PAUL SCHERRER INSTITUT

Stavros Samothrakitis & Camilla B. Larsen :: Applied Materials Group :: Paul Scherrer Institute

Laue and time-of-flight neutron diffractive imaging methods for 3D grain mapping of polycrystalline materials

ESS ILL Joint User Meeting 2022

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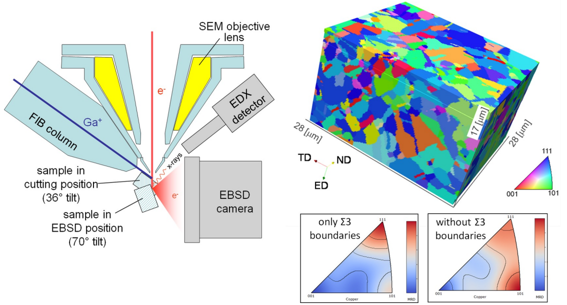
Laue 3DNDT & 6DXND

Introduction & Motivation

- **Grain orientation distribution, size and shape, among other microstructural features are directly connected to macroscopic properties (mechanical, magnetic, etc.) of polycrystalline meterials.**

- **Detailed knowledge of the crystallographic grain network, not only on the surface but also in the bulk of samples, is essential for tunning and manipulating macroscopic properties for applications.**

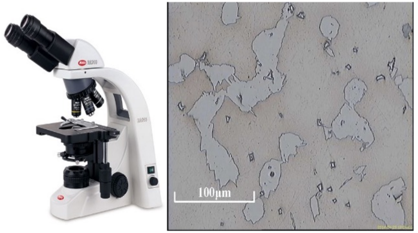
- **It is thus important to develop methods/tools that provide detailed information:**
  - **2D and 3D,**
  - **Distructive and non-distructive,**
  - **Optical light, electrons, X-rays, neutrons**



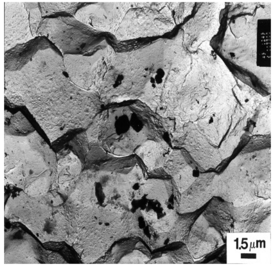
3D EBSD

Figures taken from: https://www.mpie.de/3093283/Techniques3DEBSD

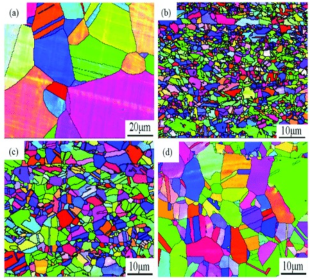
Optical Microscopy

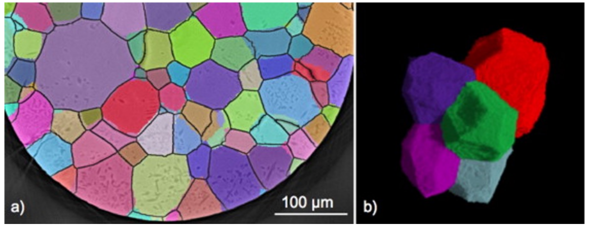


SEM



EBSD





3DXRD

Materials Science and Engineering: A, Vol. 524, (2009), Pp. 69-76

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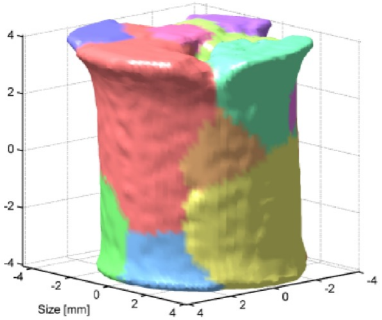
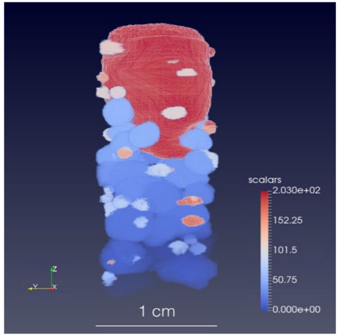
Cold neutron diffraction contrast tomography



Analyst, 2014, *139*(22), 5765-5771.



Time-of-Flight Three Dimensional Neutron Diffraction in Transmission Mode

Scientific Reports, 2017, 7(1), 9561.

Introduction & Motivation

- **Grain orientation distribution, size and shape, among other microstructural features are directly connected to macroscopic properties (mechanical, magnetic, etc.) of polycrystalline meterials.**

- **Detailed knowledge of the crystallographic grain network, not only on the surface but also in the bulk of samples, is essential for tunning and manipulating macroscopic properties for applications.**

- **It is thus important to develop methods/tools that provide detailed information:**
  - **2D and 3D,**
  - **Distructive and non-distructive,**
  - **Optical light, electrons, X-rays, neutrons**

Laue 3D neutron diffraction tomography

6D X-ray & neutron diffraction

Laue 3D Neutron Diffraction Tomography

Characteristics

FALCON - E11 - HZB

- **Not wavelength resolved ----> High Flux (also at continuous sources)**
- **Fast acquisition time (a few hours even minutes vs days)**
- **Use of simple Laue setup; easy to perform**

