Virtual Diffraction Using LAMMPS

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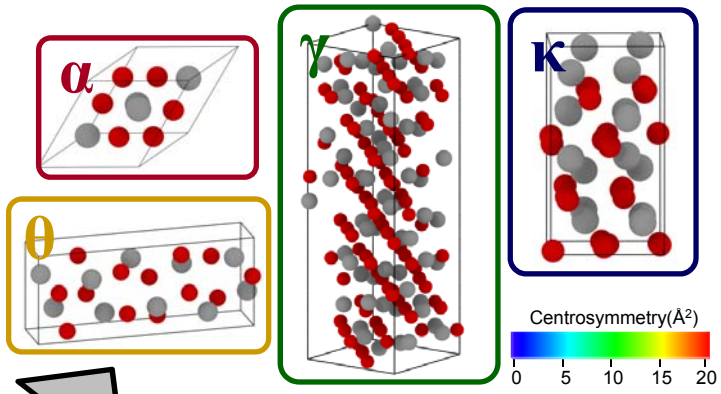
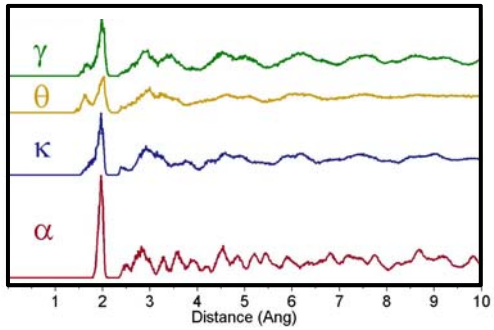
Motivation

- Identification of complex material (non-cubic) phases and phenomena within atomistic simulations is difficult.
- Experimental x-ray and electron diffraction routinely used for characterization of (complex) materials.
- Fundamental disconnect between simulation and experimental characterization tools.

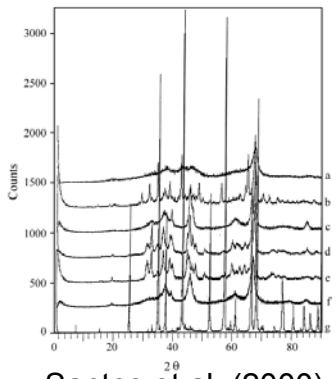
Alumina Polymorphs



Radial Distribution Functions

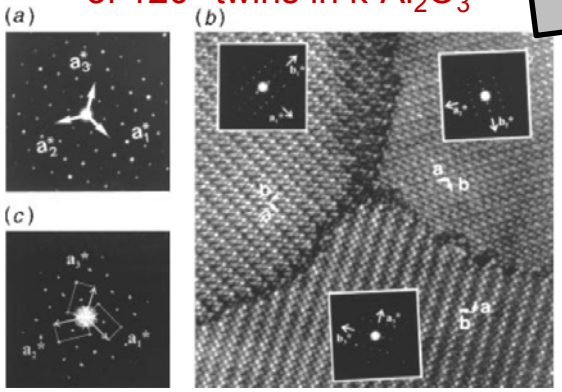


XRD of Alumina Phases



Santos et al. (2000)

SAED and HRTEM of 120° twins in κ-Al_2O_3



Ollivier et al. (1997)

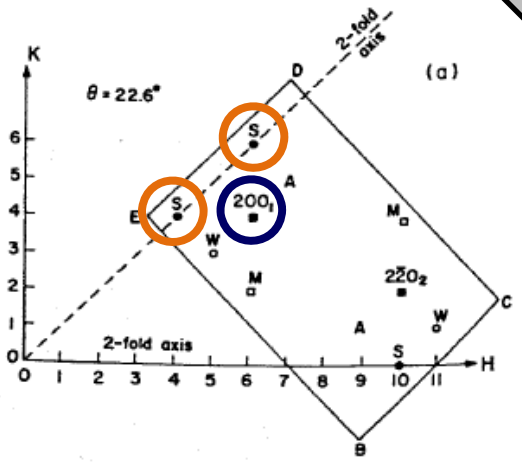
Virtual diffraction offers a bridge to directly connect atomistic simulations with experimental characterization.

Background

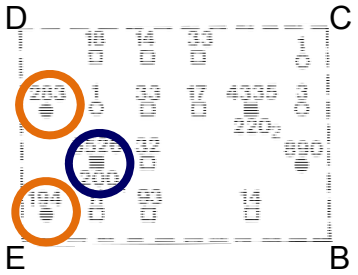
Previous studies have paved the way to the current virtual diffraction algorithm and implementation:

Twist boundaries in Au
(Bristowe and Sass, 1980)

Experimentally Observed



Computed Structure Factor

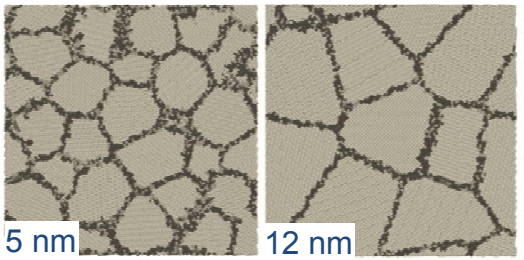


Required knowledge of crystal structure.

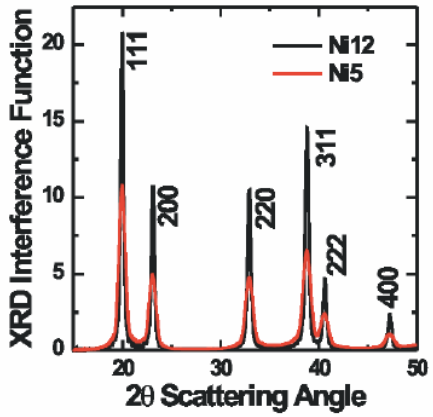
Produced only XRD patterns

Limited Implementation

Nanocrystaline Ni
(Derlet et al, 2005)

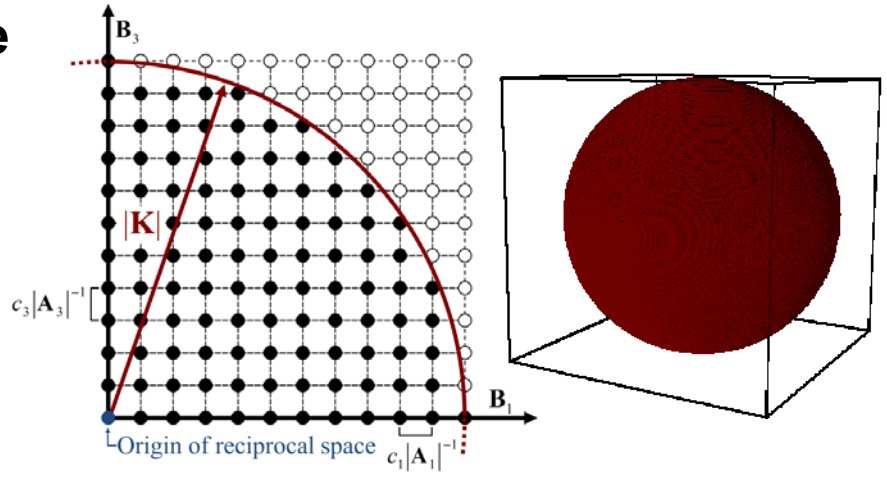


Debye Scattering Equation



Virtual Diffraction from 10,000 ft

1. **Create a mesh of reciprocal space**
   - o 3D rectilinear mesh but with spherical boundary conditions.
   - o Ultrafine resolution eliminates any need for prior knowledge of the crystal structure.



2. **Compute diffraction intensities at each point on the reciprocal space mesh using the structure factor equations**
   - o Compute structure factor for all atoms within the simulation.

