# predicting phase transition mechanisms by integrating atomistic simulations, experiments, and AI techniques
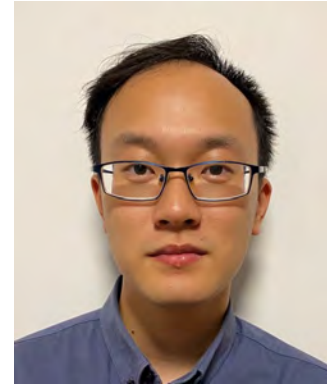


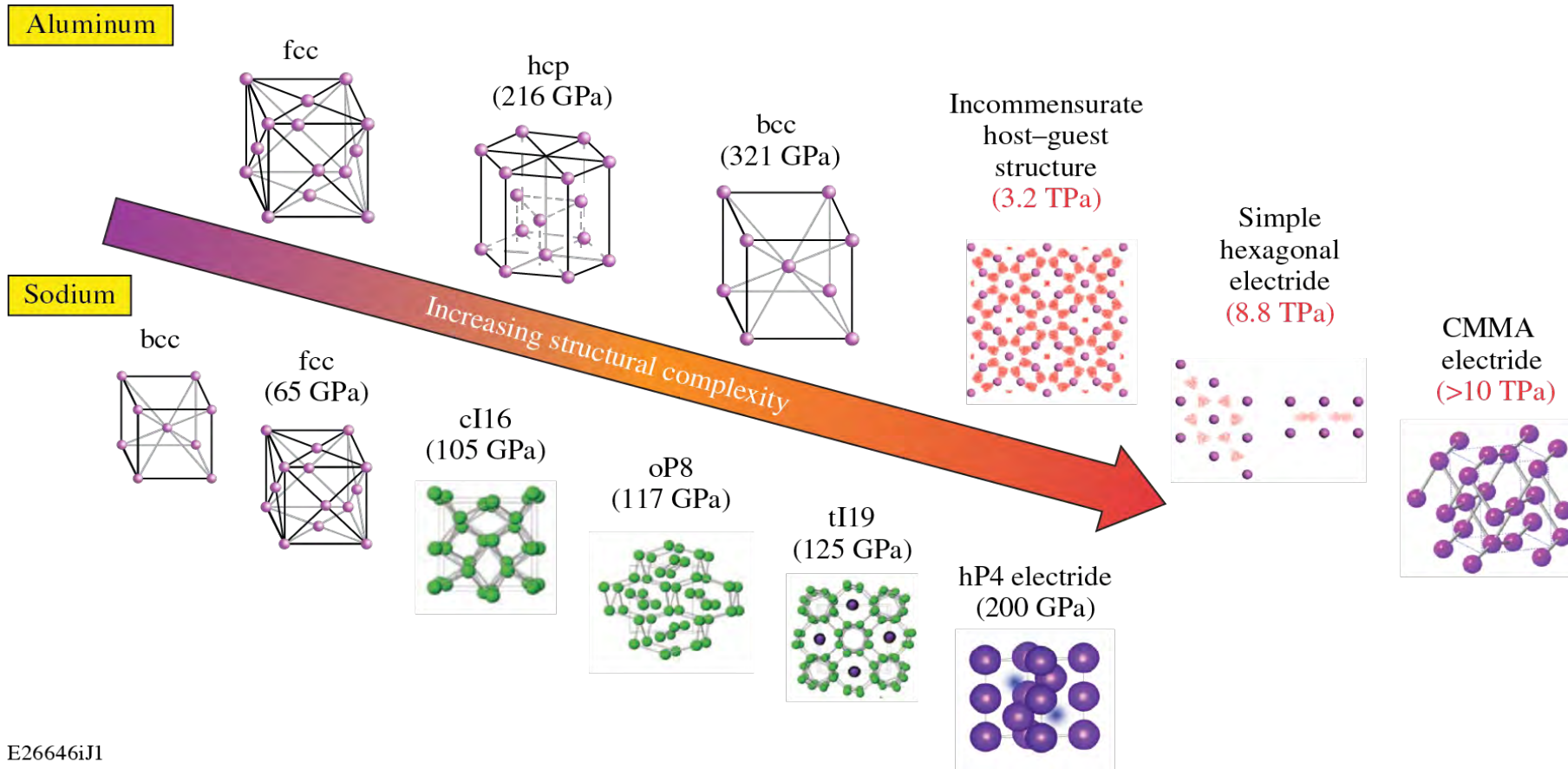Lijie He, PhD    Haomin Liu, PhD    Ali Shargh, PhD    Zhaotong Du, MS    Jerardo Salgado, PhD
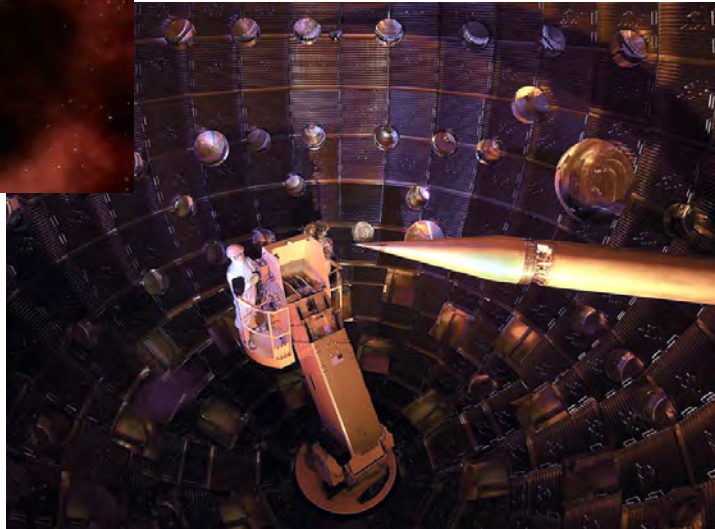
Niaz Abdolrahim
LLNL, 2022

**UNIVERSITY of ROCHESTER**

# Materials at High-Energy-Density (HED)



Aluminum

fcc

hcp
(216 GPa)

bcc
(321 GPa)

Incommensurate
host–guest
structure
(3.2 TPa)

Simple
hexagonal
electride
(8.8 TPa)

CMMA
electride
(>10 TPa)

Increasing structural complexity

Sodium

bcc

fcc
(65 GPa)

cI16
(105 GPa)

oP8
(117 GPa)

tI19
(125 GPa)

hP4 electride
(200 GPa)

E26646iJ1

# Understanding materials at HED



How are planets form

Quest for room temperature superconductivity

*Room-temperature superconductivity*
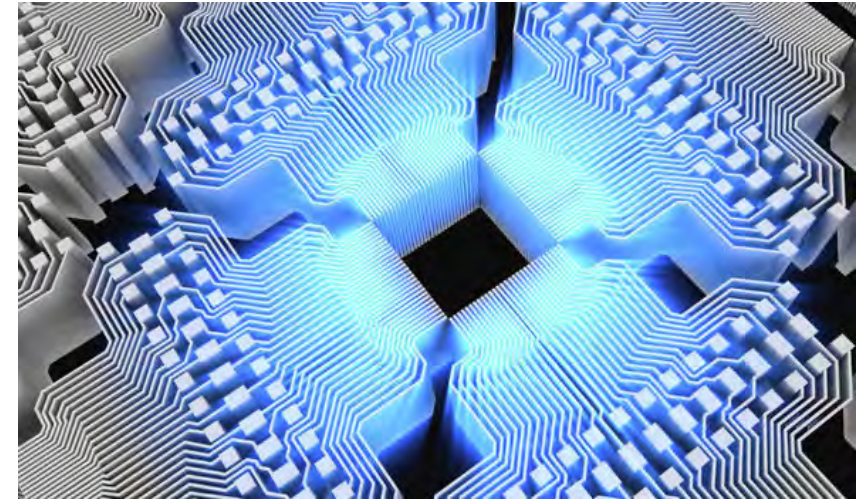
Cheap energy created from fusion and delivered over superconducting wires

National Ignition Facility experiment puts researchers at threshold of fusion ignition

Superfast quantum computers

IBM supercomputer

# HED capabilities in the U.S. and worldwide.

| Facility/End Station | Type of Machine | Energy Delivered | Peak Power | Repetition Rate | Maximum Pressures | Key physics goals |
|---|---|---|---|---|---|---|
| National Ignition Facility | Laser | 1.8-MJ UV photons | 500 TW | ~1 shot/ 3 h | to 10 TPa | New physics of H and simple elements to atomic pressure |
| Z | Pulsed power | 3.5-MJ current | 350 TW/ 26 MA | ~1 shot/ day | 0.5 TPa | Elements and matter beyond Thomas–Fermi |
| OMEGA/ OMEGA EP | Lasers | 30-kJ UV | 30 TW | ~1 shot/ 3 h | 1 TPa | New techniques for dense H and matter beyond Thomas–Fermi |
| Matter at Extreme End Station (LCLS) | X-ray laser | 1-mJ x rays + 50-J laser | 10 GW | 120 Hz | 0.3 TPa | Unwrapping complex structures and kinetic pathways to complexity |
| Dynamic Compression Sector (APS) | Laser, guns, x ray | 1-$\mu$J x rays + 100-J laser | 10 MW | 120 Hz | 0.3 TPa | Unwrapping complex structures for mid-to-high $Z$ |
| Laser Net | Lasers | to 20 kJ | to 30 TW | Several | 1 TPa | Develop structure and electronic property techniques |
| Diamond-Anvil Cell Facilities (including APS and BNL) | Static compression x ray, THz | NA | NA | Continuous | 0.5 TPa | Understand kinetic thresholds and constrain ground states |

