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Phase Transition Mechanisms in Cadmium Sulfide from X-ray Diffraction Comparisons of High-pressure Experiments and MD Simulation

J. Matthew D. Lane, Bryce Thurston, Tommy Ao, David Montes de Oca Zapiain, Mark Rodriguez, Marcus Knudson

Sandia National Laboratories, Albuquerque, NM

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APS March Meeting, Chicago, IL

U.S. DEPARTMENT OF ENERGY

NNSA

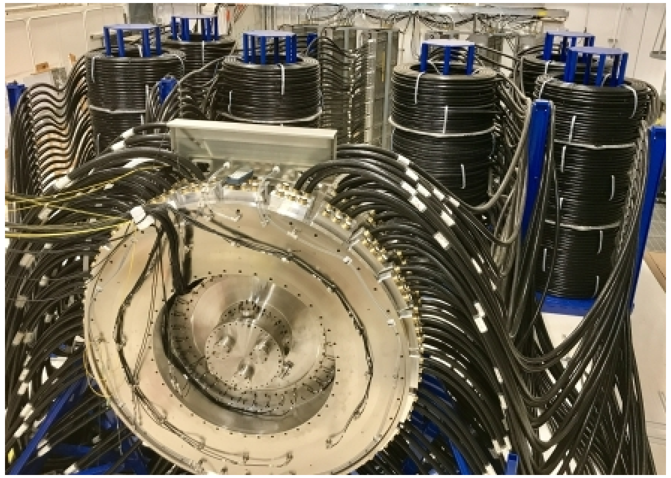
Motivation: Dynamic compression experiments

Thor and Z are pulsed-power accelerators which can drive shockless ramp waves to pressures of 10s and 100s of GPa, respectively.

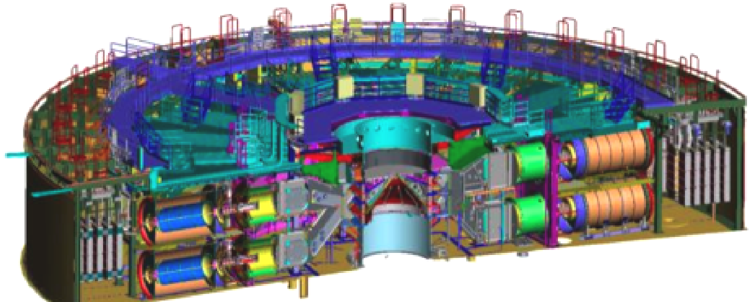
Pulsed-power drivers translate stored electrical energy into magnetically (Lorentz force) driven compression on nanosecond timescales with incredibly controlled pulse-shaping.

- Adding X-ray diffraction diagnostic to existing velocimetry (VISAR & PDV)
- Emphasis on characterizing of phase transitions, phase transition kinetics and strength mechanisms

Thor pulsed-power driver



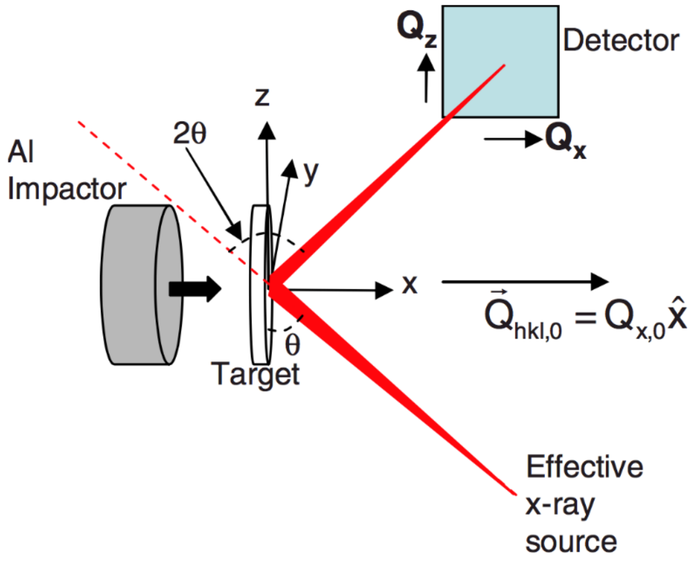
Z-machine at Sandia National Labs



33 m in diameter, 3 stories tall

22 MJ stored energy
25 MA peak current
100-600 ns rise time

X-ray diffraction geometry



Challenges for in-situ 2D XRD

Capturing diffraction data of transient compressed states

X-ray source

- Collimation (diode point source)
- Relatively broad X-ray spectra (x-ray diode)
- Reflection geometry with 20-50 um penetration

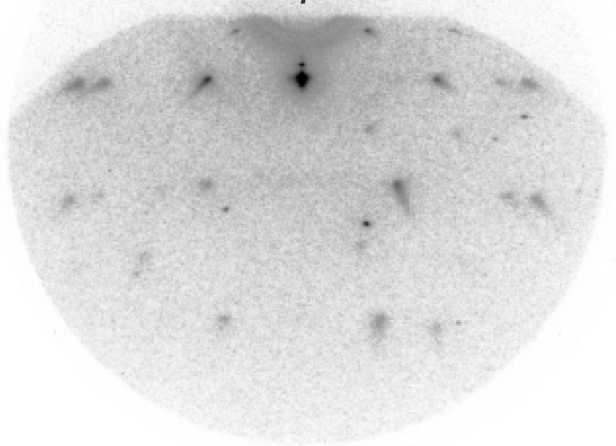
Dynamic response

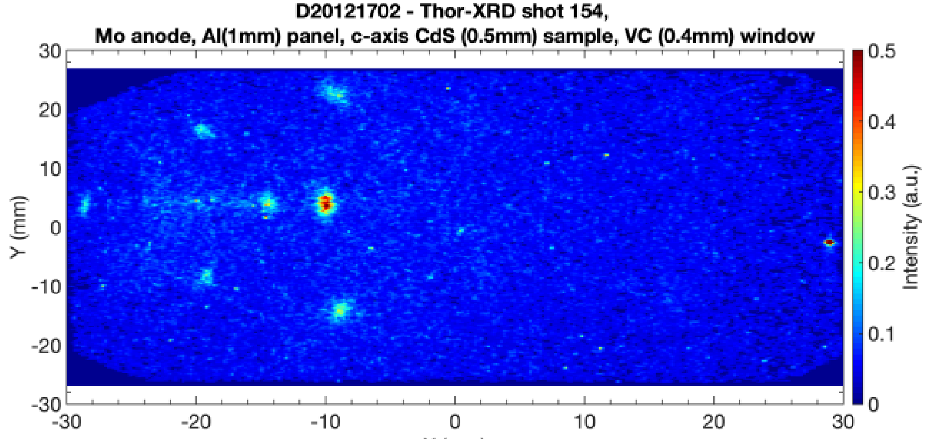
- **Data sparse**
- Non-spatial uniformity: compression, rotation
- Time evolution
- Mixed states, incomplete transformation

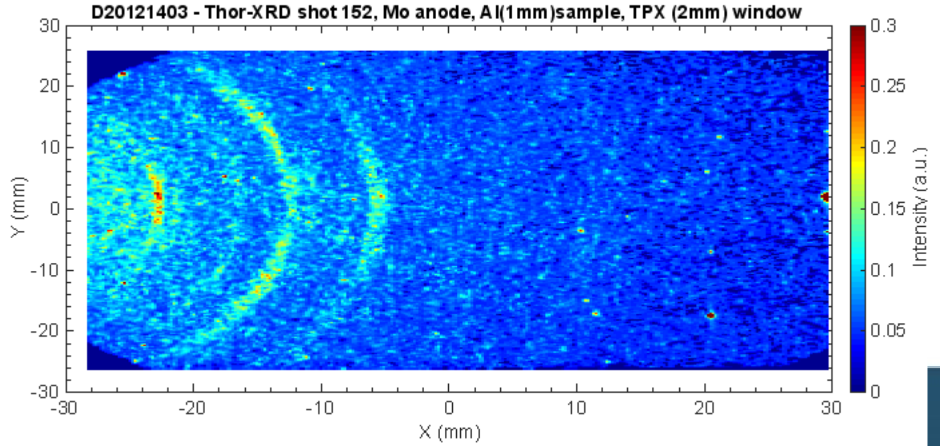
Background/Noise

- Window/tamper
- Machine produced x-ray background

18-4-063 – DCS c-axis 1.683 km/s impact

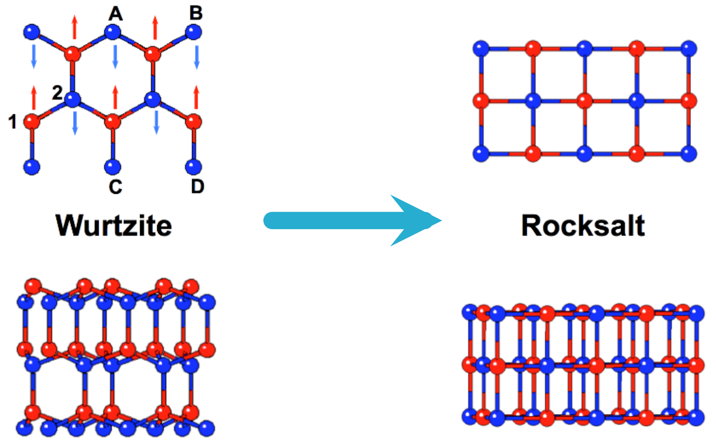






Phase transformation kinetics in cadmium sulfide (CdS)

Pressure-induced Cadmium Sulfide (CdS) phase transition from Wurtzite to Rocksalt at ~2-3 GPa



The transition is delayed near the onset pressure.