→ **Implemented into the LAMMPS MD code**

3. **Analysis and visualization of diffraction intensities to produce**
   - o X-ray diffraction line profiles (XRD)
   - o Selected area electron diffraction patterns (SAED)

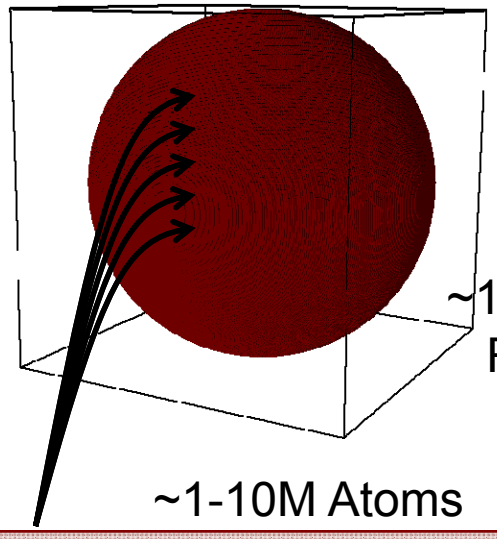
Diffraction Intensity Computation

At each reciprocal lattice node the diffraction intensity is calculated utilizing the structure factor equation,

$$I(\mathbf{K}) = \mathrm{Lp}\, F(\mathbf{K}) F^{*}(\mathbf{K})$$

Lp = Lorentz-Polarization Factor (only XRD)
F = Structure Facture
$\mathbf{K}$ = Reciprocal Lattice Point (Relp)

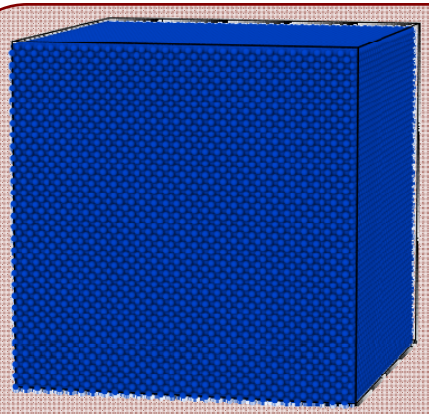
Sampled Reciprocal Space



Structure Factor:

$$F(\mathbf{K}) = \sum_{j=1}^{N} f_j \exp\left(2\pi i\, \mathbf{K} \cdot \mathbf{r}_j\right)$$

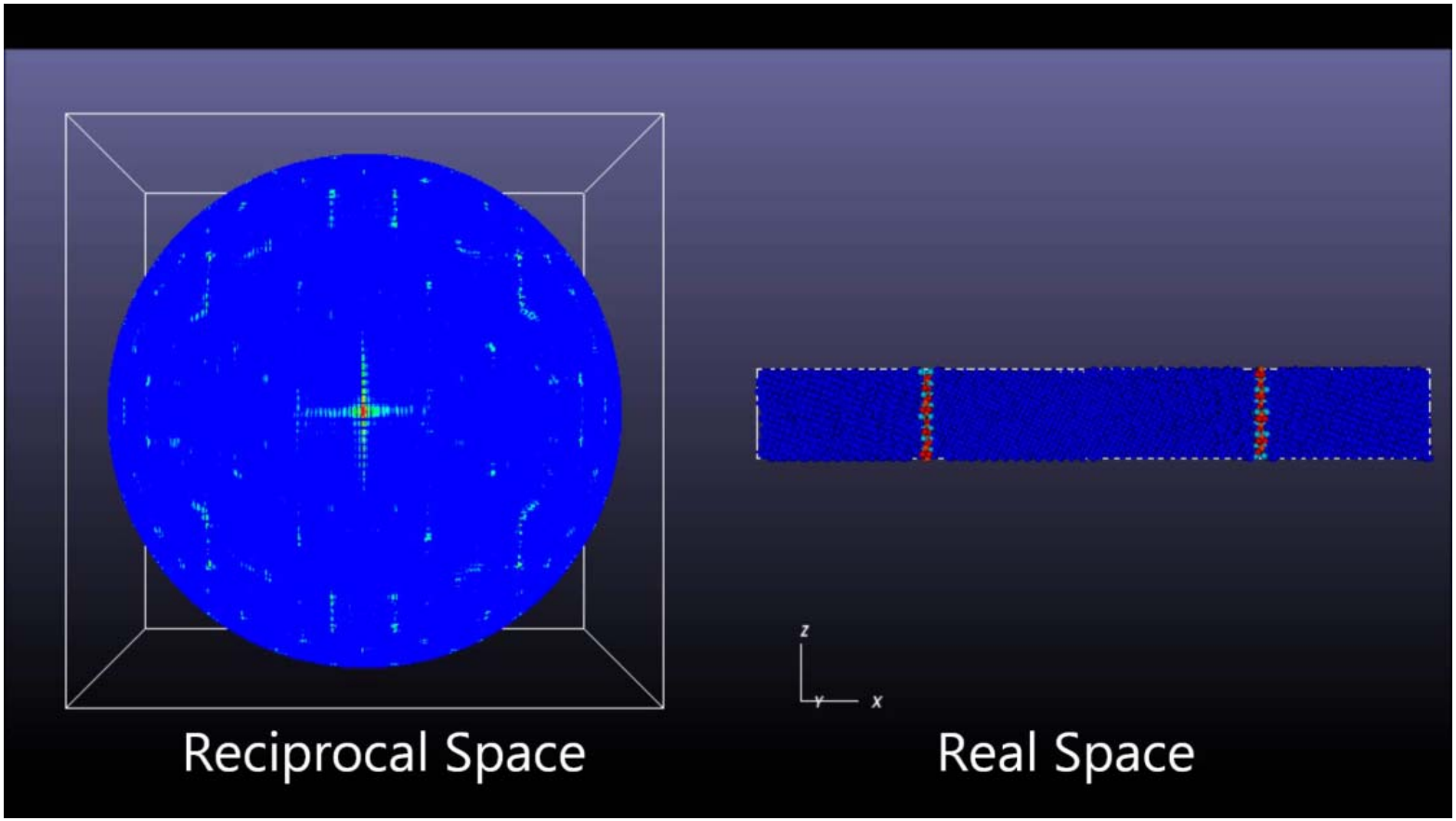
N = Number of Atoms in Simulation
f_j = Atomic Scattering Factor
$\mathbf{r}_j$ = Atom Position



All atoms are sampled at each reciprocal point.

3D Diffraction Data

- Example 3D diffraction data taken from Ni tilt grain boundary.



- How to visualize 3D diffraction data to represent experimental techniques and make unique insights?

Diffraction Theory

- Diffraction conditions satisfied when a reciprocal lattice node is located on the surface of Ewald's sphere

