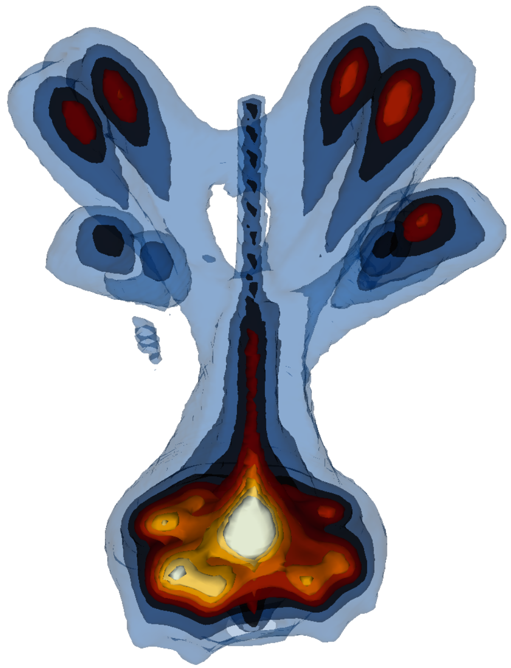
New tools for fast and reliable 3-dimensional reciprocal space mapping using area detectors

Christian M. Schlepütz
and John Hammonds



June 19, 2014
InterCAT Technical Workgroup (TWG)

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Software
- Xrayutilities: D. Kriegner, E. Wintersberger
  http://xrayutilities.sourceforge.net
- PySpec: Stuart Wilkins
  https://github.com/stuwilkins/pyspec
- Paraview: Kitware (3-D visualization)
  http://paraview.org
- ImageJ: W.S. Rasband, NIH
  http://imagej.nih.gov/imagej

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- GeoSoilEnviroCARS: National Science Foundation – Earth Sciences (EAR-0622171)

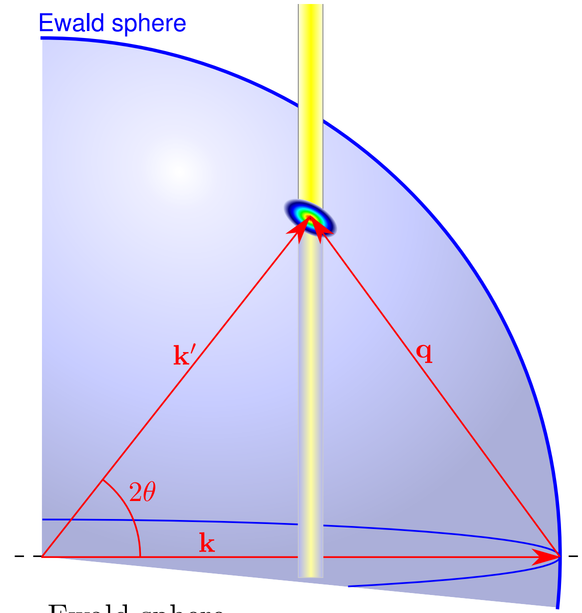
Outline

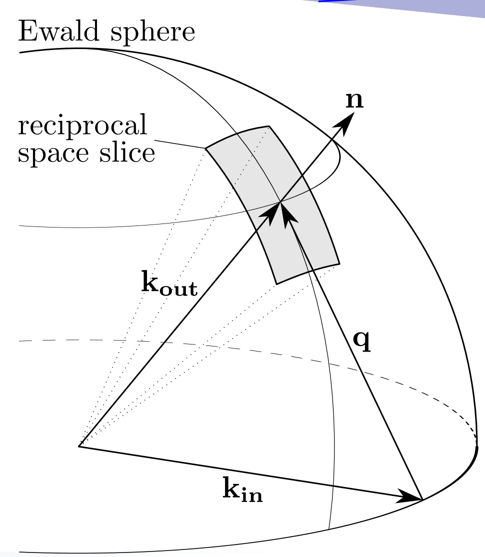
- Introduction to 3D reciprocal space mapping
- Introducing **rsMap3D**
- Examples:
  - Powder diffraction: LaB_6 standard
  - Reciprocal space maps: $BiFeO_3$ on $SrTiO_3$(001) and $LaAlO_3$(001)
  - Pole figures: Growth of Fe_2O_3 on ITO on YSZ
- Conclusions and Outlook

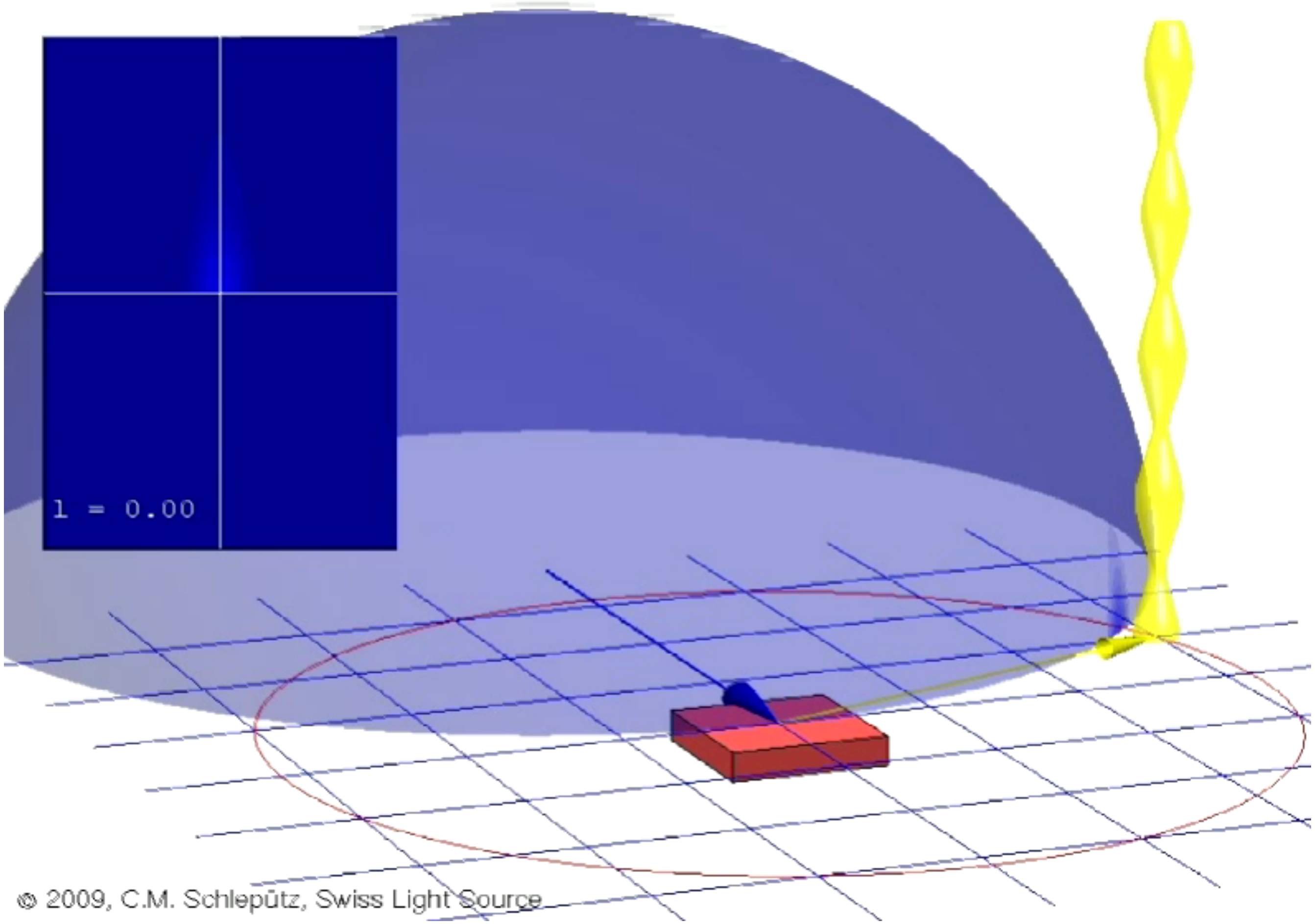
What do we “see” in a diffraction experiment?

- Elastic scattering:
  - |**k**| = |**k**’|
  - **q** = **k**’ -**k**
  - Set of all possible **q** vectors define the Ewald sphere.
- All points on the Ewald sphere are in the scattering condition.
- Detector “sees” a subset of the surface of the Ewald sphere.
- Area detectors:
  - Image contains curved 2D slice in 3D space.
  - Each pixel has distinct **Q** = (q_x, q_y, q_z)
- Sample rotation “exposes” different reciprocal space slices to Ewald sphere.

“Ewald sphere - Lab frame, instrument”
“Reciprocal space - Sample property”