To explore the multiscale nature of matter from atomic to the macro-scale, connecting experimental observations with atomistic simulations and deep learning computer vision techniques to answer key question of how atomic rearrangement through defect motion enables the bulk phase transformations in extreme solids
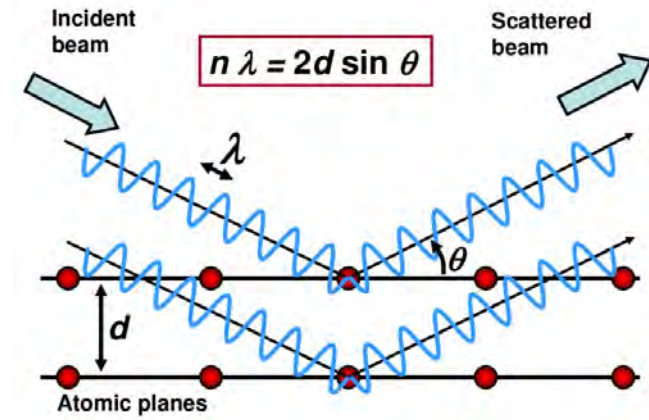
# Outline

- Phase transformation of Aluminum at high pressure (XRD analysis)

- Phase identification of water at ambient temperature (Raman, IR, and XRD analysis)

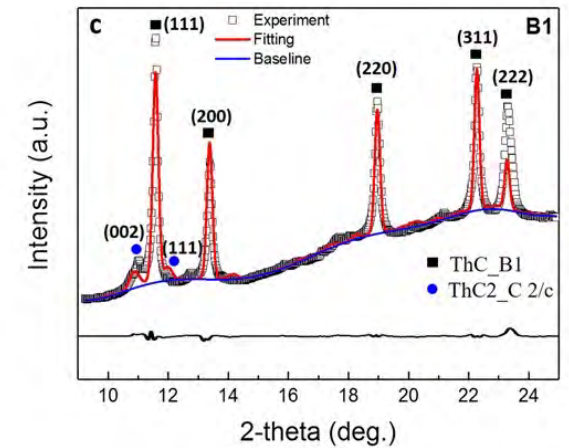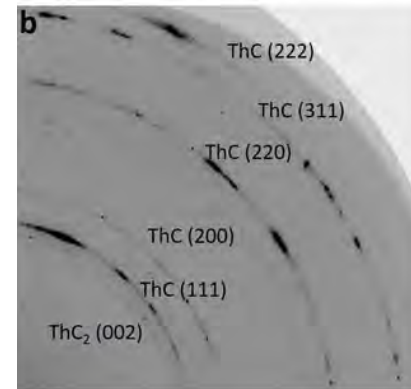- Developing AI-powered models for classification of large XRD data

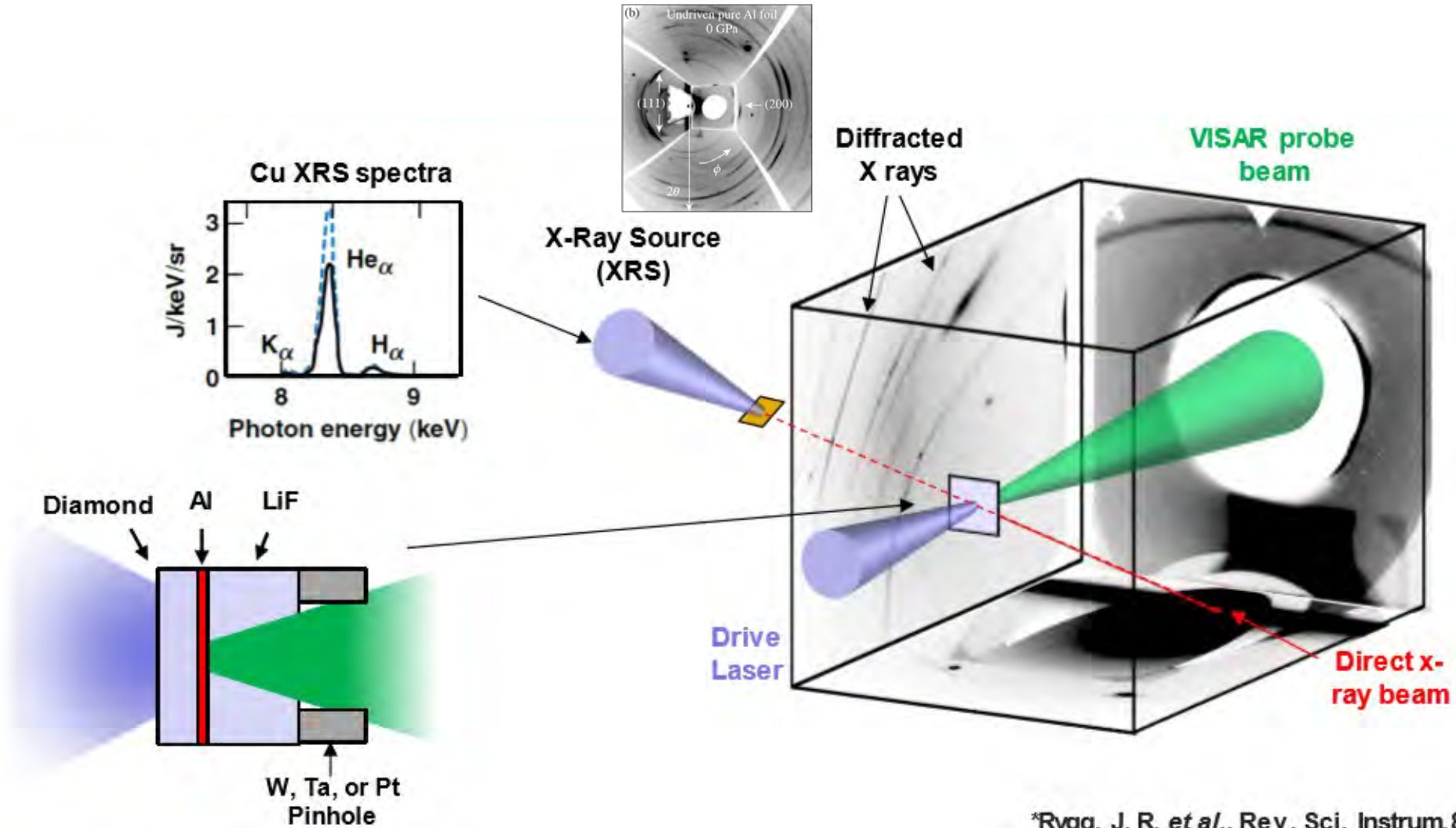# Crystal structure diagnostics
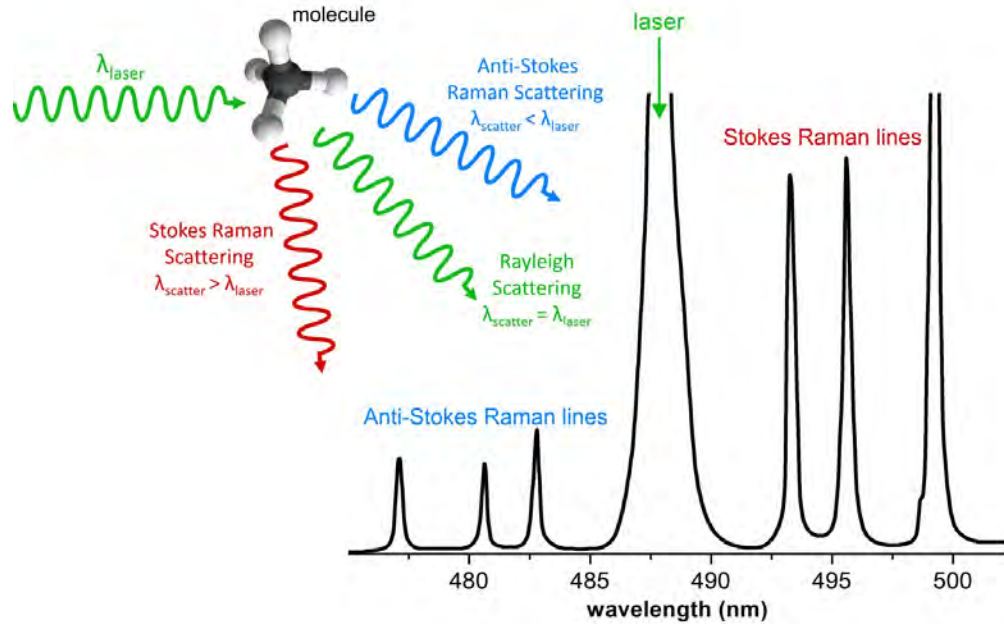


Principles of Raman spectroscopy



Principles of X-ray diffraction

$$n \lambda = 2d \sin \theta$$



XRD of Thorium monocarbide

UNIVERSITY of ROCHESTER

# Phase transformation of Aluminum-Ramp compression loading



Cu XRS spectra

X-Ray Source (XRS)