X-Ray Powder diffraction patterns

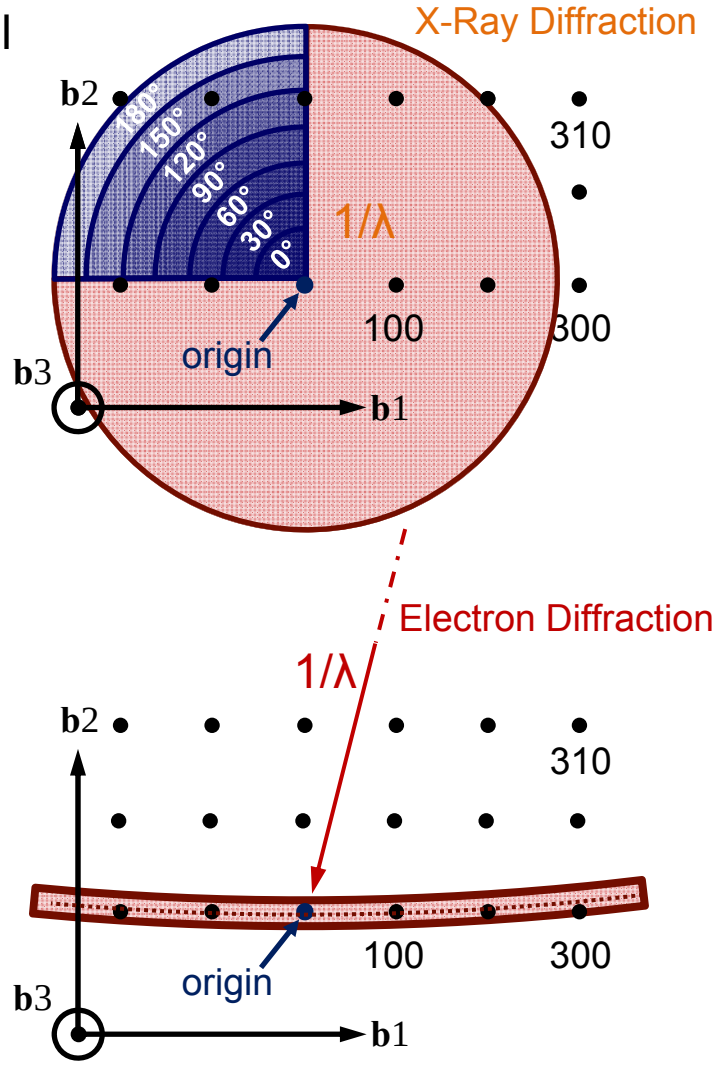
- All orientations of the crystal equally possible
- Intensity data are binned corresponding to their diffraction angle calculated using Bragg's law:

$$2\sin(\theta)/\lambda = |\mathbf{K}|$$

- Concentric spherical shell bounding angles

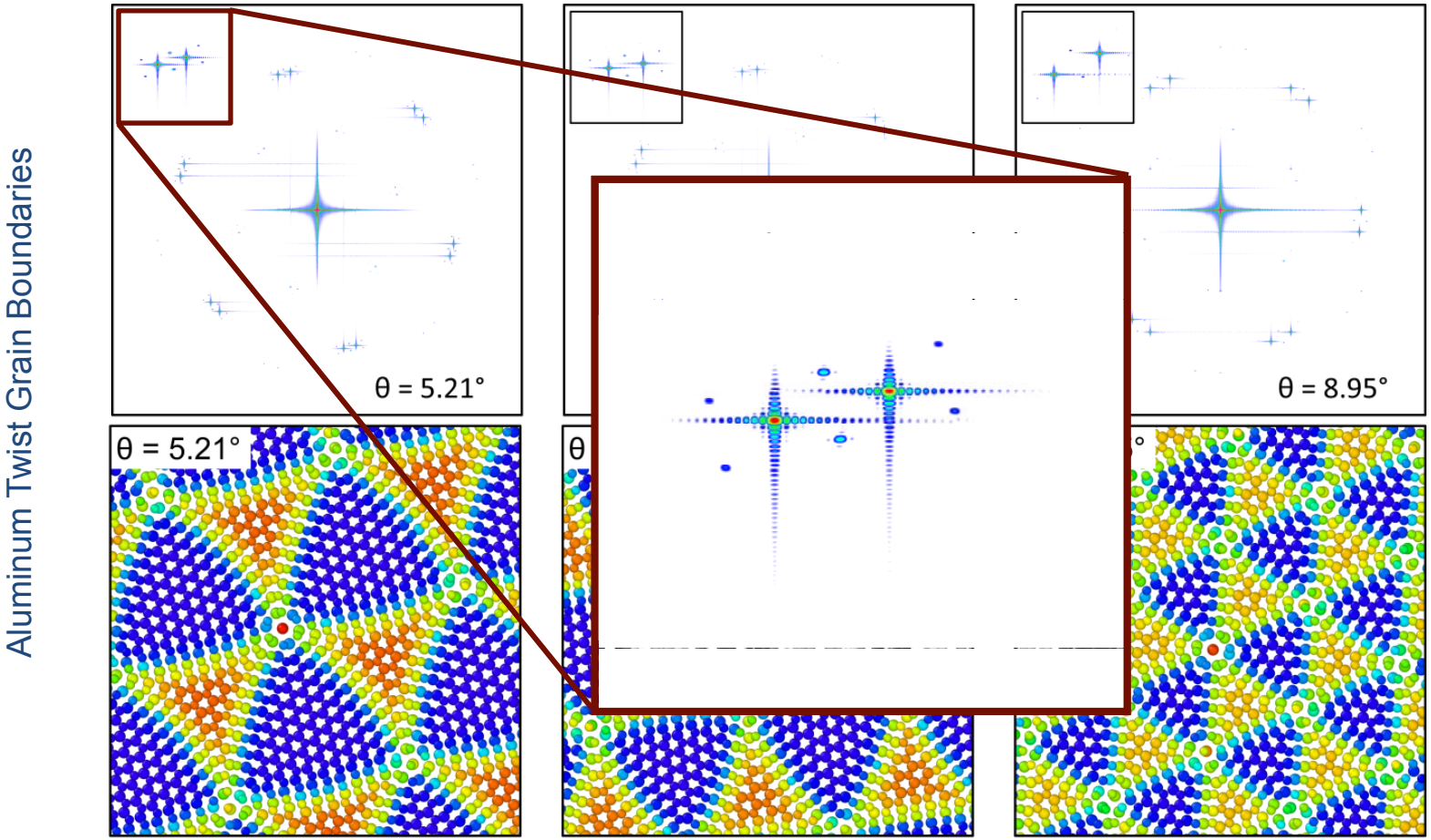
Electron diffraction patterns

- Radius of Ewald sphere much larger
- Diffraction conditions intersect a near planar slice of reciprocal space lattice



Check – Al Twist GBs

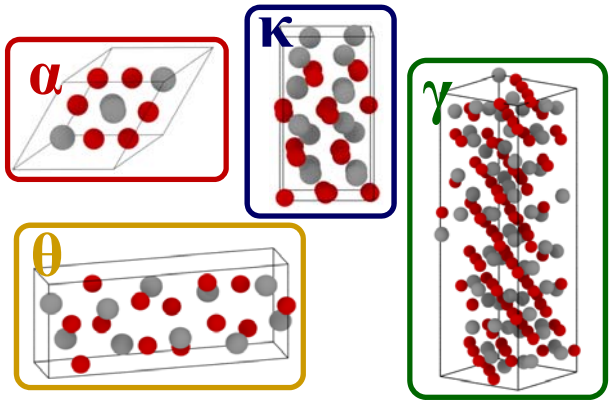
- Virtual SAED patterns clearly identify misorientation in Al {111} twist GBs.
- Secondary peaks emerge due to screw dislocation network.



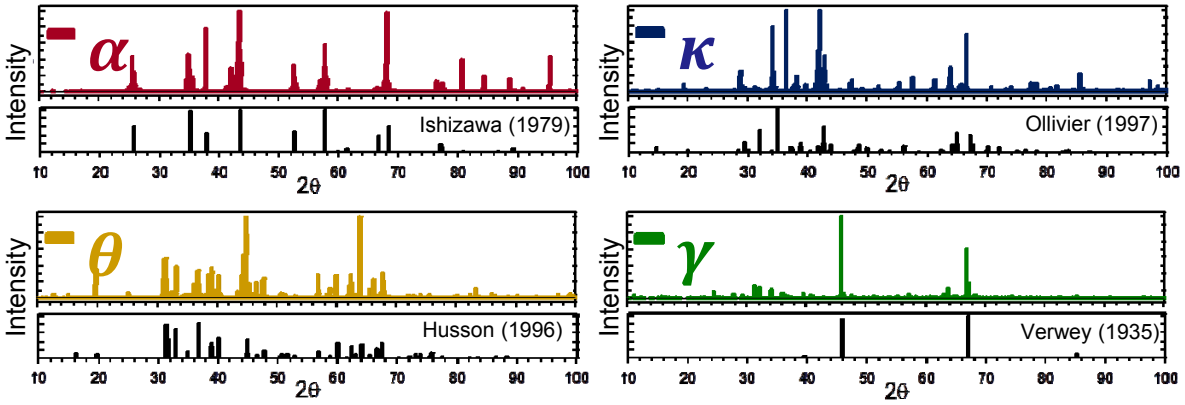
Bulk Alumina

- Virtual x-ray diffraction patterns can successfully differentiate alumina polymorphs.
- Virtual XRD patterns can be compared directly to experiments to validate the fidelity of the potential – ReaxFF.