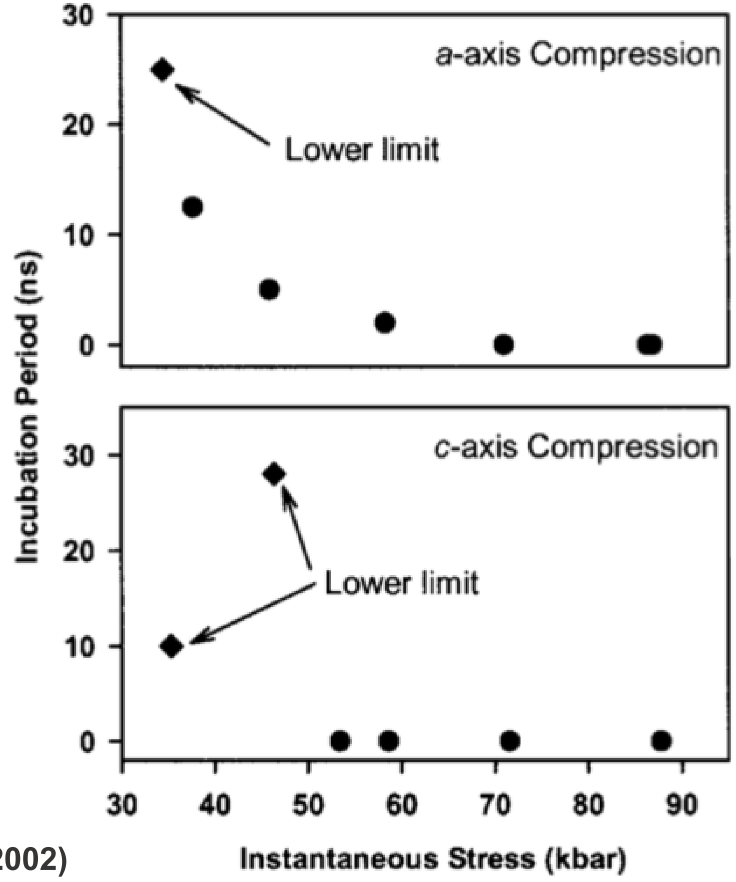
Transition kinetics depend strongly on the crystal orientation

Measurements of the two-step transition, with fast, time-resolved spectroscopy showing the first step is sub-ns

Knudson, Gupta, Kunz. Phys. Rev. B, 59, 11704 (1999)
Knudson, Gupta. Phys. Rev. Lett., 81, 2938 (1998)

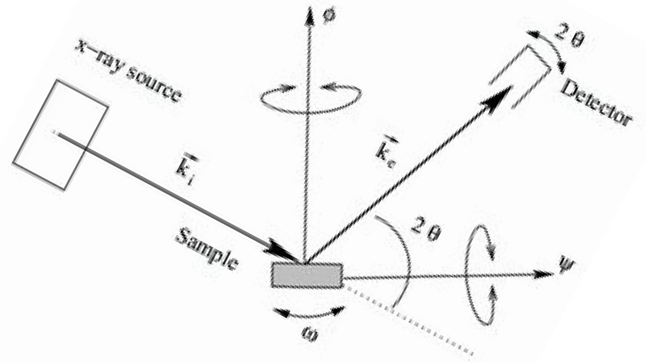
Inelastic deformation is observed before the onset of the phase transition which may influence the mechanism

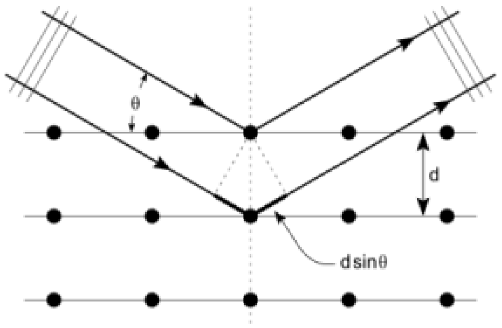
Second step in transition is delayed depending on peak stress



Knudson, Gupta, J. Appl. Phys. 91, 9561 (2002)

Simulated diffraction patterns from LAMMPS





Bragg condition: $\boldsymbol{K} = \boldsymbol{K}_B$

$$\frac{\sin(\theta)}{\lambda} = \frac{|\boldsymbol{K}|}{2} \qquad \frac{1}{d_{hkl}} = |\boldsymbol{K}_B|$$

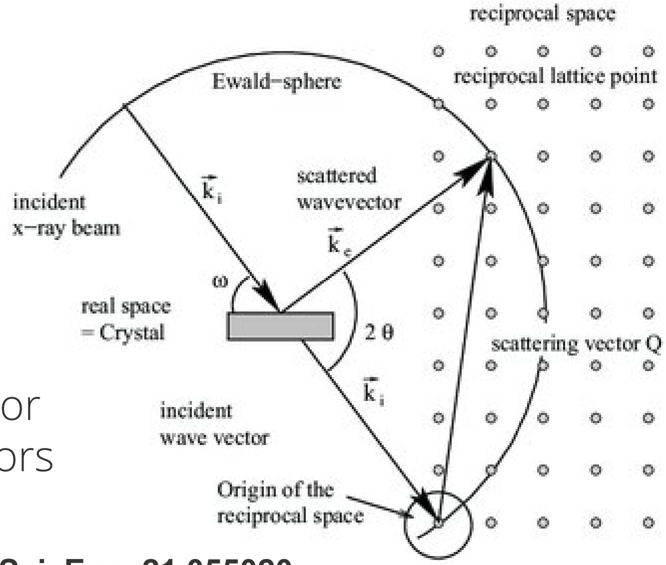
K is the wave vector difference between incident and the scattered X-ray, $K = k_e - k_i$.

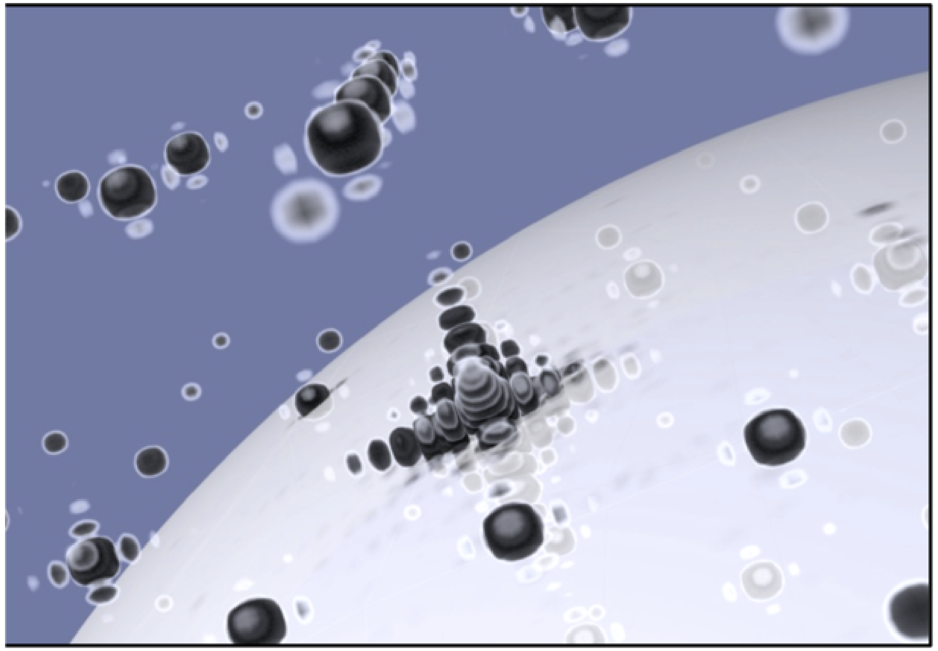
Constructing the Reciprocal space lattice in LAMMPS

$$F(\boldsymbol{K}) = \sum_{j=1}^{\#atoms} f_j(\theta) \exp\left(2\pi i \boldsymbol{K} \cdot \boldsymbol{r}_j\right)$$

$$I_x(\boldsymbol{K}) = Lp(\theta) \frac{F(\boldsymbol{K}) F^*(\boldsymbol{K})}{N}$$

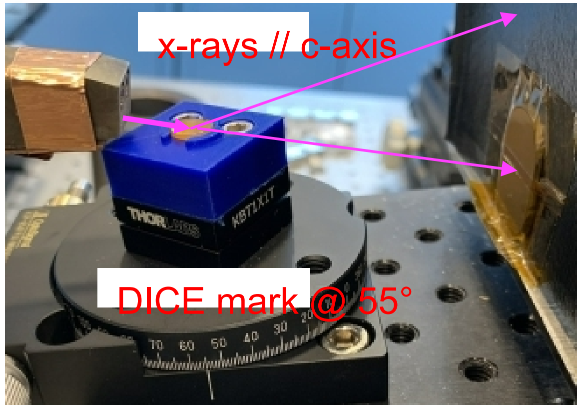
$Lp(\theta)$ is the Lorentz-polarization factor
And f_j are the atomic scattering factors





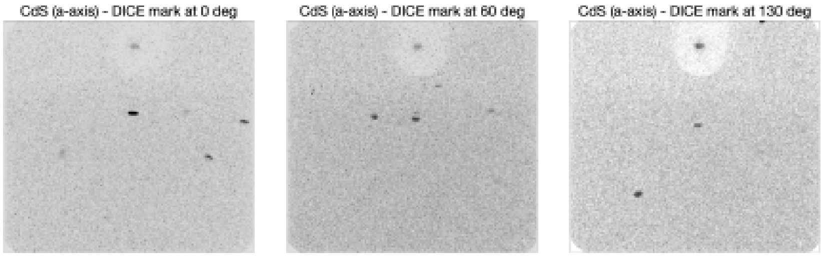
Coleman et al., 2013 Modelling Simul. Mater. Sci. Eng. 21 055020