Diffracted X rays

VISAR probe beam

Diamond   Al   LiF

W, Ta, or Pt Pinhole

Drive Laser

Direct x-ray beam

*Rygg, J. R. et al., Rev. Sci. Instrum. 83, 113904 (2012
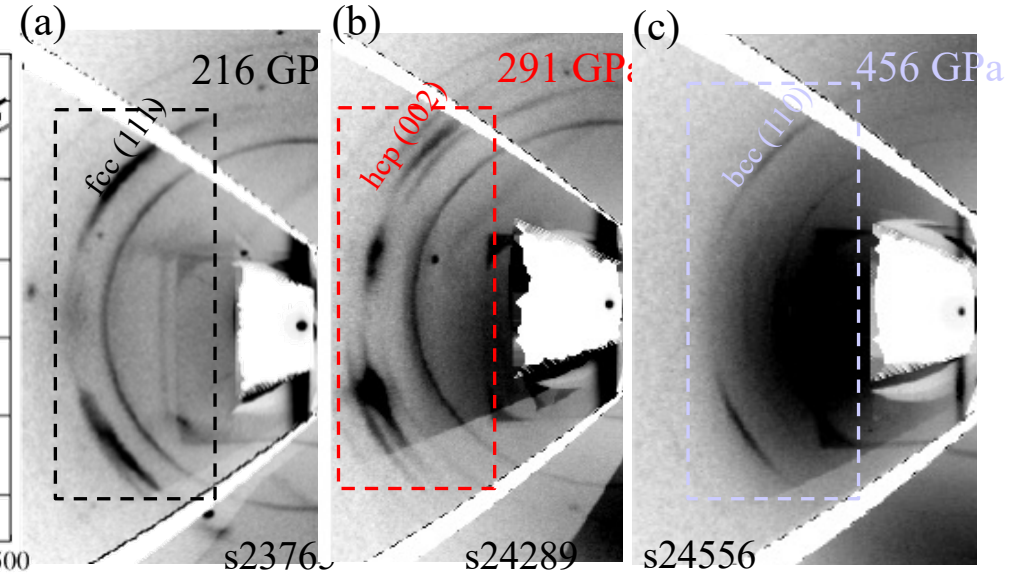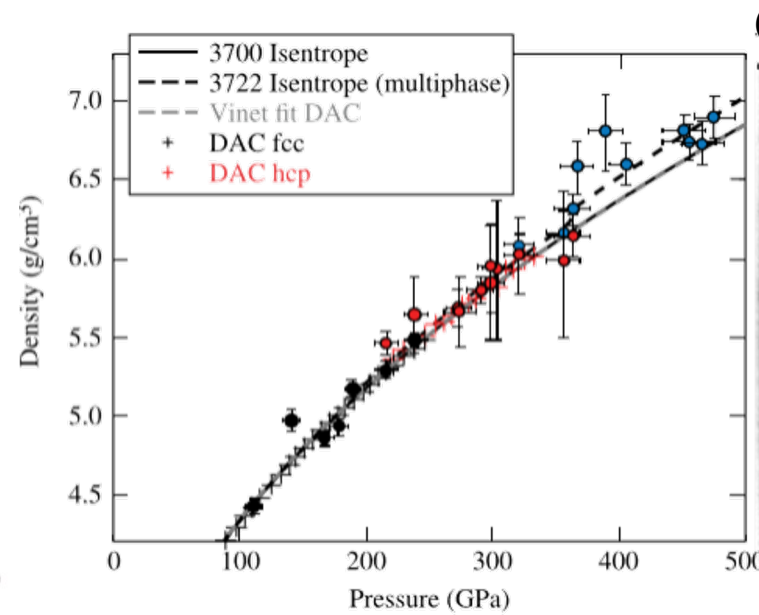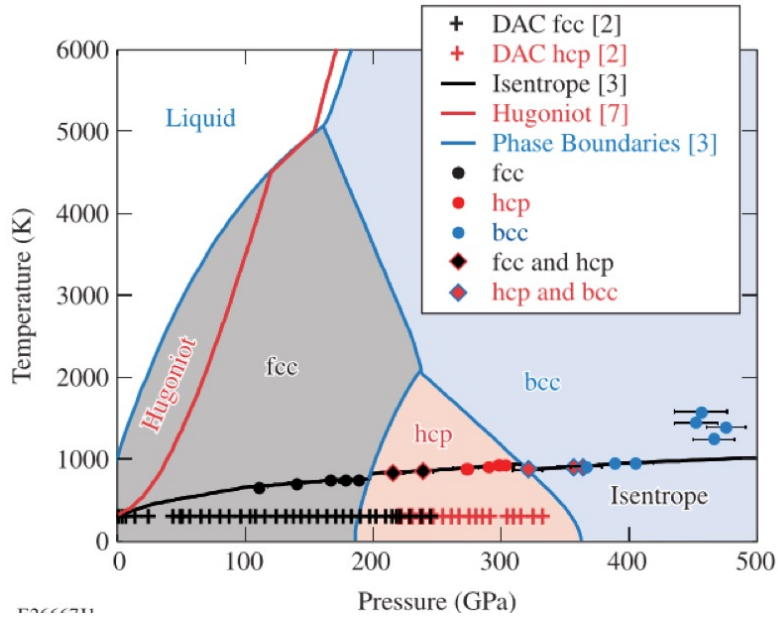
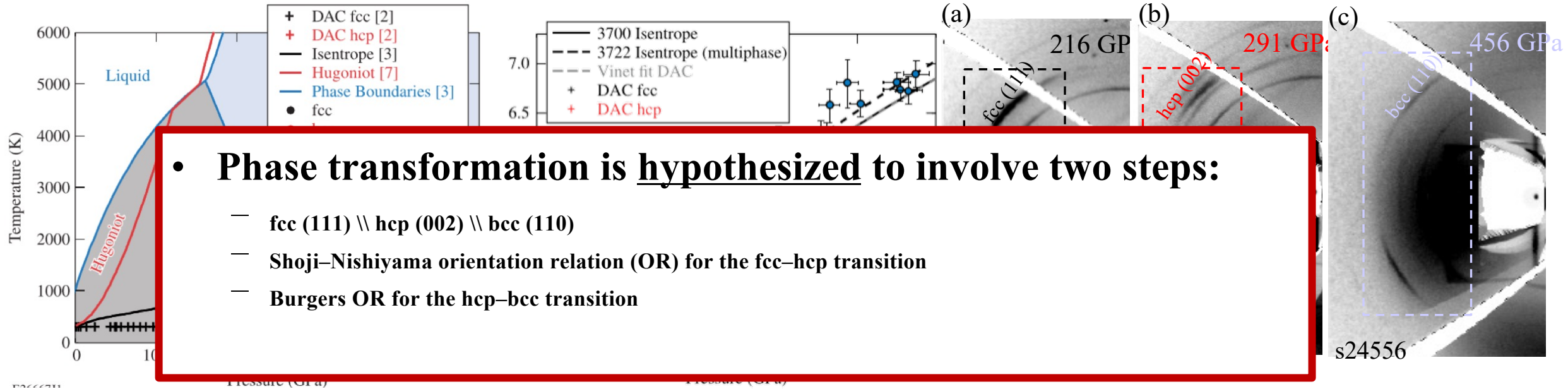# Phase transformation of Aluminum- Experimental observations







*Polsin. D. N. et al., Phys. Plasmas 25, 082709 (2018)

- Stress-density response follows Al isentrope

- In situ XRD show fcc-hcp-bcc phase transformation

# Phase transformation of Aluminum- Experimental observations



**216 GPa**    fcc (111)

**291 GPa**    hcp (002)

**456 GPa**    bcc (110)

s24556

- **Phase transformation is <u>hypothesized</u> to involve two steps:**
  - **fcc (111) \\ hcp (002) \\ bcc (110)**
  - **Shoji–Nishiyama orientation relation (OR) for the fcc–hcp transition**
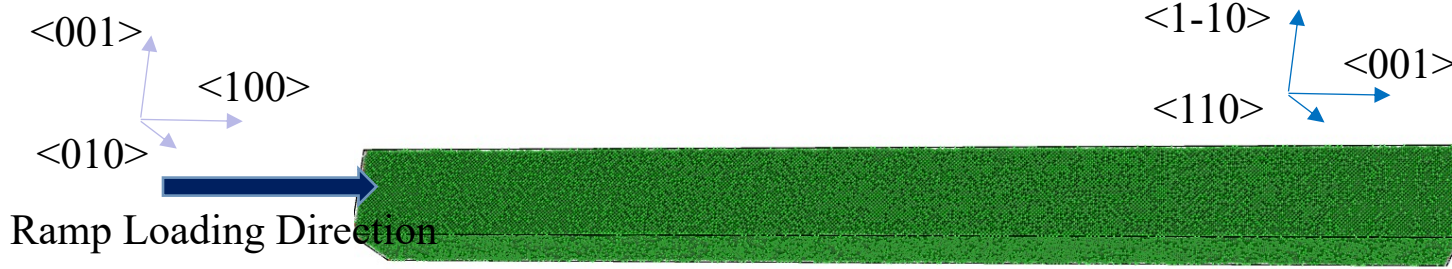  - **Burgers OR for the hcp–bcc transition**

*Polsin. D. N. et al., Phys. Plasmas 25, 082709 (2018)

- Stress-density response follows Al isentrope
- In situ XRD show fcc-hcp-bcc phase transformation

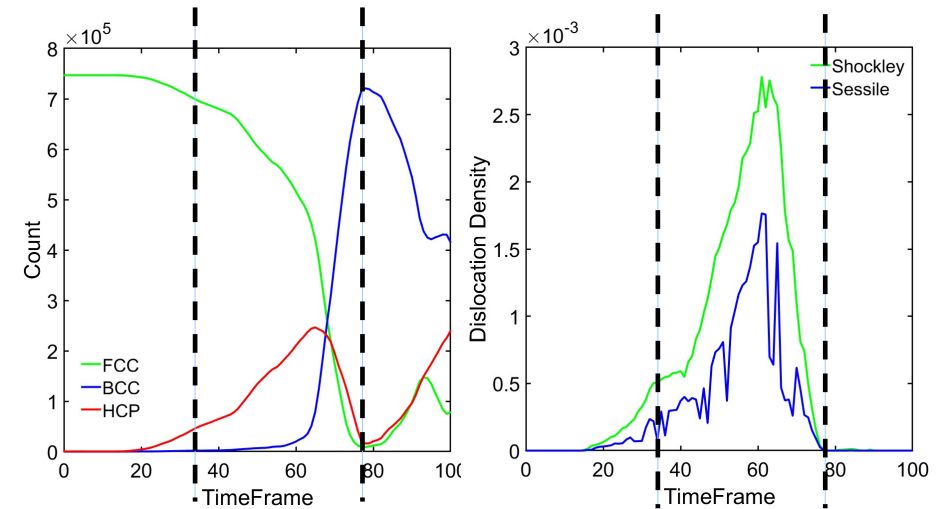# Phase transformation of Aluminum- MD simulations of ramp compression

<001>
<100>
<010>

<1-10>
<001>
<110>

Ramp Loading Direction

Right boundary is a momentum mirror, which reflect when they attempt to move through the wall.

Left boundary is the ramp, which accelerate from 0 – 6 km/s.

