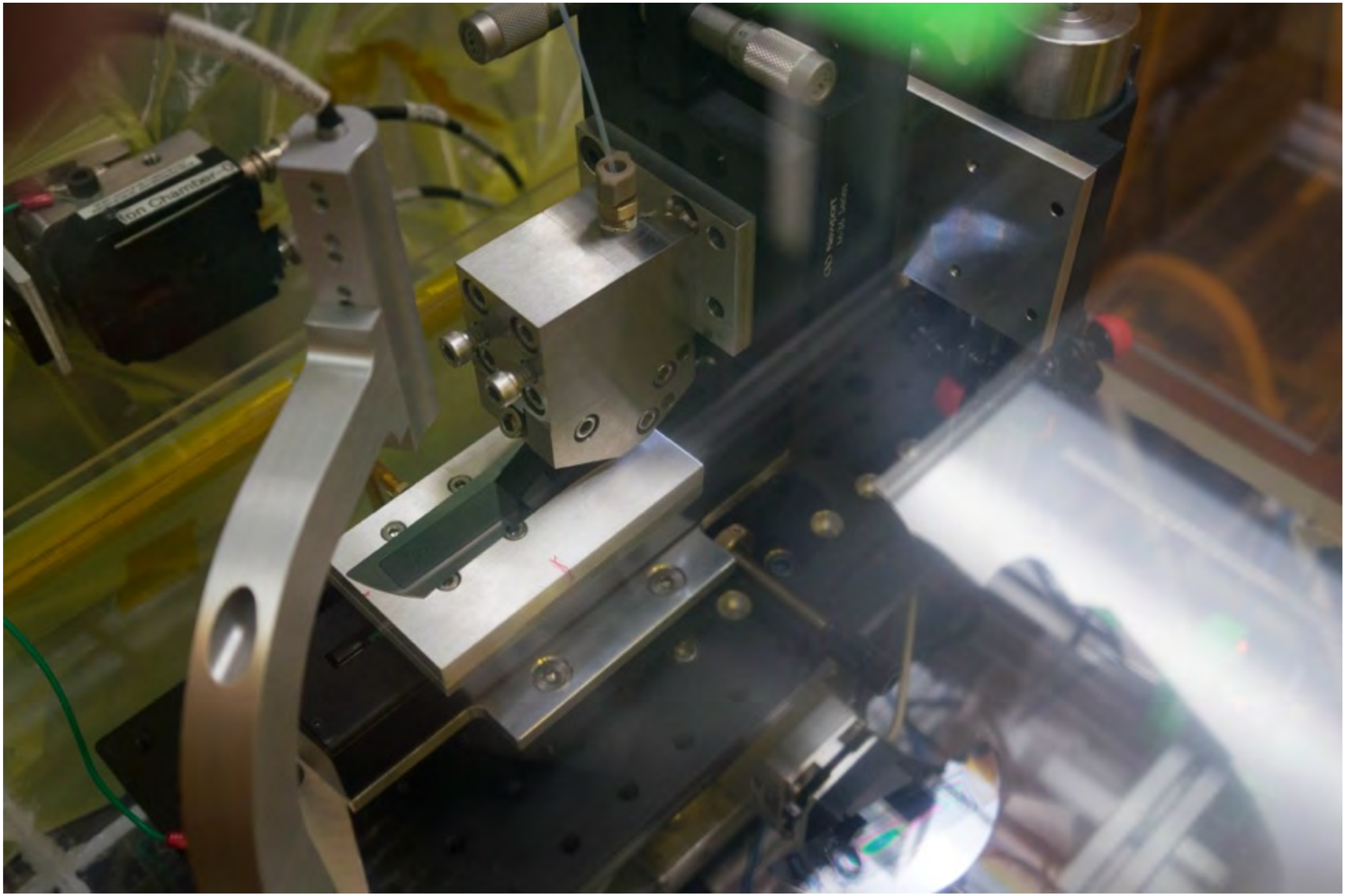
Center of Applied Mathematics for Energy Research Applications

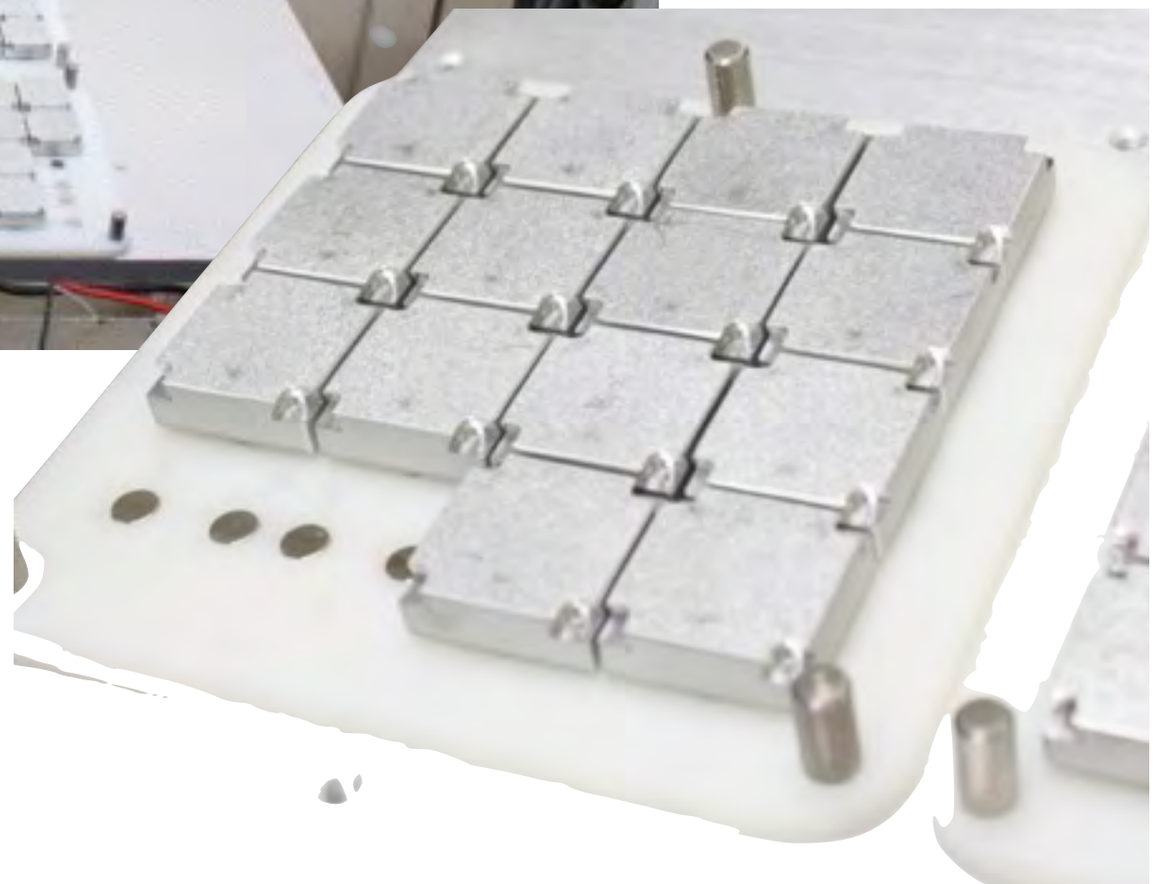
camera.lbl.gov

Beamline 7.3.3 SAXS/WAXS/GISAXS

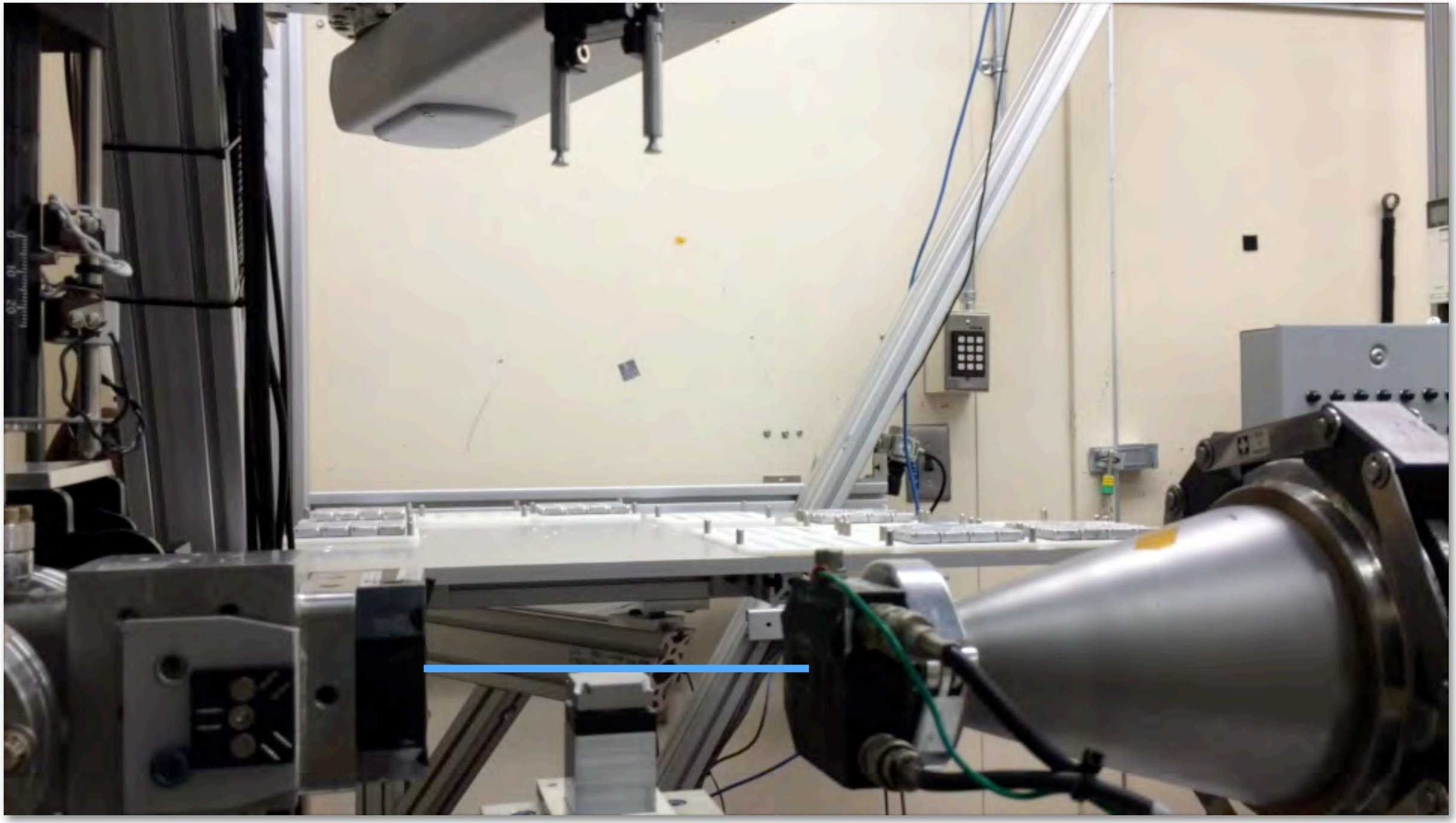


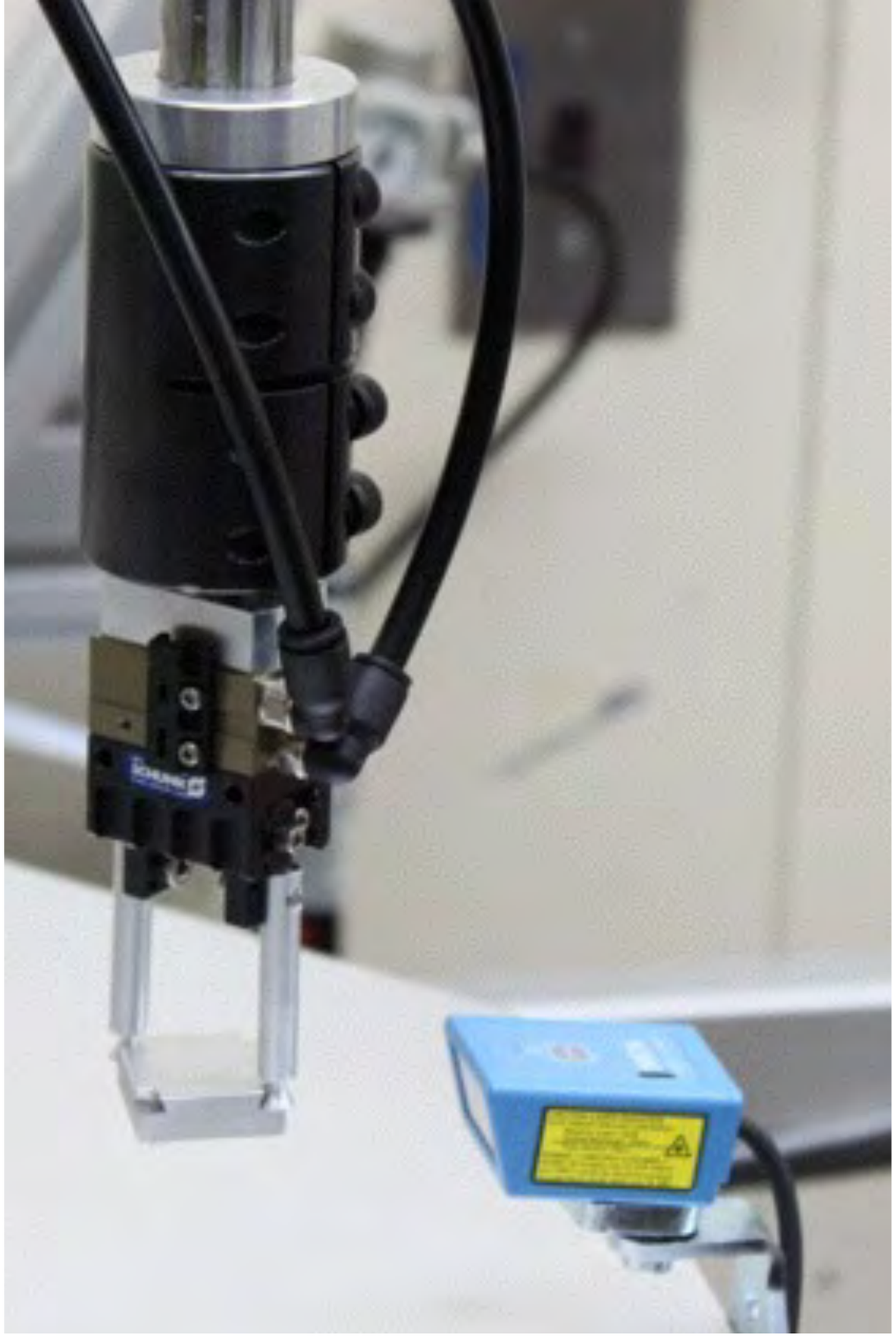

Newport
M-36 Series


Ion Chamber



RAPIDD PROPOSALS

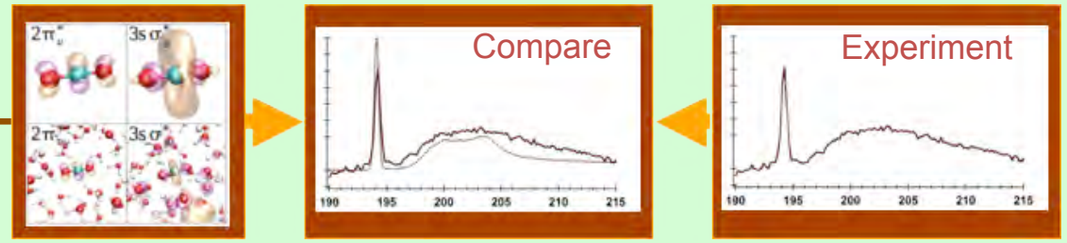




How the user sees end-to-end.

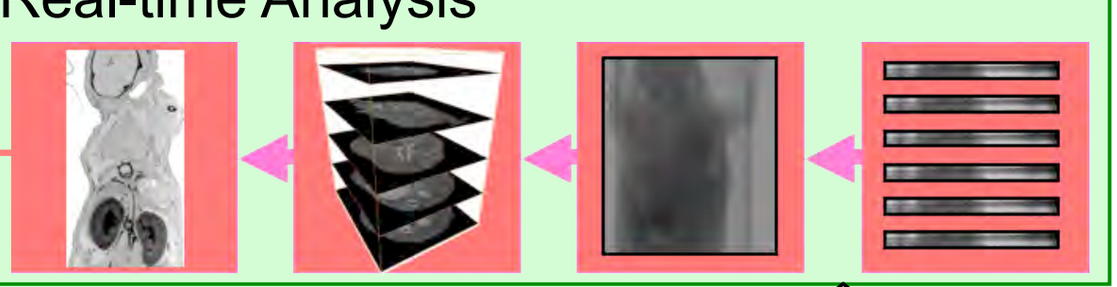
Framework

User Initiated Simulation



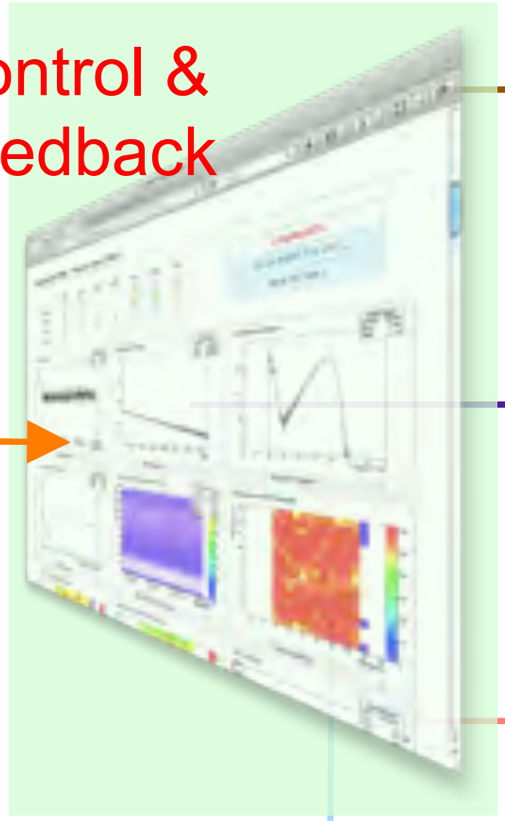
User Initiated Analysis

Real-time Analysis

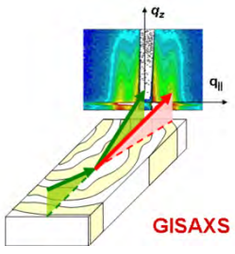


Real-time Analysis Pipeline

Control & Feedback



User



Experiment

Data Pipeline

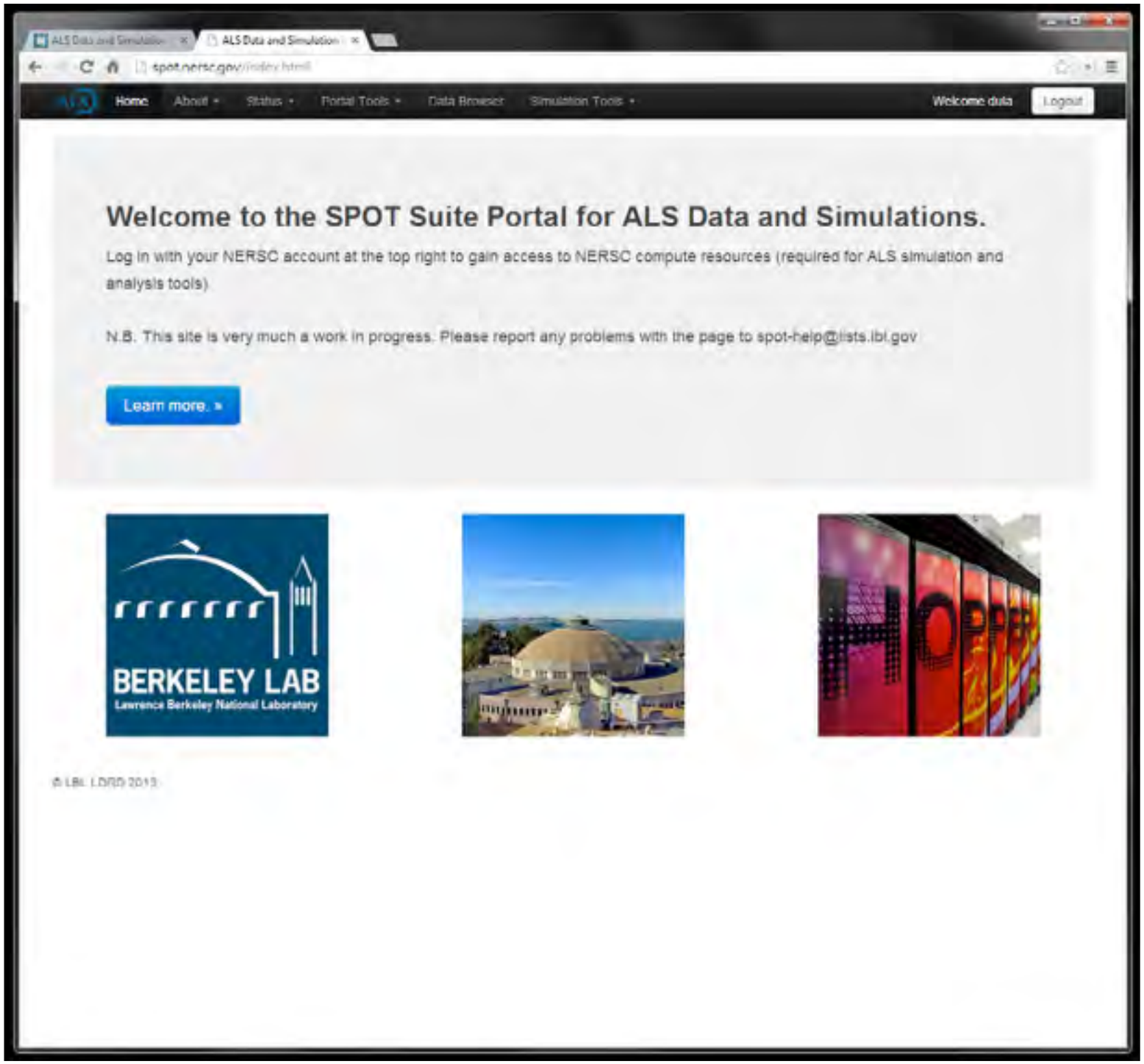
HPC Storage and Compute

ALS Data Portal

POWERED BY

NERSC ESnet CRD

- Users are asking for faster scans, but are not prepared for the consequences: they are overwhelmed by data rates/volumes
- Most don't have the background to use high performance computers
- SPOT Suite allows users to take advantage of high performance computers, to overcome their data problems

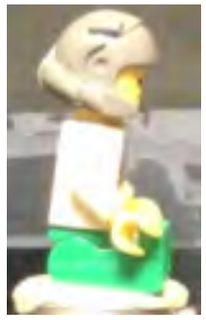


ALS Data Portal

POWERED BY NERSC ESnet CRD

Tomography Pipeline

Prep Sample



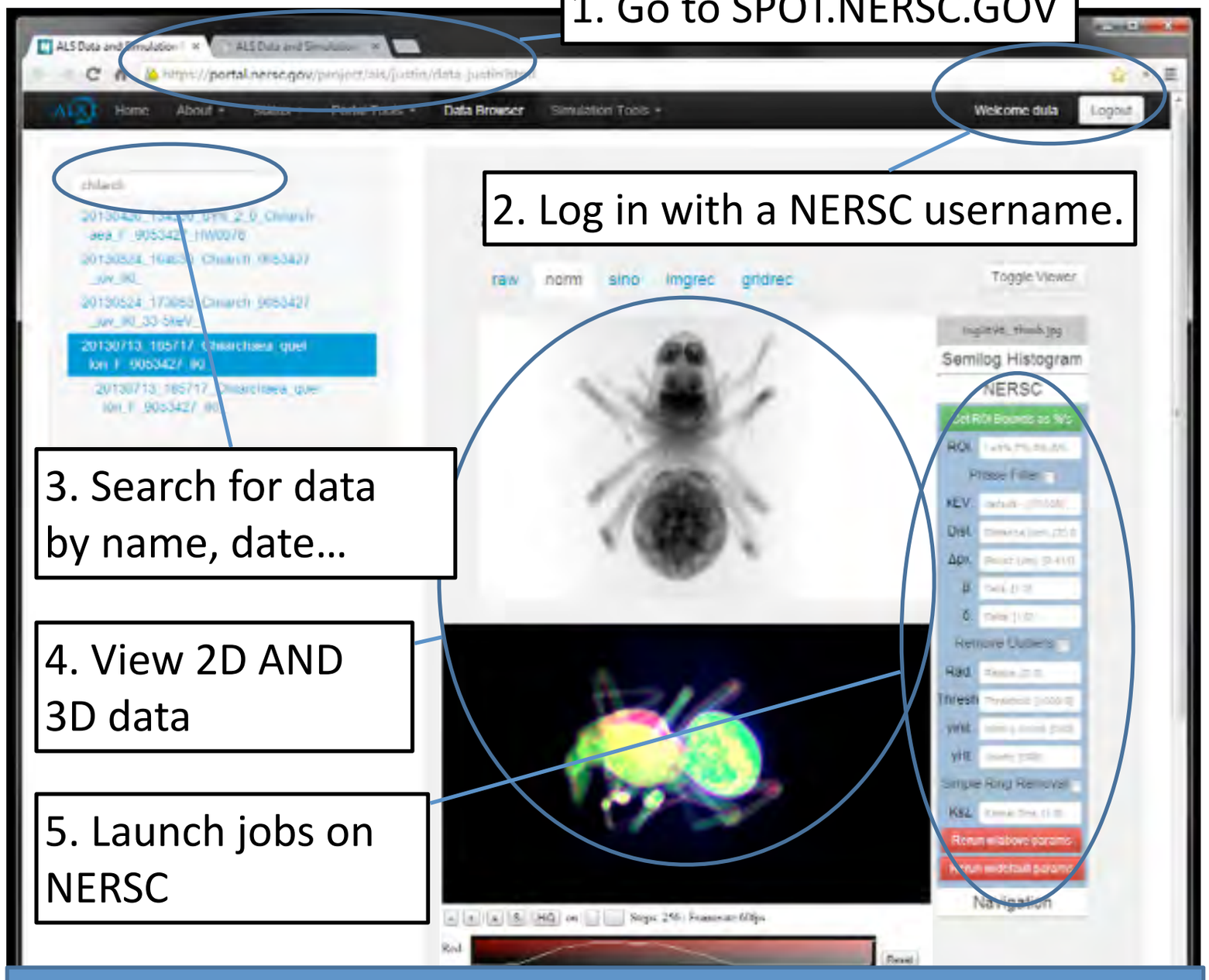
X-ray Image

Store/ Pack Data

Move Data

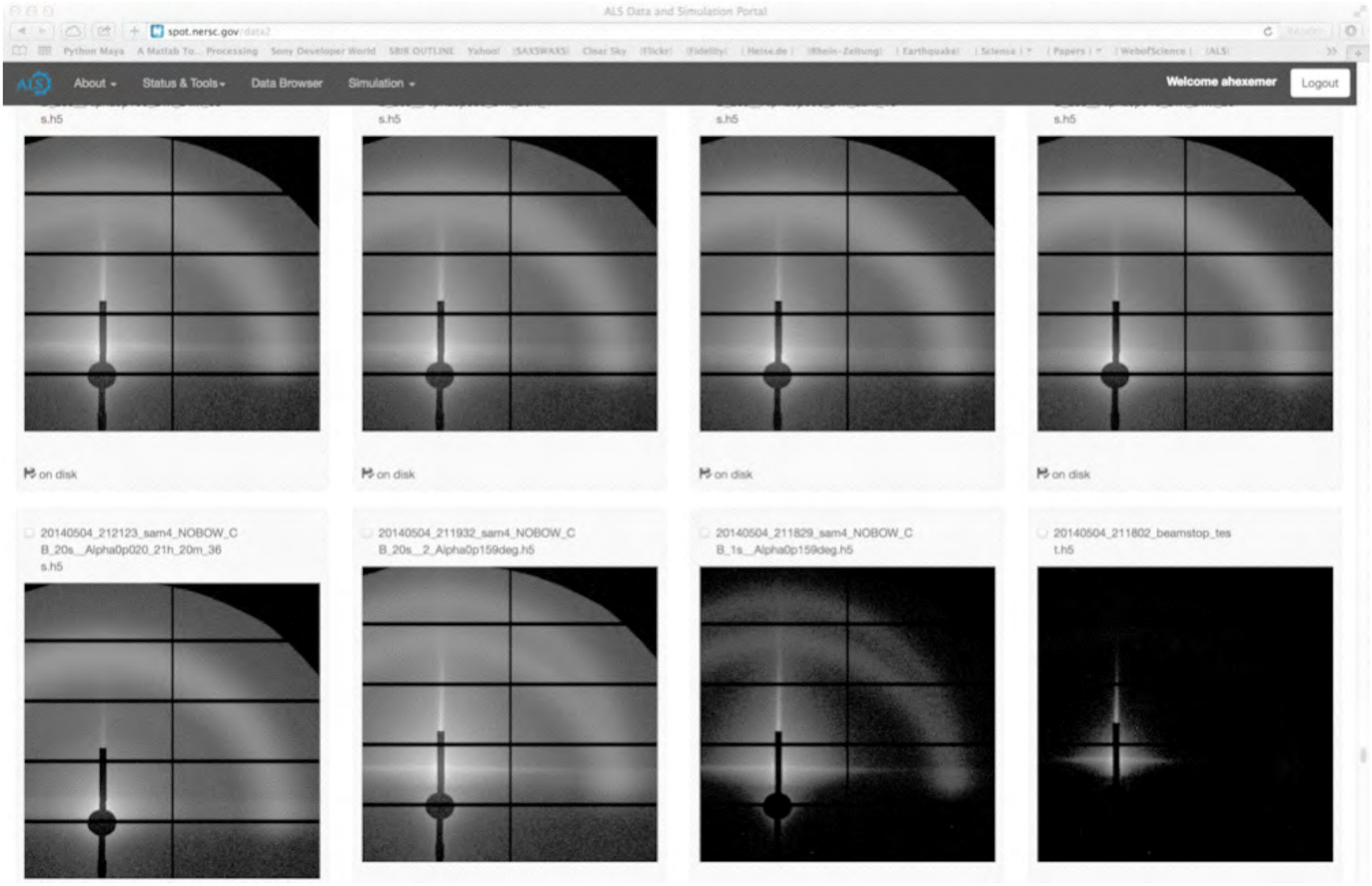
Process/ Simulate

SPOT FRAME-WORK



SPOT Suite transfers, processes, and presents data as it is collected, giving "real-time" feedback!