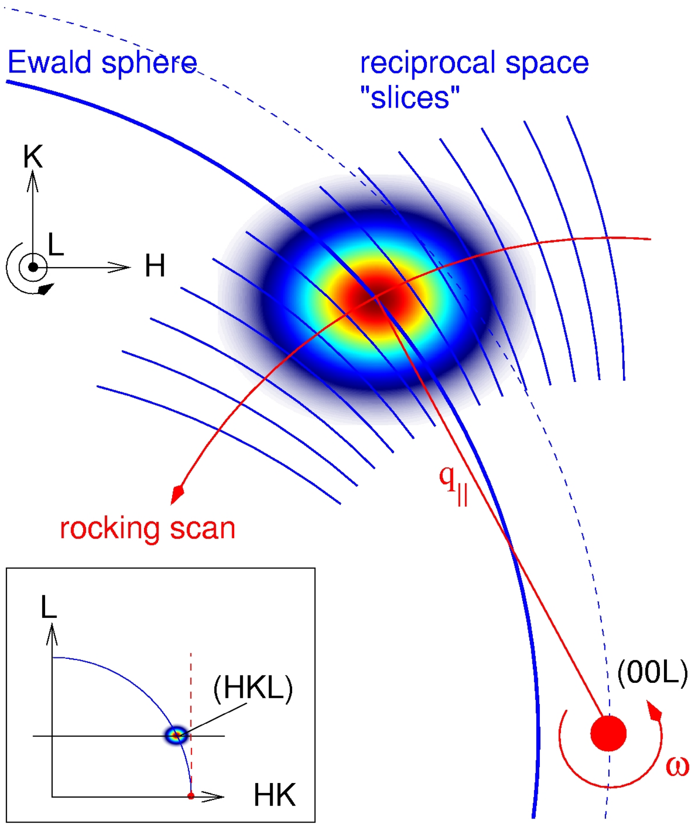




l = 0.00
© 2009, C.M. Schlepütz, Swiss Light Source

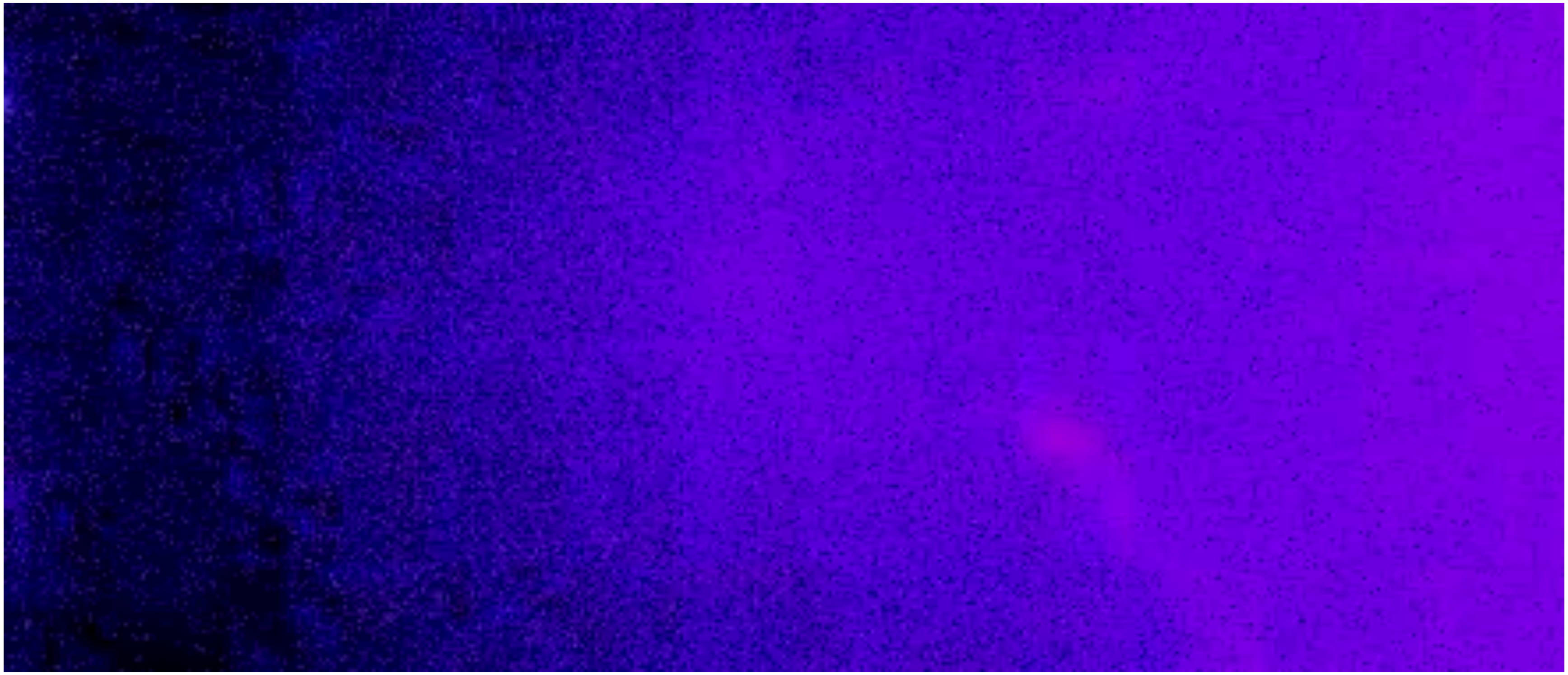
From slices to 3D reciprocal space maps (RSMs)

- Sample rotation selects different slices
  ➜ scan yields set of detector images.
- Selecting the ideal scan direction is not always easy…
  - stich multiple scans together.
- Data processing challenge:
  - Compute q_x, q_y, q_z (HKL) values for each detector pixel.
  - Map resulting set of curved reciprocal space slices to "friendly" (usually orthogonal) coordinate system.
  - Find the right way to visualize 3D data sets on paper:
    Line cuts, plane cuts, isosurfaces, point clouds, contour maps, pole figures, histograms, etc.

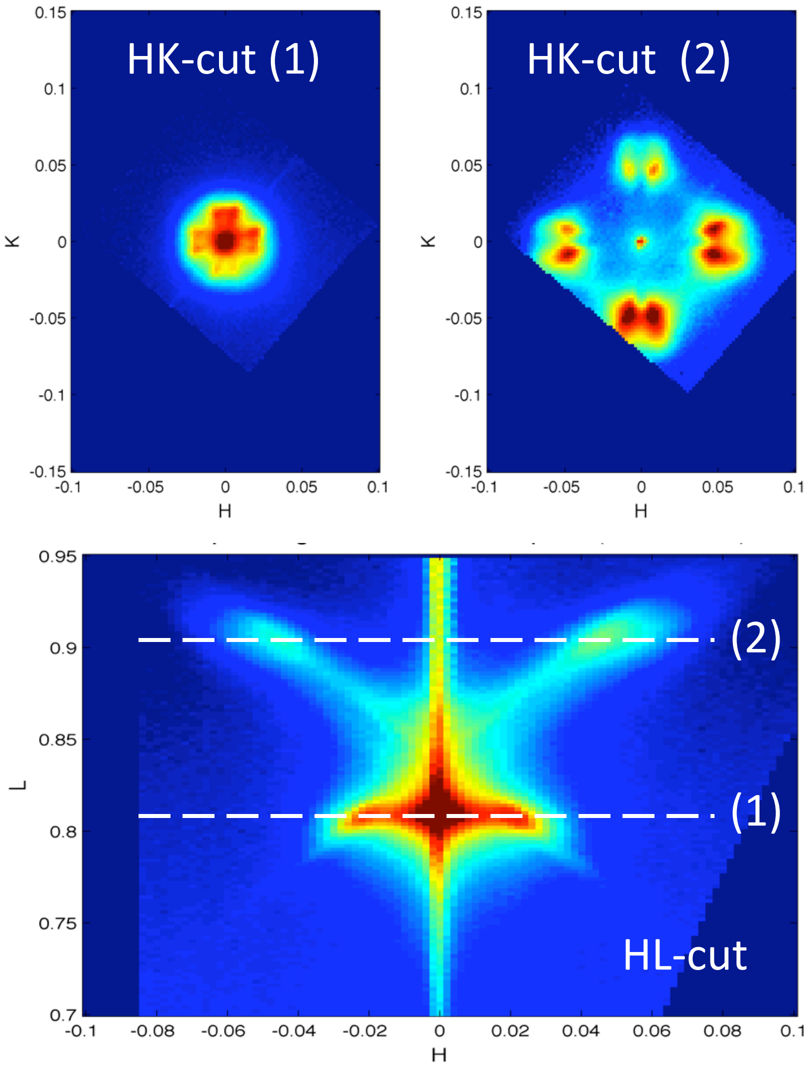


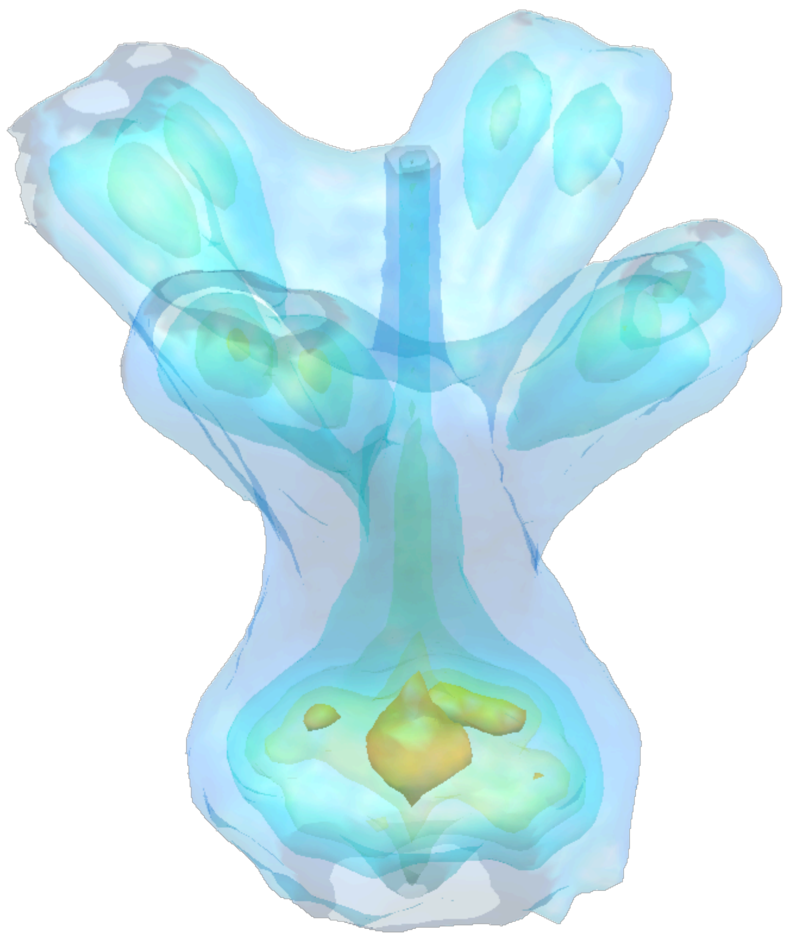
Example: raw detector data

Rocking scan (theta) of the (001) peak of thick $BiFeO_3$ on $LaAlO_3$



Example: Desired output





Semi-transparent 3D isosurfaces

Introducing rsMap3D

- Written by John Hammonds, APS Software Services Group
- Based on Matlab and Python scripts by C. Schlepuetz
- Substantial input from Jon Tischler

- SVN repository:
  https://subversion.xray.aps.anl.gov/RSM/rsMap3D/
- Documentation (work in progress):
  https://confluence.aps.anl.gov/display/RSM/SSG_000116+Reciprocal+Space+Mapping

Design goals

- Accommodate various beamline and instrument configurations
- Capable of handling different types of area and line detectors
- Can handle any form of angle scans and/or energy scans
- Platform-independent (Windows, Mac, Linux)
- Does not require any software with license fees
- No special computer hardware requirements
  - Reasonably large data sets should be analyzable on any standalone computer (≥ 8 GB RAM)
- GUI interface & scriptable

Implementation

- rsMap3D is written in Python (2.7)
- Required Python packages:
  - numpy
  - pytables
  - pyQT
  - vtk
- External python packages:
  - xrayutilities
    - http://xrayutilities.sourceforge.net
    - Dominik Kriegner, Eugen Wintersberger, BESSY
    - Performs a lot of the actual number crunching
    - heavy-duty calculation are written in C
    - code is written with multi-processing support (openmp)
  - pyspec
    - https://github.com/stuwilkins/pyspec
    - Stuart Wilkins, Brookhaven National Laboratory
    - used to read SPEC files

Implementation

- Detector and instrument configurations are stored in external configuration files (xml format)
- Can be read in through the GUI
- Instrument configuration should be fairly static for each beamline
- Detector configuration may change more frequently
- More than one detector can be included in the detector config

Instrument configuration

- Any number of sample and detector rotation axes
- Arbitrary choice of coordinate system (needs to be consistent with detector config)
- Filter and monitor corrections