FCC
HCP
BCC

fcc hidden, only hcp and bcc atoms displayed
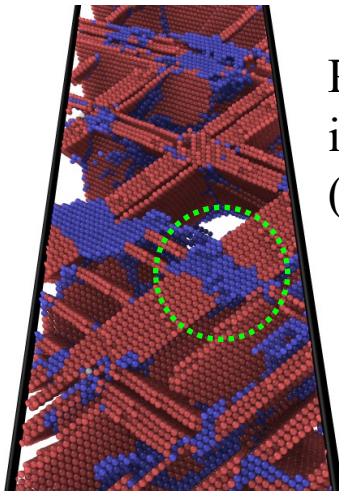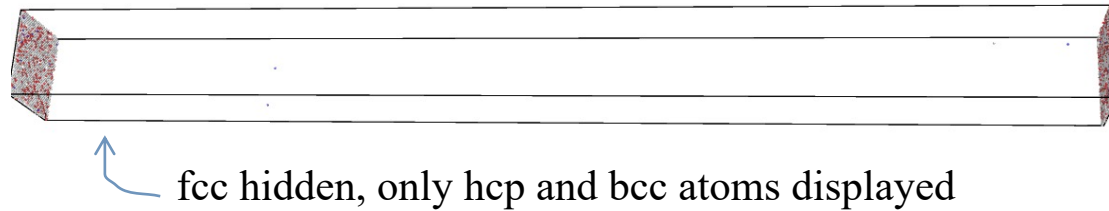
Phase transformation nucleation happens at the intersection of two stacking fault planes.
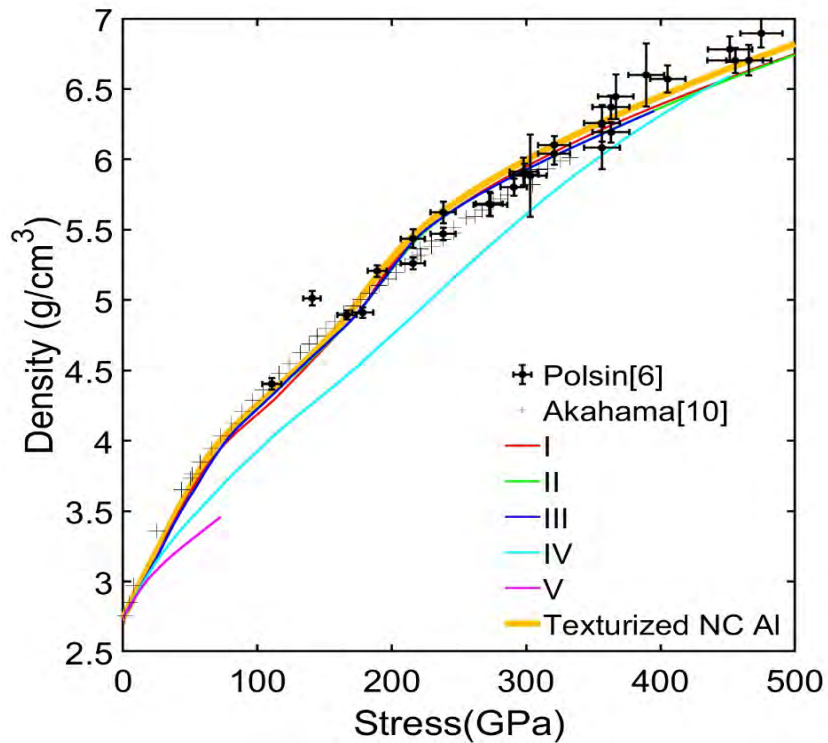(**Dislocation-assisted phase transformation**)

Dislocation density grows significantly at the time when major phase transformation happens

# Phase transformation of Aluminum- MD simulations of ramp compression

$$x_s(t_s) = \frac{1}{M} x(t), \qquad t_s = \frac{1}{M} t, \qquad v_s(t_s) = \frac{\partial x_s}{\partial t_s} = \frac{\frac{1}{M}\partial x}{\frac{1}{M}\partial t} = \frac{\partial x}{\partial t} = v(t) \qquad \frac{F_s}{F} = \frac{m_s(L_s/T_s^2)}{m(L/T^2)} = \left(\frac{\rho_s}{\rho}\right)\left(\frac{L_s}{L}\right)^2 \left(\frac{T_s}{T}\right)^{-2} = 1 \qquad \check{v}_p = \frac{v_t L}{\tau C_0^2}$$



| Setup | Length(nm) | Duration(ps) | Length Scale Factor | Time Scale Factor | Atom Count | Dimensionless strain rate |
|-------|-----------|--------------|---------------------|-------------------|------------|---------------------------|
| Polsin (Experiment) | 20000 | 10000 | 1 | 1 | N/A | 0.306 |
| I | 100 | 50 | 1/200 | 1/200 | $6.25 \times 10^5$ | 0.306 |
| II | 1000 | 500 | 1/20 | 1/20 | $6.25 \times 10^6$ | 0.306 |
| III | 2000 | 1000 | 1/10 | 1/10 | $1.25 \times 10^7$ | 0.306 |
| IV | 1000 | 250 | 1/20 | 1/40 | $6.25 \times 10^6$ | 0.612 |
| V | 2000 | 250 | 1/10 | 1/40 | $1.25 \times 10^7$ | 1.224 |
| Texturized NC | 100 | 50 | 1/200 | 1/200 | $5.63 \times 10^{6*}$ | 0.306 |