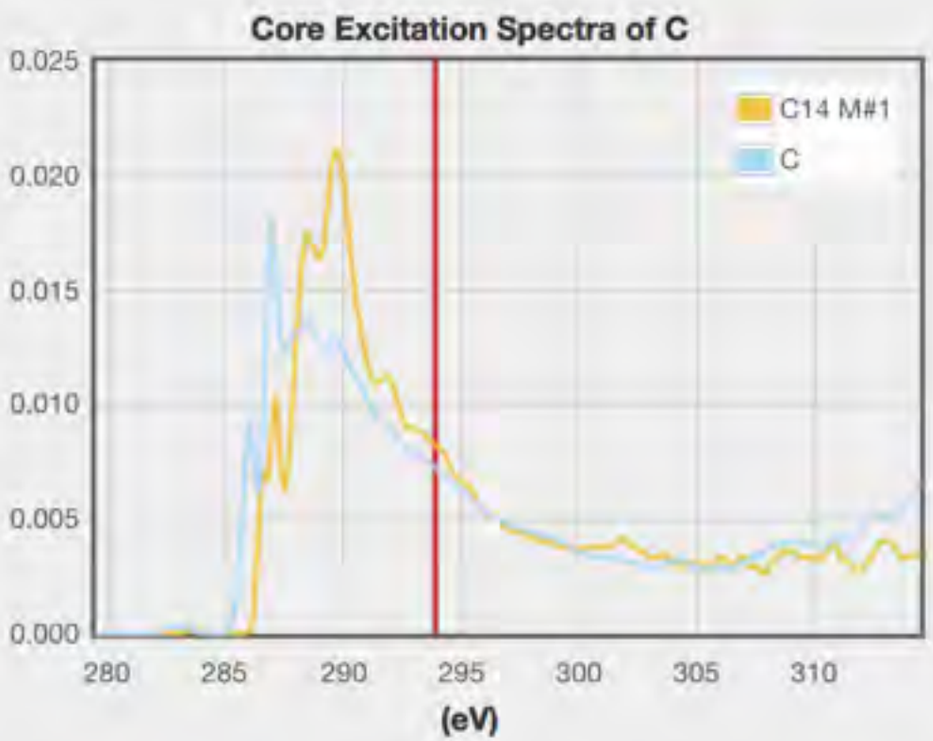
spot.nersc.gov

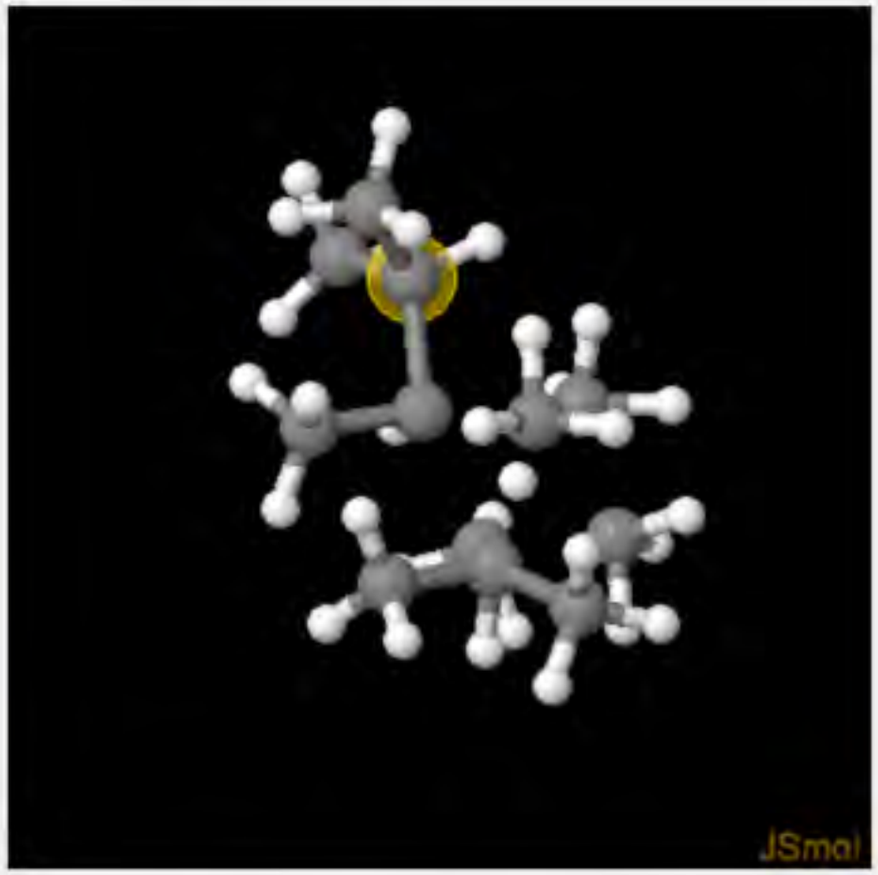


Results for MolName

Run May 6, 2014 at 4:03:57 PM PDT | Other Calculations | View Files | Delete Job

View Spectra of: C





Save Plot XCH Shift: 284.5

Show Excitation Spectrum of: ☑C ☐C2 ☐C6 ☐C7 ☐C8 ☐C9 ☐C10 ☐C11 ☐C12 ☐C13 ☑C14 ☐C15 ☐C16

Show Variation by Dimension? ☐

Show Models: ☑1

Postprocessing Options

Energy 296.98 Get States

Save This Spectra To MFTheory Database

Jmol Options:

View Model 1

Animate | Unitcell | Save IMG | POV

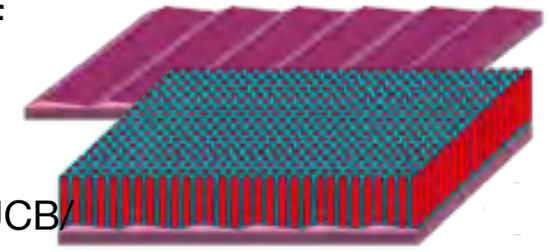
View States: Load Custom State

No contributing states.

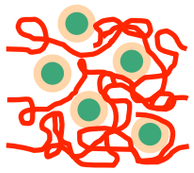
Note, these will take a few minutes to load.

Quantitative GISAXS modeling needs

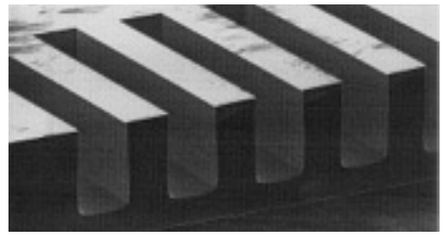
Long-range ordering of block copolymers for dense storage media
(Russell, UMass Amherst, Xu, UCB/MSD, A. Hexemer LBNL)



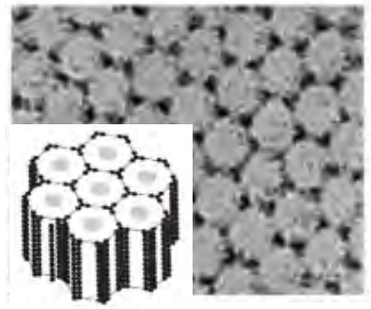
Nanoparticle/polymer composites for solar cells
(Segalman, UCB/MSD & Urban, TMF)



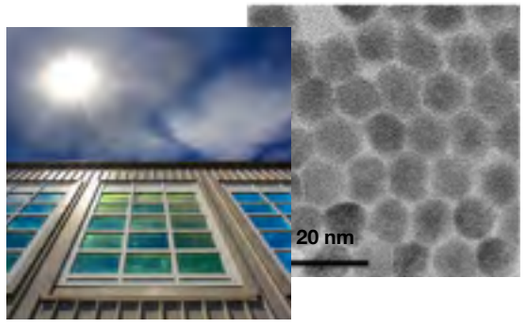
Lithographic patterning
(Soles, NIST; Ocko, BNL)



Self-assembly of nanoparticles in block copolymer thin films (Xu, UCB/MSD)

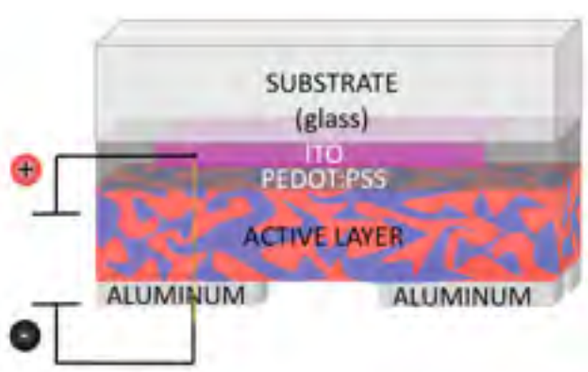


Electrochromic windows
(Milliron, TMF)

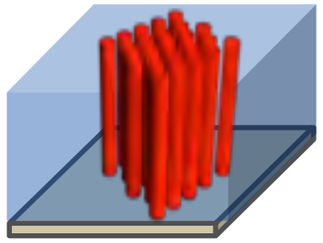


Battery electrolytes
(Balsara, UCB/MSD/EETD)

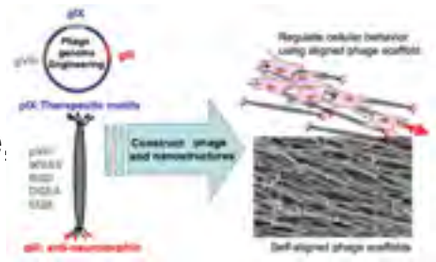
OPV BHJ materials
(McGehee, Stanford; Toney, SSRL/SLAC; Gomez, PSU; Kline, NIST; Liu, TMF; Ade, NCSU; Kramer, UCSB; Russell, UMass Amherst; Amassian, KAUST, A. Hexemer LBNL)



Composite membranes for artificial photosynthesis
(Segalman, UCB/MSD)

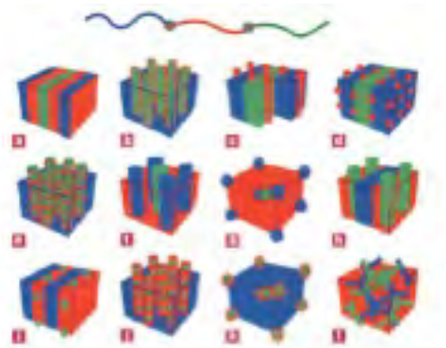


Virus nanofiber tissue engineering materials (Lee, UCB/PBD)



Block copolymer self-assembly

(Kramer, UCSB; Russell, UMass Amherst; Xu, UCB/MSD)



HIPGISAXS

camera.lbl.gov

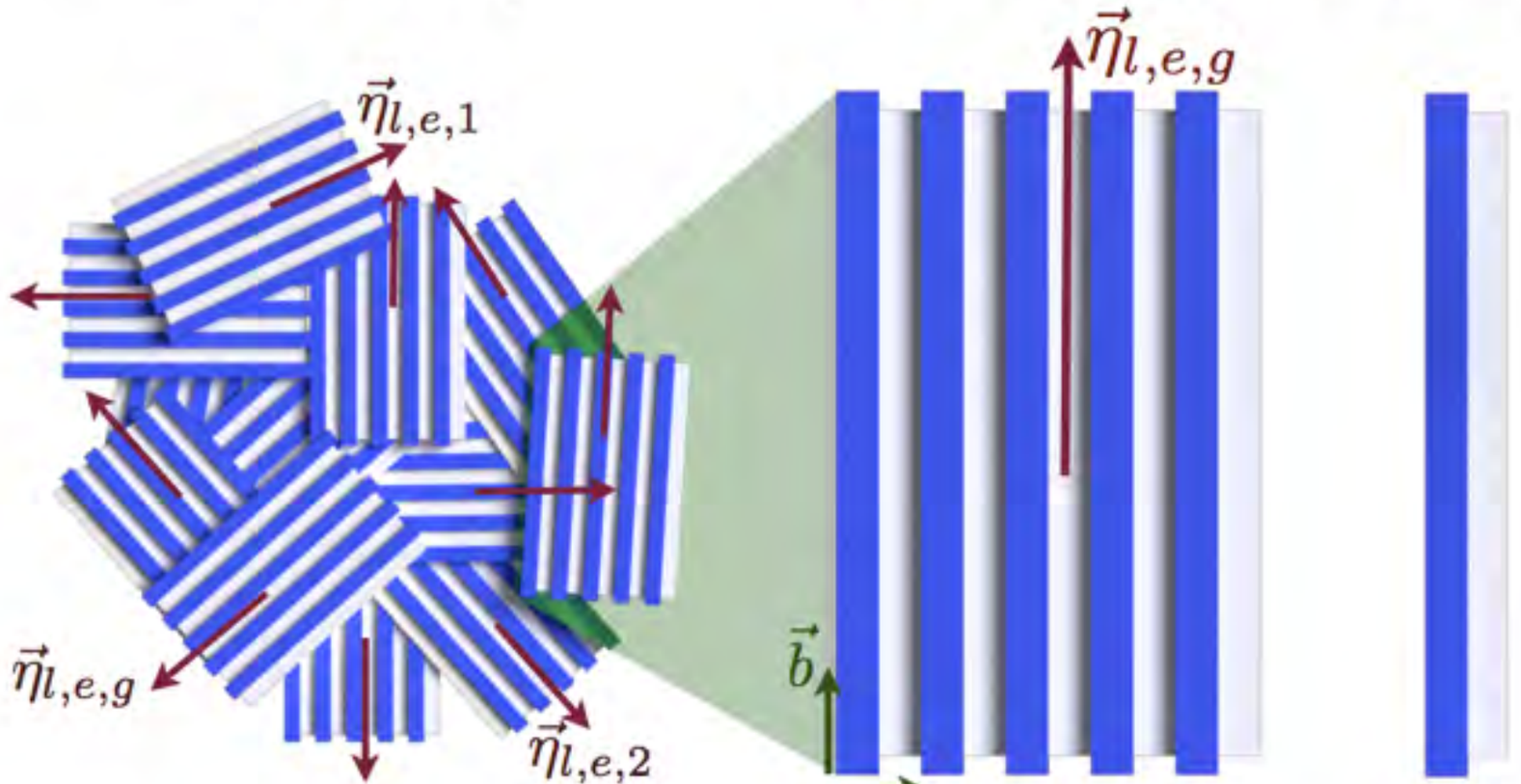
and

http://saxs-waxs-gpu2.lbl.gov

beta version online (slow)

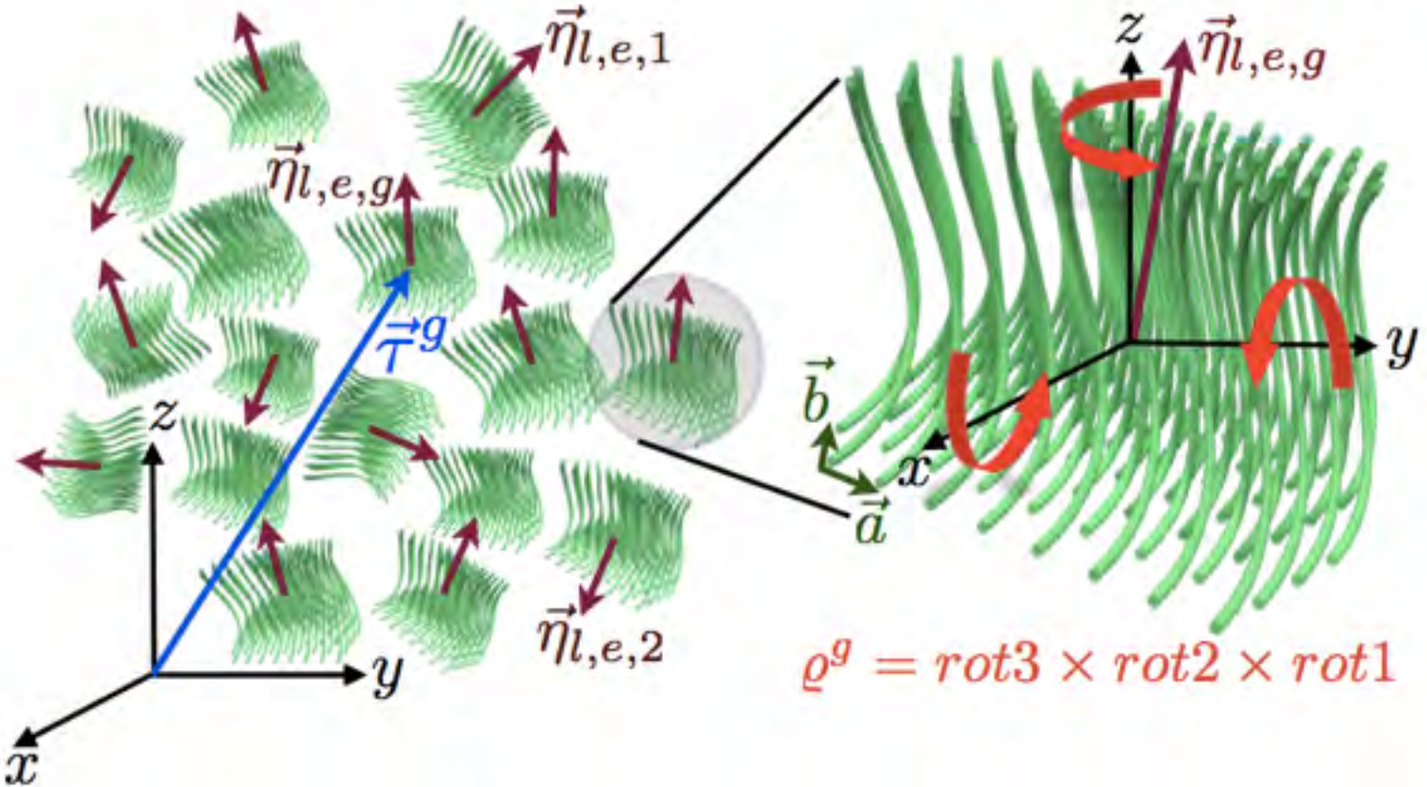
(A)

Ensemble: $\mathcal{E}_{l,e}$ Grain: $\mathcal{G}_{l,e,g}$ Shape: $\Omega_{l,e,g,s}$

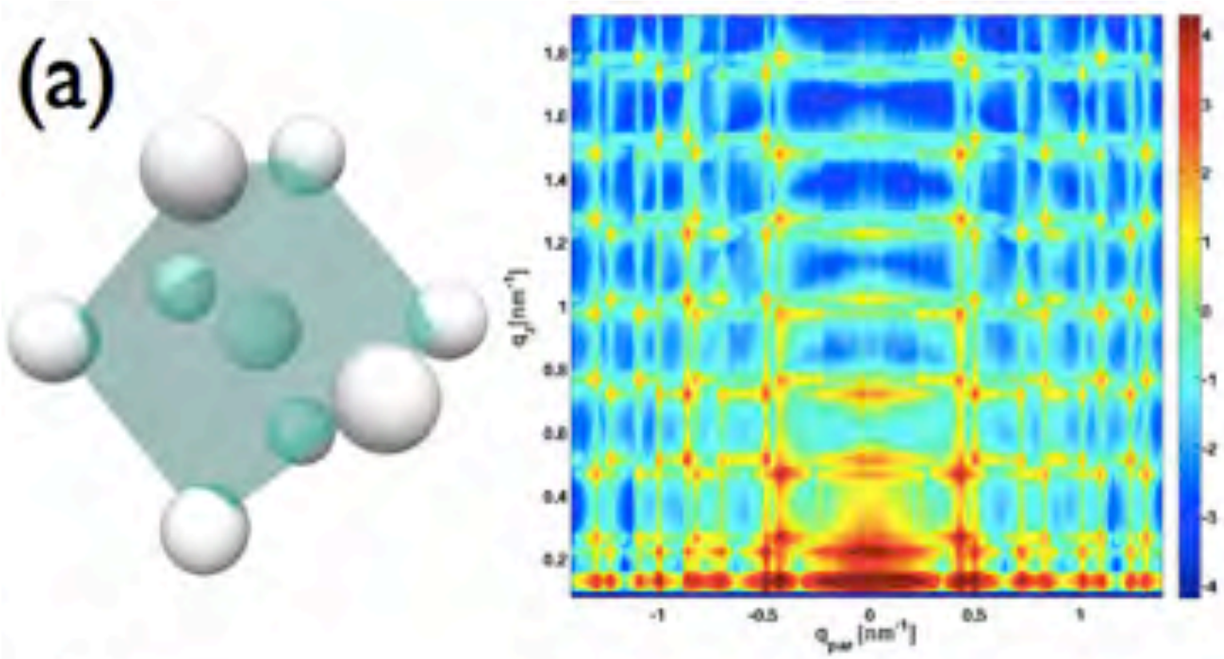


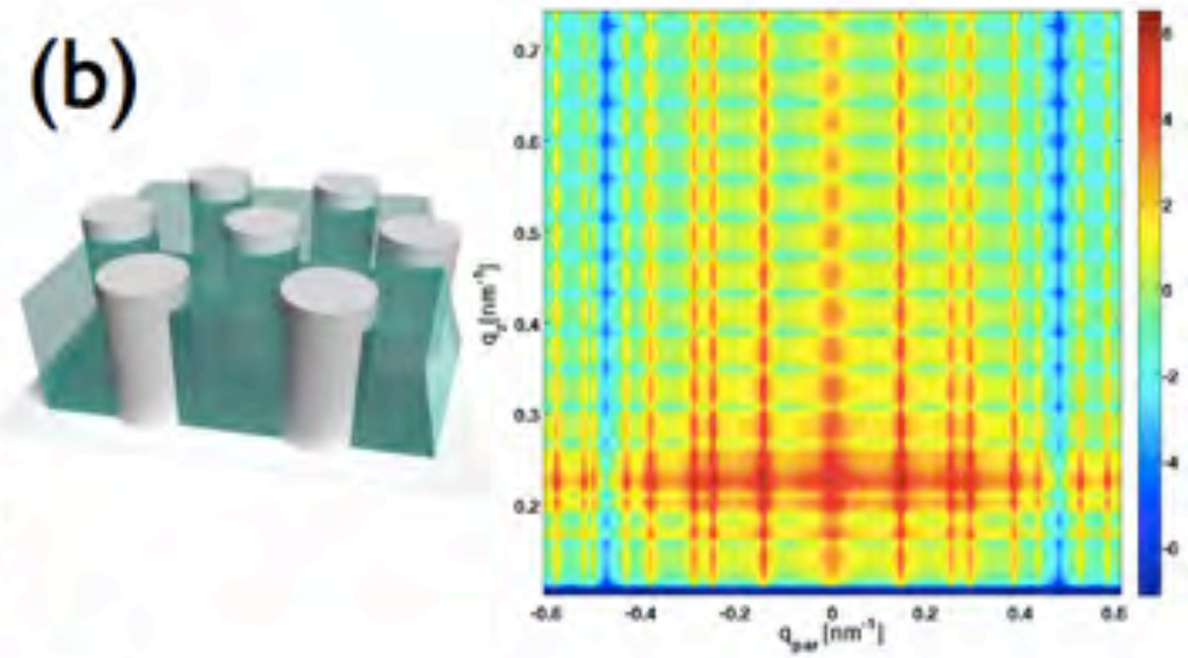
(B)

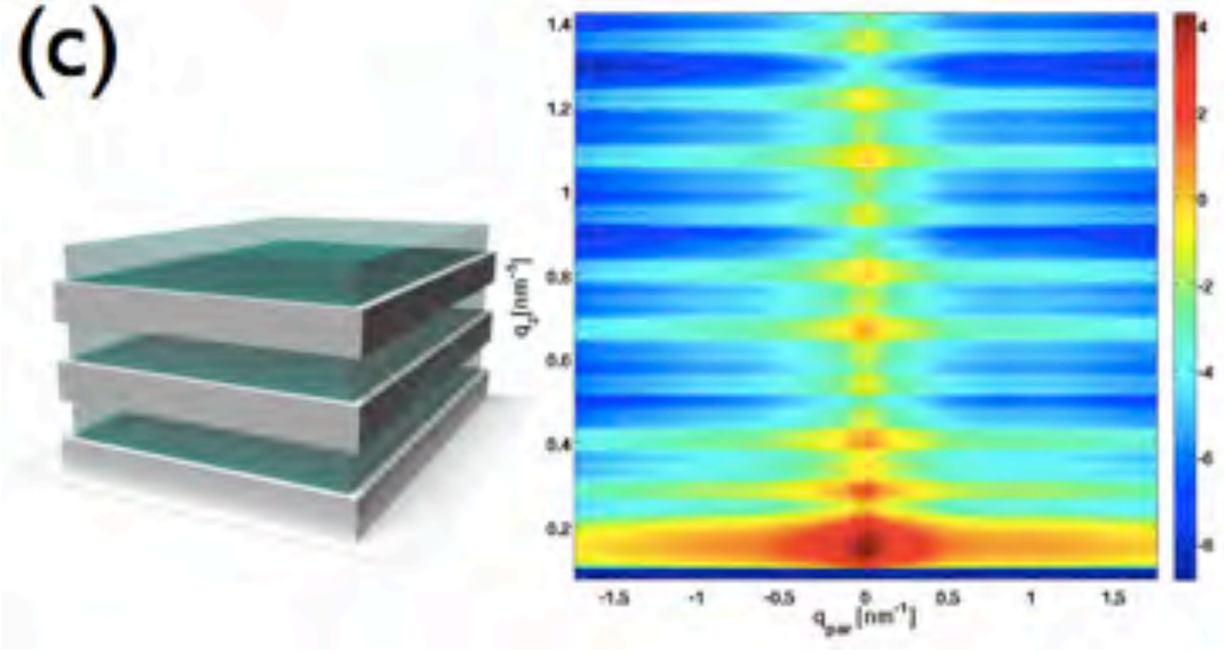
Ensemble: $\mathcal{E}_{l,e}$ Grain: $\mathcal{G}_{l,e,g}$ Shape: $\Omega_{l,e,g,s}$

