

Domain formation and planar defects in NASICON electrolyte materials

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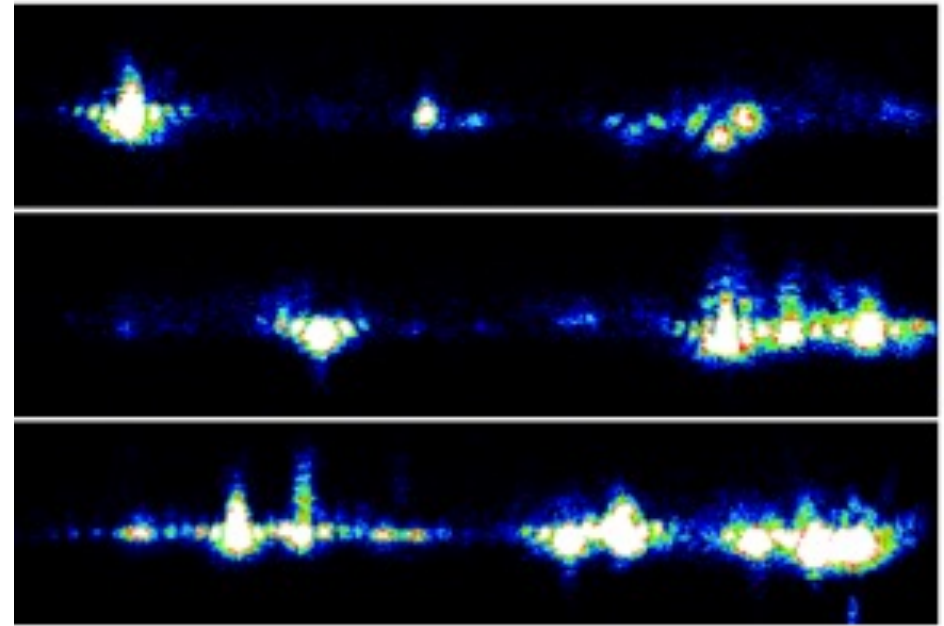
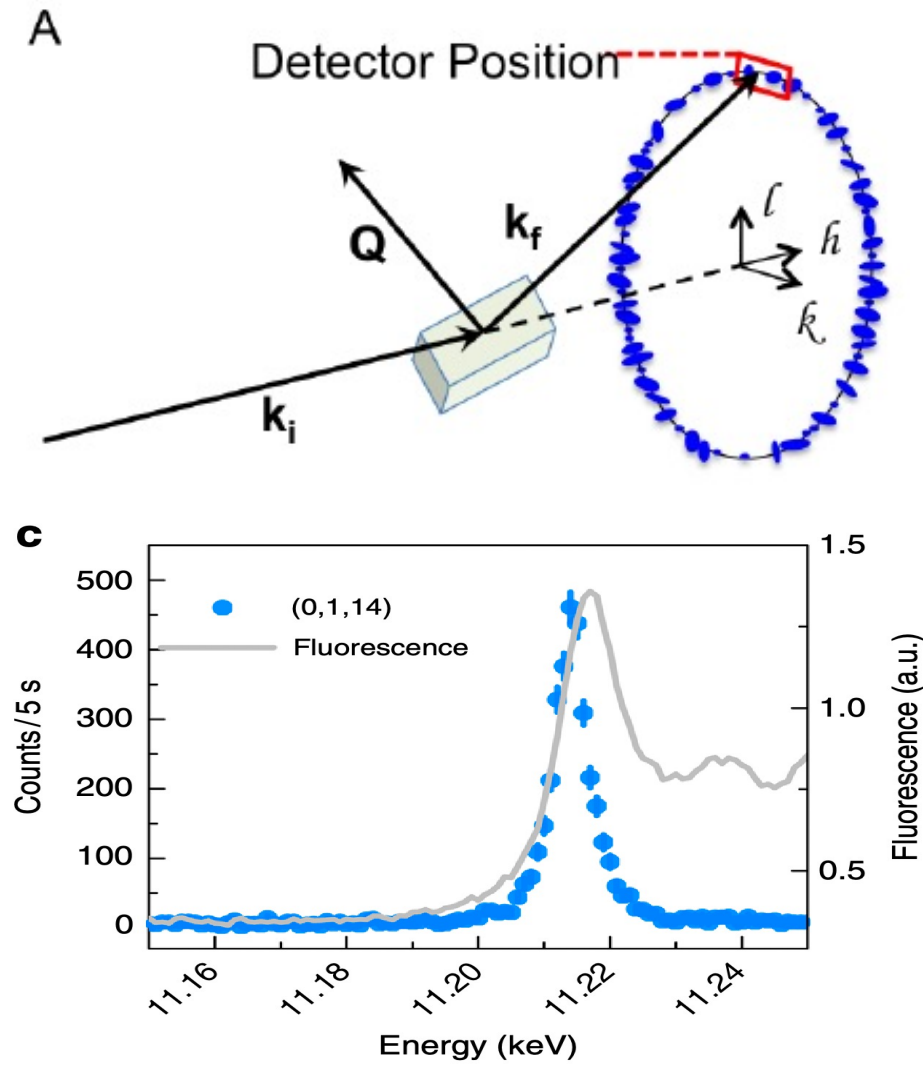
“MS-069: Crystallography of solid-state ionic conductors”

IUCr Congress, Calgary, August 2026

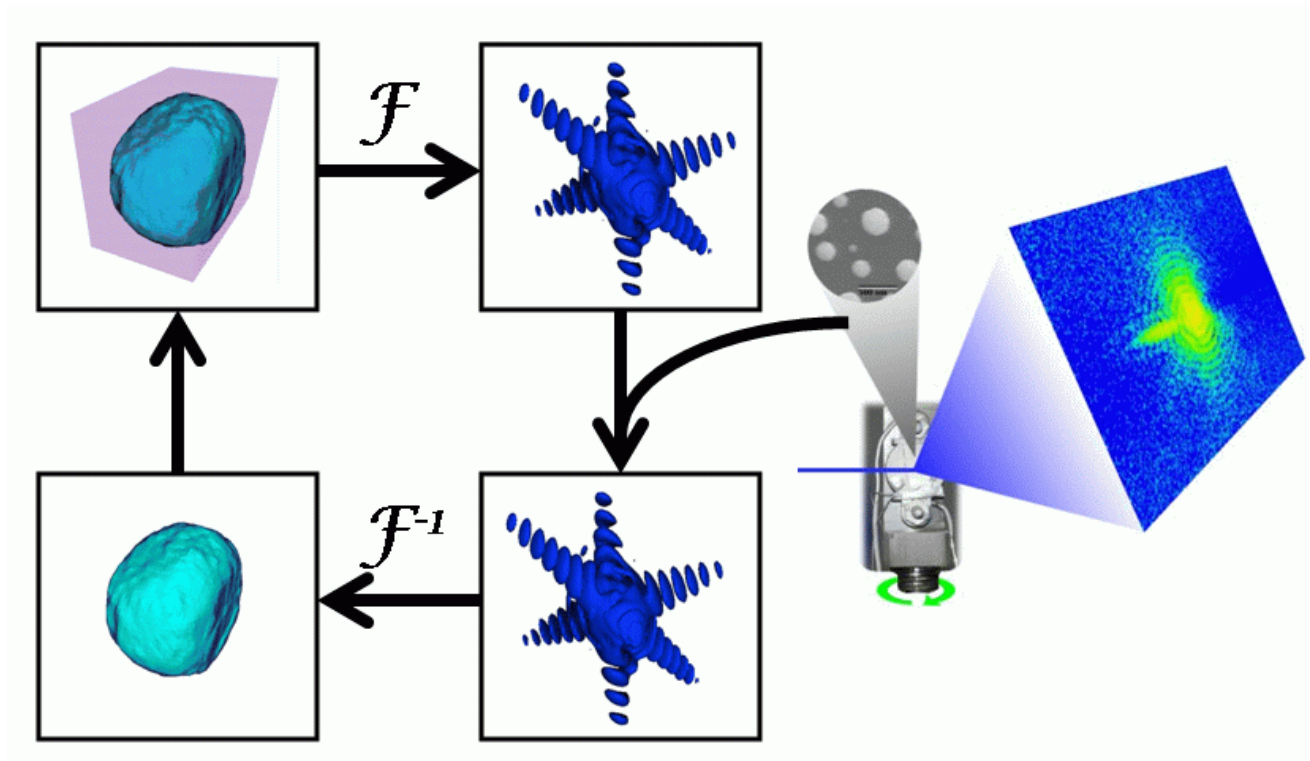
Bragg Coherent X-ray Diffraction

- Bragg Coherent Diffraction Imaging (BCDI)
- Sensitivity to strain
- Phase domain structures
- Domain counting rule
- Solid State Electrolyte Crystals
- High Entropy conduction
- Cross sectional phase images
- Phase-domain-wall conduction pathway

(Resonant) Coherent X-ray Diffraction



Generic “Error Reduction” method

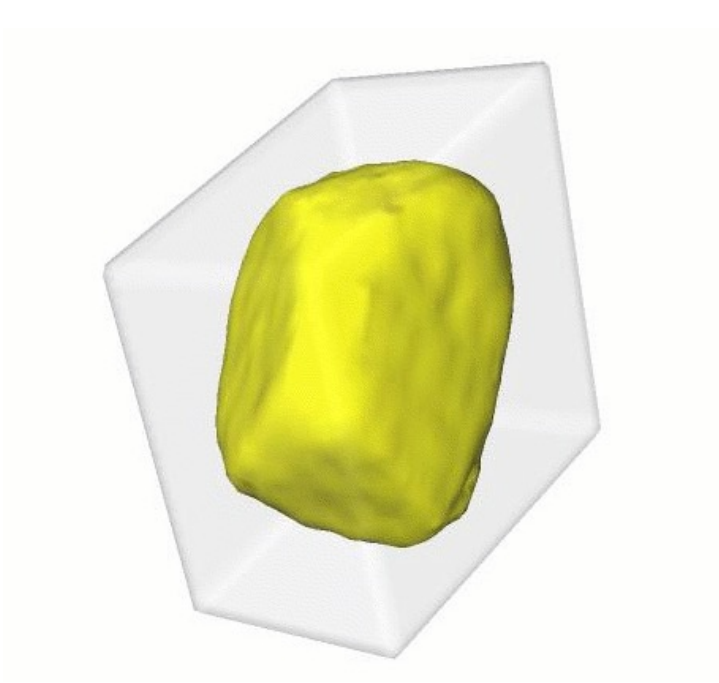
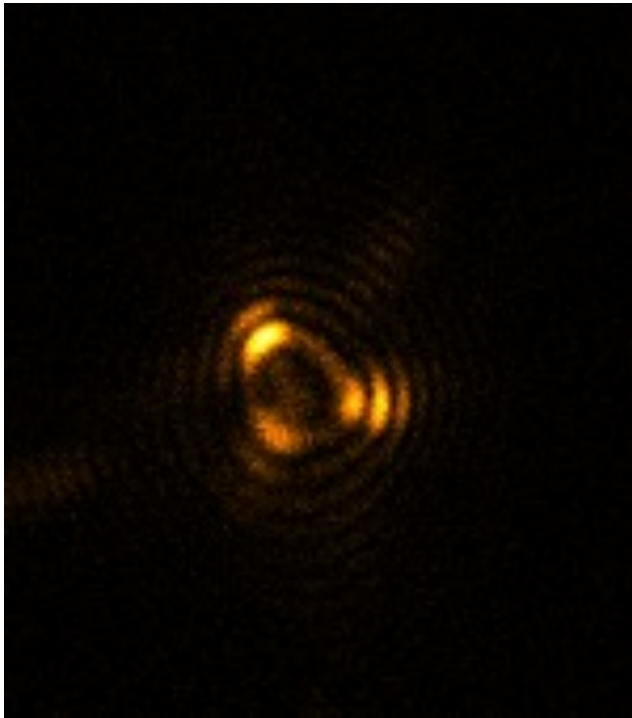