# Phase transformation of Aluminum- MD simulations of ramp compression
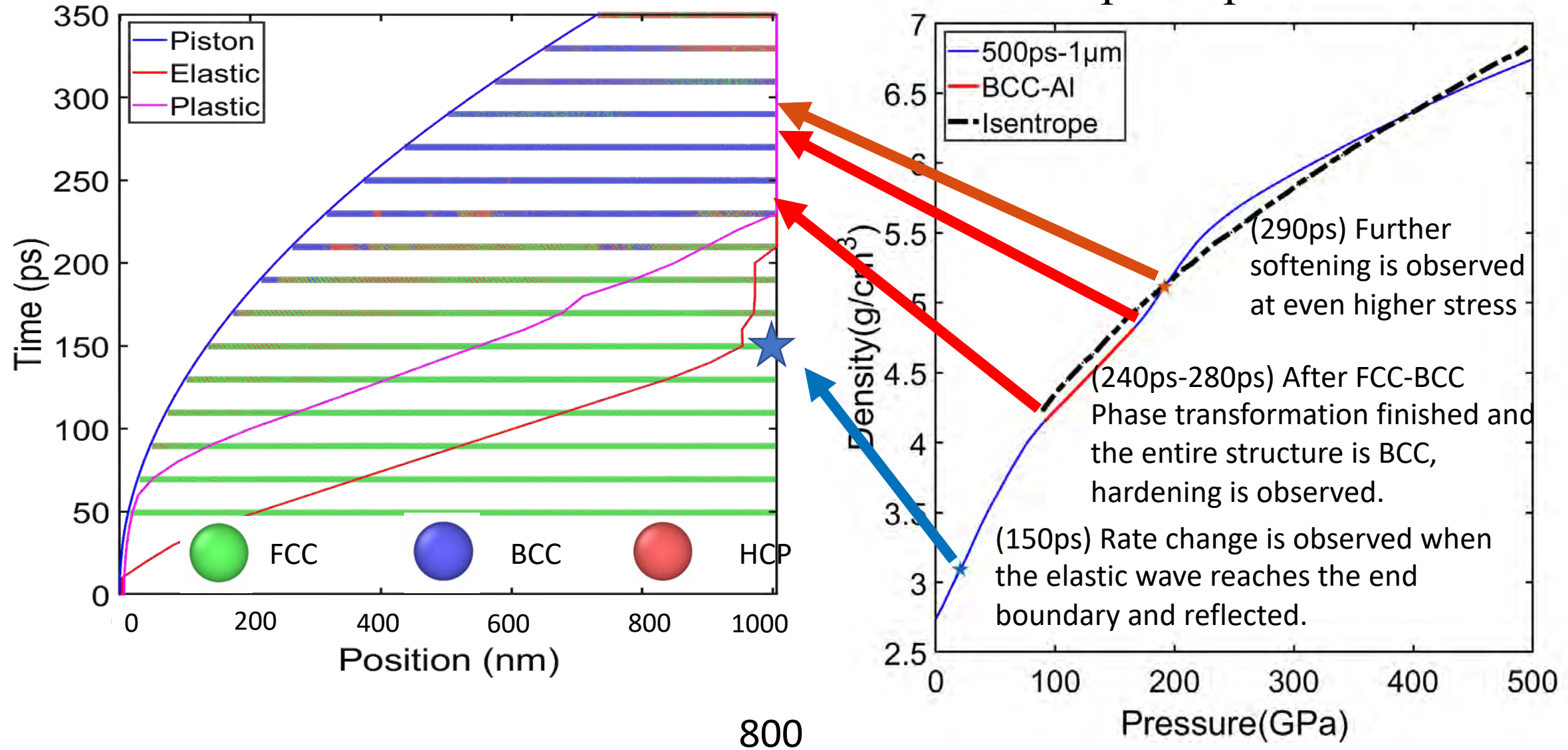
# Phase transformation of Aluminum- MD simulations of ramp compression
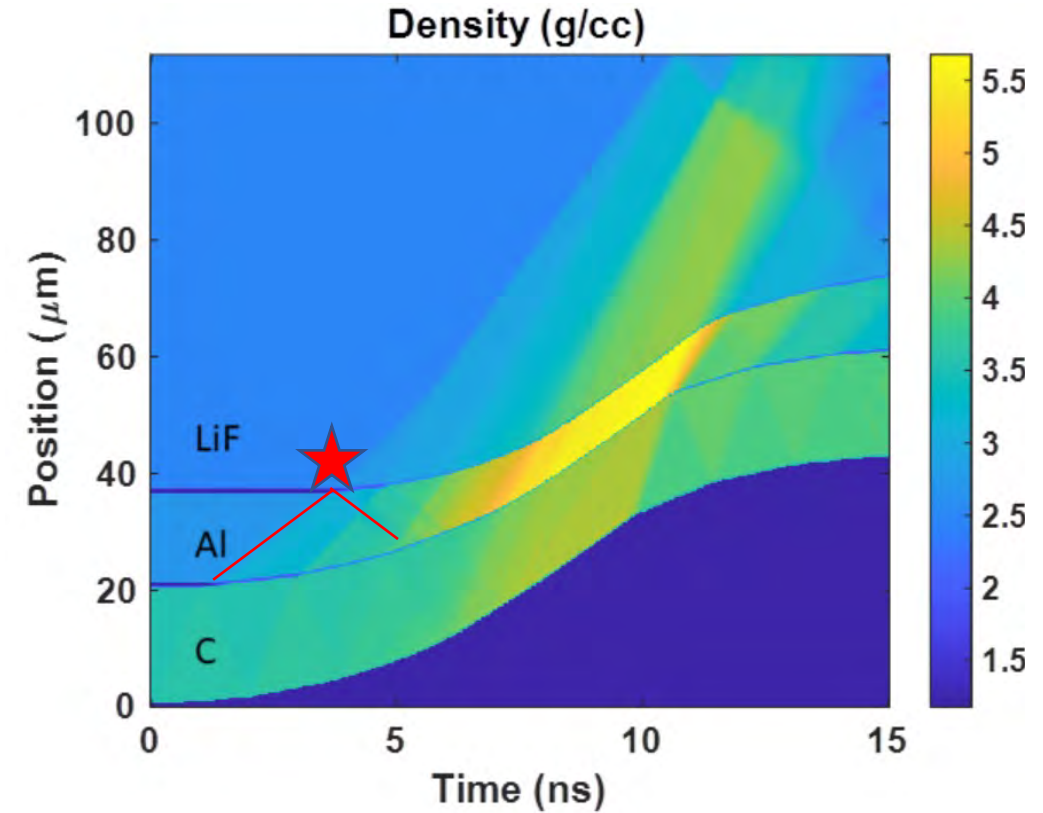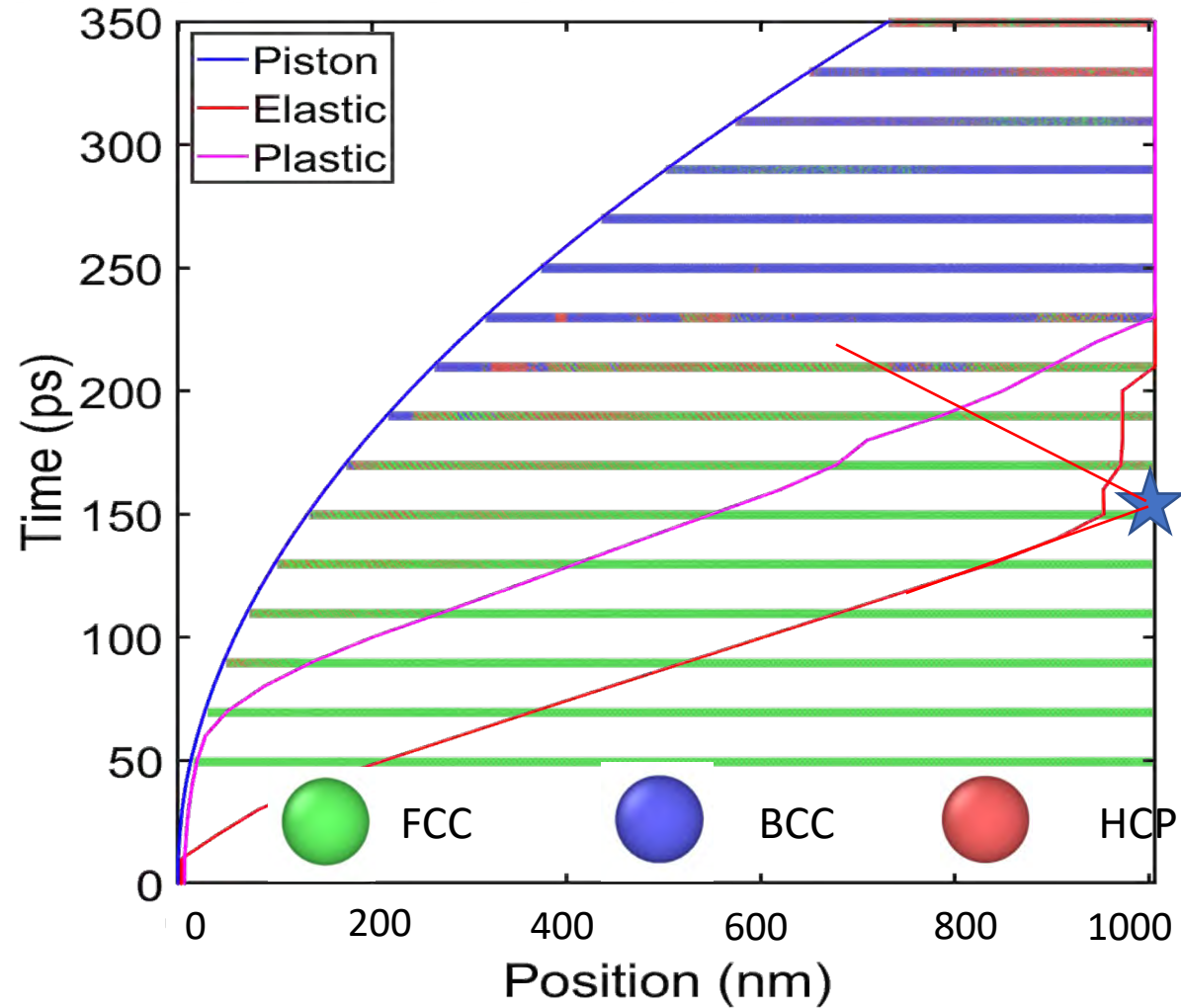
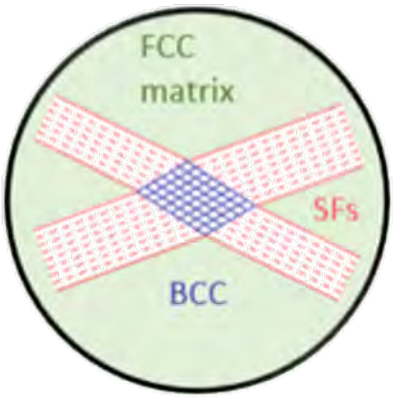

800

Elastic deformation

Micro twin formation on (111)

Stacking faults (SFs) formation on (-111) and unzipp (111) micro twins into SFs.

Phase transformation to bcc imitated at the intersecting edges between (-111) and (111) SFs.
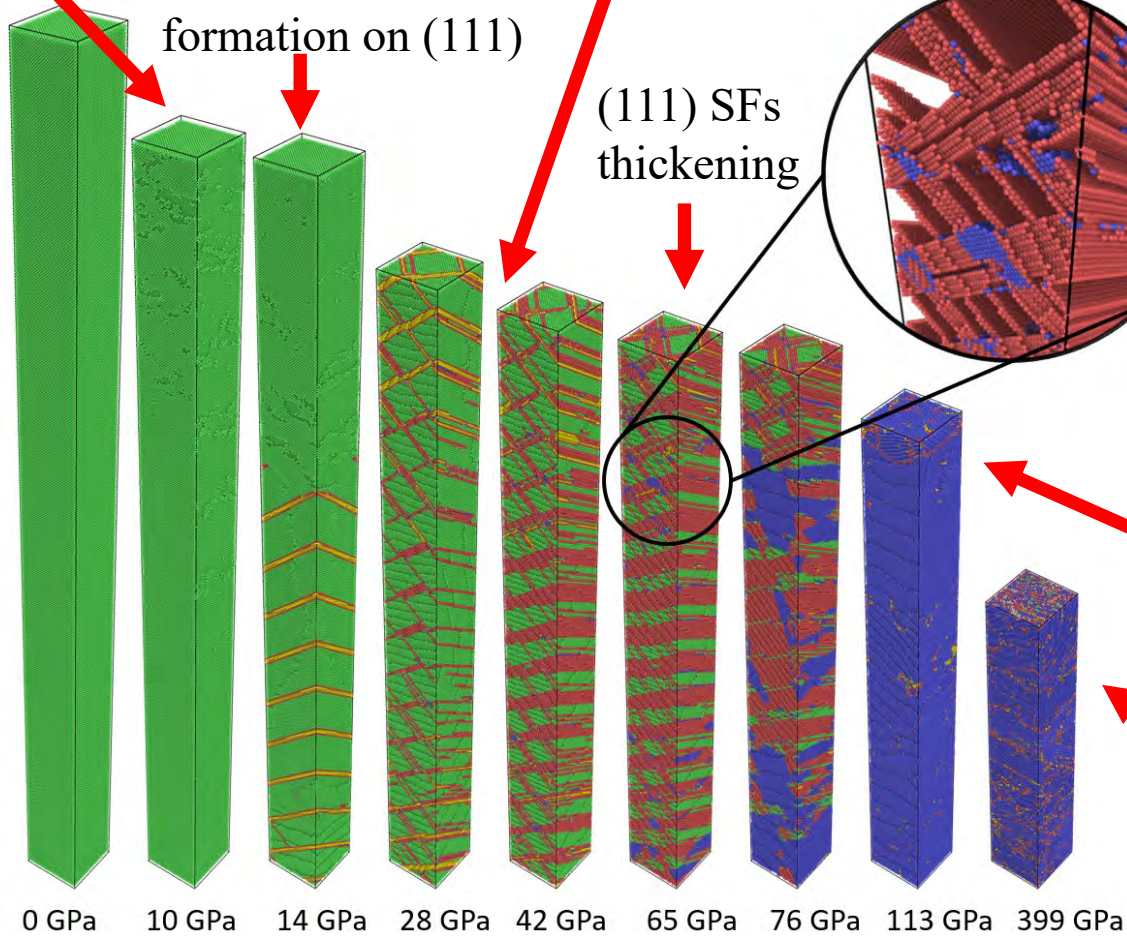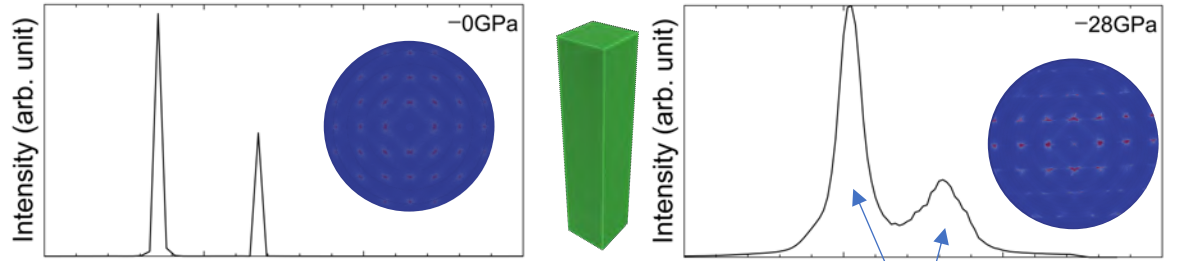
Undeformed perfect fcc Al

(111) SFs thickening

Phase transformation completed, entire structure bcc Al.