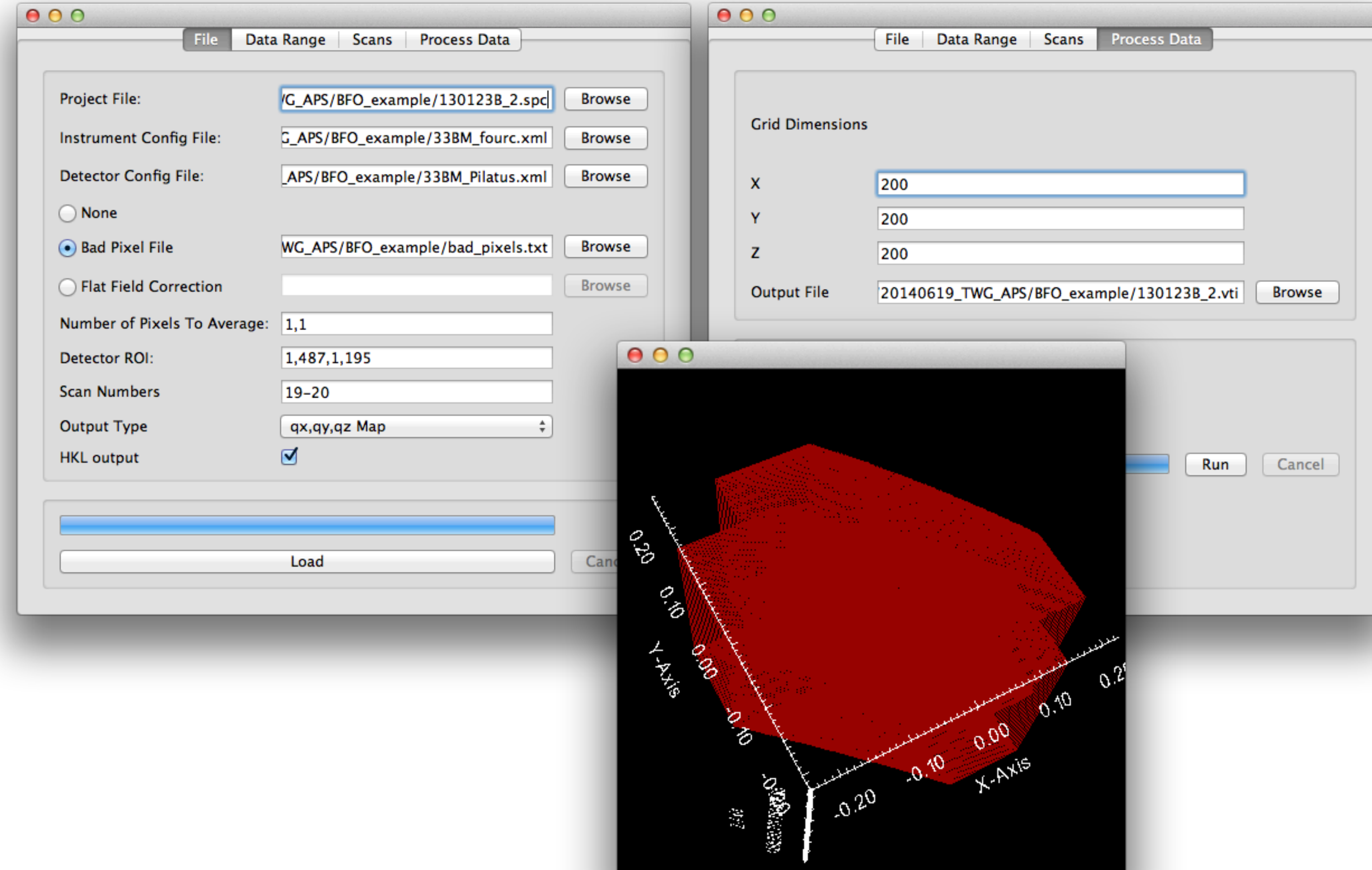
```xml
<?xml version="1.0" encoding="UTF-8" ?>
<instForXrayutils xmlns="https://subversion.xray.aps.anl.gov/RSM/instForXrayutils">
    <dateWritten>Wed, Mar 5, 2014</dateWritten>
    <timeWritten>14:36:13.5 (-6)</timeWritten>
    <!-- Define the sample circles -->
    <sampleCircles numCircles="3">
       <circleAxis number="1" specMotorName="theta" directionAxis="z-"/>
       <circleAxis number="2" specMotorName="chi" directionAxis="y+"/>
       <circleAxis number="3" specMotorName="phi" directionAxis="z-"/>
    </sampleCircles>
    <!-- Define the detector circles as a series of axes. -->
    <detectorCircles numCircles="1">
       <circleAxis number="1" specMotorName="X2mtheta" directionAxis="z-"/>
    </detectorCircles>
    <!-- Define reference directions -->
    <primaryBeamDirection>
        <axis number="1">0</axis>
        <axis number="2">1</axis>
        <axis number="3">0</axis>
    </primaryBeamDirection>
    <inplaneReferenceDirection>
        <axis number="1">0</axis>
        <axis number="2">1</axis>
        <axis number="3">0</axis>
    </inplaneReferenceDirection>
    <sampleSurfaceNormalDirection>
        <axis number="1">1</axis>
        <axis number="2">0</axis>
        <axis number="3">0</axis>
    </sampleSurfaceNormalDirection>
    <!-- Set the counter names for monitor and filter corrections -->
    <monitorName scaleFactor="1"></monitorName>
    <filterName scaleFactor="1">None</filterName>
</instForXrayutils>
```

Detector configuration

- Supports any number of detectors (Identified by ID)
- rsMap3D currently only handles one detector at a time

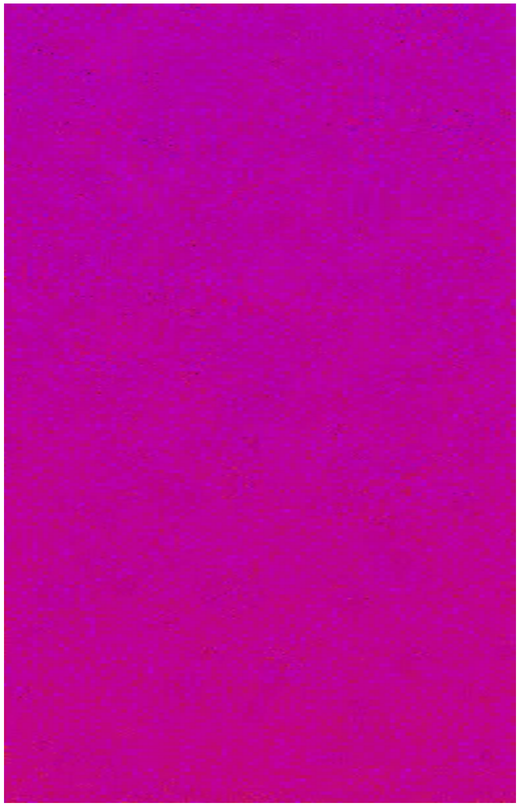
```xml
<?xml version="1.0" encoding="UTF-8"?>
<detectorGeometryForXrayutilities xmlns="https://subversion.xr
    <dateWritten></dateWritten>
    <timeWritten></timeWritten>

    <Detectors>
       <Detector>
            <pixelDirection1>x-</pixelDirection1>
            <pixelDirection2>z+</pixelDirection2>
            <centerChannelPixel>200 95</centerChannelPixel>
            <Npixels>487 195</Npixels>
            <size unit="mm">83.764 33.54</size>
            <distance unit="mm">778.51</distance>
            <ID>Pilatus</ID>
            <note></note>
        </Detector>
    </Detectors>
</detectorGeometryForXrayutilities>
```

The GUI



File
Data Range
Scans
Process Data
Project File:
/G_APS/BFO_example/130123B_2.spc
Browse
Instrument Config File:
G_APS/BFO_example/33BM_fourc.xml
Detector Config File:
_APS/BFO_example/33BM_Pilatus.xml
None
Bad Pixel File
WG_APS/BFO_example/bad_pixels.txt
Flat Field Correction
Number of Pixels To Average:
1,1
Detector ROI:
1,487,1,195
Scan Numbers
19-20
Output Type
qx,qy,qz Map
HKL output
Load
Grid Dimensions
X
200
Y
200
Z
200
Output File
20140619_TWG_APS/BFO_example/130123B_2.vti
Run
Cancel
Y-Axis
X-Axis

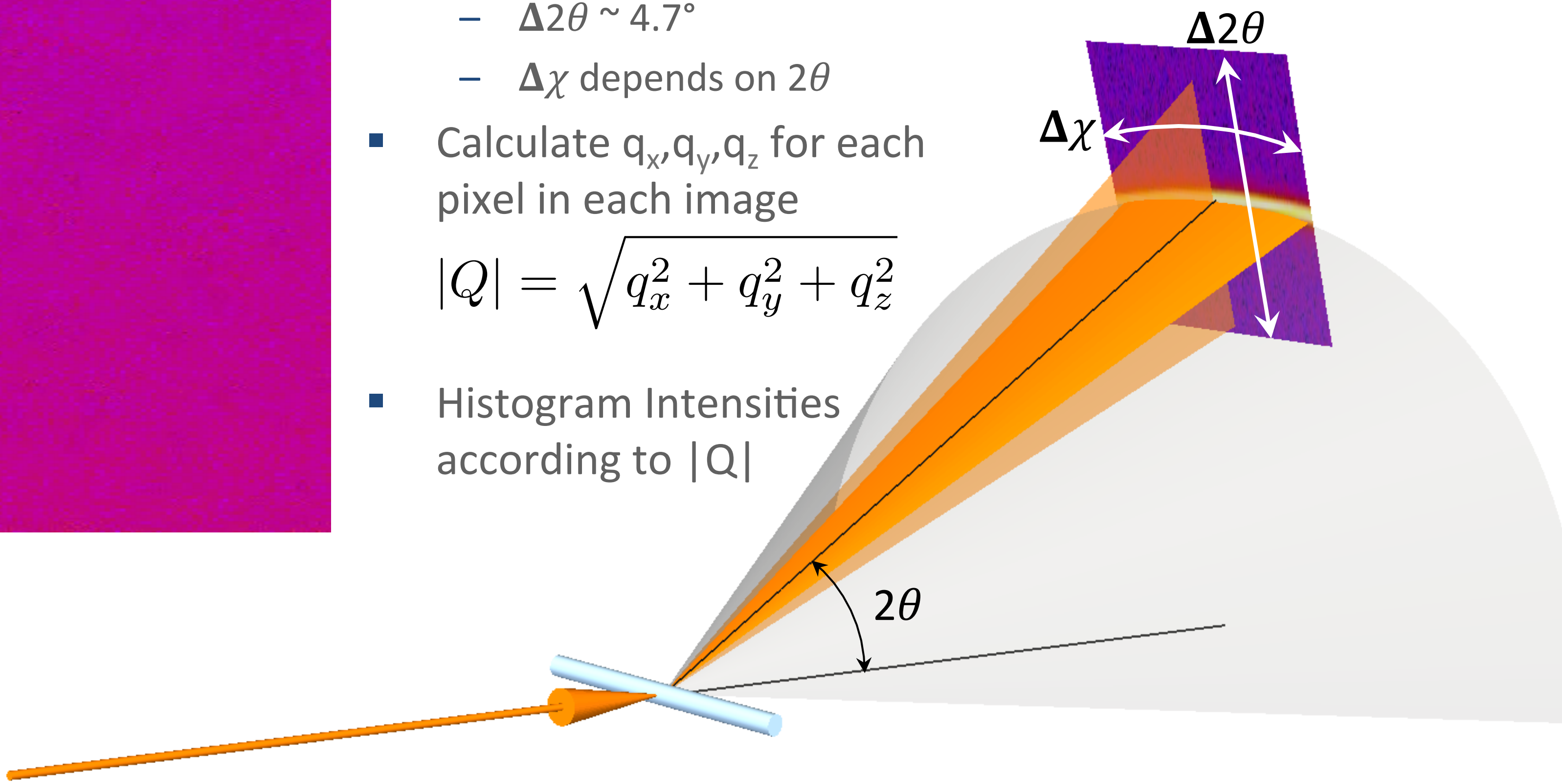
Powder diffraction with an area detector



- Detector "sees" a finite range of $\Delta 2\theta$, $\Delta\chi$ for each 2θ position.
- Example: @ 1m detector distance (using Pilatus 100K):
  - $\Delta 2\theta$ ~ 4.7°
  - $\Delta\chi$ depends on 2θ
- Calculate q_x,q_y,q_z for each pixel in each image