J. R. Fienup Appl. Opt. 21 2758 (1982)

R. W. Gerchberg and W. O. Saxton Optik 35 237 (1972)

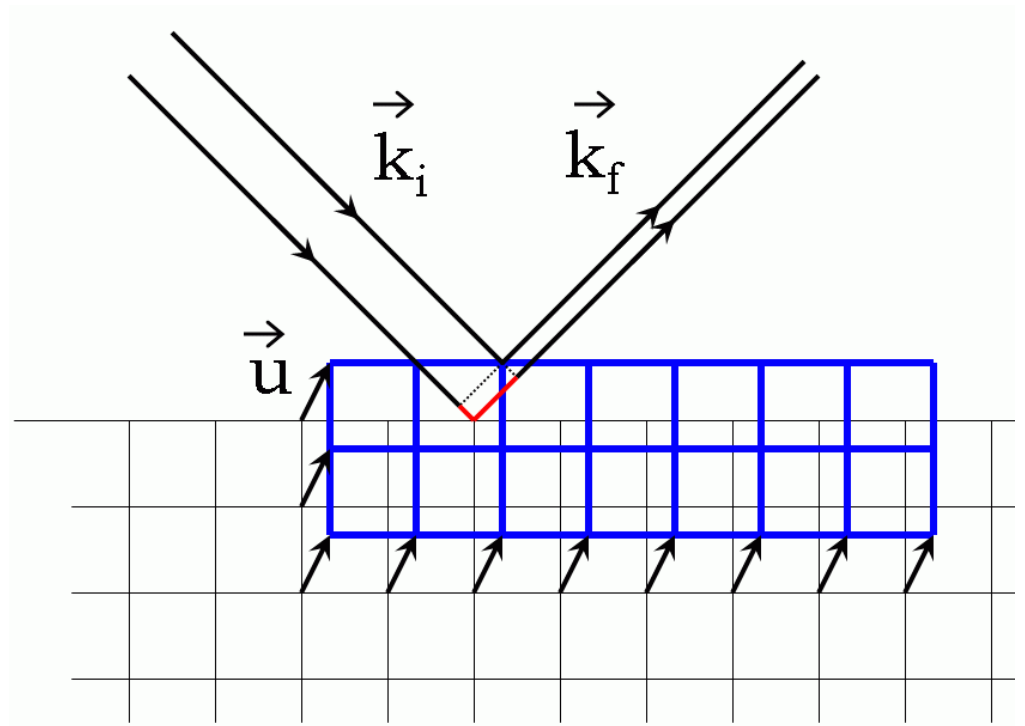
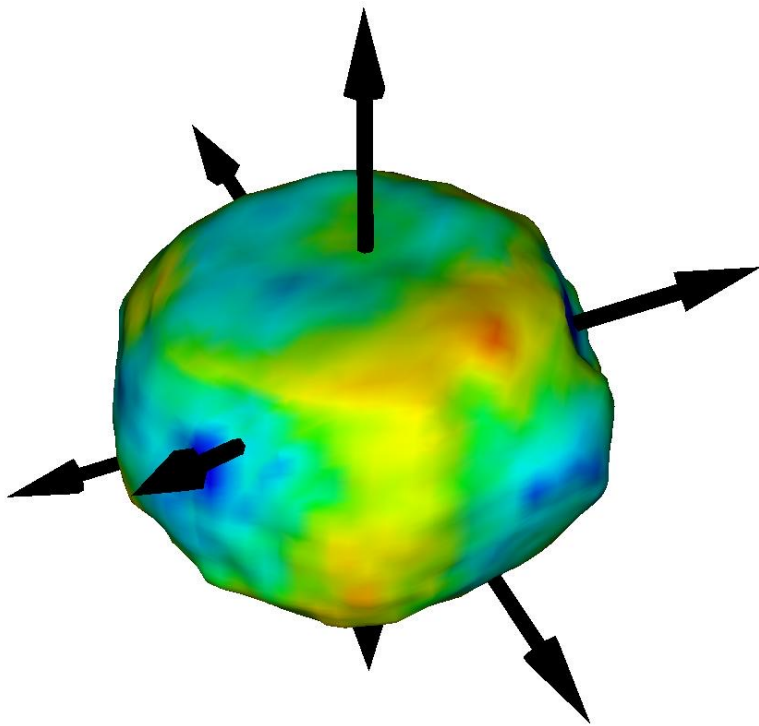
Gold nanocrystal reconstruction

showing support used for 20 HIO followed by 10 ER



Phase isosurface showing residual strain

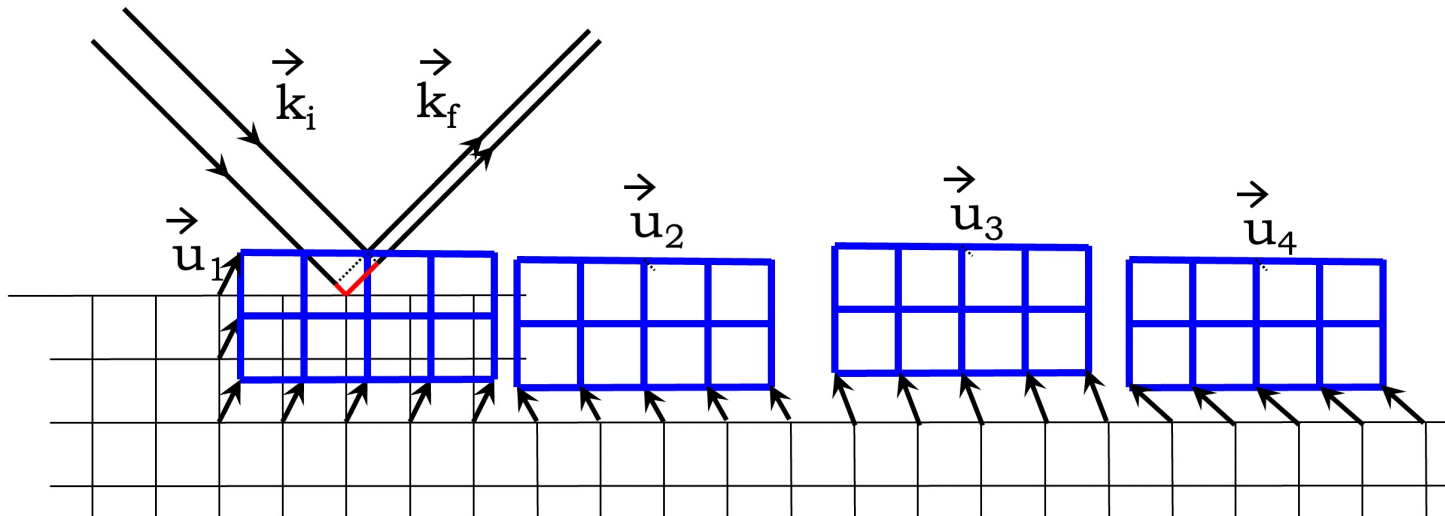
$$\Delta\phi = \mathbf{k}_f \cdot \mathbf{u} - \mathbf{k}_i \cdot \mathbf{u} = \mathbf{Q} \cdot \mathbf{u}$$



Sensitivity to domain offsets

$$\Delta\phi = \mathbf{k}_f \cdot \mathbf{u} - \mathbf{k}_i \cdot \mathbf{u} = \mathbf{Q} \cdot \mathbf{u}$$

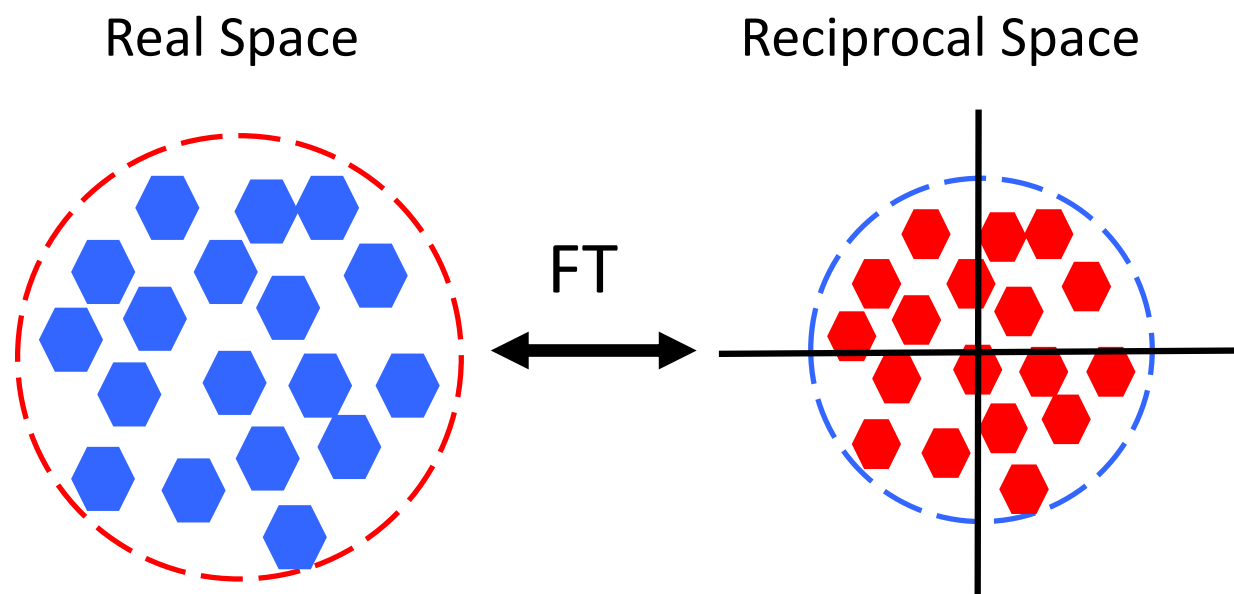
Difficult convergence to unique solution