<u>Comparison with Cold neutron diffraction contrast tomography:</u>

Monochromatic neutron beam with 3 full rotations

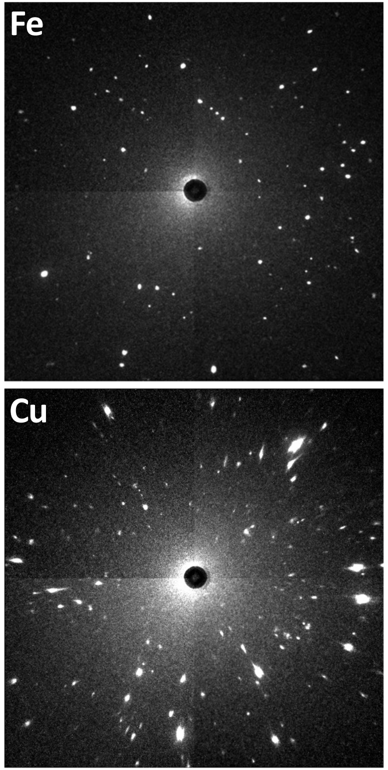
+ Flux
+ Information

White THERMAL neutron beam with a single rotation

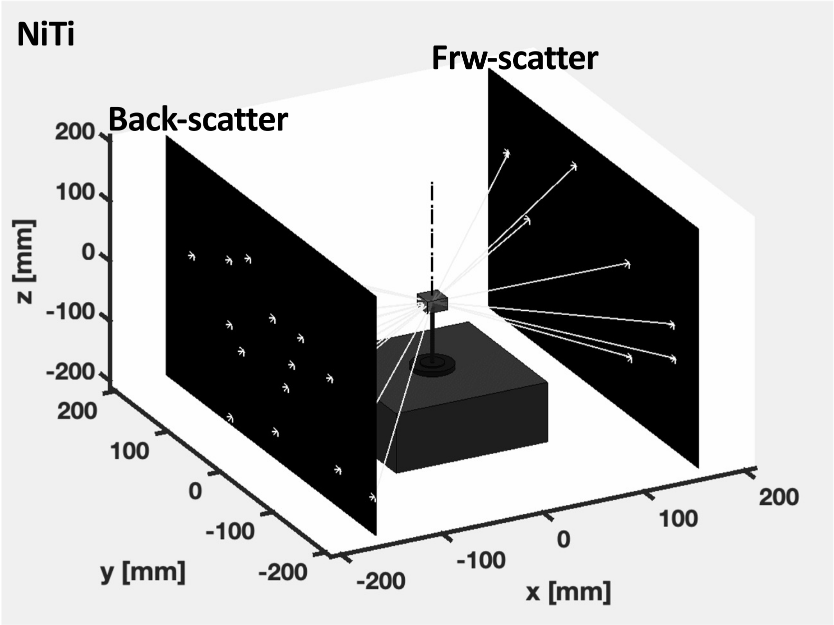
Laue 3D Neutron Diffraction Tomography

Characteristics

FALCON - E11 - HZB



Tomographic Measurements



Laue 3D Neutron Diffraction Tomography

Characteristics

In a diffraction experiment the main equation that provides information is the <u>Bragg equation:</u>

$$2d\cdot\sin\theta = n\lambda$$

Cubic:

$$\frac{1}{d^2} = \frac{h^2+k^2+l^2}{a^2}$$

Tetragonal:

$$\frac{1}{d^2} = \frac{h^2+k^2}{a^2} + \frac{l^2}{c^2}$$

Hexagonal:

$$\frac{1}{d^2} = \frac{4}{3}\left(\frac{h^2+hk+k^2}{a^2}\right) + \frac{l^2}{c^2}$$

In polychromatic case ($\lambda_1 < \lambda < \lambda_2$), the recorded data is integrated over the full wavelength range available and thus information is lost.

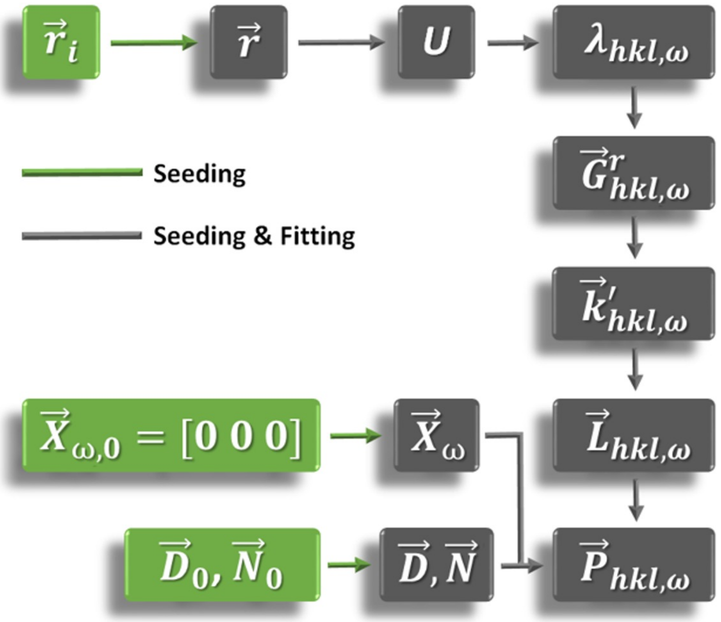
$$2\cancel{d}\cdot\sin\theta = n\cancel{\lambda}$$

Laue 3D Neutron Diffraction Tomography

Characteristics

How to solve the problem and find potential solutions ???

Use of a *forward model* !!



What is required?

- Sample information: lattice parameters and space group that will be used with Mantid to generate the samples *hkl* planes and the corresponding d-spacing (also the multiplicity and structure factors squared).
- Possible grain orientations, originally input as Rodrigues vectors, r, and then transformed into orientation matrices, U, (total number of orientations are user defined but the values are confined by the fundamental zone of the crystal system).
- Possible grain position, originally assumed at (0,0,0) and refined later.
- The omega range, detector parameters, and wavelength range.

Laue 3D Neutron Diffraction Tomography

Original Development

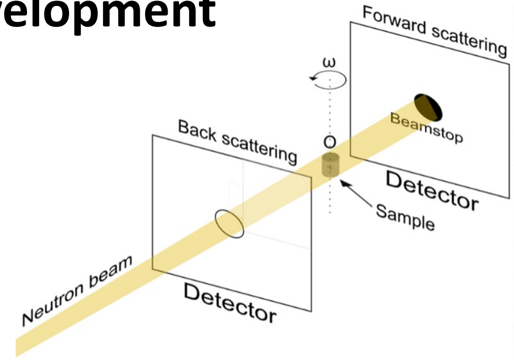
SCIENTIFIC REPORTS

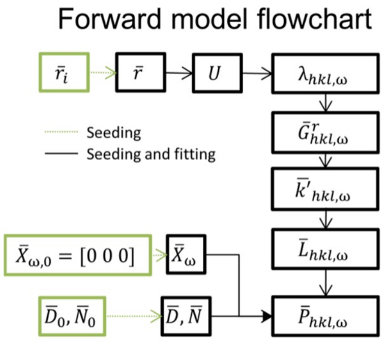
Article | Open Access | Published: 18 March 2019

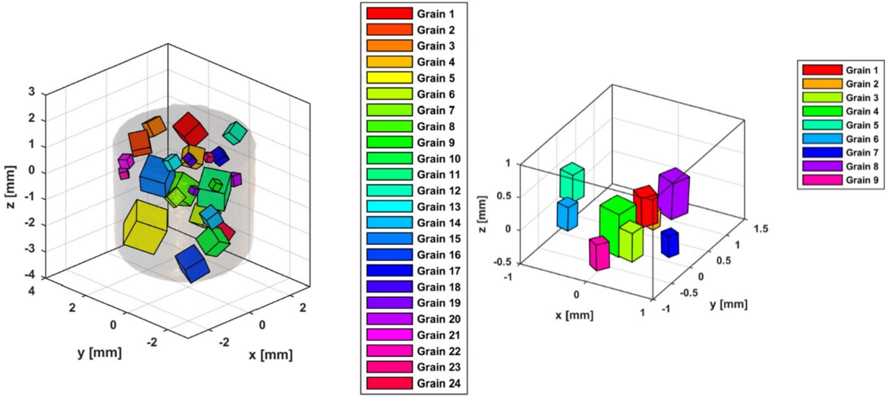
Laue three dimensional neutron diffraction

