What new things can we do with CDI at high coherence sources?

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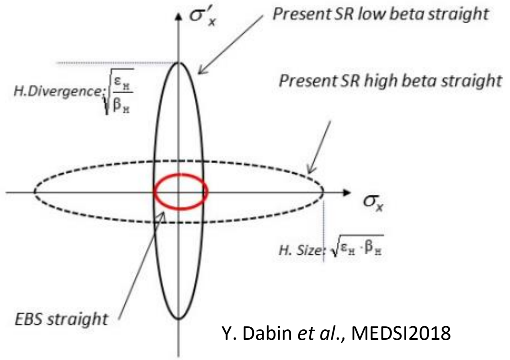
Introduction

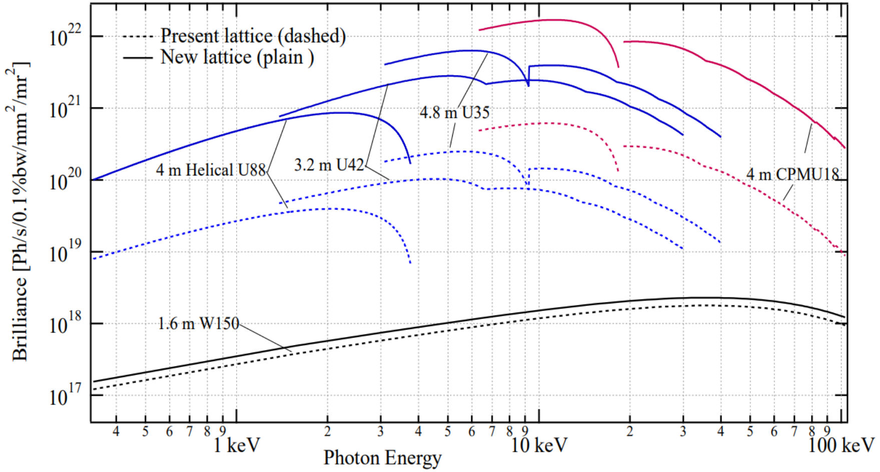
User perspective talk tailored towards the EBS and next generation storage rings

- EBS is 1-2 orders of magnitude more brilliance/coherent flux – Great!
- 10-100 times more coherent photons doesn't directly give us access to much inherently new science. We are going to have to think harder than that...
- Seeking to stimulate discussion surrounding CDI at diffraction limited storage rings
  - Identifying areas where new science might be possible
  - Challenges



Y. Dabin *et al.*, MEDSI2018

ESRF-EBS report



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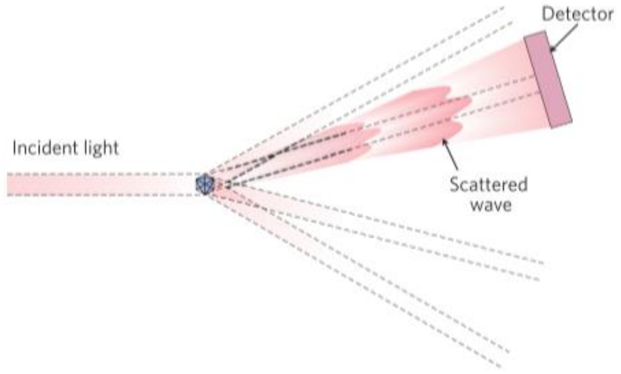
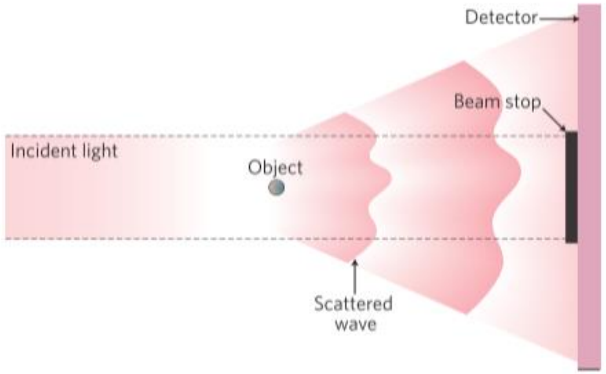
Overview

- Larger coherence volume/larger samples?
- Complications due to increased coherence
- Scanning modalities
    - Energy scanning, high energy CDI, event-mode data acquisition, e-beam scanning, multi-modal data
- CDI of the future for non-expert users
    - Data pipelines, diffraction patterns vs images, data volume and compression

Bragg CDI



Zeroth order/transmission CDI



Chapman & Nugent, Nat. Phot., 2010

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Larger coherence volume/larger samples?

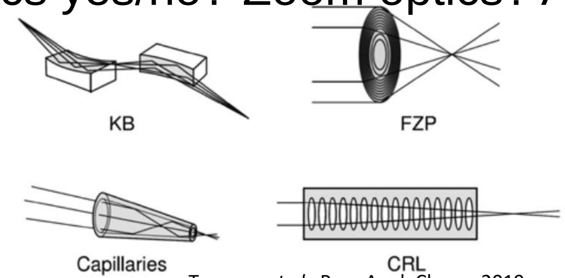
Zeroth order geometry:

- $\xi_T = \frac{\lambda z}{2\sigma}$ Smaller source/lower emittance results in an increase in coherence length (horizontal coherence approaching current coherence in the vertical direction)

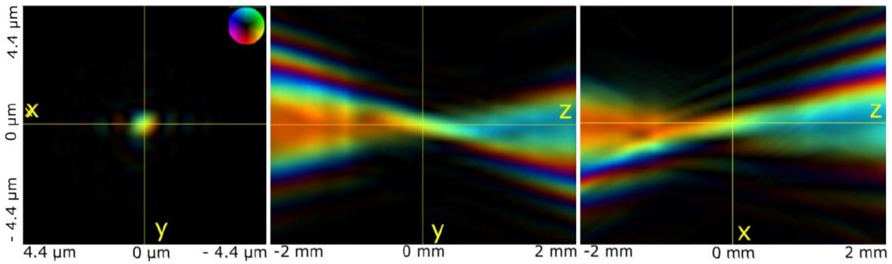
- Coherent flux increases by a factor of 10-100. - Already pushing the limits of current commercial detector technology (dynamic range, pixel size, deadtime, readout time)

  - Increased heat load to mono, optics, sample: Cooling vs stability, radiation damage

- Focusing optics yes/no? Zoom optics? Achromatic? Focused or defocused, Working distance…



Tenzano *et al.*, Pure Appl. Chem., 2019



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Larger coherence volume/larger samples?

Nanobeams:

- Easy to oversample
- Small probe beneficial for XFM etc.
- Beam divergence reduces need for high dynamic range detectors

- Fast motion, many frames for large areas
- Require careful environment control + closed loop positioning

Microbeams/unfocussed:

- Larger areas in fewer frames
- Slower radiation damage/better heat dissipation
- Optics are simpler and potentially more stable

- $z = \frac{2a\Delta x}{\lambda}$ Oversampling of the probe

Parameters	Distance
1 µm @ 9 keV, 55 µm pixel	0.8 m
10 µm @ 9 keV, 55 µm pixel	8 m
10 µm @ 30 keV, 55 µm pixel	26.6 m

Not possible at many existing beamlines

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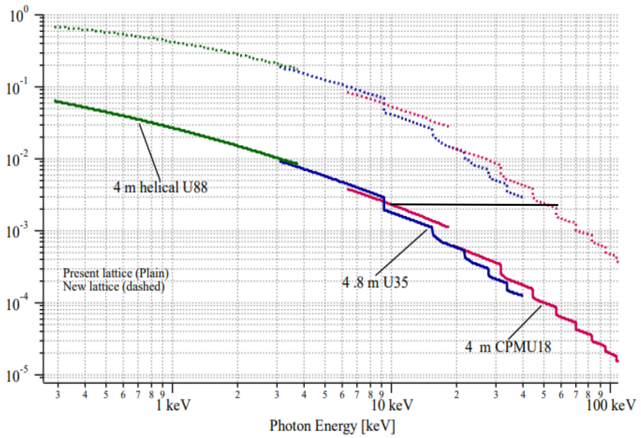
Larger coherence volume/larger samples?

Zeroth order geometry:

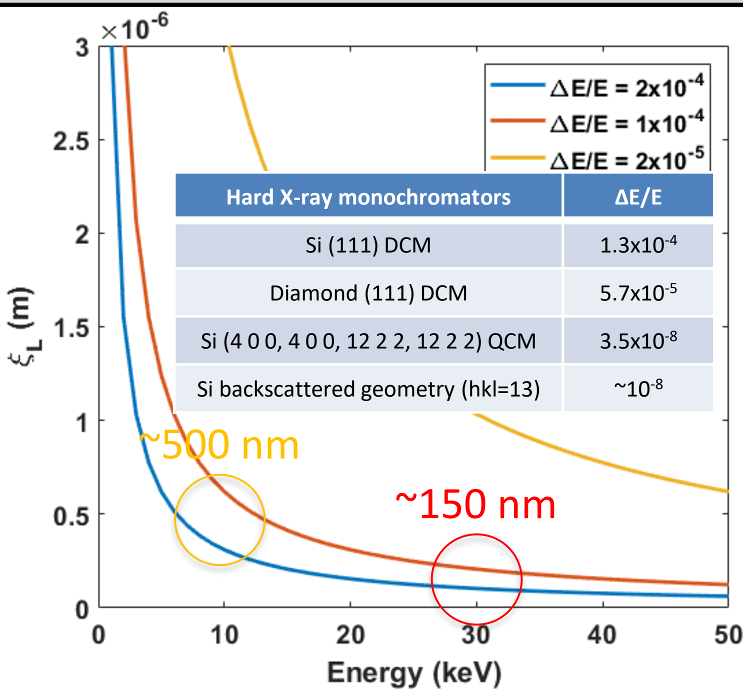
- $\xi_L = \frac{\lambda^2}{2\Delta\lambda}$ (not dependent on source size)
- Probed region of the sample needs to:

  ~ satisfying single scattering approximation

  ~ fit inside the coherence volume (relaxed)



ESRF-EBS whitepaper



Hard X-ray monochromators	ΔE/E
Si (111) DCM	1.3×10^{-4}
Diamond (111) DCM	5.7×10^{-5}
Si (4 0 0, 4 0 0, 12 2 2, 12 2 2) QCM	3.5×10^{-8}
Si backscattered geometry (hkl=13)	~10^{-8}

- EBS = larger ξ_T at high energy (difficult to achieve large ξ_L at high energies)
  - Continue to develop mathematic approaches to partial coherence/work on multi-slice/include dynamical diffraction
  - Design higher energy resolution monochromators (higher order reflections, multi-bounce, backscattered geometry, Si>Ge>Diamond etc…)

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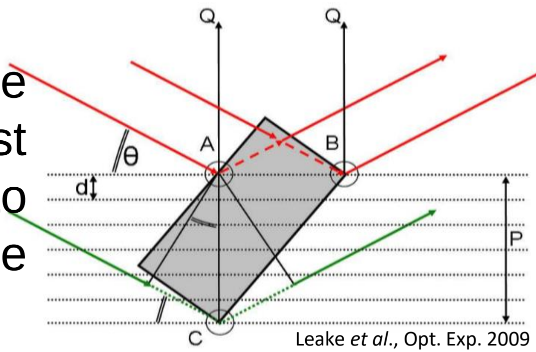
Larger coherence volume/larger samples?

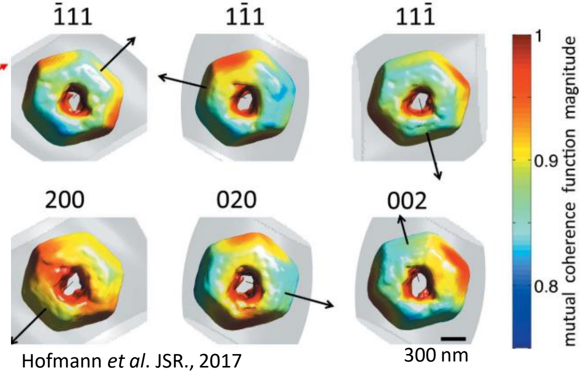
Bragg geometry:

Parameters	Distance
1 µm @ 9 keV, 55 µm pixel	0.8 m
10 µm @ 9 keV, 55 µm pixel	8 m
10 µm @ 30 keV, 55 µm pixel	26.6 m

- Isolated crystals: oversampling for large probes is hard, achieving this off the optical axis for high angles is even harder! Sample fixation is also going to be hard
- 10-100 times faster data collection will be amazing for Bragg ptychography (assuming that these photons can be measured). Bragg ptycho reconstruction quality should surpass isolated crystal reconstructions
- BCDI still limited by the OPLD exceeding the coherence volume i.e. the projection of the longest axis of the sample onto Q (AC). Currently limited to hundreds of nm thick samples at high incidence angle.



Leake *et al.*, Opt. Exp. 2009



Hofmann *et al.* JSR., 2017

Making faster measurements is a great enabler for CDI. There are even more exciting possibilities at diffraction limited storage rings in terms of newly accessible scanning modalities. There will be some challenges though…

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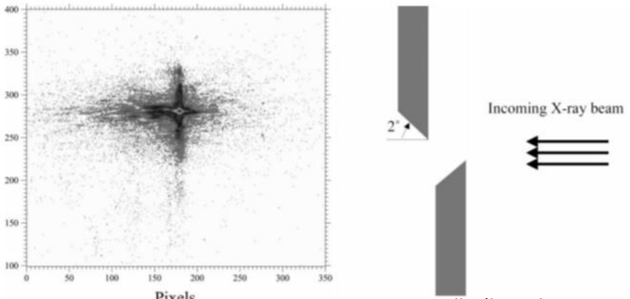
Complications due to increased coherence

- Beamline components in the beam path will diffract more and impart structure/spurious scatter: Annoying for CDI beamlines and quite challenging for beamlines possessing many optical components
    - e.g. Structure from slits or contamination on windows/mirrors/lenses



Le Bolloc'h et al., JSR, 2002

- Traditionally non-coherence beamlines (2/3rds of beamlines) may need assistance in resuming regular operation and with combining their standard technique with CDI for multimodal measurements
    - Spinning diffuser (destroy the coherence, what a shame!)
    - Minimize the components in the beam e.g. optics, windows, slits (soft edges/clean up slits)

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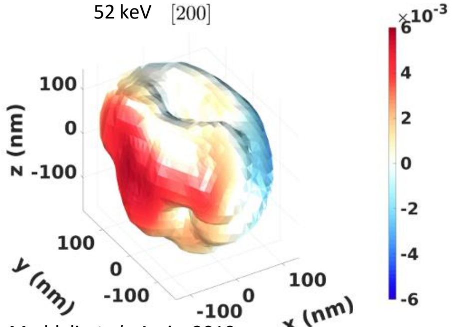
Scanning modalities

Higher dimensional scans

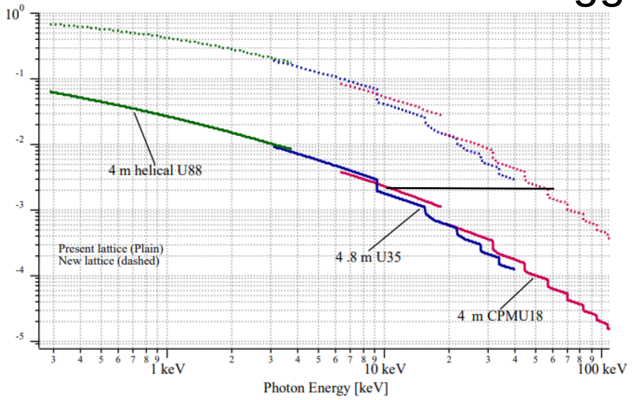
- Increased coherent flux means that scans can be faster and more samples could be measured
- Time resolved e.g. pump-probe, slowly evolving states (hours-µs)
- Tomography: As spatial resolution improves, projections through complex objects becomes insufficient

High energy CDI

- Upgrade brings a dramatic increase in coherence at higher energies. No longer limited to performing typical experiments below 15 keV
- Buried structures, more adsorption edges
- More accessible reflections in the Bragg geometry



Maddali *et al.*, Arxiv, 2019



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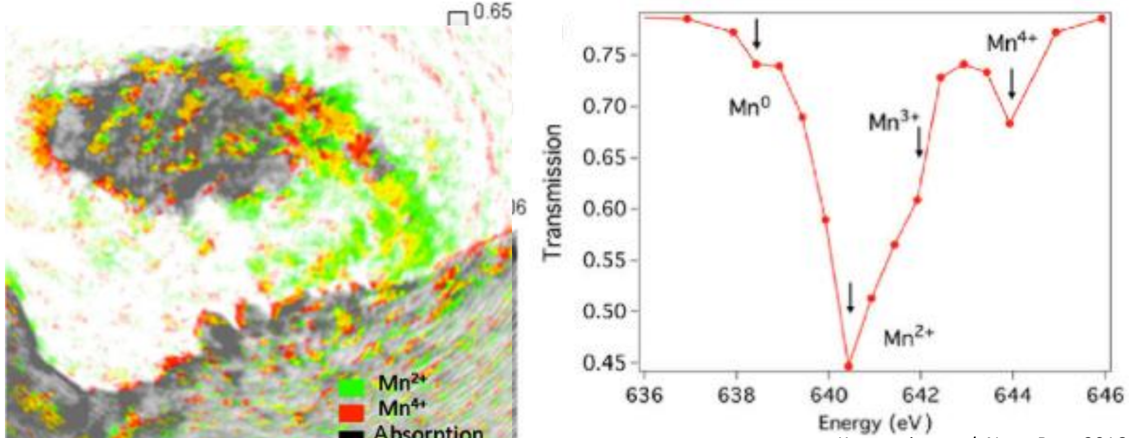
Scanning modalities

Energy scanning in transmission geometry

- XAS/XANES spectrum for every pixel in ptychographic scan (near neighbour distance, coordination, electronic state and chemistry)
- What about the phase information?

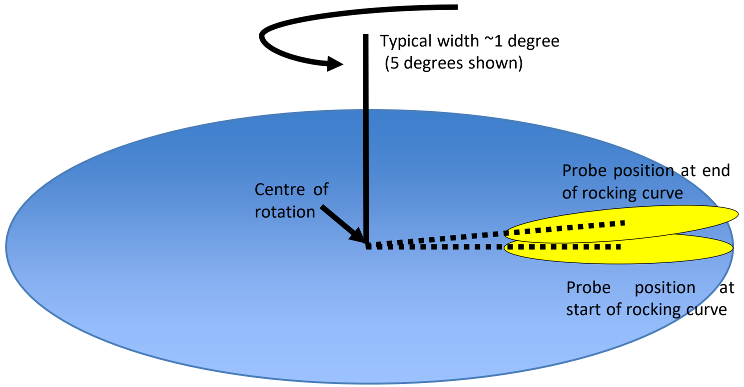
In situ growth of Mn-Co/polyprrol, a Pt free O_2-reduction catalyst



Kourousias *et al.* Nano Res., 2016

Energy scanning in Bragg geometry

- Replace the need to rock the sample: eliminates issues with changes in the projected sample volume
- Improves sample stage stability



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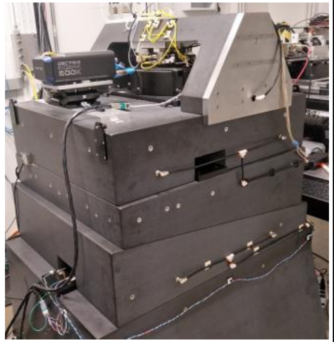
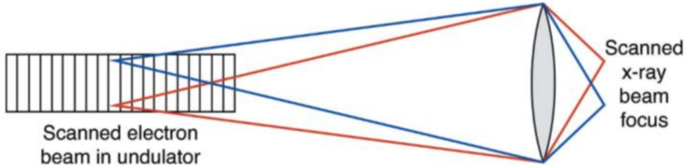
Scanning modalities

Optics scanning and e-beam scanning

- Sample stages are heavy/hard to move rapidly and accurately. Velociprobe scans the optics



- The stored beam could be scanned resulting in a movement of the source position. Due to demagnification would need to move the e-beam by a few mm to scan μm’s



de Jonge *et al.*, JSR., 2014

Event-mode data acquisition

125 years

Now
Event-mode?

- Missed detector triggers are problematic for large datasets at high frame rates

- Incorporated CDI acquisition in progress signal into data stream (XFM beamline of the AS)

- A step further? Adopt particle physics approaches to data streams where a photon event is recorded rather than ‘taking a picture’ at each sample position

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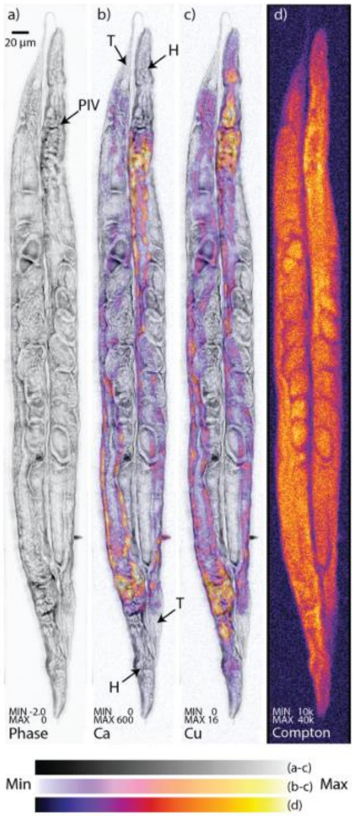
Scanning modalities (last 2)

Multi-modal data

- Microscopy users should want complementary CDI with their measurements
- What do we do with TB/hour: Analyse on-the-fly

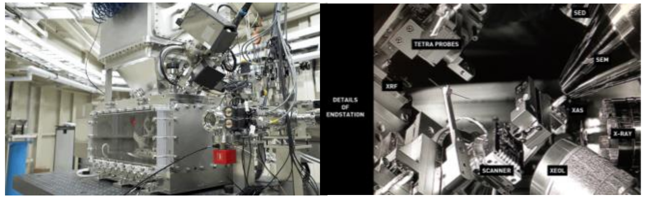
½ million diffraction patterns in a single 0.1mm^2 scan with 50 nm pixels (3 hour scan, 22 minutes to phase) 2x10^3 Gy



Jones & Phillips *et al.*, Chem. Com., 2018

Why stop at X-rays?

- X-ray + Electron microscope
- Reduction in scan time reduces long term stability requirements. Can an X-ray beam be delivered to a FIB-SEM or similar? TPS already has one!



21A XND @ Taiwan Photon Source http://tpsbl.nsrrc.org.tw

- What other instruments can simultaneously provide complementary information?

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CDI for non-expert users

- CDI is already becoming more of imaging workhorse than ‘art’. This general trend will continue.
- How can we make sure that non-experts:
  - Understand the information that they receive
  - Leave with their answer not the raw data
- Data pipelines
- Diffraction patterns vs microscopy images
  - Also addresses impracticality of storing huge datasets for long periods of time

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Summary

- Lots of exciting new possibilities
  - Multi-modal data acquisition, high coherence at many beamlines
  - Shorter scan times allows higher dimensional datasets e.g. *in operando*, over adsorption edges, as a function of pump delay time etc.
- Reduce overheads (not just acquisition overheads)
- Improve phasing schemes
- Consideration of data density, format, analysis, interpretation, storage
- Continue to work on improving detectors (less deadtime, faster readout, higher dynamic range)

Engineering Science: Hofmann Group

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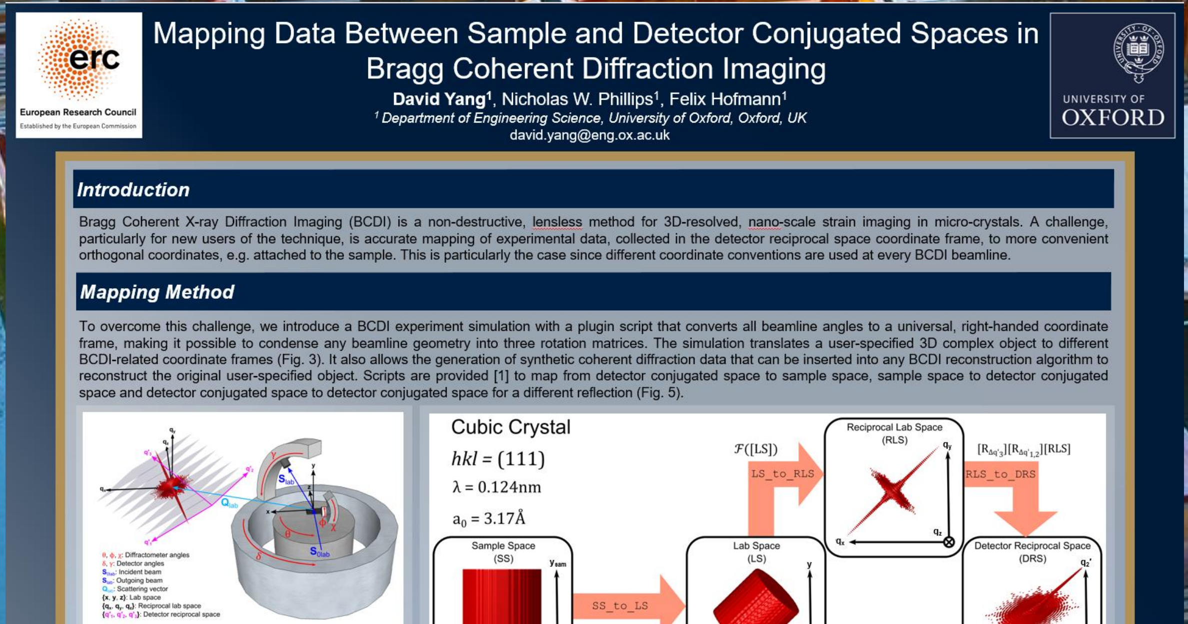
Felix Hofmann: First talk tomorrow

Bragg coherent diffractive imaging: an everyday microscopy tool?



Mapping Data Between Sample and Detector Conjugated Spaces in Bragg Coherent Diffraction Imaging

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Introduction

Bragg Coherent X-ray Diffraction Imaging (BCDI) is a non-destructive, lensless method for 3D-resolved, nano-scale strain imaging in micro-crystals. A challenge, particularly for new users of the technique, is accurate mapping of experimental data, collected in the detector reciprocal space coordinate frame, to more convenient orthogonal coordinates, e.g. attached to the sample. This is particularly the case since different coordinate conventions are used at every BCDI beamline.

Mapping Method

To overcome this challenge, we introduce a BCDI experiment simulation with a plugin script that converts all beamline angles to a universal, right-handed coordinate frame, making it possible to condense any beamline geometry into three rotation matrices. The simulation translates a user-specified 3D complex object to different BCDI-related coordinate frames (Fig. 3). It also allows the generation of synthetic coherent diffraction data that can be inserted into any BCDI reconstruction algorithm to reconstruct the original user-specified object. Scripts are provided [1] to map from detector conjugated space to sample space, sample space to detector conjugated space and detector conjugated space to detector conjugated space for a different reflection (Fig. 5).

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