I. K. Robinson, Synchrotron Radiation News **37** 11 (2024)

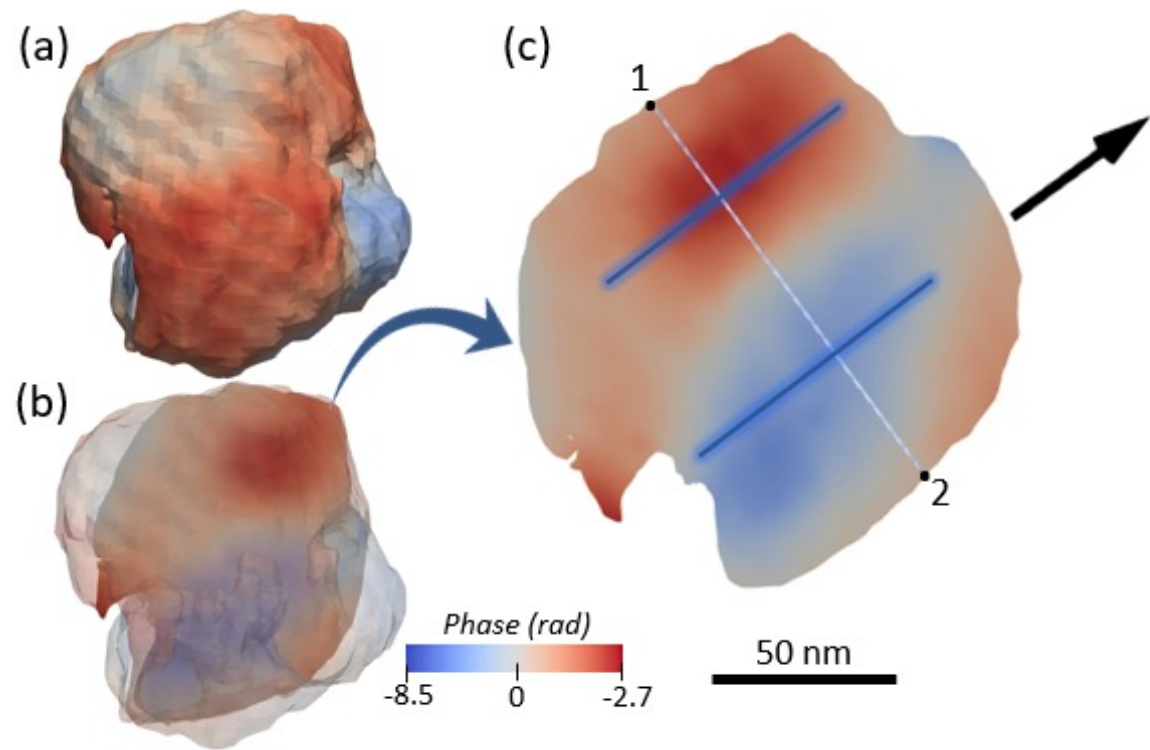
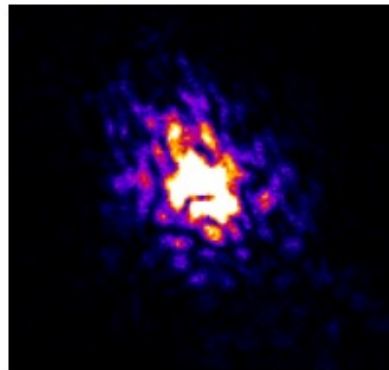
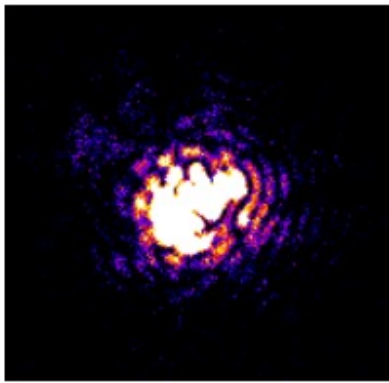
Domain Counting in Coherent Diffraction

Ian Robinson et al, J. Supercond. and Novel Magnetism **33** 99 (2020)



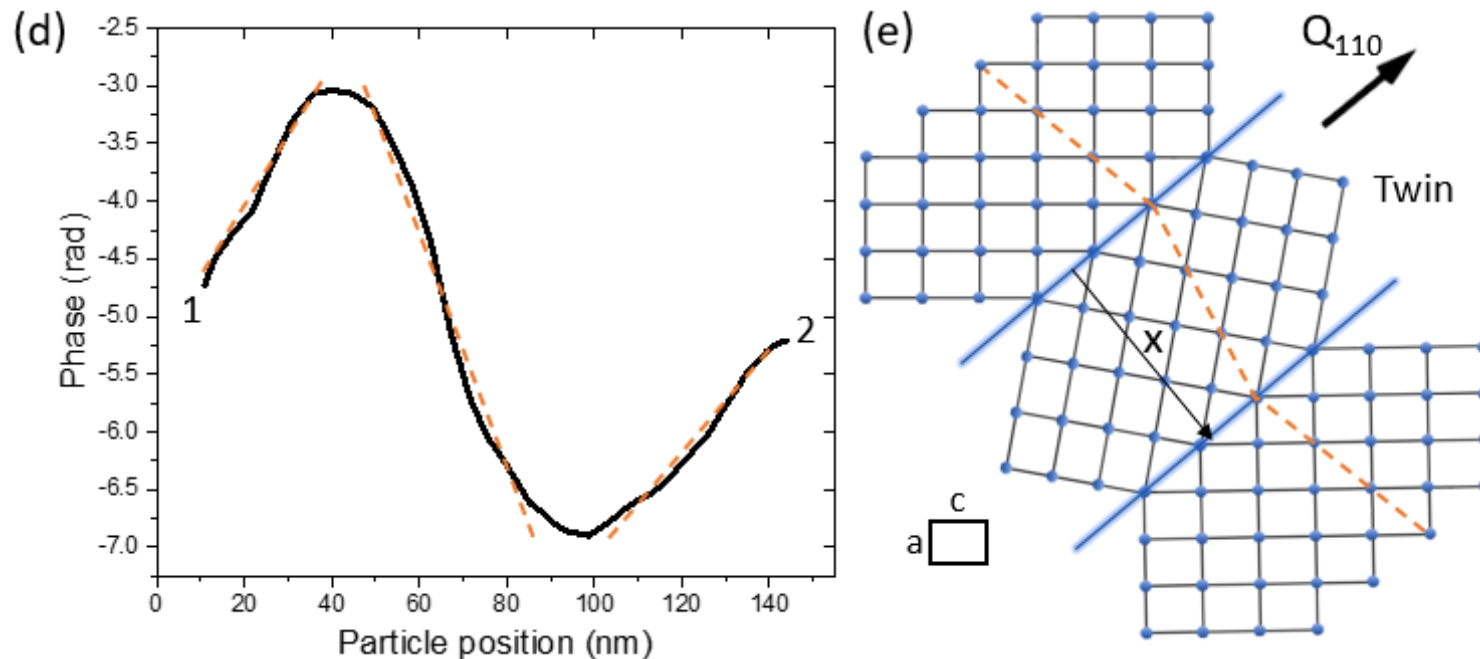
Nano BaTiO₃ MDielectric Material in MLC Capacitors

Tetragonal small-angle grain boundaries



Nanoscale Merohedral Twinning

90° grain boundaries energetically favored



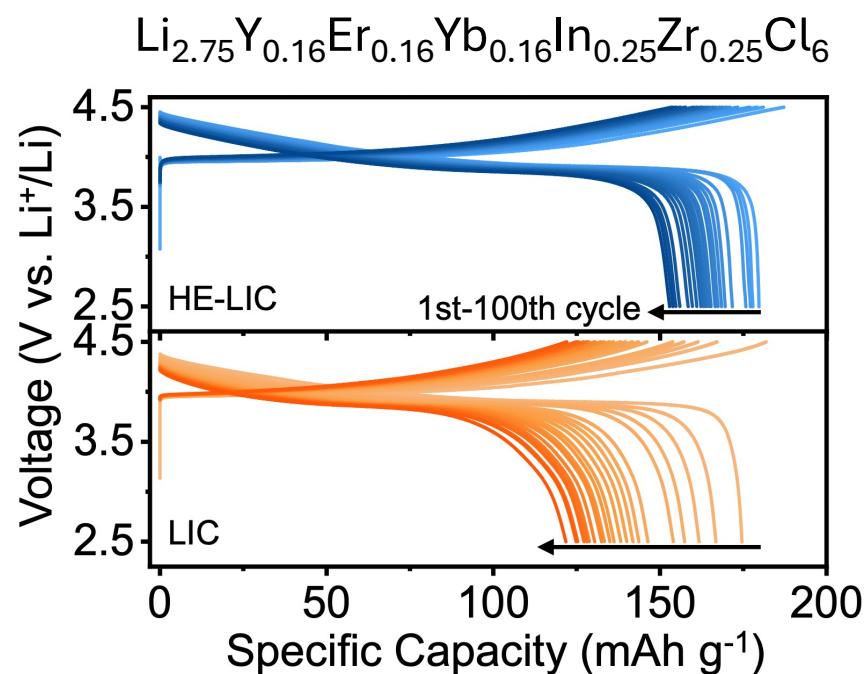
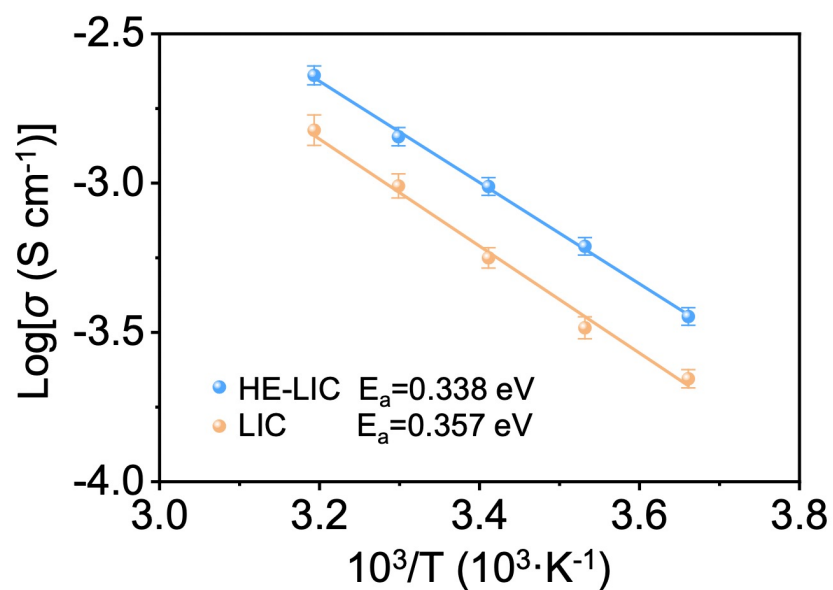
$c/a = 1.0035$ for observed change of slope

Ana F. Suzana et al, Adv. Func. Materials 2208012 (2023)