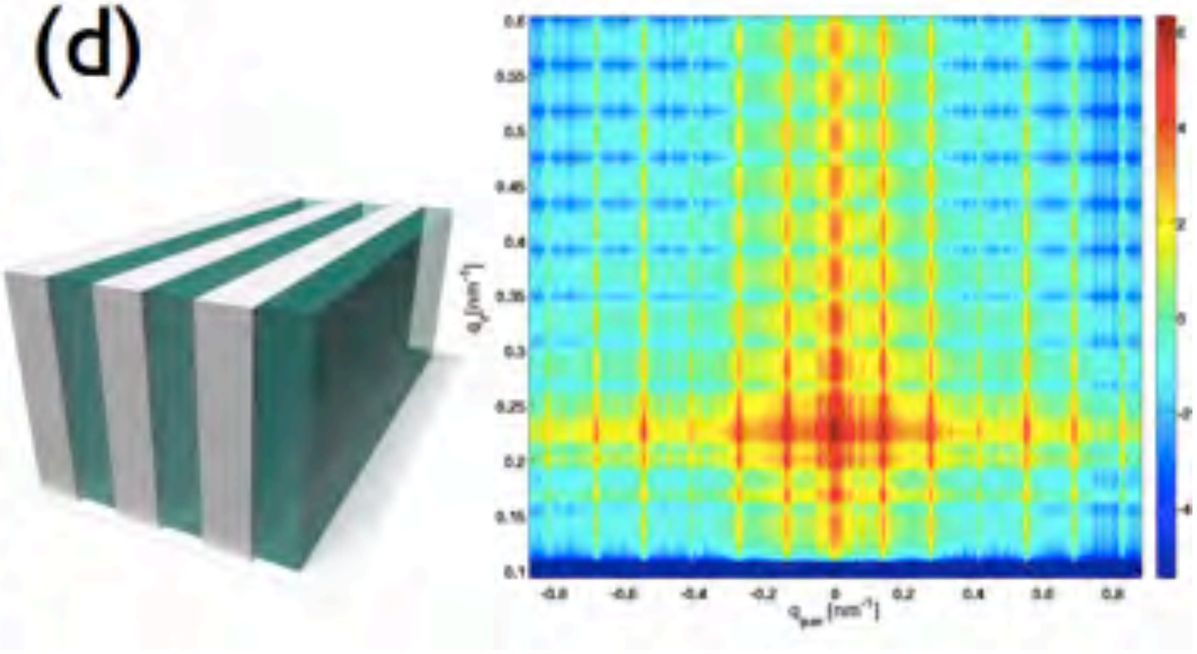


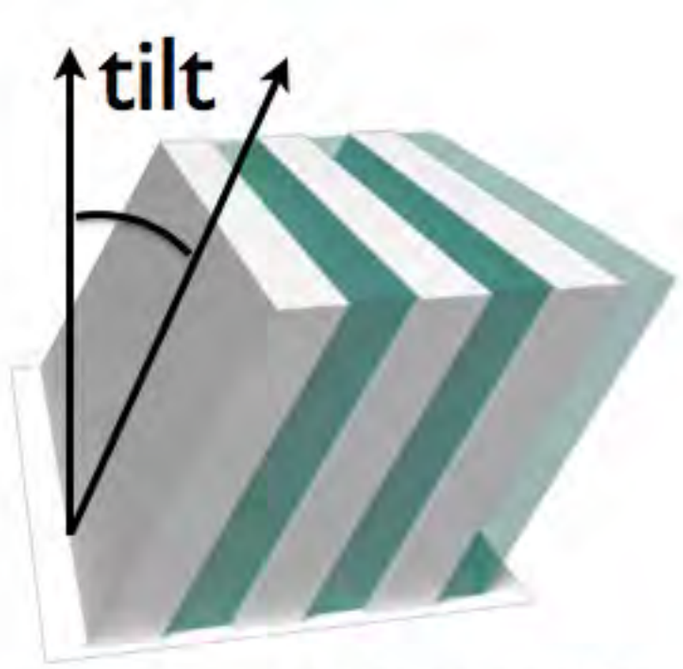
Some Simple Block Copolymer structures



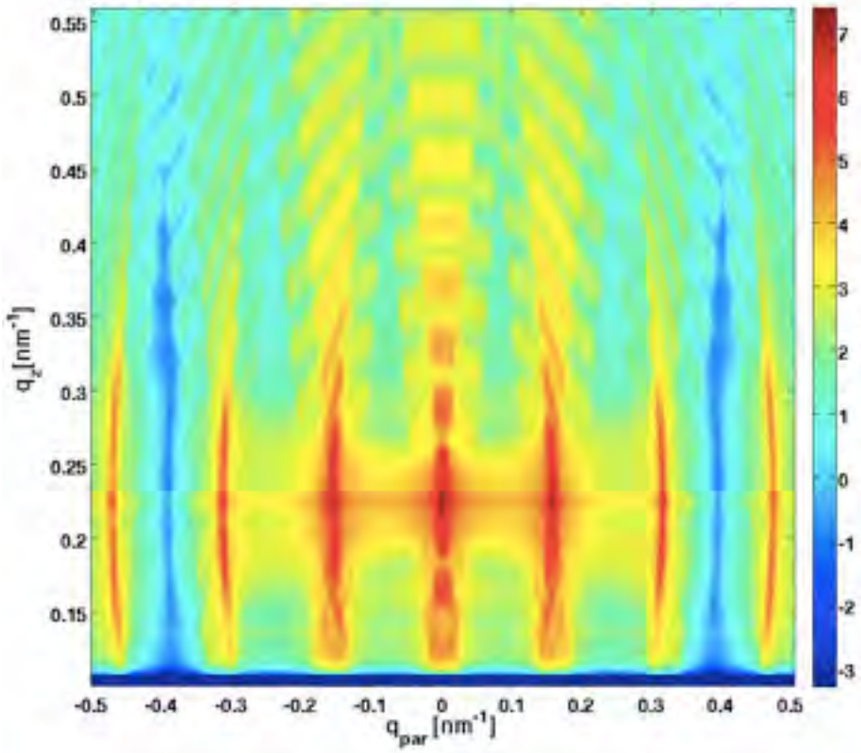




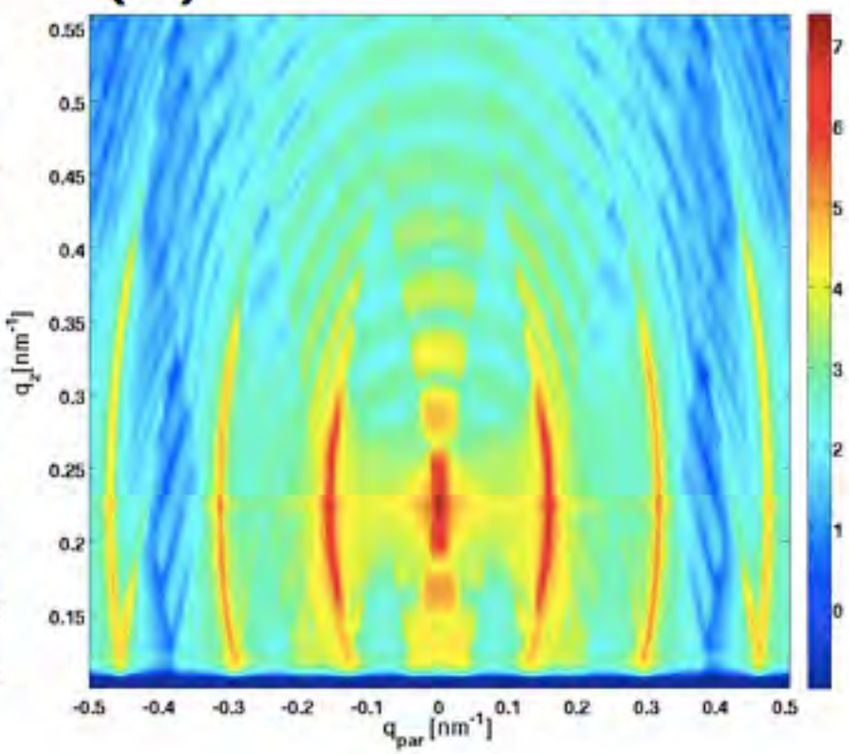




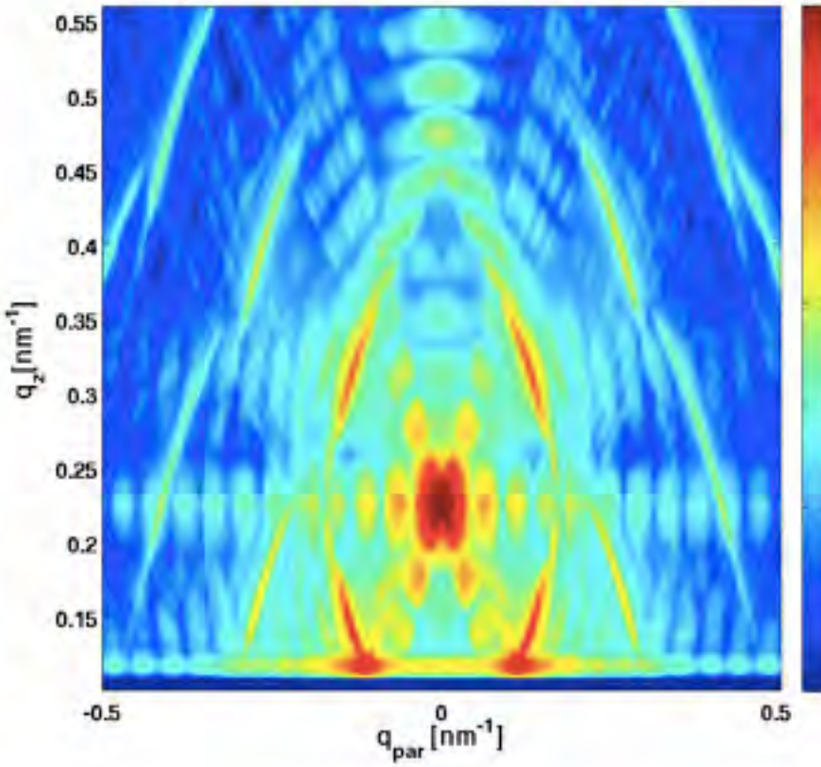
(a) tilt = $0° \pm 10°$



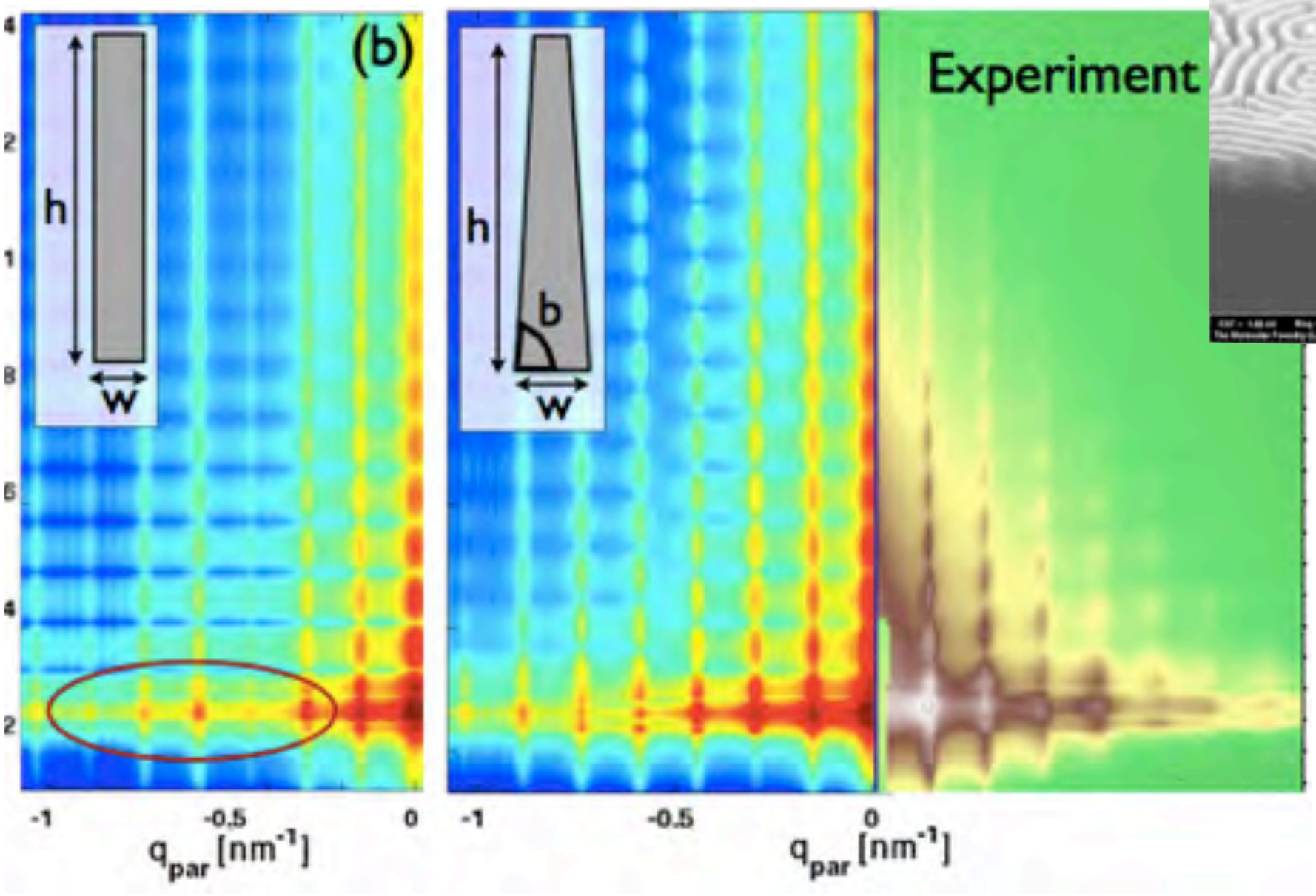
(b) tilt = $0° \pm 20°$

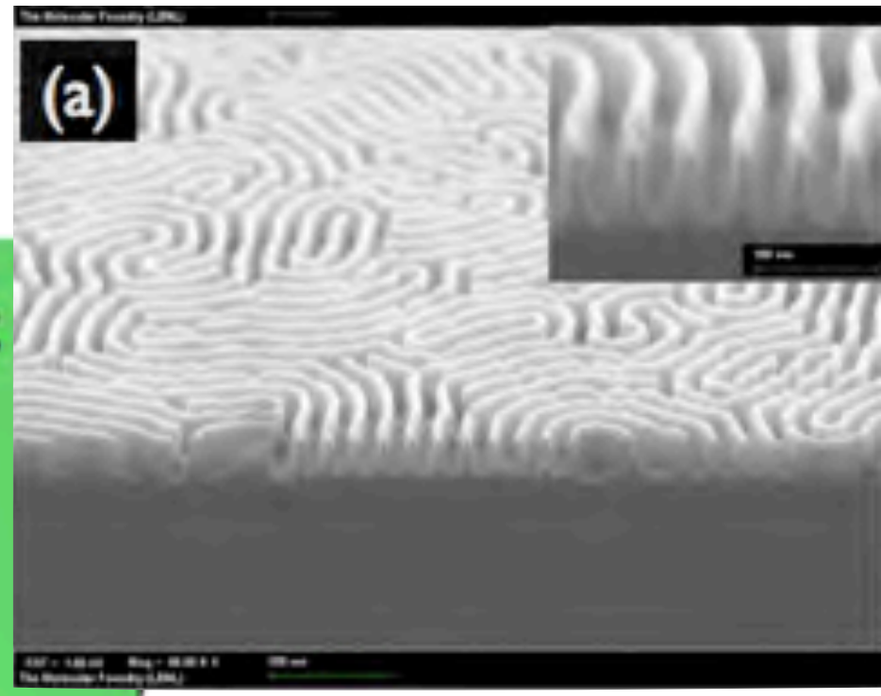


(c) tilt = $35° \pm 10°$

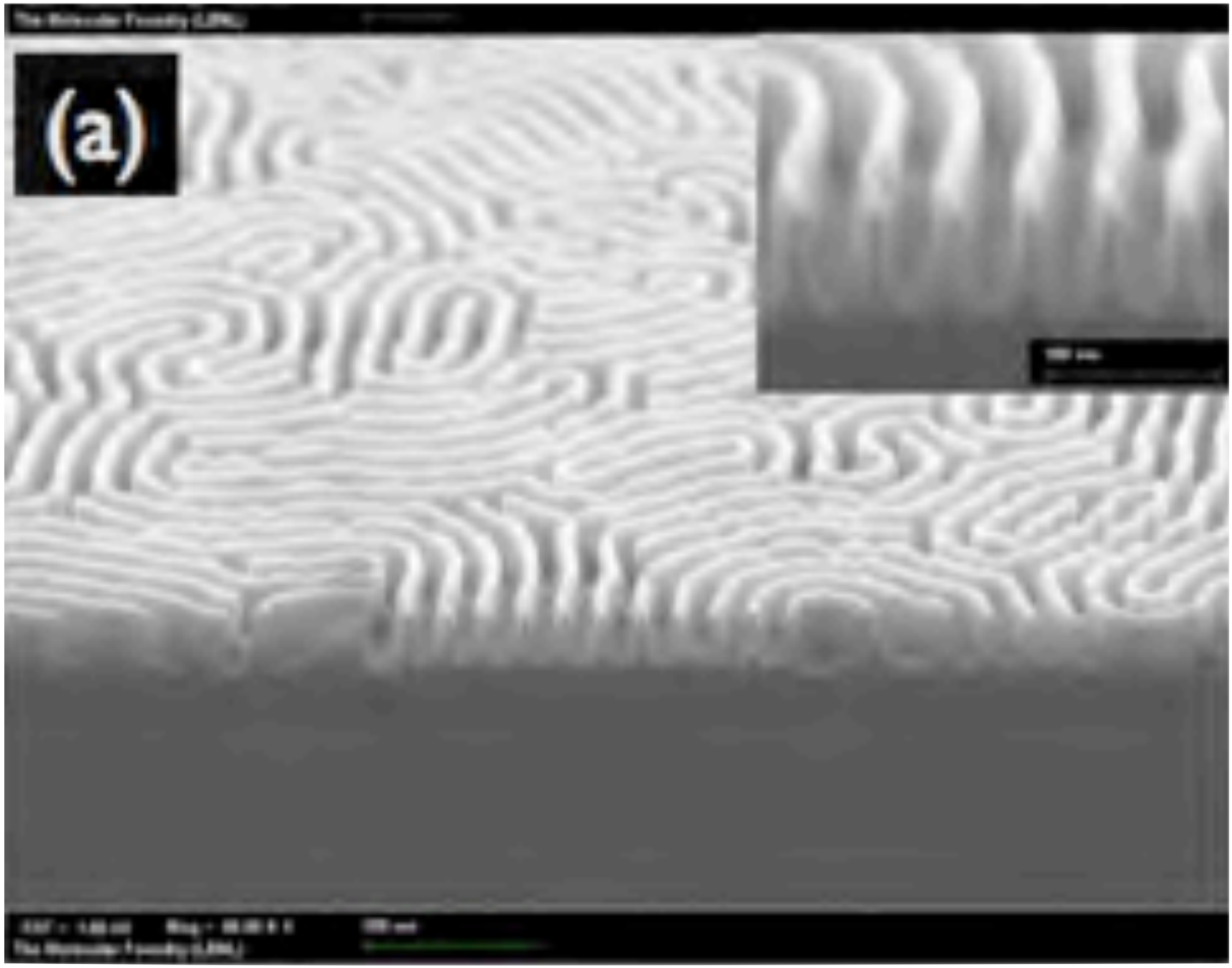


Computed GISAXS images for a “fingerprint” Si grating sample at incident angle $\alpha_i = 0.15°$





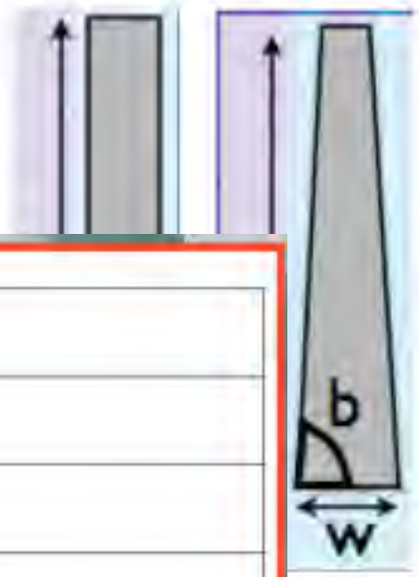
(b) Simulated GISAXS pattern for a rectangular model of the grating cross section with width and height ranges of nm and nm, respectively. (c) Simulated GISAXS pattern (*left*) using a shape with a trapezoidal cross section.


(a)

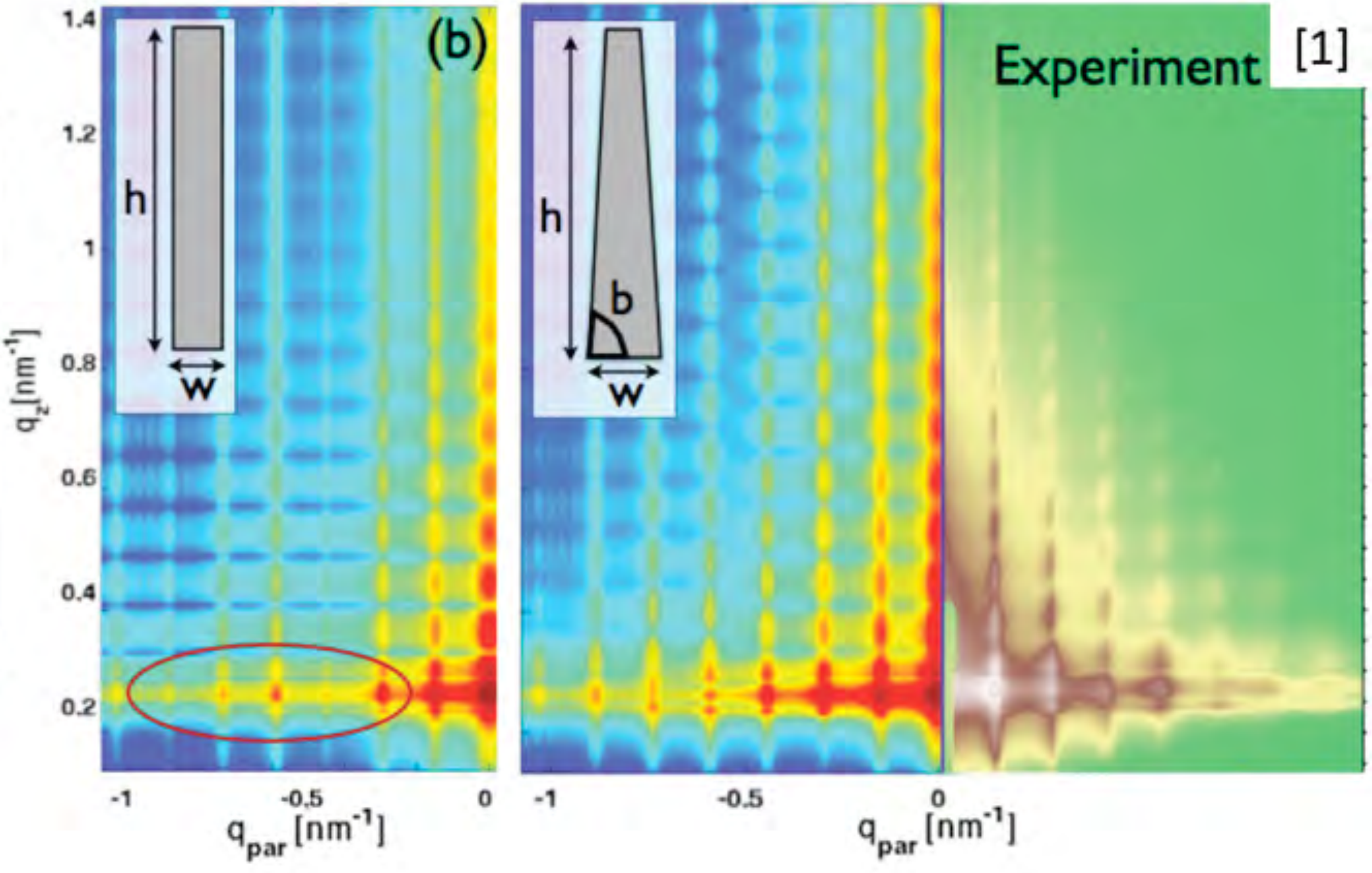
Si grating pattern (Cont'd)

```
hipGisaxsInput = {
shape = {
key = "s1", # a un
name = truncpyr
param={ type="heig
param={ type="widt
param={ type="leng
param={ type="base
},
layer = {
key = "substr", #
order = -1 , # spe
refindex = { delta
}
```

Shape	id	parameters
Box	box	"xsize", "ysize" , "height"
Cylinder	cylinder	"radius", "height"
Sphere	sphere	"radius"
Truncated pyramid	truncpyr	"xsize", "ysize" , "height", "baseangle"
Truncated cone	truncone	"radius", "height", "baseangle"
3-fold prism	prism3	"edge", "height"
6-fold prism	prism6	"edge", "height"
Sawtooth (prism along x)	sawtooth	"xsize", "ysize", "height", "baseangle"



[1] Xiaodan Gu, Zuwei Liu, Ilja Gunkel, S. T. Chourou, Sung Woo Hong, Deirdre L. Olynick, and Thomas P. Russell. *Advanced Materials* (2012)

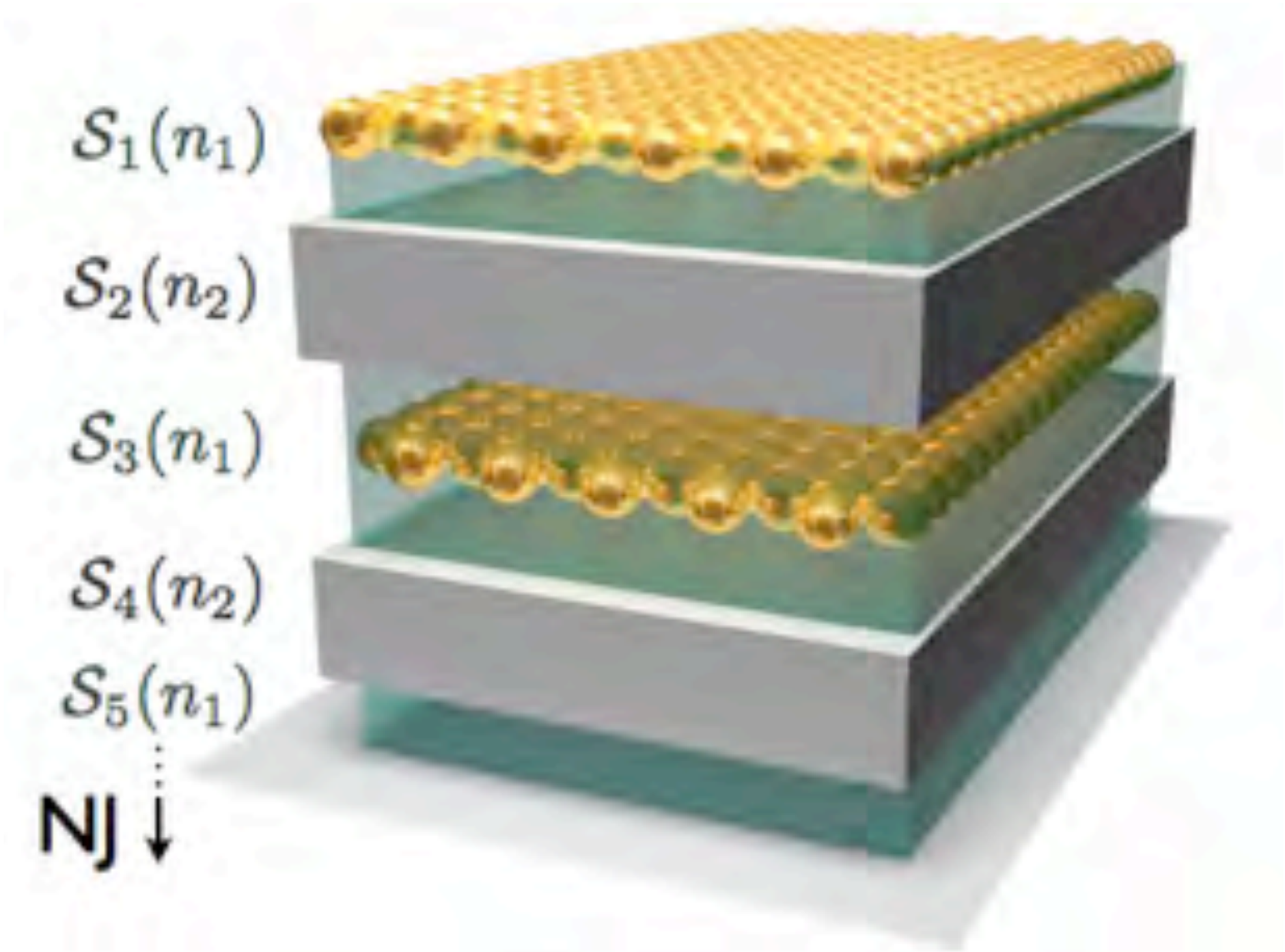

(b)
h
w
h
b
w
Experiment
[1]
1.4
1.2
1
0.8
0.6
0.4
0.2
q_z [nm^{-1}]
-1
-0.5
0
q_{par} [nm^{-1}]
-1
-0.5
0
q_{par} [nm^{-1}]

Si grating pattern (Cont'd)

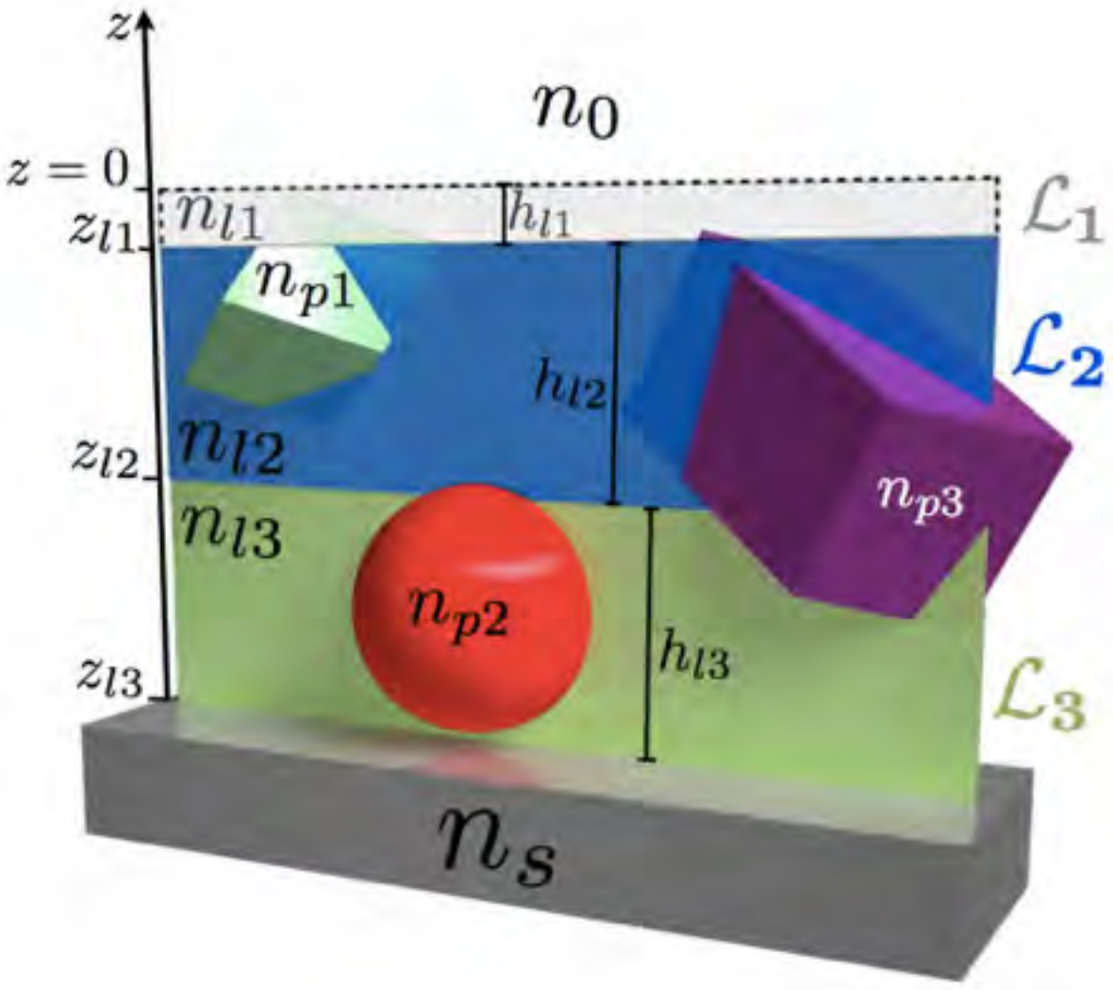
```
structure = {
key = "st1",
grain = {
shape:key = "s1", # key of the relevant shape
refindex = { delta = 4.88e-06, beta = 7.37e-08 }, # refractive index parameters of grain
layer:key = "", # key of the relevant layer
lattice = { type="cubic" } , # lattice forming grain
scaling = 43, # scaling factor for lattice vectors
repetition = [ 1 8 1 ] # shape repetitions in x, y and z
},
ensemble = {
maxgrains = [ 20 20 1 ], # maximum number of grains along x, y and z
distribution = "random", # spatial domain distr. in irrad. Volume (see list)
orientations = {
stat="range", # domain orientation distr. (see list)
rot1={ axis="x", angles=[ -10 10 ] }, # first rotation to apply to domain
rot2={ axis="z", angles=[ 0 360 ] } # second rotation to apply to domain
}
}
},
```

[1] Xiaodan Gu, Zuwei Liu, Ilja Gunkel, S. T. Chourou, Sung Woo Hong, Deirdre L. Olynick, and Thomas P. Russell. *Advanced Materials* (2012)

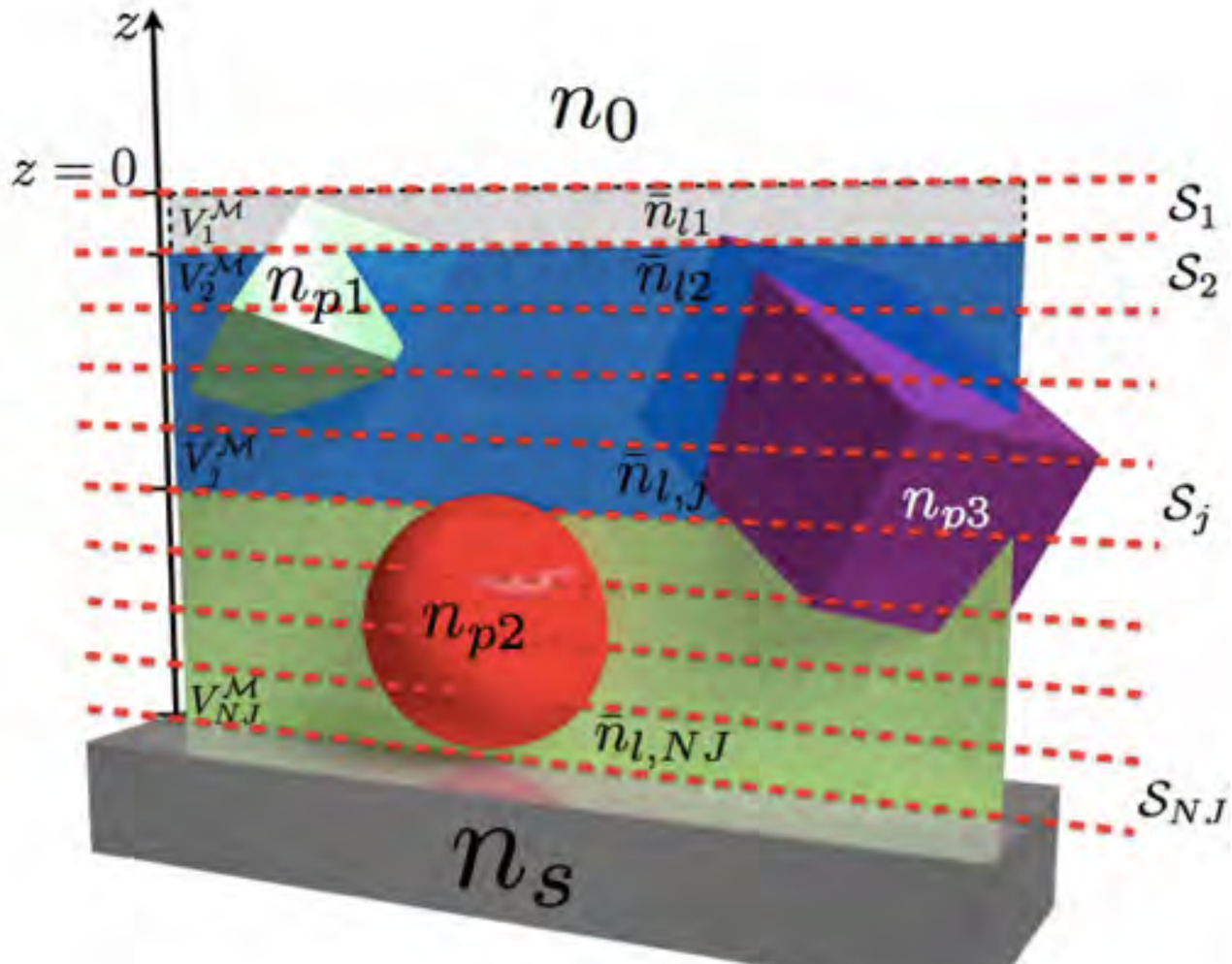
Hexagonally packed spherical nanoparticle assemblies in a block copolymer lamellar film.



incident angle $\alpha_i = 0.12°$ and 14 layers.