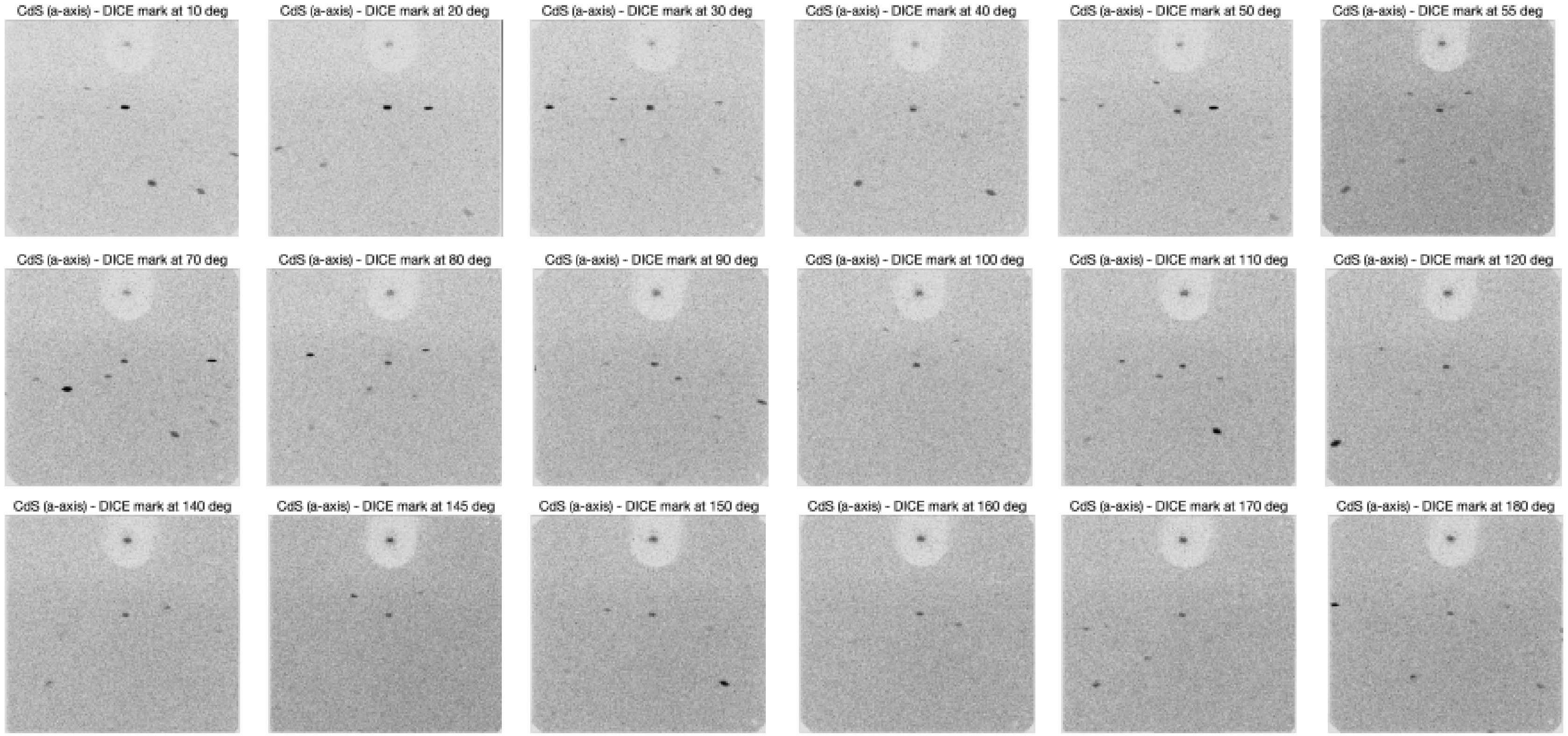
Simulations comparable to Experiments

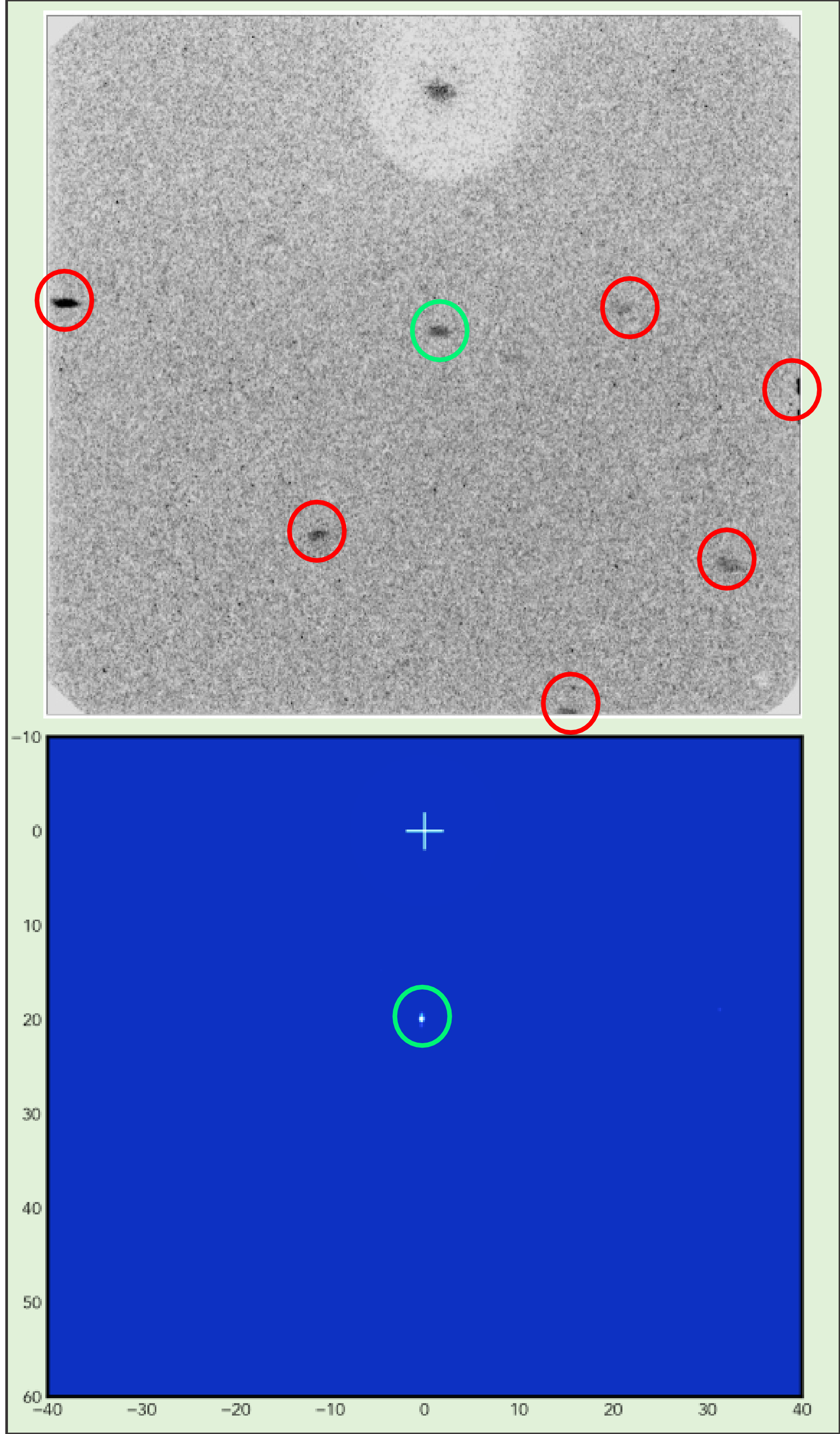


Thor diode data: Tom Ao, Dane Morgan

Ambient 180 rotation of CdS with a Molybdenum 17.6 keV k-α line source



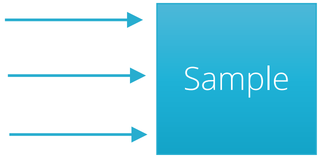


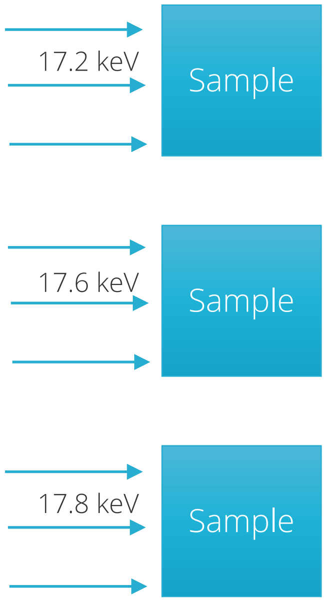


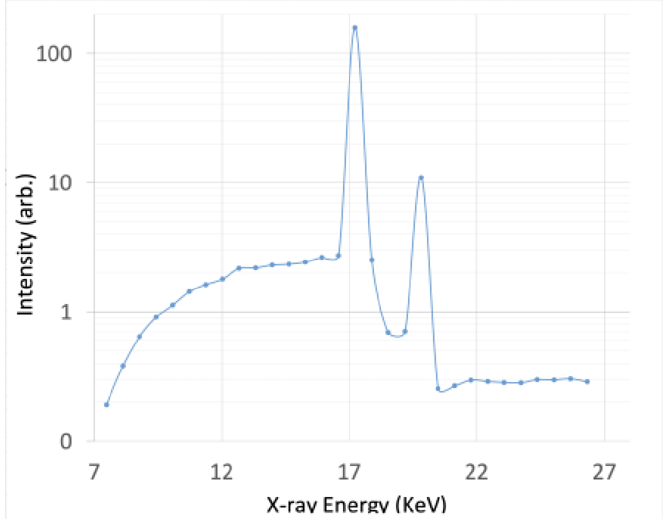
Simulations comparable to Experiments

Broad spectrum (polychromatic source) *characterizations for each source: Molybdenum, Tungsten, Syncrotron (DCS)*

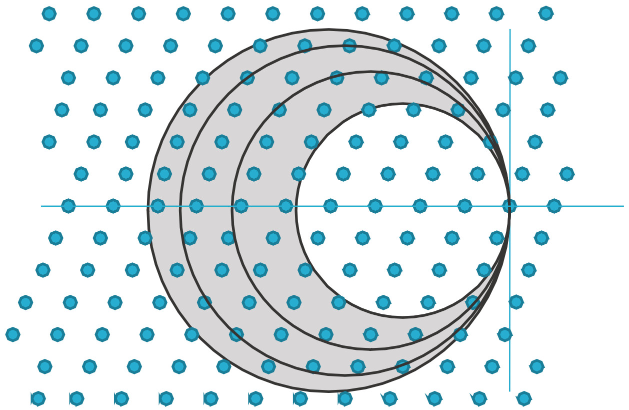
X-ray spectrum is source and filter dependent