I. K. Robinson, MS-69, IUCr 2026

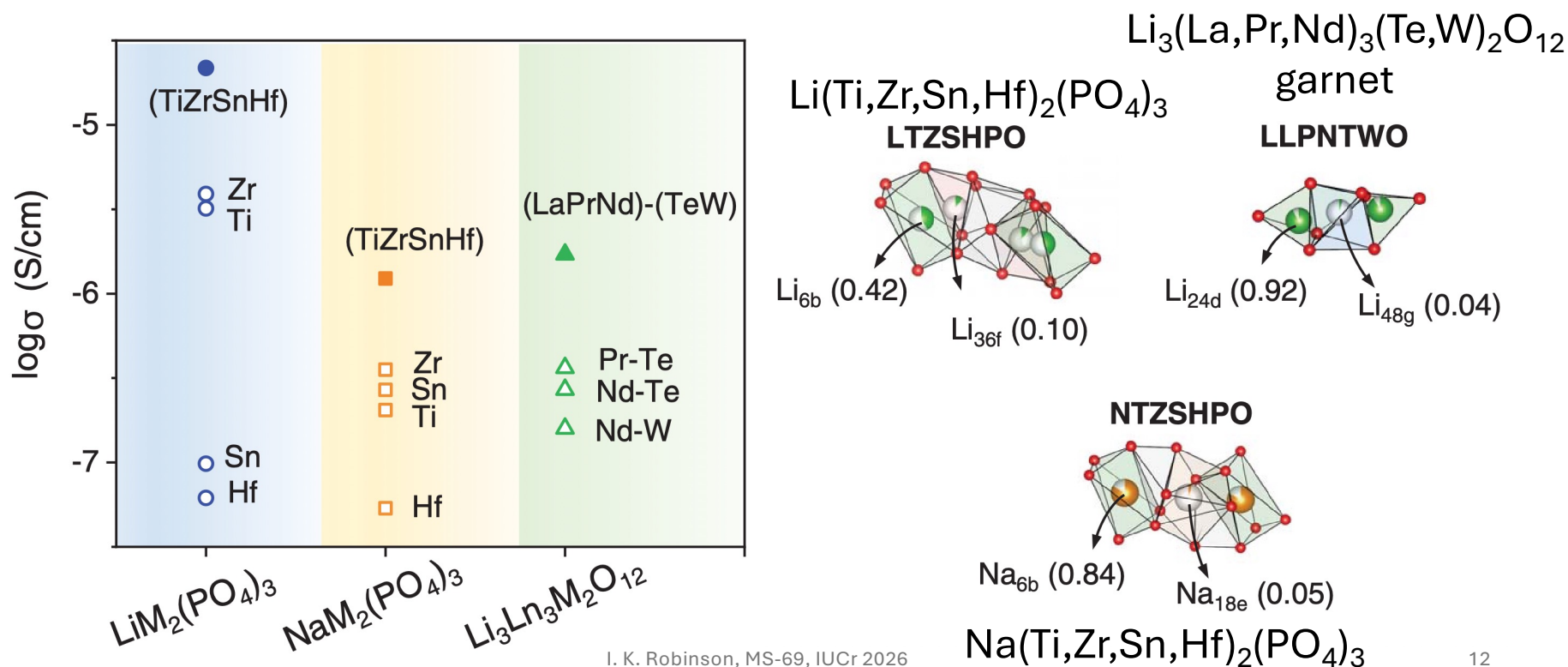
High-Entropy enhancement of halide Li_3InCl_6 SSE

Z. Song., W. Luo, Nat. Comms **15** 1481 (2024)



High-Entropy enhancement of Li-NASICON SSE

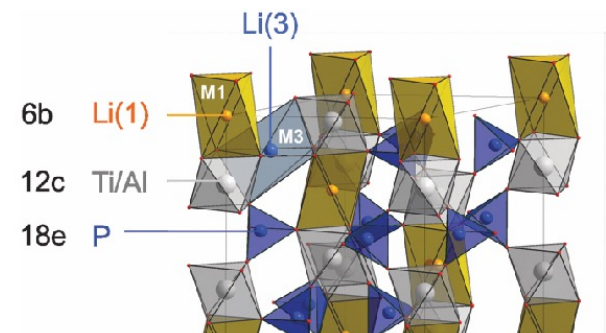
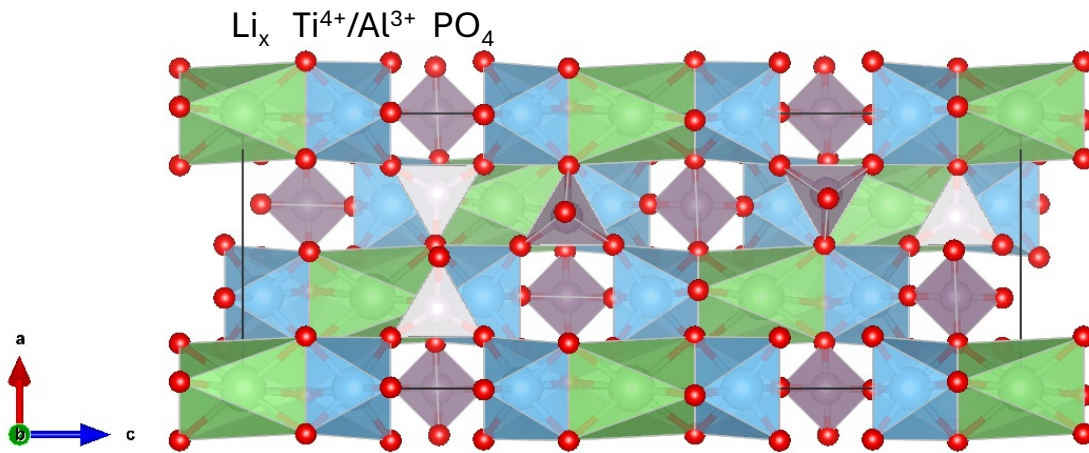
Y. Zeng, et al. Science **378** 1320 (2022)



I. K. Robinson, MS-69, IUCr 2026

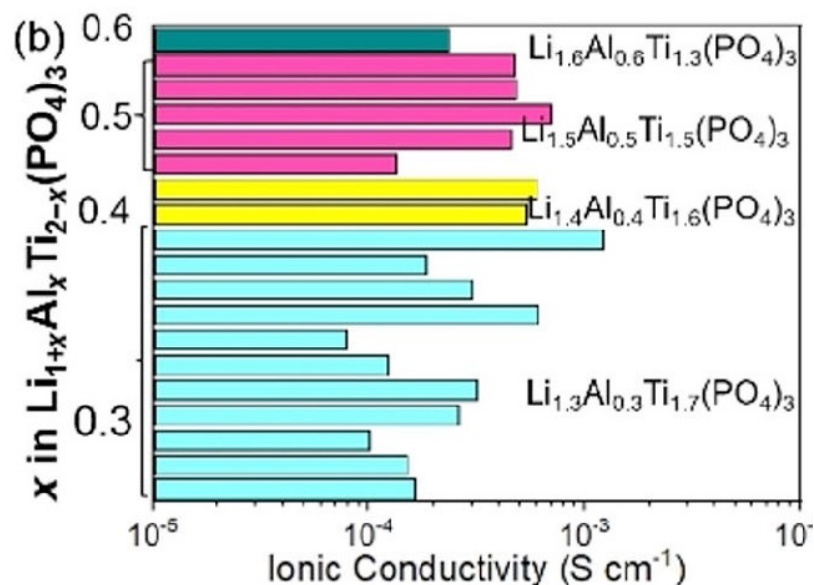
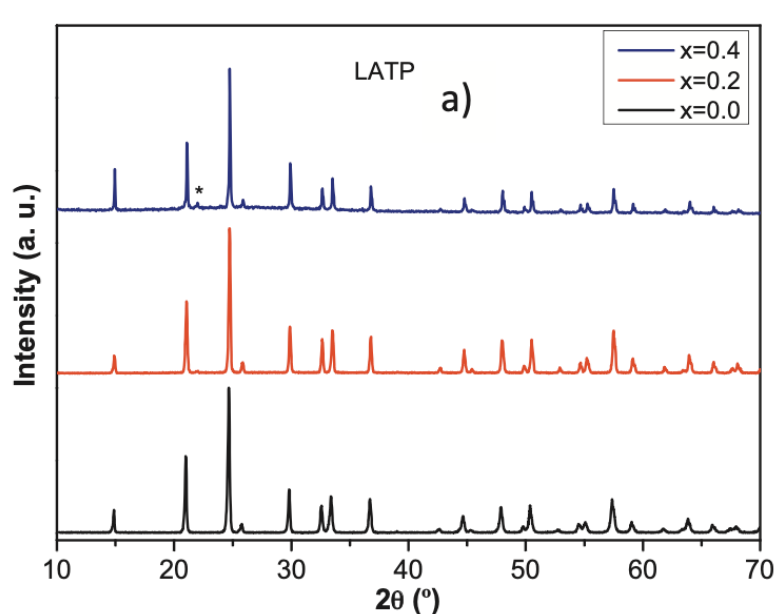
Systematic series with increasing substitution

Low Entropy	$\text{LiTi}_2(\text{PO}_4)_3$	$\sigma = 10^{-6} \text{ S/cm}$
Medium Entropy	$\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$	$\sigma \sim 10^{-4} \text{ S/cm}$
High Entropy	$\text{Li}_3\text{Ca}_{0.5}\text{Ti}_{0.5}\text{Zn}_{0.5}\text{Sn}_{0.5}(\text{PO}_4)_3$	$\sigma \sim 10^{-3} \text{ S/cm}$



V. Epp et al, Phys. Chem. Chem. Phys. **17** 32115 (2015)
“Li stuffing” model

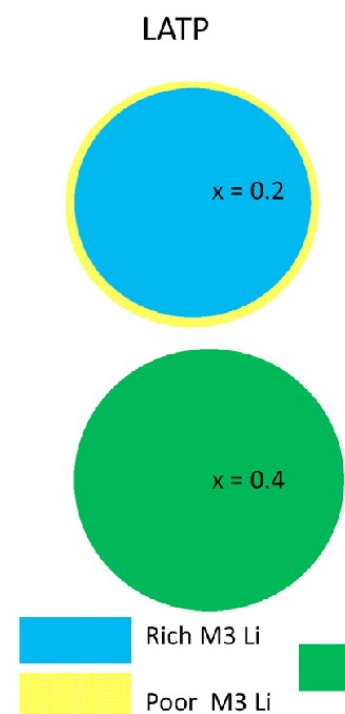
Possible inhomogeneity induced by Al doping in LTP



x	a	c
0	8.5112	20.8479
0.2	8.5020	20.8180
0.4	8.4975	20.7921

R. DeWees, H. Wang, Chem. Sus. Chem. 12 3713 (2019)

K. Arbi, et al, J. Eur. Ceram. Soc. 35, 1477 (2015)



BCDI test of role of chemical diversity in Li ion conduction

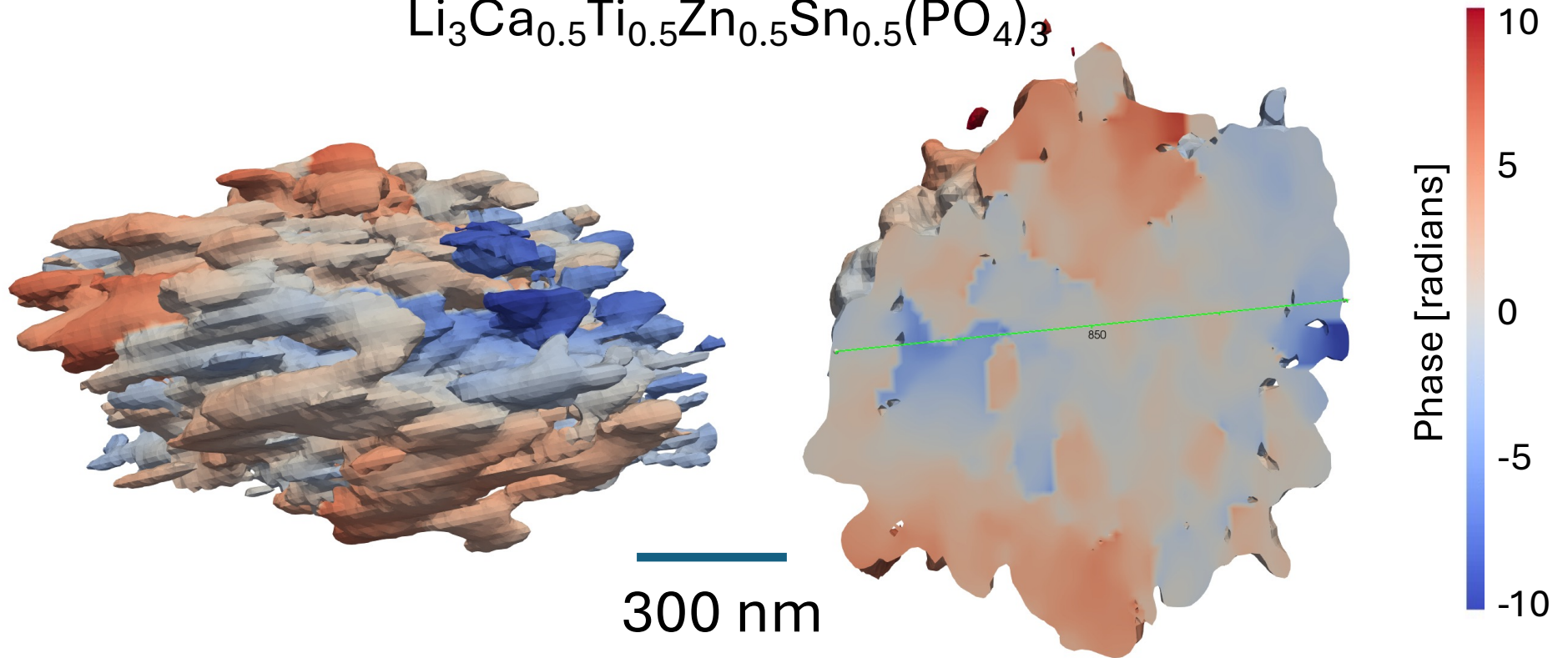
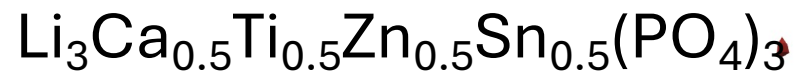
Sample preparation:

- Solid State synthesis by firing of stoichiometric mixtures.
- Ball milling
- Pressing into pellets
- Sintering (optional)

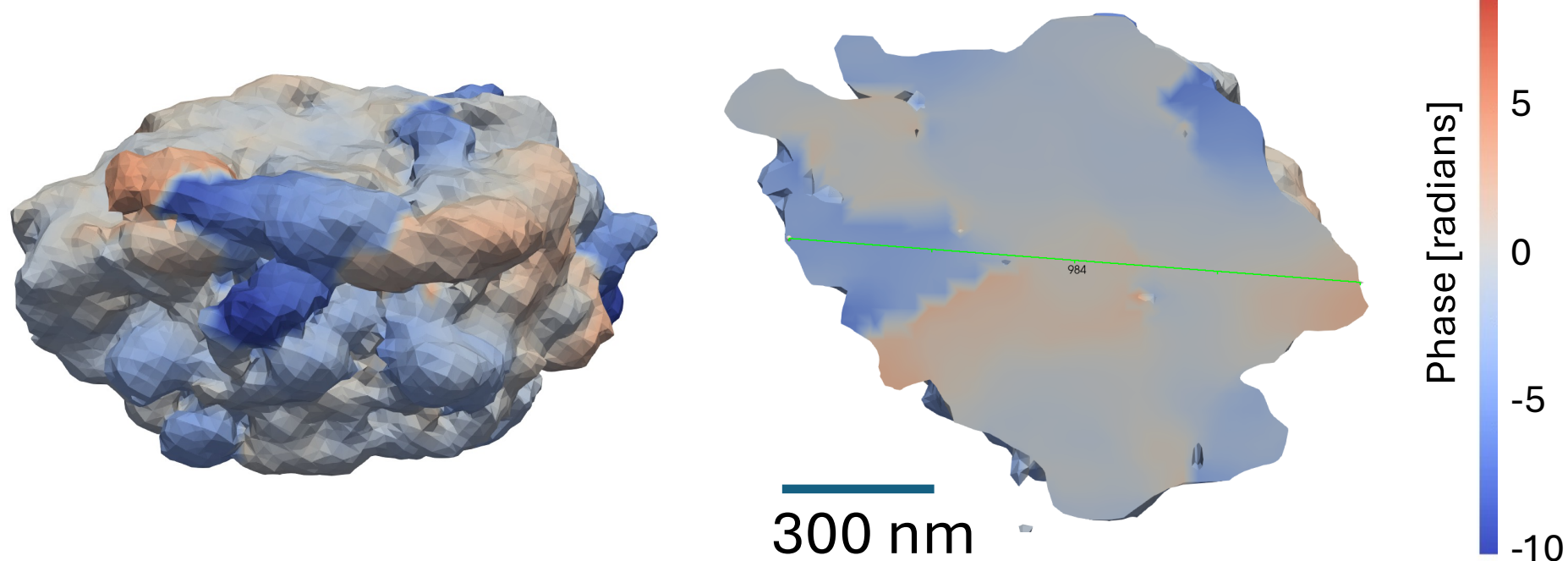
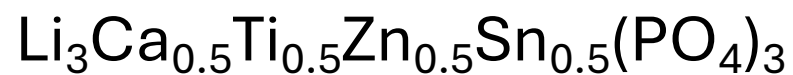
Possible mechanisms:

- Full thermodynamic equilibrium
- Random site occupation
- Surface segregation
- Local demixing/dealloying
- Second phase formation
- Pore formation
- Percolation pathways
- Localization at crystal defects

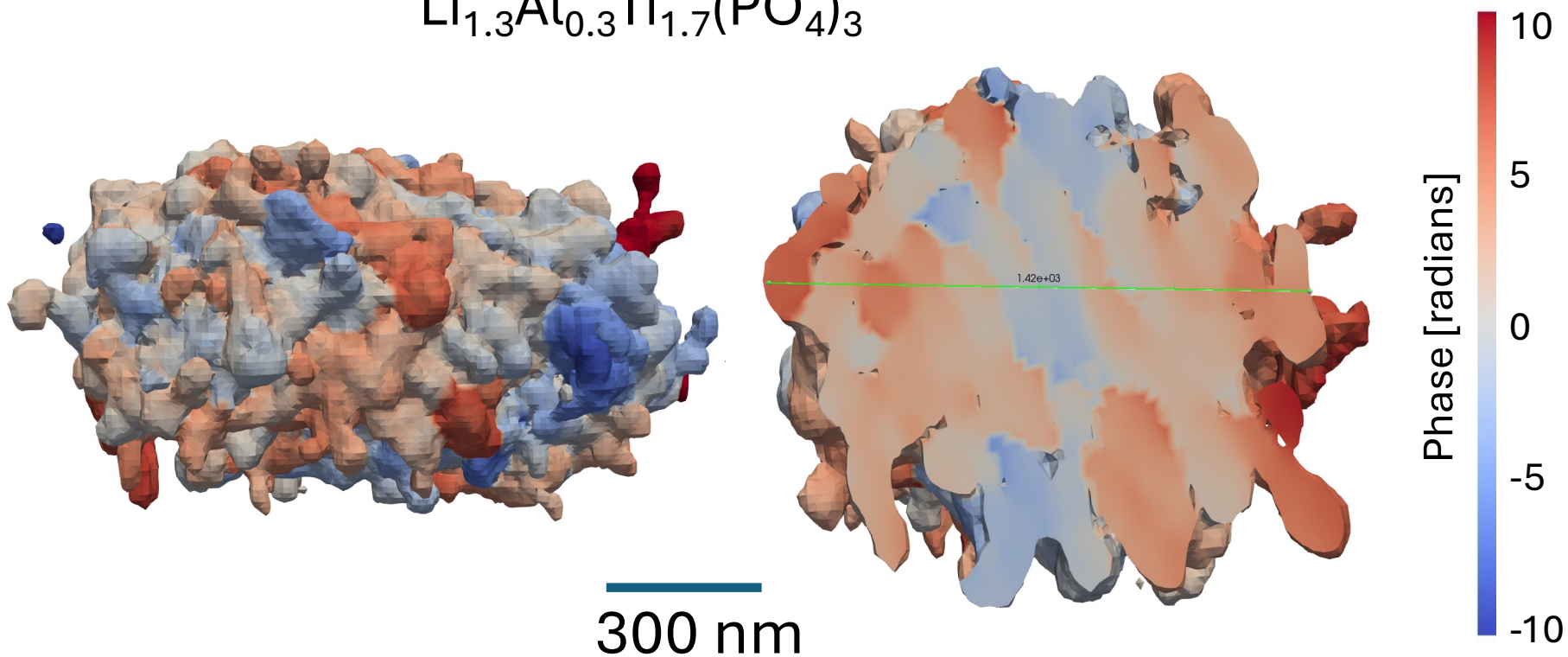
CM3-68F High-Entropy $\text{Li}_3\text{Ti}_2(\text{PO}_4)_3$ SSE



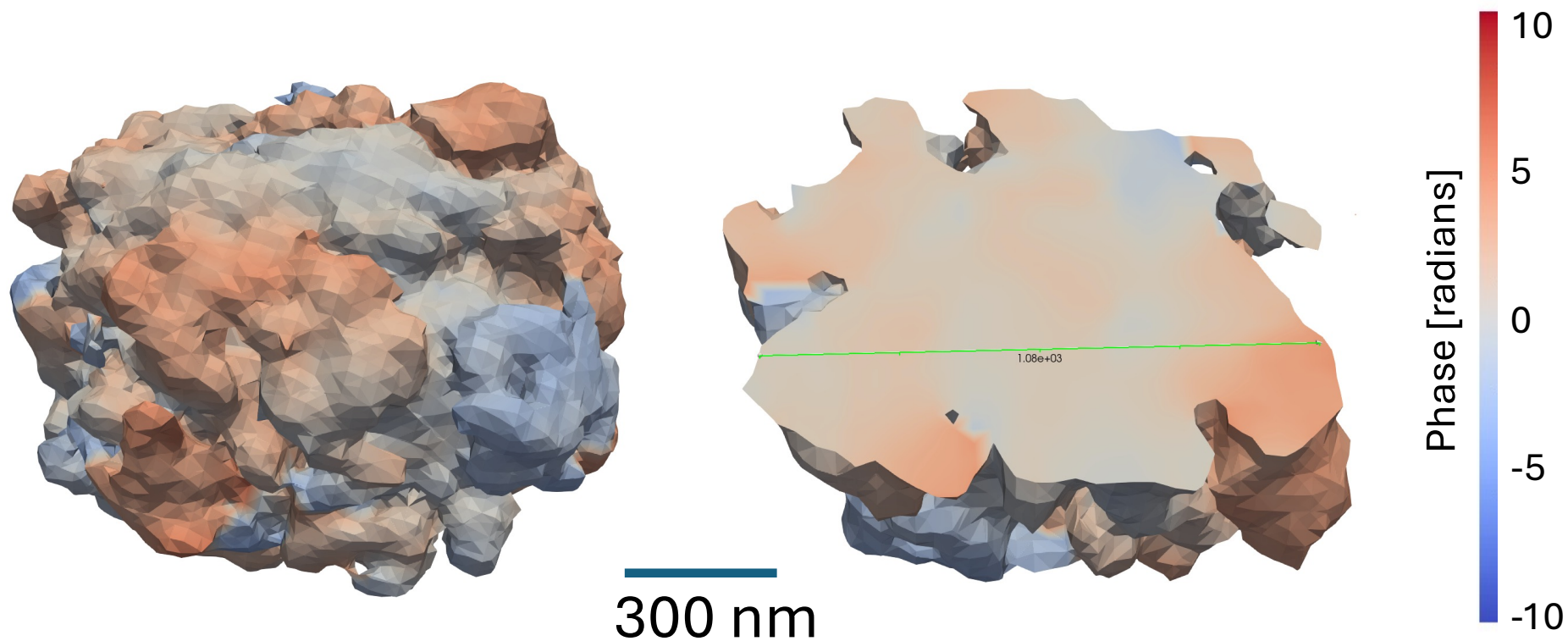
CM3-41 High-Entropy $\text{Li}_3\text{Ti}_2(\text{PO}_4)_3$ SSE



CM2 Medium-Entropy $\text{Li}_3\text{Ti}_2(\text{PO}_4)_3$ SSE

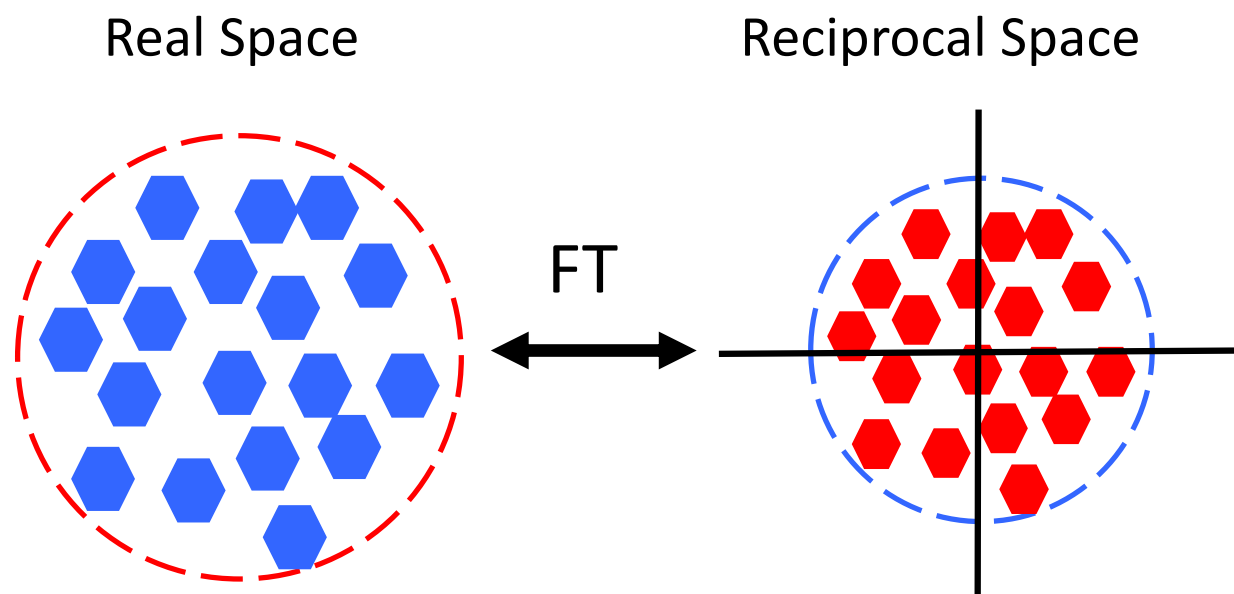


CM1-221 Low-Entropy $\text{Li}_3\text{Ti}_2(\text{PO}_4)_3$ SSE



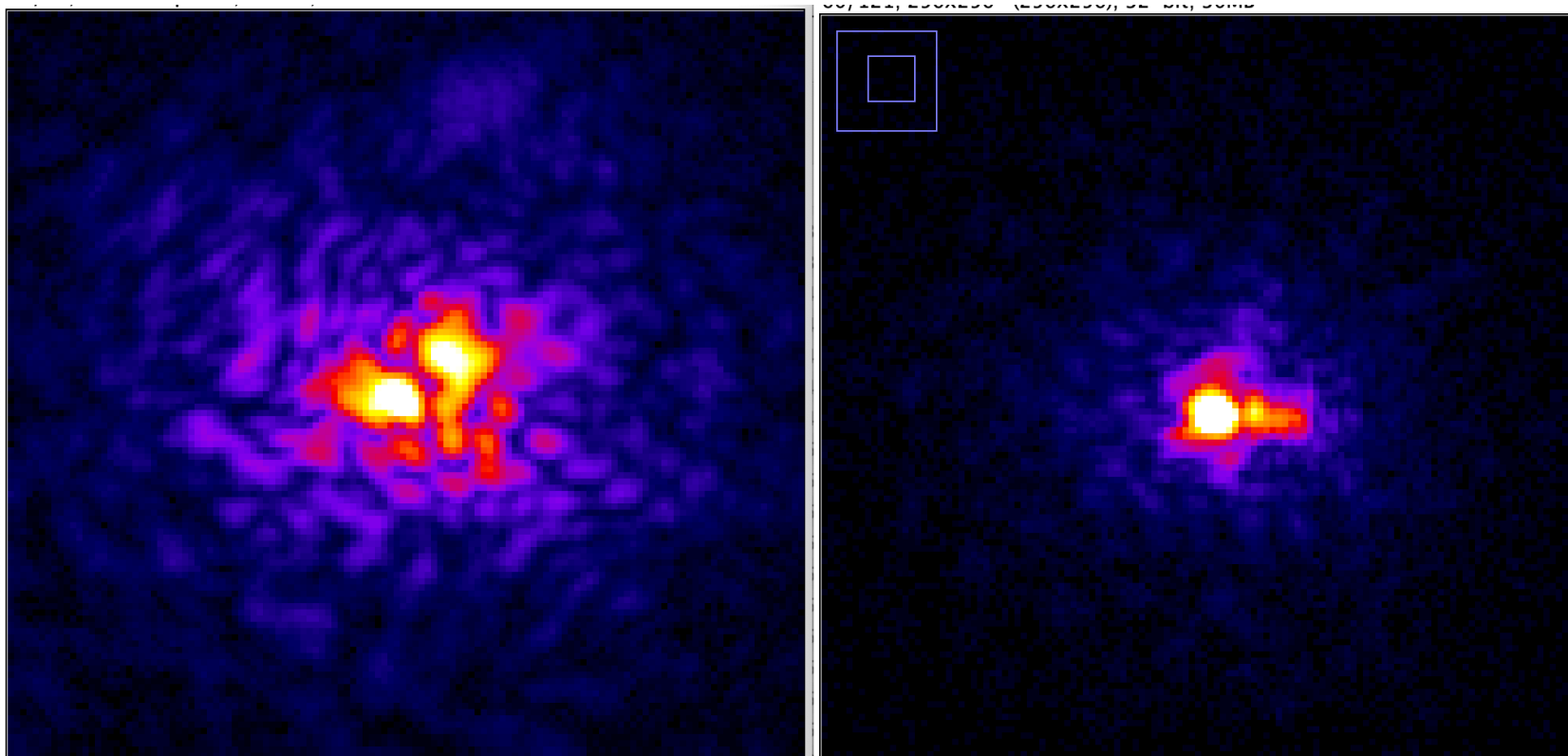
Domain Counting in Coherent Diffraction

Ian Robinson et al, J. Supercond. and Novel Magnetism **33** 99 (2020)