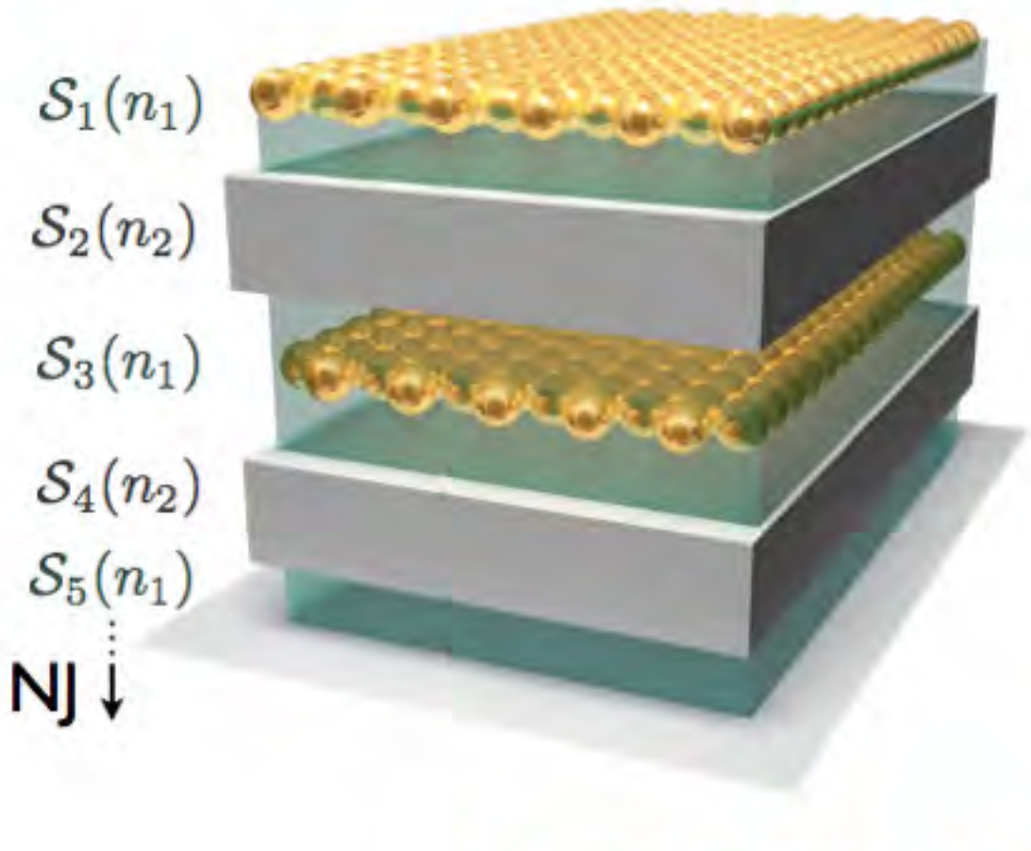

z
n_0
$z=0$
z_{l1}
n_{l1}
h_{l1}
$\mathcal{L}_1$
n_{p1}
$\mathcal{L}_2$
h_{l2}
z_{l2}
n_{l2}
n_{p3}
n_{l3}
n_{p2}
h_{l3}
z_{l3}
$\mathcal{L}_3$
n_s

(a)

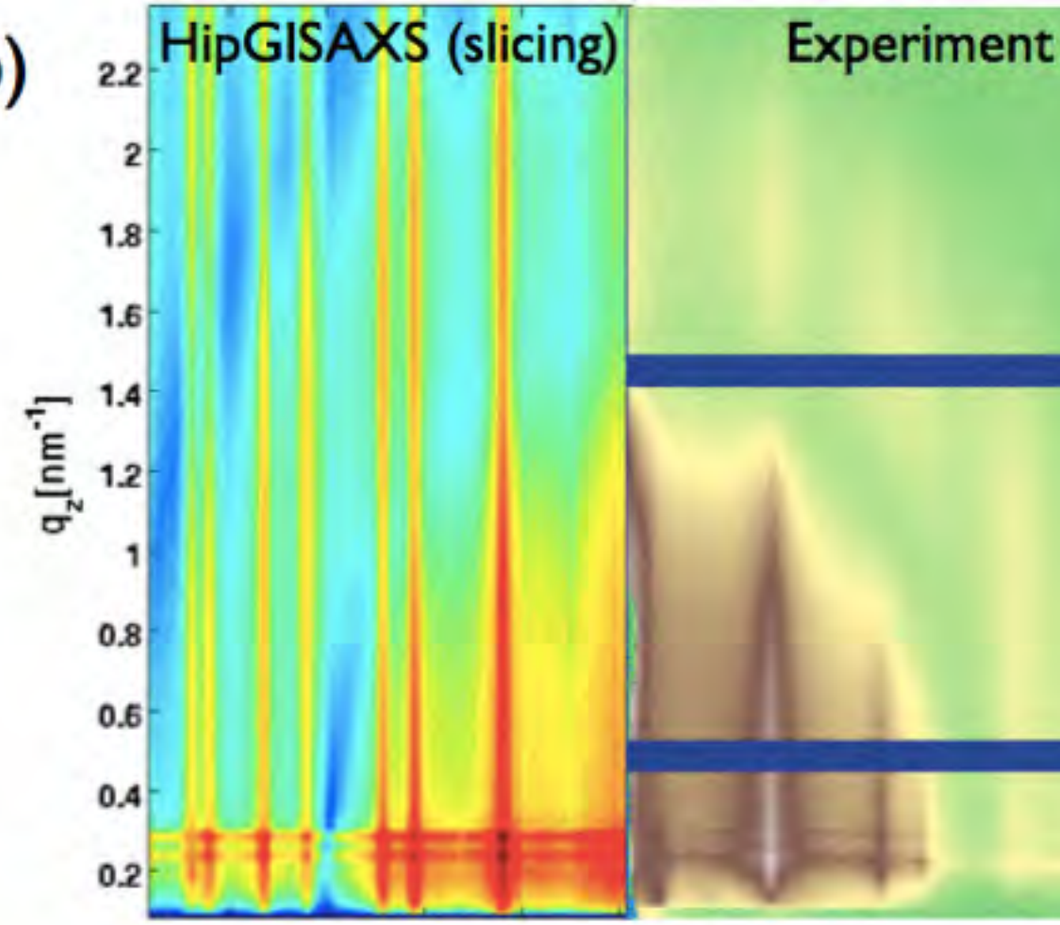

z
n_0
$z=0$
$V_1^{\mathcal{M}}$
$\bar{n}_{l1}$
$\mathcal{S}_1$
$V_2^{\mathcal{M}}$
n_{p1}
$\bar{n}_{l2}$
$\mathcal{S}_2$
$V_j^{\mathcal{M}}$
$\bar{n}_{l,j}$
n_{p3}
$\mathcal{S}_j$
n_{p2}
$V_{NJ}^{\mathcal{M}}$
$\bar{n}_{l,NJ}$
$\mathcal{S}_{NJ}$
n_s

(b)

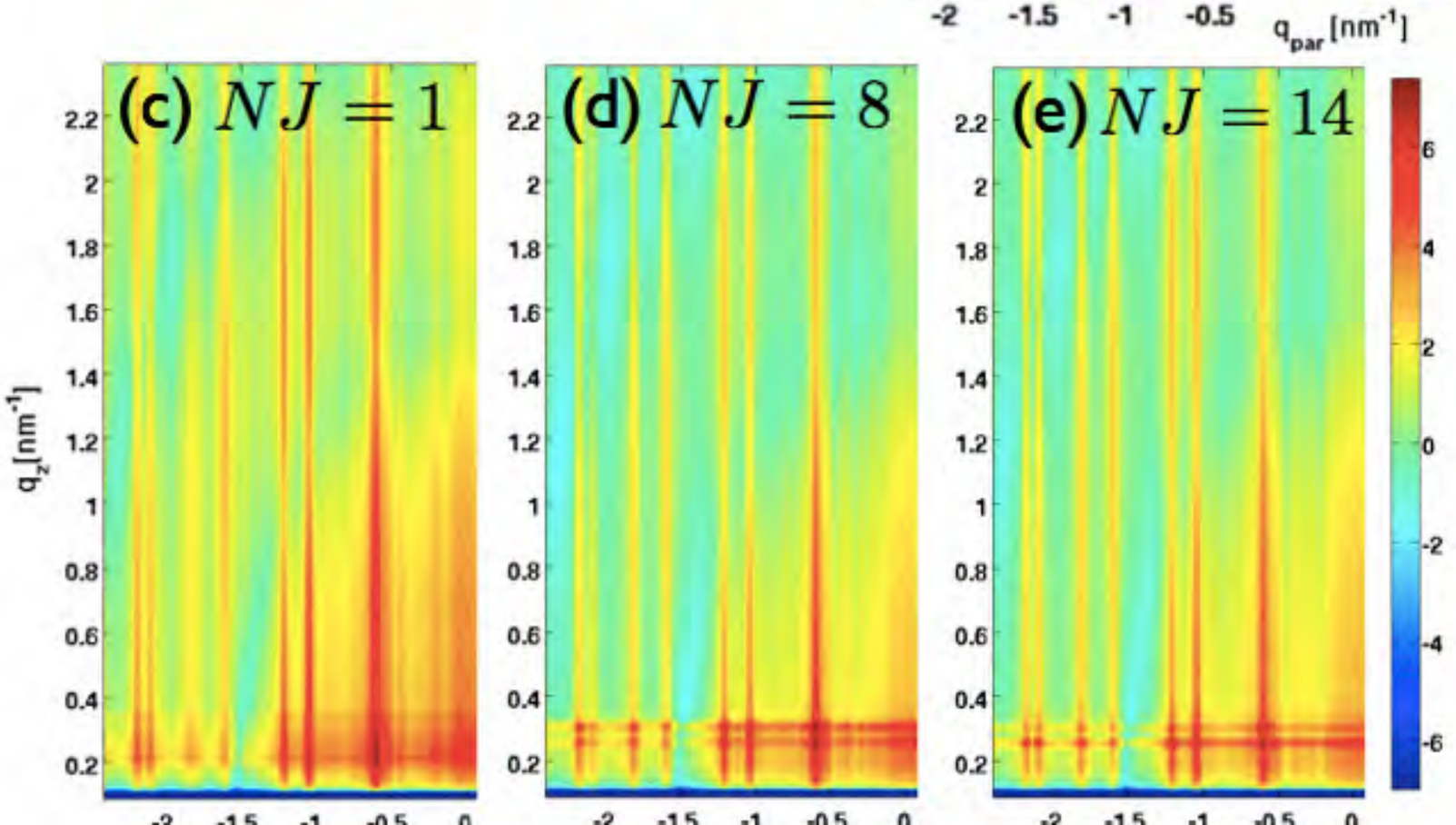
(a)


$\mathcal{S}_1(n_1)$
$\mathcal{S}_2(n_2)$
$\mathcal{S}_3(n_1)$
$\mathcal{S}_4(n_2)$
$\mathcal{S}_5(n_1)$
NJ

(b)

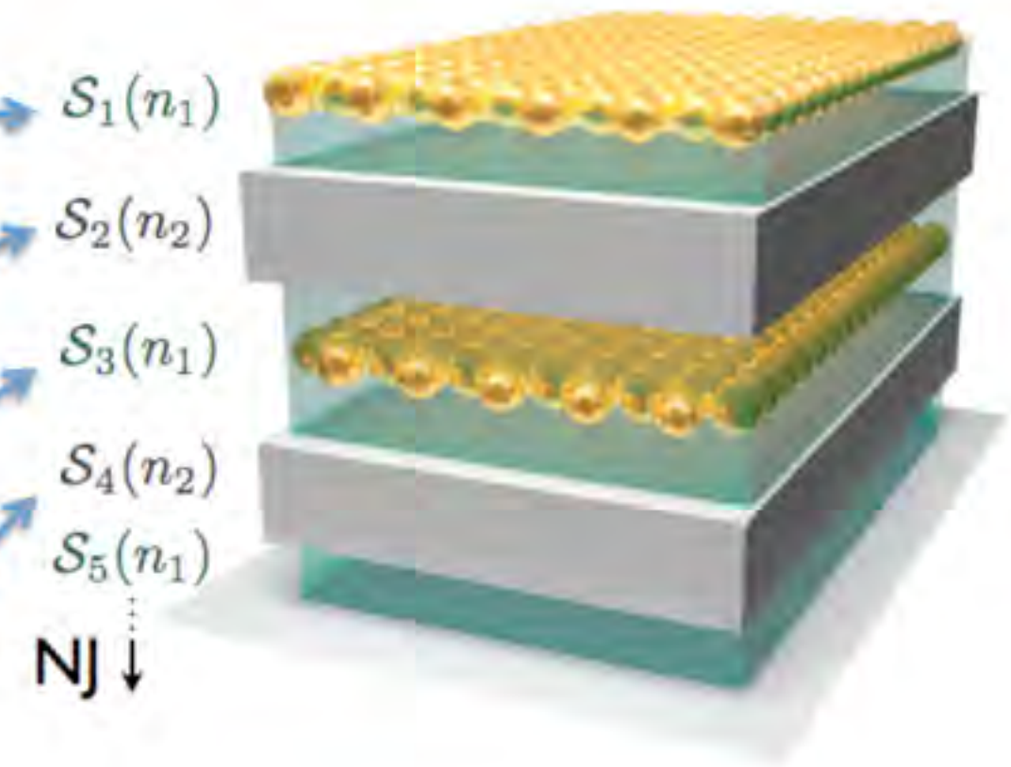

HipGISAXS (slicing)
Experiment
q_z[nm^{-1}]

$\alpha_i = 0.2°$


(c) $NJ = 1$
(d) $NJ = 8$
(e) $NJ = 14$
q_z[nm^{-1}]
q_{par} [nm^{-1}]

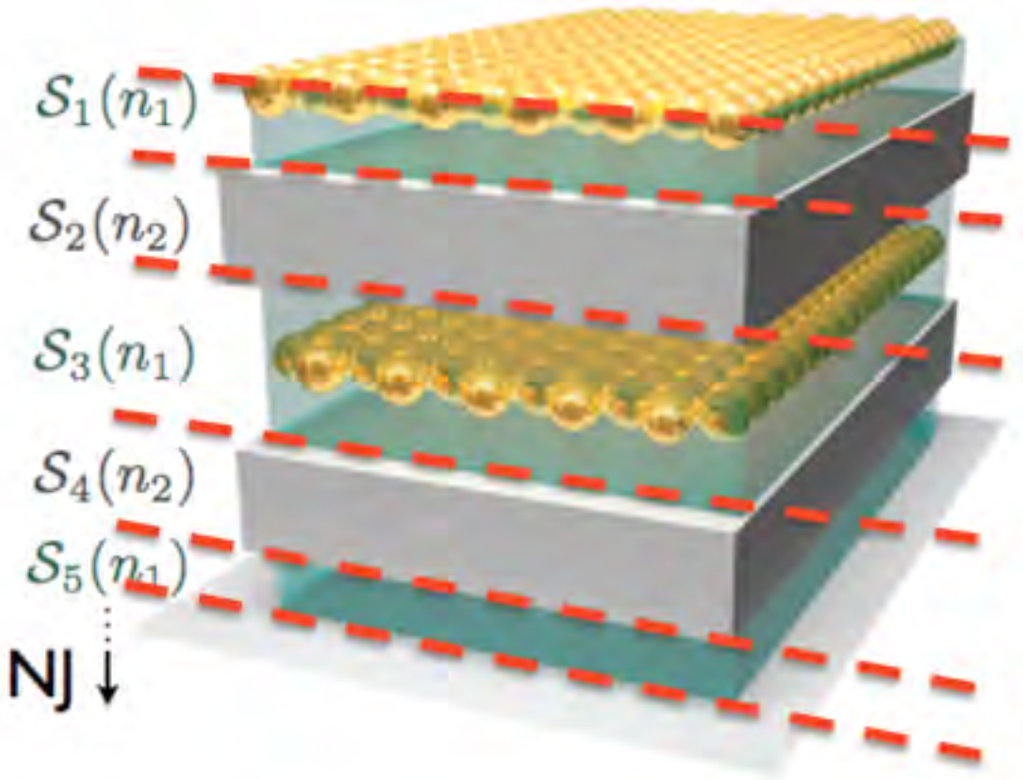
Slicing: Au nanopart. assemblies in PS-*b*-P4VP thin films

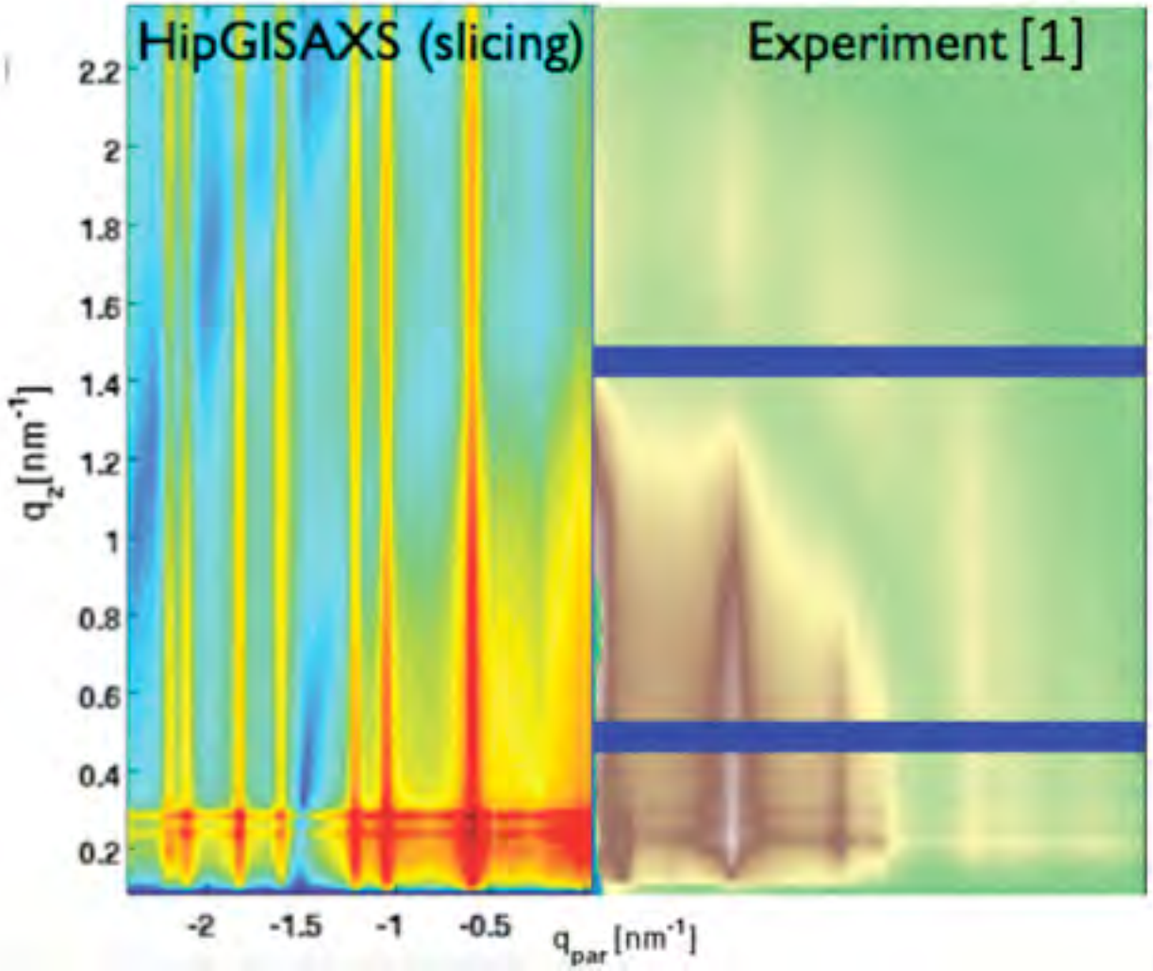
```
hipGisaxsInput = {
shape = {
key = "s1", # a unique key to identify this shape in this file
name = "sphere" , # code name of the shape (see list)
param={ type="radius", min=6.0, max=6.2, stat="gaussian", p1=6.1, p2=0.2, nvalues=10 }
},
layer = {
key = "substr", # special key for case of the substrate layer infinite in lower half space
order = -1 , # special order code for the substrate layer
refindex = { delta=4.88e-06, beta=7.37e-08 } # Si layer
},
layer = {key = "l1",
order = 1 ,
thickness=10,
refindex = { delta=4.87e-06, beta=4.89e-09 } # P4VP layer
},
layer = {key = "l2",
order = 2 ,
thickness=12,
refindex = { delta=2.48e-06, beta=2.25e-09 } # PS layer
},
layer = {key = "l3",
order = 3 ,
thickness=10,
refindex = { delta=4.87e-06, beta=4.89e-09 } # P4VP layer
},
layer = {key = "l4",
order = 4 ,
thickness=12,
refindex = { delta=2.48e-06, beta=2.25e-09 } # PS layer
},
```



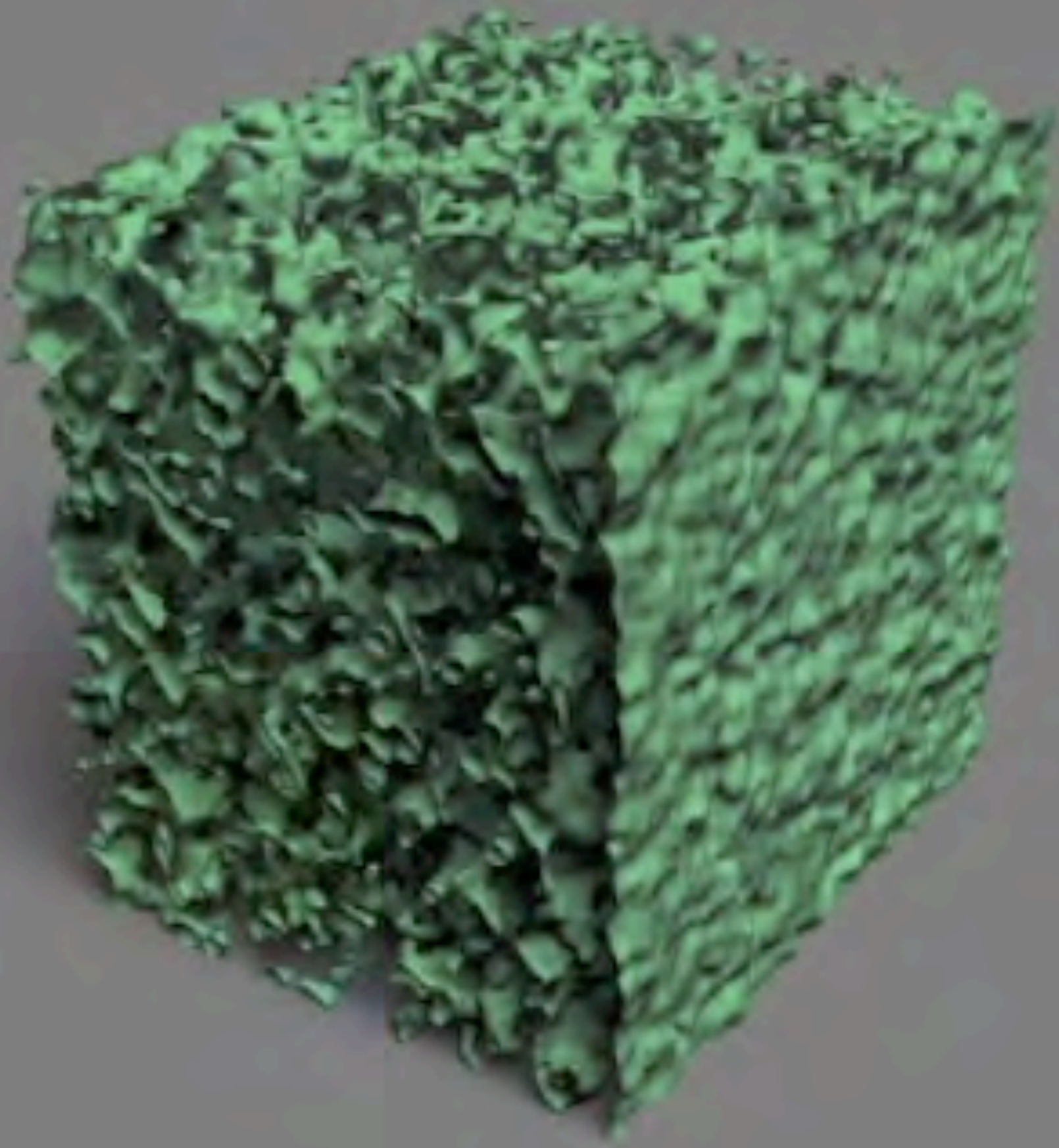
Slicing: Au nanopart. assemblies in PS-*b*-P4VP thin films (Cont'd)

```
instrumentation = {# there is one set of configuration; the elements do not need to appear together
scattering = {
expt = "gisaxs",
alphai ={ min=0.2 }, # incidence angles
photon ={ value=10000, unit="ev" }, # photon energy
},
},
computation = {
method="dwba", # theorical approach (see list)
nslices = 5
}
}
```

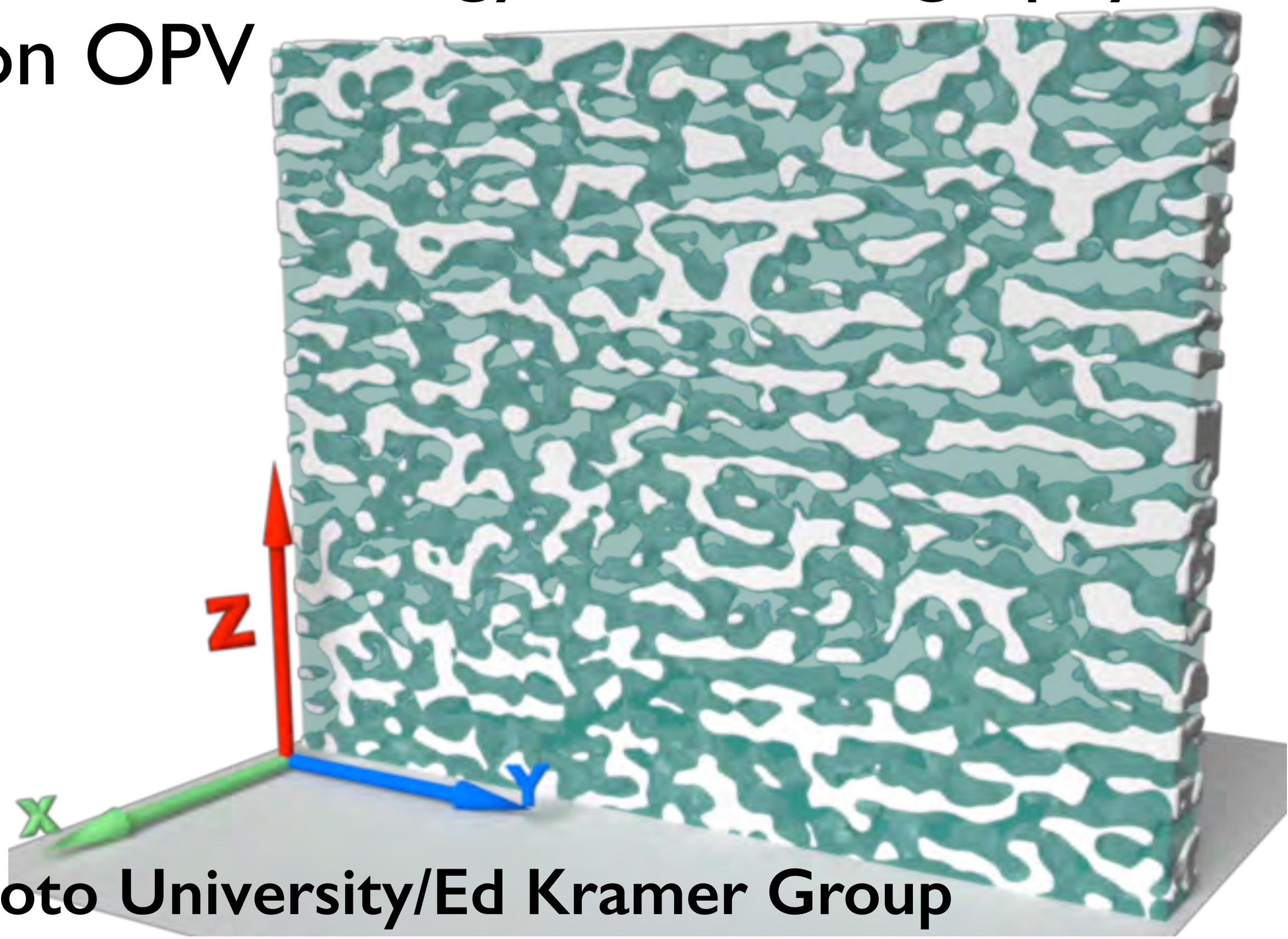




[1] Joseph Kao,et al. *Nano Lett.*, 12:2610–2618 (2012)



Electron energy loss tomography on OPV



Kyoto University/Ed Kramer Group

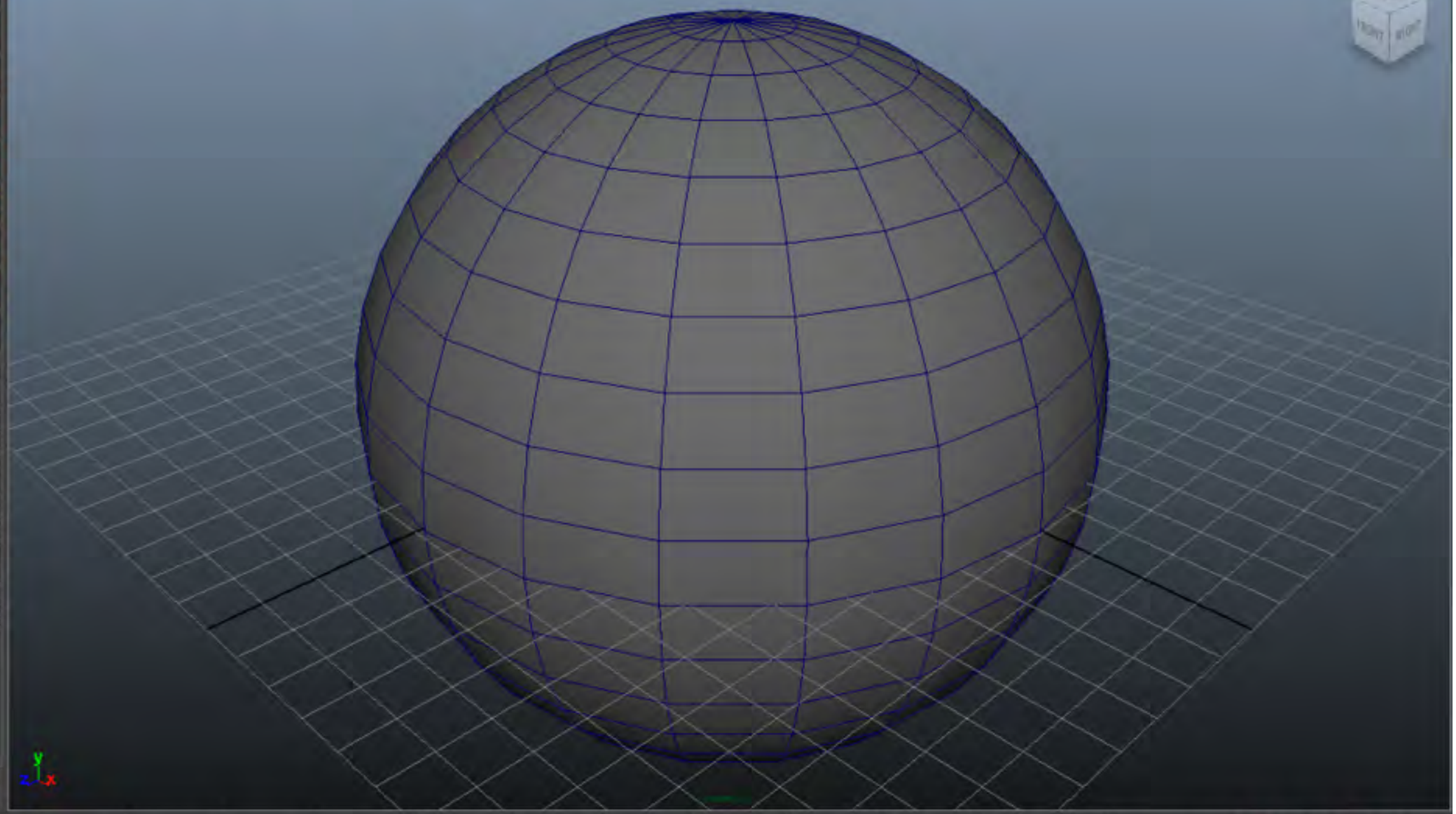
Autodesk Maya 2011 Hotfix 3 x64: untitled*

Polygons

X: Y: Z:

General | Curves | Surfaces | Polygons | Subdivs | Deformation | Animation | Dynamics | Rendering | PaintEffects | Toon | Muscle | Fluids | Fur | Hair | nCloth | Custom