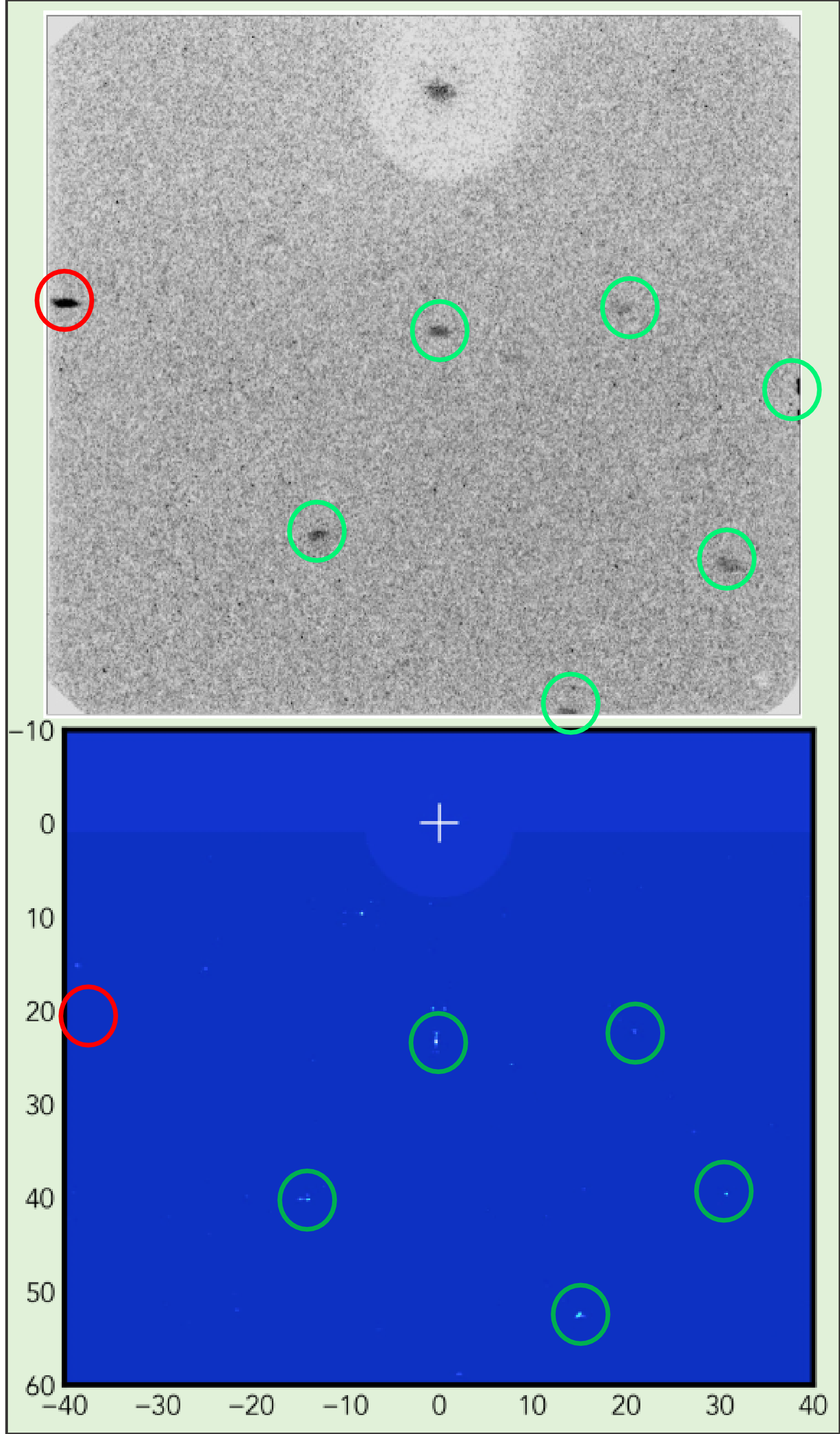




Generalization of Ewald Sphere



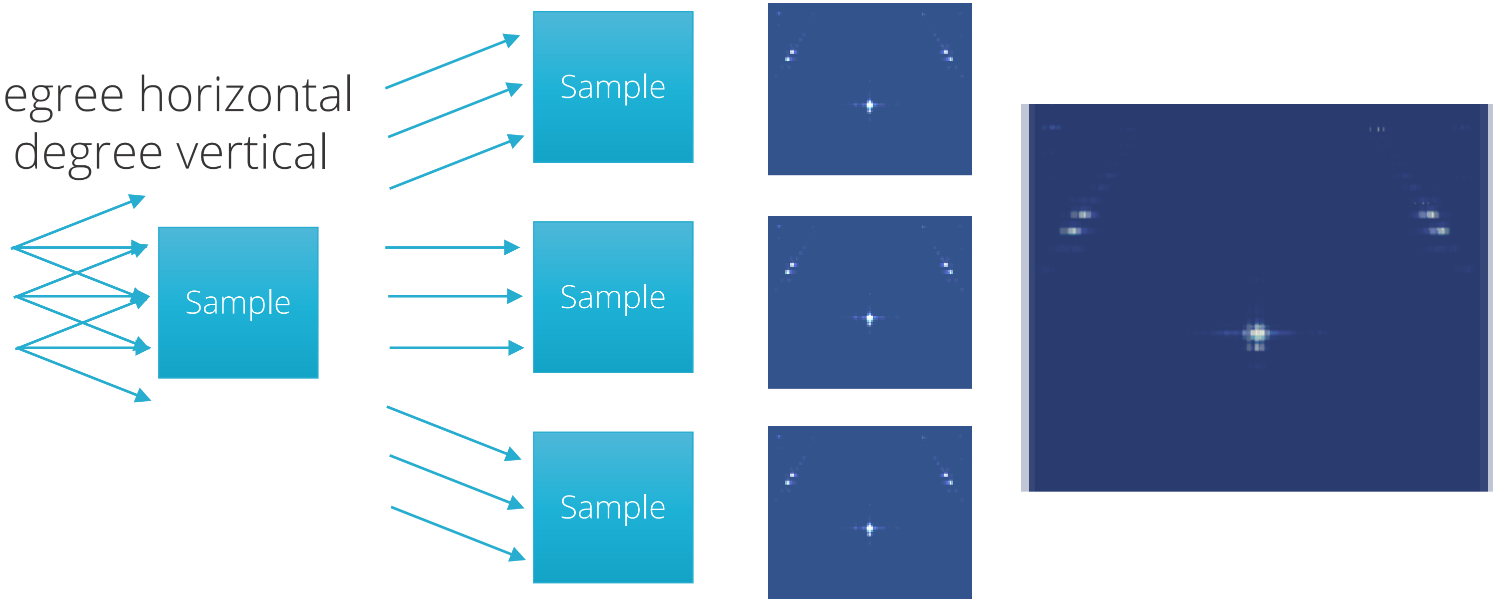
The spectral sum is constructed by numerical integration of the wavelength contributions from, Kα, Kβ and Brems. This is completely generalizable to any source spectrum.

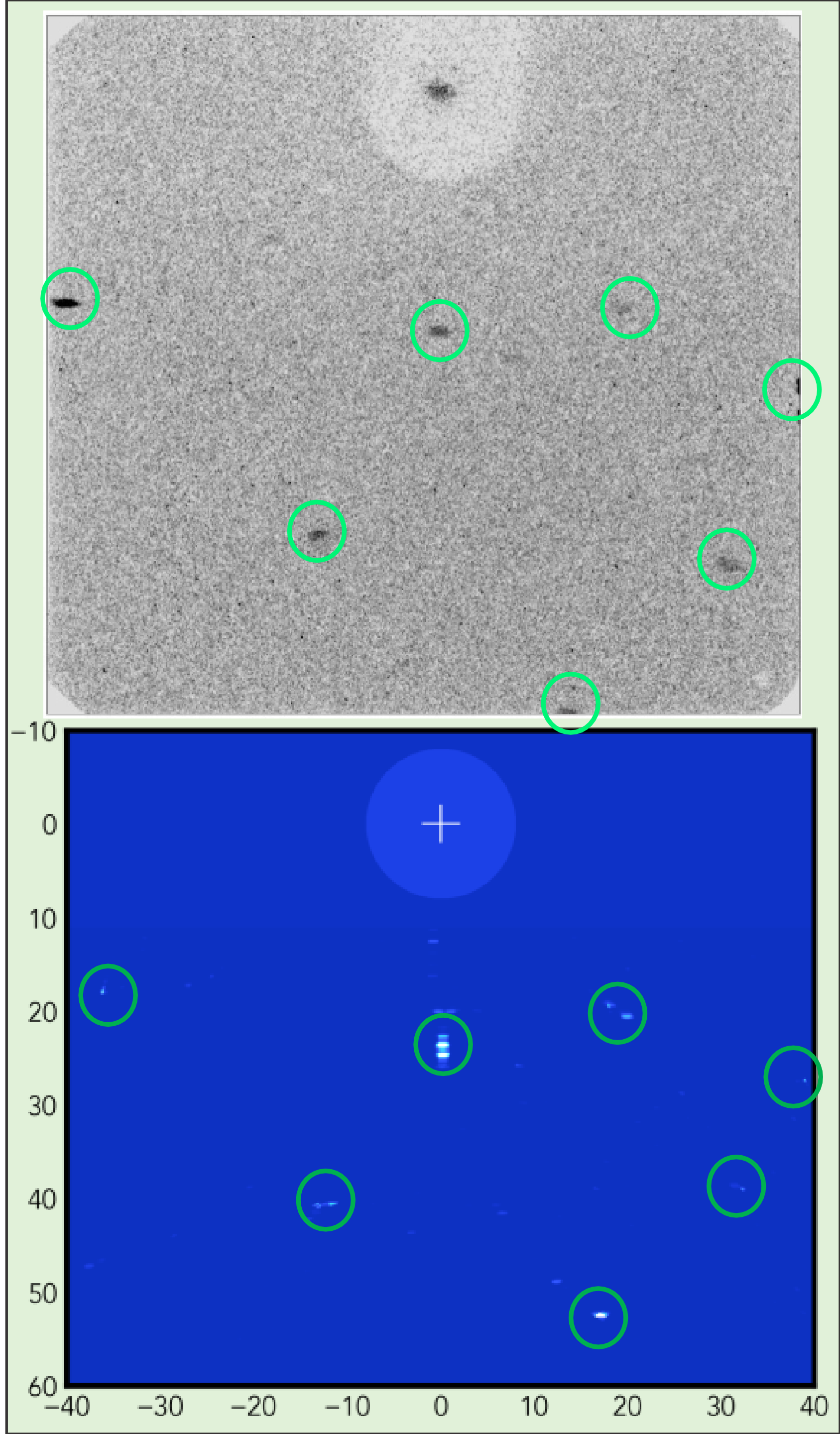


Simulations comparable to Experiments

Angular spread (imperfect collimation) *measured for experimental source and summed from individual orientations*

1 degree horizontal
0.5 degree vertical

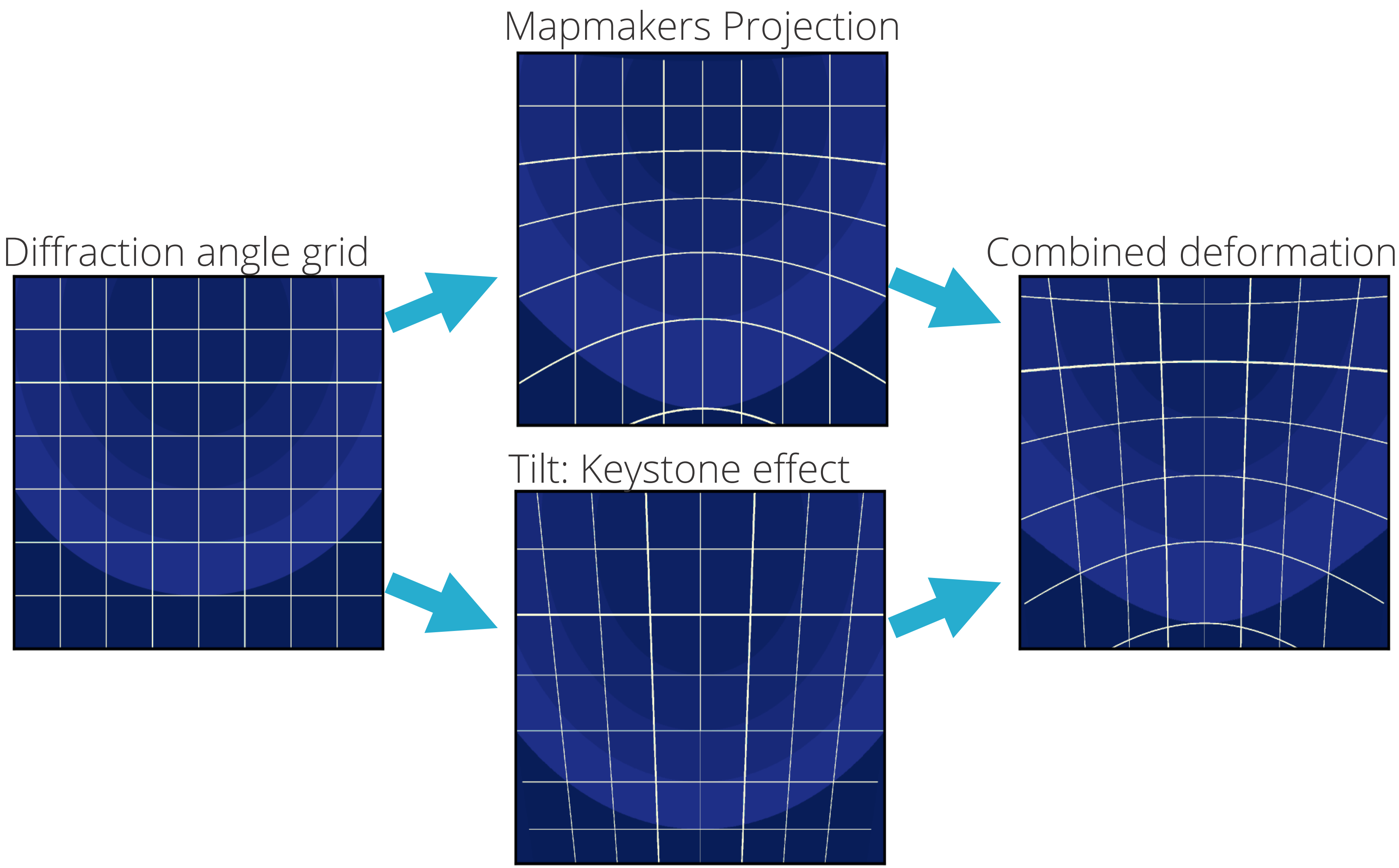


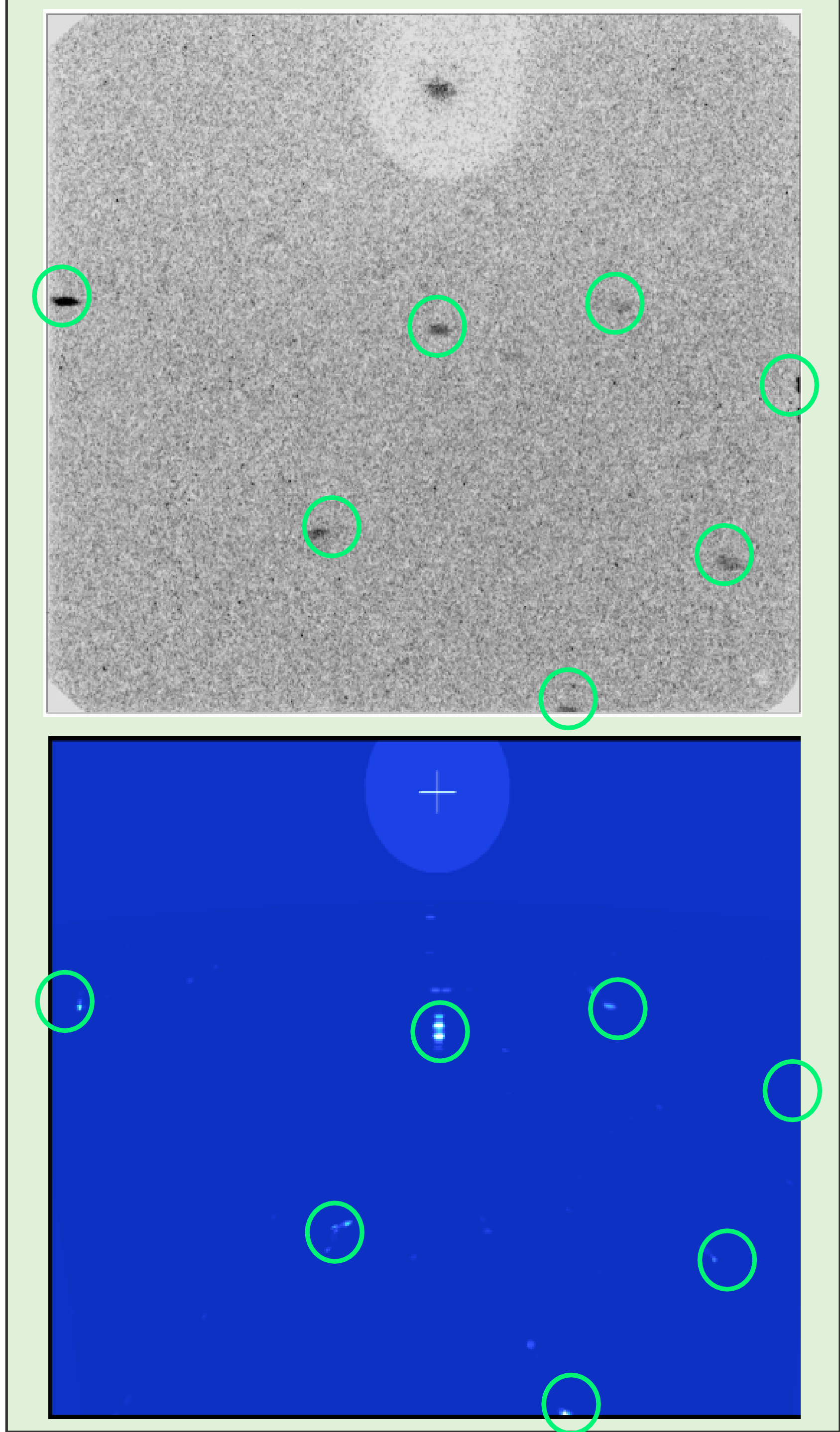


This is not a blur, "effective" spread model it is constructed by superposition just as in the experiments.

Simulations comparable to experiments

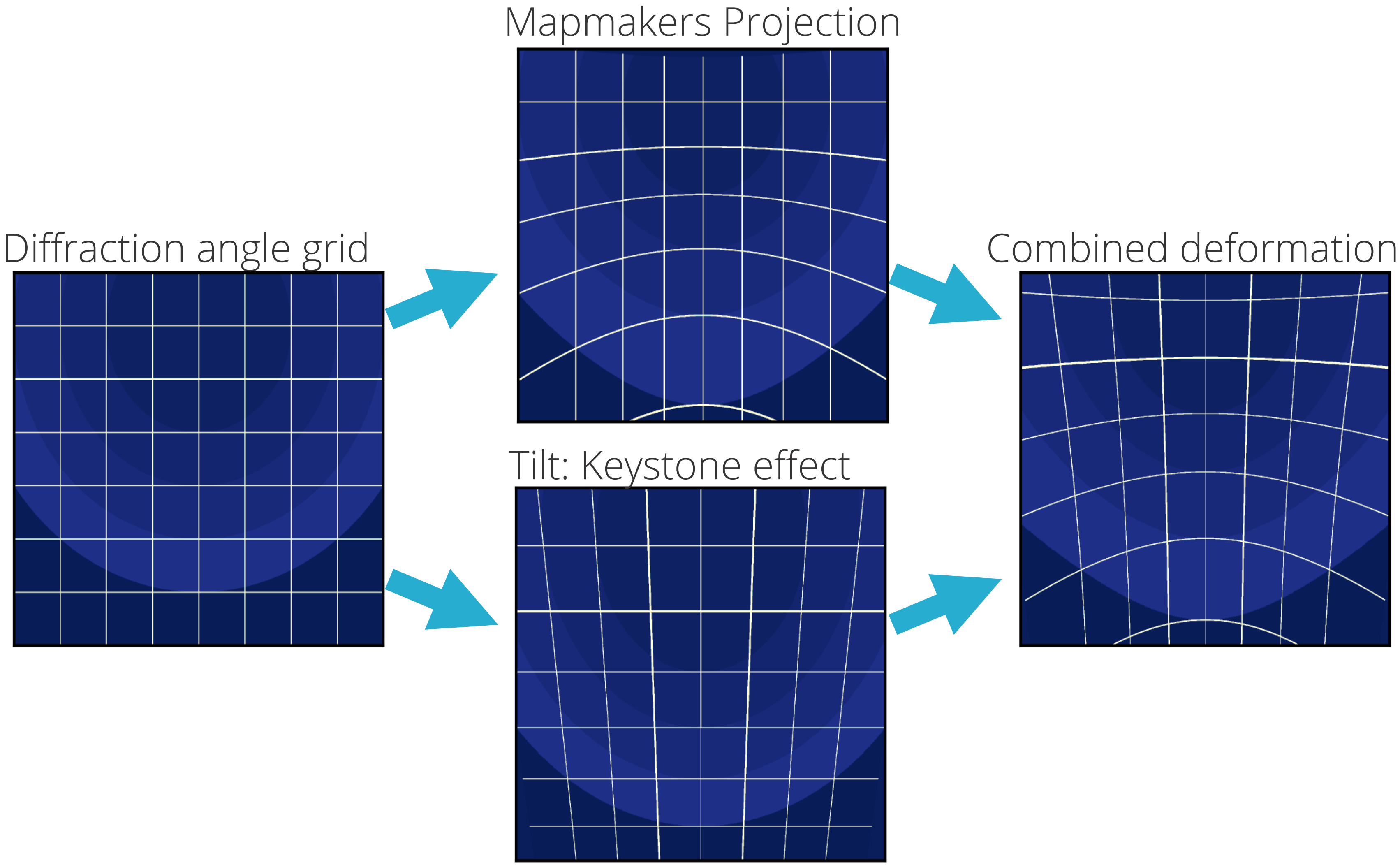
Analytical deformation calculation *for projection of pattern onto flat image plate (IP)*

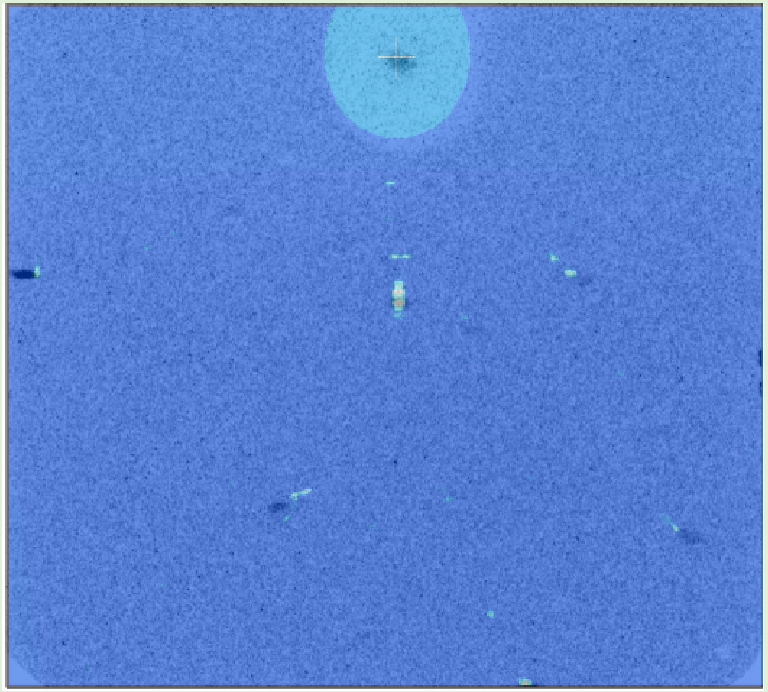




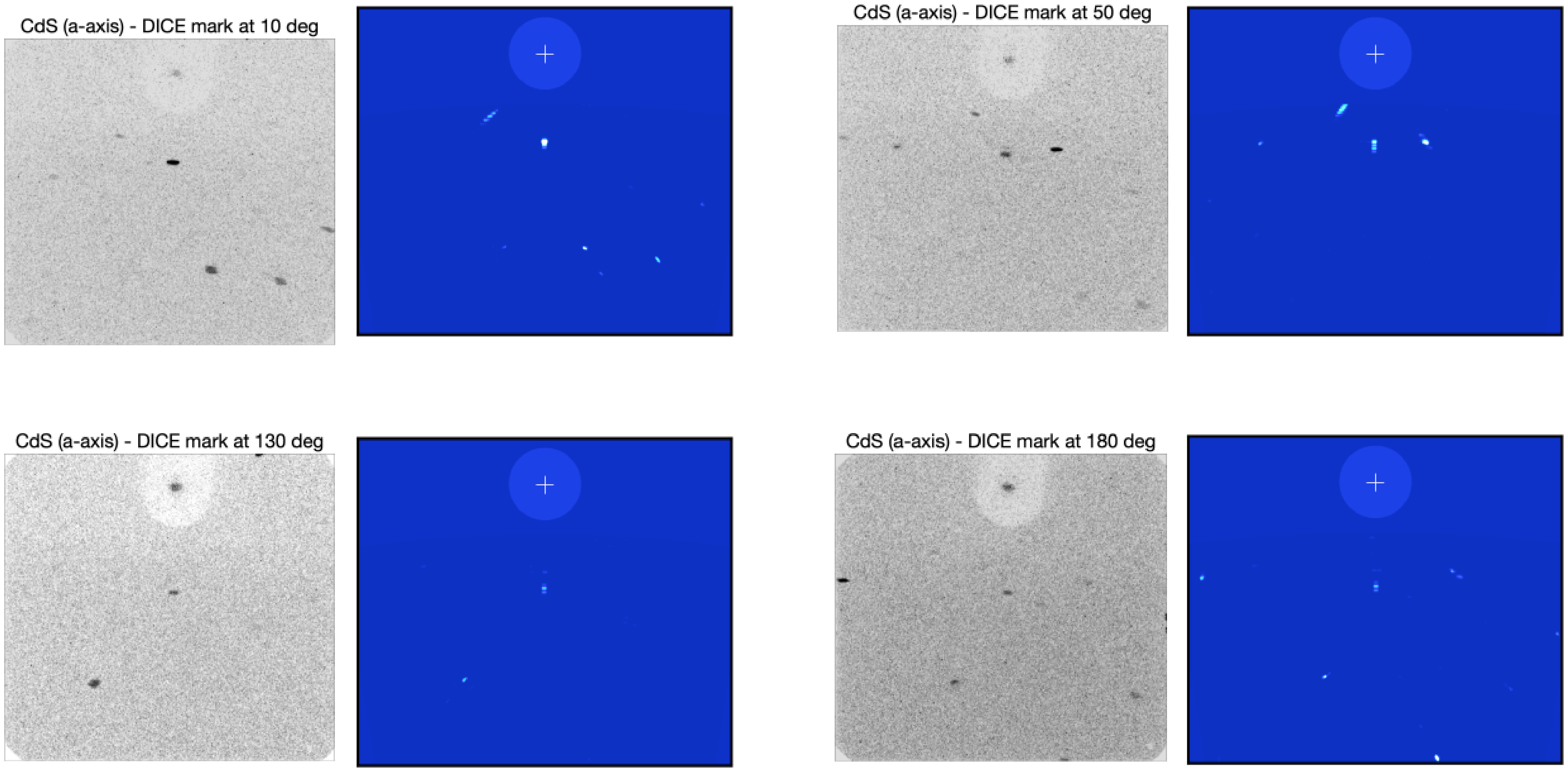
Simulations comparable to experiments

Analytical deformation calculation *for projection of pattern onto flat image plate (IP)*





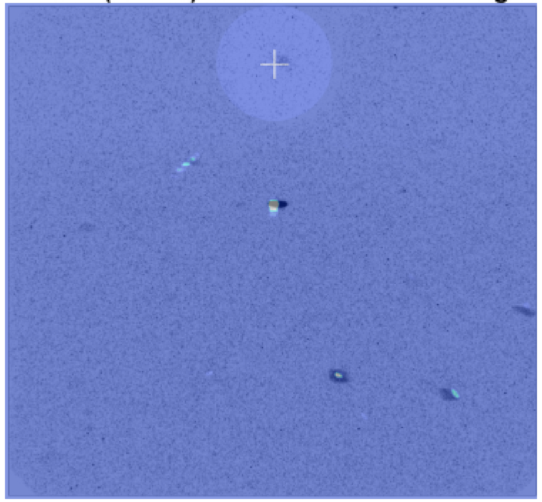
Robust prediction of the whole rotation



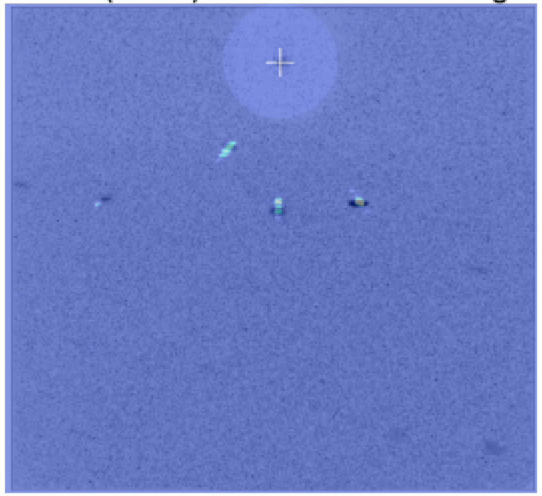
This is a single rotation about an axis, without individual fitting.

Robust prediction of the whole rotation

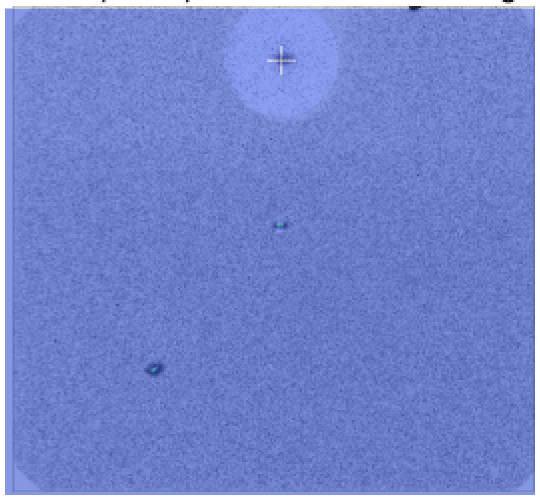
CdS (a-axis) - DICE mark at 10 deg



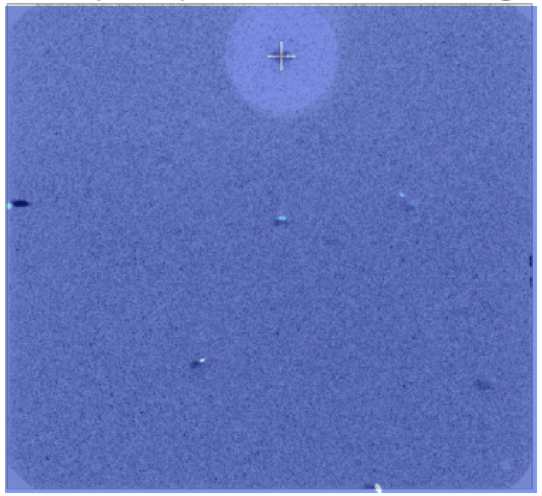
CdS (a-axis) - DICE mark at 50 deg



CdS (a-axis) - DICE mark at 130 deg

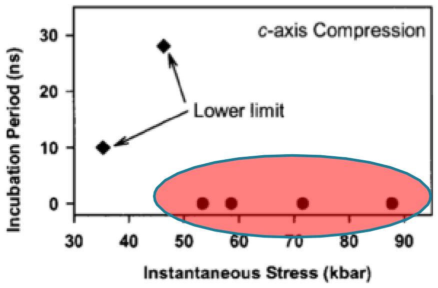


CdS (a-axis) - DICE mark at 180 deg



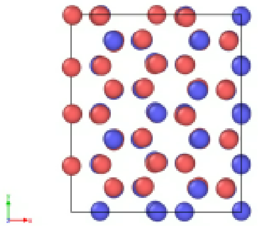
This is a single rotation about an axis, without individual fitting.

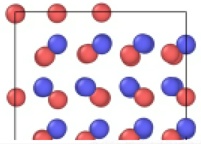
DCS Experiments capture high-pressure c-axis mechanism