Al_2O_3 Polymorphs



X-ray Diffraction

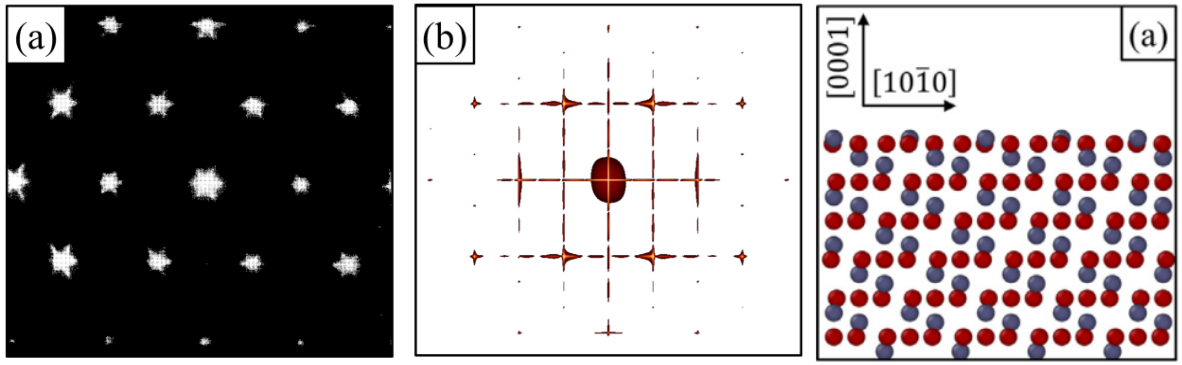


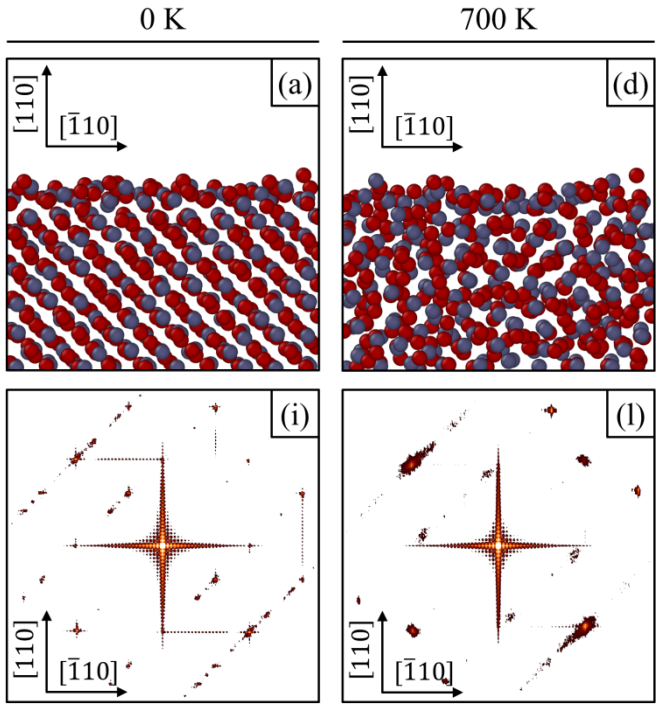
- o A generic tool for structural identification in simulations of complex materials.
- o Coupled with optimization routine, can become a route to provide atomic resolution of experimental diffraction patterns – including defects.

Alumina Surfaces

- Virtual SAED patterns can verify the correct orientation of surface simulations.

α-Al_2O_3 (0001) Surface





- Virtual SAED identify substantial surface reconstructions still have underlying structure.

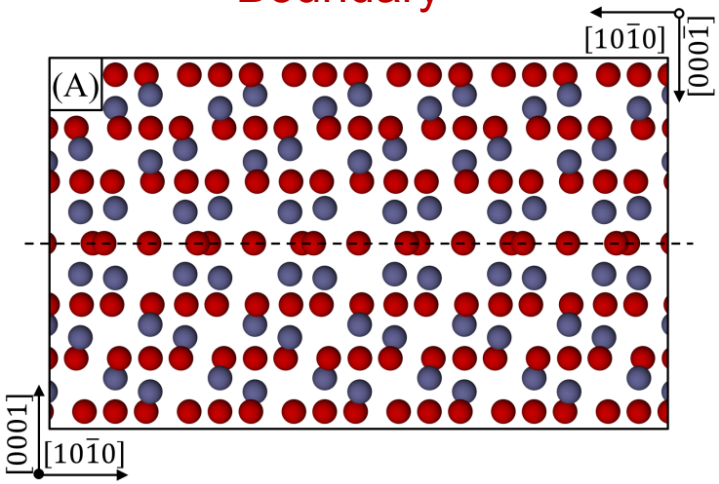
- o Providing atomic detail to smearing of diffraction peaks can help associate to specific strain field.
- o Tracking 'motion' of peaks to identify phase transformation mechanisms.

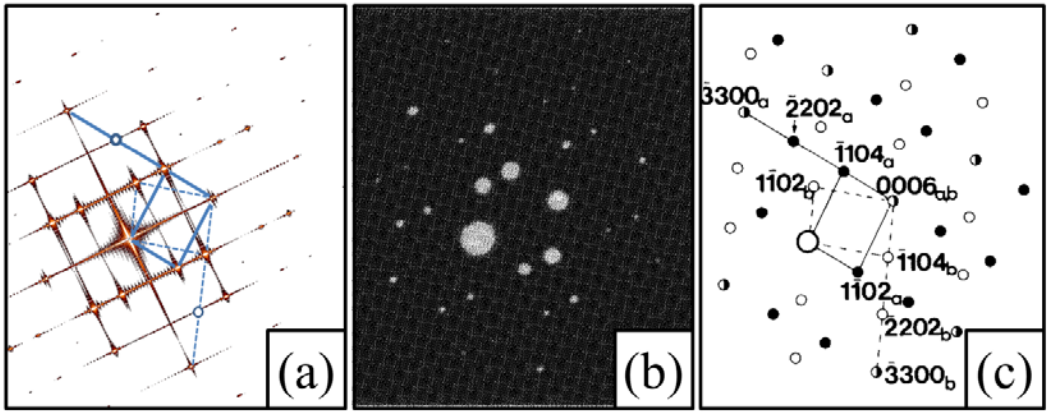
Alumina Interfaces

- Virtual SAED patterns help identify misorientation in Al_2O_3 GB simulations.
- Simulated patterns are matched to experimental (when available) to provide further validation of the structure.

- Virtual SAED patterns aided the construction of complex interfaces.
- Optimal orientation relationship evaluated by overlapping diffraction peaks.