View | Shading | Lighting | Show | Renderer | Panels



1 2 4 6 8 10 12 14 16 18 20 22 24 1.00

1.00 1.00 1 24 24.00 48.00 No Anim Layer No Character Set

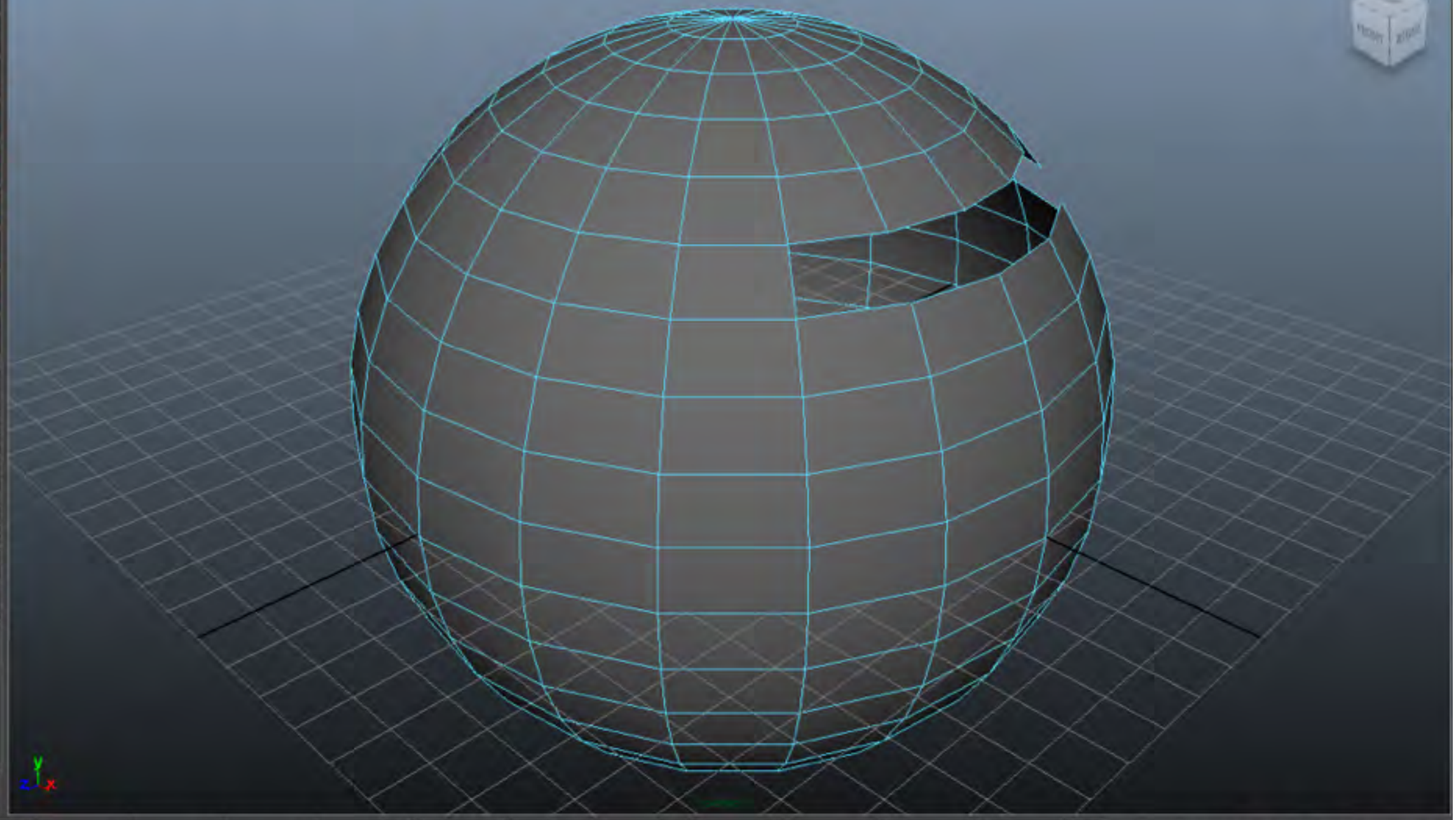
MEL

Autodesk Maya 2011 Hotfix 3 x64: untitled*

Polygons

General Curves Surfaces Polygons Subdivs Deformation Animation Dynamics Rendering PaintEffects Toon Muscle Fluids Fur Hair nCloth Custom

View Shading Lighting Show Renderer Panels



1 2 4 6 8 10 12 14 16 18 20 22 24 1.00

1.00 1.00 1 24 24.00 48.00 No Anim Layer No Character Set

MEL

Select handle objects (RMB for more info)

Form factor

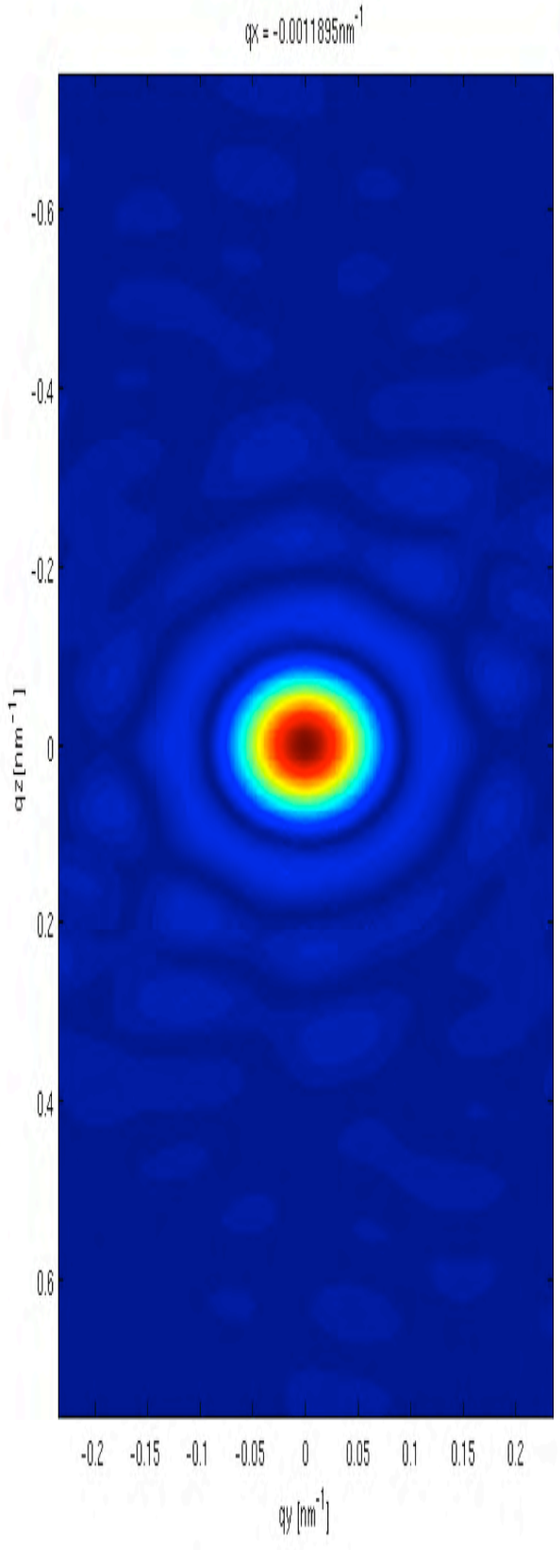
$$F(q) = \int_v e^{i\, q\cdot r}\, d^3 r$$

$$F(q) = -\frac{1}{q^2}\int_s \frac{\partial e^{i\, q\cdot r}}{\partial n} d^2 r$$ Green' theorem

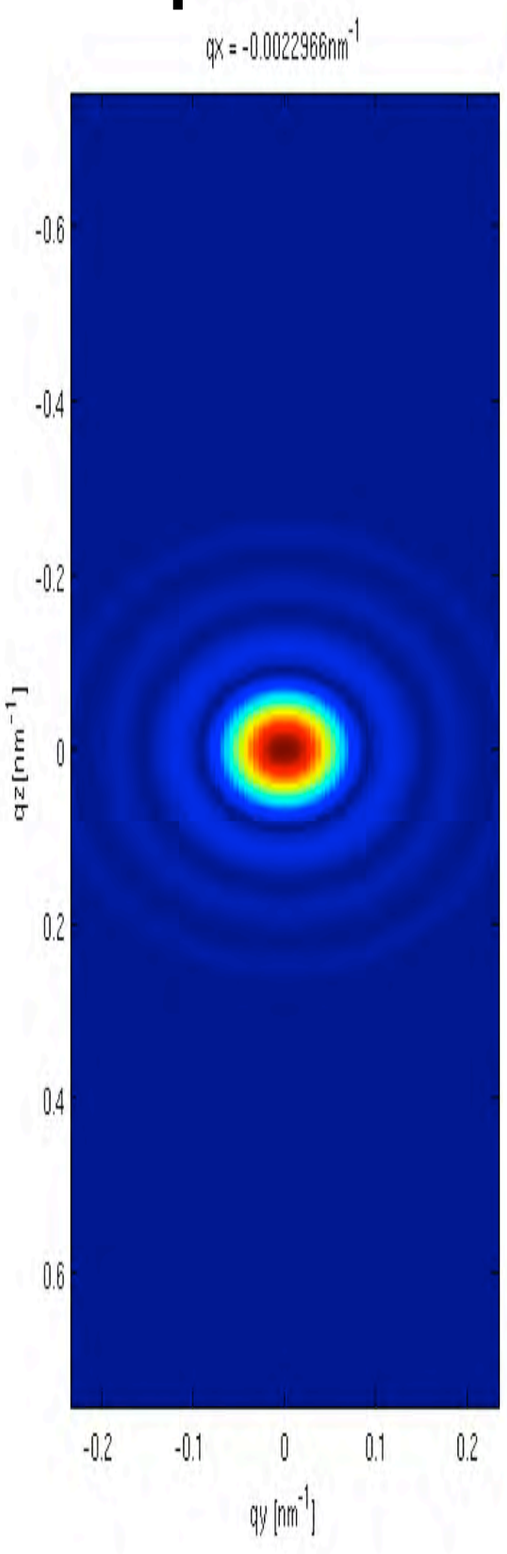
$$\approx -\frac{i}{q^2}\sum_{t=1}^{NTr} \vec{q_{nt}} e^{i\vec{q}.\vec{r_t}}\, s_t$$

do what we can analytical

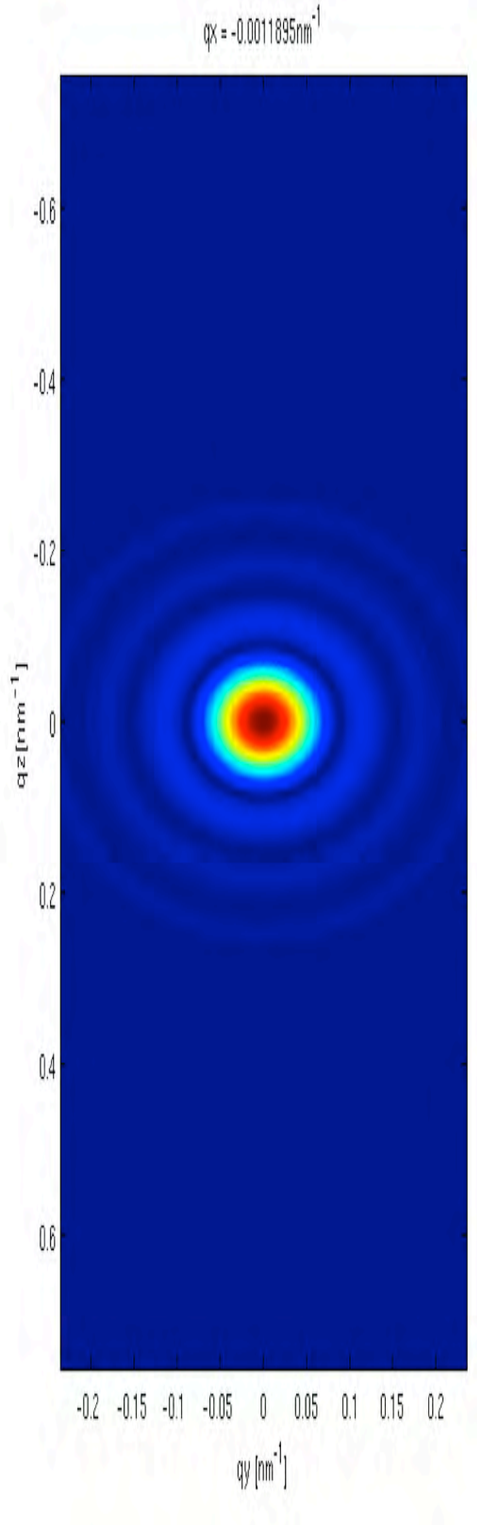
Sphere Form Factor



Low resolution:
40 triangles

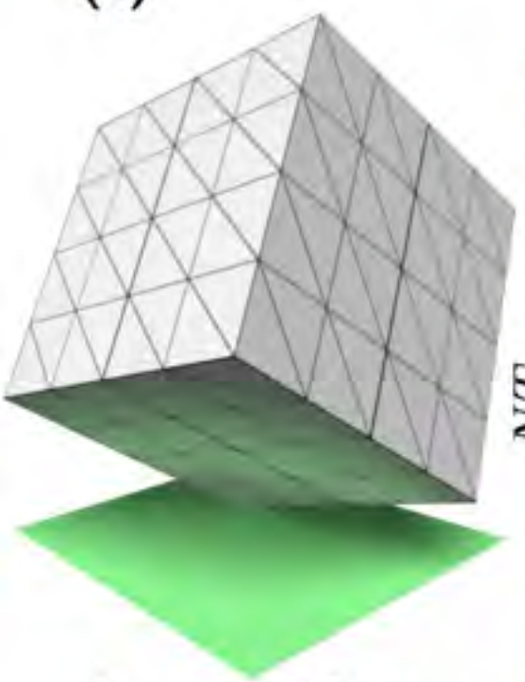


High resolution:
1200 triangles

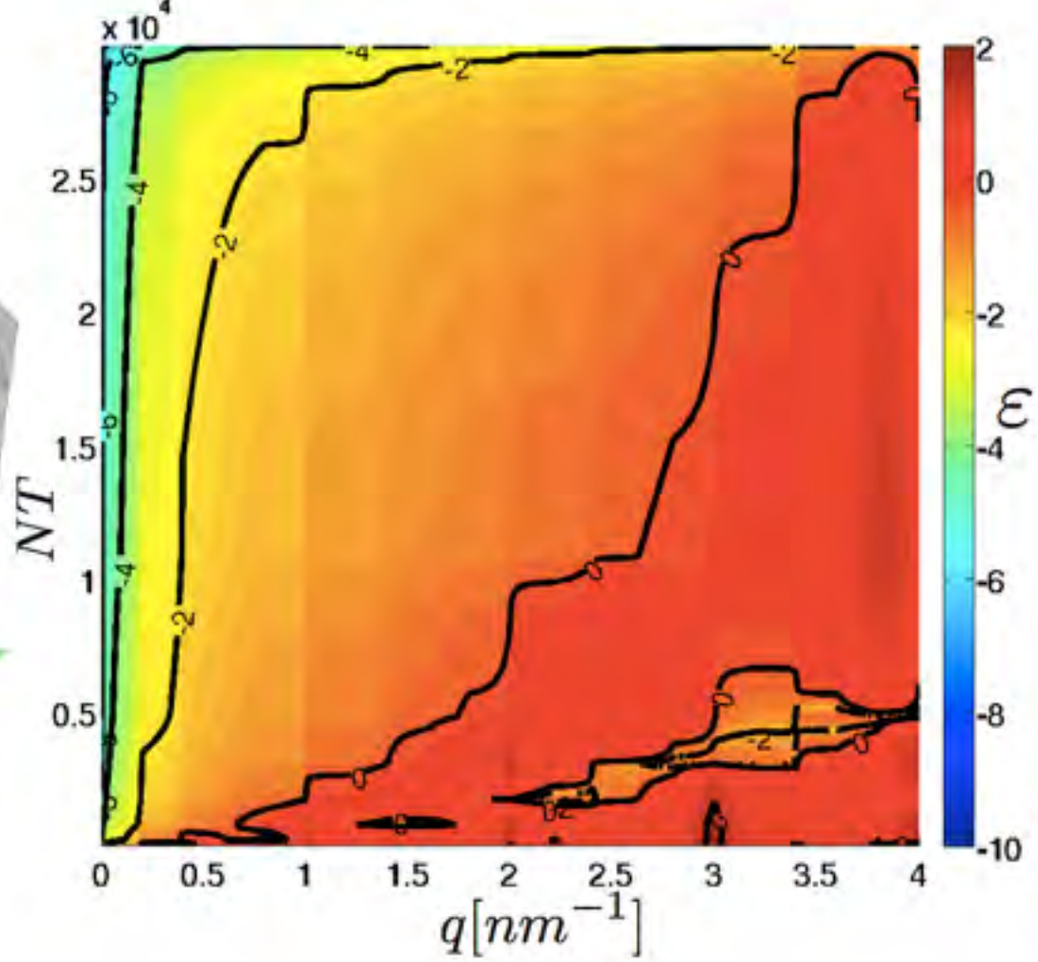


Analytical

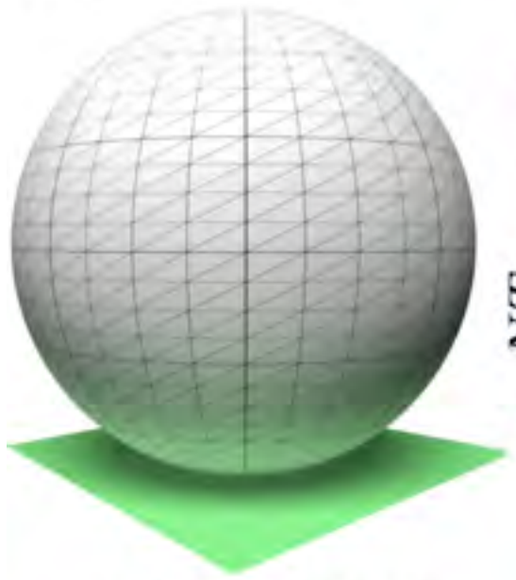
(a)



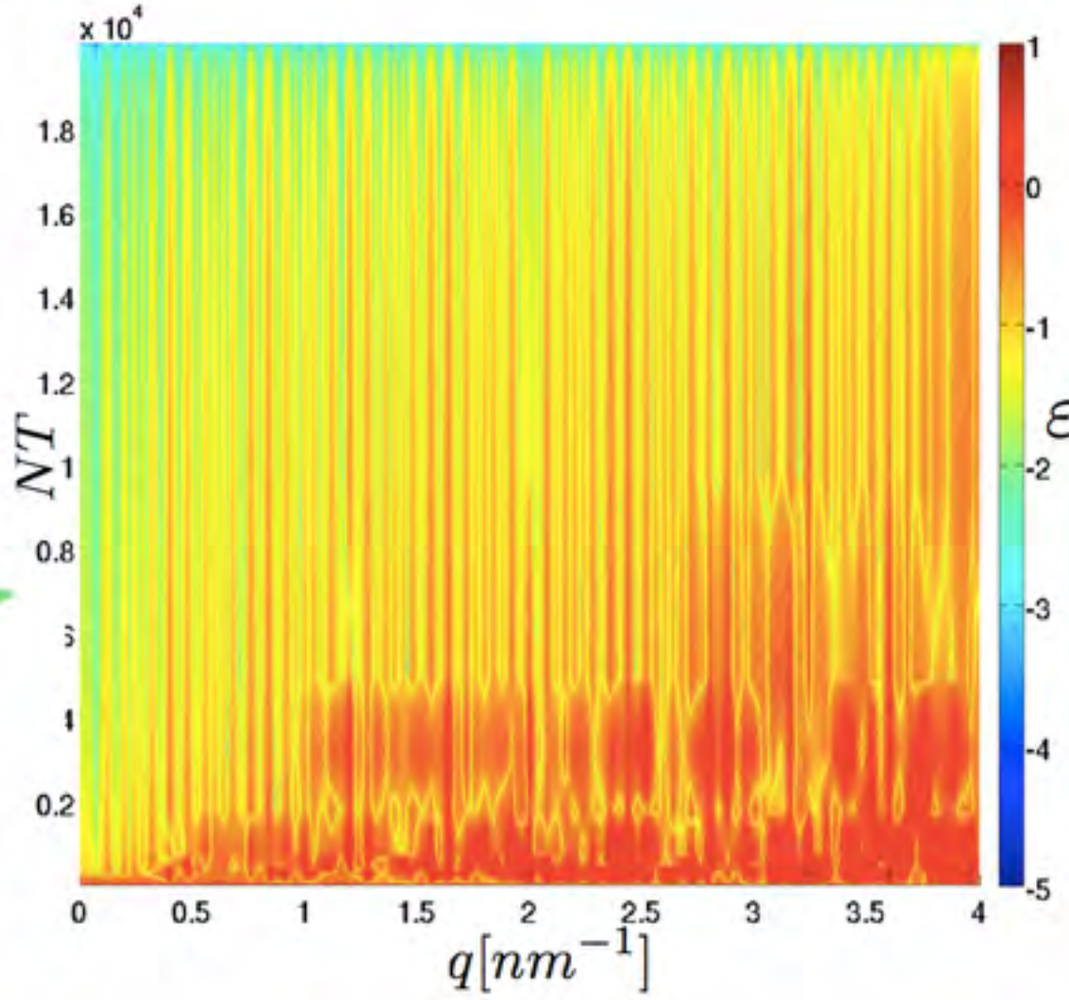
length=50nm

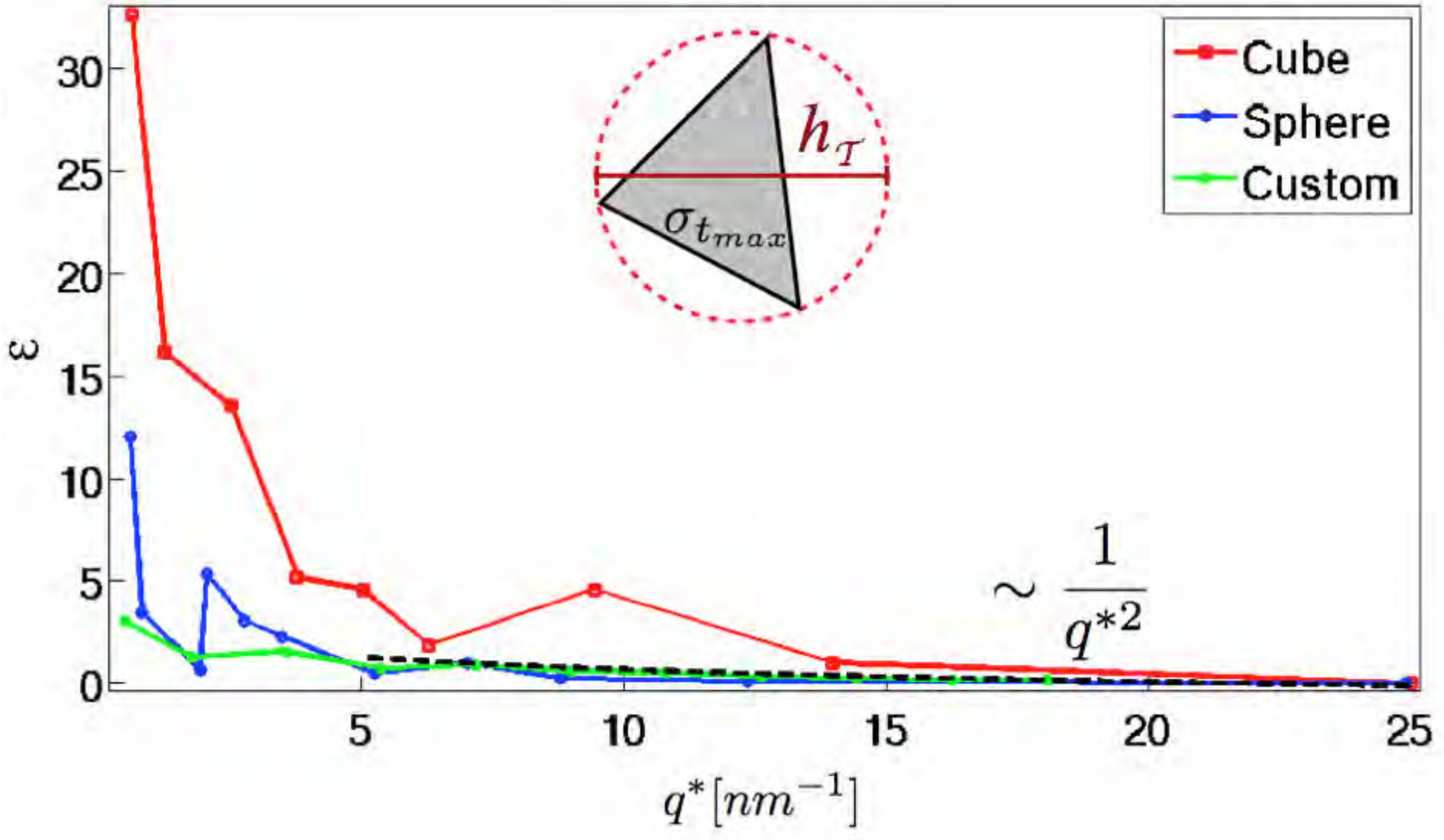


Error of triangulation versus analytical



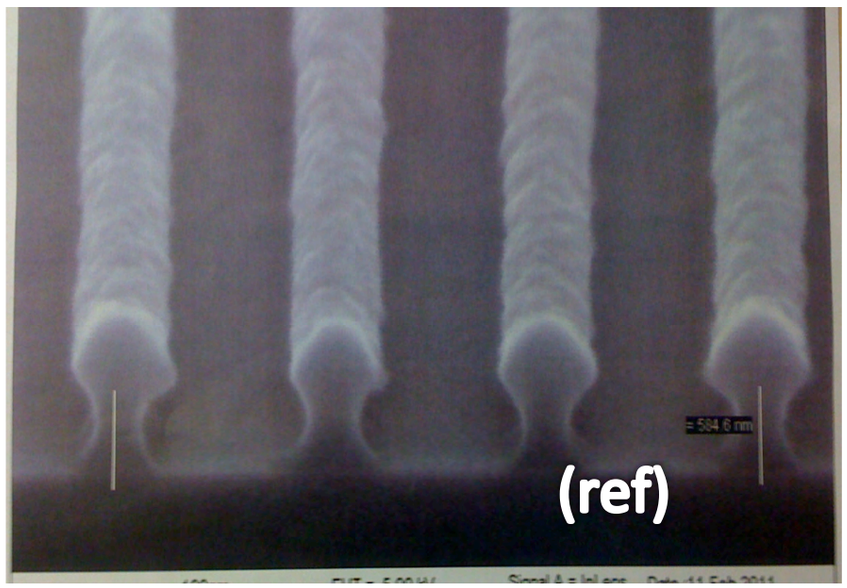
raduis=25nm



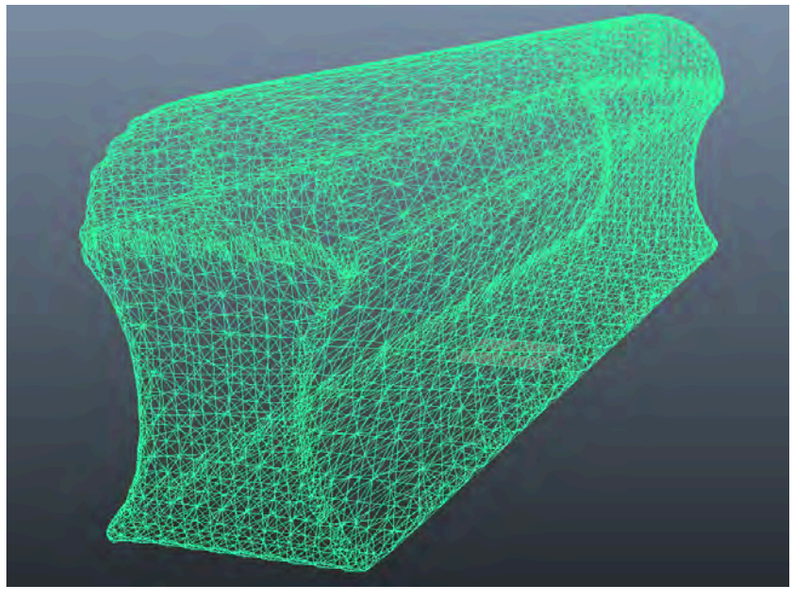

Cube
Sphere
Custom
h_T
$\sigma_{t_{max}}$
ω
30
25
20
15
10
5
0
5
10
15
20
25
$\sim \frac{1}{q^{*2}}$
$q^*[nm^{-1}]$