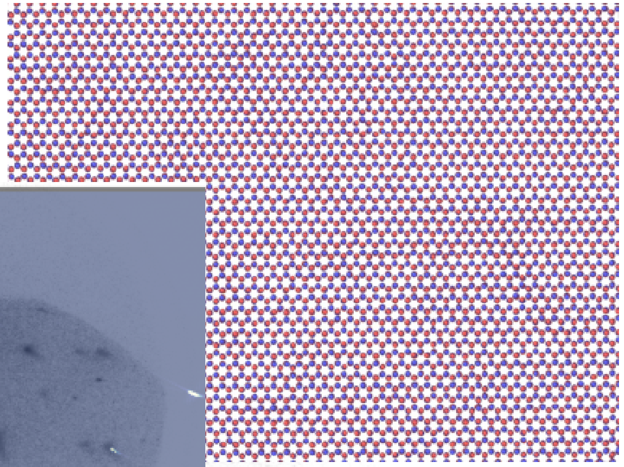


DCS 18-4-063
c-axis (4 deg off a-axis)
1.683 km/s

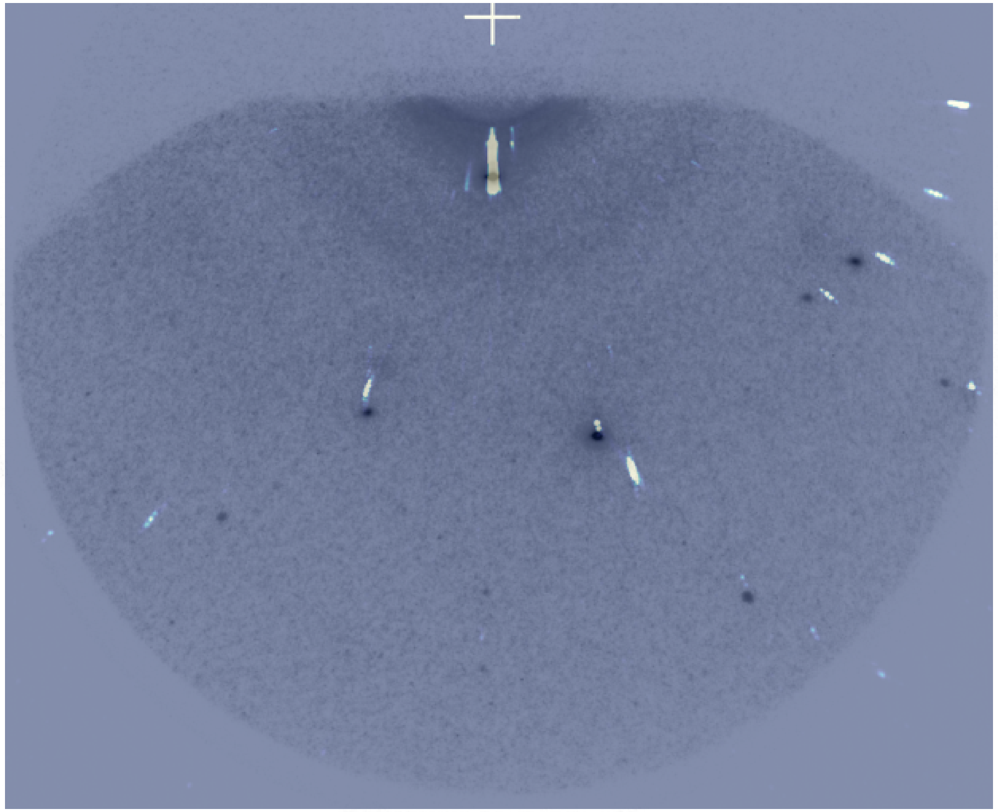
DCS data: Marcus Knudson
Lane, et al. SCCM AIP Conf. Proc. 2272, 100016 (2019)



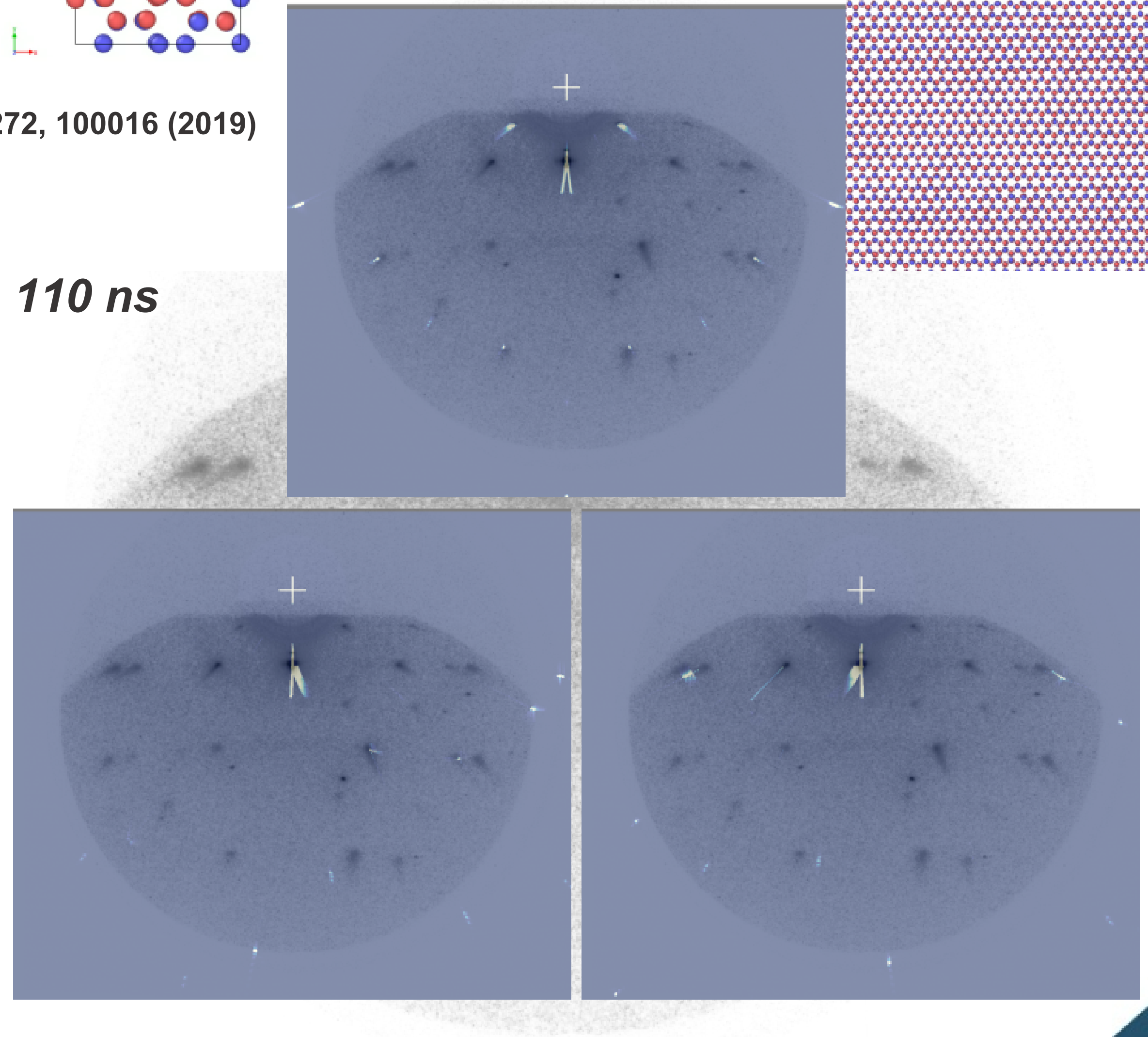




-43 ns

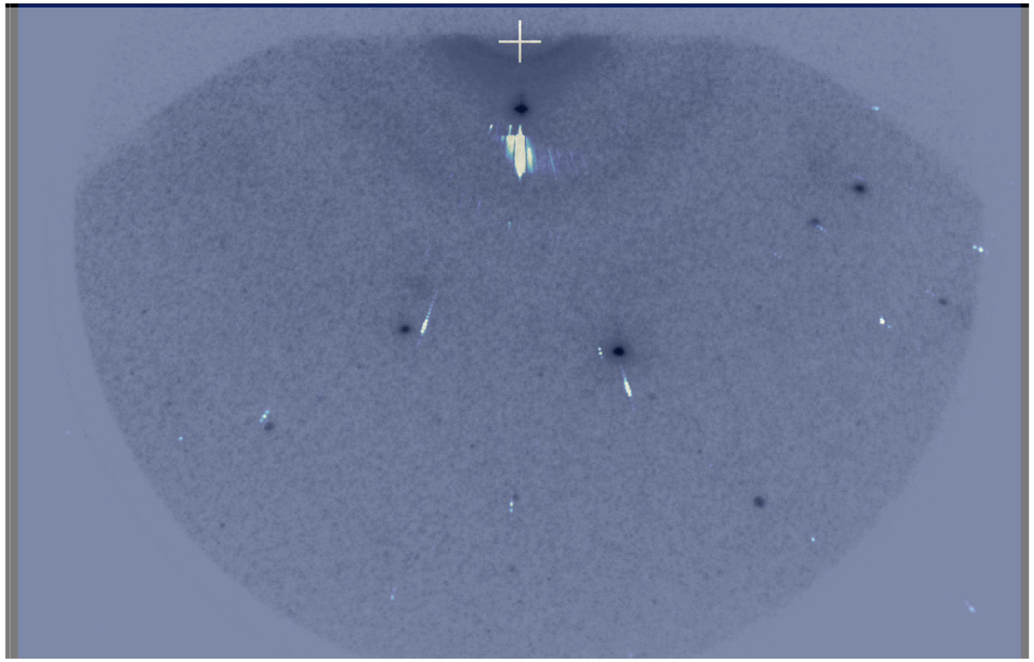


110 ns

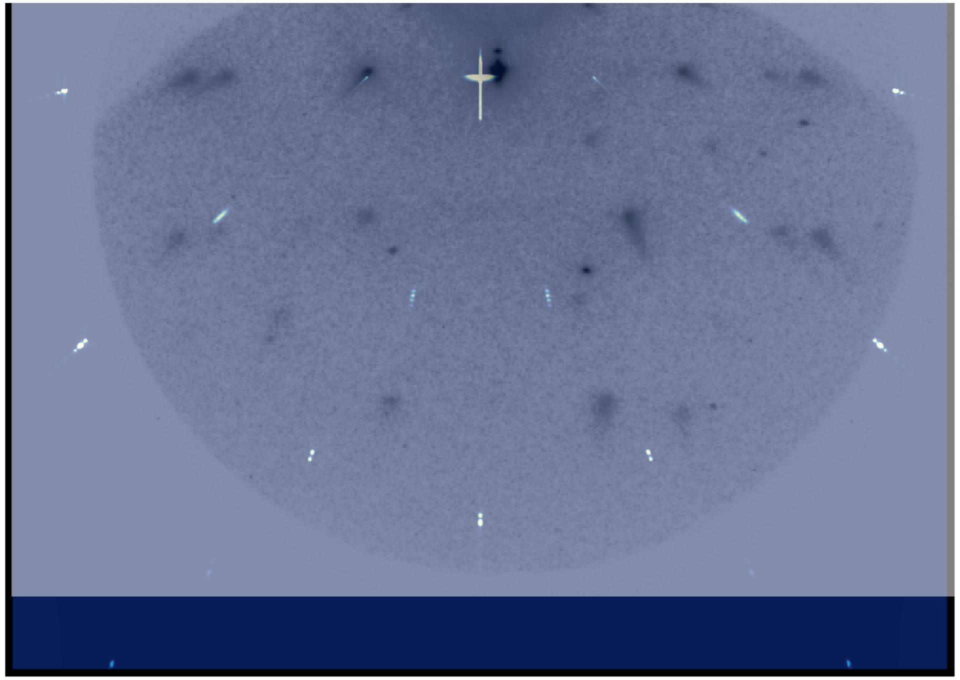


Pattern identification (currently excruciatingly manual)

CdS Wurtzite Structure



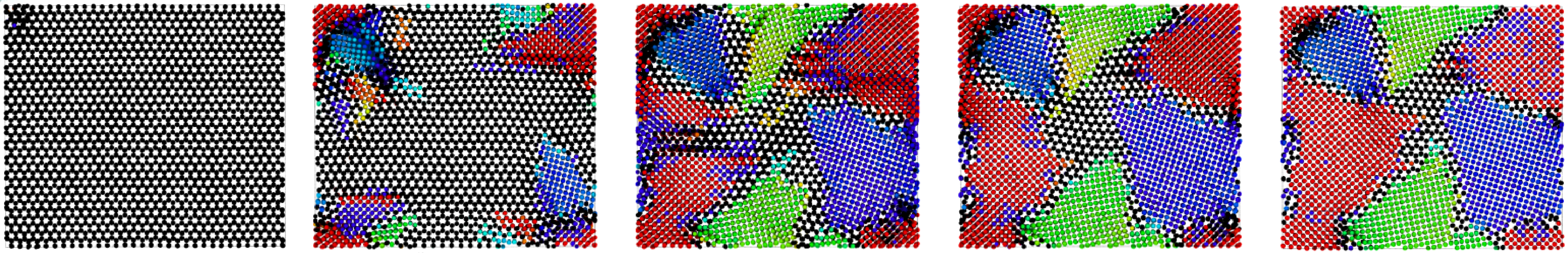
CdS Rock Salt Structure

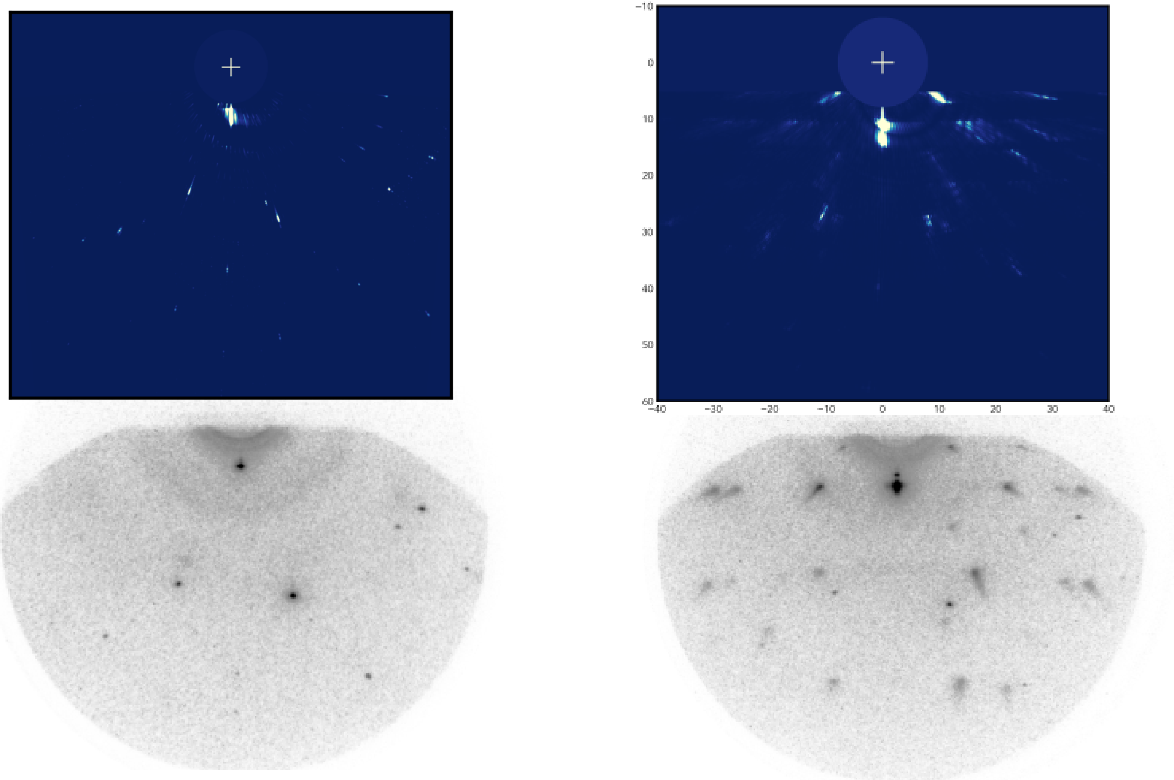


Rather: We are currently working to **apply machine learning** approaches to automate and improve this manual grid search to allow **effectively a 2D Rietveld refinement approach** using simulated data.

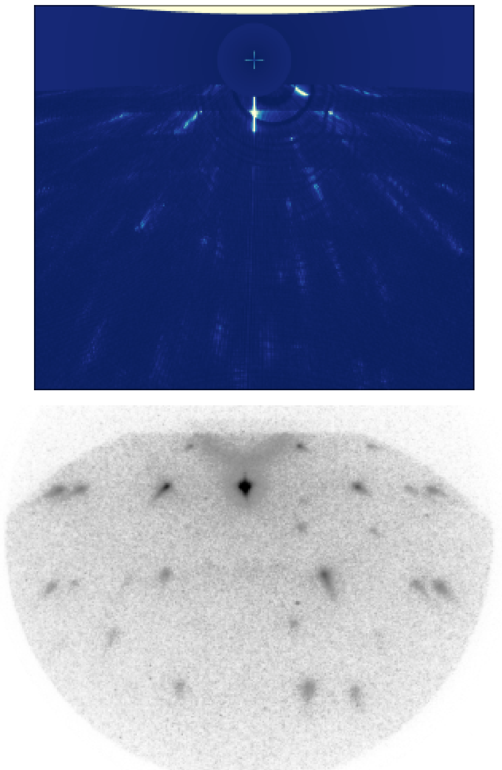
Time lapse XRD with mixed state simulations – high pressure c-axis

Lane, et al. SCCM AIP Conf. Proc. 2272, 100016 (2019)

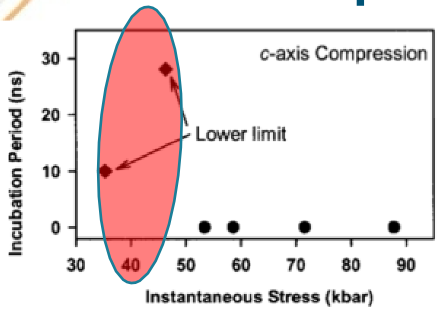




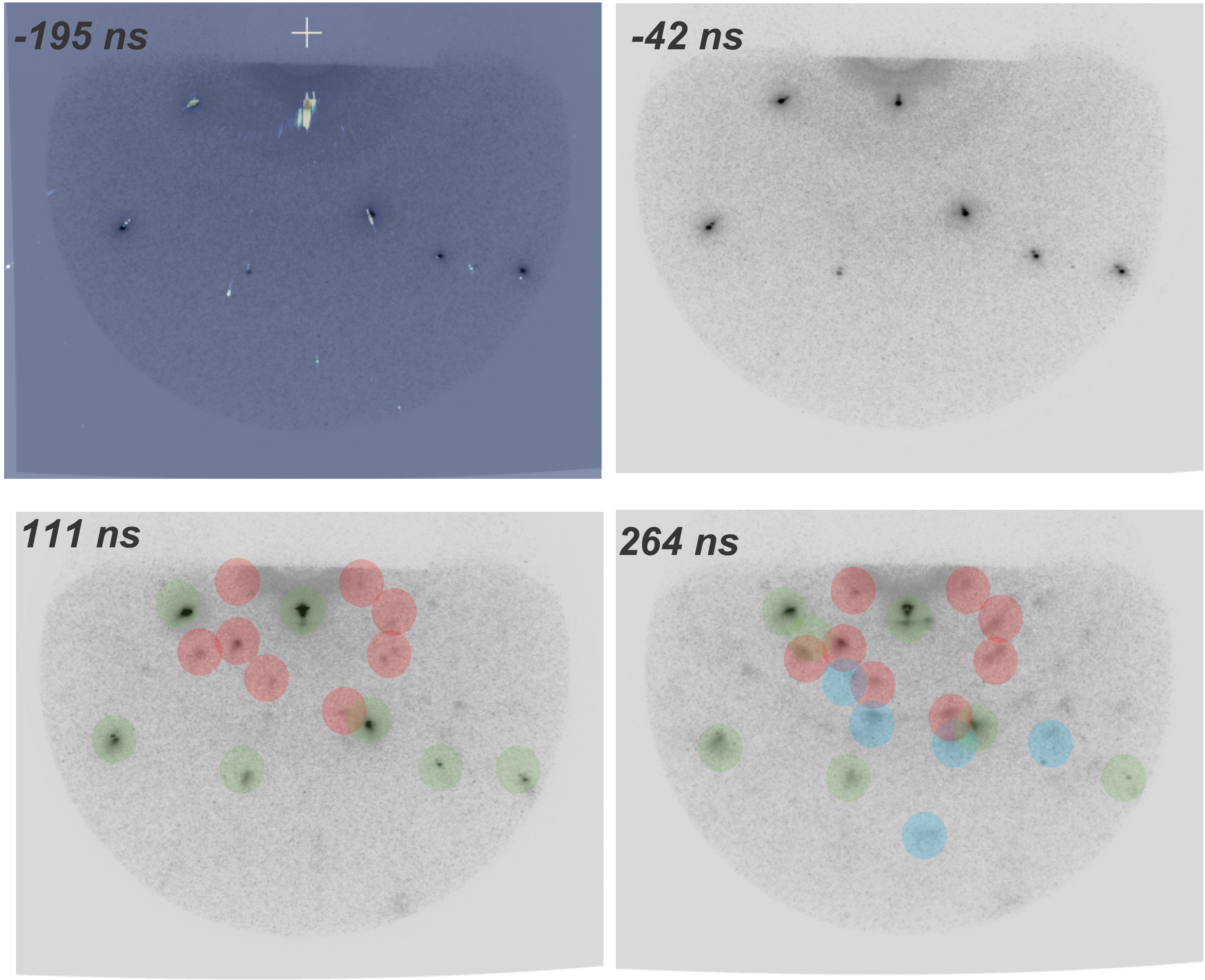
X-ray diffraction from atomistic simulation will allow realistic characterization of mixed states, partial phase transitions, twinning and other deformation mechanisms



Low pressure c-axis shock loading



DCS 19-4-074
c-axis (-7 deg off a-axis)
1.334 km/s



Wurtzite residual

Well defined intermediate

Possible Rock Salt

Conclusions and Future Directions

Realistically simulating 2D XRD patterns requires

- incorporating X-ray spectra, beam spread, & projection deformation
- LAMMPS reciprocal space calculation from any atomistic configurations

Here we used this new LAMMPS capability to simulate CdS compression and phase transformation on the Thor pulsed power driver and Dynamic Compression Sector gun

This work enables even more powerful new quantitative analysis of diffraction patterns

- incorporate machine learning to this now data rich analysis
- Allows 2D Reitveld refinement to optimize data extraction
- Allows incorporation of intensity variation in addition to spot location