Further plasticity in bcc Al.

0 GPa   10 GPa   14 GPa   28 GPa   42 GPa   65 GPa   76 GPa   113 GPa   399 GPa
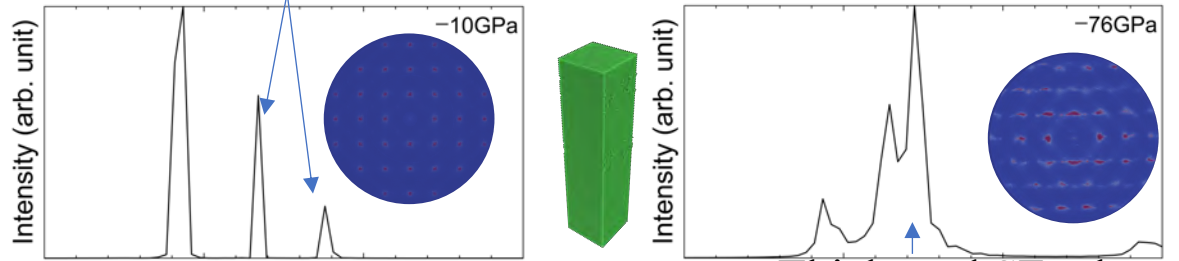
FCC   Twinned-FCC   BCC   HCP

# Phase transformation of Aluminum- Virtual XRD and SAED



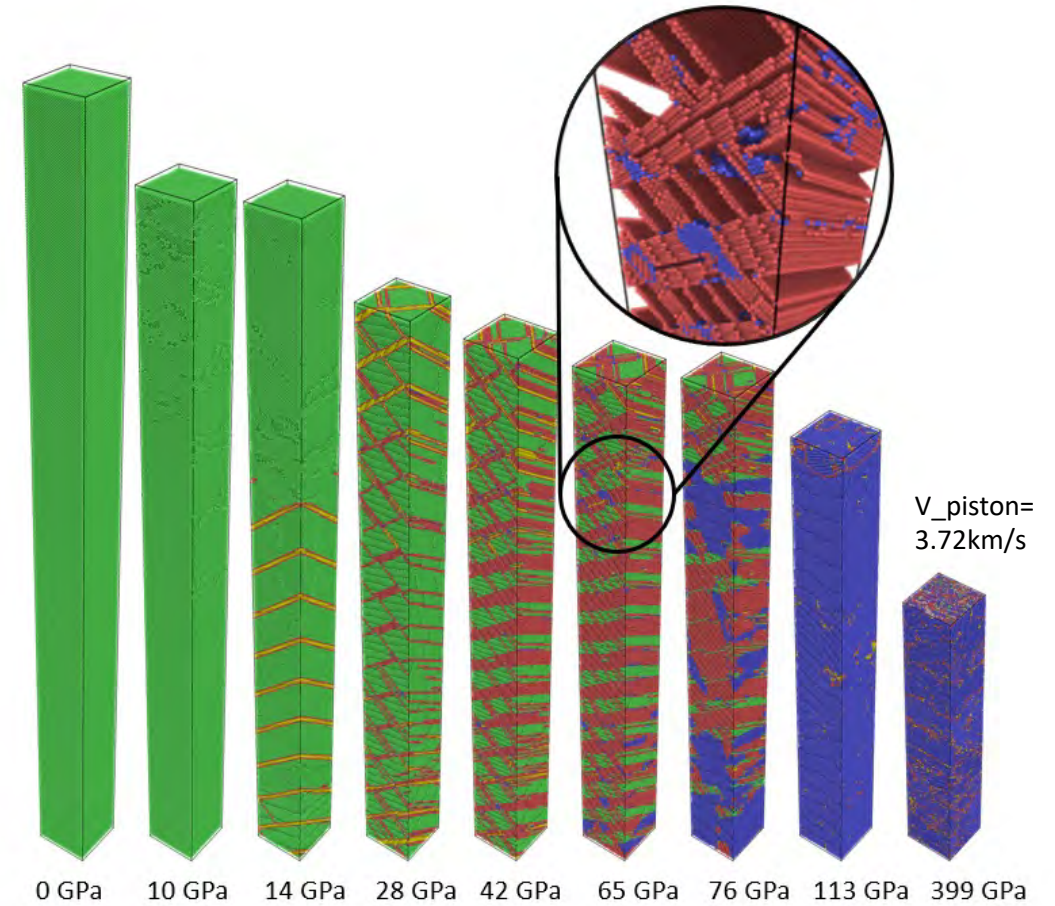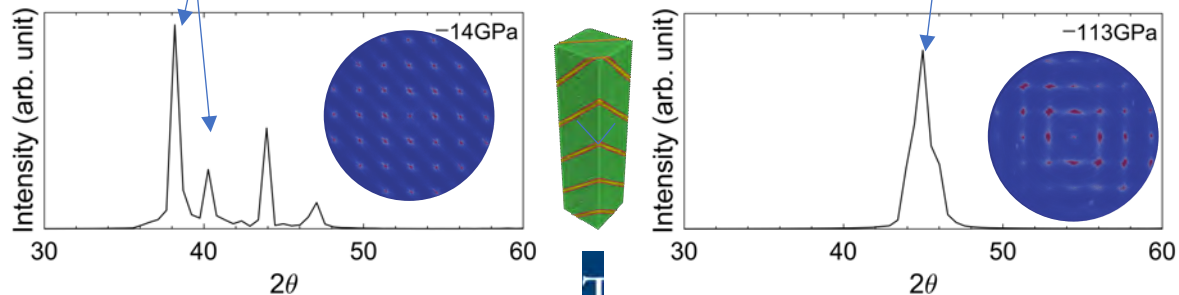{200} splitting due to elastic deformation along loading direction

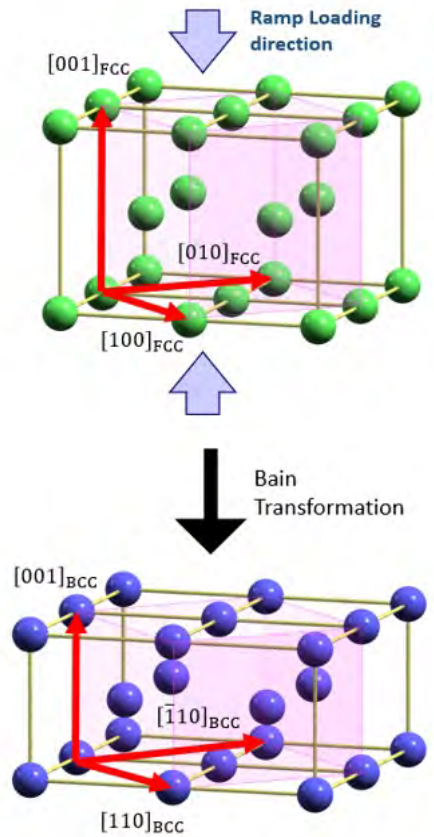Peaks broadening and relative shifting due to SFs

{111} splitting due to inhomogeneous straining caused by micro twin

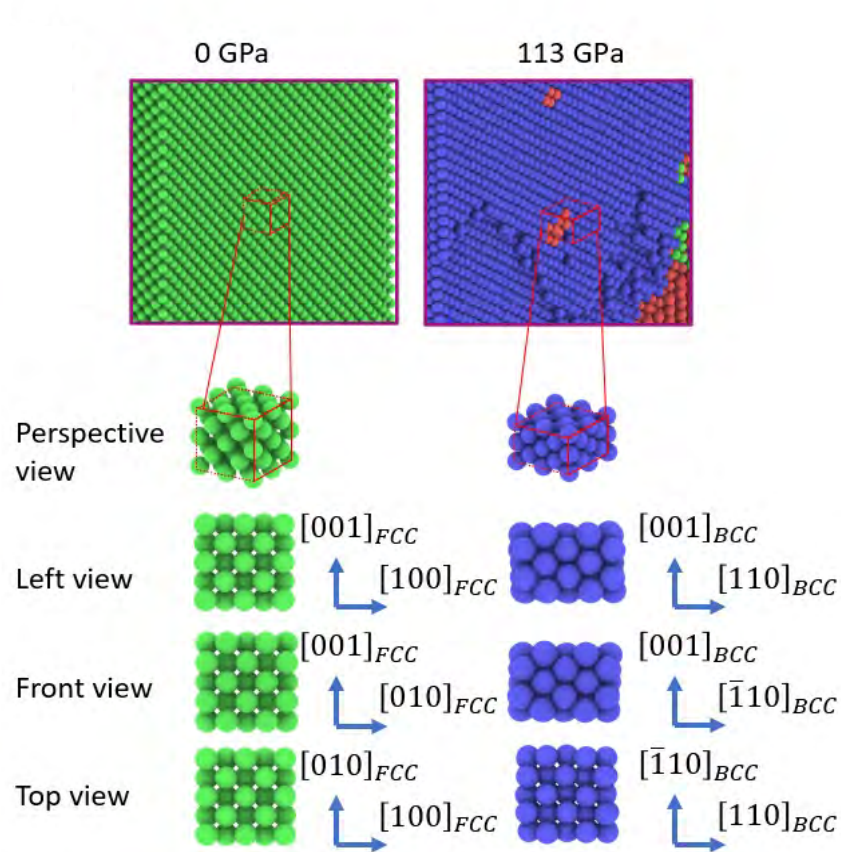Thickened SFs show hcp signature

Transformed structure show bcc signature

V_piston= 3.72km/s

0 GPa    10 GPa    14 GPa    28 GPa    42 GPa    65 GPa    76 GPa    113 GPa    399 GPa
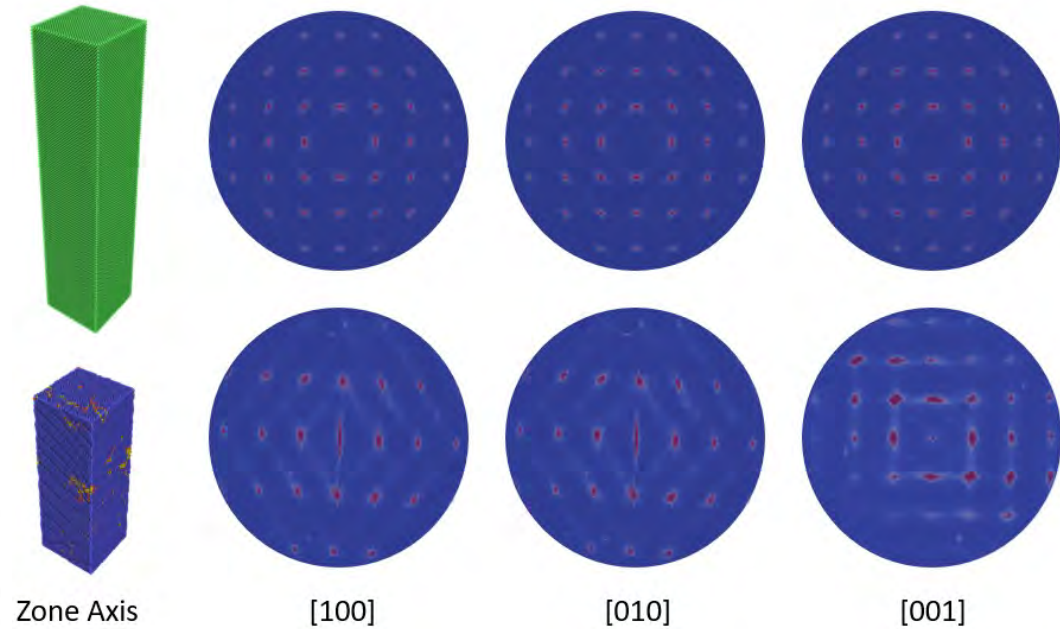
FCC    Twinned-FCC    BCC    HCP

# Phase transformation of Aluminum- Bain transformation
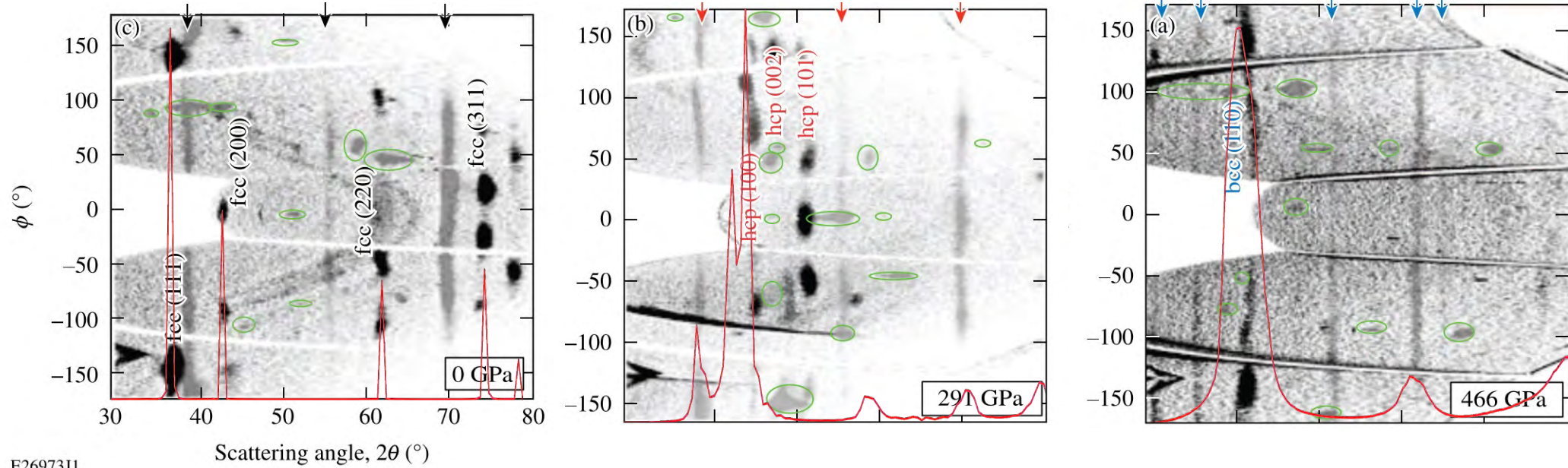
Path illustration

**Polyhedral template matching**
lattice orientation calculation

**Virtual SAED pattern**

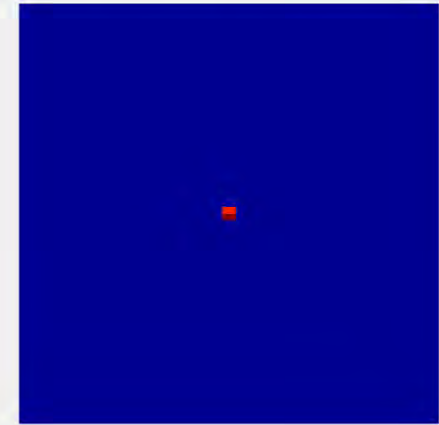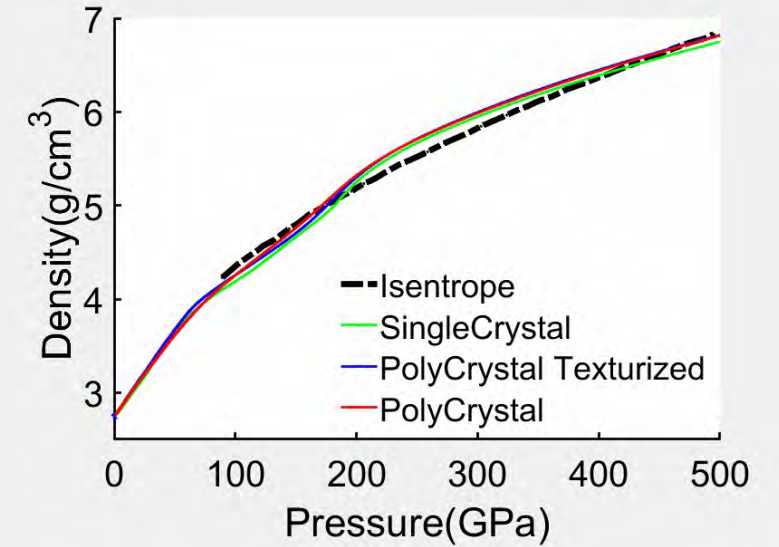# Phase transformation of Aluminum- comparison with experiments



E26973J1

| XRD | Stress at which the 1st figure (fcc) is taken | Stress at which the 2nd figure (hcp) is taken | Stress at which the 3rd figure (bcc) is taken |
|---|---|---|---|
| Experiment | 0 GPa | 291 GPa | 466 GPa |
| Simulation | 0 GPa | 76 GPa | 399 GPa |

*Polsin. D. N. et al., Phys. Plasmas 25, 082709 (2018)

UNIVERSITY *of* ROCHESTER

# Phase transformation of Aluminum- Texturized nanocrystalline (NC) sample

- Average grain size: 15 nm

- [001]-oriented FCC texturized

- Dimension is 30.37 nm × 30.37 nm × 100 nm, and a scaling factor of 1/200 is used
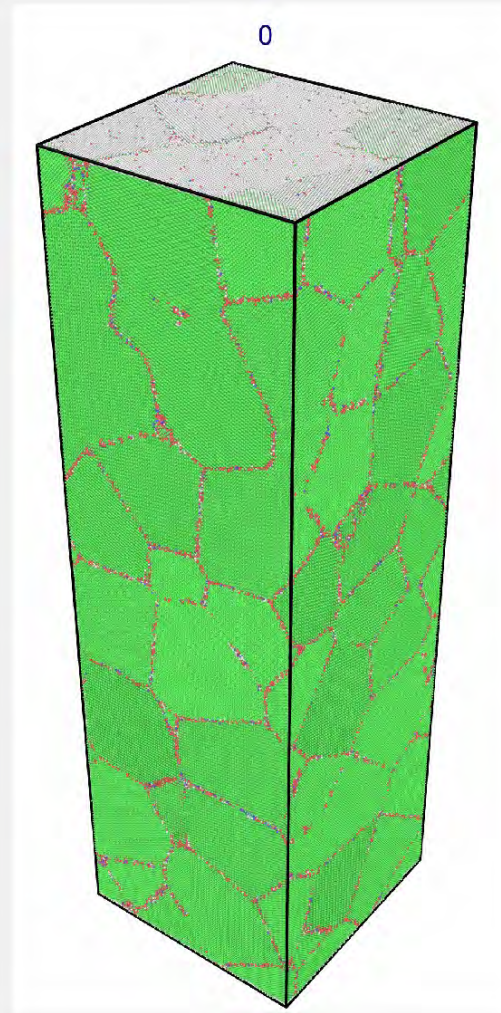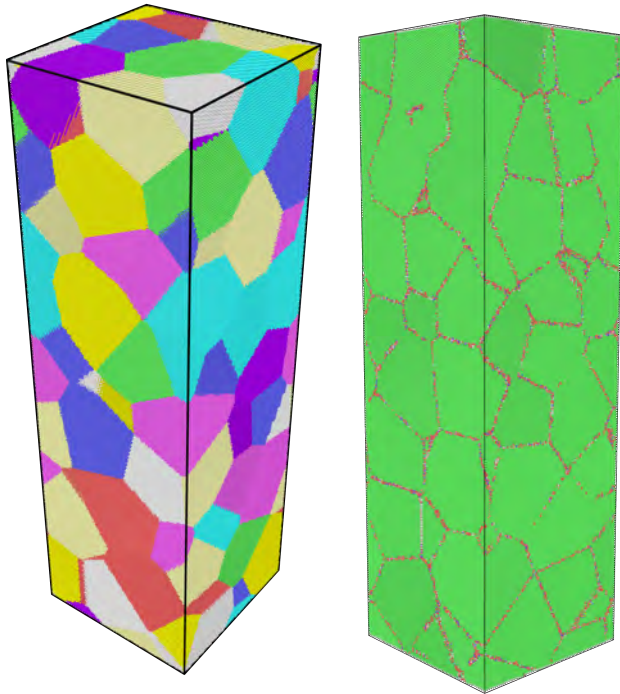
# Phase transformation of Aluminum- Texturized nanocrystalline (NC) sample