… More complex morphologies

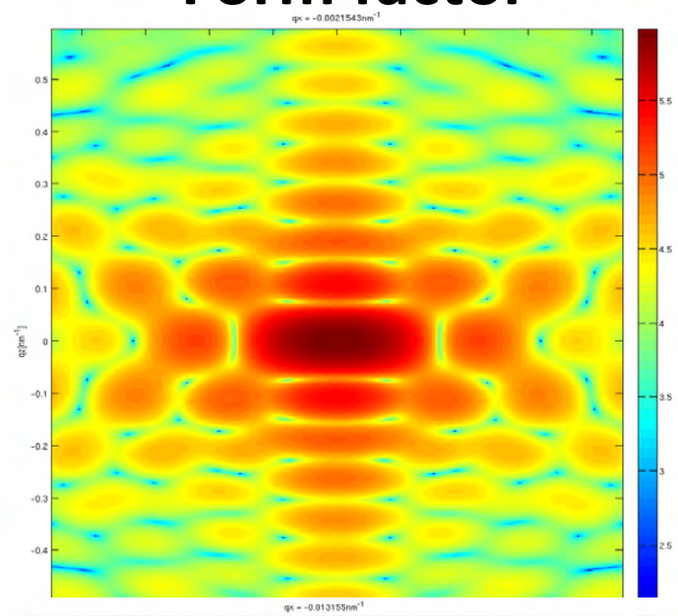
Actual sample

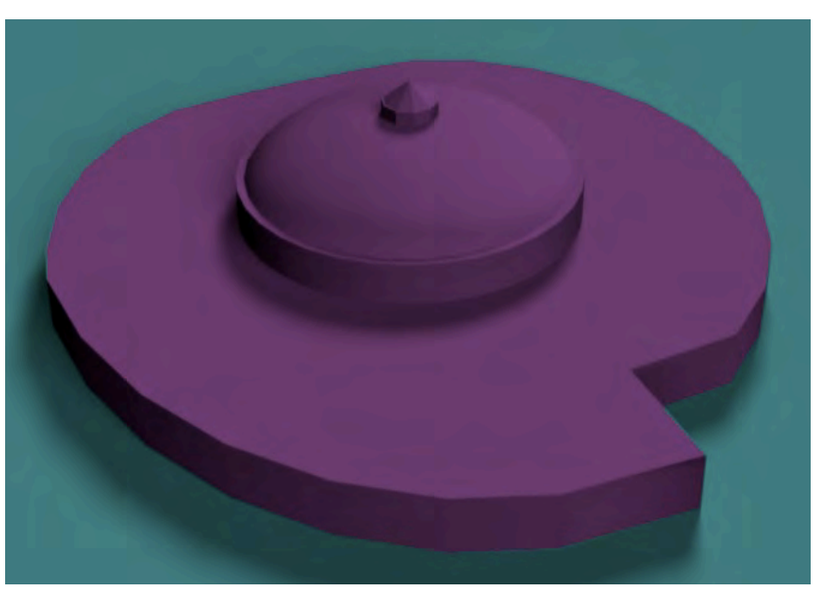


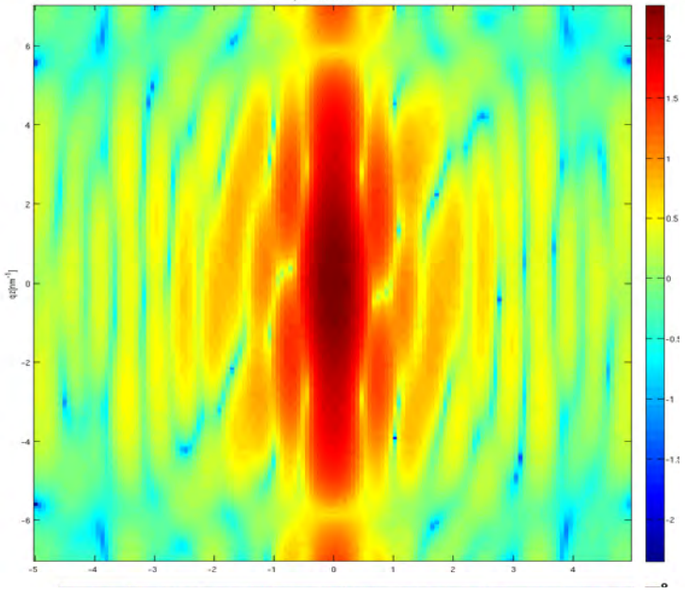
Generated discrete shape model



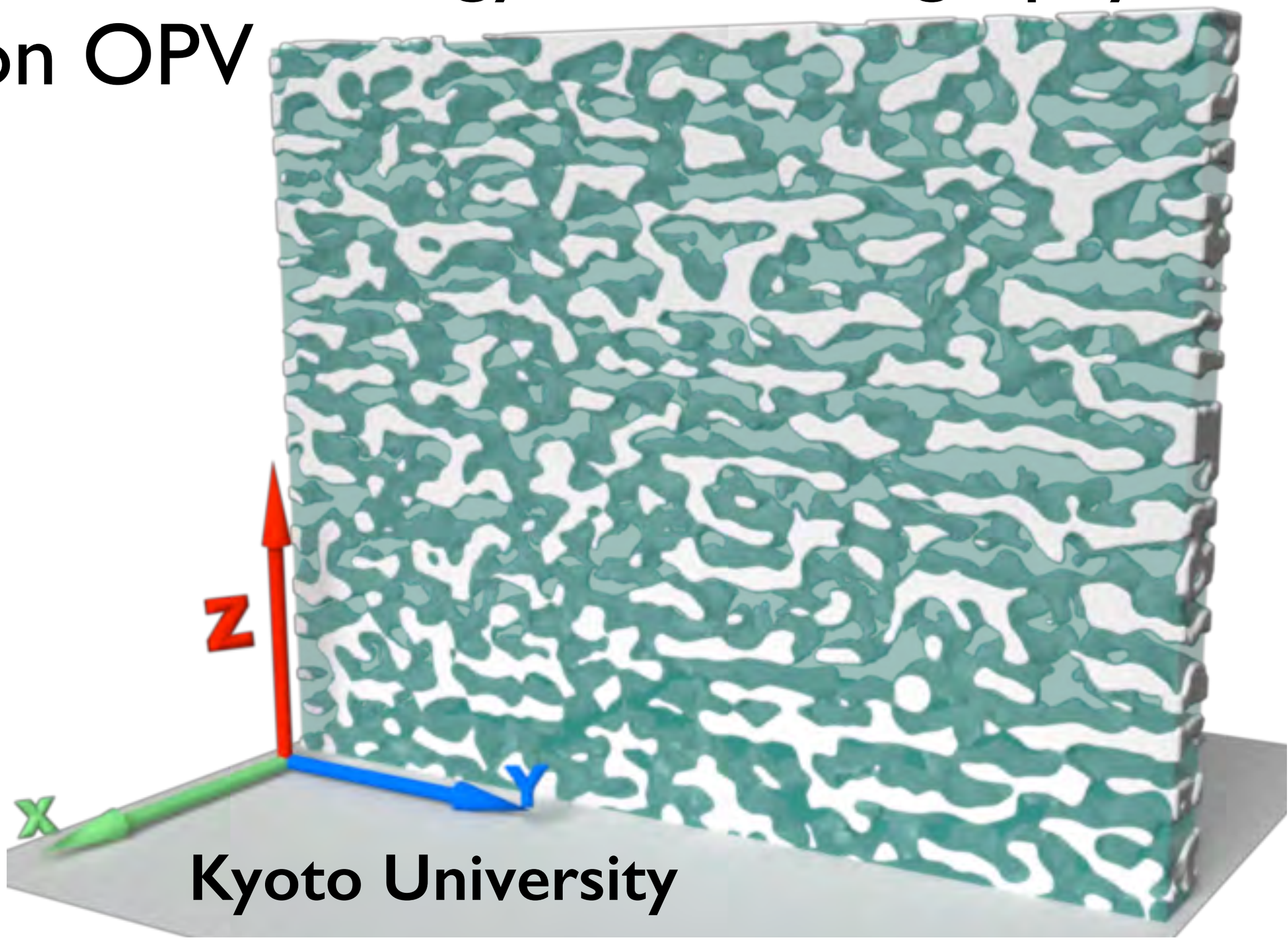
Form factor





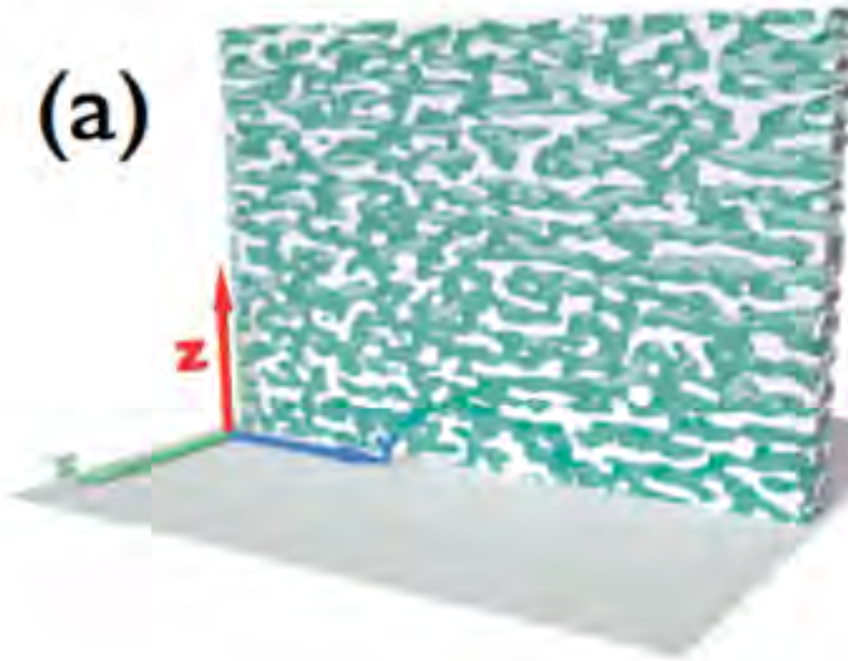


Electron energy loss tomography on OPV



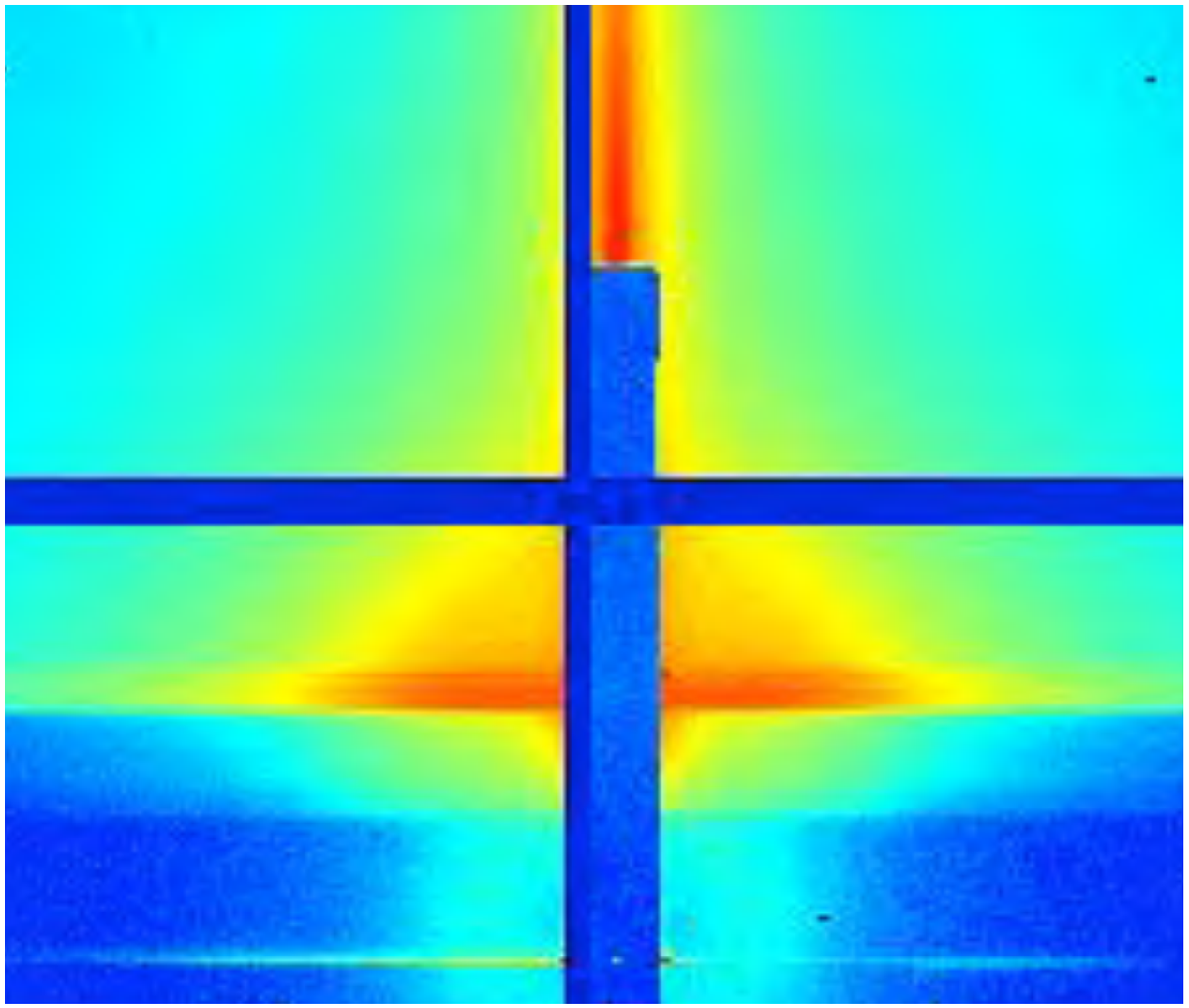
Kyoto University

Custom shape: OPV sample

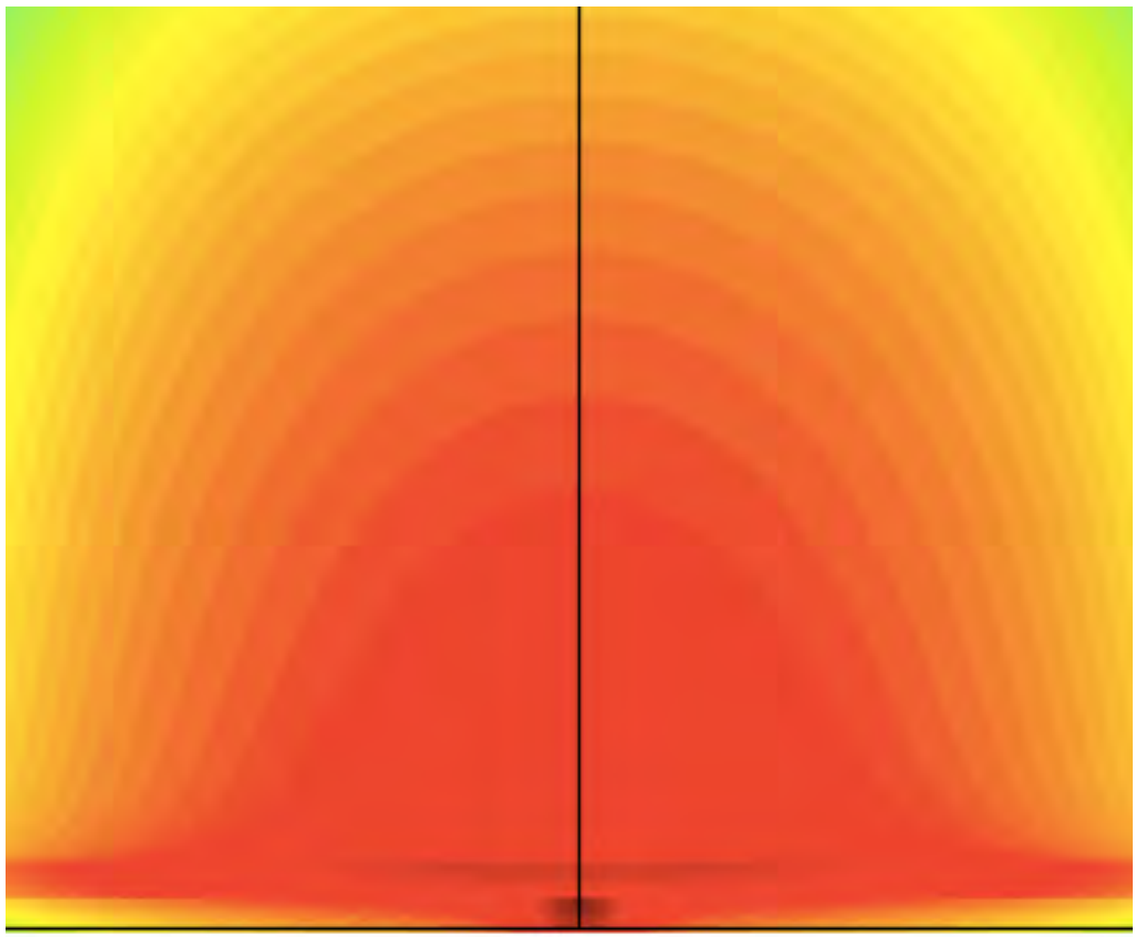



```
hipGisaxsInput = {
shape = {
key = "s1", # a unique key to identify this shape in this file
name = "opv.hdf5" , # code name of the shape (see list)
},
layer = {
key = "substr", # special key for case of the substrate layer infinite in lower half space
order = -1 , # special order code for the substrate layer
refindex = { delta=4.88e-06, beta=7.37e-08 }
}
layer = {
key = "l1", # a unique key
order = 1, # order of this layer (top = 1 to bottom);
thickness = 800, # layer thickness in nm
refindex = { delta = 4.8e-06, beta = 4.2e-08 } # layer refractive index parameters
},
```

GISAXS from sample set



GISAXS simulation from tomography set



q_{par}

MacPyMOL

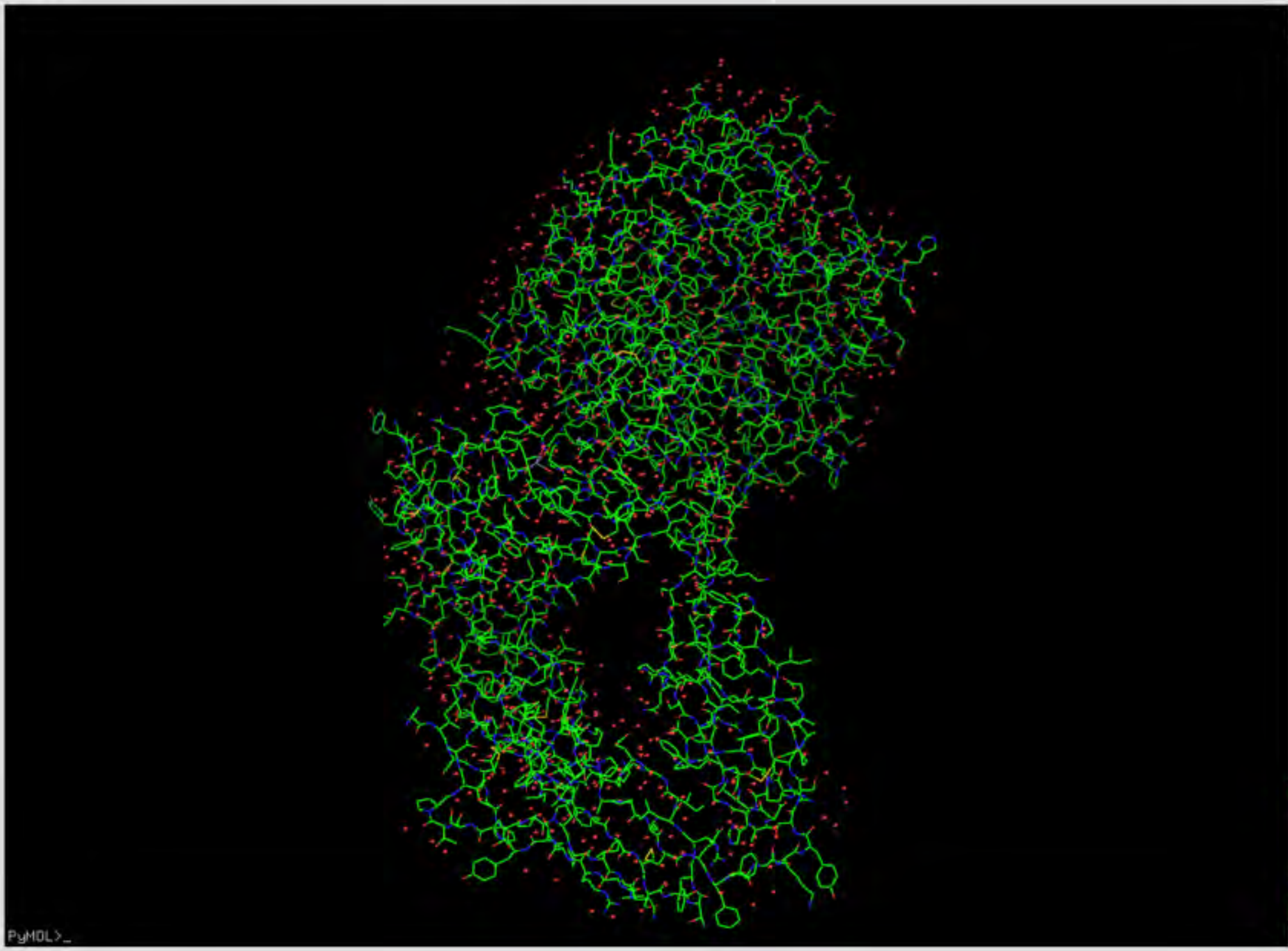
```
COMPND    5 MUTATION: YES;
COMPND    6 MOL_ID: 2;
COMPND    7 MOLECULE: CR2/CD121/C3D/EPSTEIN-BARR VIRUS RECEPTOR;
COMPND    8 CHAIN: B, C;
COMPND    9 FRAGMENT: SEQUENCE DATABASE RESIDUES 21-153
 ObjectMolecule: Read secondary structure assignments.
 ObjectMolecule: Read crystal symmetry information.
 Symmetry: Found 18 symmetry operators.
 CmdLoad: "/Users/alexander_hexemer/Downloads/pdb1ghq.ent" loaded as "pdb1ghq".
```

Reset | Zoom | Draw | Ray | Rock

Unpick | Hide Sele | Get View

|< | < | Stop | Play | > | >| | MClear

PyMOL>



all A S H L C
pdb1ghq A S H L C

Mouse Mode 3-Button Viewing
Buttons L M R Wheel
Keys Rota Move MovZ Slab
ShFt +Box -Box Clip MovS
Ctrl +/- PkAt Pk1 MvSZ
CtSh Sele Orig Clip MovZ
SnglClk +/- Cent Menu
DblClk Menu - PkAt
Selecting Residues
Frame [1/ 1] 25/sec

MacPyMOL

```
COMPND    5 MUTATION: YES;
COMPND    6 MOL_ID: 2;
COMPND    7 MOLECULE: CR2/CD121/C3D/EPSTEIN-BARR VIRUS RECEPTOR;
COMPND    8 CHAIN: B, C;
COMPND    9 FRAGMENT: SEQUENCE DATABASE RESIDUES 21-153
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 ObjectMolecule: Read crystal symmetry information.
 Symmetry: Found 18 symmetry operators.
 CmdLoad: "/Users/alexander_hexemer/Downloads/pdb1ghq.ent" loaded as "pdb1ghq".
```

Reset | Zoom | Draw | Ray | Rock

Unpick | Hide Sele | Get View

|< | < | Stop | Play | > | >| | MClear

PyMOL>

```
save myscene.obj
```

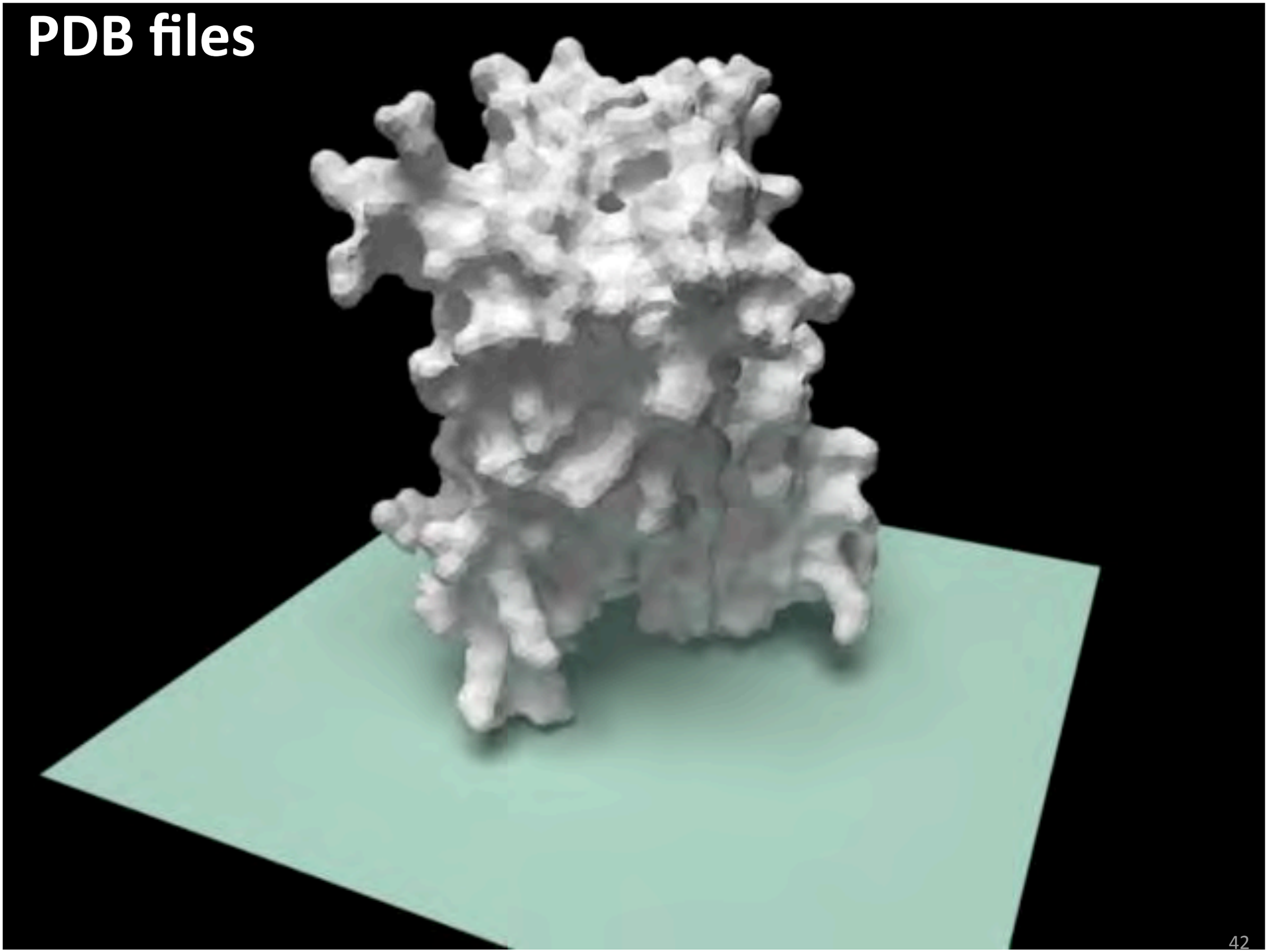
all A S H L C

pdb1ghq A S H L C

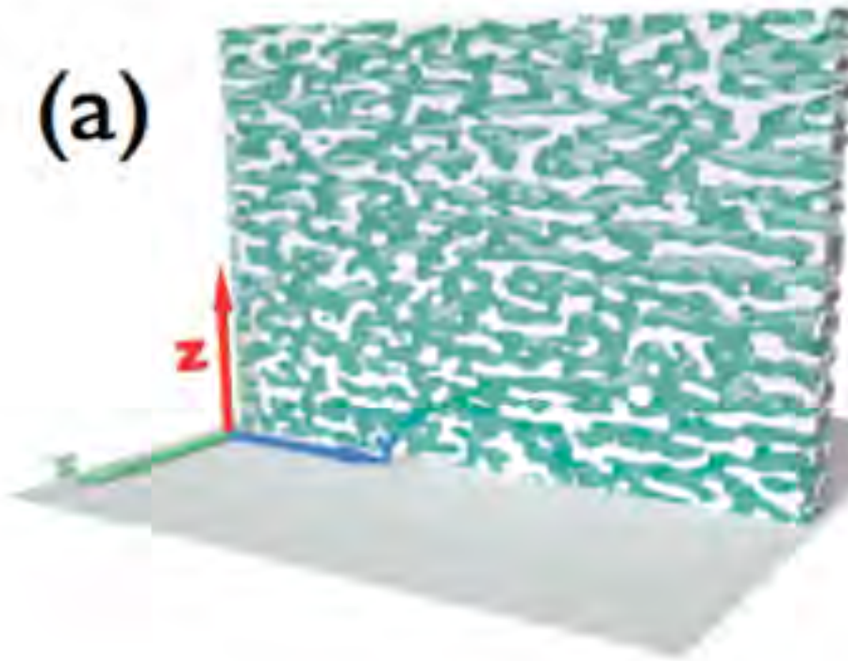
Mouse Mode 3-Button Viewing
Buttons L M R Wheel
& Keys Rota Move MovZ Slab
Shft +Box -Box Clip MovS
Ctrl +/- PkAt Pk1 MvSZ
CtSh Sele Orig Clip MovZ
SnglClk +/- Cent Menu
DblClk Menu - PkAt
Selecting Residues
Frame [1/ 1] 0/sec

PyMOL>_

PDB files



Custom shape: OPV sample




```
hipGisaxsInput = {
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name = "opv.hdf5" , # code name of the shape (see list)
},
layer = {
key = "substr", # special key for case of the substrate layer infinite in lower half space
order = -1 , # special order code for the substrate layer
refindex = { delta=4.88e-06, beta=7.37e-08 }
}
layer = {
key = "l1", # a unique key
order = 1, # order of this layer (top = 1 to bottom);
thickness = 800, # layer thickness in nm
refindex = { delta = 4.8e-06, beta = 4.2e-08 } # layer refractive index parameters
},
```
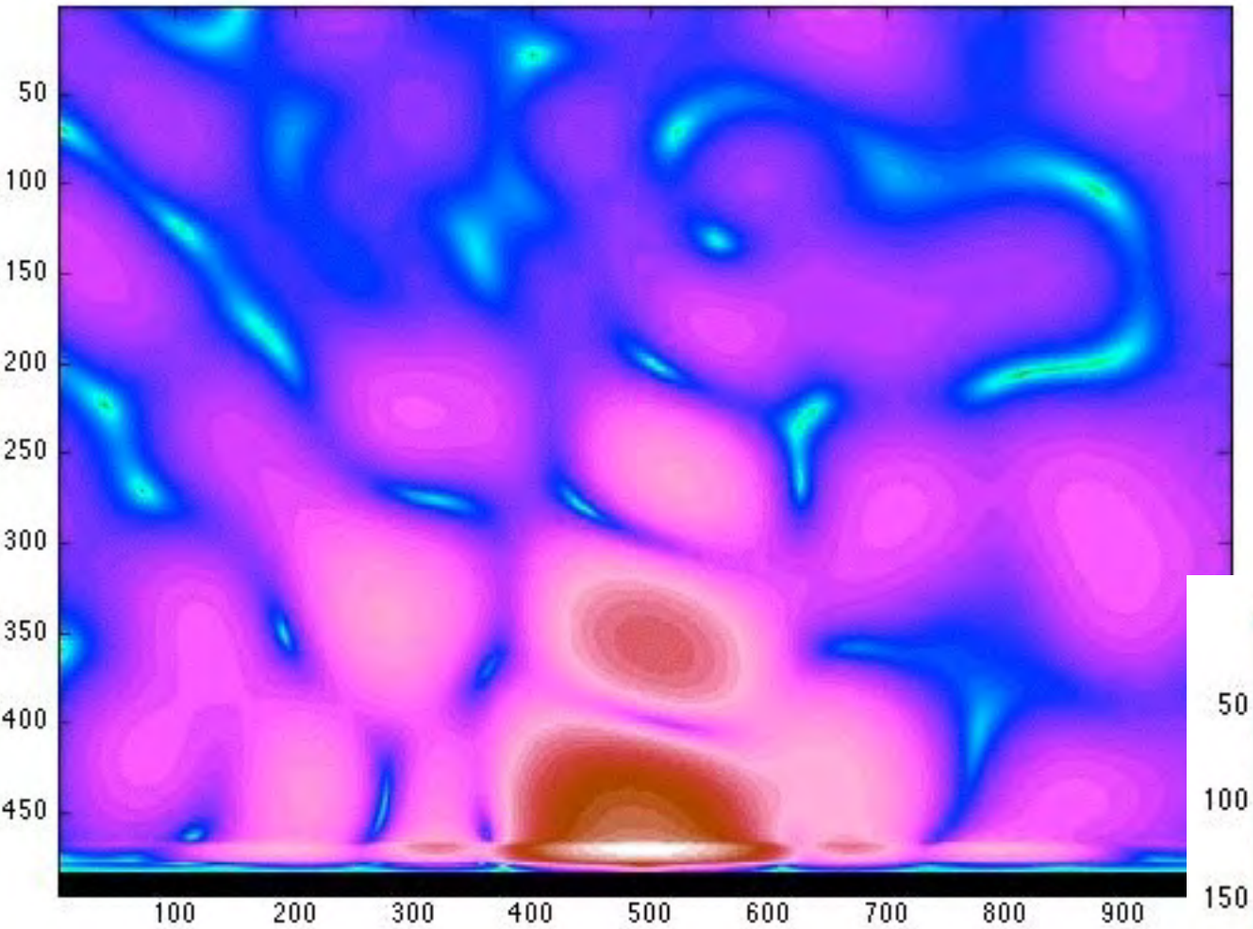
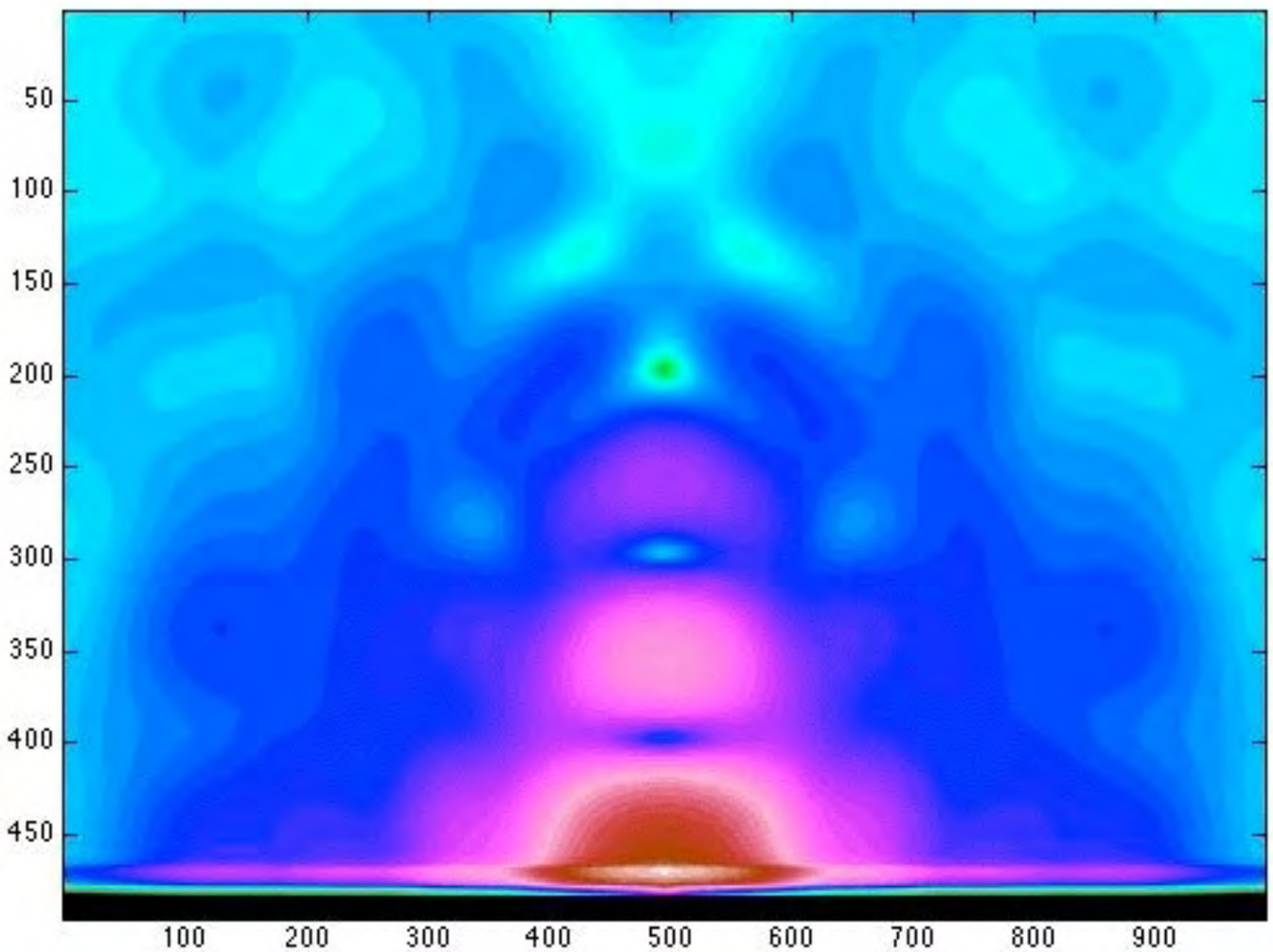
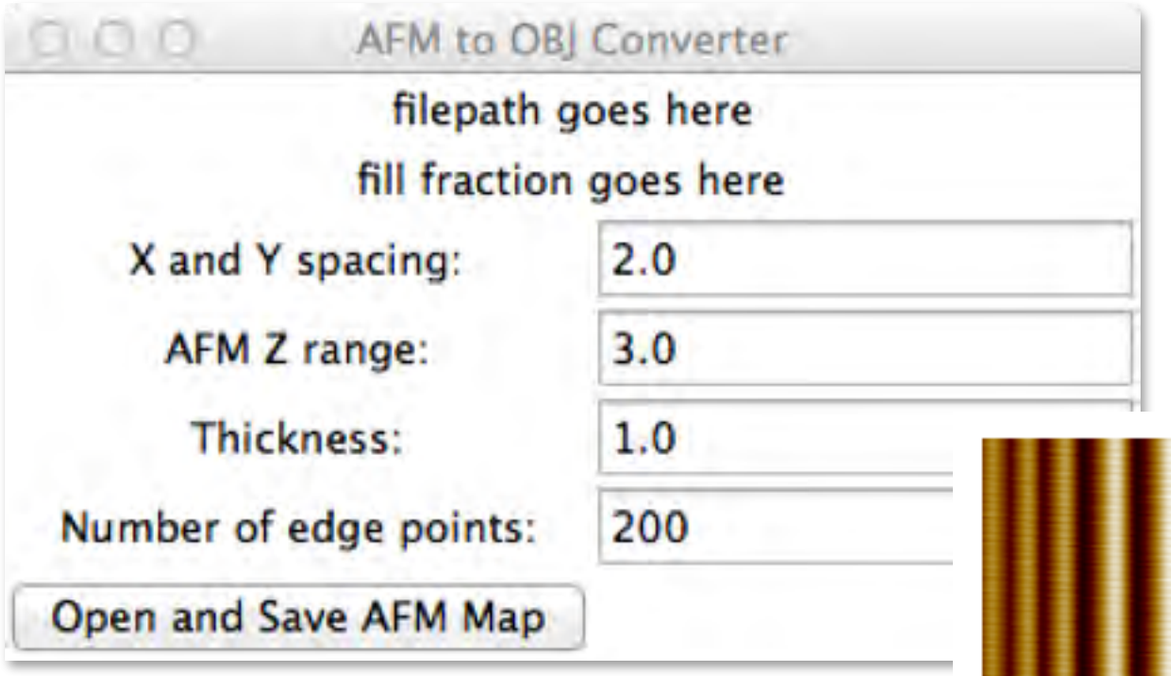