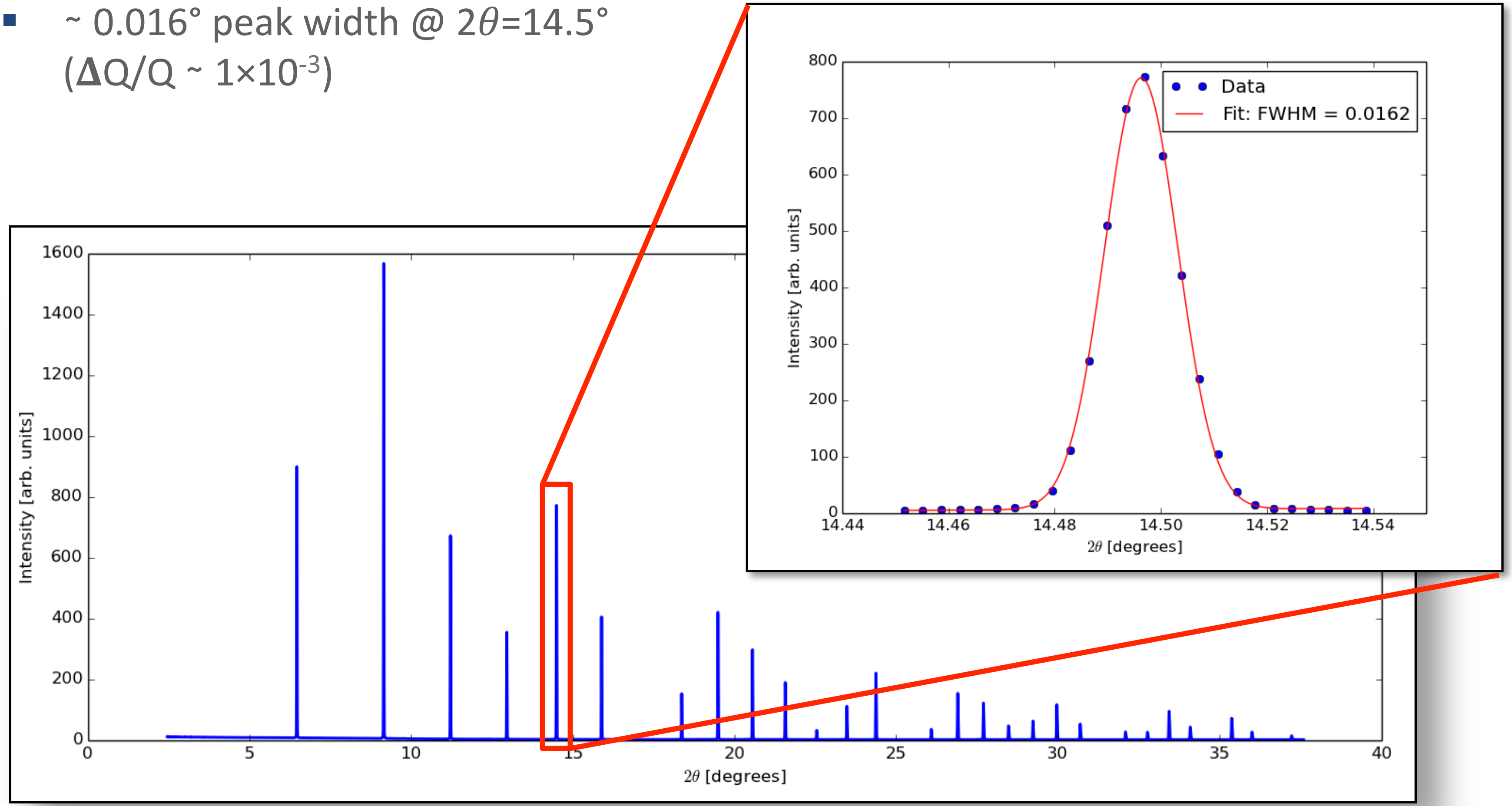
$$|Q| = \sqrt{q_x^2 + q_y^2 + q_z^2}$$

- Histogram Intensities according to |Q|



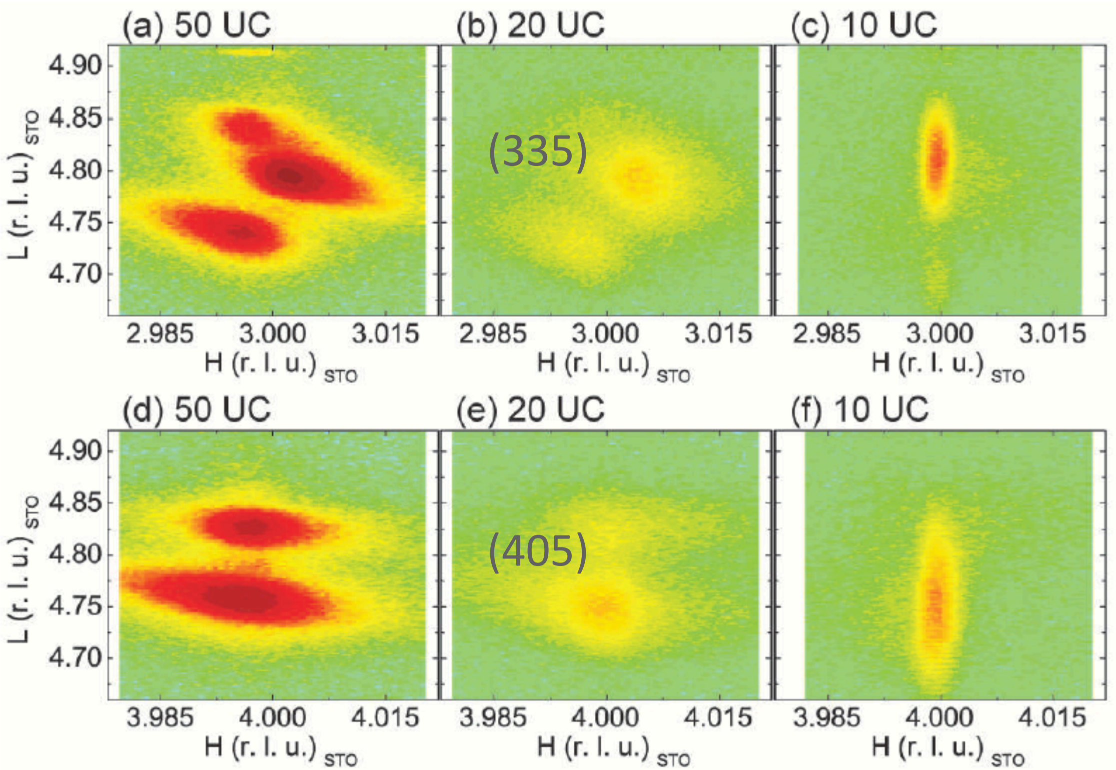
LaB_6 standard

- 400 second scan (400 points of 1 second)
- 5 – 35 degrees (nominal)
- ~ 0.016° peak width @ 2θ=14.5°
  ($\Delta Q/Q$ ~ 1×10^{-3})



$BiFeO_3$ thin films on $SrTiO_3$

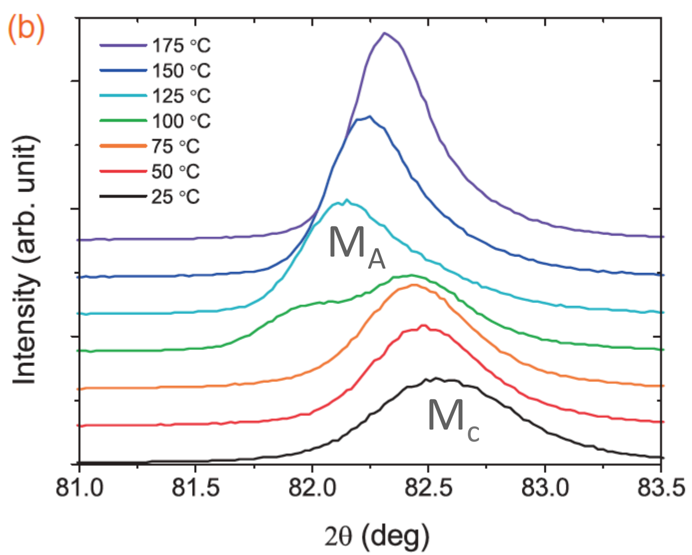
- Thicker film:
  - 3-fold splitting of peaks with H = K
  - 2-fold splitting of peaks with H=0 or K=0
  - ➔ M_A monoclinic
- Thin film:
  - single peak
  - sharper in-plane
  - ➔ Tetragonal
- Phase Transition!

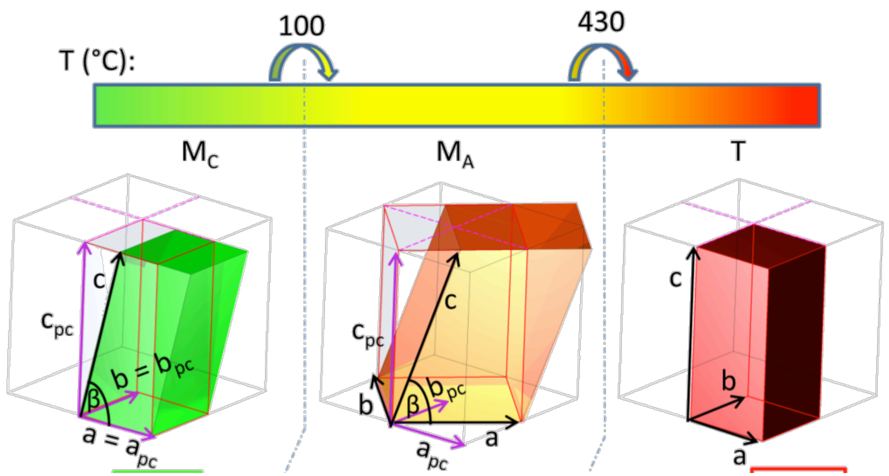


Y. Yang *et al.*, *APL Materials* **1,** 052102 (2013).

$BiFeO_3$ on $LaAlO_3$ (001)

- Subject to large compressive strain: ~4.6%
- Phase transitions with temperature: M_C → M_A → T
- T-phase has giant c/a ~ 1.24
- M_A and M_C are also T-like with giant c/a

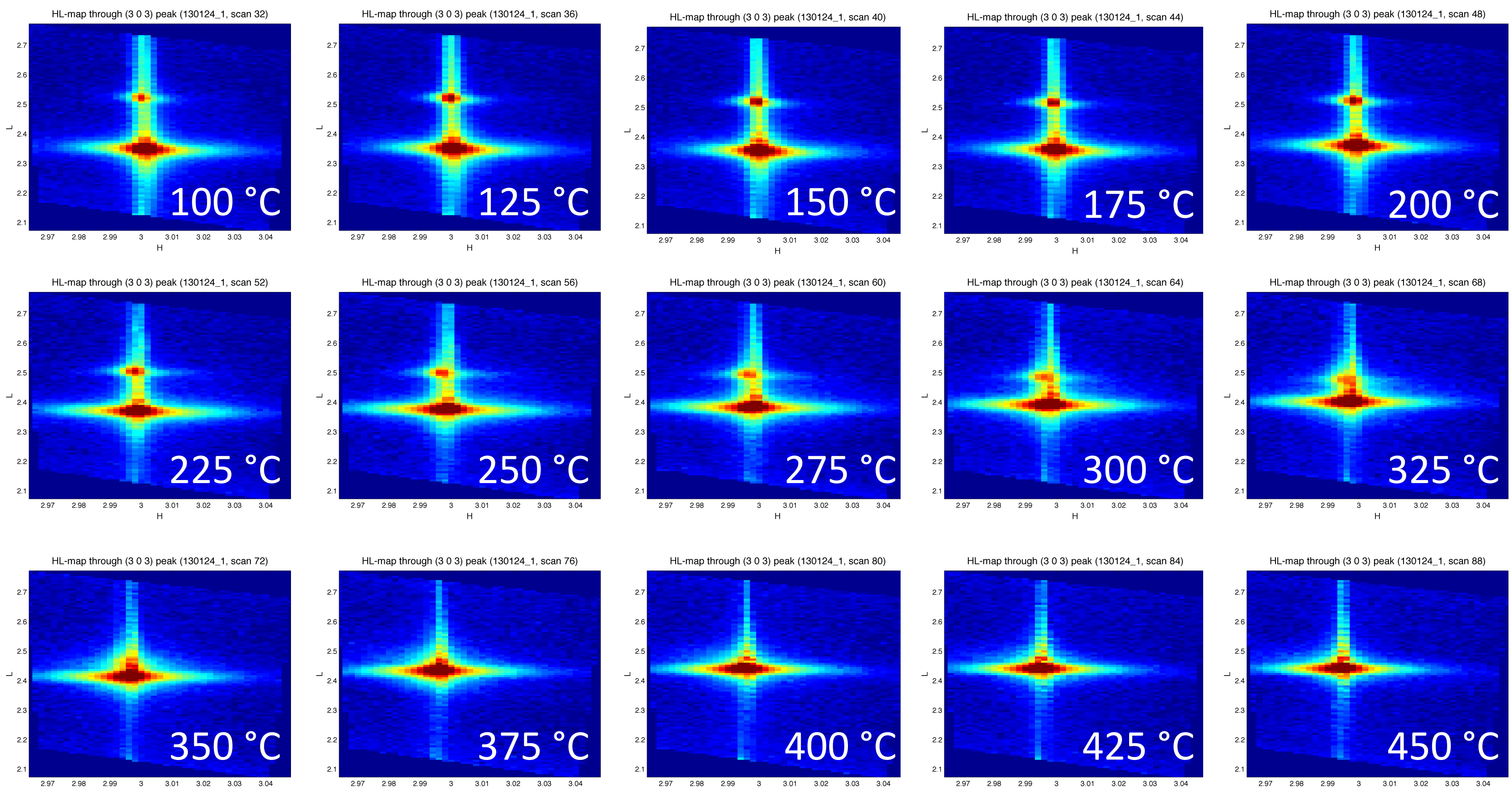




W. Siemons *et al.*, *Appl. Phys. Express* **4,** 095801 (2011).

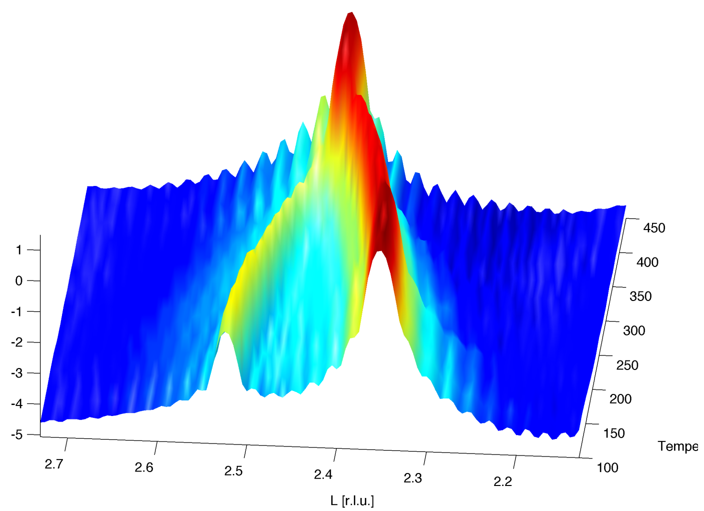
Temperature dependence

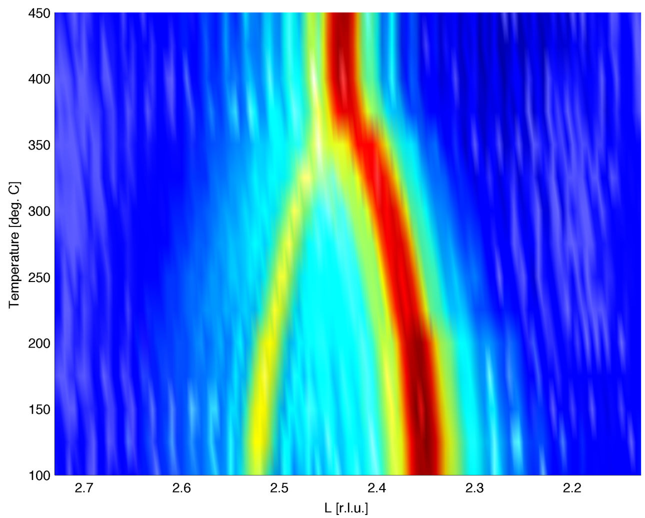
- $(303)_{pc}$ peak of BFO
- 18 nm thick film
- 100 °C – 450 °C



M_A to T phase transition

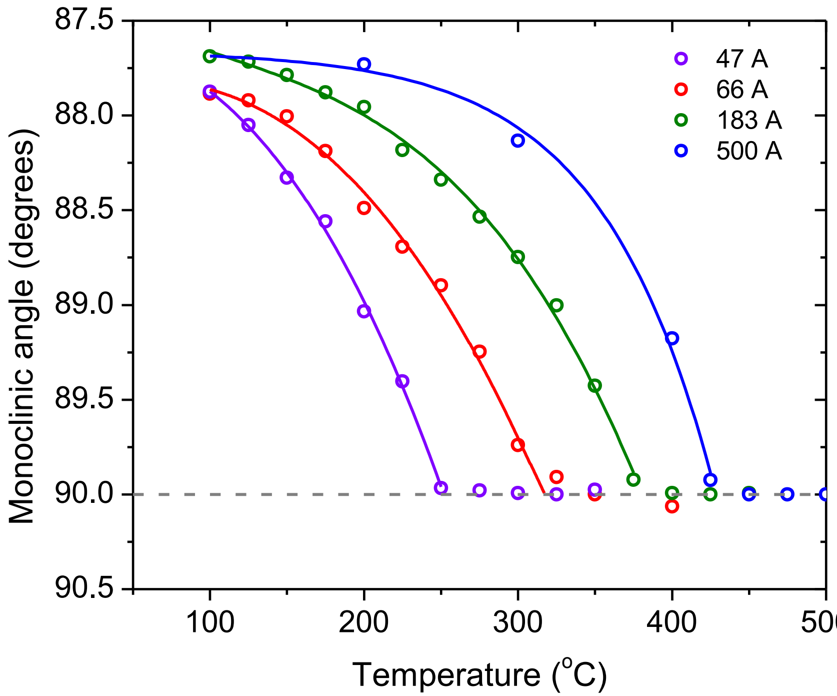
- Take line profile along L (out-of-plane) at H,K = (3,0).
- Peak splitting changes as a function of temperature.
- Amount of splitting yields monoclinic tilt angle.
- Single peak indicates absence of monoclinic tilts ➔ formation of tetragonal structure.

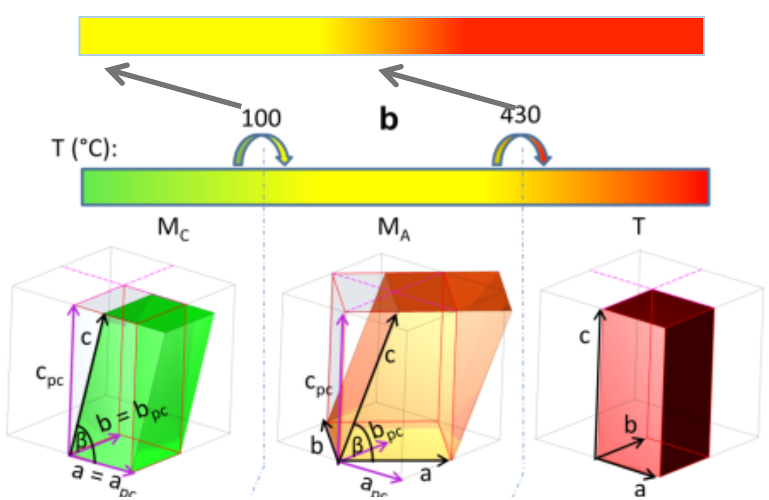




Phase transition temperature vs. film thickness

- Decreasing the film thickness decreases the transition temperature for the M_A to T phase transition.
- Same is true for the M_C to M_A transition.
- The whole structural phase diagram seems to be pushed to lower temperatures.

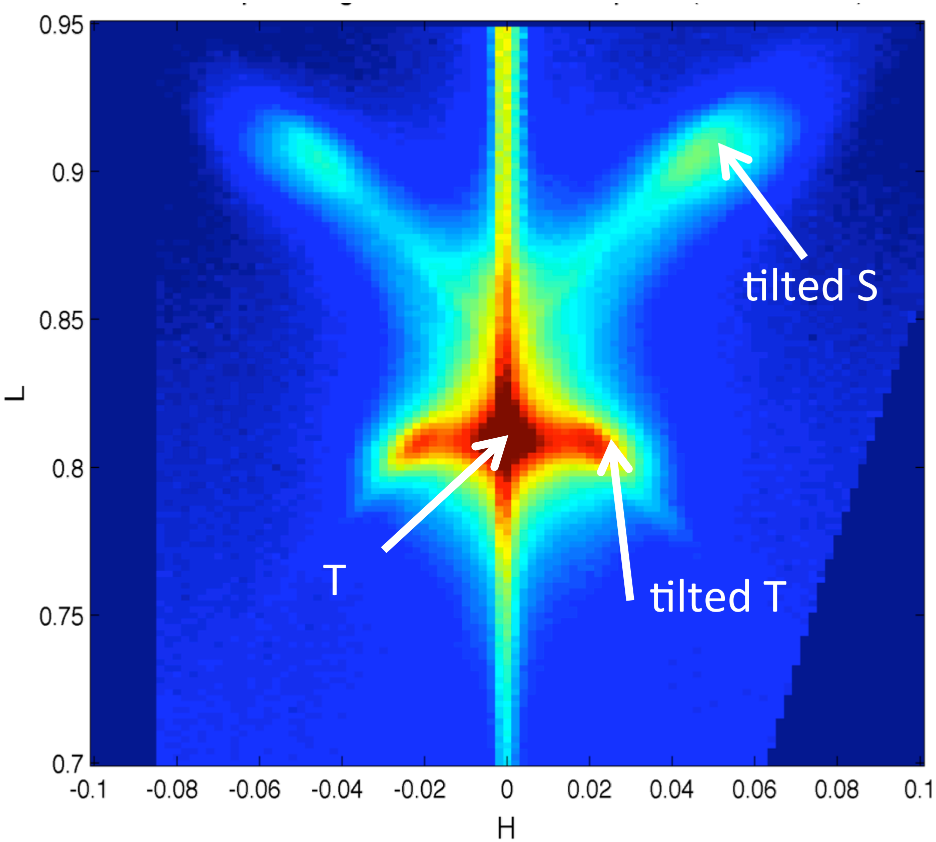




Courtesy of W. Siemons

A new phase!

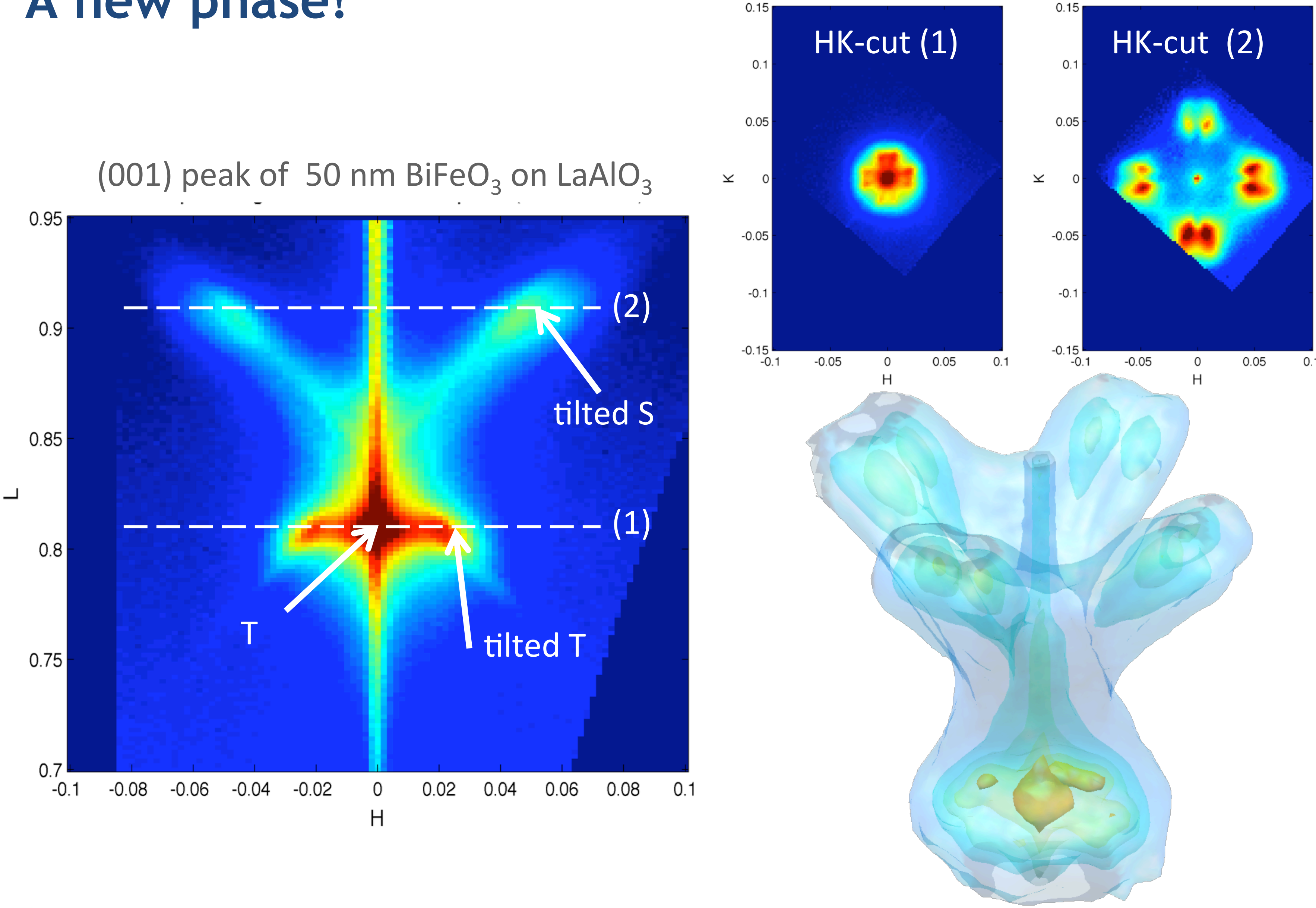
(001) peak of 50 nm $BiFeO_3$ on $LaAlO_3$



- New intermediate tilted "S" phase.
- First discovered by H. Christen *et al*.
- c/a ~ 1.09 (T: c/a ~ 1.24)
- Only a tilted variant is present, no peak in the θ-2θ direction.
- Up to 8 "S"-domains.
- T-like M_A phase is also partially tilted.
- Streaks connect tilted "T" and "S".
- Gradual changeover from tilted variant of "T" to "S"?
- Microdiffraction: "S" is only present at domain boundaries.

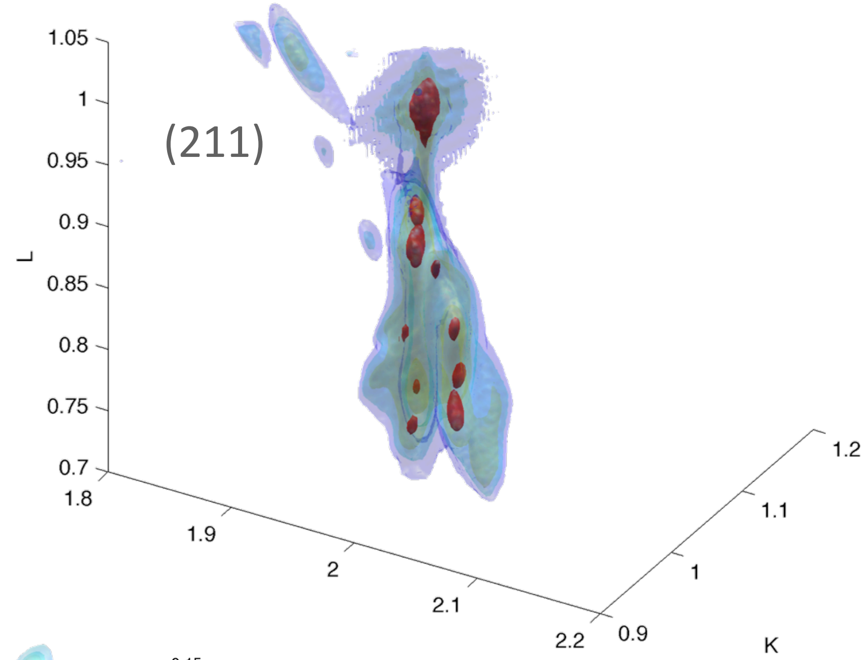
A new phase!

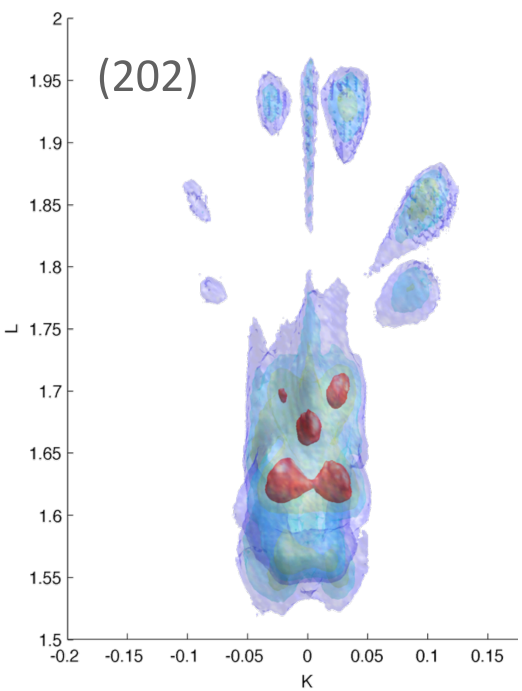
(001) peak of 50 nm $BiFeO_3$ on $LaAlO_3$

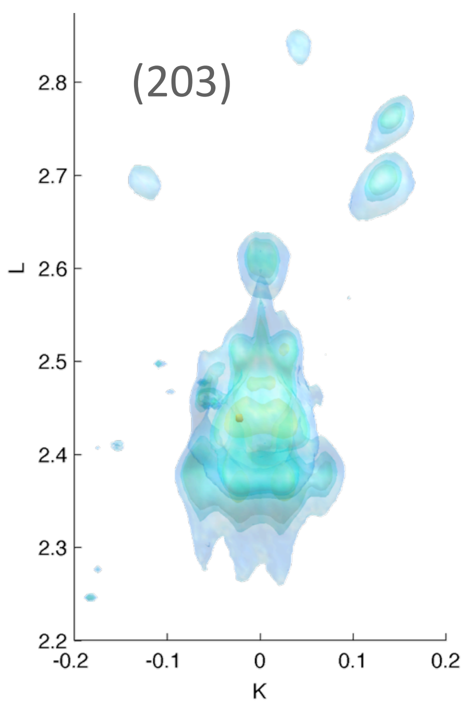


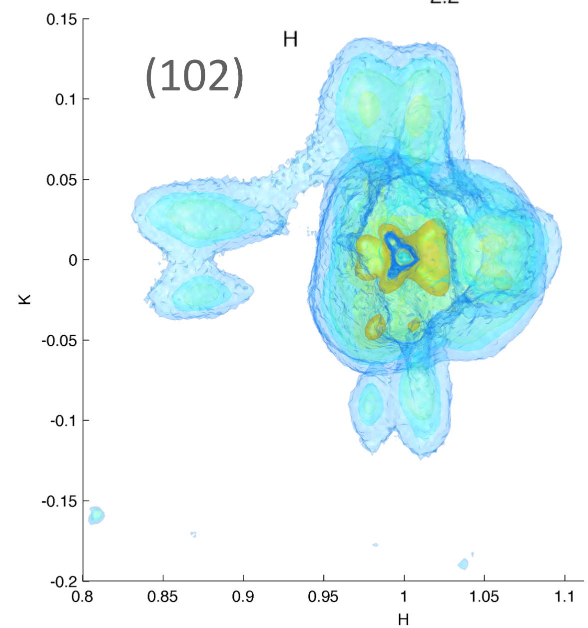
Welcome to the Zoo!

- Over 300 peak positions extracted.
- Refined triclinic unit cell parameters in presence of 8 domains.
- work in progress…