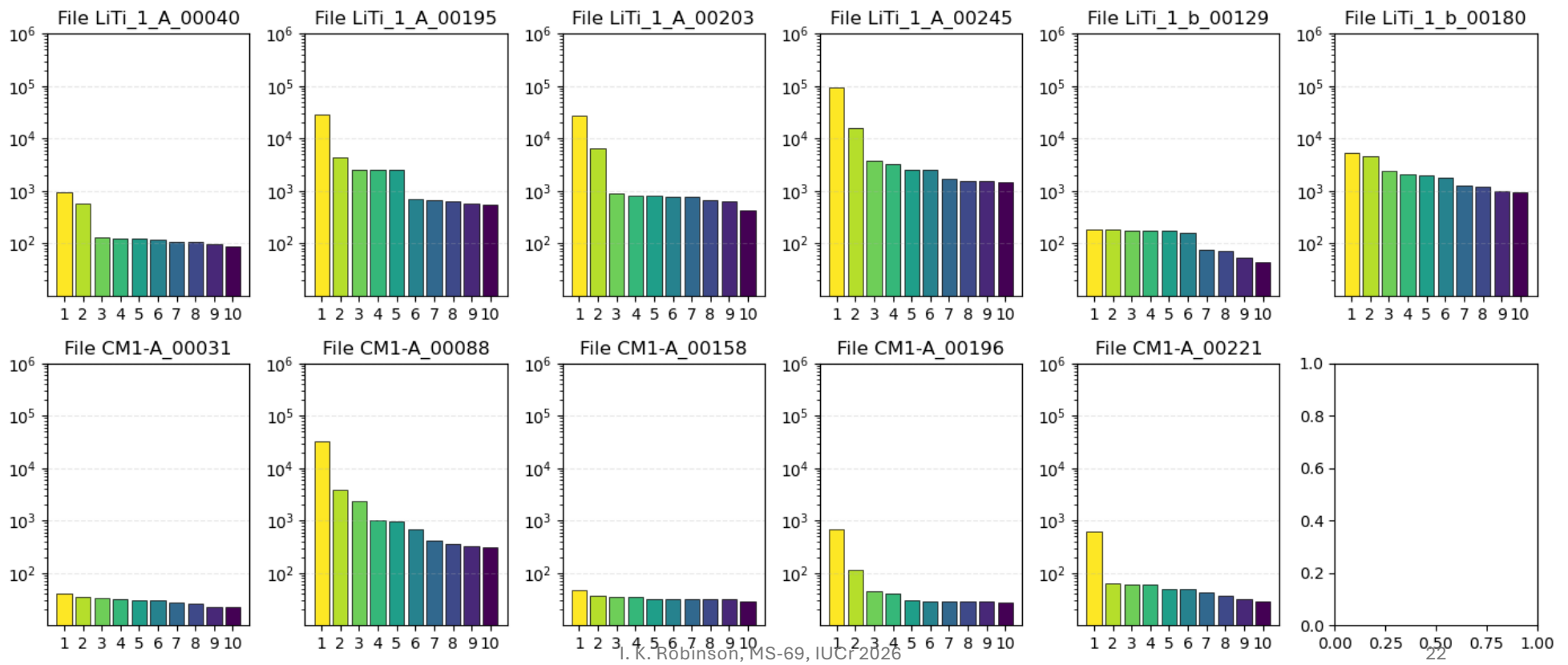


Compare High and Low Diversity Diffraction

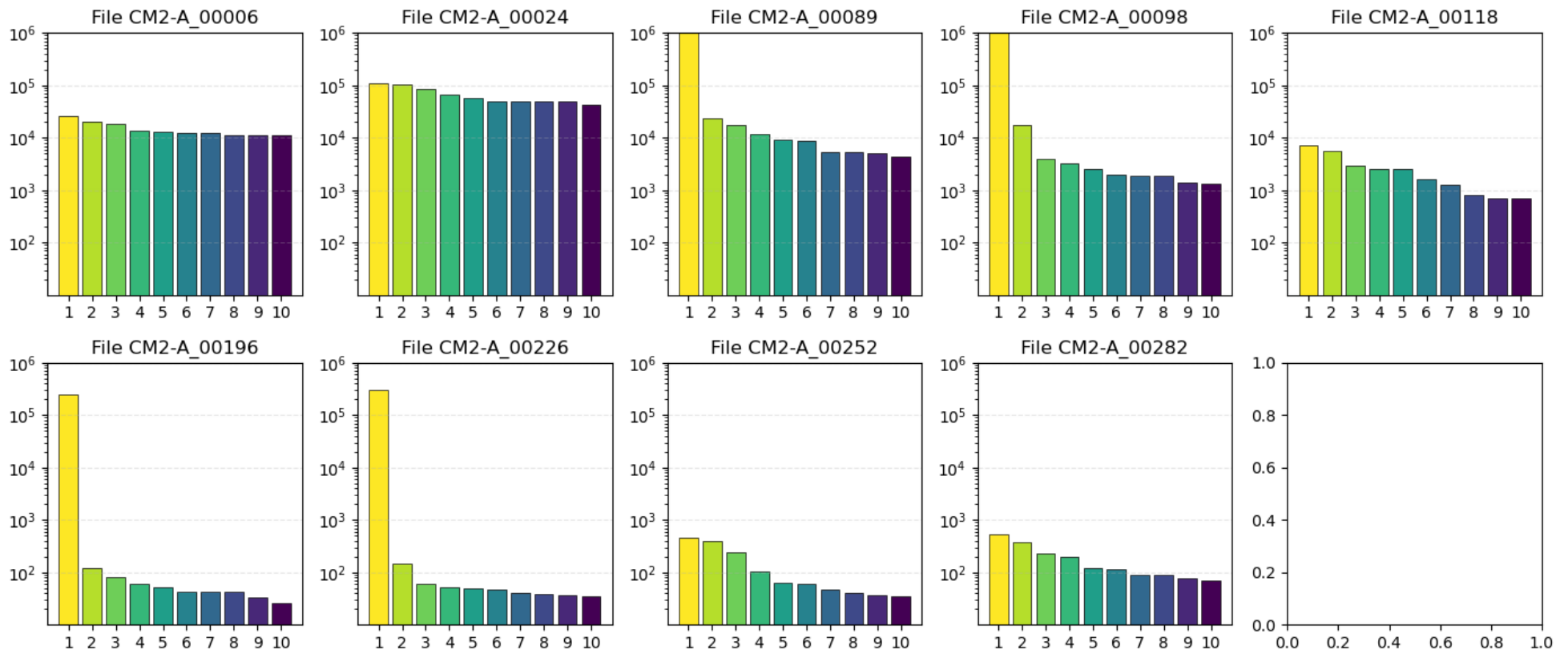
$\text{Li}_3\text{Ca}_{0.5}\text{Ti}_{0.5}\text{Zn}_{0.5}\text{Sn}_{0.5}(\text{PO}_4)_3$ and $\text{LiTi}_2(\text{PO}_4)_3$



CM1 (low diversity) speckle peak intensity histograms

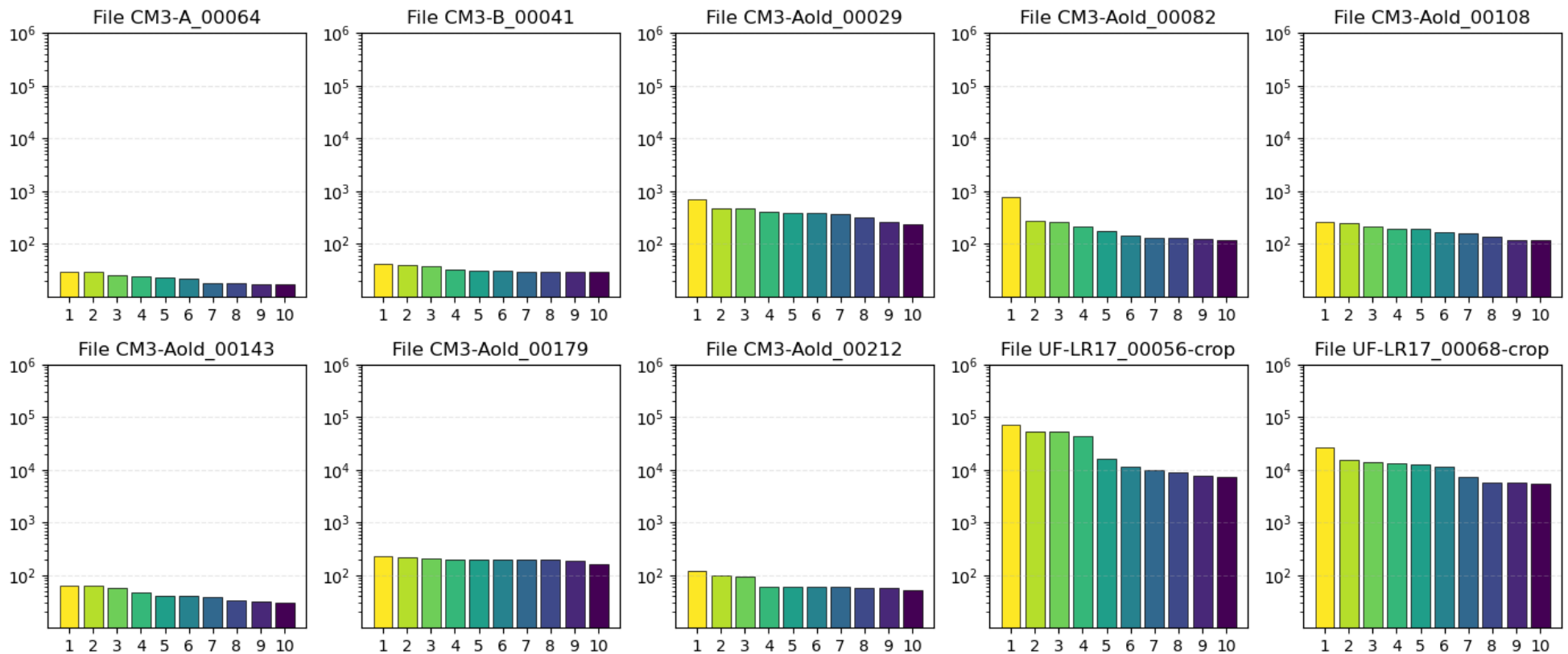


CM2 (medium diversity) speckle peak intensity histograms



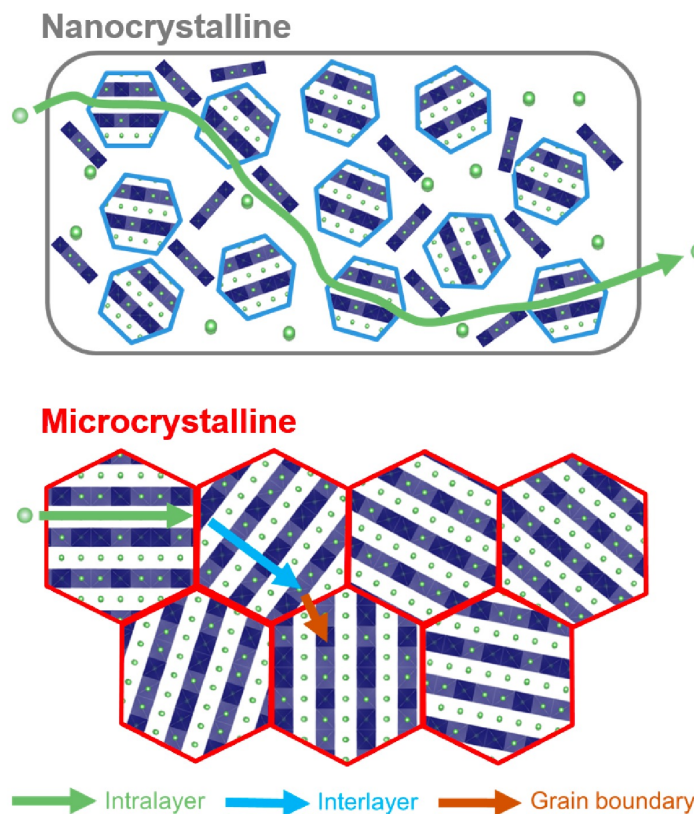
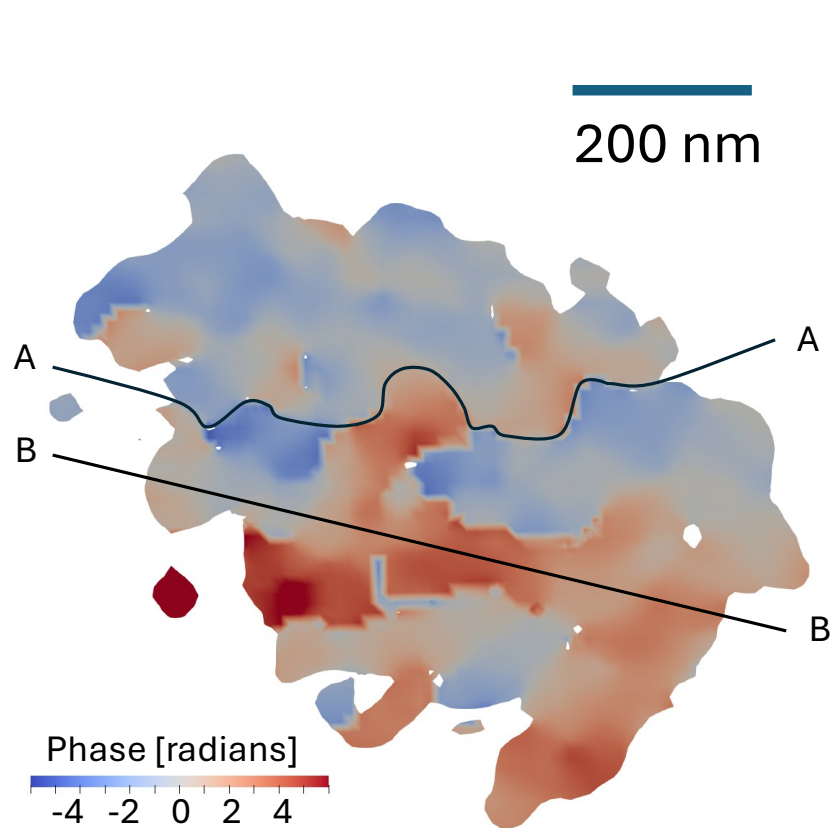
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CM3 (high diversity) speckle peak intensity histograms



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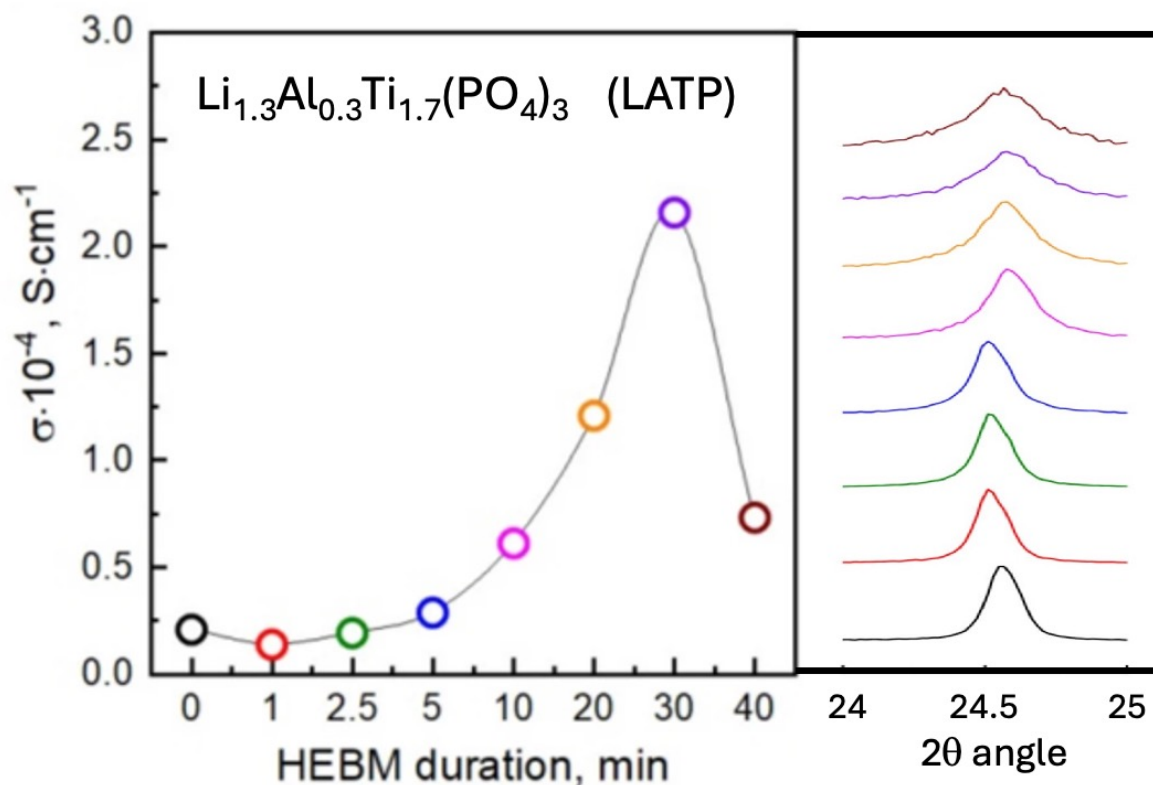
Domain-boundary conduction pathway



$\text{Li}_{3(1+x)}\text{AlP}_2$ J. Hu, et al J. Mater. Chem. A (2025)
I. K. Robinson, MS-69, IUCr 2026

Conductivity and XRD profile of LATP

113 Bragg peak as a function of high-energy ball-milling (HEBM) time
A.A. Shindrov et al, Ionics **31** 1351–1360 (2025)



I. K. Robinson, MS-69, IUCr 2026

Coherent X-ray Diffraction Imaging of SSE Domains

- Bragg Coherent Diffraction Imaging (BCDI)
- Sensitivity to strain
- Phase domain structures
- Domain counting rule
- Solid State Electrolyte Crystals
- High Entropy conduction
- Cross sectional phase images
- Phase-domain-wall conduction pathway
- Possible formation role of ball milling

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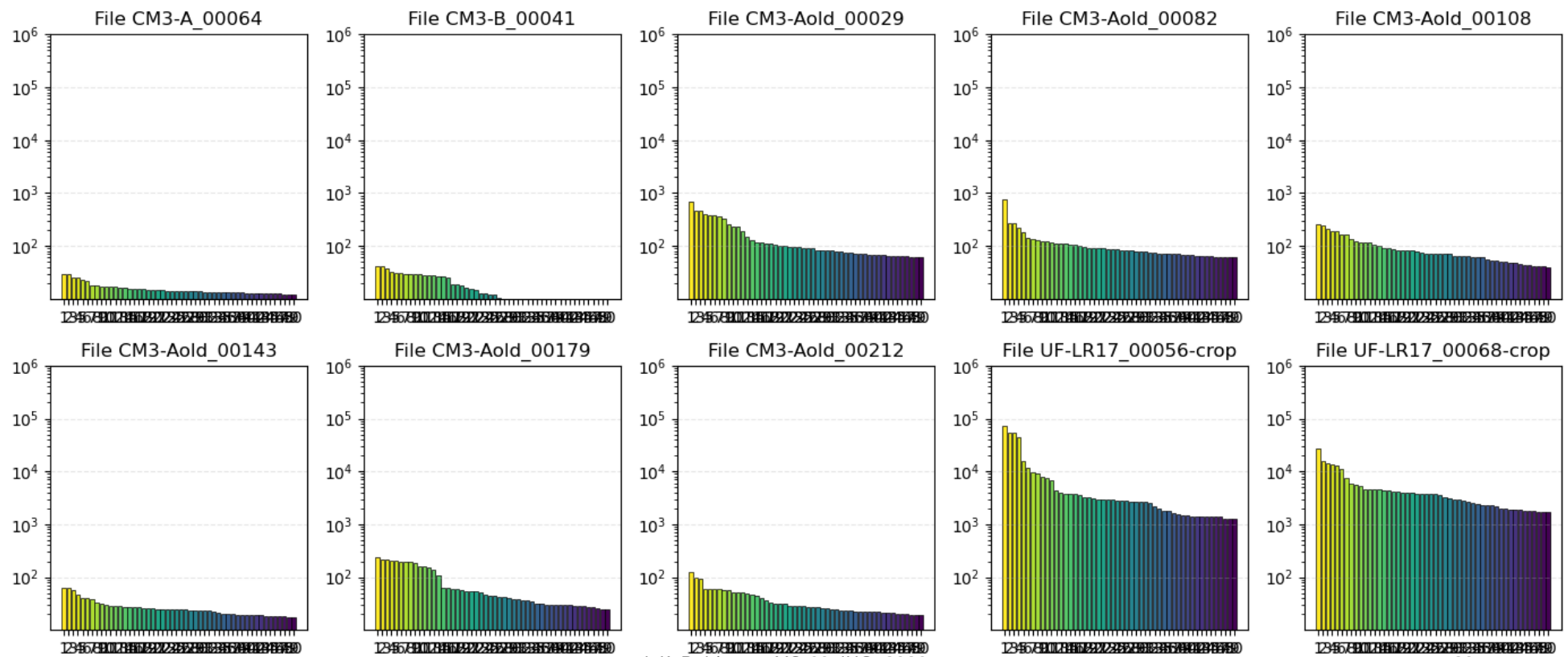


Fabian Westermeier
Wojciech Roseker



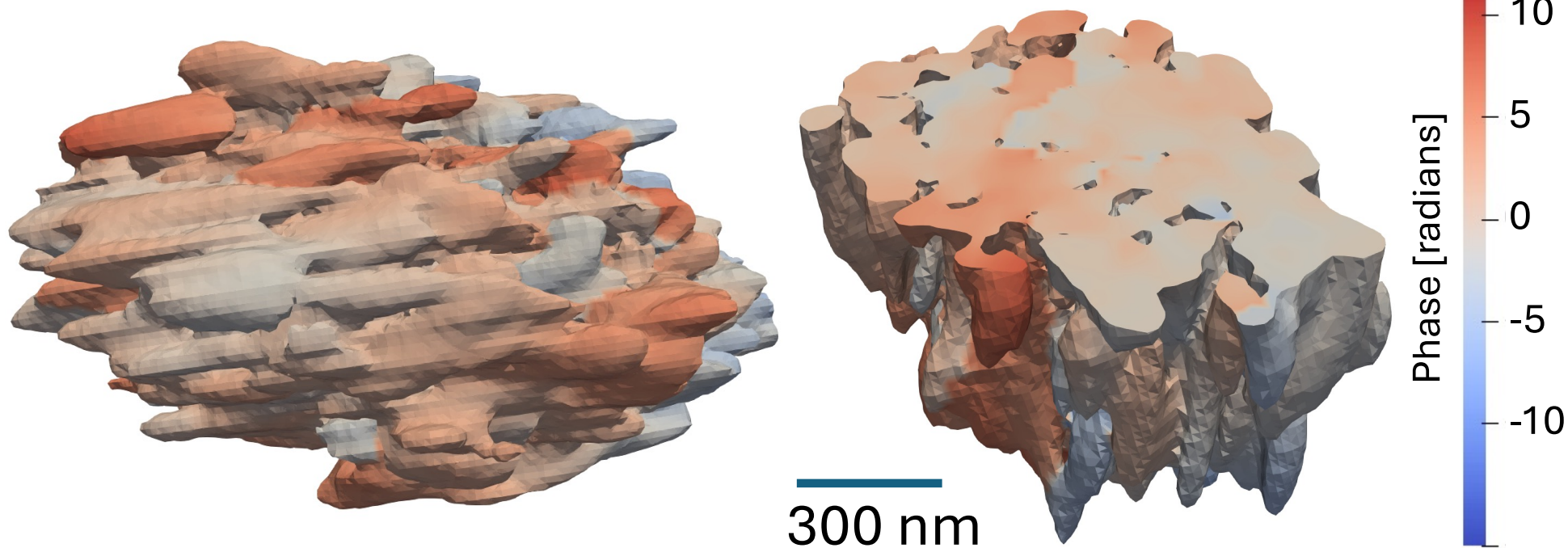
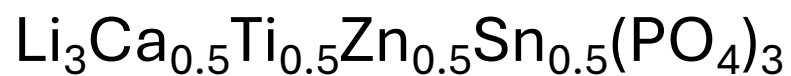
Dina Sheyfer
LuXi Li

CM3 (3-2A) speckle peak intensity histograms



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CM3-68 High-Entropy $\text{Li}_3\text{Ti}_2(\text{PO}_4)_3$ SSE



CM3-68 High-Entropy $\text{Li}_3\text{Ti}_2(\text{PO}_4)_3$ SSE