Single protein single orientation inside homogeneous membrane



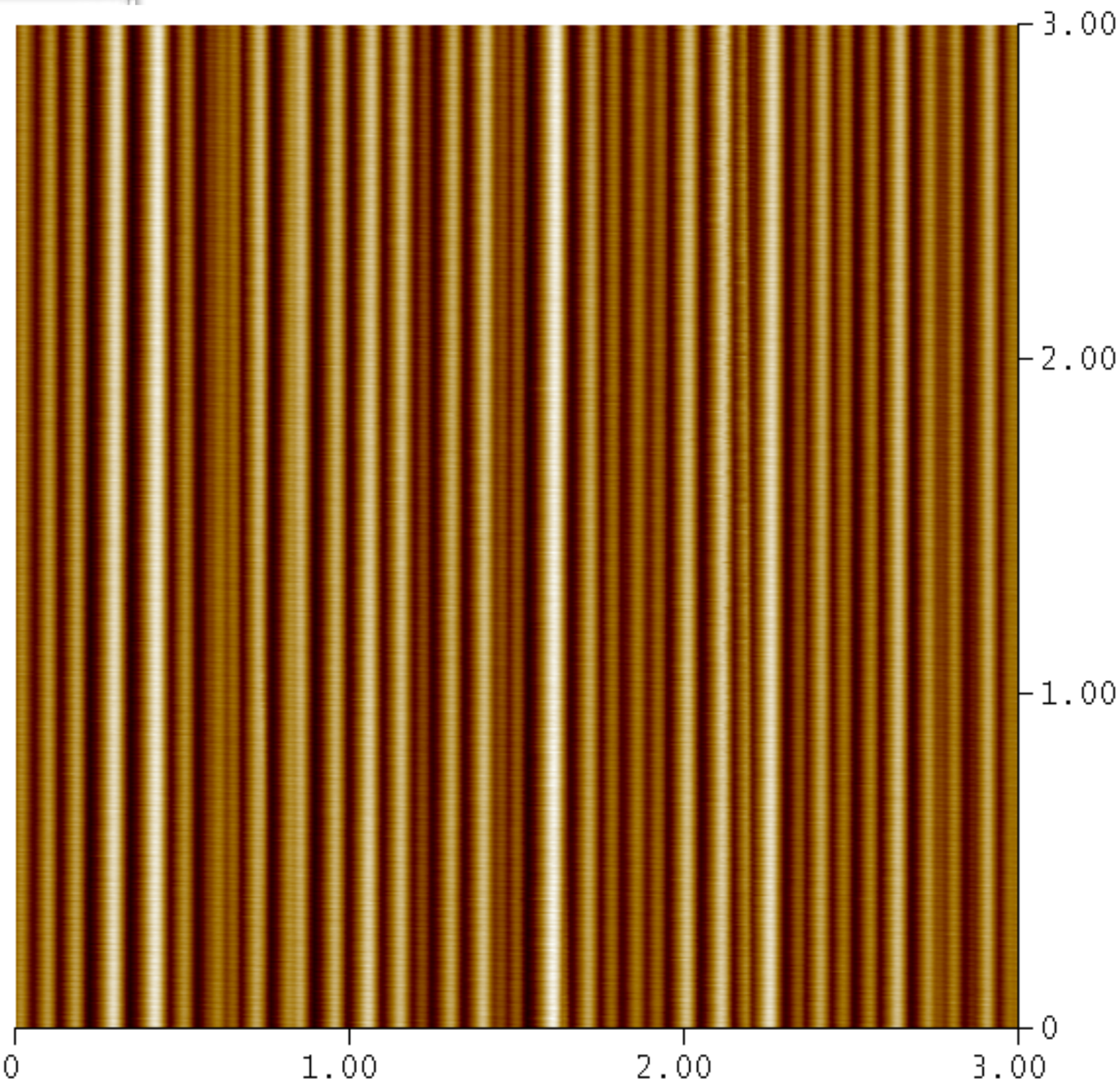
Single protein radial averaged orientation inside homogeneous membrane



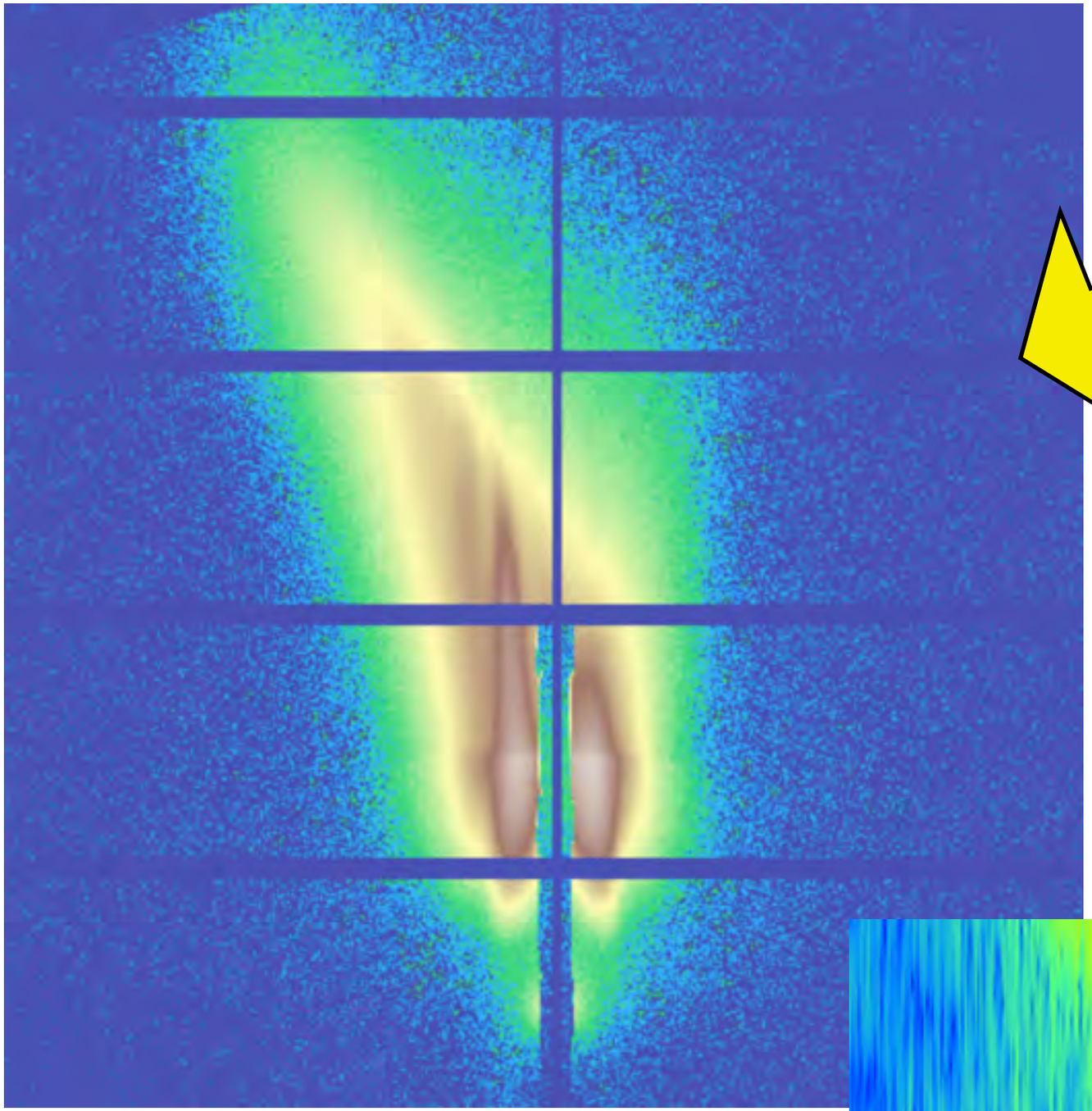
Combine different techniques

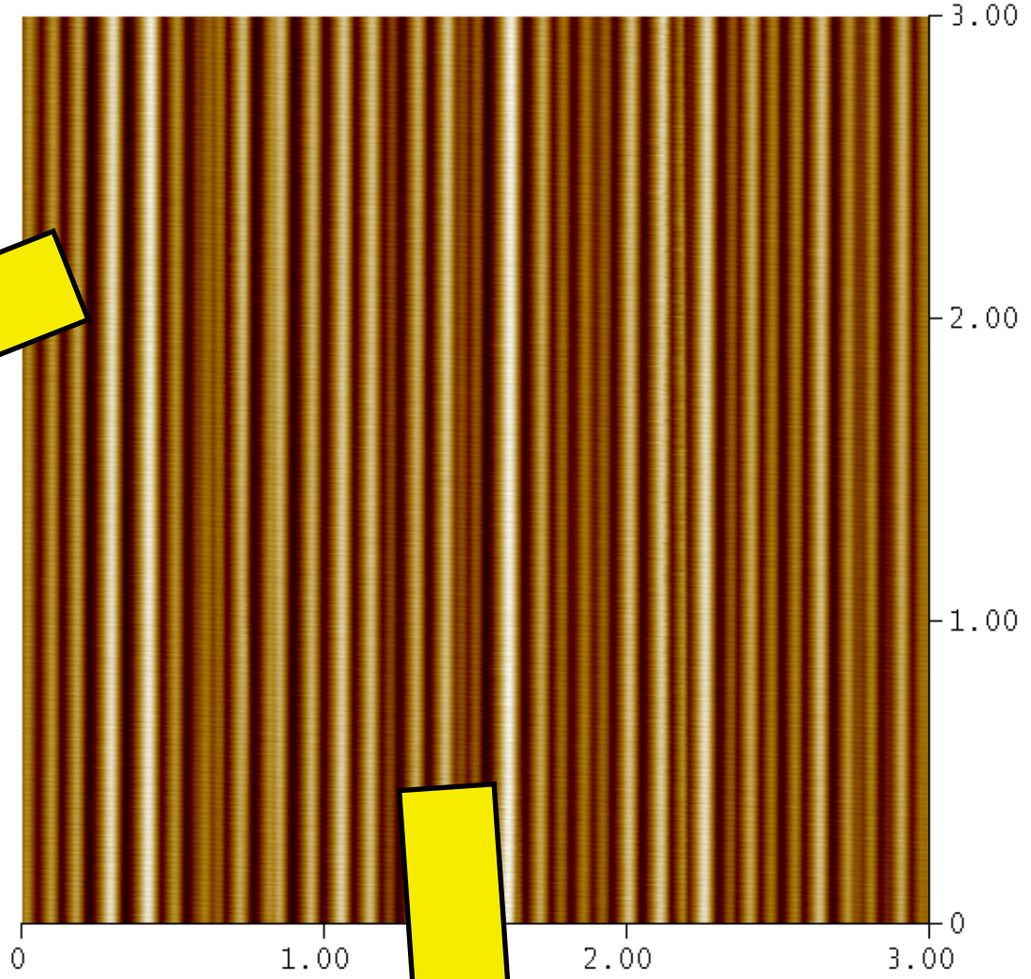


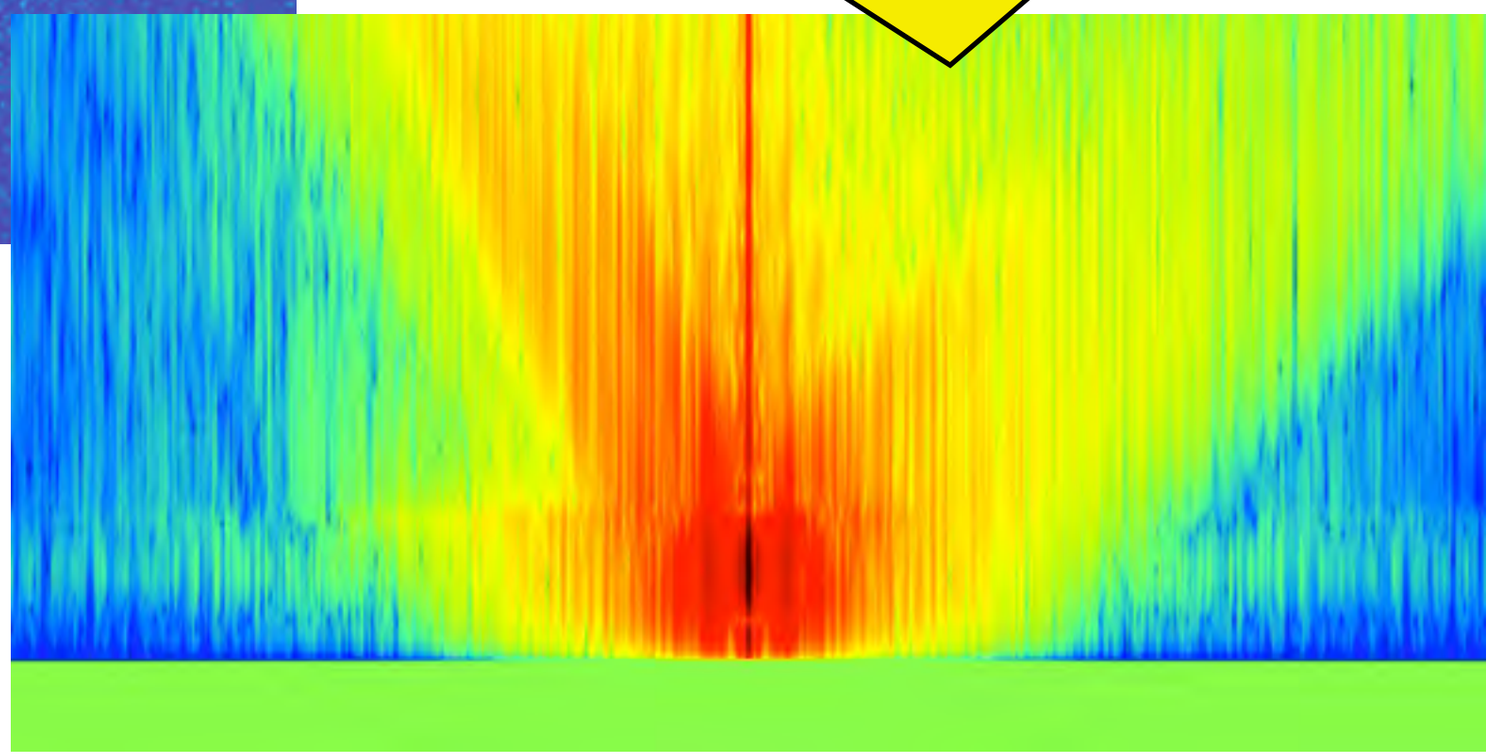
Collaboration with Cornell



SFM data of a sapphire substrate

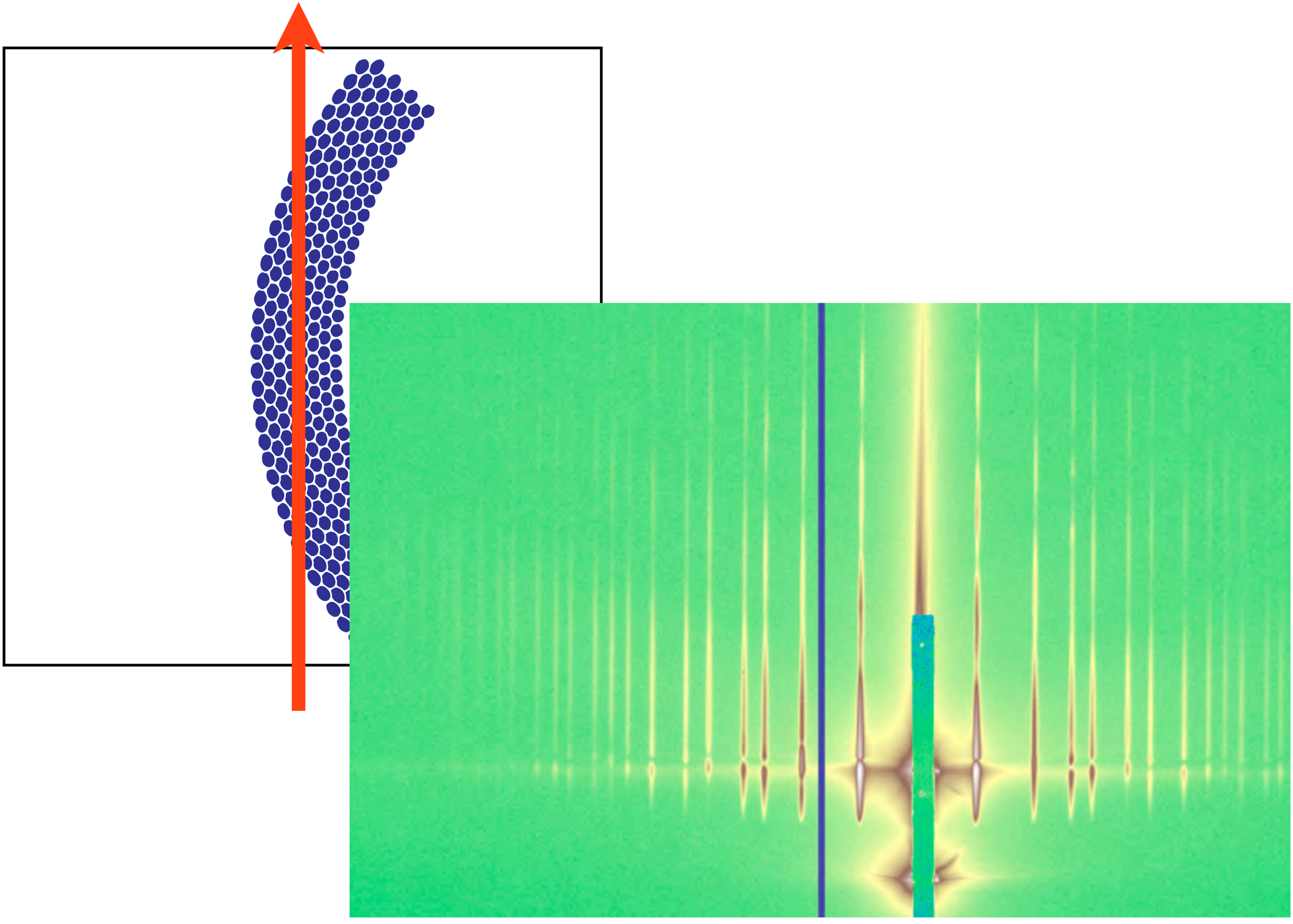




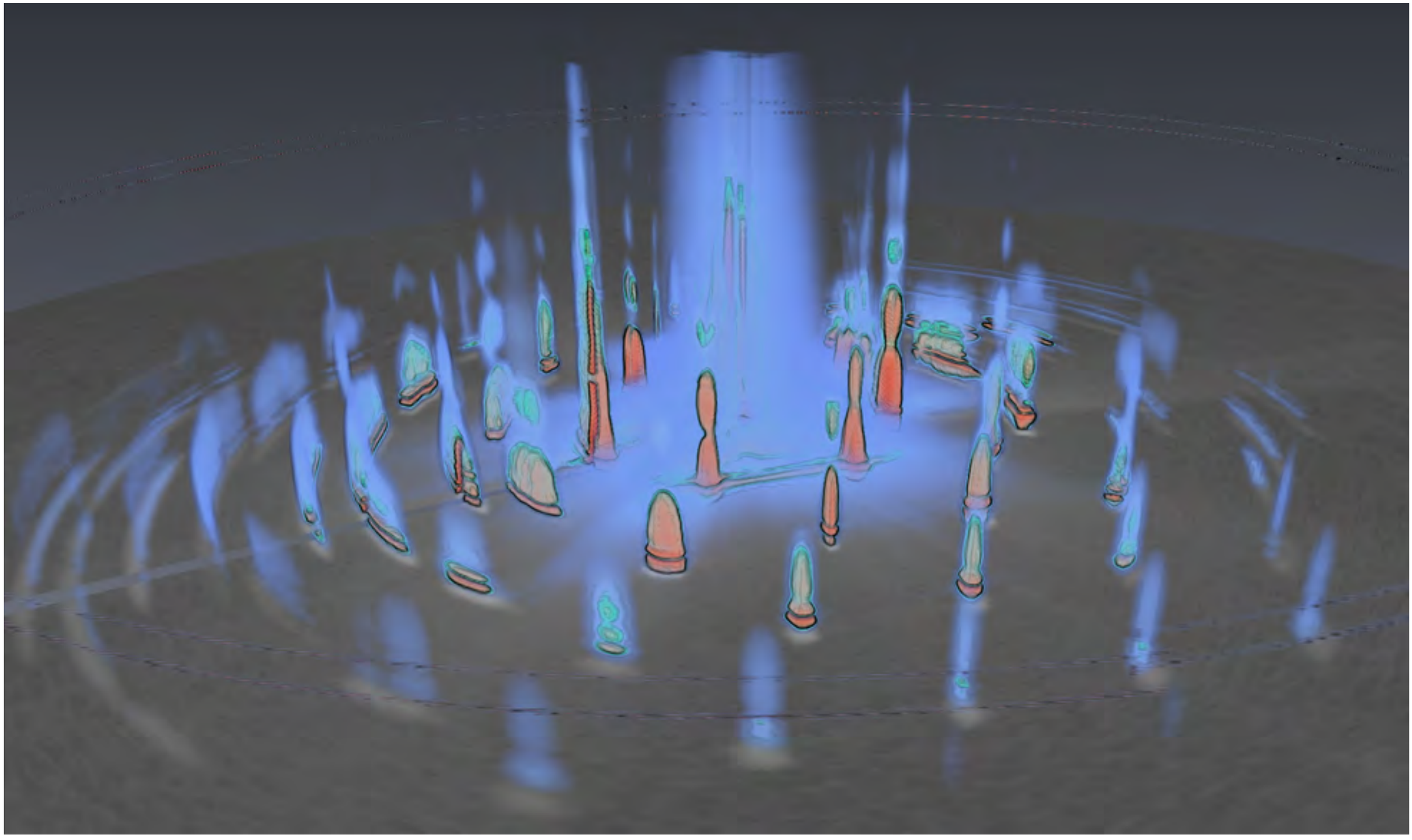


collaboration with
Eric Grohn (Cornell)

Many frames datasets



Frame Assembly



3d volume rendering for many frame data sets

Fitting Data

Parameter Fitting Approach

Let $\vec{X}$ be the vector of (continuous) sample parameters

$I(q_y, k_z^i, k_{z0}^f; \vec{X})$ is the computed scattered intensity for sample parameters $\vec{X}$

1D Relative error w.r.t. the experimental intensity at k_{z0}^f given k_z^i :

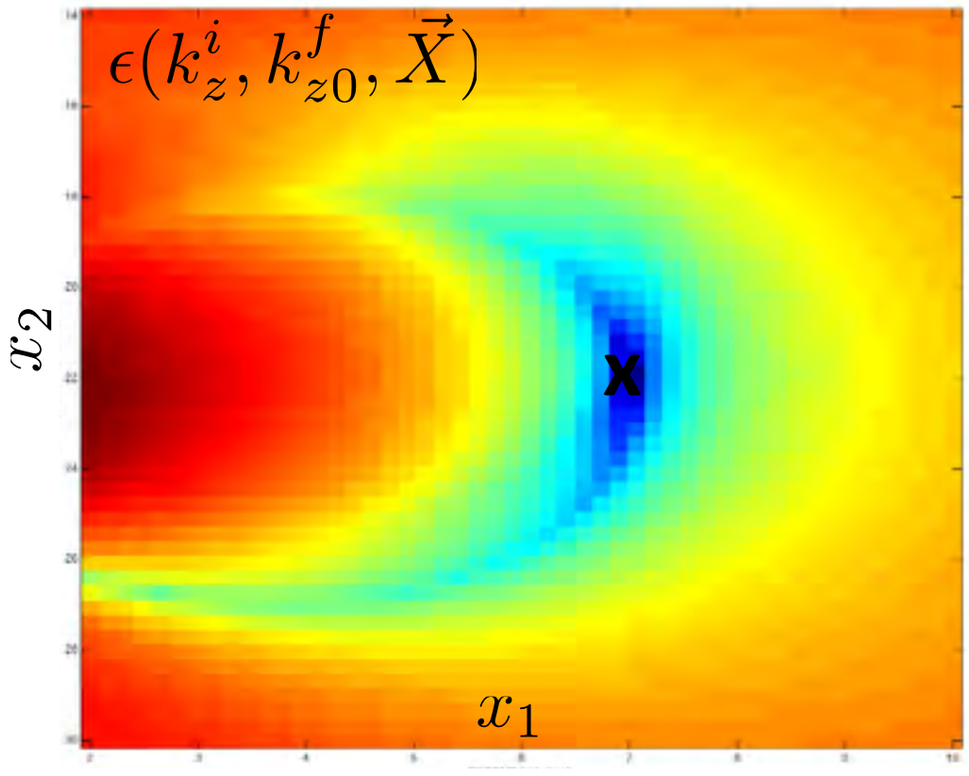
$$\epsilon(k_z^i, k_{z0}^f, \vec{X}) = \left[\int \left| \frac{I(q_y, k_z^i, k_{z0}^f; \vec{X}) - I_{exp}(q_y, k_z^i, k_{z0}^f)}{I_{exp}(q_y, k_z^i, k_{z0}^f)} \right|^2 dq_y \right]^{\frac{1}{2}}$$

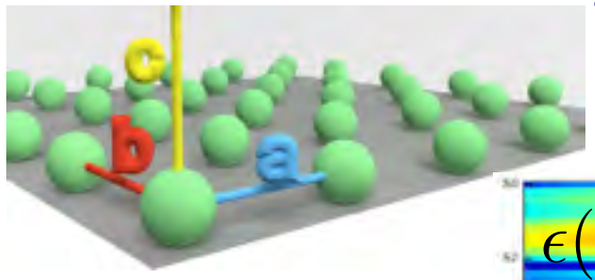
Solve: $\vec{X}_{cv}(k_z^i, k_{z0}^f) = \underset{\vec{X}}{\operatorname{argmin}} \{\epsilon(k_z^i, k_{z0}^f, \vec{X})\}$

Spheres with Size Distribution:

$(x_1, x_2) = (\bar{R}, V)$

avg. rad. std. dev.