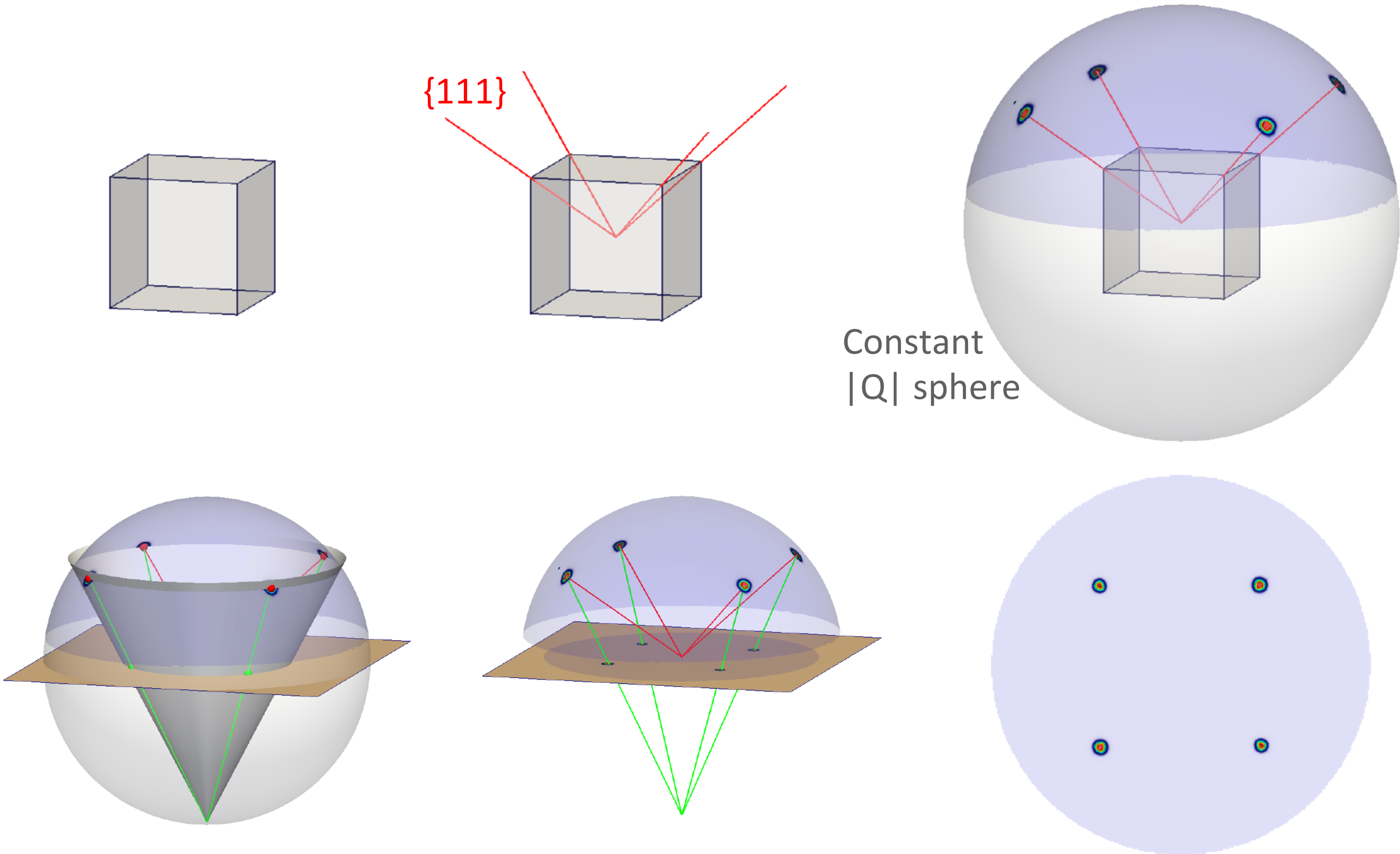




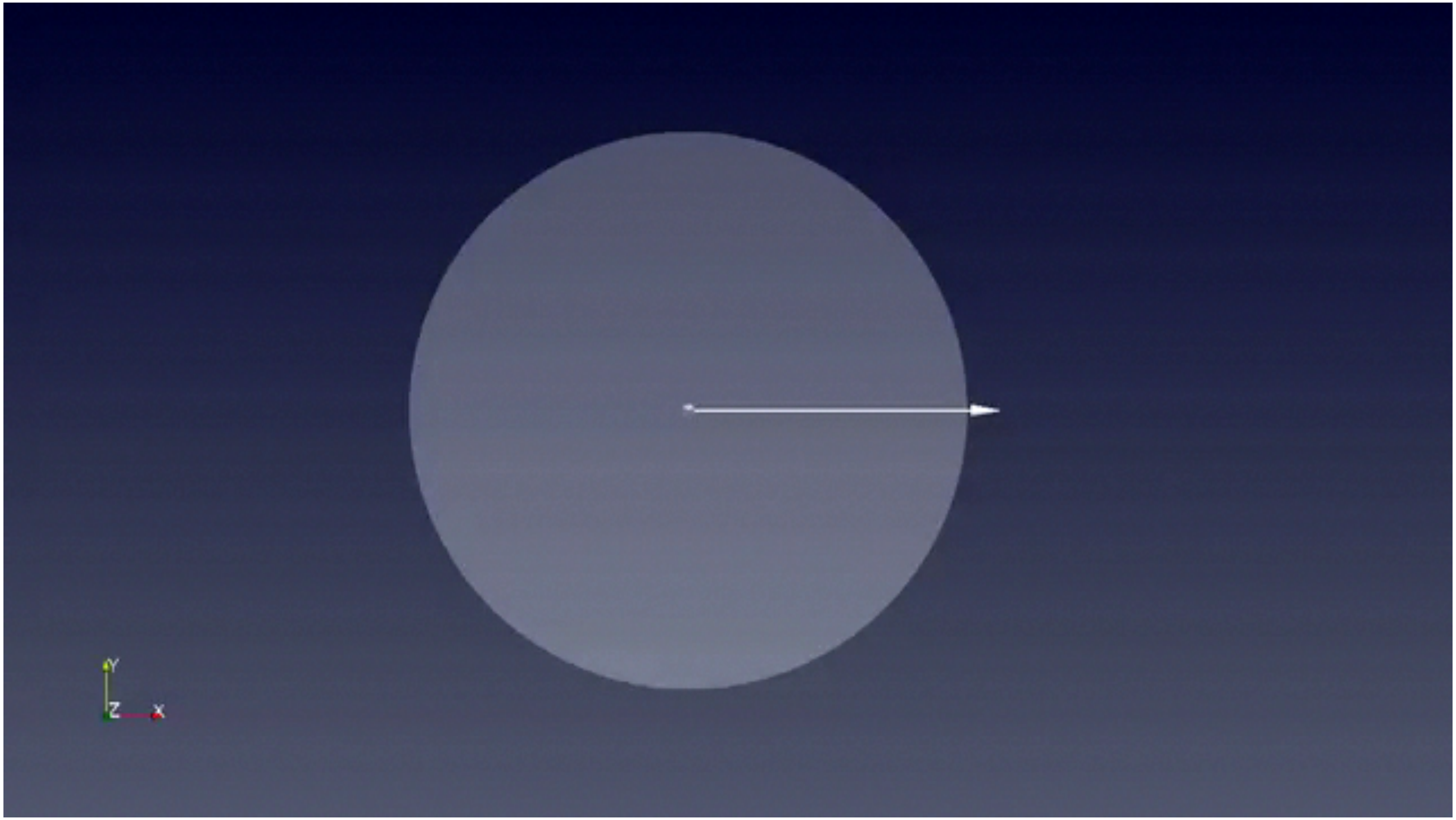




X-ray Pole Figures: Example YSZ(100)

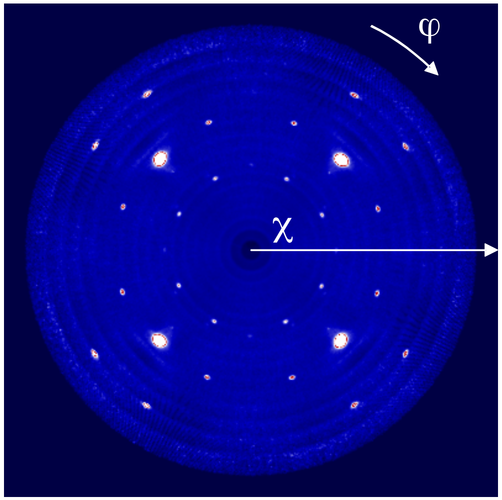


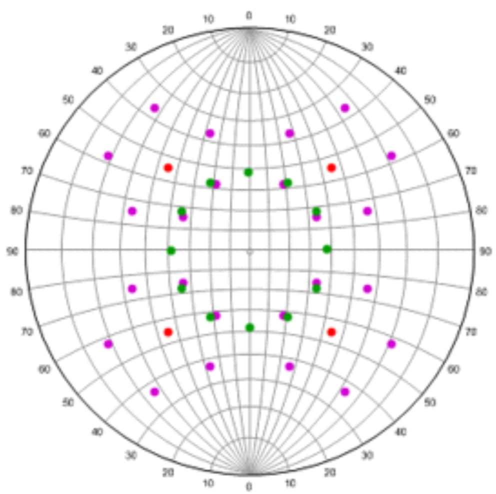
Working with 3D pole figure data



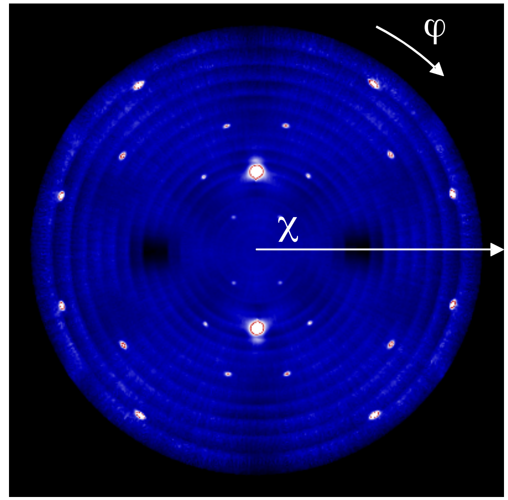
Pole Figures: Sample between *d* = 2.45-2.75 Å

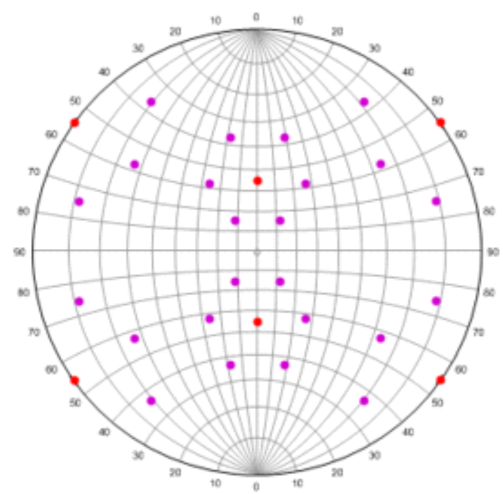
||YSZ(100)



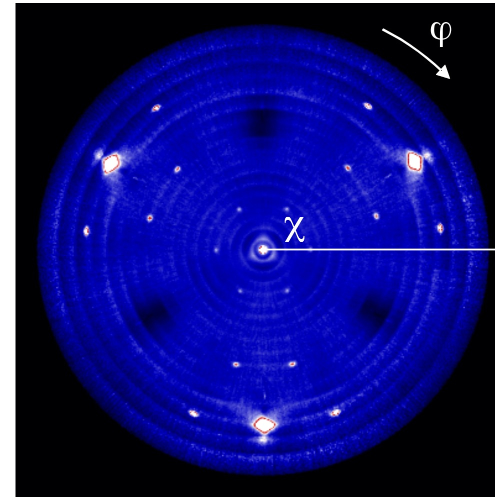


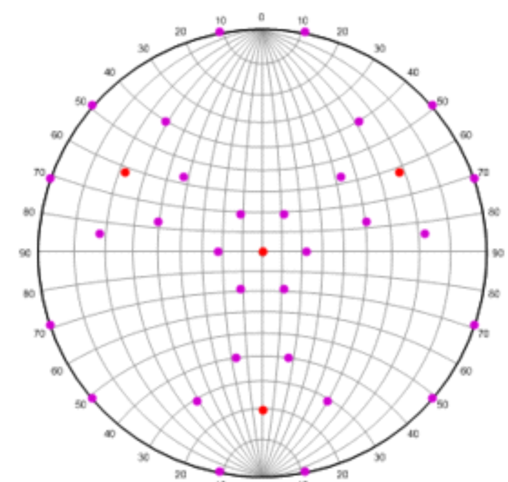
||YSZ(110)