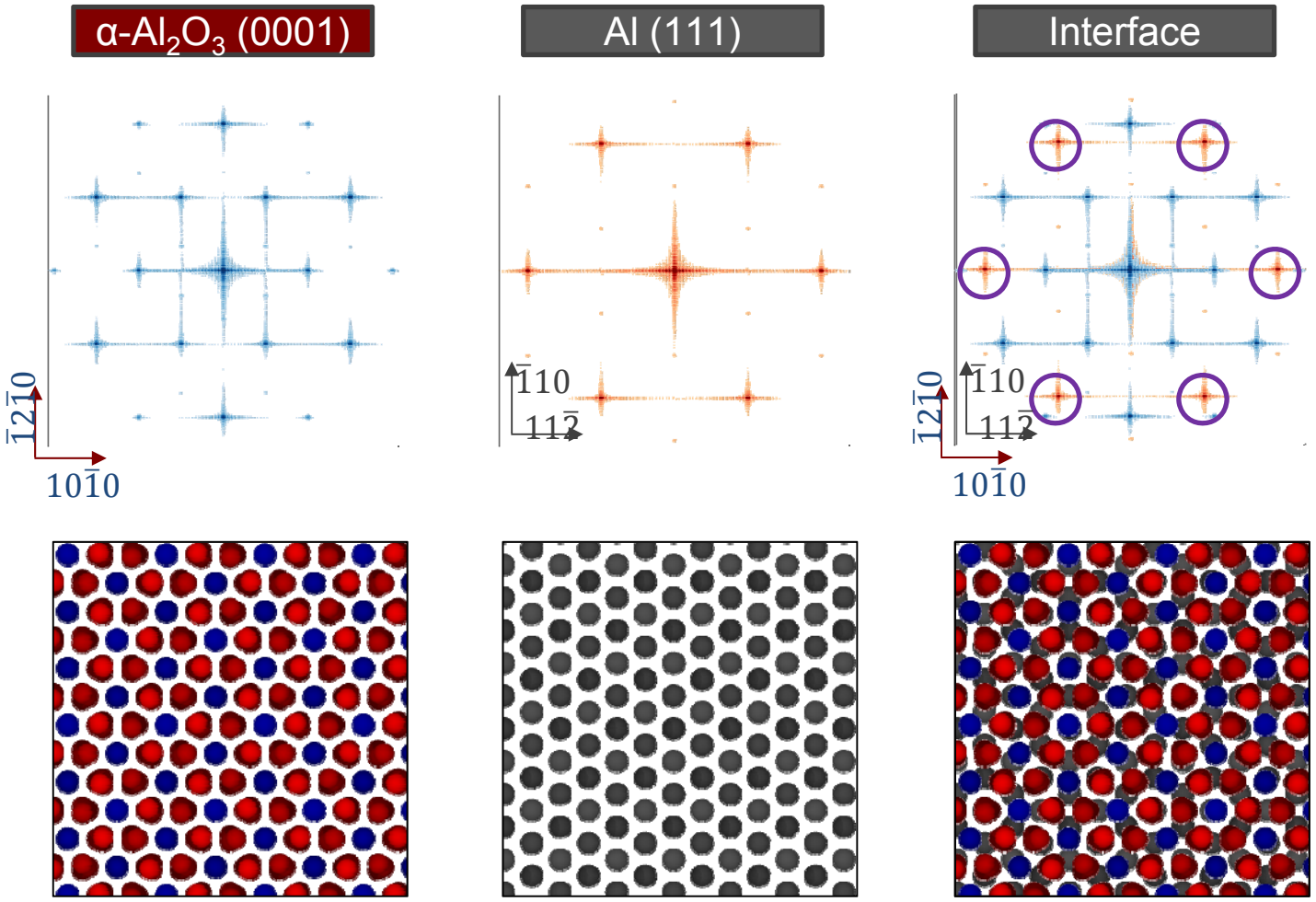
α-Al_2O_3 Basal Twin Boundary





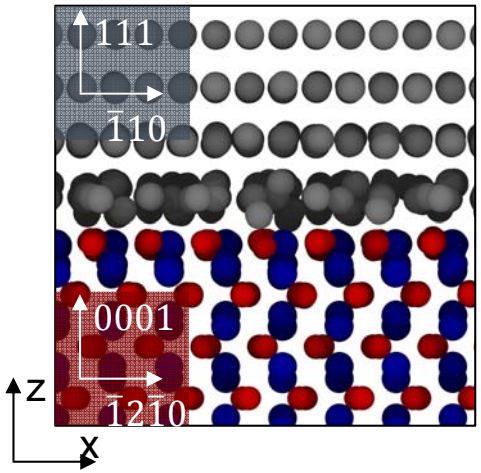
α-Al_2O_3 (0001) / Al (111) Orientation Relationship

- Lower interface energy found at maximum overlapping diffraction peaks.

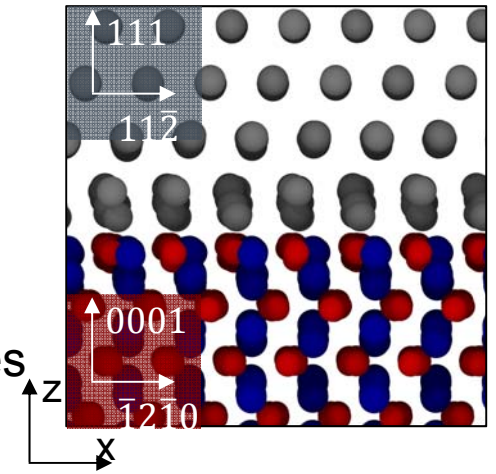


α-Al_2O_3 (0001) / Al (111)

Initial Orientation



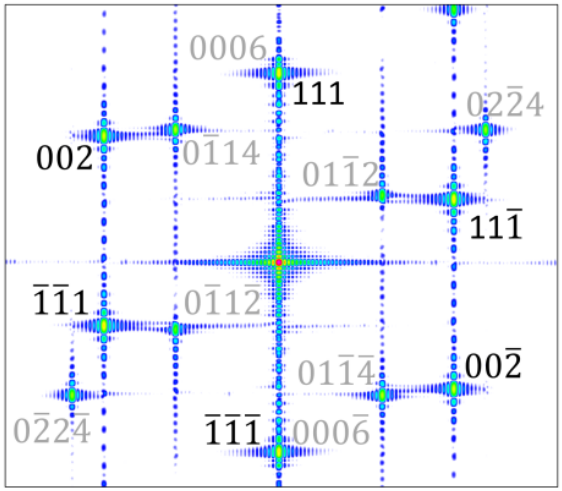
Optimized Orientation

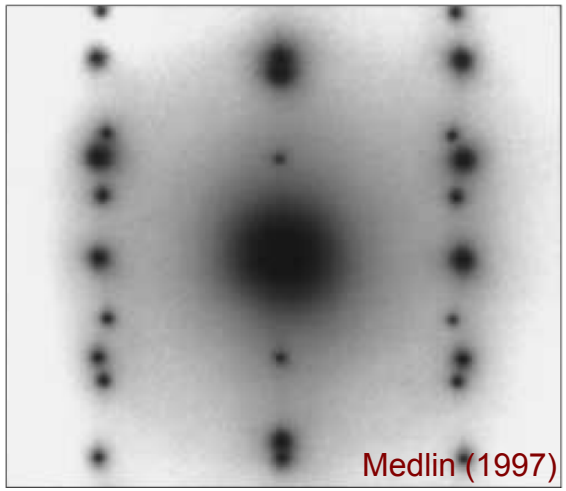


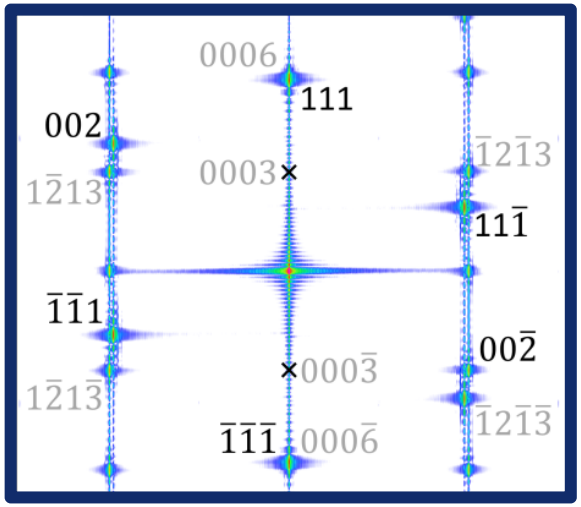
Work of adhesion using ReaxFF agree with DFT prediction

- Initial - 0.40 J/m^2
- Optimized - 0.82 J/m^2
- DFT: 1.06 J/m^2 Siegel (2002)

Optimized orientation SAED agrees with experimental.

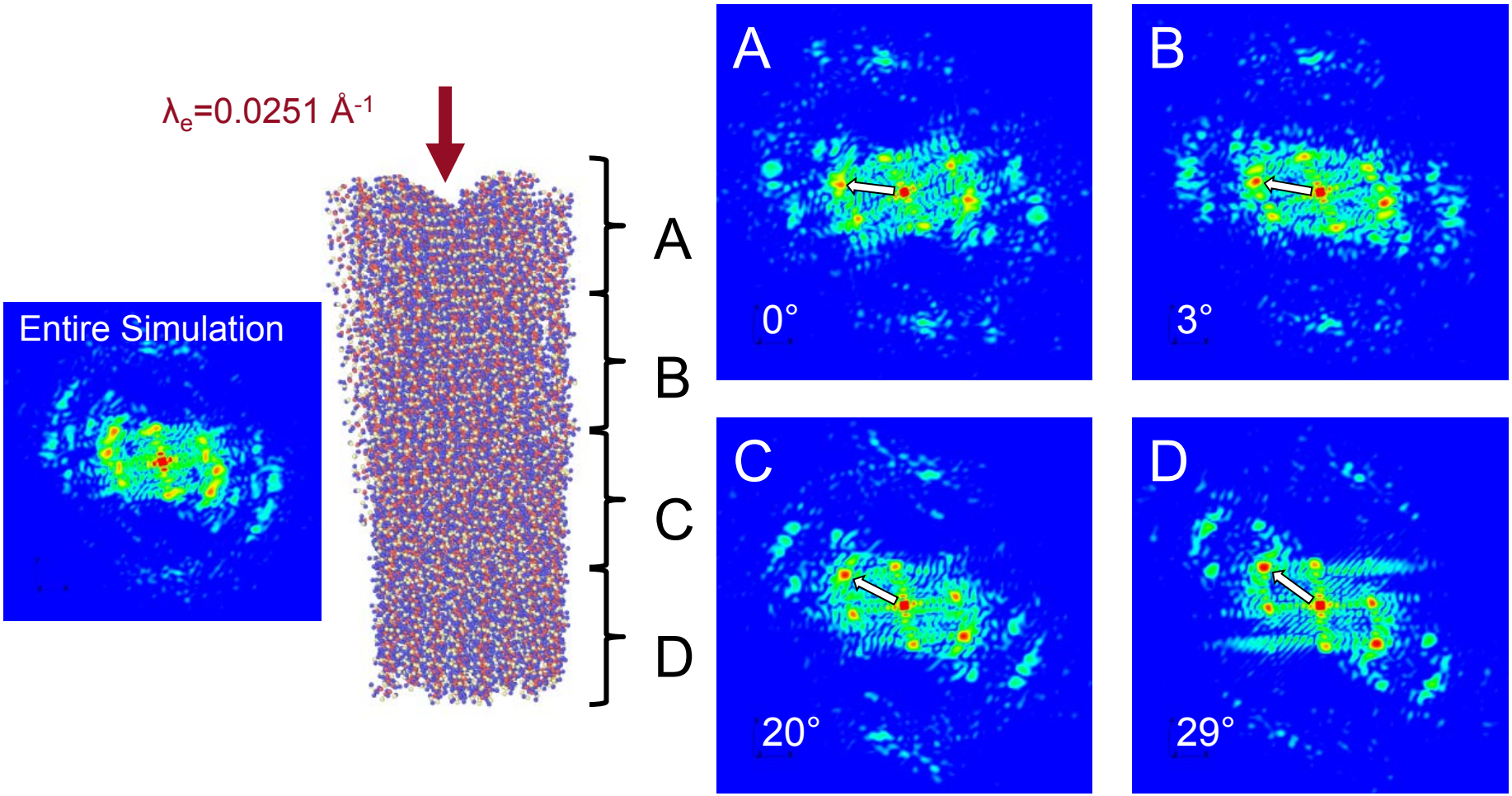






Cellulose Fiber Rotation

- Virtual SAED patterns used to characterize cellulose fibers.
- Peak shifts indicate a rotation of the cellulose matrix.



Wrap Up

Conclusions

- Implementation of XRD and SAED virtual diffraction methods open new routes for greater coupling between experiments and simulations.

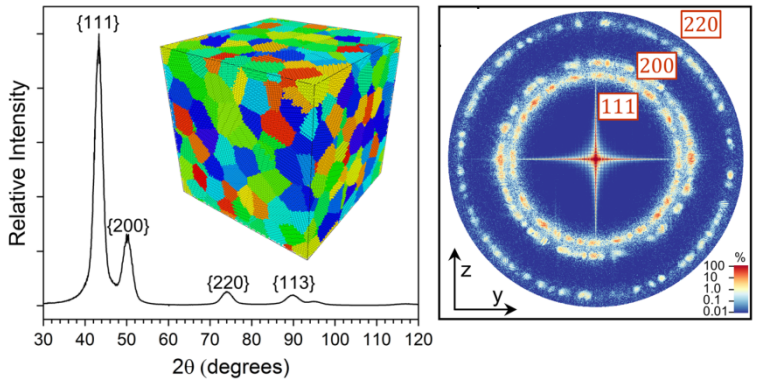
- Designed to be sufficiently generic for any material system and is adaptable to multimillion atom simulations.

Future Outlooks

- Using virtual diffraction to identify mechanisms of phase transformation and grain growth.

- Investigating the coupling between simulation length scales using peak broadening.

Nanocrystalline Metals



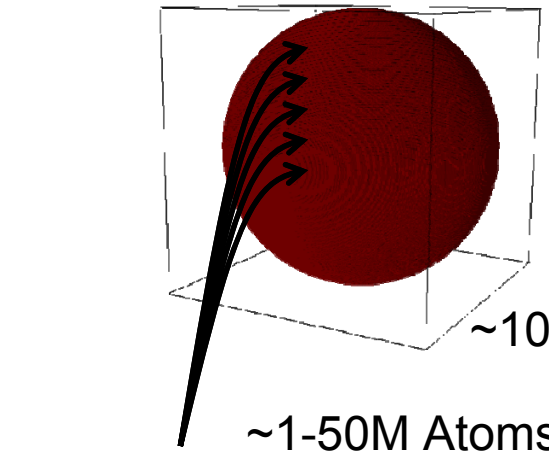
Thank You

Other Slides:

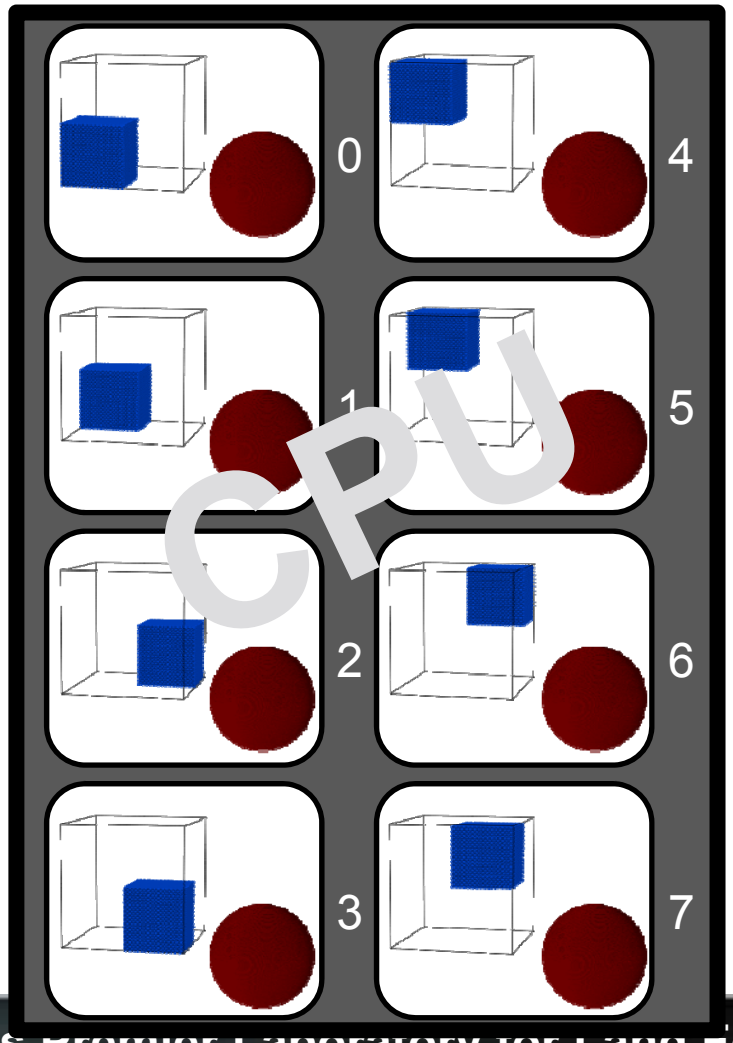
1) Parallelization
2) Diffraction commands

MPI parallelization over real space

- LAMMPS - Spatial decomposition of atoms parallelized over MPI processes
- Each process must have a copy of the reciprocal space mesh

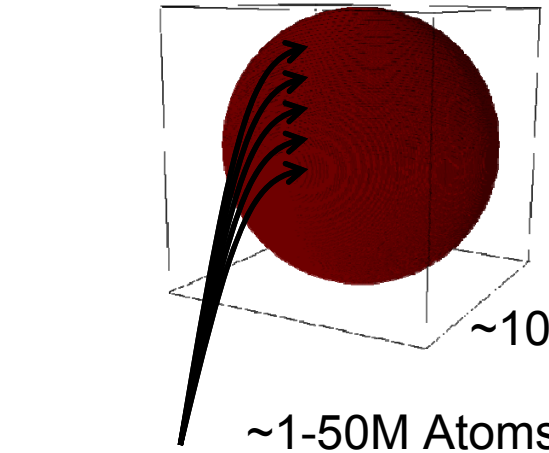


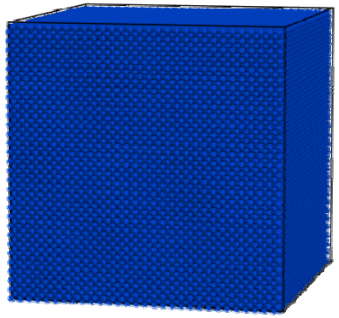
Very Memory Intensive



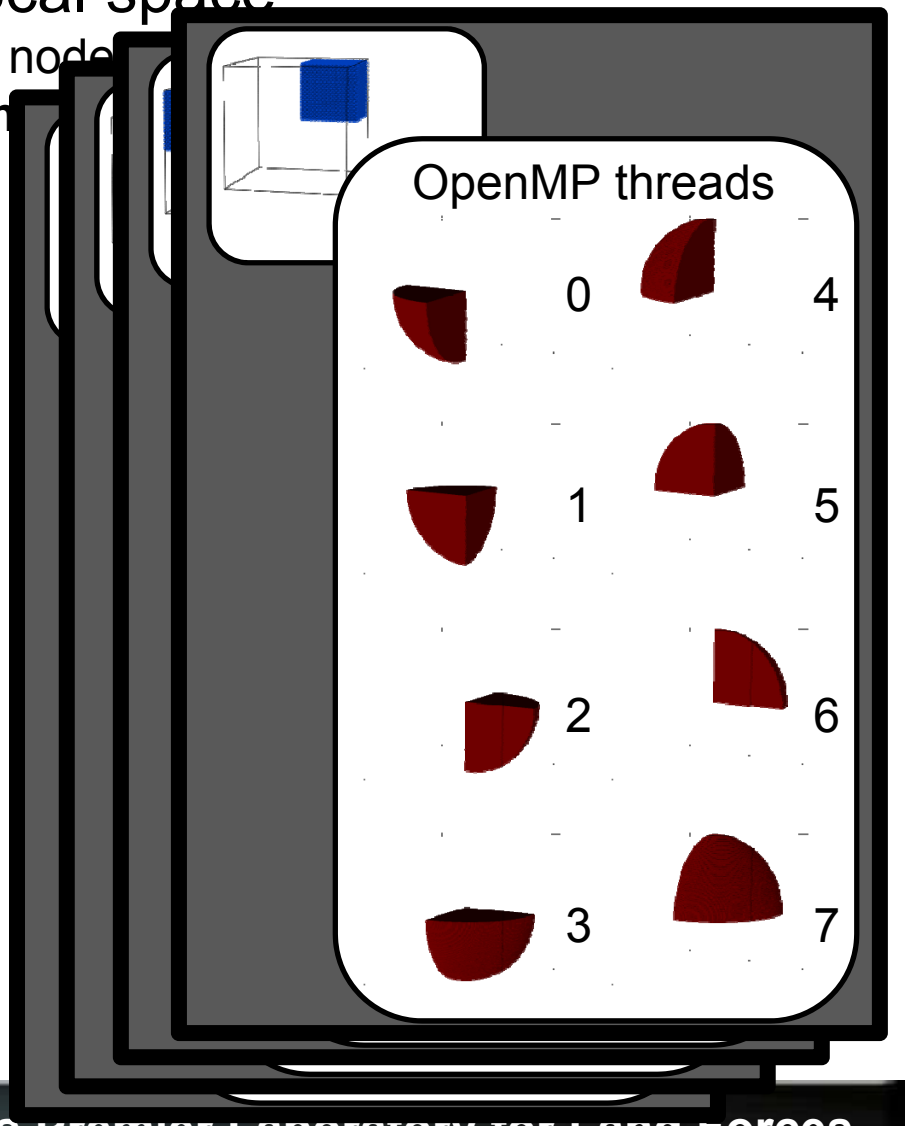
OpenMP parallelization of reciprocal space

- Reduce the number of MPI processes per node
- **Decompose reciprocal space with Open**
- MPI Parallelization of atoms across multiple CPU nodes