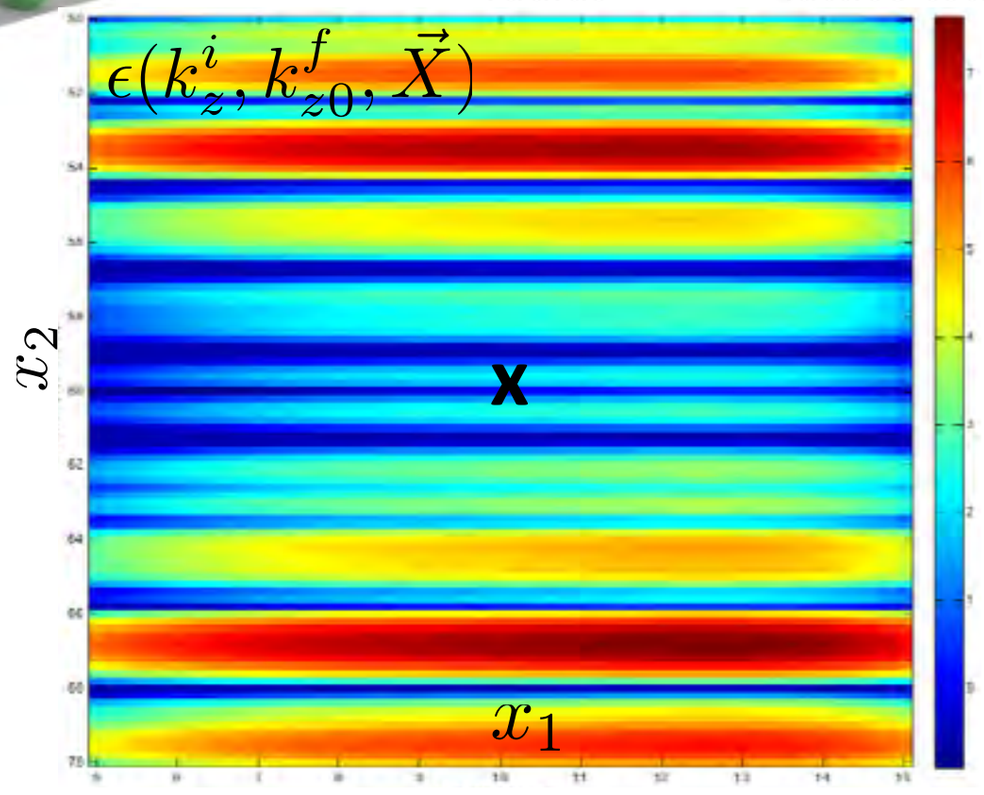


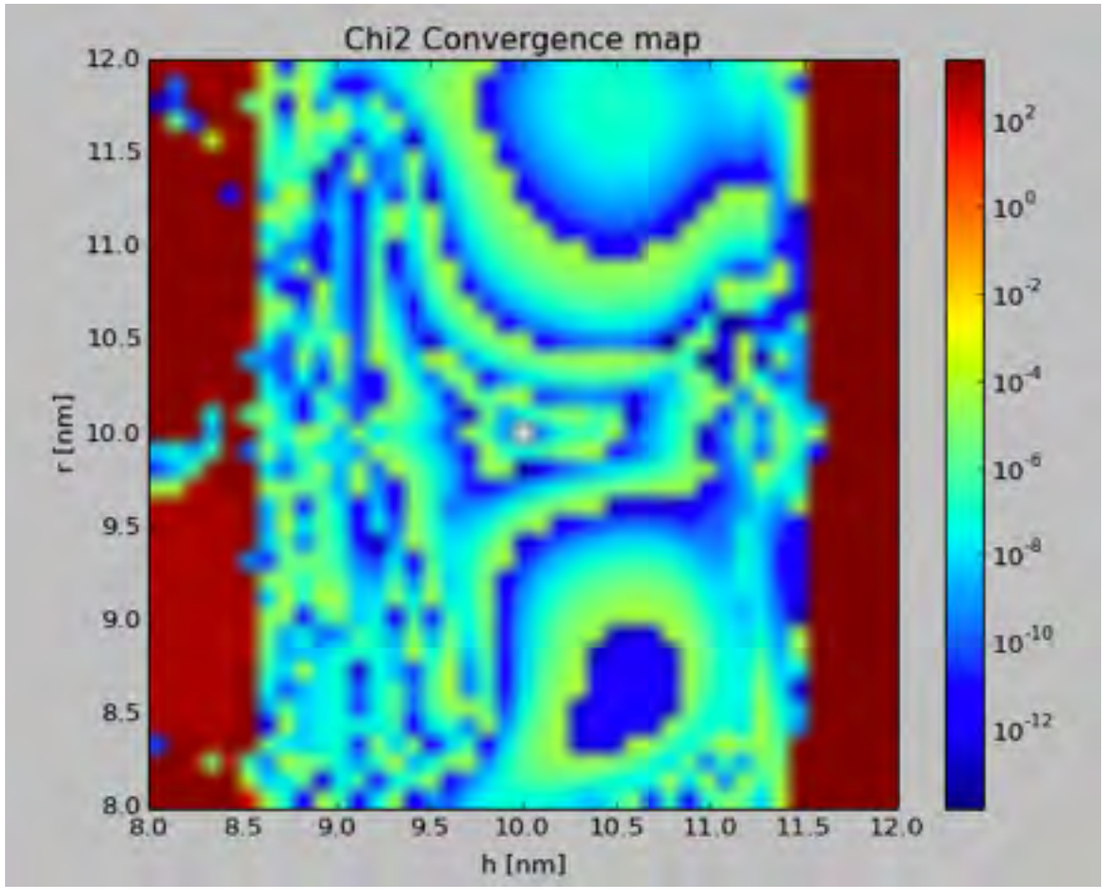
Error function locally "smooth", however, multiple nearby local minima for periodic lattice.

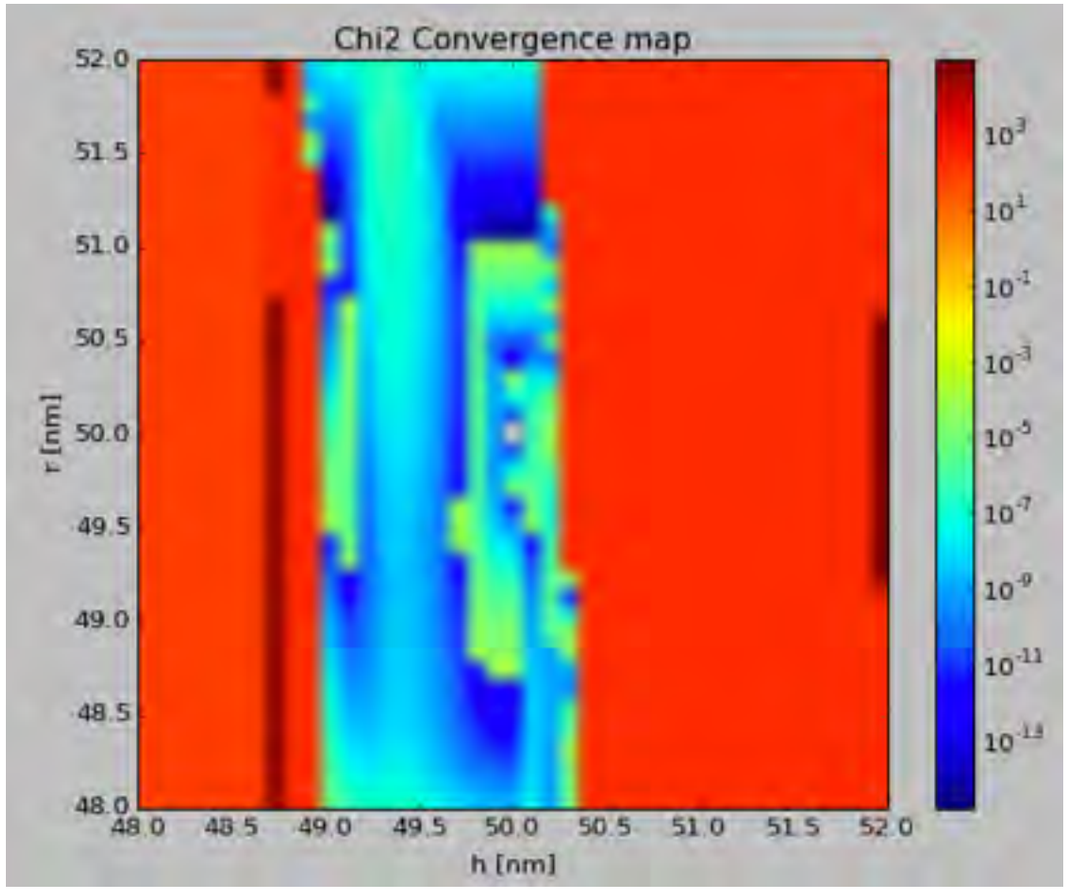
Spheres in a Cubic Lattice:

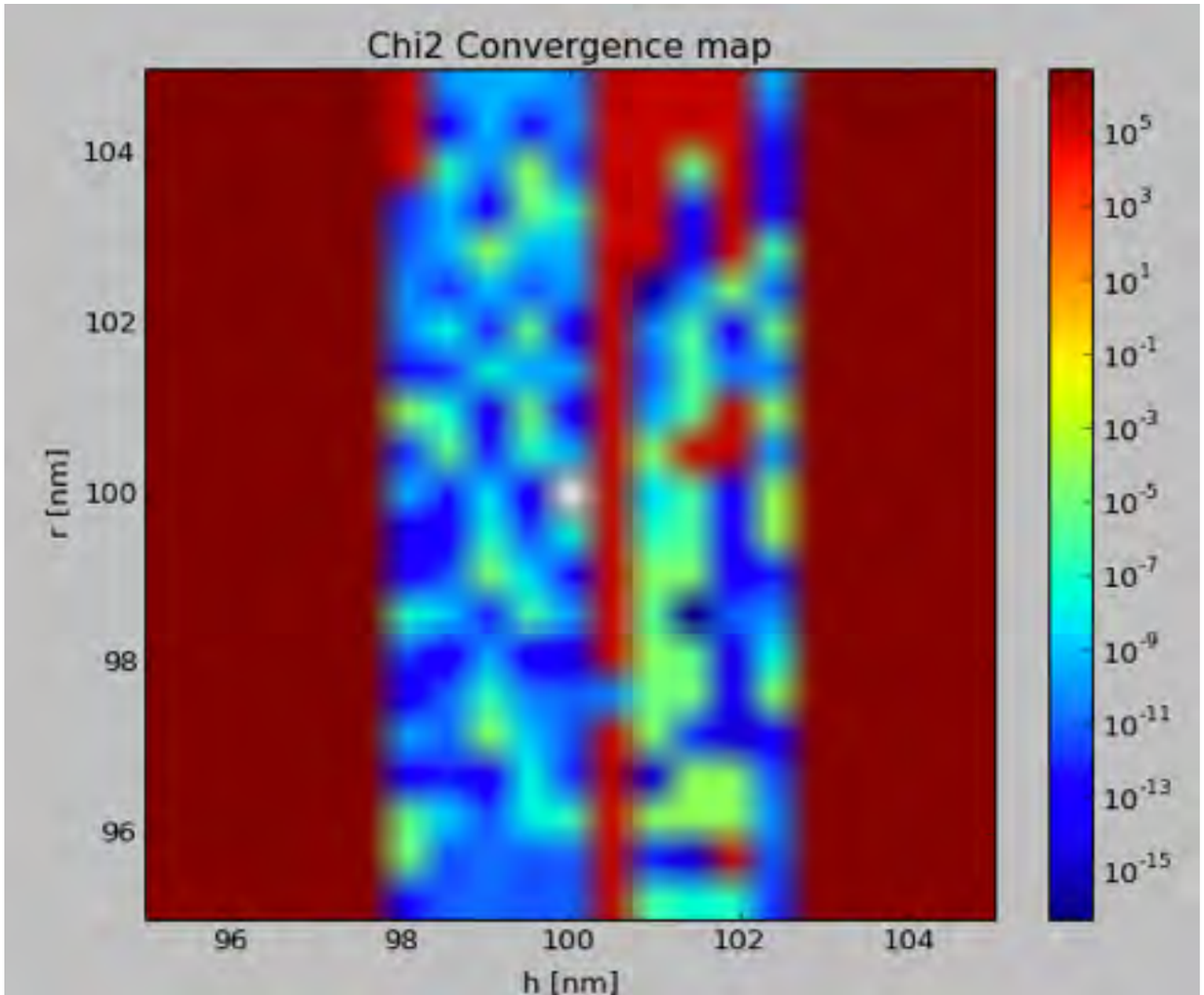
$(x_1, x_2) = (R, a)$

radius lattice cst.

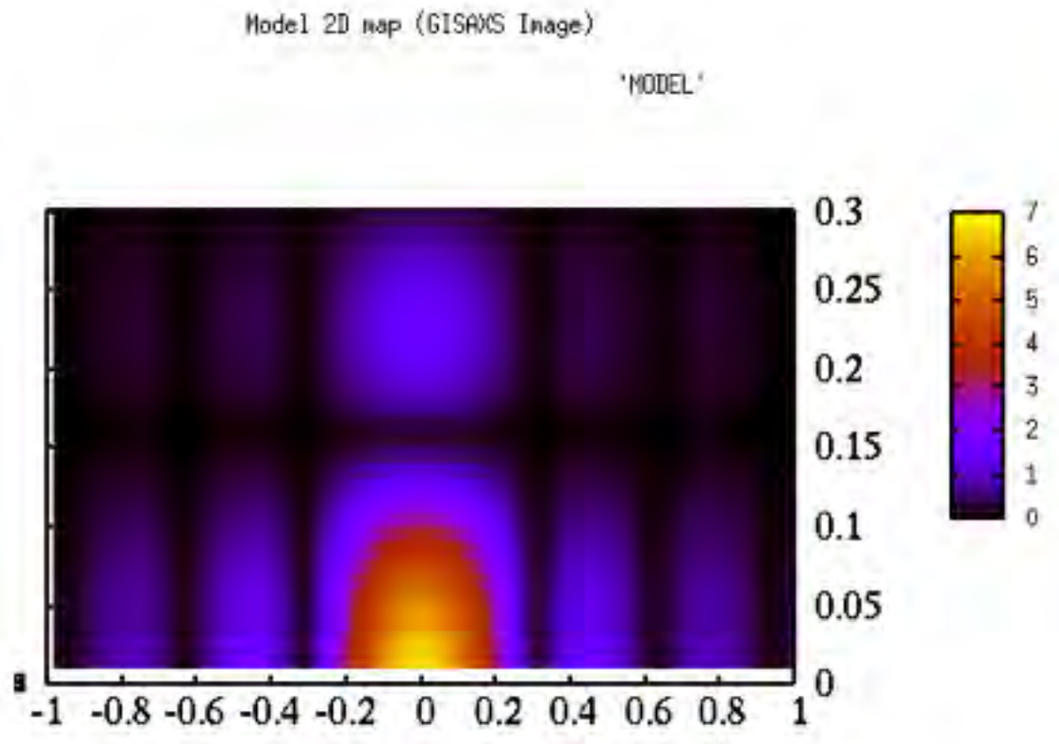


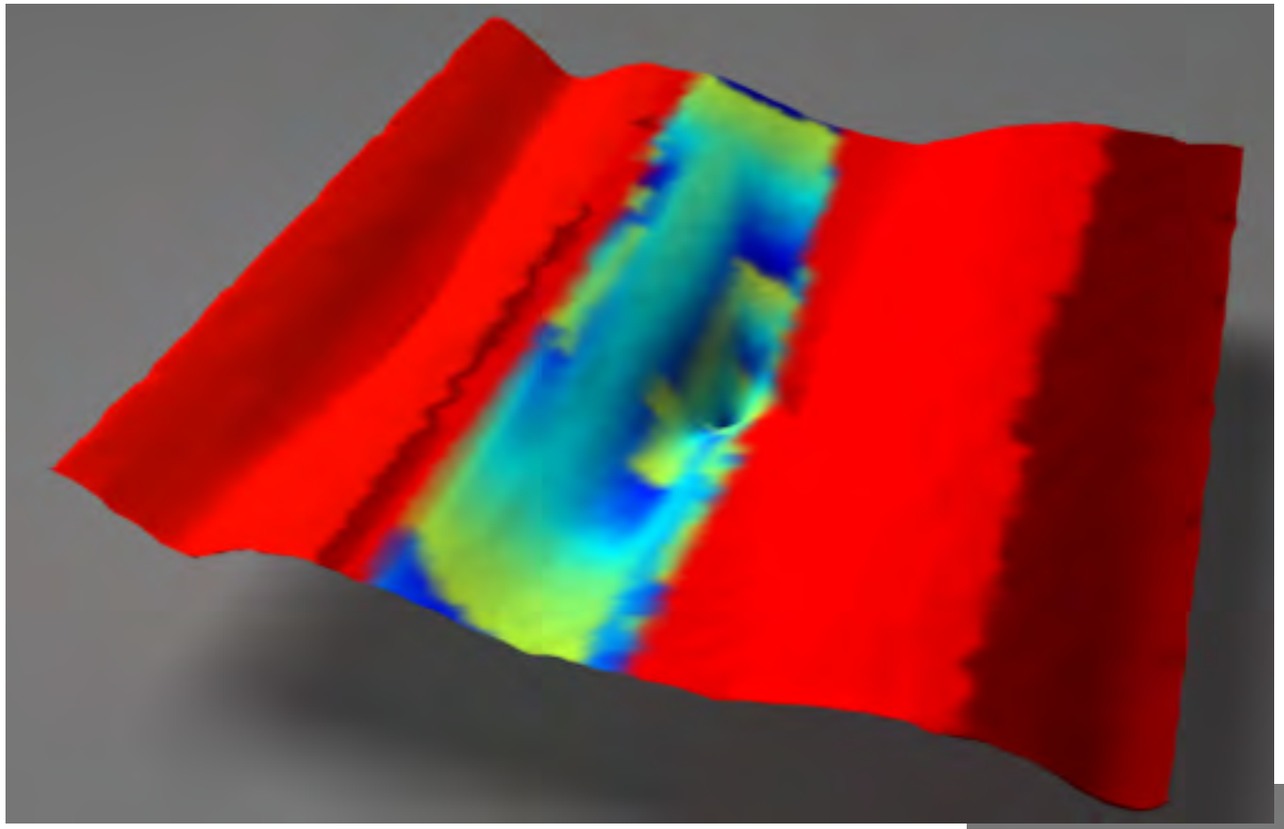


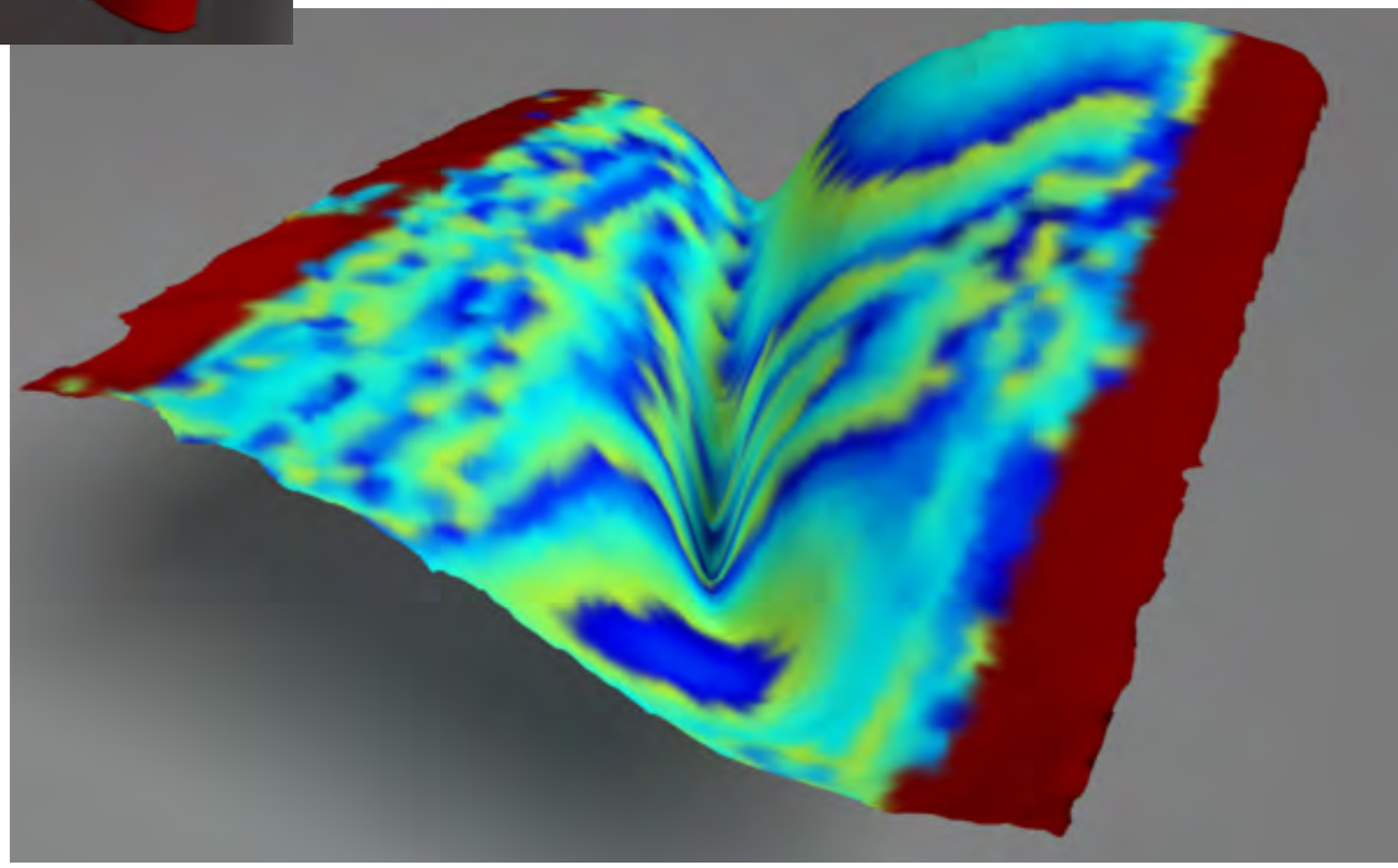




Fitting of a cylinder with different initial condition

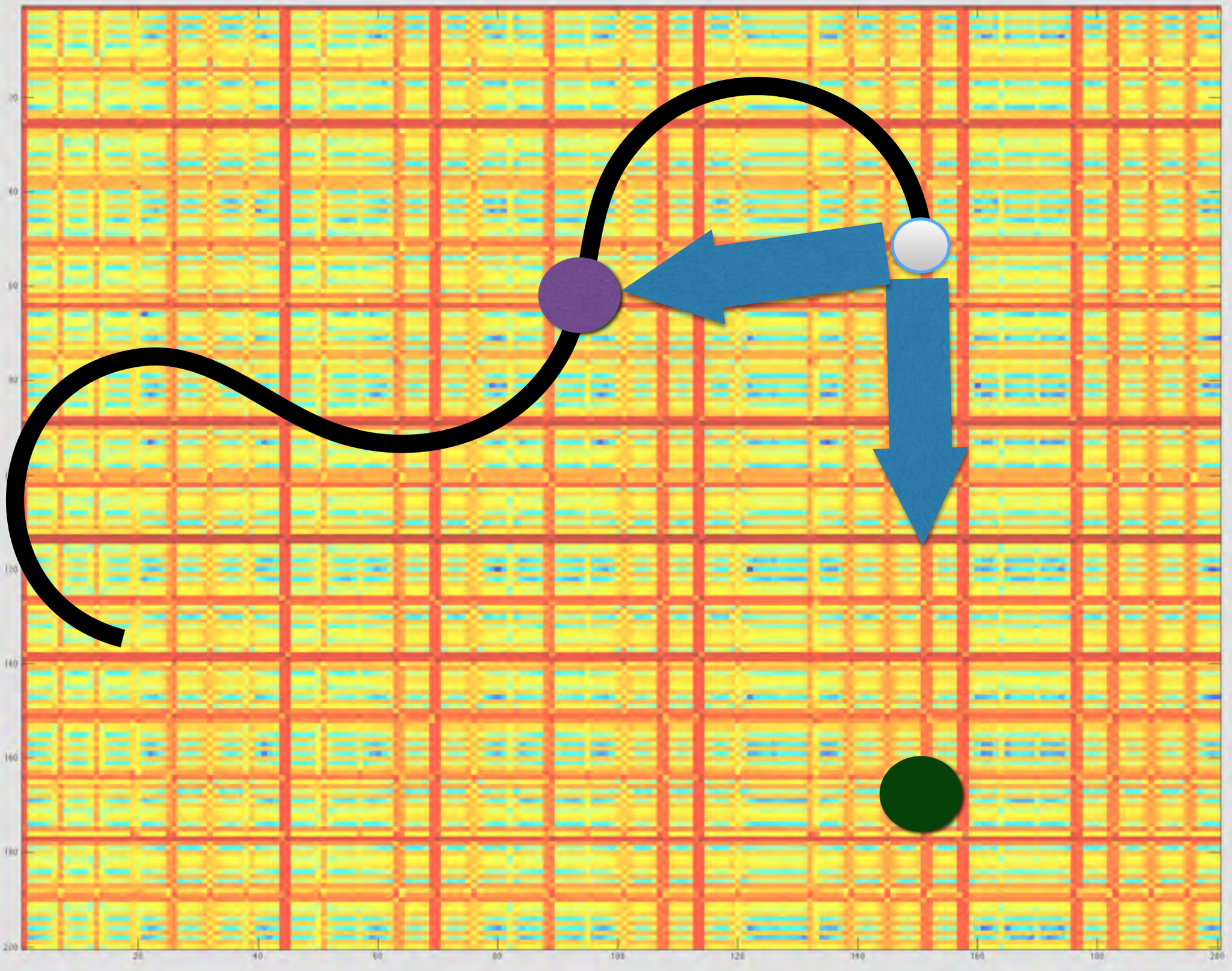






Brute force

Particle Swarm Optimization



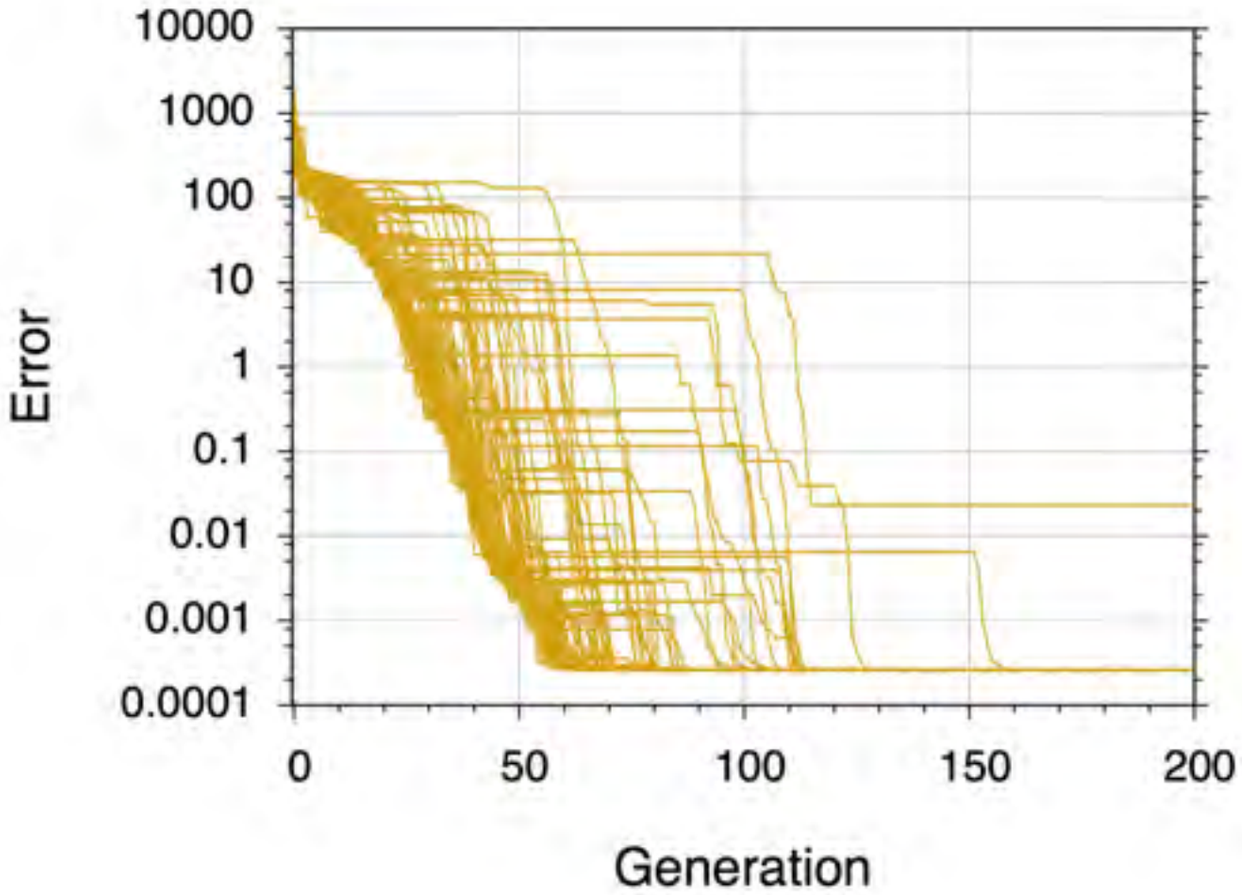
- The Common PSO Algorithm

test fit up to 10 parameters

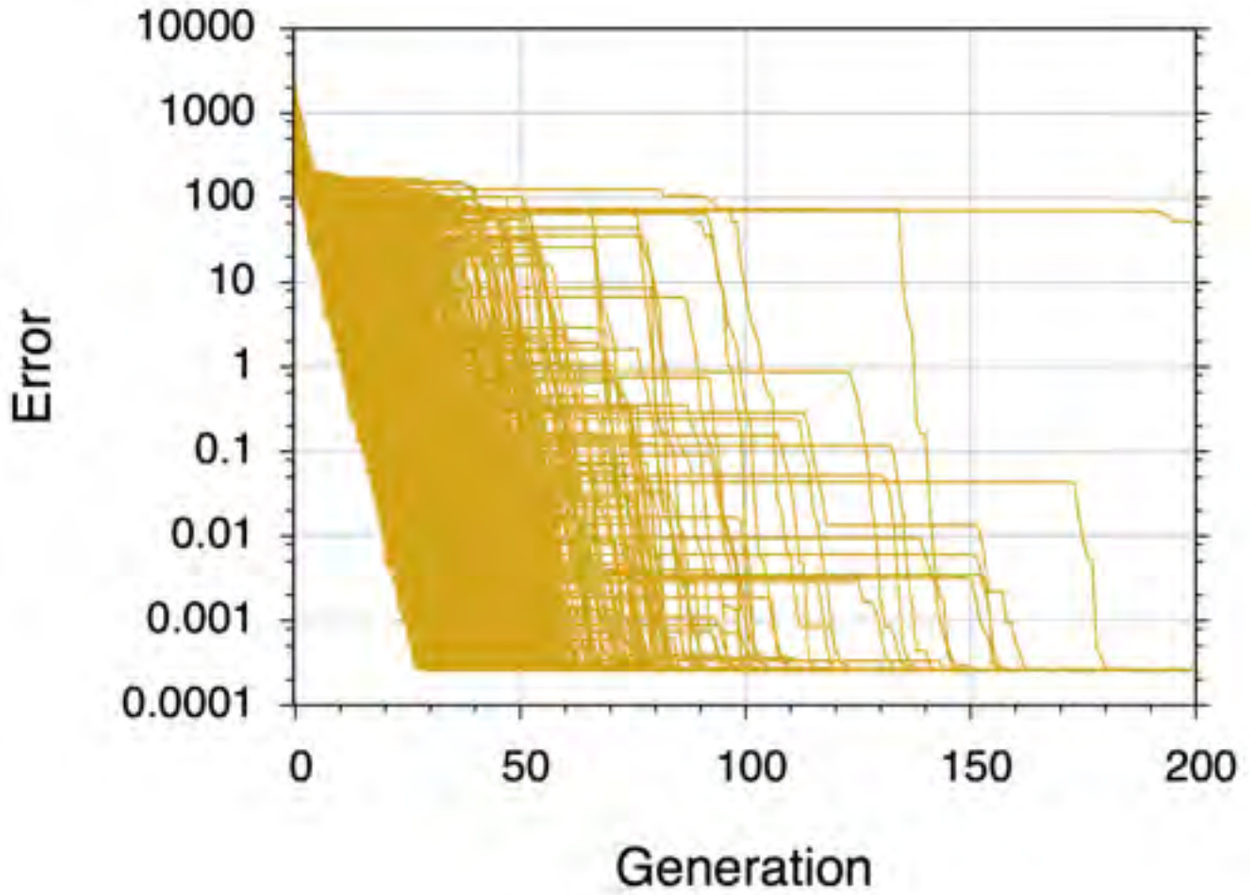
$$v_i(k+1) = \phi(k)\, v_i(k) + \alpha_1 \left[\gamma_{1i}\left(p_i - x_i(k)\right)\right] + \alpha_2 \left[\gamma_{2i}\left(G - x_i(k)\right)\right]$$

- ϕ - Inertia function
- $\alpha 1,2$ – Acceleration constants

100 agents



1000 agents



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Shapes

Key: s1

Name: cylinder

Origin vector: 0 0 0

Z-tilt: 0

XY-rotation: 0

Parameter List +

Parameter Details:

Type: radius

Minimum: 20

Maximum: 120

Number of values: 50

Statistic: gaussian

Mean: 100

Std deviation: 10

Parameter Details:

Type: height

Minimum: 150

Maximum: 0

Number of values: 1

Statistic: single

Mean: 0

Std deviation: 0

Layers

Key: l1

Order: -1

Thickness: -1

Refractive index: Delta: 9.107e-1 Beta: 2.466e-1

Structures

Key: st1

Grain

Layer key: l1

Lattice: