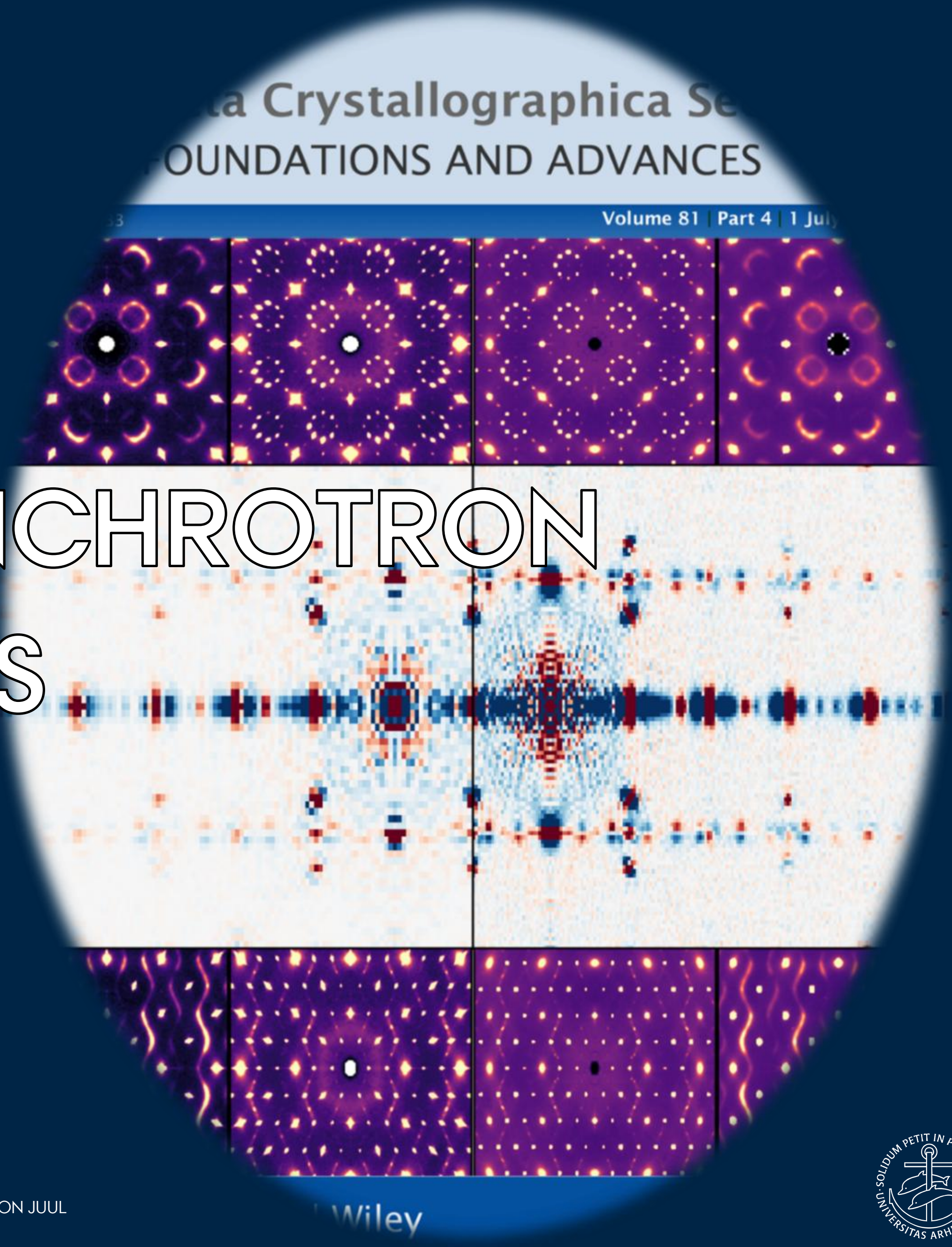
3D-ΔPDF: From Synchrotron to In-house Sources

Karl Juul
Department of Chemistry, Aarhus University



3D-ΔPDF

3-Dimensional
Single crystals

Difference
Between the total and average PDFs

Pair Distribution Function
How pairs of atoms are distributed (placed) in space

Map with distribution of atom-pairs that differ from the average structure

3D-ΔPDF

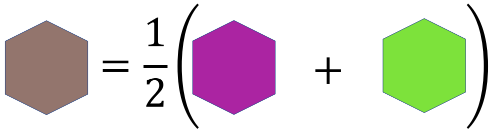
3-Dimensional
Single crystals

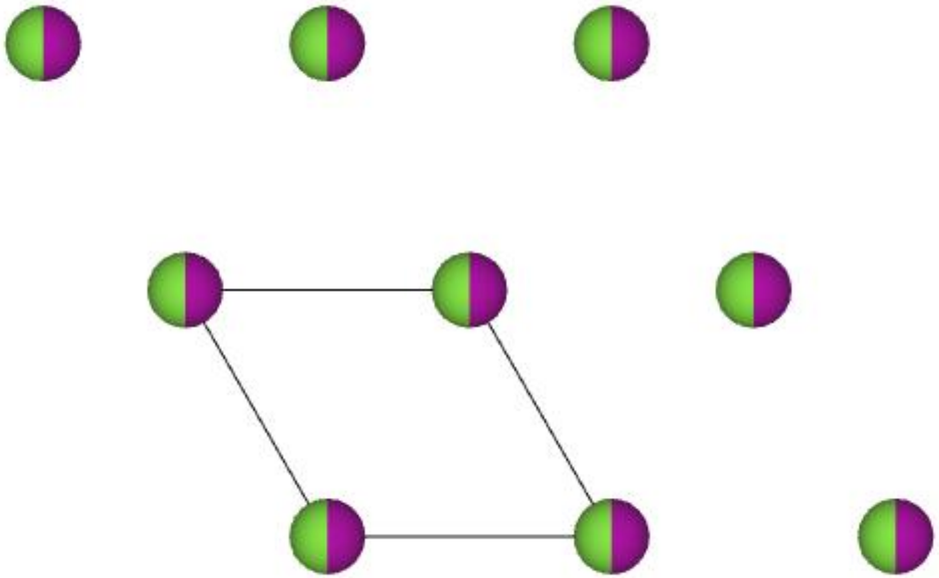
Difference
Between the total and average PDFs

Pair Distribution Function
How pairs of atoms are distributed (placed) in space

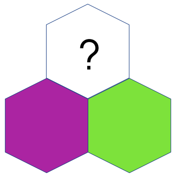
$$I_{\text{tot}}(\boldsymbol{q}) = I_{\text{Bragg}}(\boldsymbol{q}) + I_{\text{diffuse}}(\boldsymbol{q})$$
$$\text{3D}-\text{PDF}(\boldsymbol{r}) = P(\boldsymbol{r}) + \text{3D}-\Delta\text{PDF}(\boldsymbol{r})$$
$$\text{3D}-\Delta\text{PDF}(\boldsymbol{r}) = \mathcal{F}^{-1}[I_{\text{diffuse}}(\boldsymbol{q})]$$

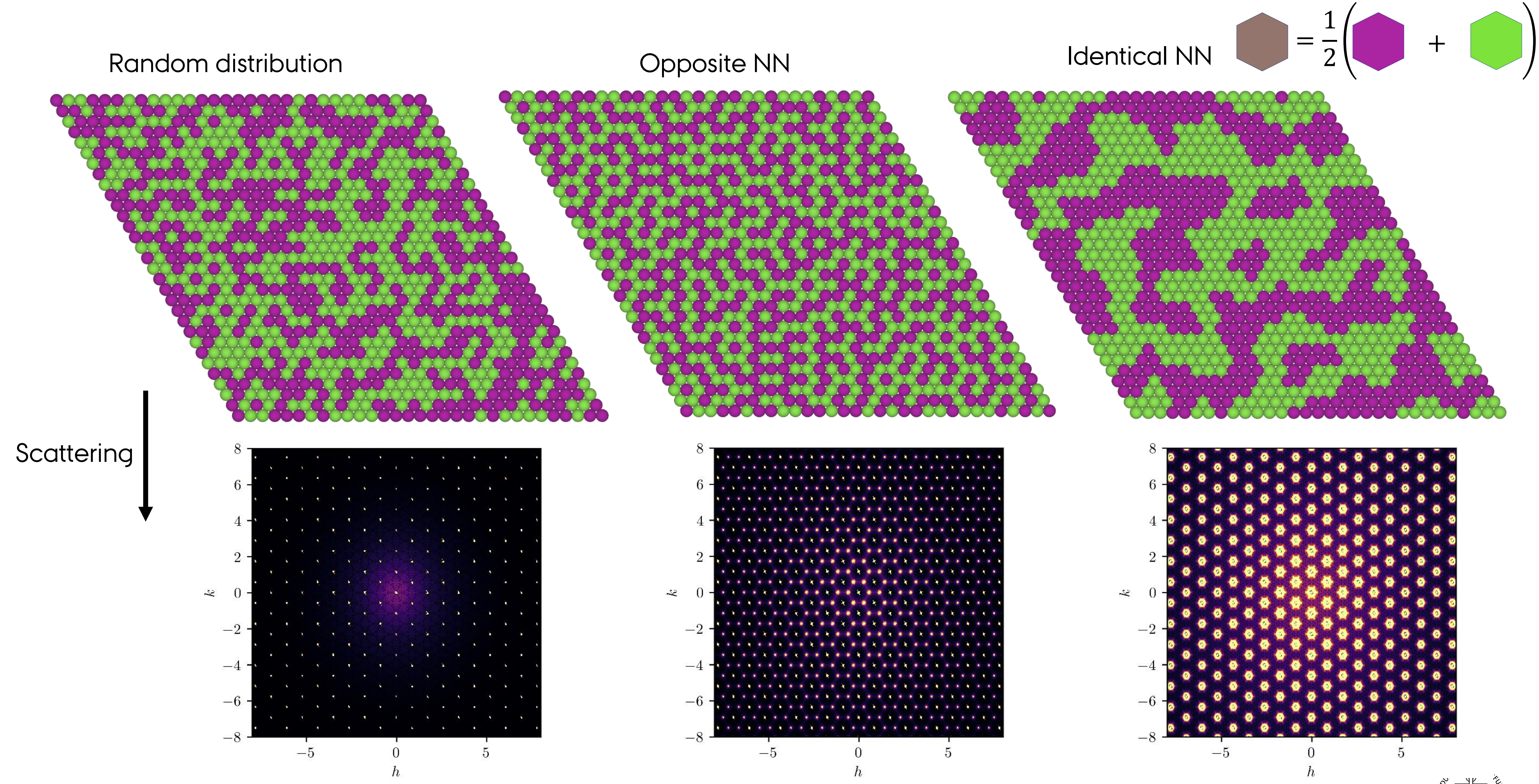
TRIANGULAR ISING (A)FM



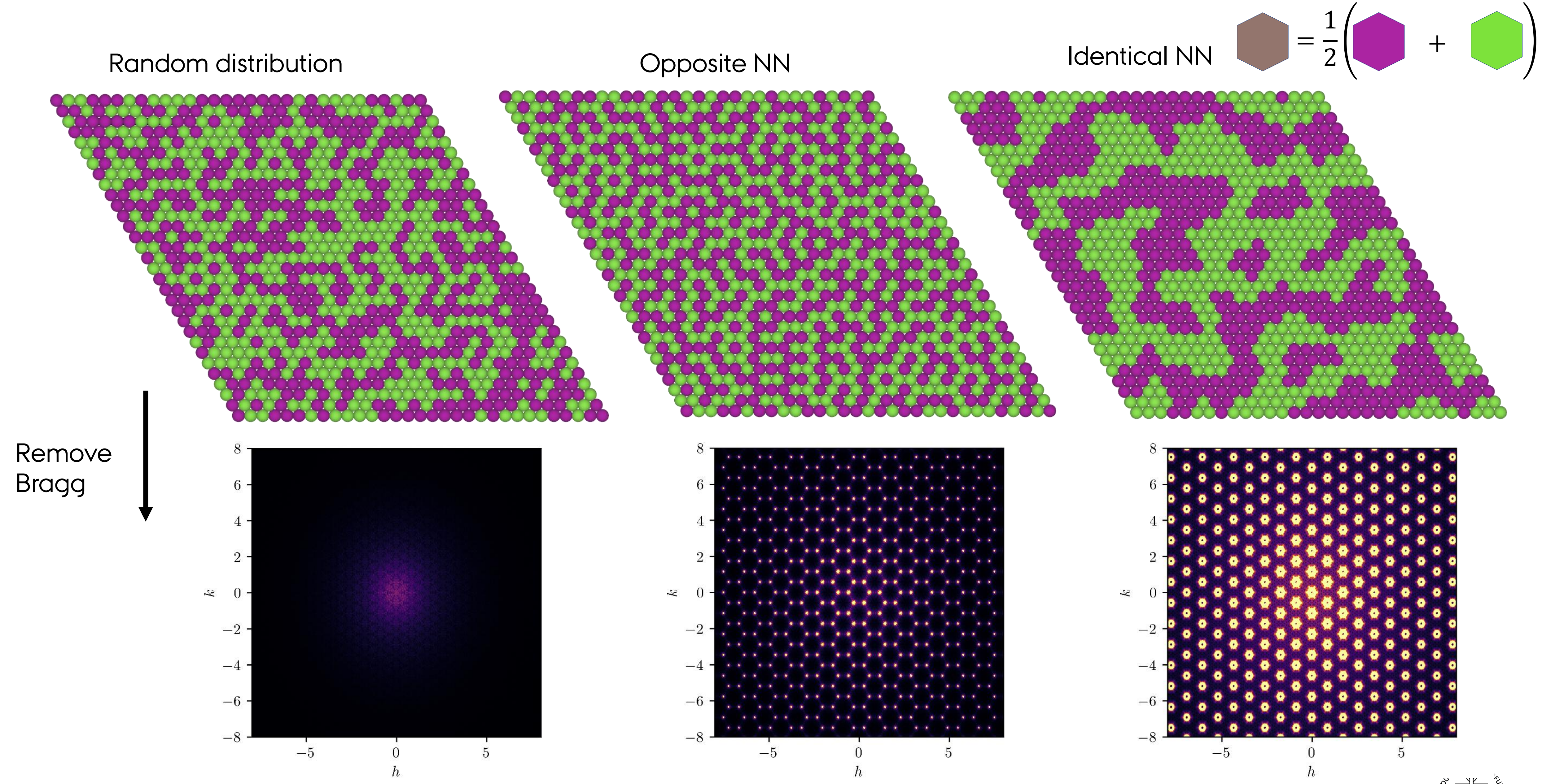


1. Random distribution
2. Identical nearest neighbours
3. Opposite nearest neighbours

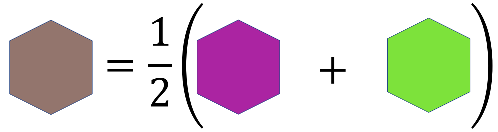




Identical NN
= 1/2 (+)
Random distribution
Opposite NN
Scattering
k
h
8
6
4
2
0
−2
−4
−6
−8
−5
0
5

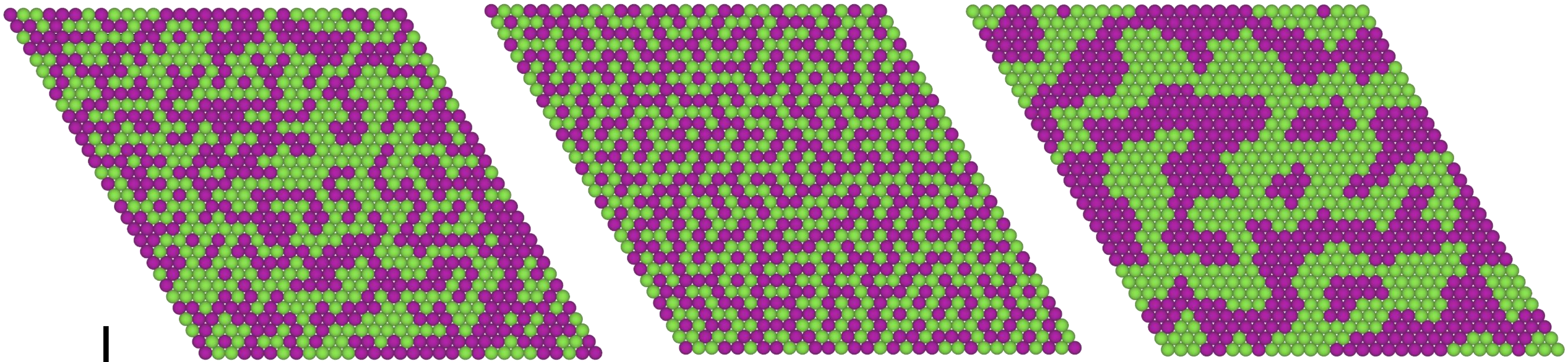

Random distribution
Opposite NN
Identical NN
= 1/2 (+)
Remove Bragg
8
6
4
2
0
−2
−4
−6
−8
k
−5
0
5
h

Identical NN

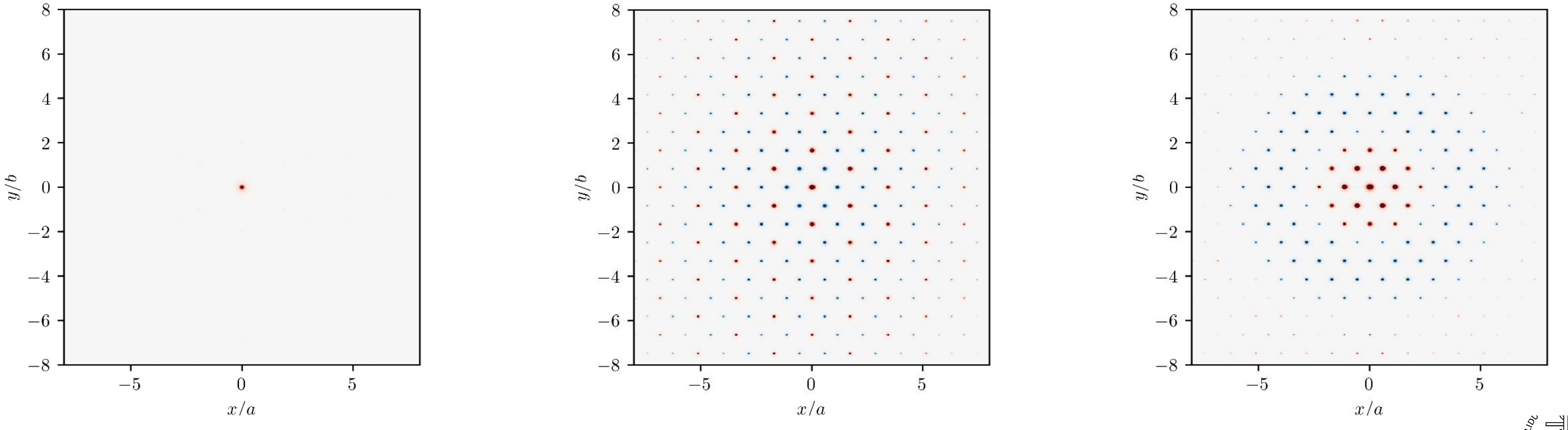


Random distribution

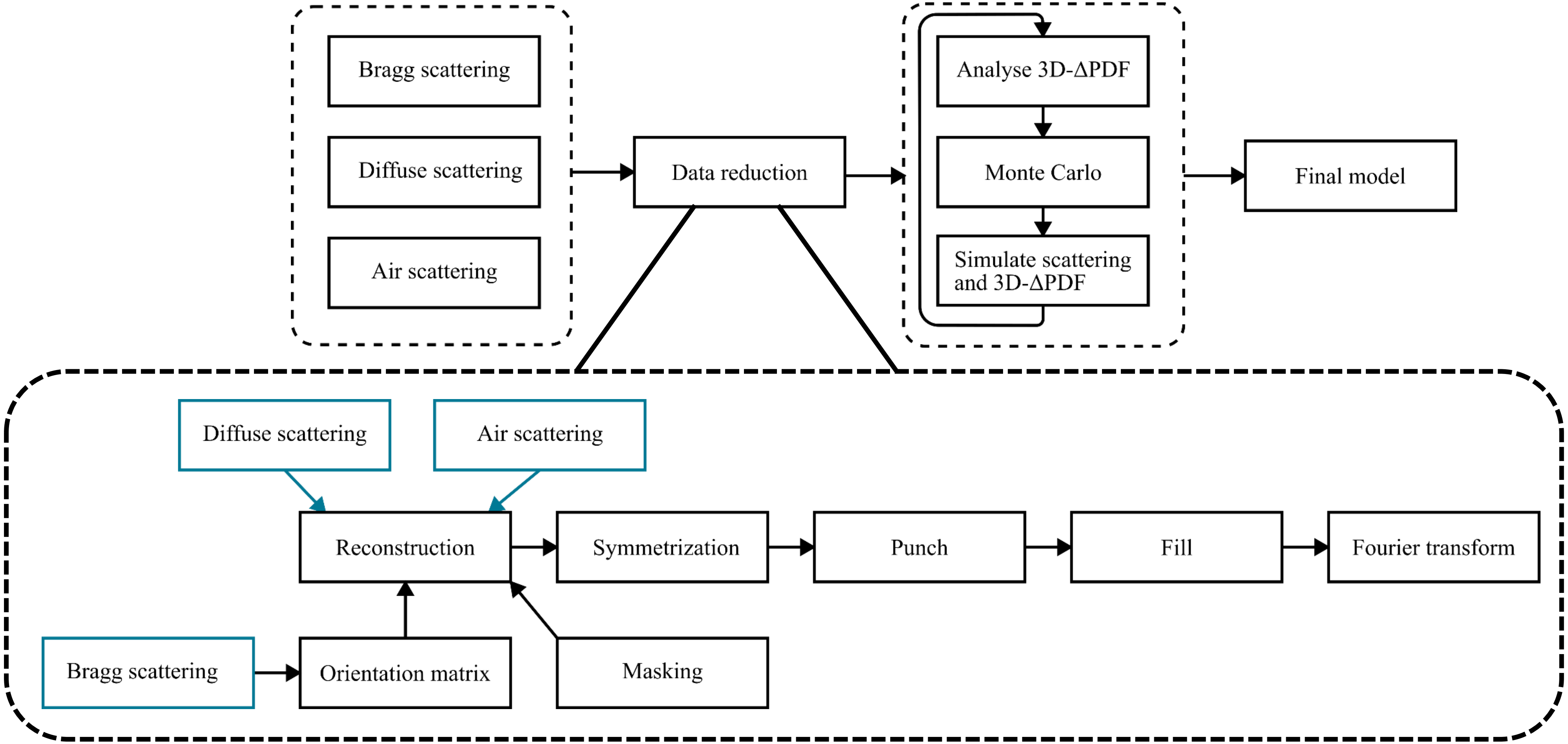
Opposite NN



Fourier Transform

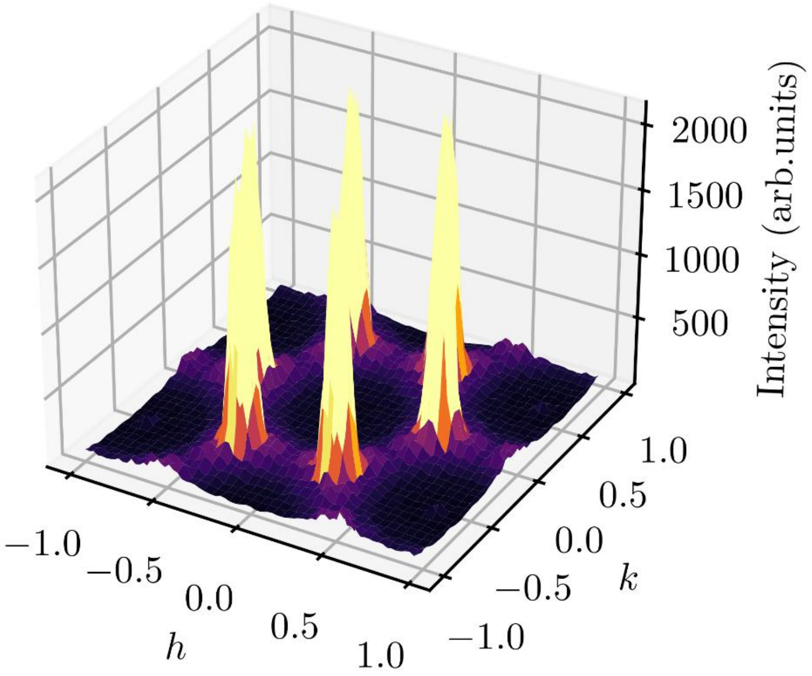


THE WORKFLOW

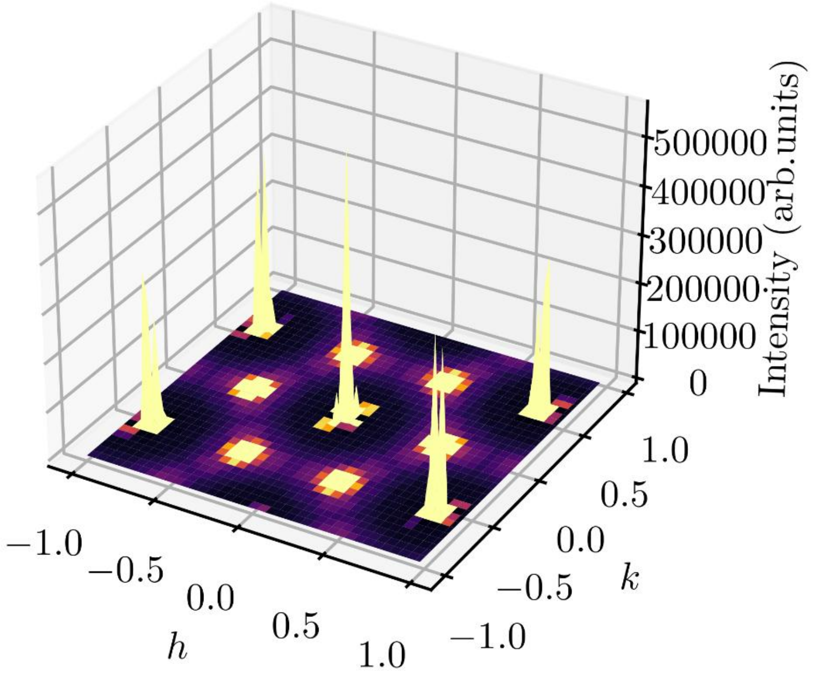


A CONSIDERATION OF SCALE

Diffuse scattering



Total scattering



Diffuse scattering is very weak

***Historically* Data is measured at synchrotron sources**

THE FIRST IN-HOUSE DIFFUSE SCATTERING

RESEARCH

CRYSTALLINE ORDERING

Truchet-tile structure of a topologically aperiodic metal–organic framework

Emily G. Meekel[1], Ella M. Schmidt[1,2], Lisa J. Cameron[3], A. David Dharma[3], Hunter J. Windsor[3], Samuel G. Duyker[3,4], Arianna Minelli[1], Tom Pope[5], Giovanni Orazio Lepore[6], Ben Slater[5], Cameron J. Kepert[3*], Andrew L. Goodwin[1*]

research papers

Acta Cryst B

STRUCTURAL SCIENCE
CRYSTAL ENGINEERING
MATERIALS

ISSN 2052-5206

Direct interpretation of the X-ray and neutron three-dimensional difference pair distribution functions (3D-ΔPDFs) of yttria-stabilized zirconia

Ella Mara Schmidt,[a,b*] Reinhard B. Neder,[c] James D. Martin,[d] Arianna Minelli,[b] Marie-Hélène Lemée[e] and Andrew L. Goodwin[b]

BENCHMARKING STUDY

research papers

FOUNDATIONS ADVANCES

ISSN 2053-2733

Benchmarking 3D-ΔPDF analysis using in-house X-ray sources

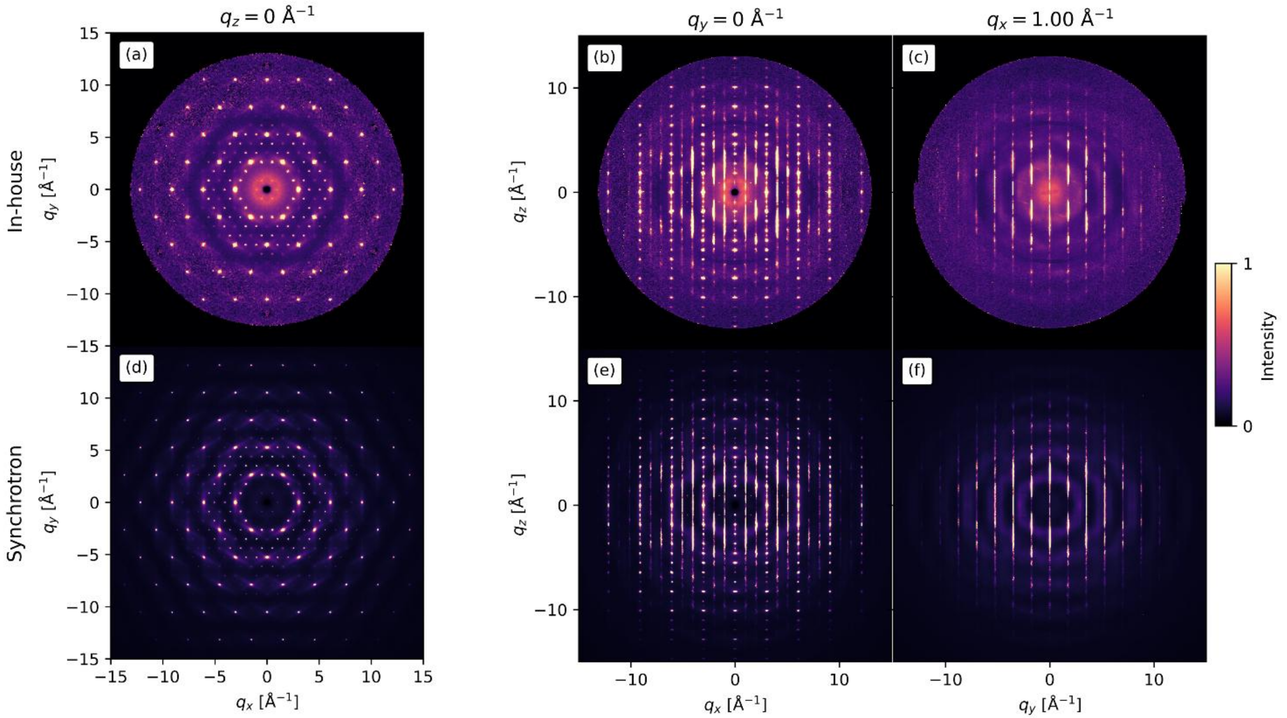
Karl O. R. Juul, Kristoffer A. H. Støckler and Bo B. Iversen*

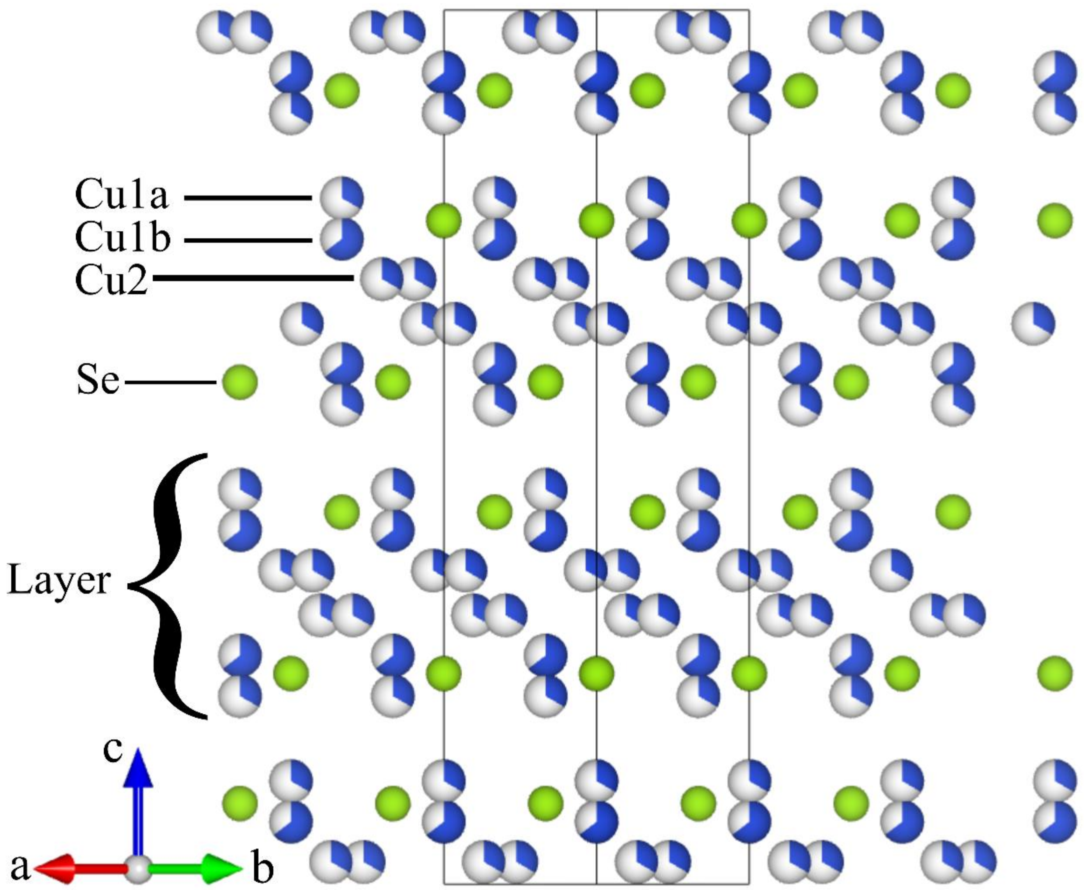
Center for Integrated Materials Research, Department of Chemistry and iNANO, Aarhus University, Aarhus, DK-8000, Denmark. *Correspondence e-mail: bo@chem.au.dk

Compound	Diffuse intensity	Diffuse location	Synchrotron reference
Cu_2Se	Strong	Away from Bragg	Roth & Iversen, 2019
$Nb_{1-x}CoSb$	Weak	Away from Bragg	Roth *et al.*, 2021
InTe	Weak	On Bragg	Støckler *et al.*, 2024

Acta Cryst. (2025). A**81**, 254–268

CASE STUDY: Cu_2Se





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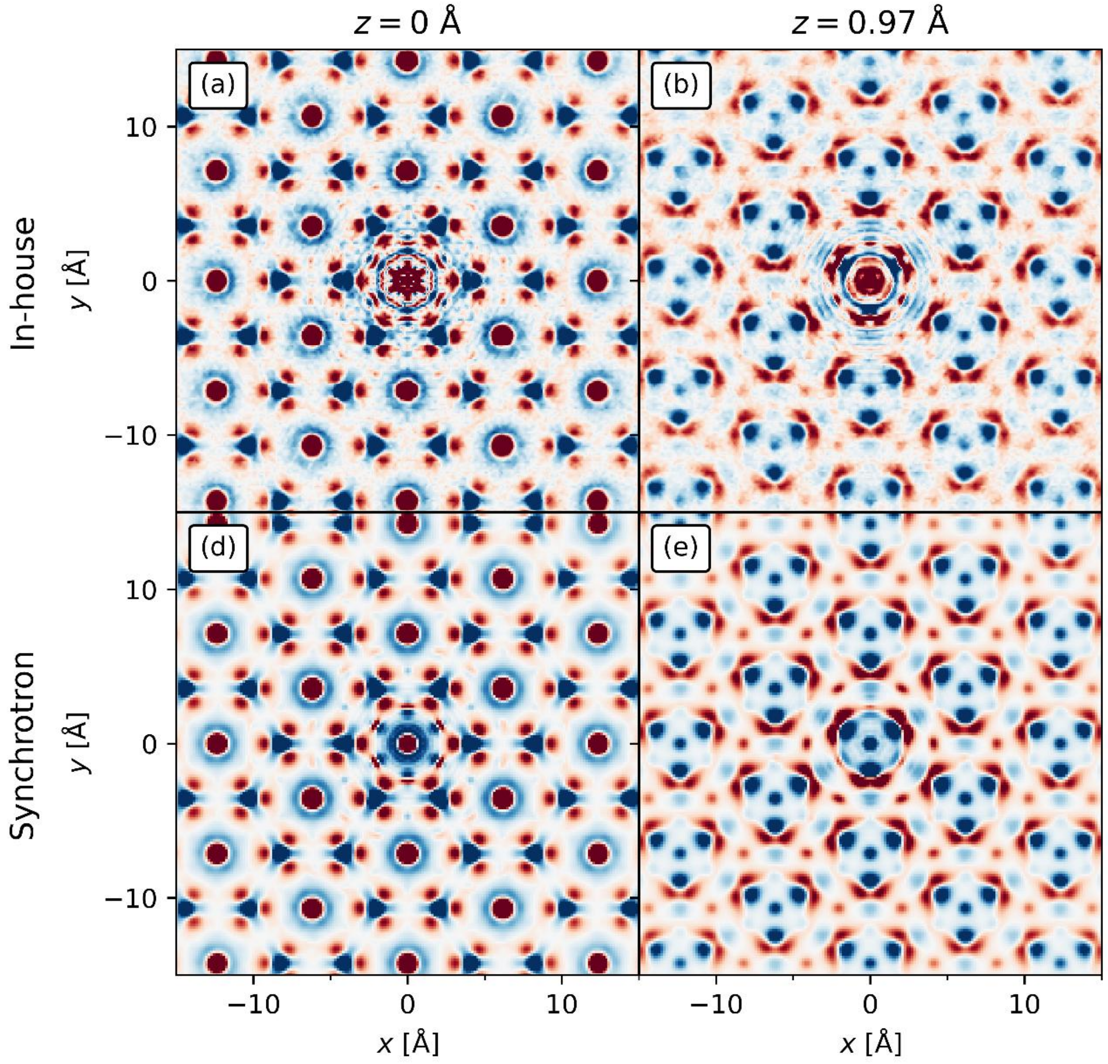

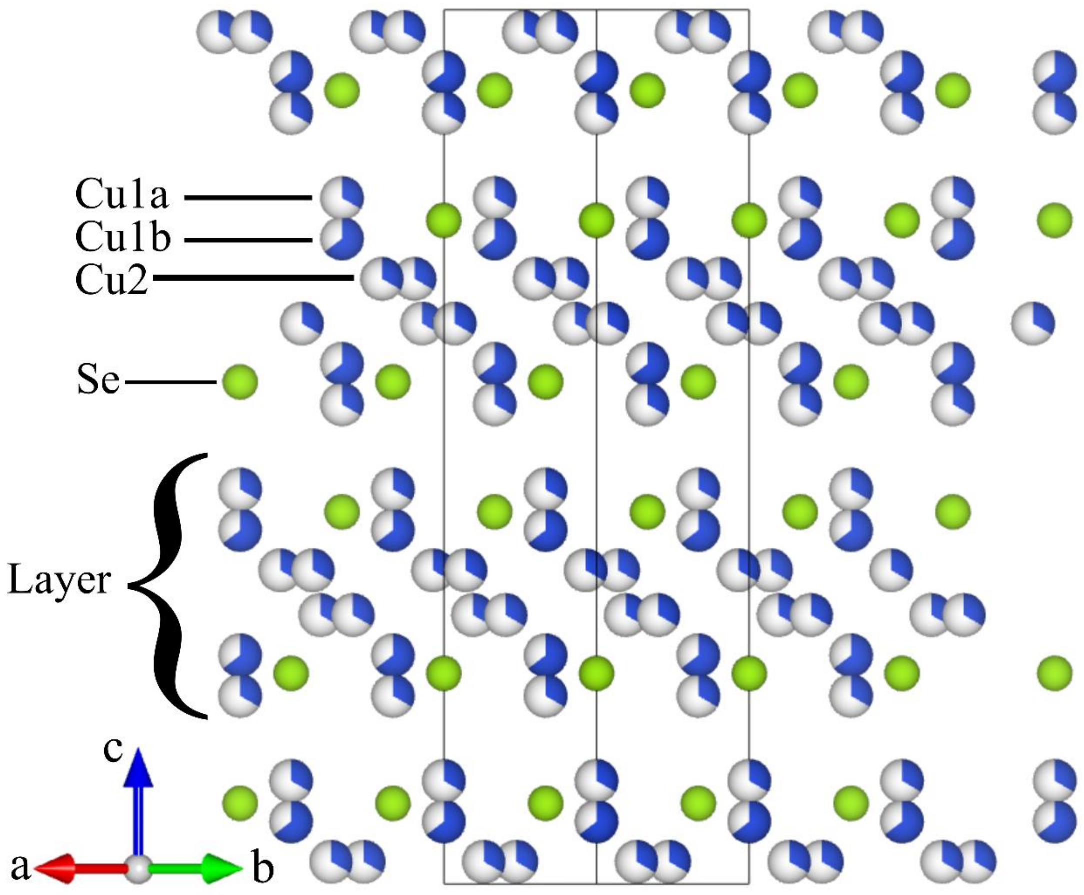

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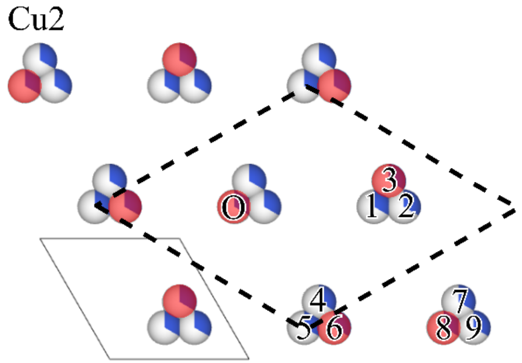
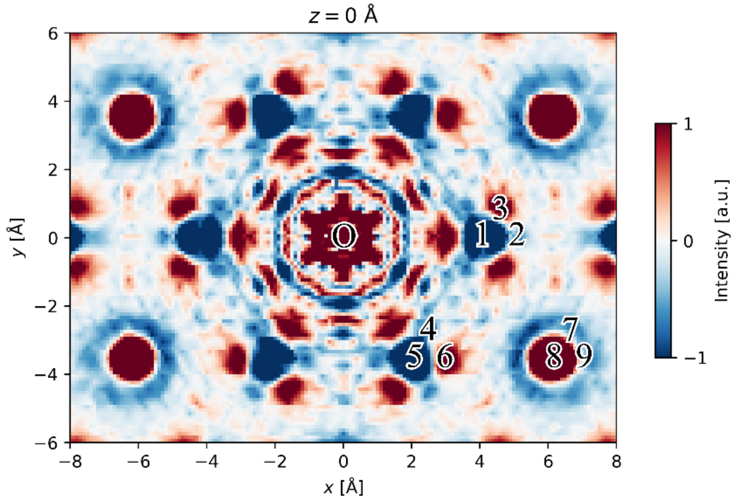
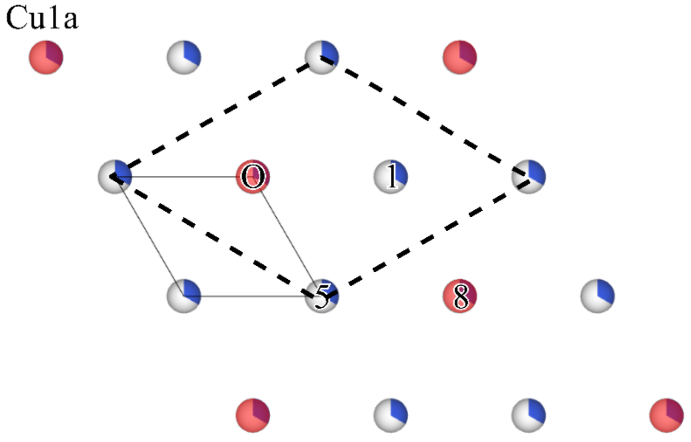
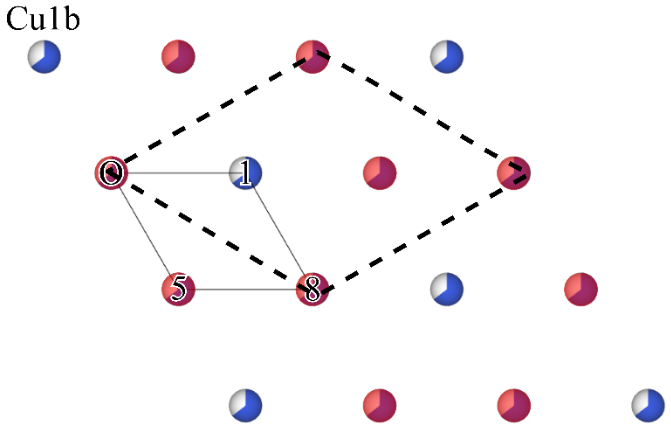




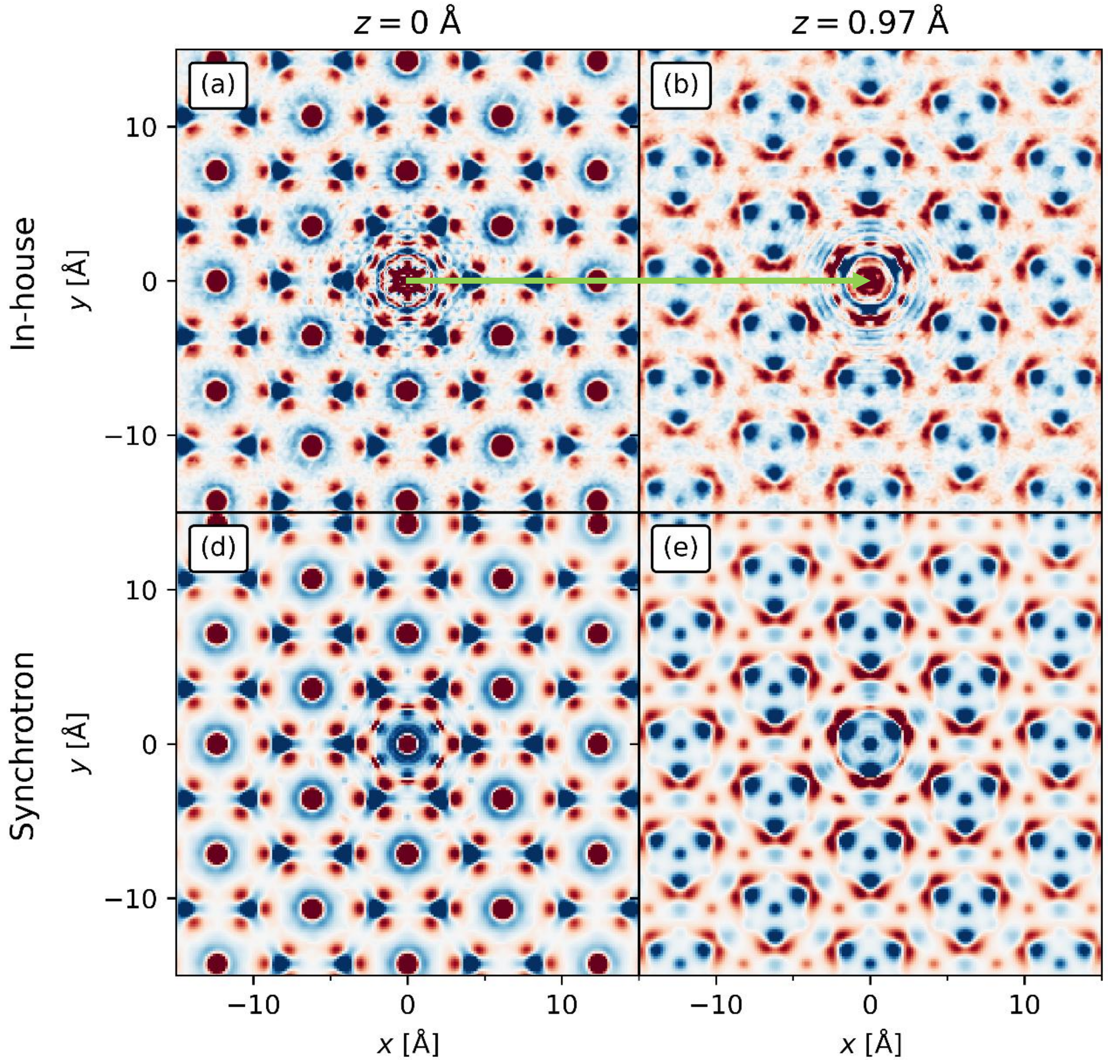


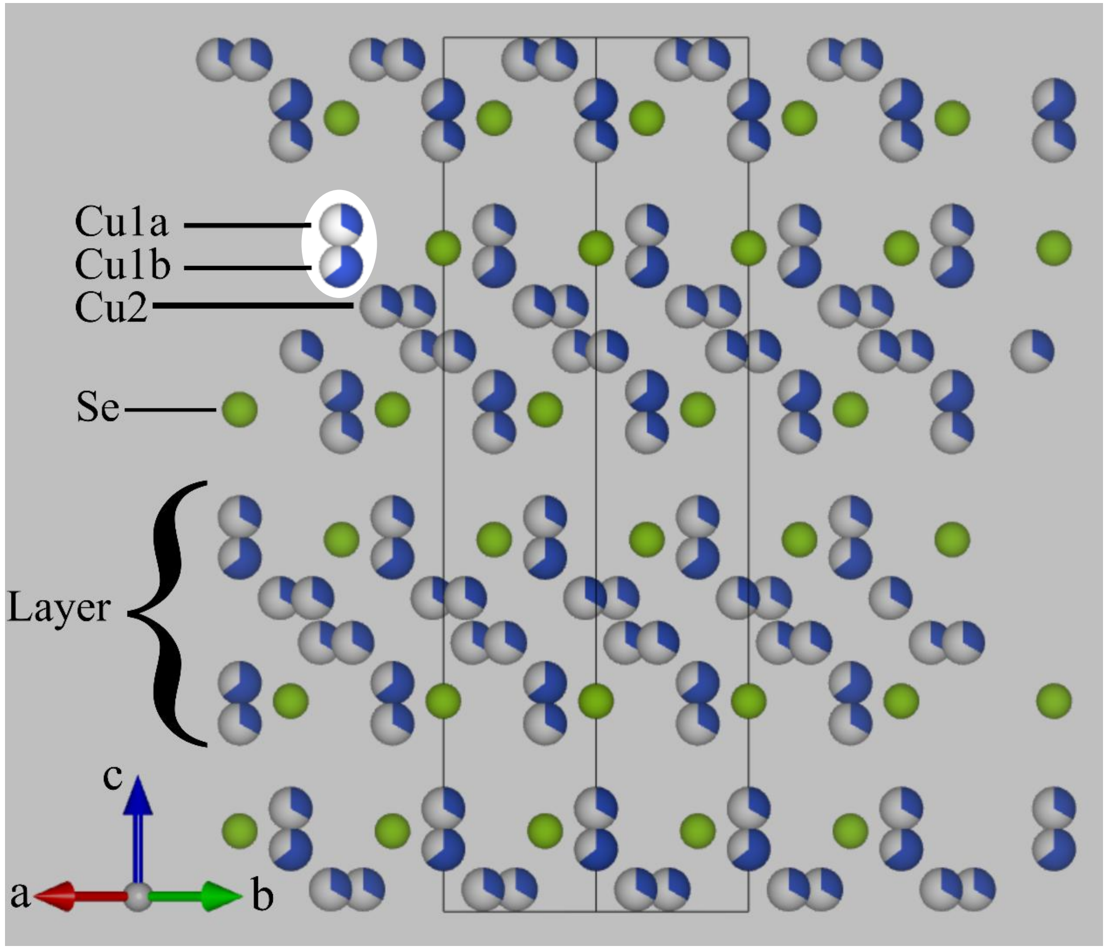


$$\text{peak integral} = \sum_{i,j|\boldsymbol{r}_{ij}=\boldsymbol{r}} \delta Z_i \delta Z_j$$

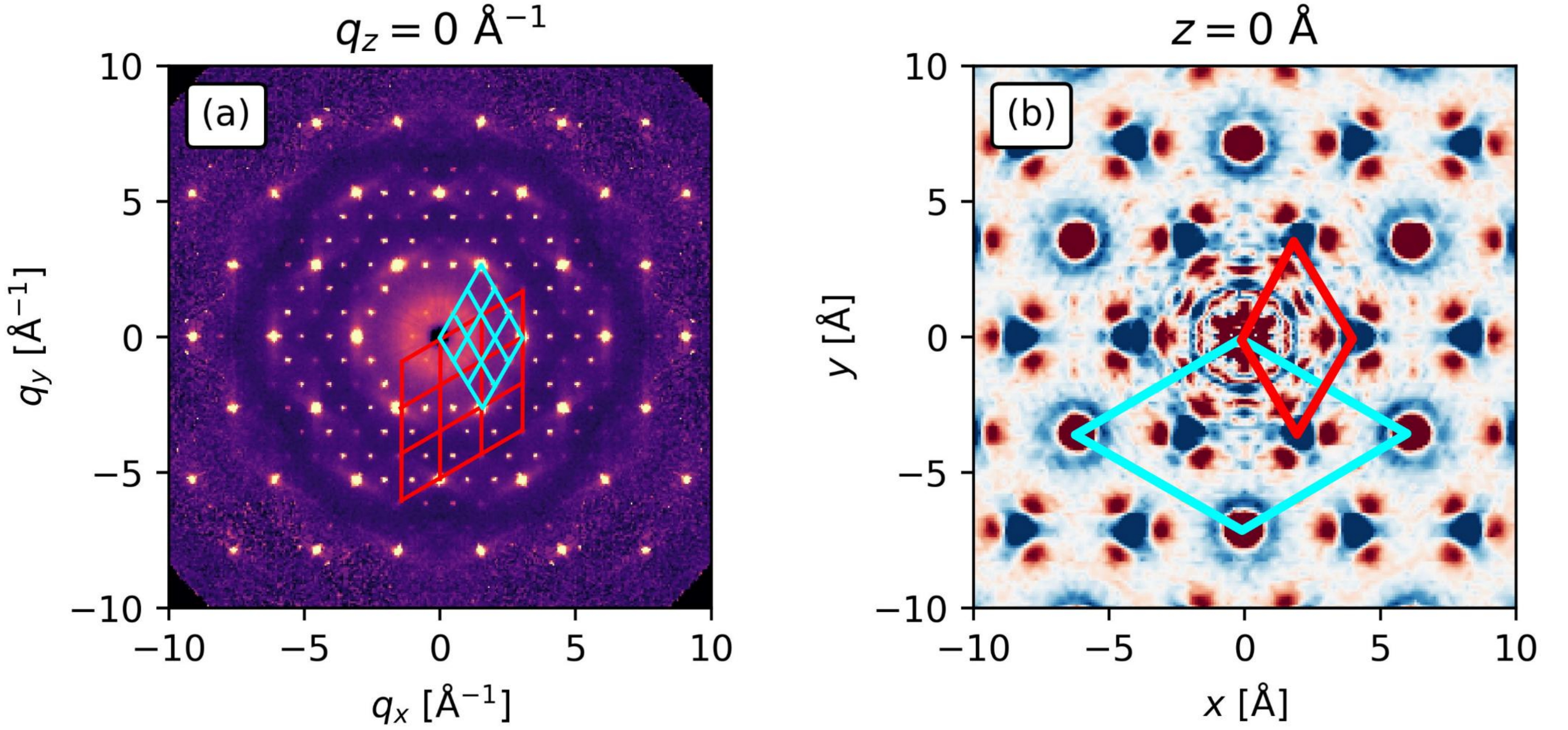
$$\begin{aligned} P(5) &= \delta Z_{\text{occ}}^2 + 2\delta Z_{\text{occ}}\delta Z_{\text{vac}} \\ &= \left(\left(\frac{1}{3}\right)^2 + 2\frac{1}{3}\cdot\frac{-2}{3}\right) Z_{\text{Cu}}^2 \\ &= -\frac{1}{3} Z_{\text{Cu}}^2 \end{aligned}$$

CASE STUDY: Cu_2Se

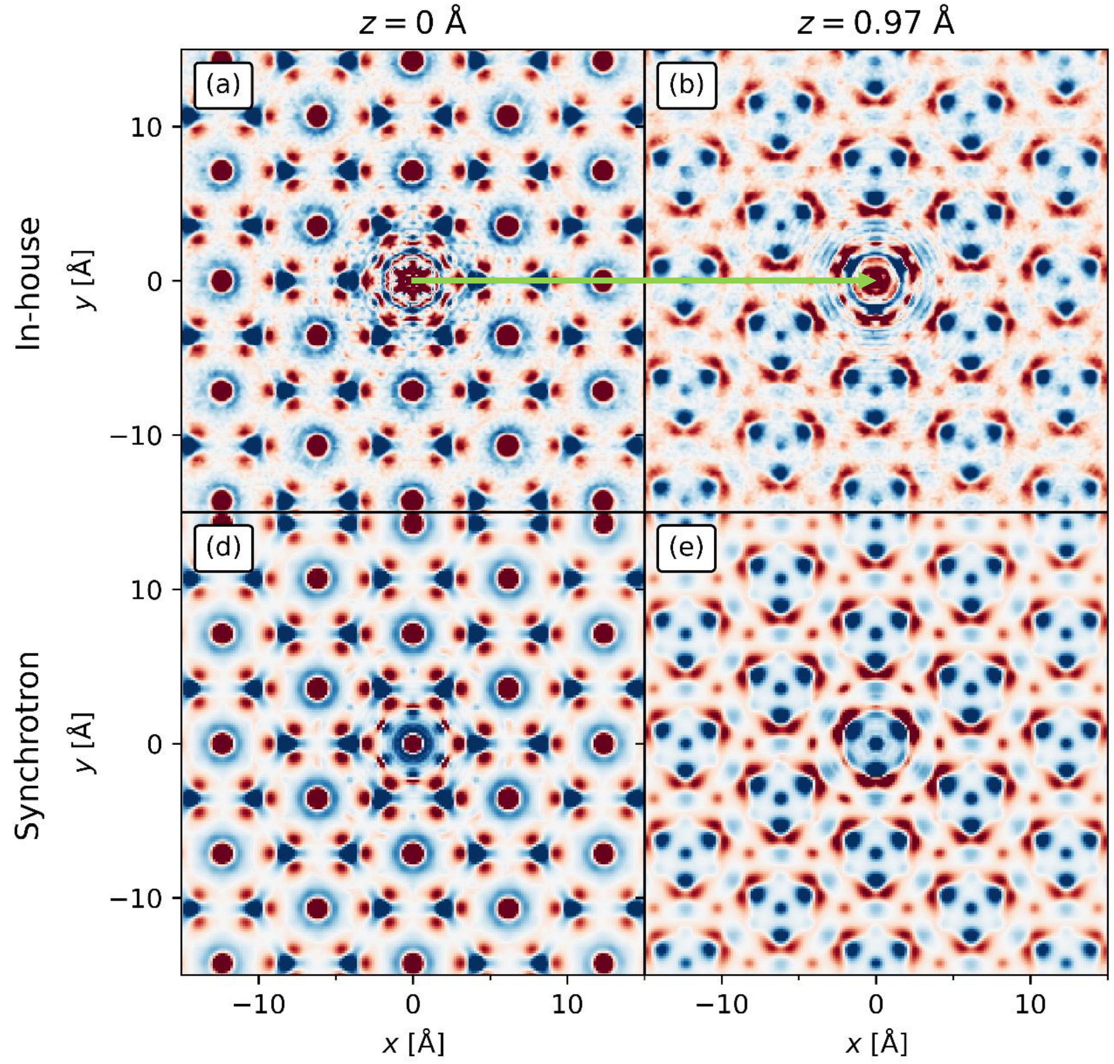


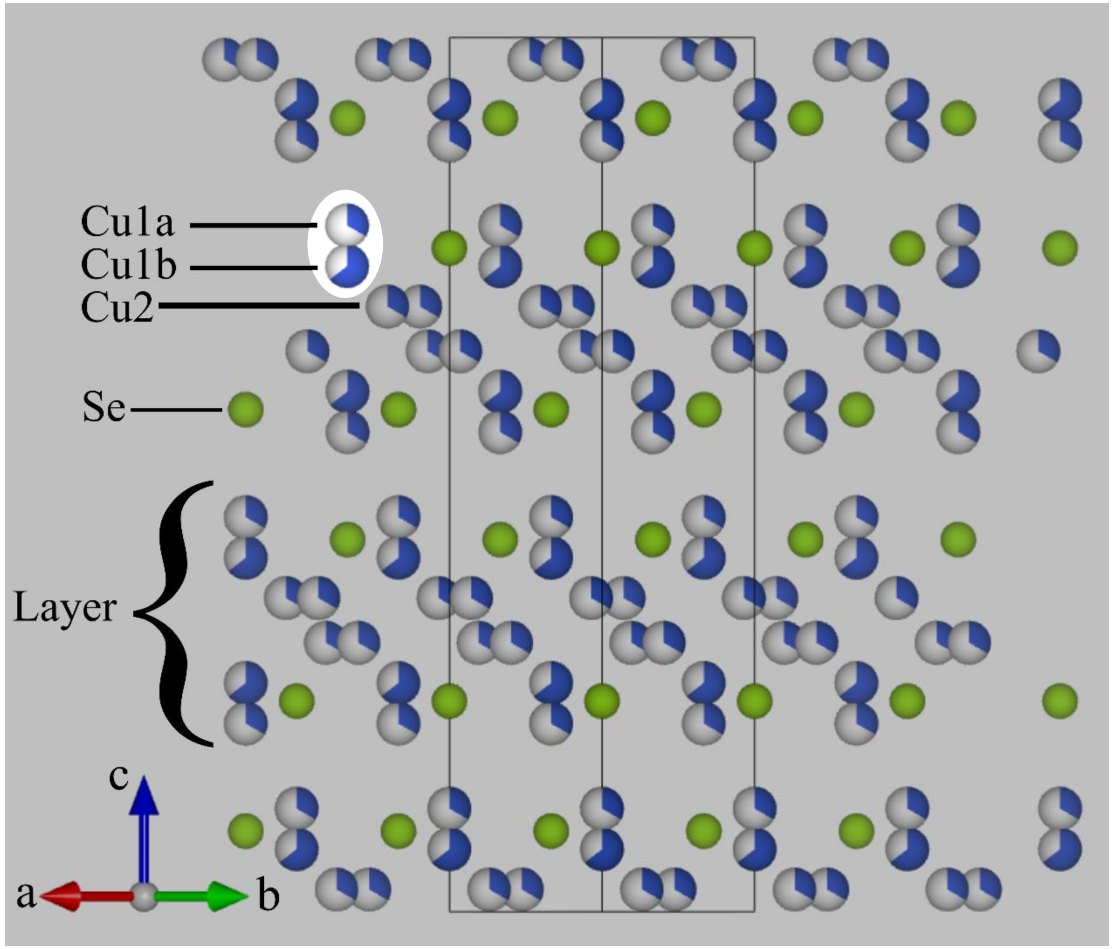


CASE STUDY: Cu_2Se

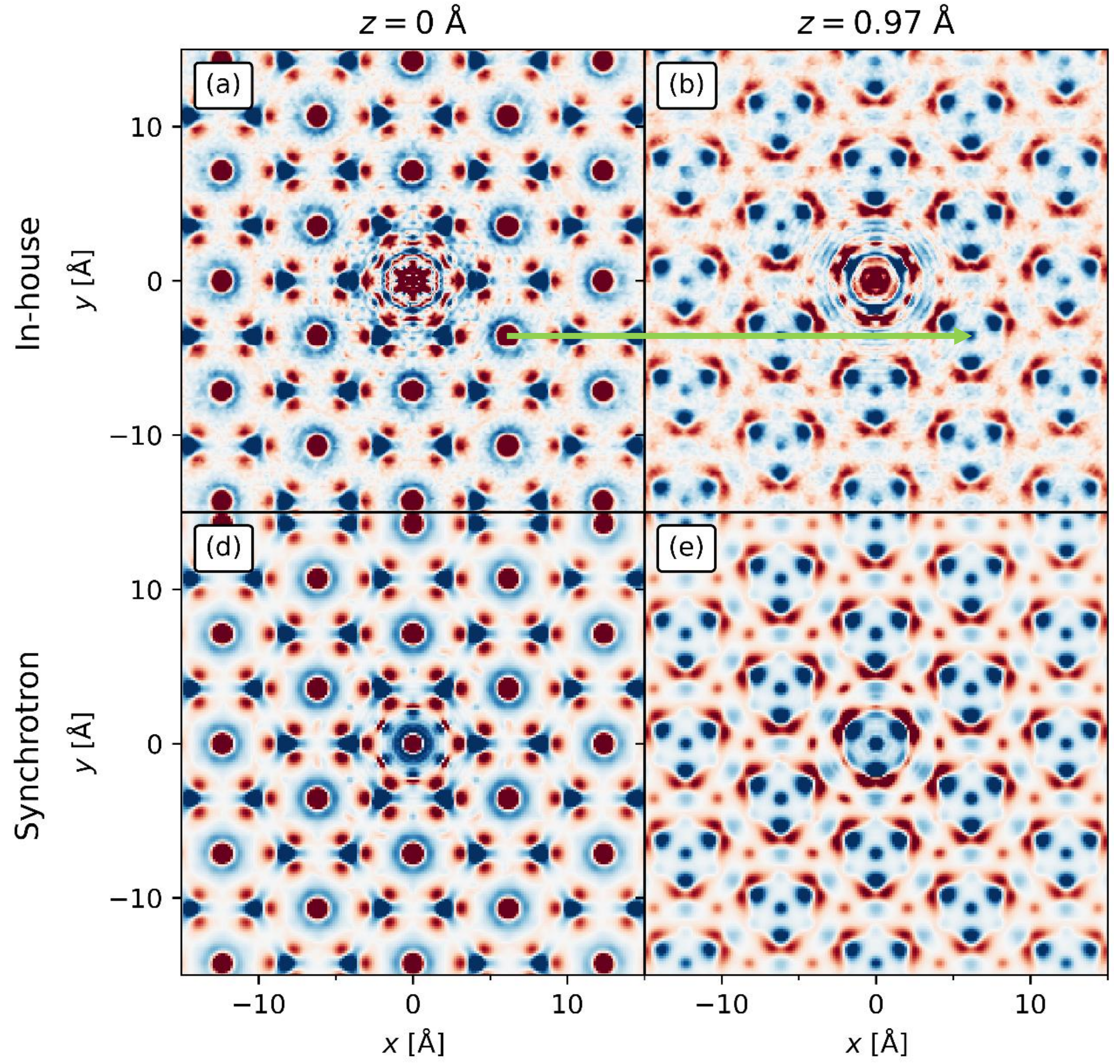


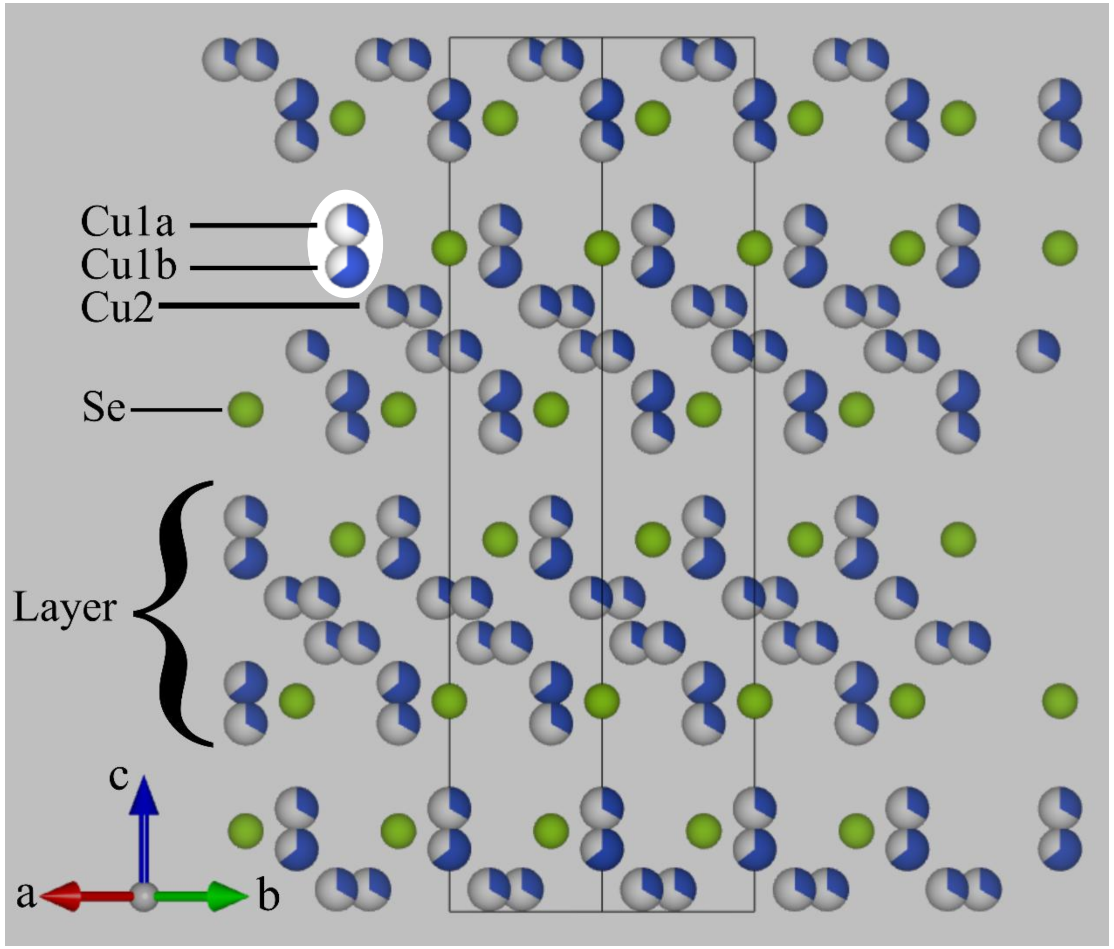
CASE STUDY: Cu_2Se



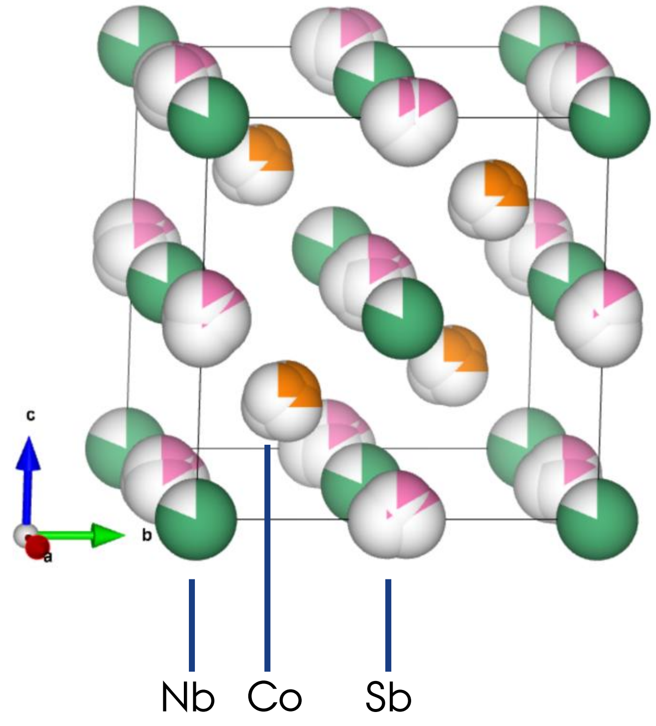


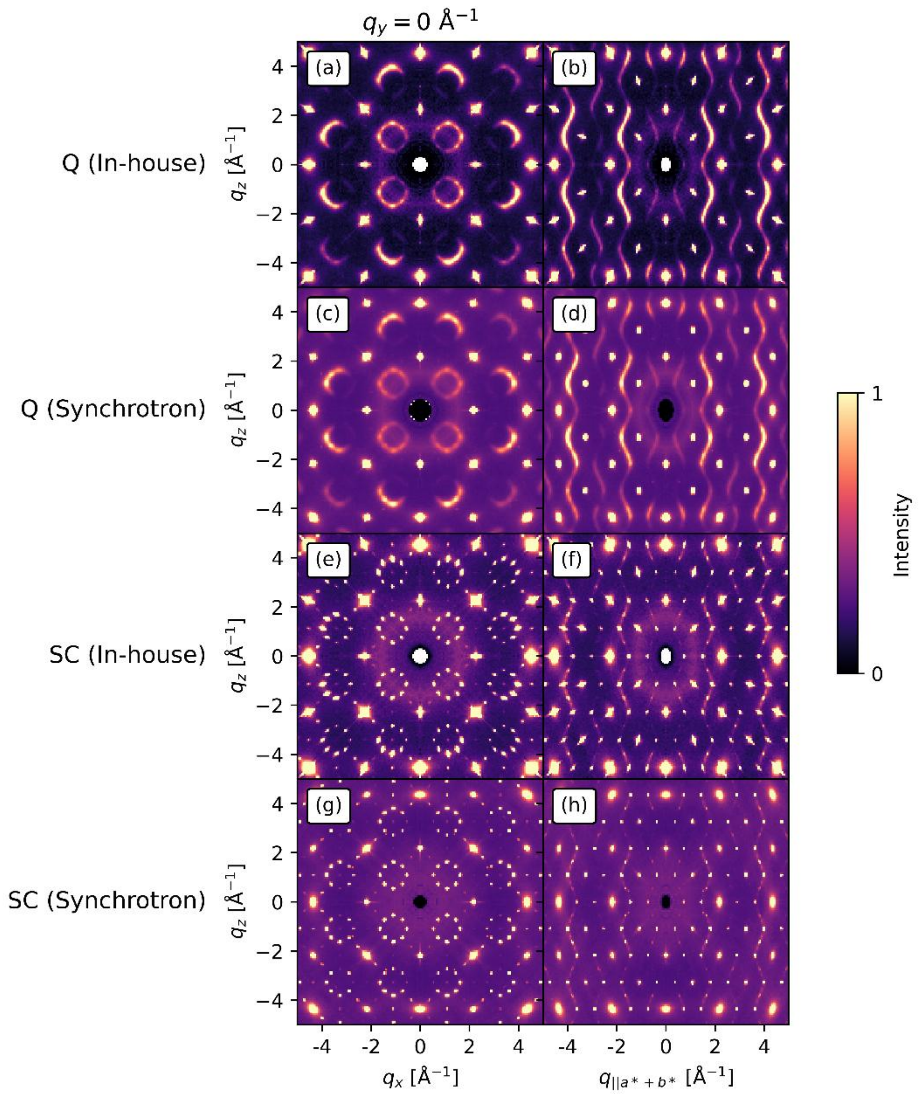
CASE STUDY: Cu_2Se

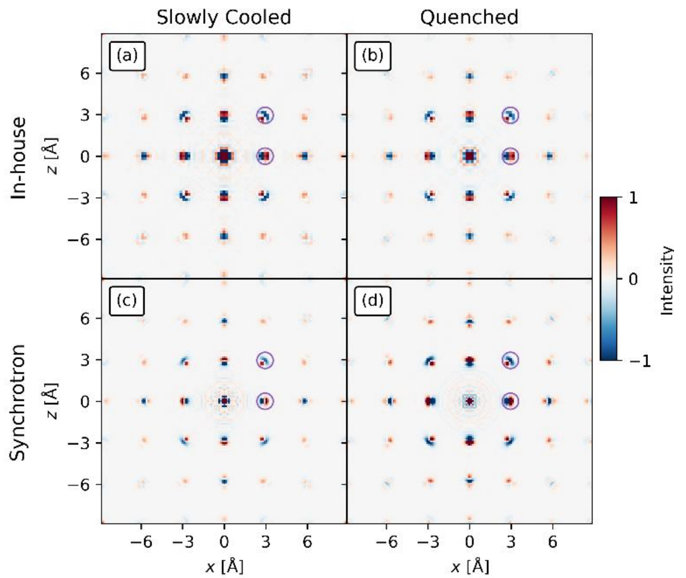




CASE STUDY: $Nb_{1-x}CoSb$







$$\text{peak integral} = \sum_{i,j|\boldsymbol{r}_{ij}=\boldsymbol{r}} \delta Z_i \delta Z_j$$

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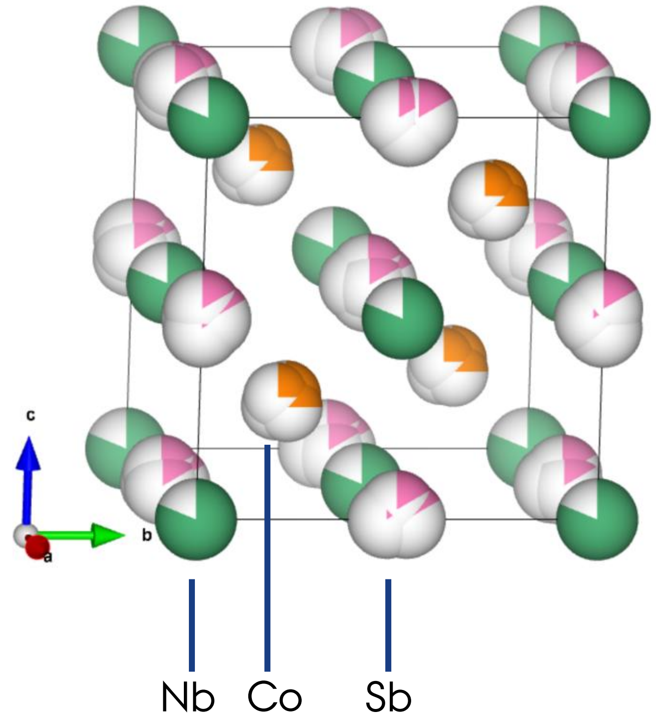

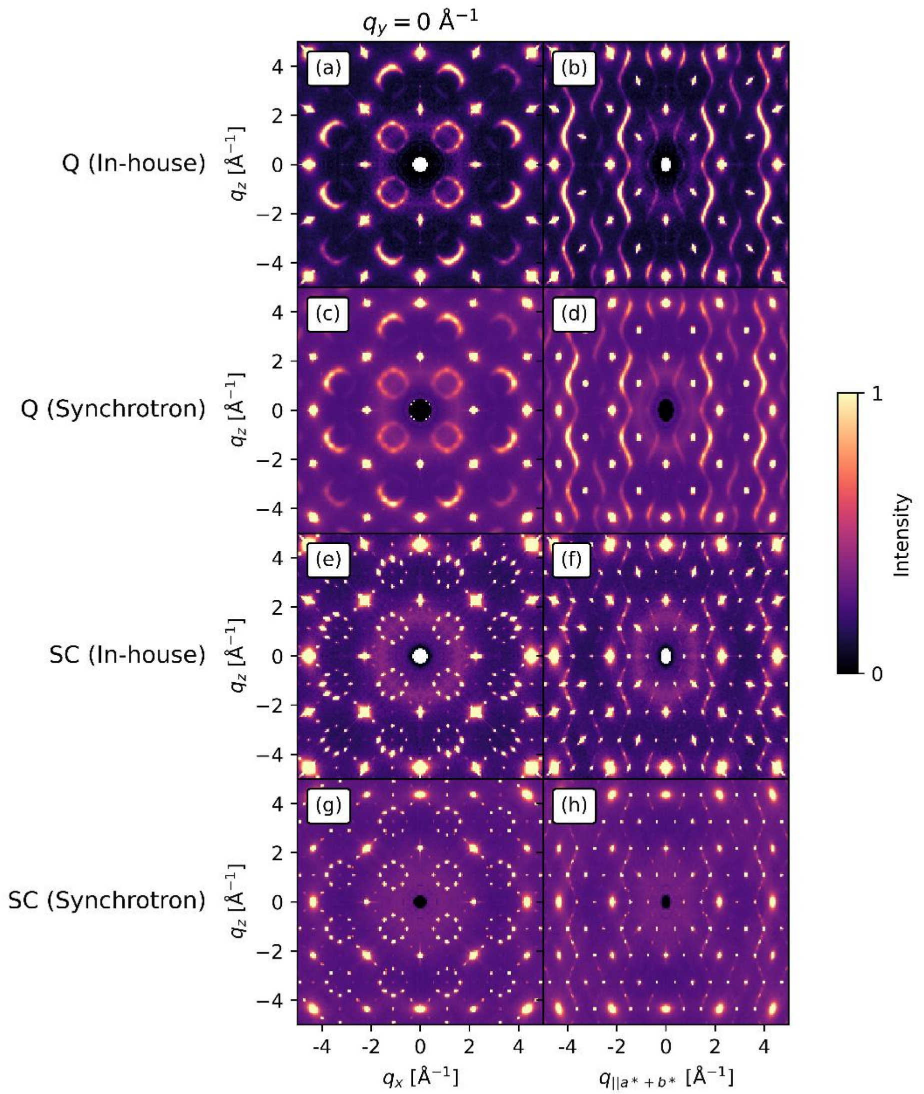

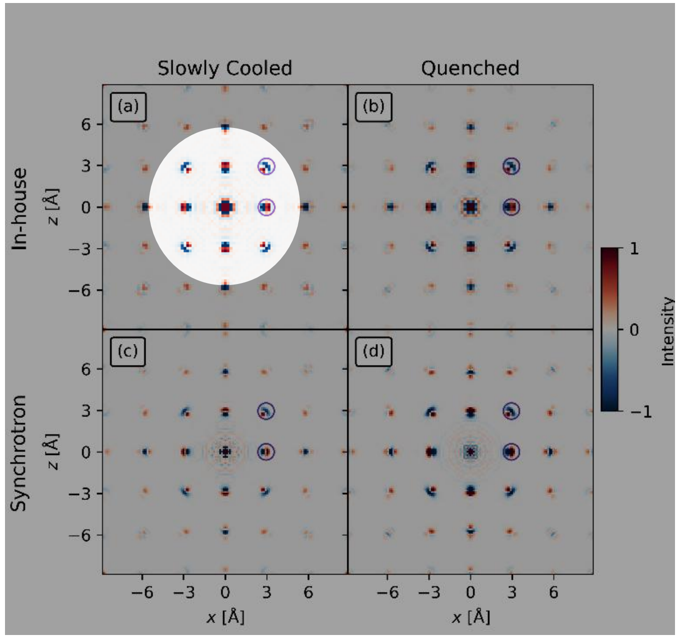

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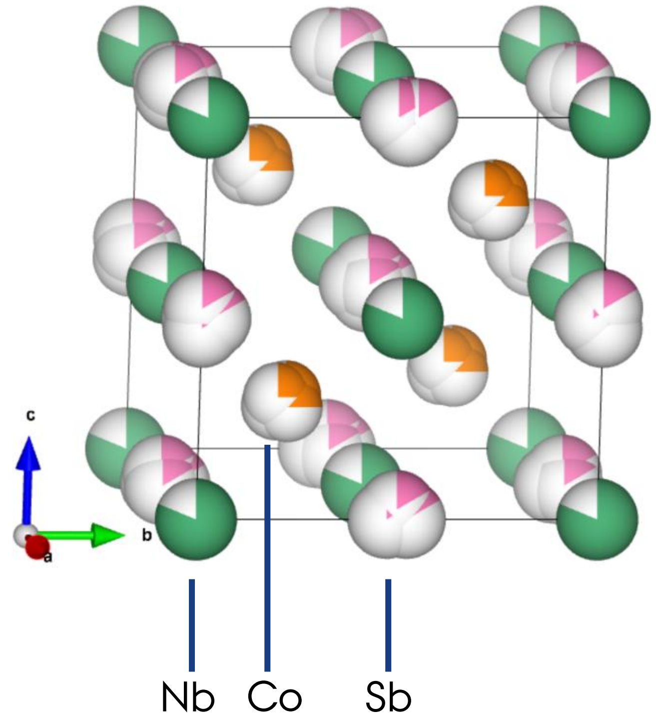
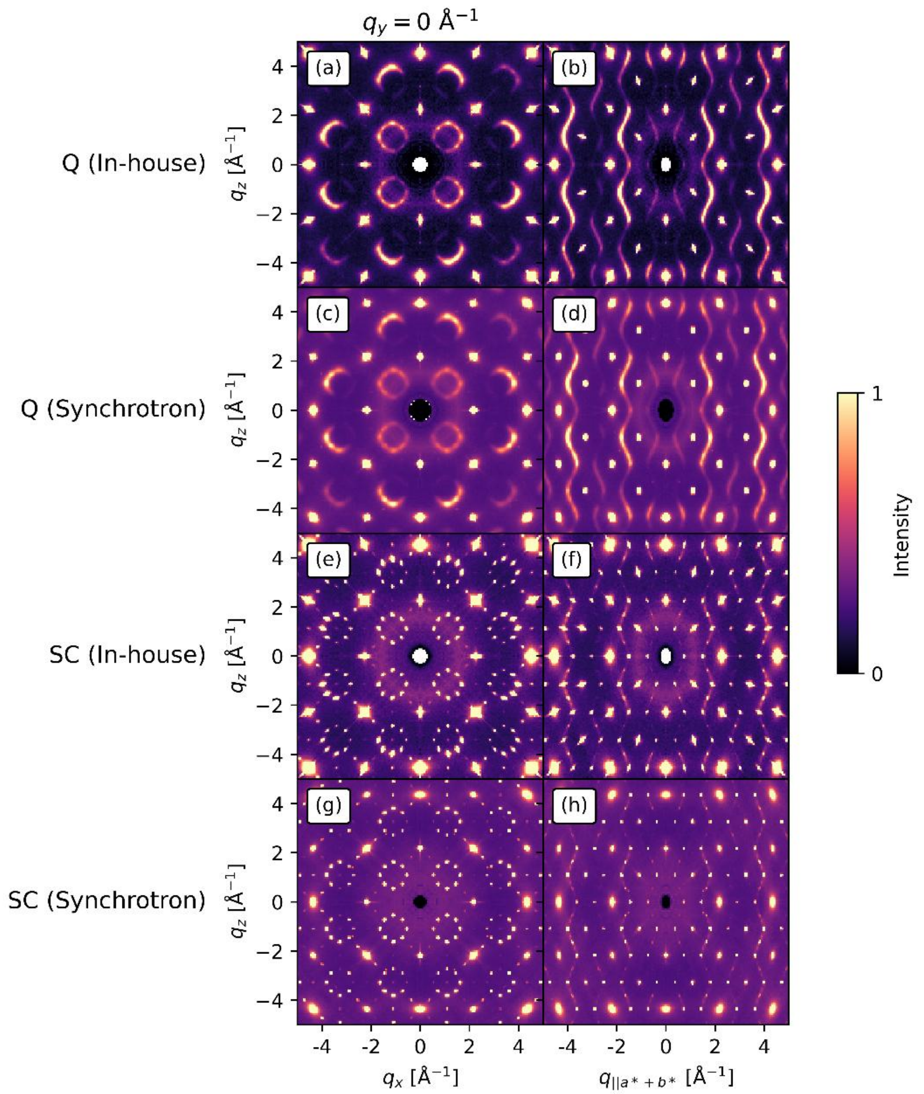
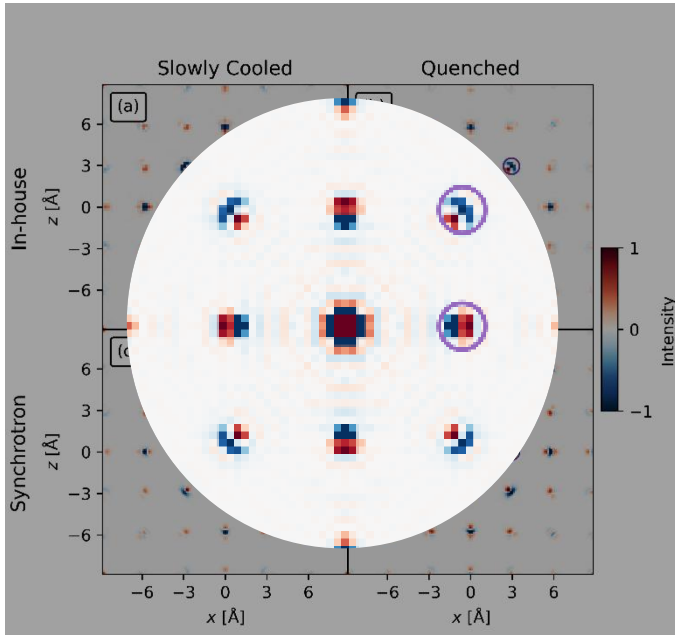
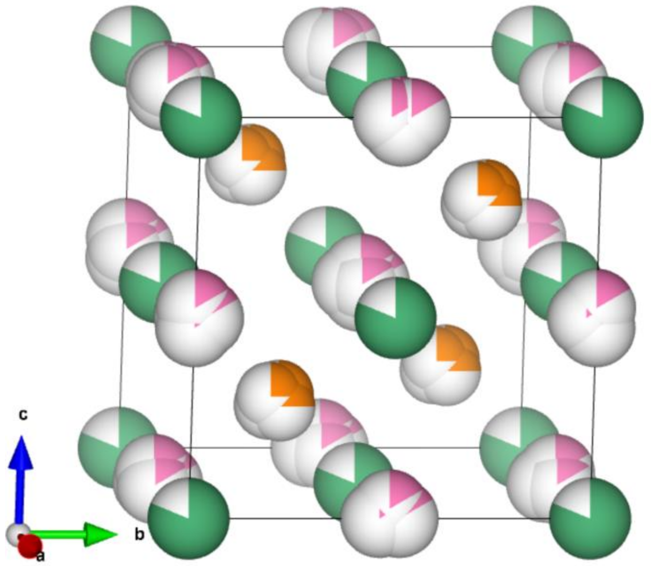


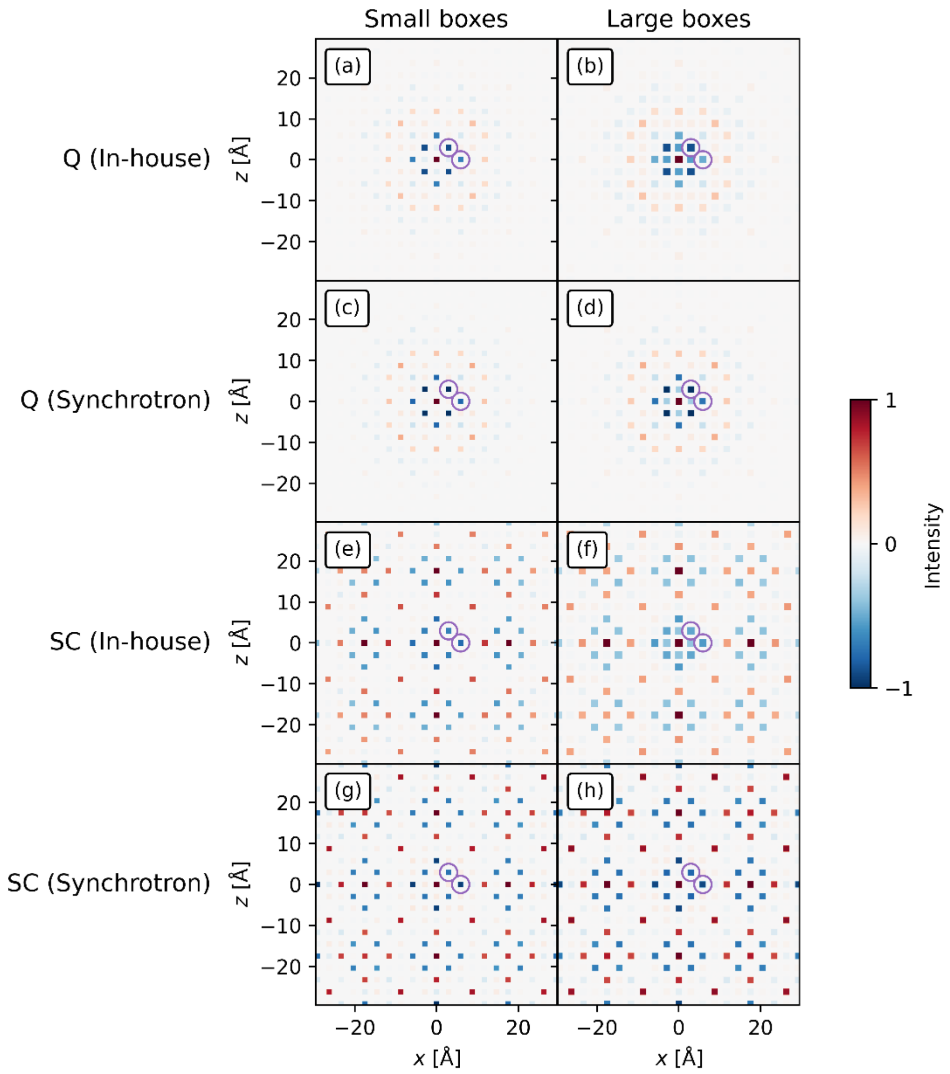


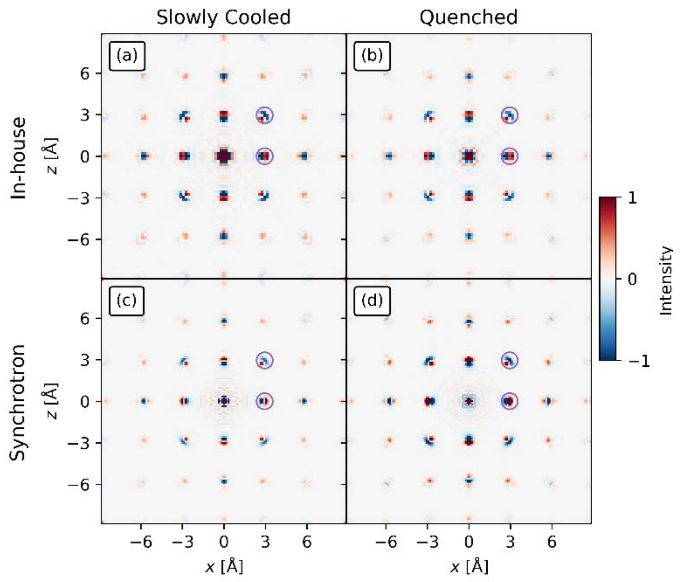


$$\text{peak integral} = \sum_{i,j|\boldsymbol{r}_{ij}=\boldsymbol{r}} \delta Z_i \delta Z_j$$

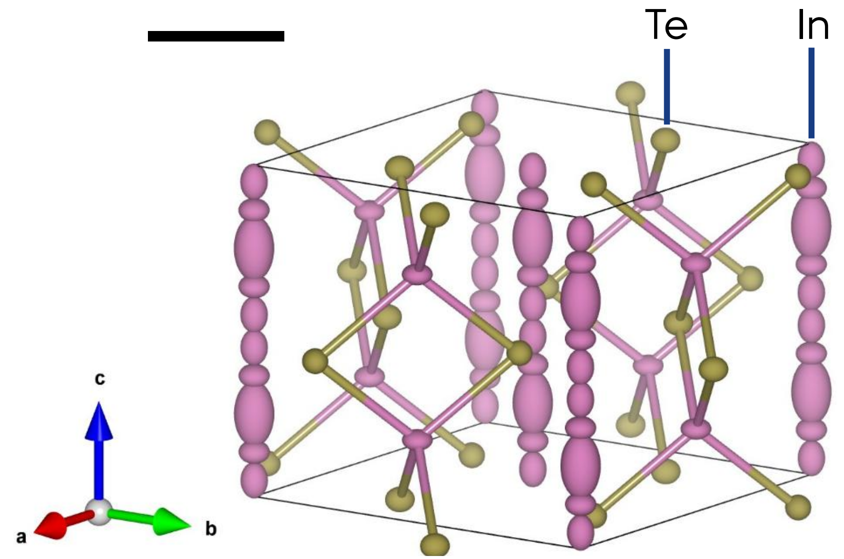
CASE STUDY: $Nb_{1-x}CoSb$

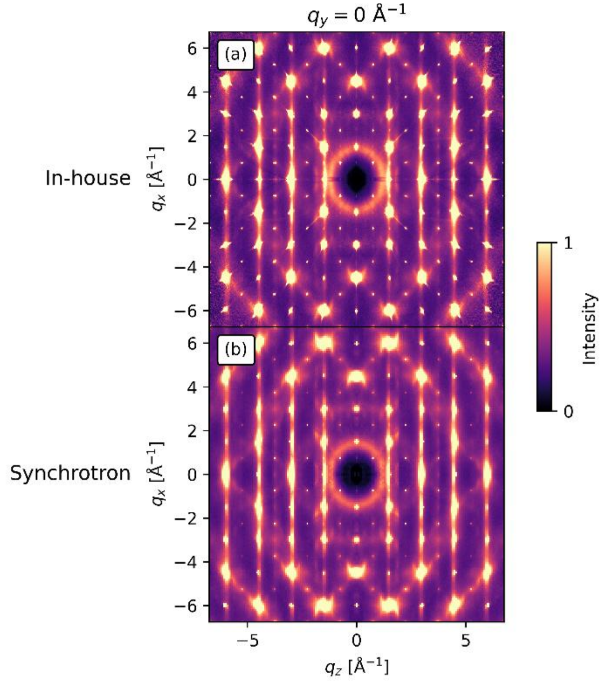


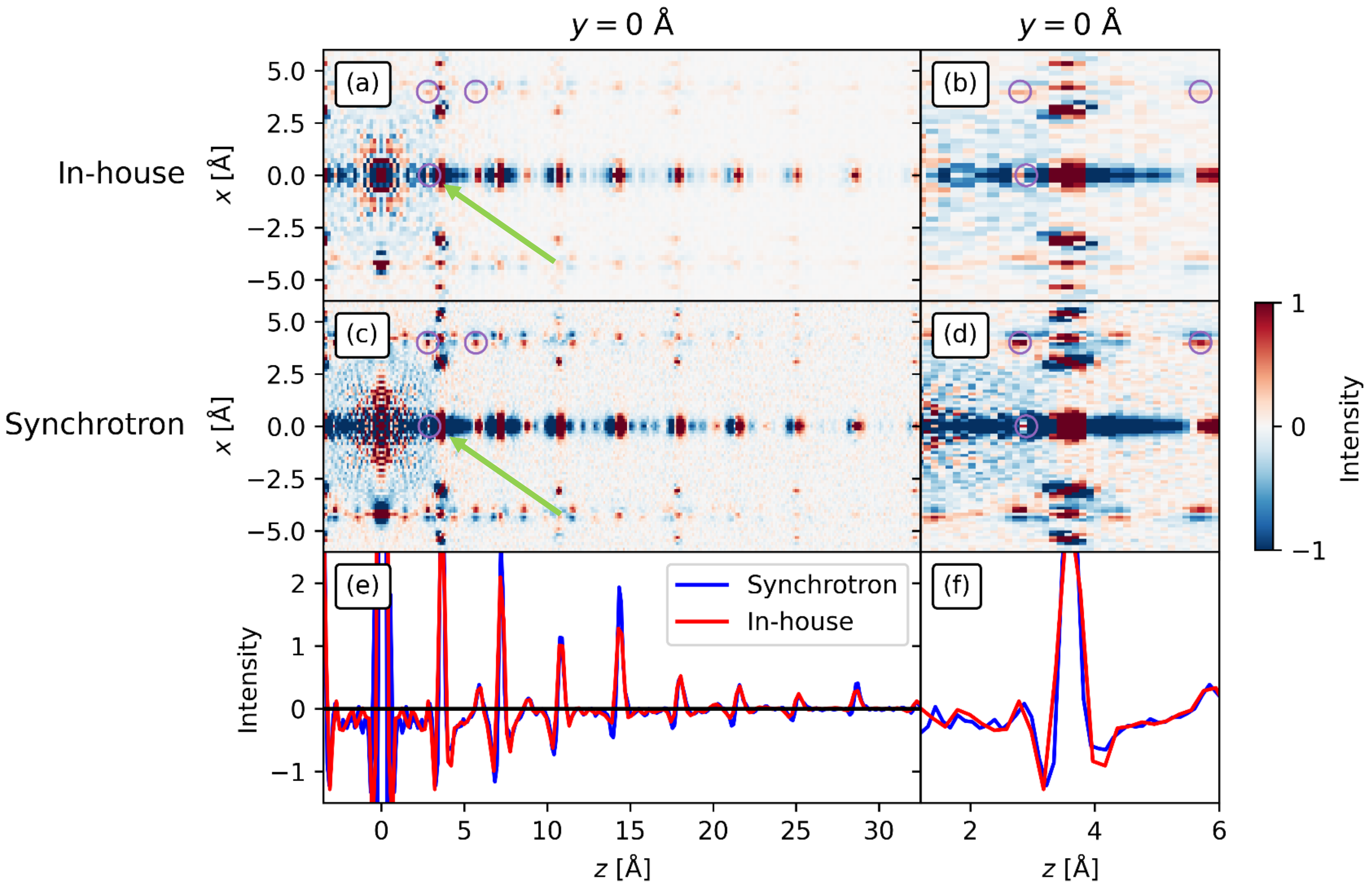




CASE STUDY: InTe







BENCHMARK CONCLUSION

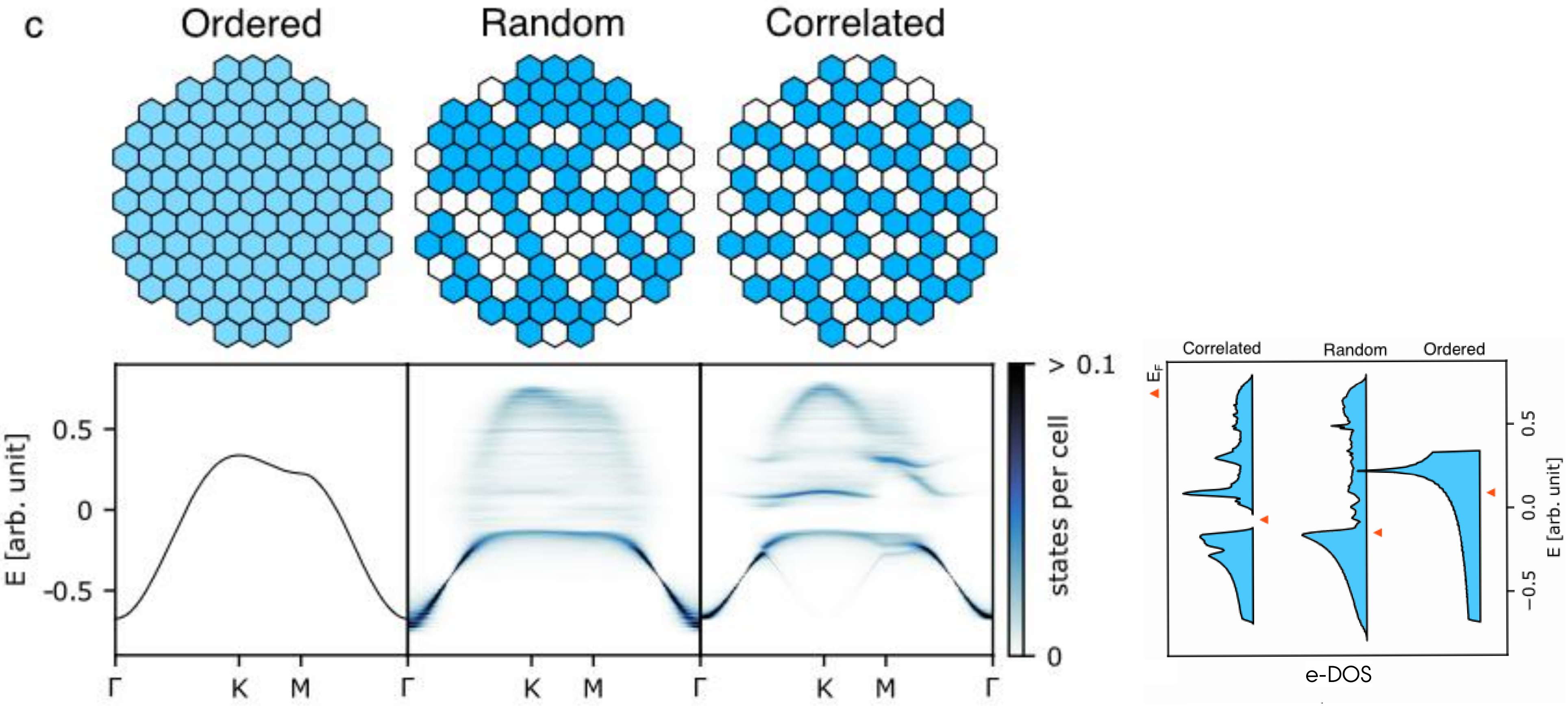
- We **succeeded** in solving the correlated disorder **in Cu_2Se and $Nb_{1-x}CoSb$** using only **inhouse** data but **did not succeed for InTe**.

- **Care** should be taken when **selecting parameters** (like punch size) on **both inhouse and synchrotron data**
- Generally, it is **possible to derive the same conclusions** from inhouse data

	Synchrotron	Inhouse
Accessibility	Less accessible	Very accessible, but with acquisition and maintenance costs.
Radial broadening due to lack of monochromaticity	No	Yes
Beam divergence	Very small	Yes
Resolution	Very good	Medium (acceptable)
Signal-to-noise ratio	Very high/high	Medium/Low
Duration of measurement	Hours (very high flux)	Hours/days (medium/low flux)
Temperature	Can often measure at very low temperatures	Can measure at moderately low temperatures
Difference between elemental sources	-	Higher atomic number gives better resolution, but lower scattering intensities (longer measurements)
Fluorescence?	Change wavelength and/or use fluorescence suppression on detector	Change source if possible and/or use fluorescence suppression on detector

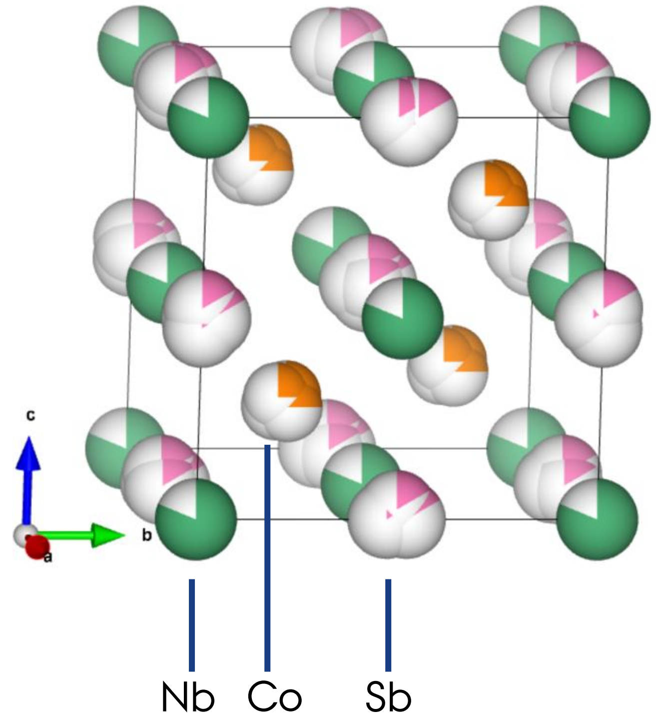
Applications of disorder

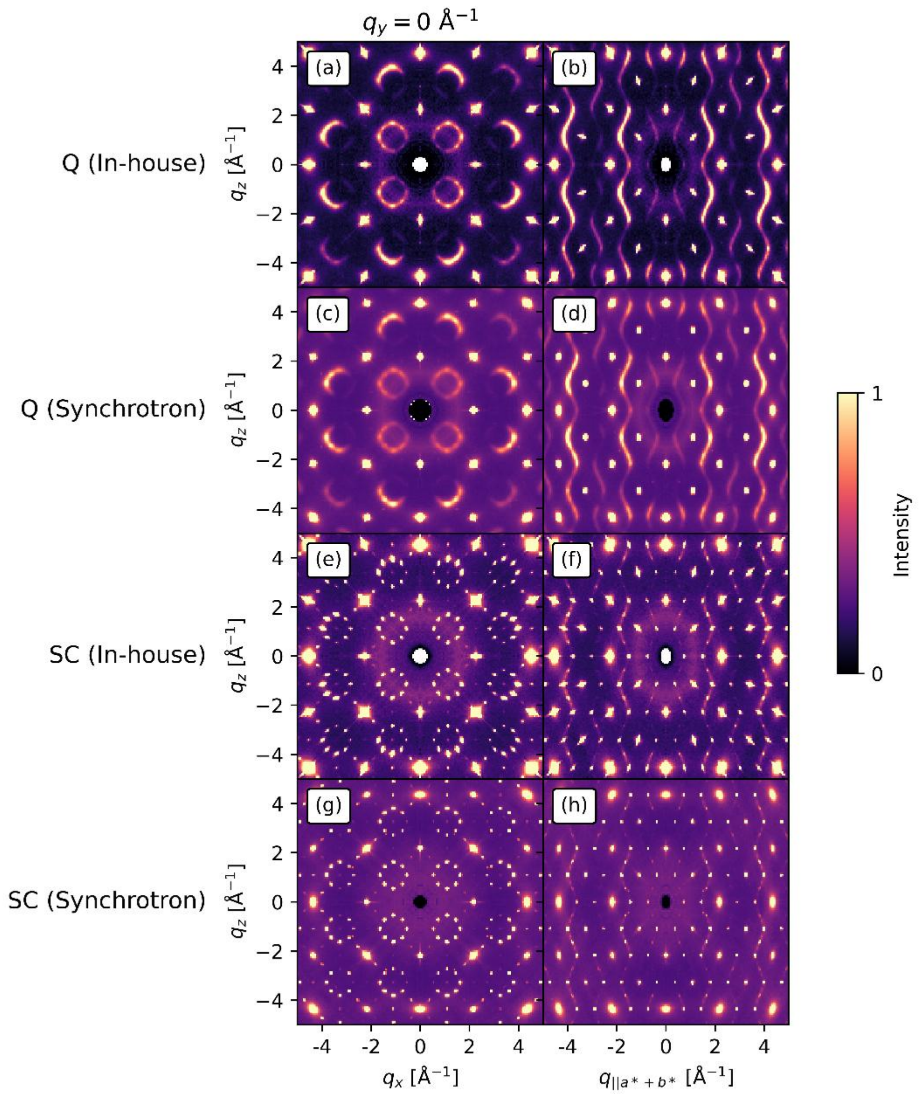
ENGINEERING BAND STRUCTURES

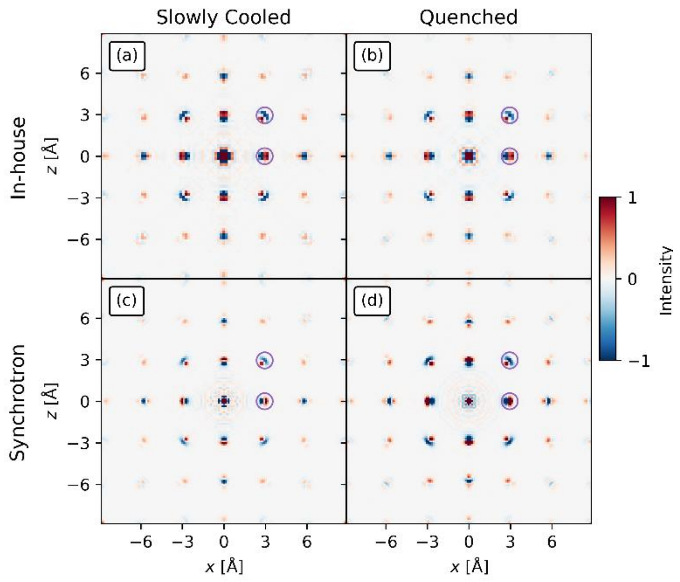


Figures (modified) from Roth & Goodwin (2023). *Nature Commun.*, **14**, 4328

TUNING DISORDER AT SYNTHESIS







$$\text{peak integral} = \sum_{i,j|\boldsymbol{r}_{ij}=\boldsymbol{r}} \delta Z_i \delta Z_j$$

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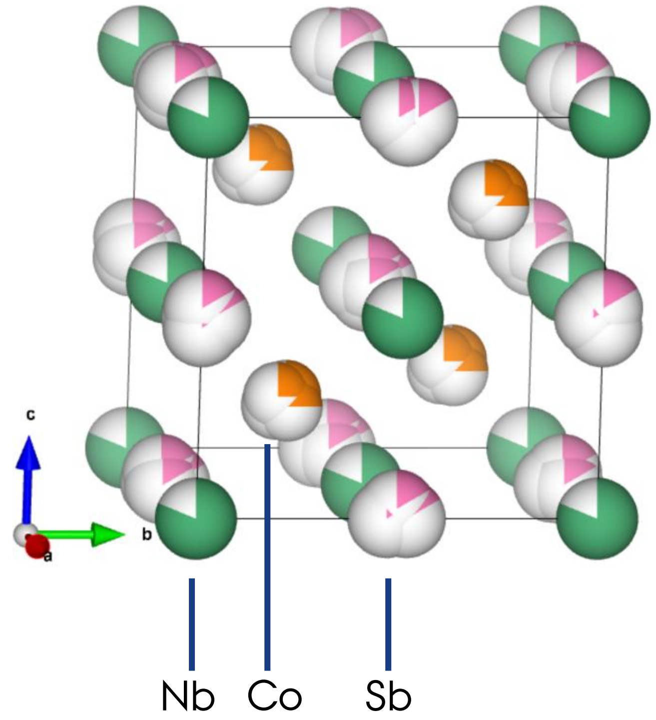

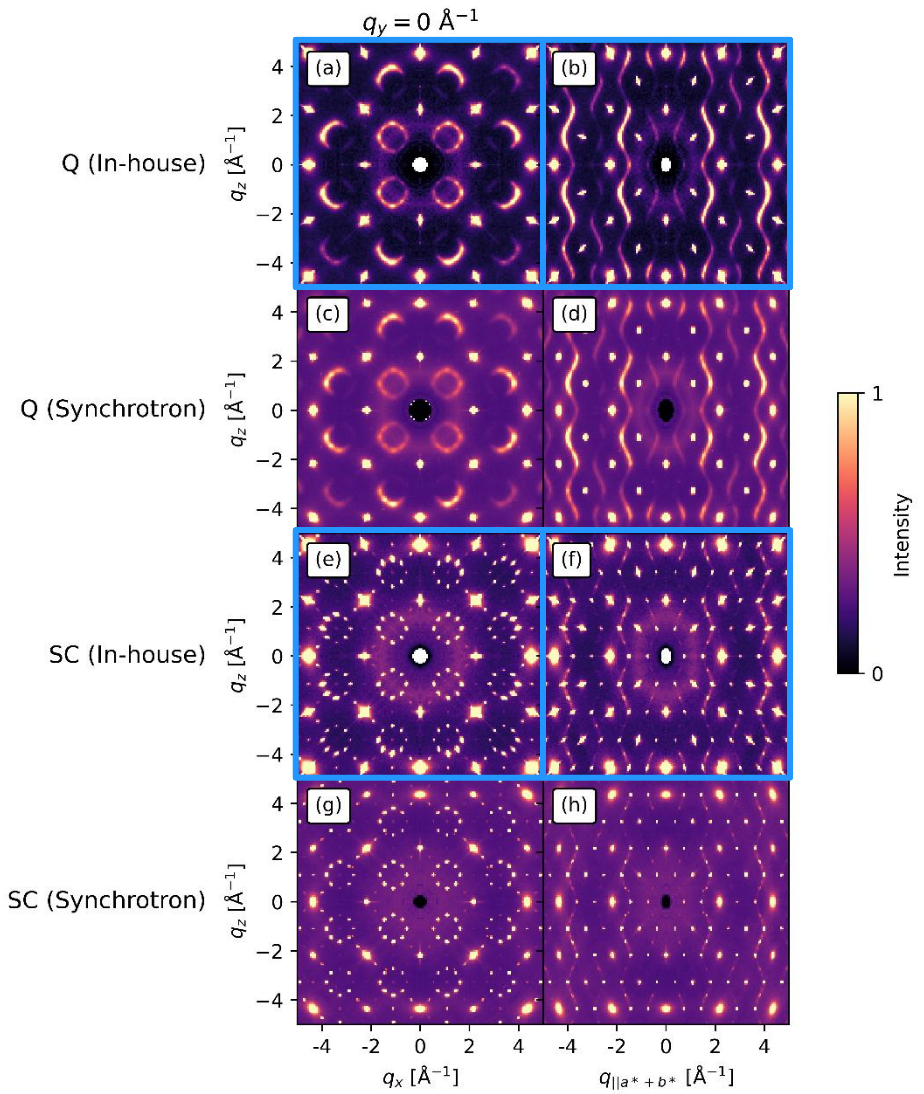

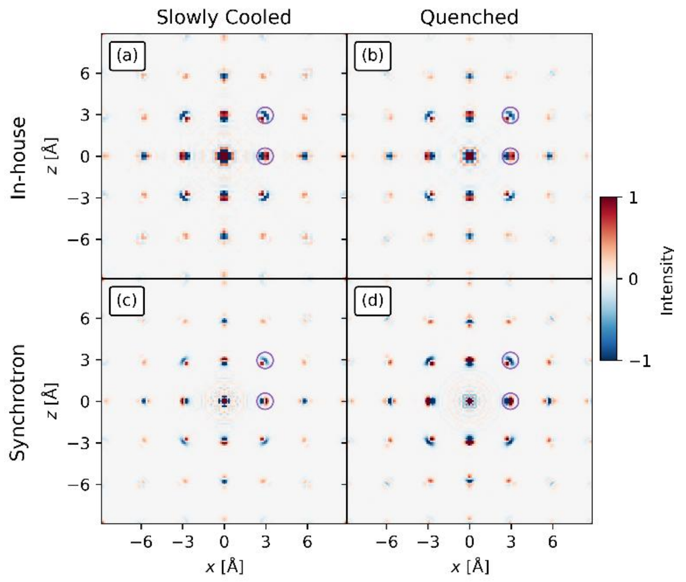

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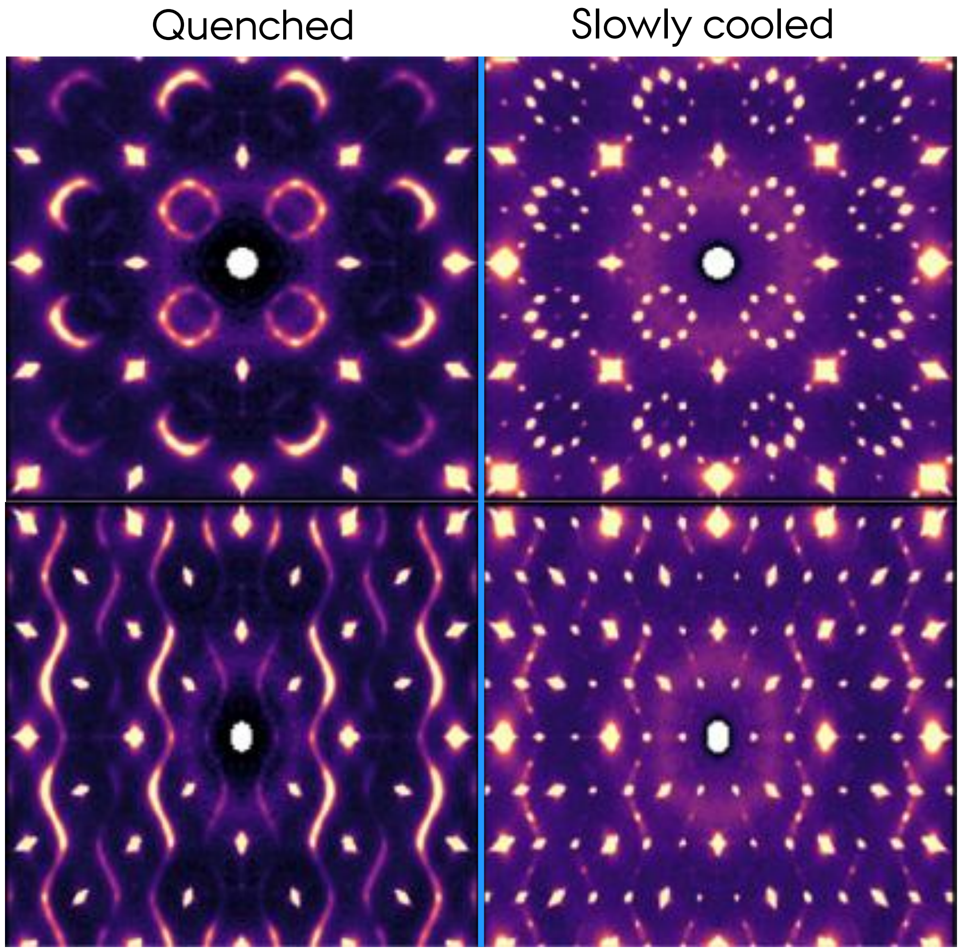
TUNING DISORDER AT SYNTHESIS

$Nb_{1-x}CoSb$



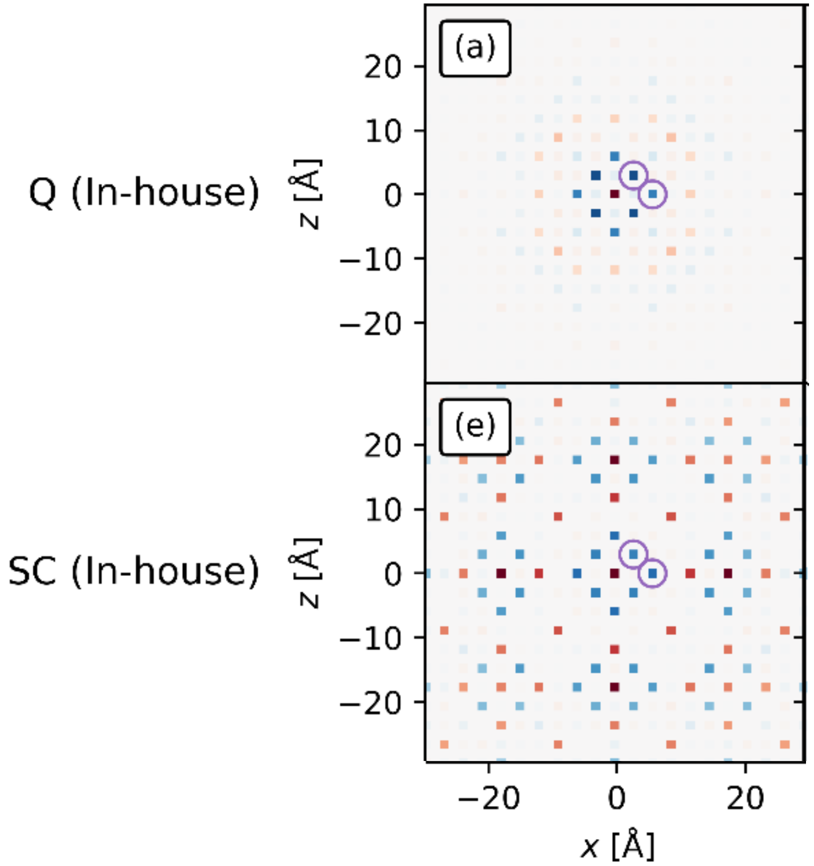


Figure adapted from front page,
Acta Cryst. A, July 2025

REVERSIBLE TRANSITIONS BETWEEN DISORDERED STATES

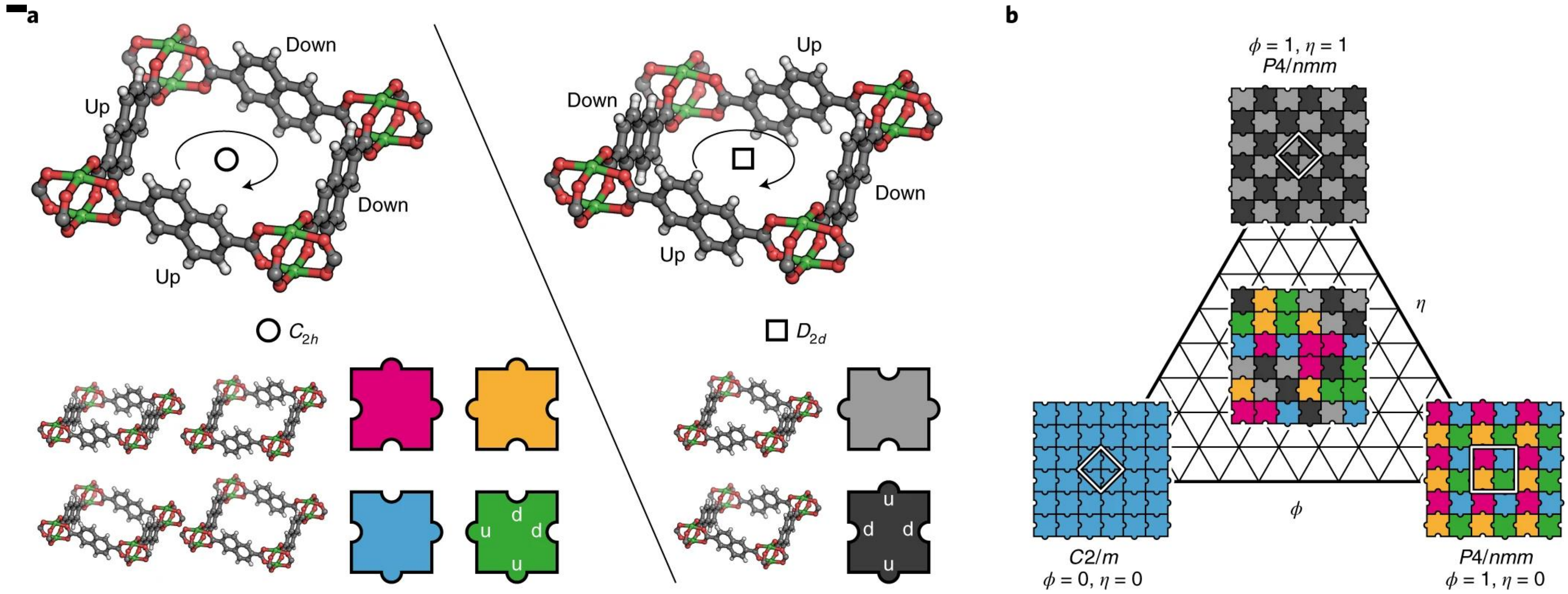


Figure from Ehrling *et al.* (2021). *Nature Chem.*, **13**, 568-574

REVERSIBLE TRANSITIONS BETWEEN DISORDERED STATES

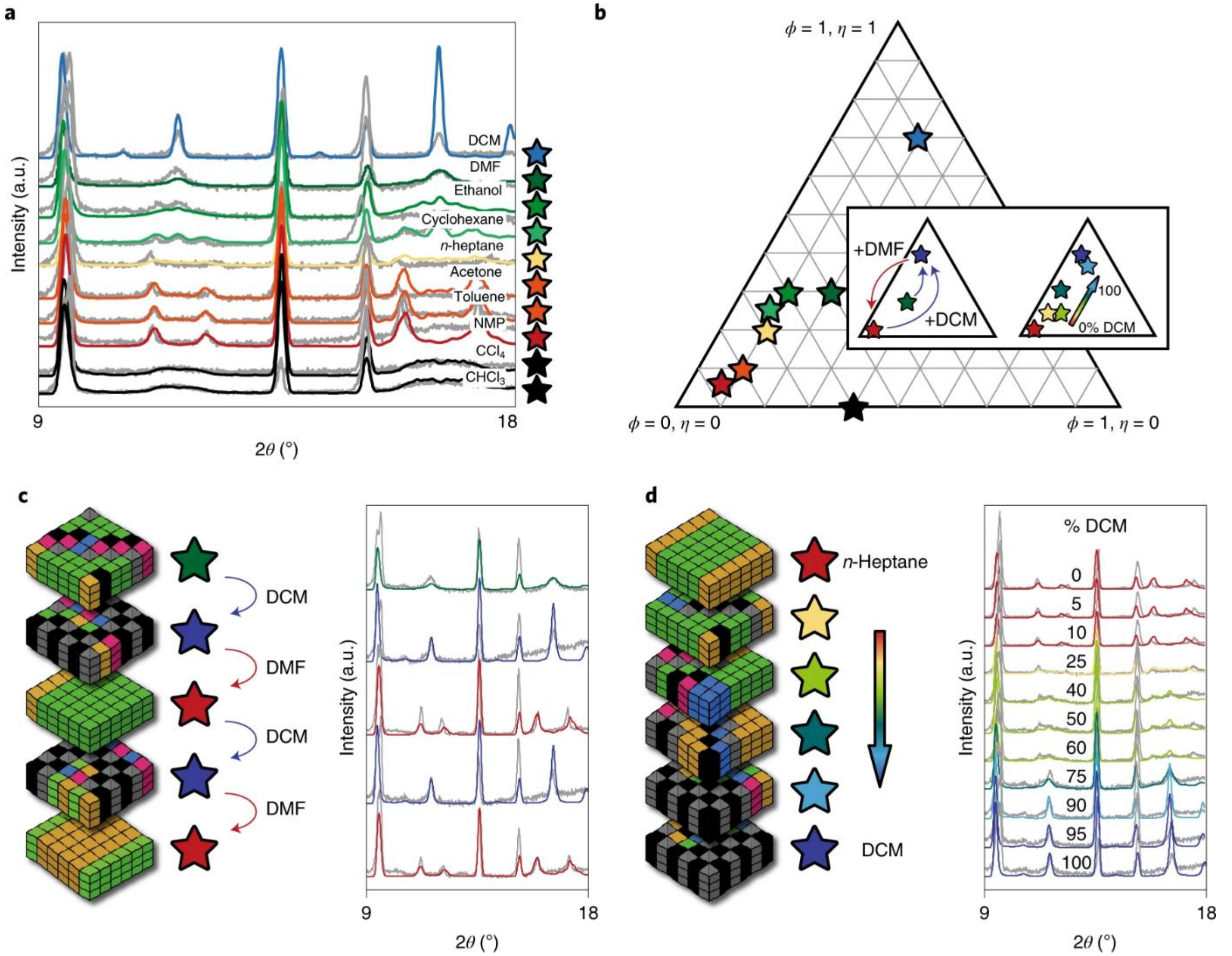


Figure from Ehrling *et al.* (2021). *Nature Chem.*, **13**, 568-574

ACKNOWLEDGEMENTS

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Bo Brummerstedt Iversen
Kristoffer Andreas Holm Støckler
Andrew Leslie Goodwin

Jacob Rixen Østerbro
Viktoria Kaznelson
Emilie Skytte Vosegaard
Jacob Svane

And you! For your attention.

Danmarks Grundforskningsfond
Danish National Research Foundation

DanScatt

CENSEMAT
center for sustainable energy materials

DANMARKS FRIE FORSKNINGSFOND
INDEPENDENT RESEARCH FUND DENMARK

SOLIDUM PETIT IN PROFUNDIS · UNIVERSITAS ARHUSIENSIS ·

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Roth & Goodwin (2023). *Nature Commun.*, **14**, 4328

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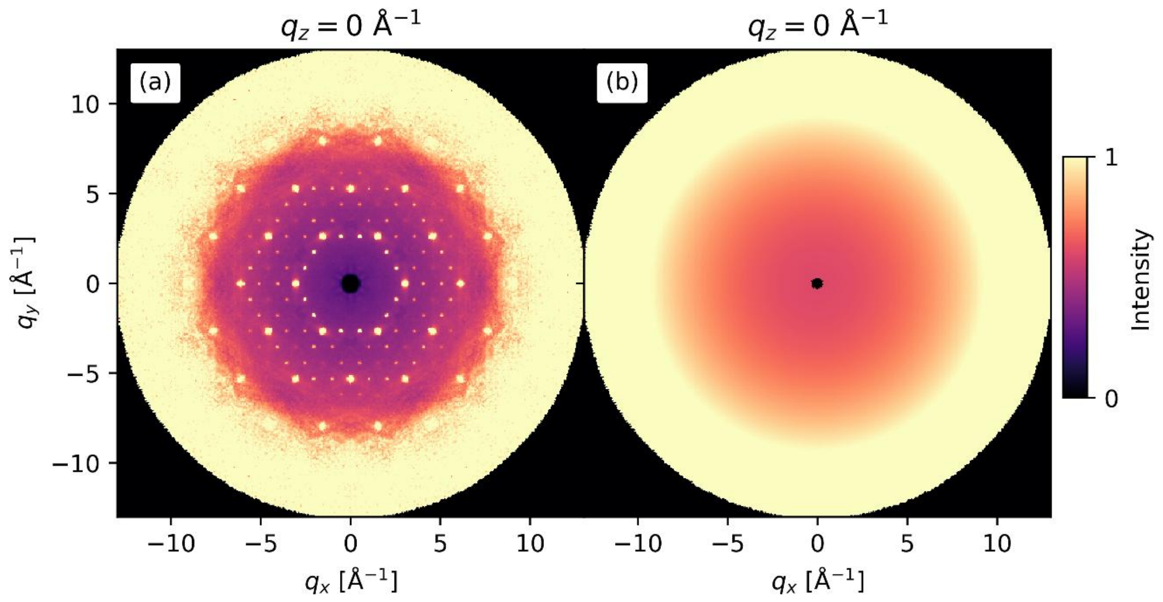
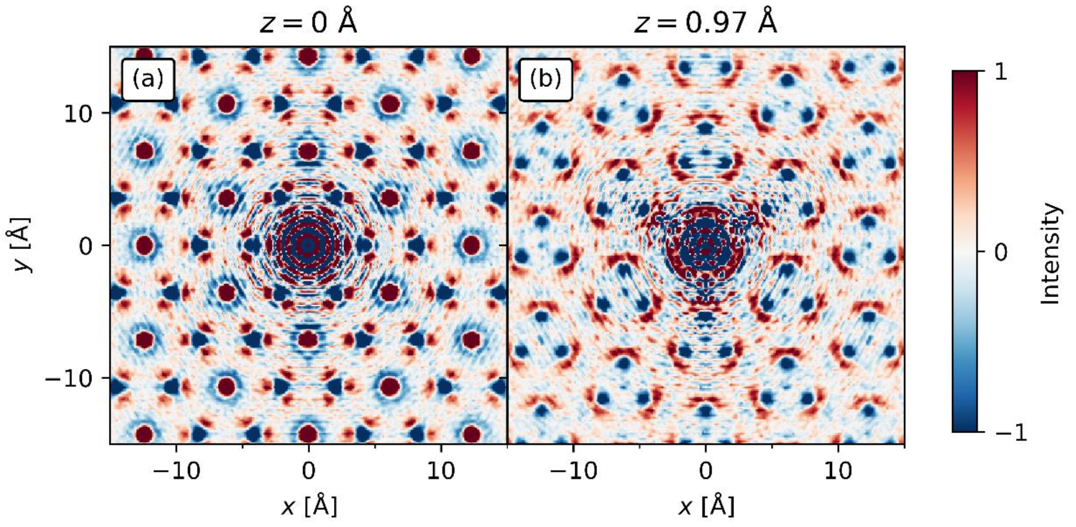
THE EFFECT OF FLUORESCENCE





INSTRUMENT SPECIFICATIONS

Instrument	Rigaku XtaLAB Synergy-S
X-ray tube	PhotonJet-S microfocus sealed tube
Radiation	Mo Kα
Detector	HyPixArc-100°
Geometry	Kappa-goniometer