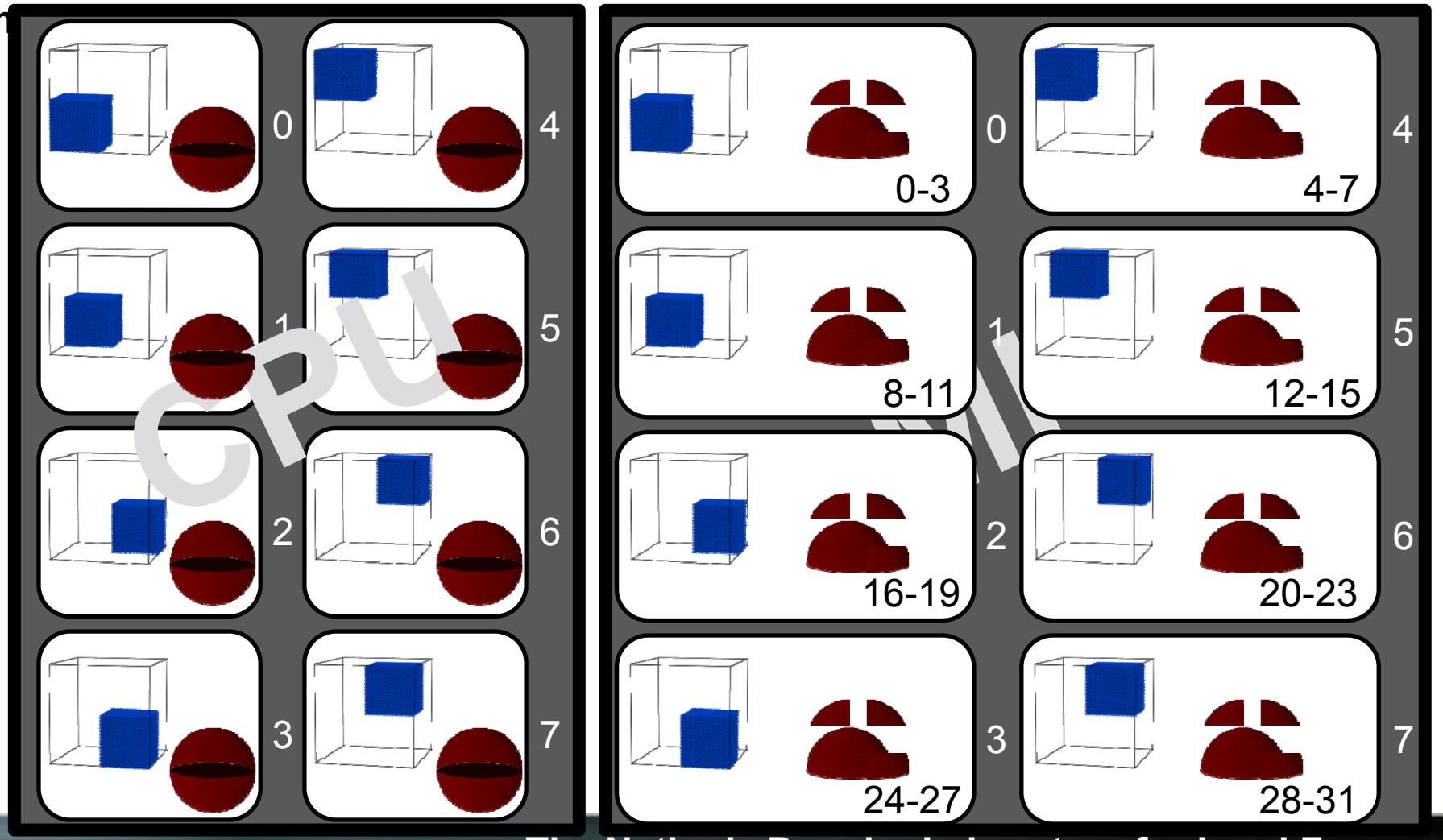


Preventative for MD computations



Using MIC accelerators in addition to CPU

- **Offloading a percentage of the computation for concurrent computation of in**



compute ID group-ID xrd lambda type1 type2 ... typeN keyword value ...

ID / group-ID are documented in compute command

xrd	Style name of this compute command
lambda	Wavelength of incident radiation (length units)
type1 type2 ... typeN	Chemical symbol of each atom type

Keywords

zero or more keyword/value pairs may be appended keyword = **2Theta** , **c** , **LP** , **manual** , or , **echo**

2Theta value	*Min2Theta Max2Theta* (Minimum and maximum 2 theta range to explore in either degrees or radians)
c values	c_1 c_2 c_3 (Parameters to adjust the spacing of the reciprocal lattice nodes in the h, k, and l directions respectively)
LP value	*1 or 0* (Switch to apply Lorentz-polarization factor)
manual	flag to use manual spacing of reciprocal lattice points based on the values of the **c** parameters
echo	flag to provide extra output for debugging purposes

Examples:

compute 1 all xrd 1.541838 B O 2Theta 0.087 0.87 c 1 1 1 LP 1 echo

compute 2 all xrd 1.541838 C O 2Theta 10.0 100.0 c 0.05 0.05 0.05 LP 1 manual

2Theta value	*Min2Theta Max2Theta* (Minimum and maximum 2 theta range to explore in either degrees or radians)
c values	c_1 c_2 c_3 (Parameters to adjust the spacing of the reciprocal lattice nodes in the h, k, and l directions respectively)
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2Theta values can be inputted as radians or degrees. The output value will match the style of the input.

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LP 1 will turn on Lorentz-polarization factor and apply a scaling to the diffraction intensity.

2Theta value	*Min2Theta Max2Theta* (Minimum and maximum 2 theta range to explore in either degrees or radians)
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compute ID group-ID saed lambda type1 type2 ... typeN keyword value ...

ID / group-ID are documented in compute command

saed	Style name of this compute command
lambda	Wavelength of incident radiation (length units)
type1 type2 ... typeN	Chemical symbol of each atom type

Keywords

zero or more keyword/value pairs may be appended keyword = **Kmax** , **Zone** , **dR_Ewald** , **c** , **manual** , **echo**

Kmax value	Maximum distance explored from reciprocal space origin (inverse length units)
Zone values	z_1 z_2 z_3 (Zone axis of incident radiation) If z_1=z_2=z_3=0 all reciprocal space will be meshed up to **Kmax**
dR_Ewald value	Thickness of Ewald sphere slice intercepting reciprocal space (inverse length units)
c values	*c1 c2 c3* (Parameters to adjust the spacing of the reciprocal lattice nodes in the h, k, and l directions respectively)
manual	flag to use manual spacing of reciprocal lattice points based on the values of the **c** parameters
echo	flag to provide extra output for debugging purposes

Examples:

compute 1 all saed 0.0251 C H N O Kmax 1.70 Zone 0 0 1 dR_Ewald 0.01 c 0.5 0.5 0.5

compute 2 all saed 0.0251 Al O Kmax 1.70 Zone 0 0 0 c 0.05 0.05 0.05 manual echo

Kmax value	Maximum distance explored from reciprocal space origin (inverse length units)
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Focusing the calculation on a particular slice of reciprocal space will reduce the computational costs.

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compute 2 all saed 0.0251 Al O Kmax 1.70 Zone 0 0 0 c 0.05 0.05 0.05 manual echo

Focusing the calculation on a particular slice of reciprocal space will reduce the computational costs.

When using the manual flag, the **c** values define the spacing used in reciprocal space. This allows precise control of the mesh.

Kmax value	Maximum distance explored from reciprocal space origin (inverse length units)
Zone values	z_1 z_2 z_3 (Zone axis of incident radiation) If z_1=z_2=z_3=0 all reciprocal space will be meshed up to **Kmax**
dR_Ewald value	Thickness of Ewald sphere slice intercepting reciprocal space (inverse length units)
c values	*c1 c2 c3* (Parameters to adjust the spacing of the reciprocal lattice nodes in the h, k, and l directions respectively)
manual	flag to use manual spacing of reciprocal lattice points based on the values of the **c** parameters
echo	flag to provide extra output for debugging purposes

fix ID group-ID ave/histo/weights Nevery Nrepeat Nfreq lo hi Nbin value1 value2 ... keyword args …

ID / group-ID are documented in compute command

ave/histo/weights	Style name of this fix command
Nevery	Use input values every this many timesteps
Nrepeat	# of times to use input values for calculating averages
Nfreq	calculate averages every this many timesteps
lo,hi	low/high bounds within which to histogram
Nbin	# of histogram bins
Value1	parameter over which the histogram is created

Keywords

Weights *value2* parameter (same size as Value1) to weight histogram

Note, this has the same functionality as fix ave/histo but has the special keyword weights to compute a weighted histogram.

fix ID group-ID saed/vtk Nevery Nrepeat Nfreq c_ID.. keyword args ...

ID / group-ID are documented in compute command

saed/vtk	Style name of this fix command
Nevery	Use input values every this many timesteps
Nrepeat	# of times to use input values for calculating averages
Nfreq	calculate averages every this many timesteps
c_ID	saed compute ID

Keywords

zero or more keyword/value pairs may be appended keyword = **file**, **ave**, **start** , **file**, or **overwrite**

Note, this has the same functionality as fix ave/time but is specially modified for compute saed to output into the 3rd generation vtk image data format for use in parallelized visualization software (i.e., Paraview or Visit).

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