Marc Raventós, Michael Tovar, Marisa Medarde, Tian Shang, Markus Strobl, Stavros Samothrakitis, Ekaterina Pomjakushina, Christian Grünzweig & Søren Schmidt

Scientific Reports **9**, Article number: 4798 (2019) | Cite this article



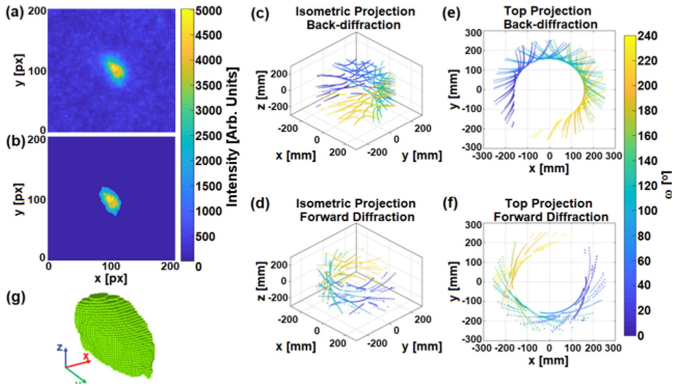


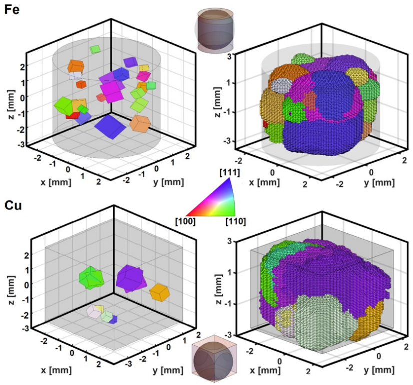


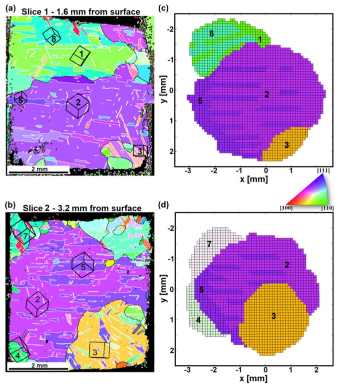
Laue 3D Neutron Diffraction Tomography

Grain Morphology Reconstruction

ASTRA toolbox
+
NVIDIA CUDA
+
vs







SCIENTIFIC REPORTS

Article | Open Access | Published: 28 February 2020

Grain morphology reconstruction of crystalline materials from Laue three-dimensional neutron diffraction tomography

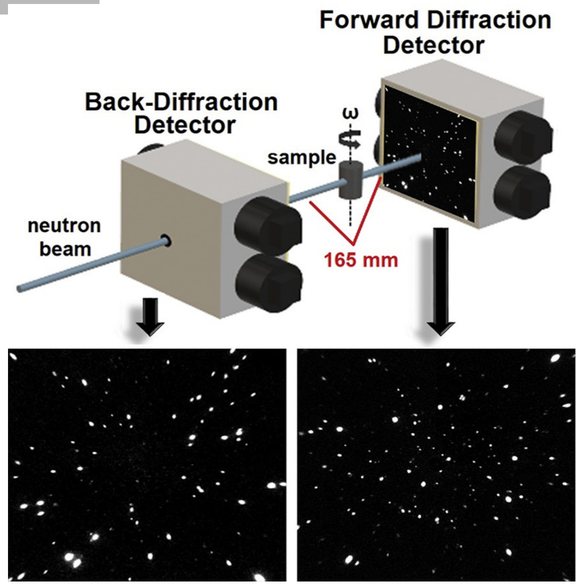
Stavros Samothrakitis, Marc Raventós, Jan Čapek, Camilla Buhl Larsen, Christian Grünzweig, Michael Tovar, Marina Garcia-Gonzalez, Jaromír Kopeček, Søren Schmidt & Markus Strobl

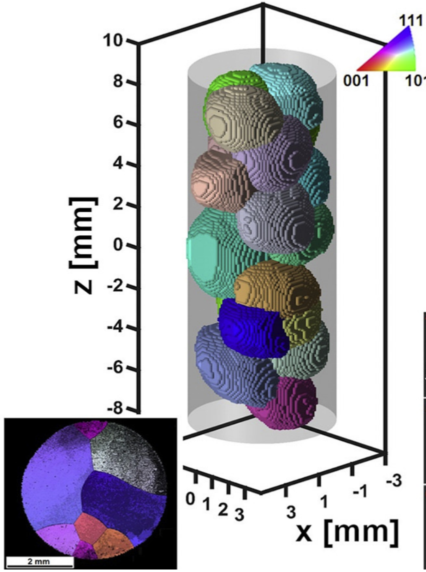
Scientific Reports **10**, Article number: 3724 (2020) | Cite this article

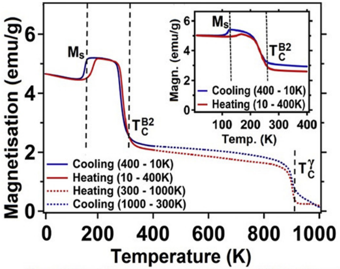
Laue 3D Neutron Diffraction Tomography

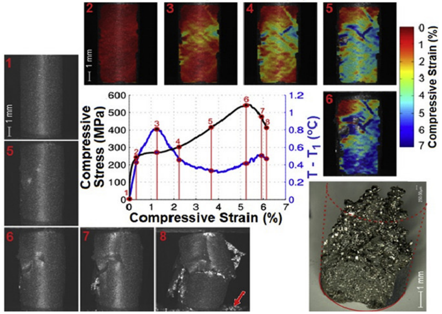
Applications

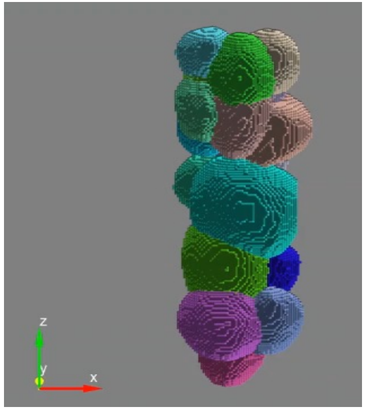
A multiscale study of hot-extruded CoNiGa ferromagnetic shape-memory alloys









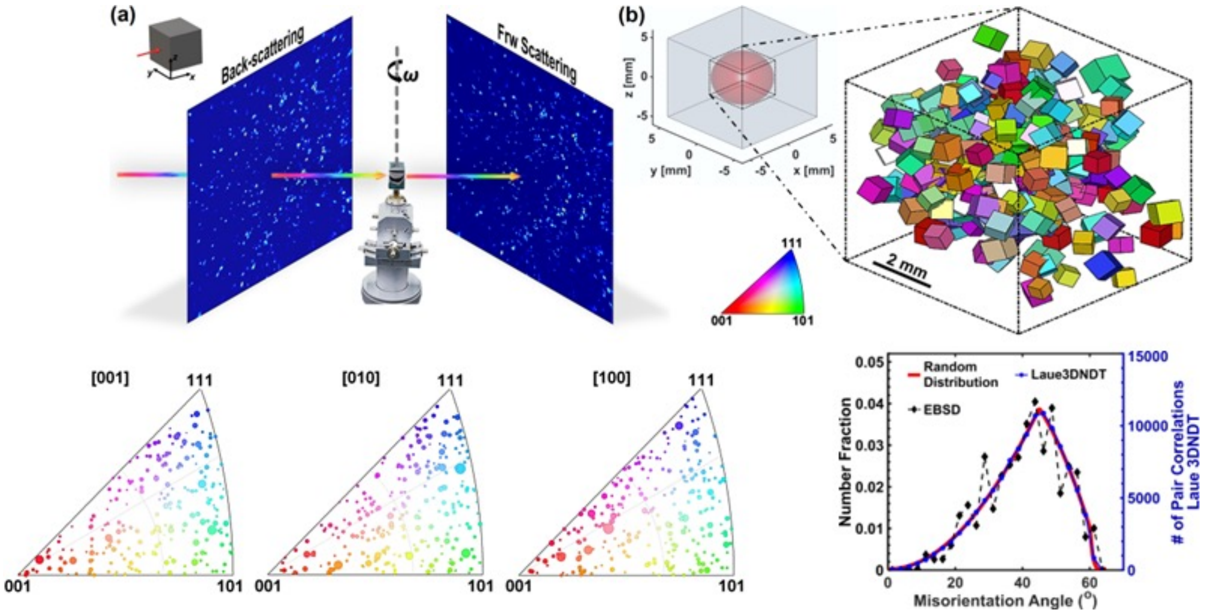


Materials & Design 196 (2020) 109118

Laue 3D Neutron Diffraction Tomography

Applications

Microstructural characterization through grain orientation mapping with Laue three-dimensional neutron diffraction tomography



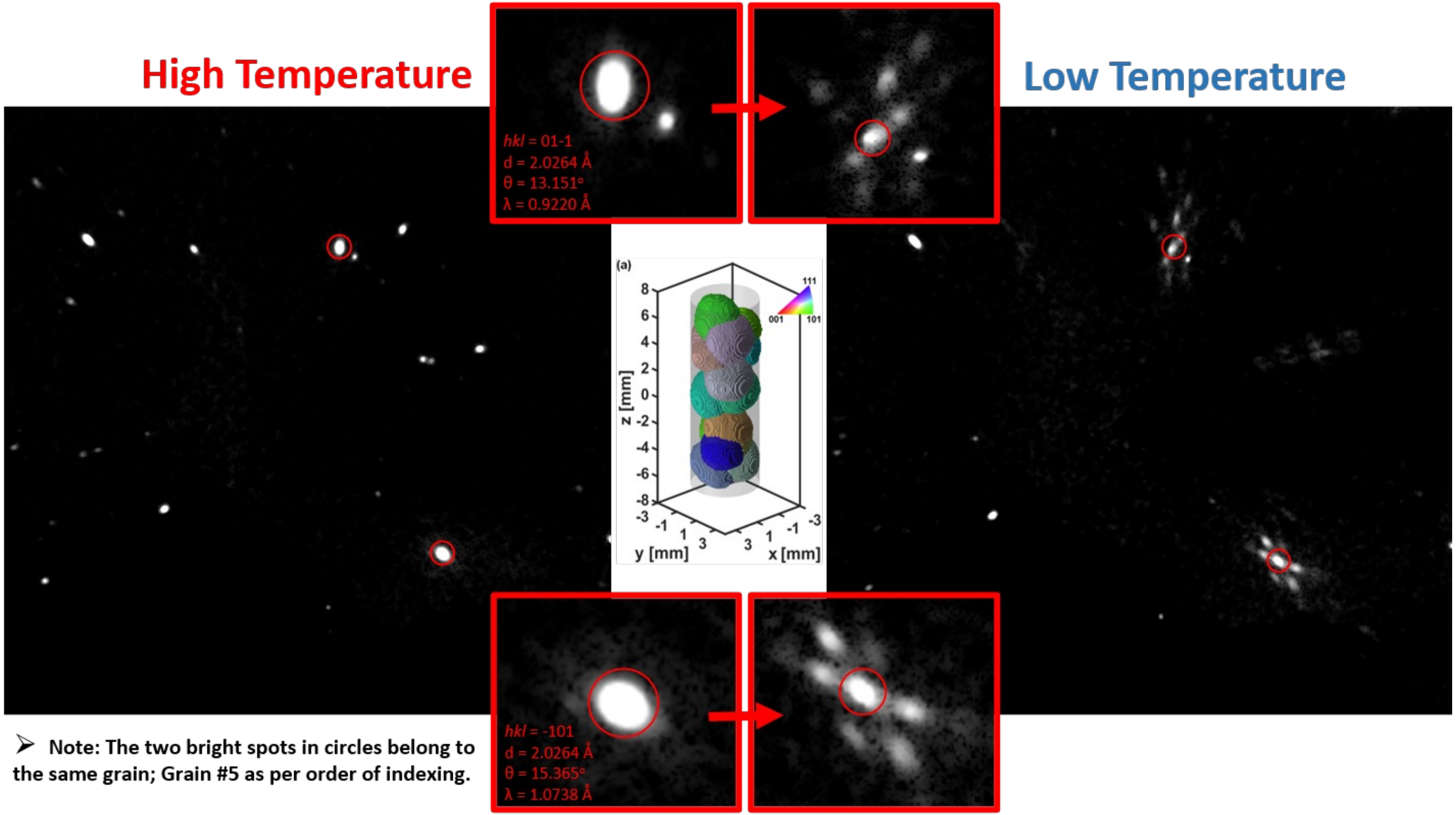
Materials Today Advances 15 (2022) 1002582

Laue 3D Neutron Diffraction Tomography

Applications

High Temperature

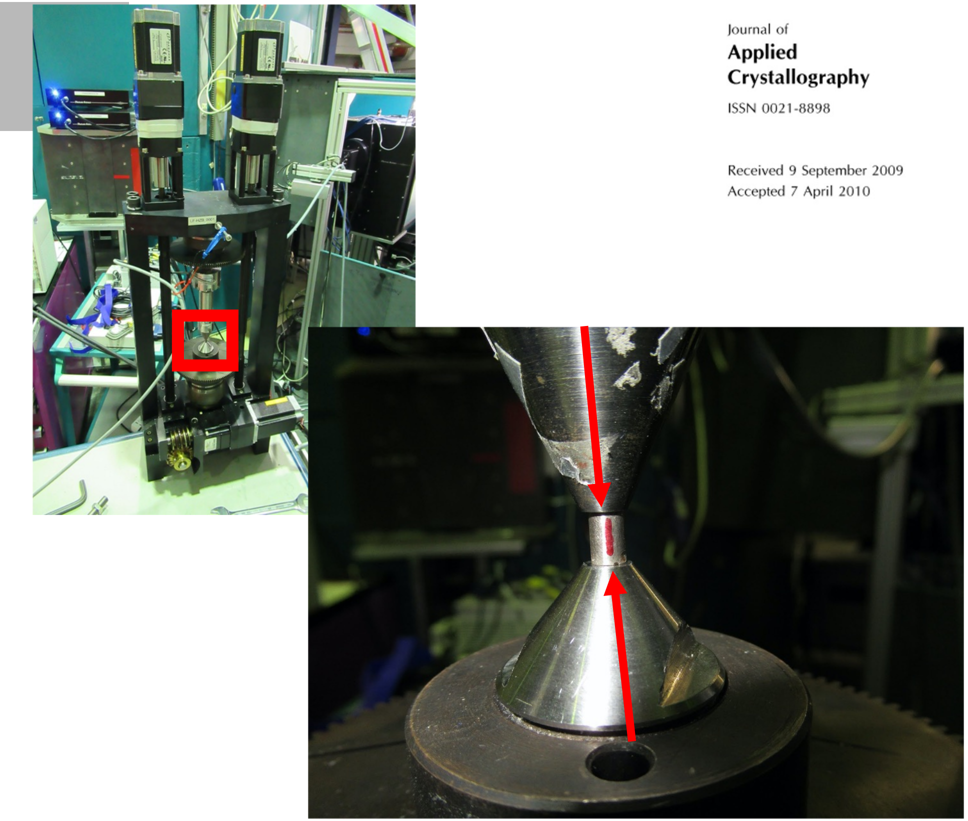
Low Temperature



➢ Note: The two bright spots in circles belong to the same grain; Grain #5 as per order of indexing.

Laue 3D Neutron Diffraction Tomography

Applications



Journal of
Applied
Crystallography
ISSN 0021-8898

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Accepted 7 April 2010

Determining grain resolved stresses in polycrystalline materials using three-dimensional X-ray diffraction

Jette Oddershede,[a*] Søren Schmidt,[a] Henning Friis Poulsen,[a] Henning Osholm Sørensen,[a] Jonathan Wright[b] and Walter Reimers[c]

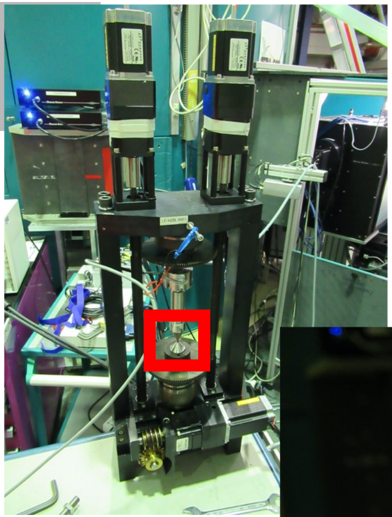
[a]Centre for Fundamental Research: 'Metal Structures in Four Dimensions', Riso DTU, DK-4000 Roskilde, Denmark, [b]ESRF, Grenoble, France, and [c]TU Berlin, Germany. Correspondence e-mail: jeto@risoe.dtu.dk

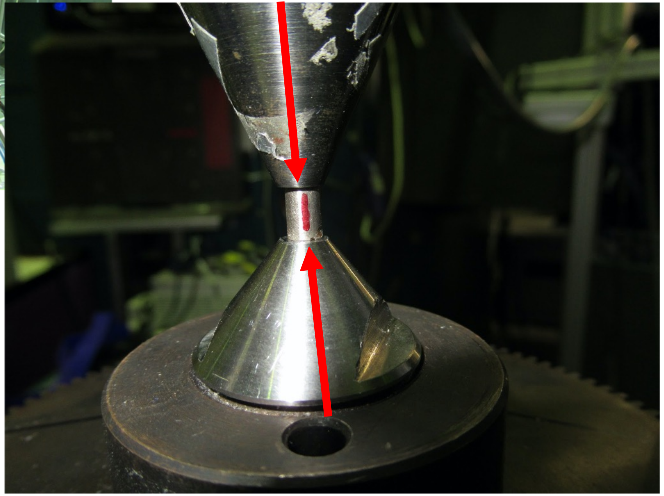
$$\underbrace{\boldsymbol{\Gamma}_{ij}^{-1}\overline{\mathbf{G}}_{ij}}_{\text{Experimental}} - \underbrace{\frac{\lambda}{2\pi}\mathbf{U}_i\mathbf{B}_i\overline{\mathbf{G}}_{hkl,ij}}_{\text{Theoretical}}$$

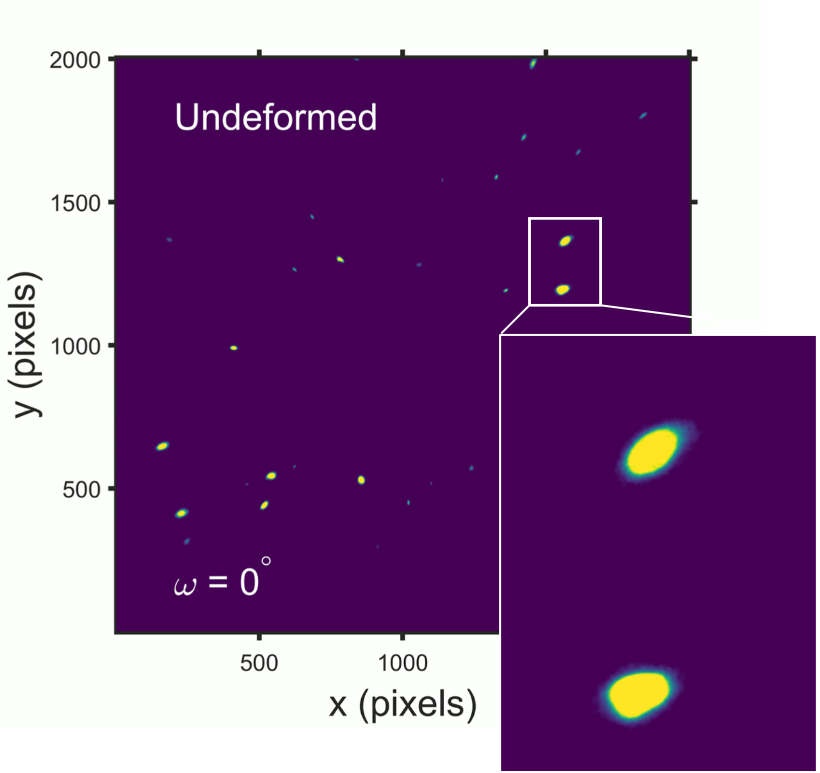
Experimental   Theoretical

Laue 3D Neutron Diffraction Tomography

Applications

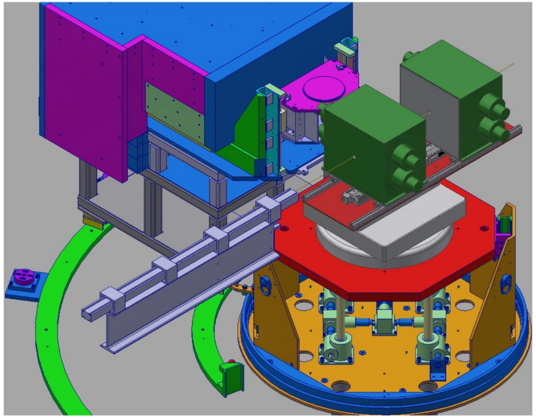


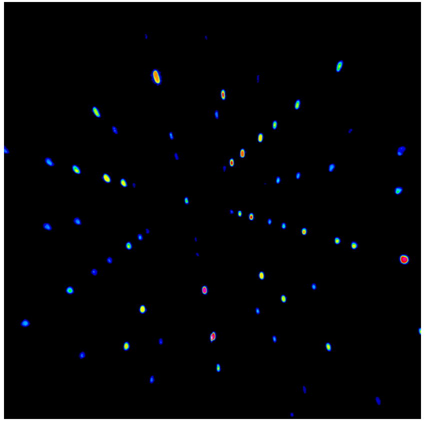


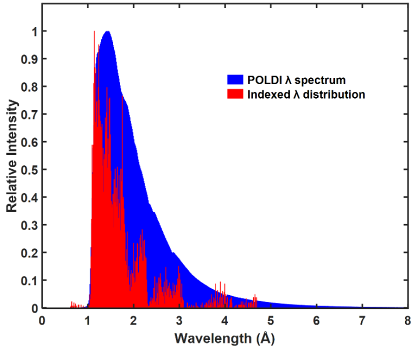


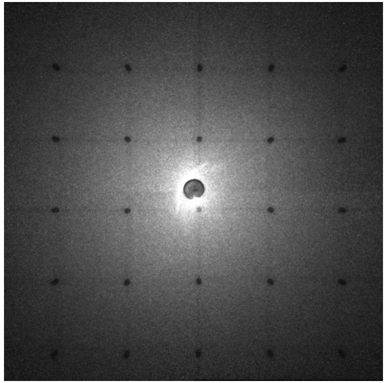
Laue 3D Neutron Diffraction Tomography

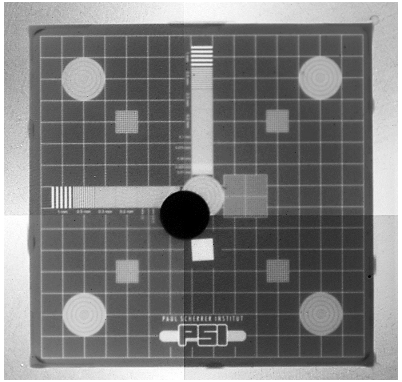
Future @ PSI









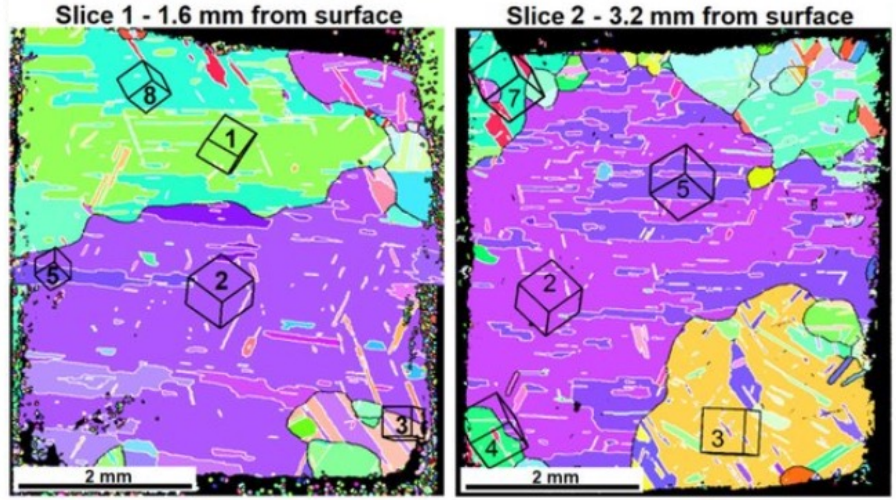


Funding: SDSC 4th call "Robust and scalable Machine Learning algorithms for Laue 3-Dimensional Neutron Diffraction Tomography" (2021).

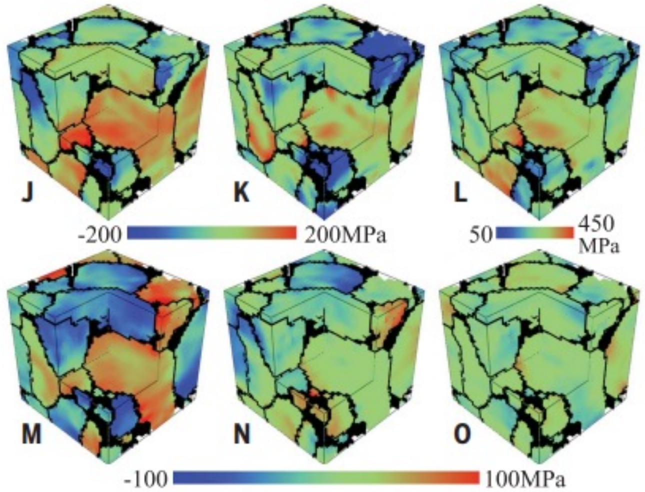
Future Development: 6DXND Tomography

6DXND: 6-Dimensional X-ray and Neutron diffraction

- **Why 6D: 3D (direct space) + 3D (orientation space)**
- **Motivation: Grain-resolved → subgrain spatial resolution**
- **Potentially better spatial resolution than Laue 3DNDT, but requires more experimental information (wavelength)**



Adapted from [S. Samothrakitis *et al., Scientific Reports 10, 3724 (2020)*]

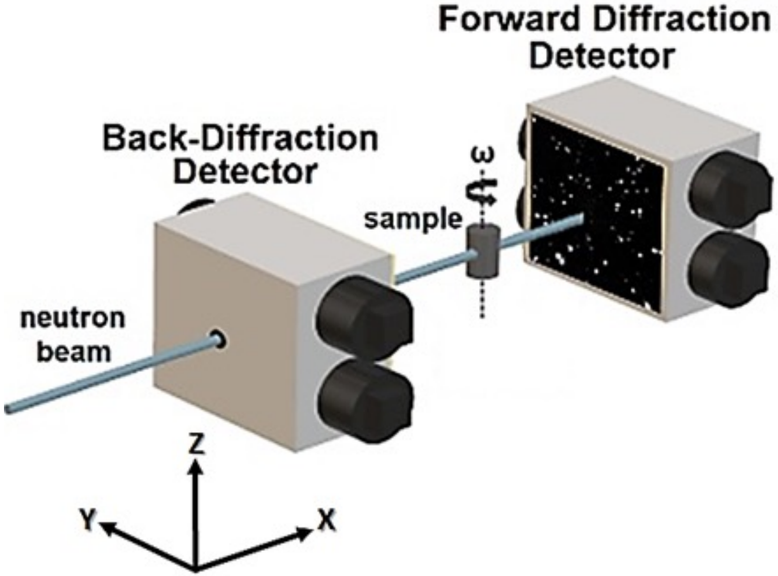


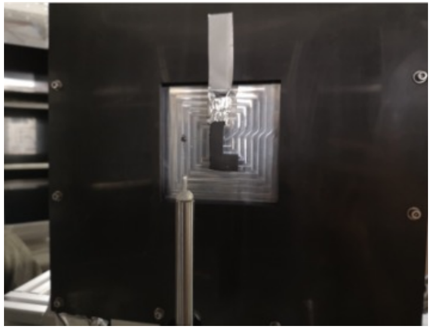
From [Y. Hayashi *et al., Science 366, 1492 (2019)*]

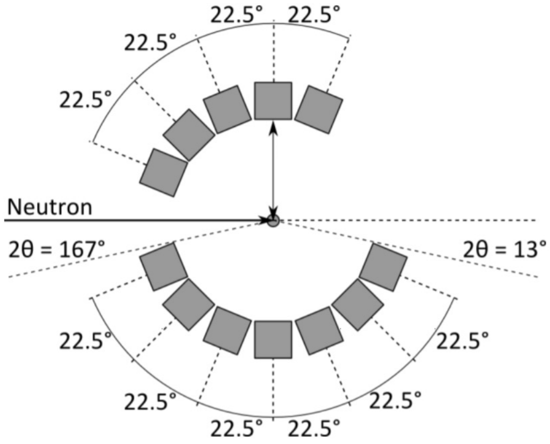
6DXND Tomography: Experimental setup

Similar experimental setup to Laue 3DNDT

- **Sample fully illuminated by beam**
- **Collect diffraction images at different sample rotations ω**







High resolution orientation distribution function (HRODF)

A discrete spherical x-ray transform of orientation distribution functions using bounding cubes

I G Kazantsev, S Schmidt and H F Poulsen

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E-mail: kazantsev1@netzero.net

Received 15 November 2008, in final form 9 August 2009
Published 16 September 2009
Online at stacks.iop.org/IP/25/105009

Abstract
We investigate a cubed sphere parametrization of orientation space with the aim of constructing a discrete voxelized version of the spherical x-ray transform. For tracing the propagation of a unit great circle through the $\mathbb{S}^3$ partition subsets, the frustums of the cubed sphere, a fast procedure is proposed. The circle's parts in each frustum are gnomonically mapped into line segments inside the bounding cubes. The line segments constitute a convex polygon with vertexes indicating frustum exit–entry points. Thus the problem of system matrix calculation is reduced to the tracing of line segments within rectangular voxel arrays partitioning the bounding cubes. Hence algebraic reconstruction techniques can be used in a comprehensive way for orientation distribution function estimation from diffraction data.

Materials Science Forum Vols. 702-703 (2012) pp 536-539
Online available since 2011/Dec/06 at www.scientific.net

doi:10.4028/www.scientific.net/MSF.702-703.536

HIGH RESOLUTION ORIENTATION DISTRIBUTION FUNCTION

Søren Schmidt[1,a] , Nicolai Fog Gade-Nielsen[2,b] , Martin Høstergaard[2] , Bernd Dammann[2,c] , and Ivan G. Kazantsev[3,d]

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[2]Dept. of Informatics and Mathematical Modelling, Technical University of Denmark, Denmark.

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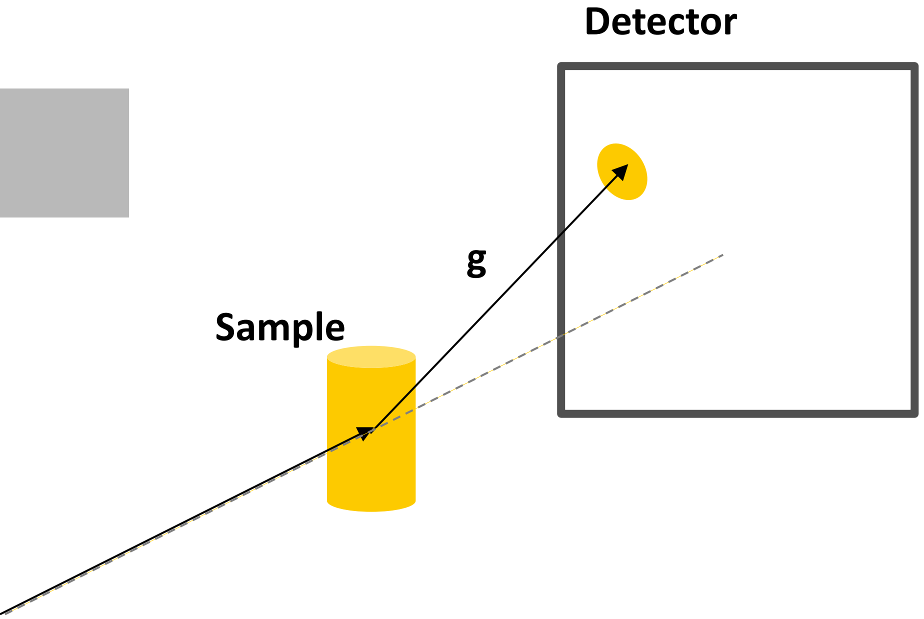
Keywords: ODF, x-ray, high resolution, inverse problems.

Abstract. A new method for reconstructing a High Resolution Orientation Distribution Function (HRODF) from X-ray diffraction data is presented. It is shown that the method is capable of accom-

6DXND: Mathematical formalism



g: experimental scatter vector
h: theoretical (unrotated) scatter vector

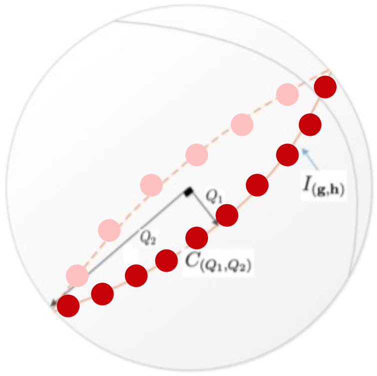
Determine orientation q such that h maps onto g

Use quaternion formalism:

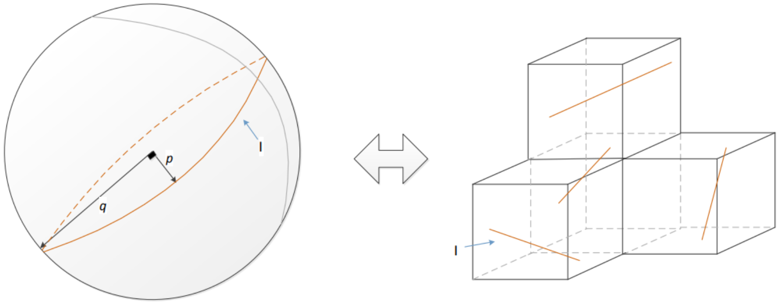
$$q = (q_0; q) = (\cos\frac{\alpha}{2}; \vec{n}\sin\frac{\alpha}{2})$$

Set of orientations is a sphere:

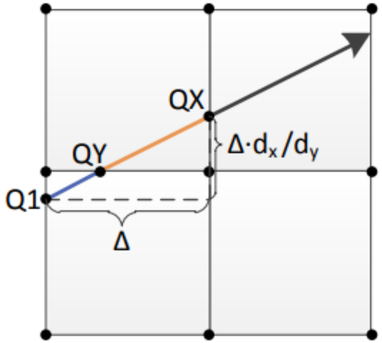
$$|\mathbf{q}| = \sqrt{q_0^2 + |\vec{q}|^2} = 1$$



6DXND: Mathematical formalism

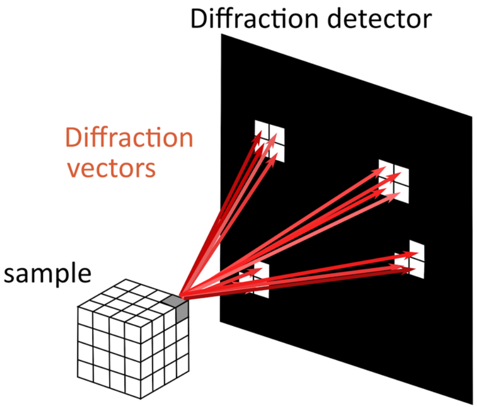


- **Set of solutions can be represented as straight lines**
  - **Problem reduces to solving a set of linear equations**
  - **Can be solved fast on gpus**
  - **Orientation space is discretized, and possible solutions explored through raytracing**

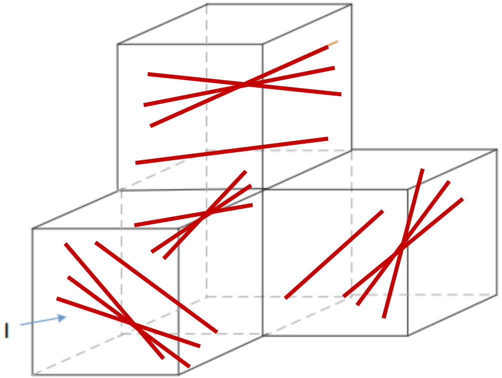


6DXND: Algorithm

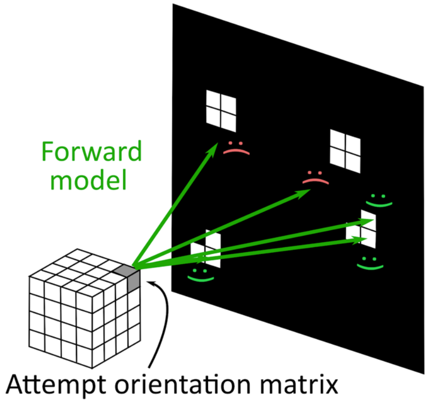
Step 1: Compute diffraction vectors



Step 2: Determine candidate orientations



Step 3: Completeness check



6DXND: Input and Output

Input: Binarized list of lit pixels per wavelength

ω	x	y
0	1	1
0	1	2
0	1	3
0	2	2
0	2	3
0	2	4
⋮	⋮	⋮
ω_N	N	N

λ_1 … λ_i

Output: List of orientations and completeness per voxel

```
VOXEL (0, 0, 0) COUNT 1
Orientation:
[[     0.9158573,    -0.0323292,      0.4002003]
 [    -0.3969138,     0.0773878,      0.9145876]
 [    -0.0605385,    -0.9964768,      0.0580442]]
Completeness: 99.07%
VOXEL (1, 0, 0) COUNT 1
Orientation:
[[     0.9158573,    -0.0323292,      0.4002003]
 [    -0.3969138,     0.0773878,      0.9145876]
 [    -0.0605385,    -0.9964768,      0.0580442]]
Completeness: 99.07%
```

6DXND: Conclusions and outlook

6DXND advantages:

- **Sub-grain level spatial resolution**
- **Spot overlap not issue**
- **Fast – Simulations shown take ~3 h on single graphic card (rtx 2060) ~15 min on small cluster**

To be done:

- **Robustness tests and further developed visualization tools**

0.1 deg step:

5 deg step:

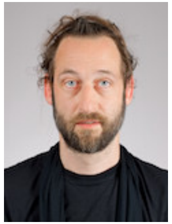
Acknowledgments

EUROPEAN SPALLATION SOURCE

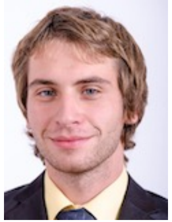

S. Schmidt


R. Woracek

PAUL SCHERRER INSTITUT


M. Strobl


E. Polatidis


J. Capek


M. Raventos

SDSC


T. Kacprzak

HZB Helmholtz Zentrum Berlin


M. Tovar

Thank you!

Questions?