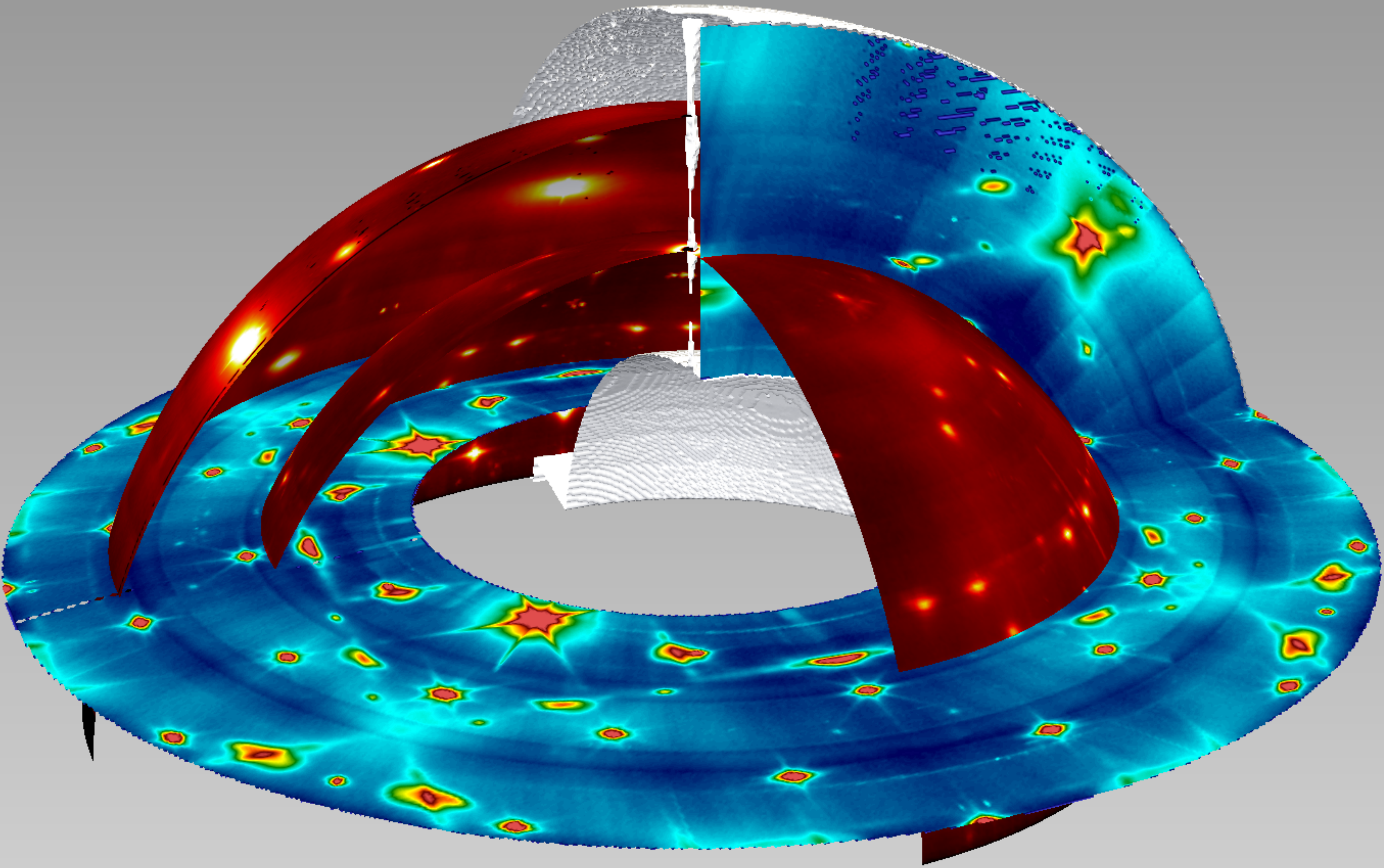


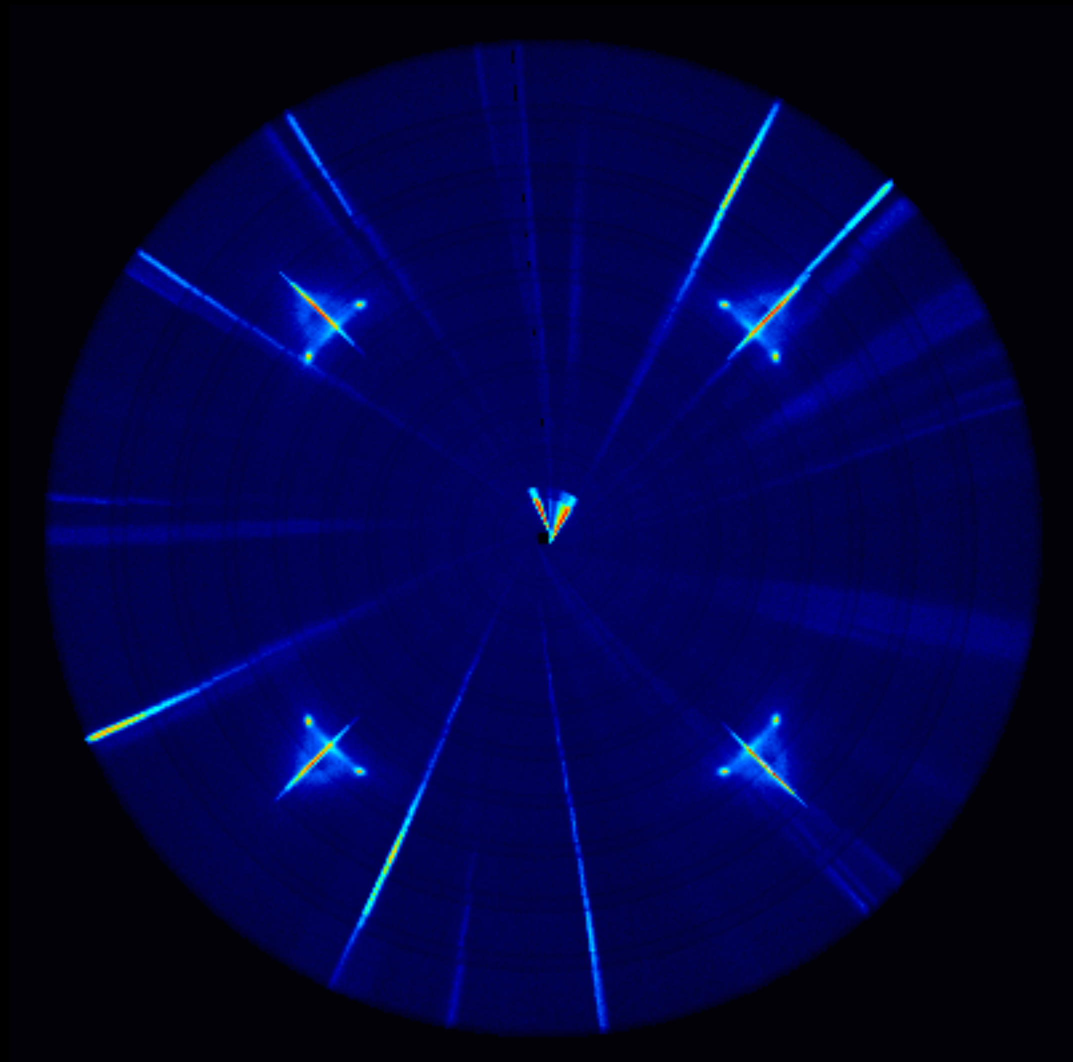
||YSZ(111)





● α-$Fe_2O_3\{10\bar{1}4\}$ ● β-$Fe_2O_3\{222\}$ ● ITO{123}





Challenges and outstanding issues

- Limitations imposed by current version of xrayutilities:
  - Limited support for arbitrary detector orientations
  - No support for arbitrary diffractometer rotation directions (must currently be along coordinate axes)
  - No consistent mechanism to mask out bad pixels
  - No easy way to keep track of per pixel (detector) or per voxel corrections and statistics
  - No straightforward support for energy scans.
- Missing support for non-orthogonal unit cells when representing data in HKL coordinates.
- Need more options for output formats
- Include powder diffraction analysis in the GUI?
- Need well-defined and standardized input data format that contains all necessary information:
  - HDF5
  - NeXuS
  - other?
  - input from other beamlines welcome!
- Simplify installation procedures for users (xrayutilities is difficult to compile at times)
- User documentation! Wiki?
- Start on libraries for data analysis based on volume sets

Conclusions

- 3D Reciprocal space exploration:
  - Powerful tool for structure investigations.
  - Fast data acquisition with area detectors.
  - “Normal scans” give 3D volume “for free”.
  - Identical setup for powder diffraction, pole figures, RSMs, etc.

- **rsMap3D**
  - Great support from Software Services Group (SSG), and John Hammonds in particular
  - Tested very successfully on a variety of different systems and use cases on 3 different beamlines (33-BM-C, 33-ID-D, 13-BM-C)
  - Ready for “Guinea Pig” users
  - Pilot user feedback will help to improve functionality and fix bugs
  - John Hammonds (SSG) is working on expanding the scope of the tool to deal with energy scans at 34-ID-E.
  - Involve other beamlines that could benefit from this tool

Pilot users wanted!

33-BM-C offers "crystallography service" in the 2014-2 cycle (May – August):

- My staff time – no extra proposals needed
- Short measurements that are:
  - Difficult to impossible to do in the lab
  - "Fast and easy" to do at the Synchrotron with an area detector
  - Answer important questions to move ahead with research on any project
  - Well defined scope of the measurements
- Dates:
  - Mon, June 2 (16:00) - Tue, June 3, (08:00)
  - Mon, June 9 (16:00) - Tue, June 10, (08:00)
  - Wed, June 18 (16:00) - Mon, June 23 (08:00)
  - Wed, July 16 (16:00) - Fri, July 18 (08:00)
  - Mon, July 28 (16:00) - Tue, July 29 (08:00)
  - Maybe: Tue, July 1 (08:00) – Thu, July 4 (08:00)

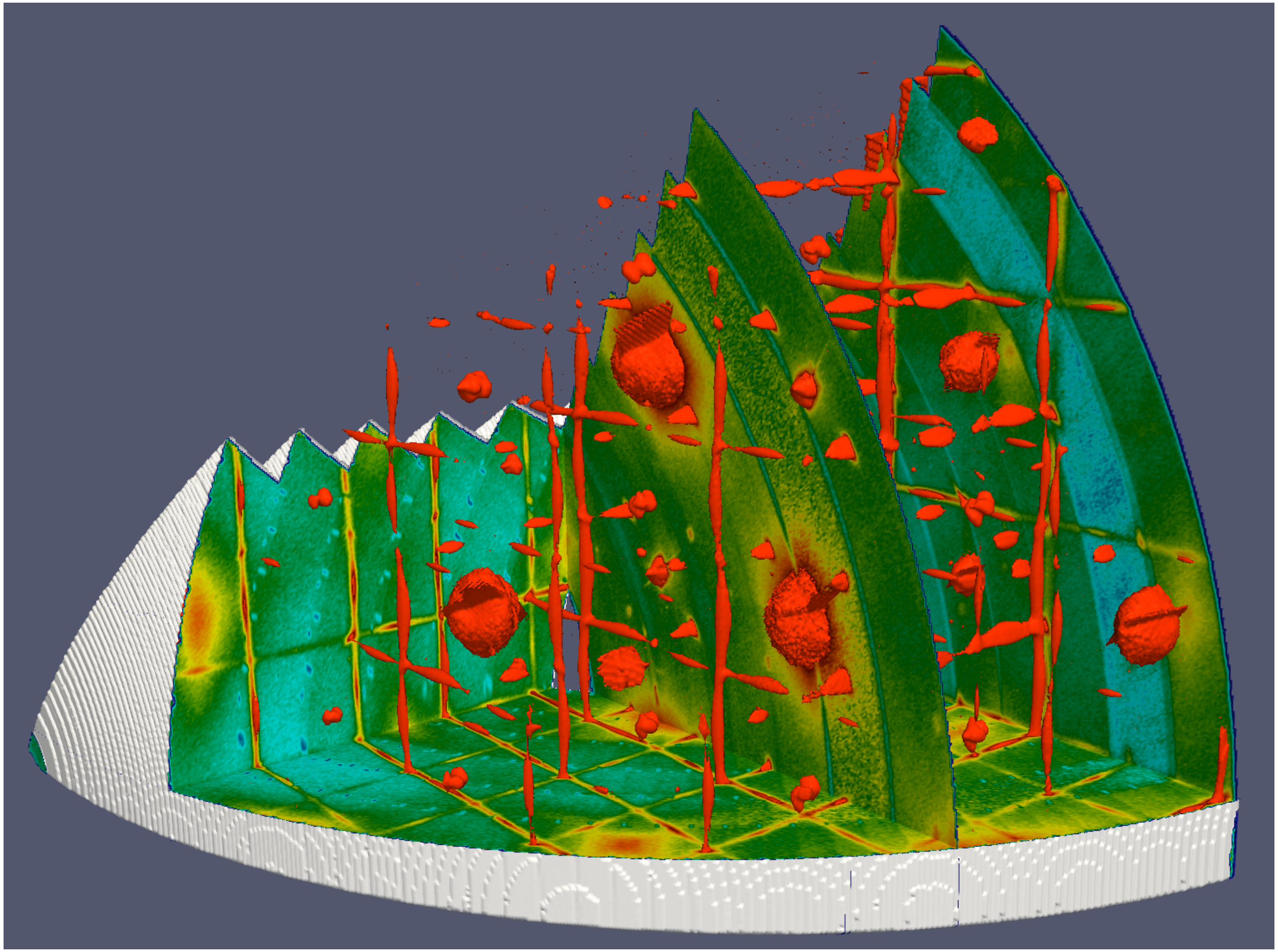
Contact me: cschlep@aps.anl.gov

Thank you!

Questions?

Contact me: cschlep@aps.anl.gov

Gallery



Gallery