# Phase transformation of Aluminum- Dislocation assisted Bain Transformation

**Experiment observation**

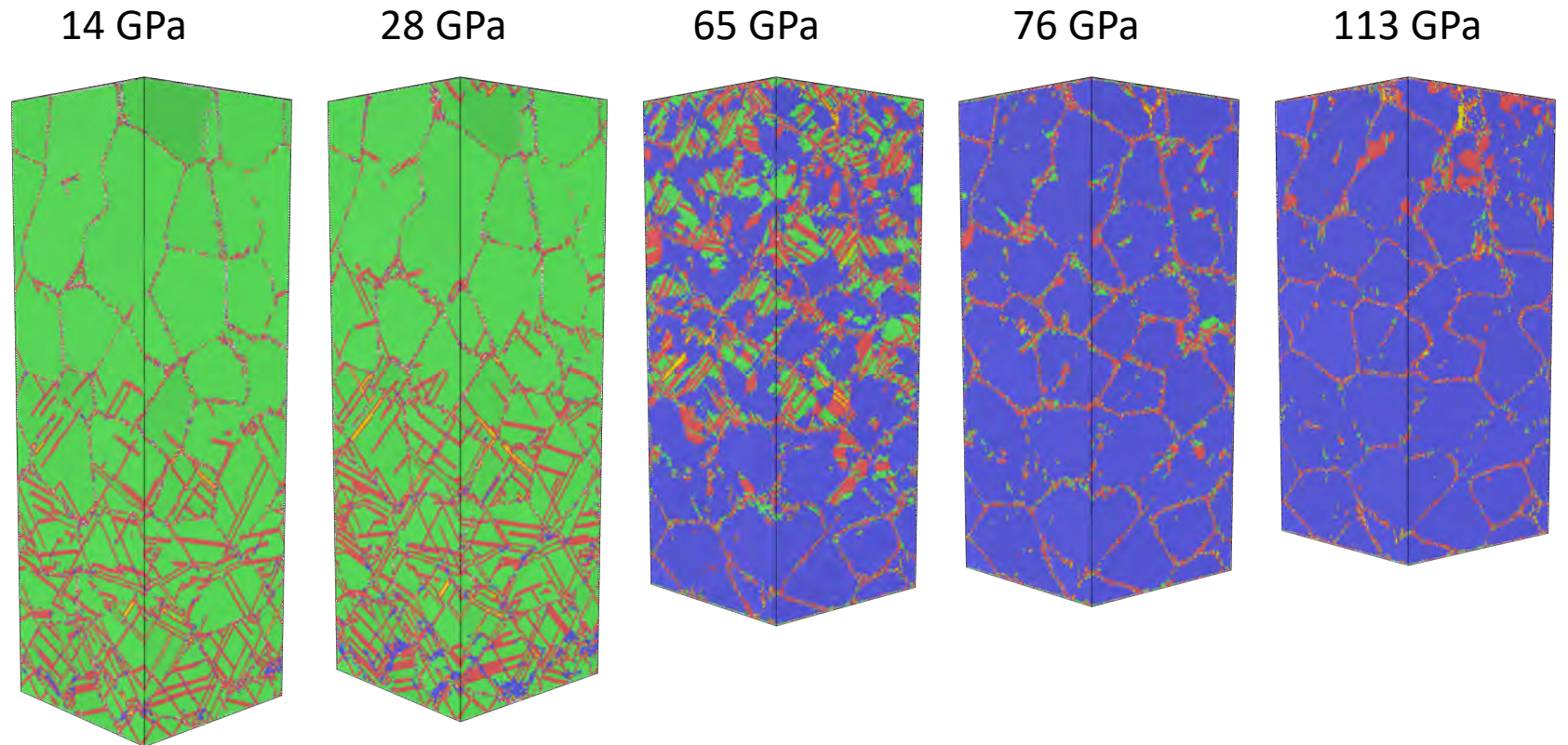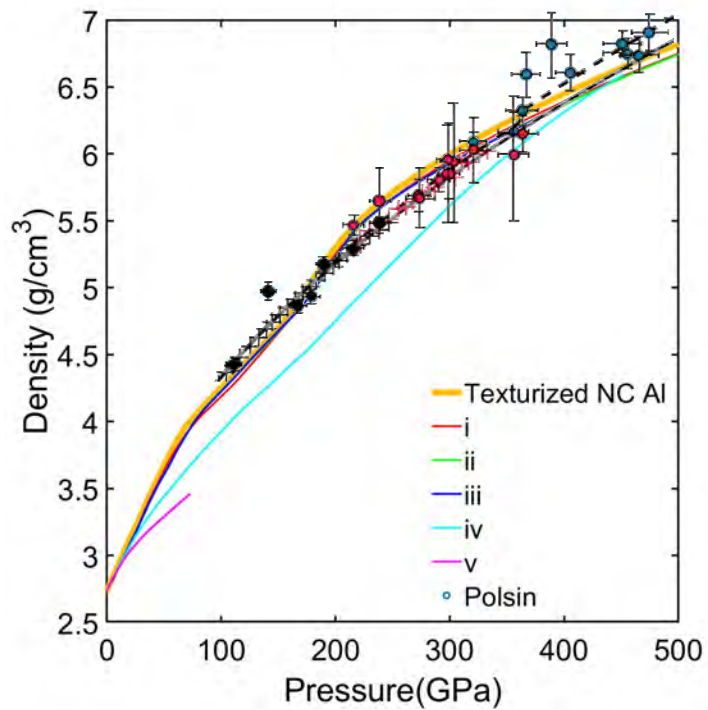- Norm to $(200)_{HCP}$ have angle of 45-50 degrees to the fiber axis

- Norm to $(110)_{BCC}$ have angle of ~50 degrees to the fiber axis

- The lattice constant of bcc is reported ~2.43 at 466 GPa

**Simulation observation**

- For stacking fault, basal plane $(001)_{HCP}//(111)_{FCC}$, therefore normal to the $(002)_{HCP}$ have angle of 54.7 degree to the fiber axis.

- For bcc in the structure, the Bain transformation predicts a 45 degree between the (110) plane and the fiber axis.

- The lattice constant of BCC is 2.33 at 400 GPa.

# Phase transformation of Aluminum- Dislocation assisted Bain Transformation

**Experiment observation**

- Norm to $(200)_{HCP}$ have angle of 45-50 degrees to the fiber axis

- Norm to $(110)_{BCC}$ have angle of ~50 degrees to the fiber axis

- The lattice constant of BCC is reported ~2.43 at 466 GPa

**Simulation observation**

- For stacking fault, basal plane $(001)_{HCP}//(111)_{FCC}$, therefore normal to the $(002)_{HCP}$ have angle of 54.7 degree to the fiber axis.

hydrostatic pressure favors transformations that result in a negative volumetric change and hinders those that result in a positive volumetric change

SEHITOGLU et al., Metallurgical and Materials Transactions,1996

# Phase identification of water at ambient temperature
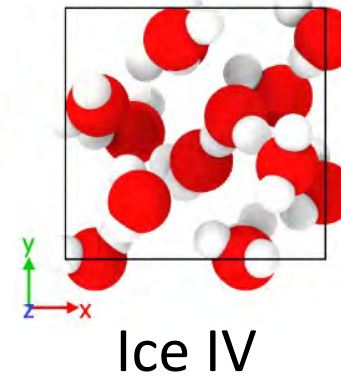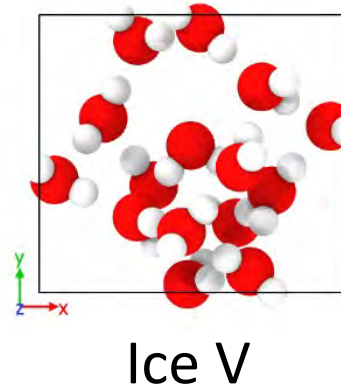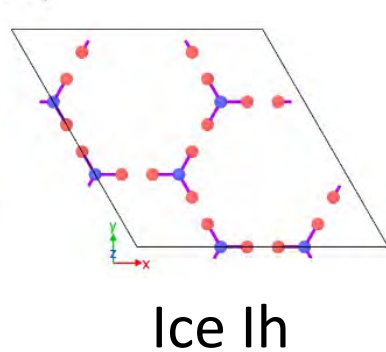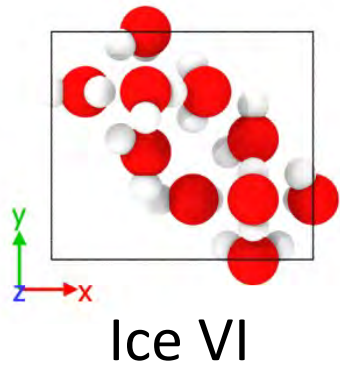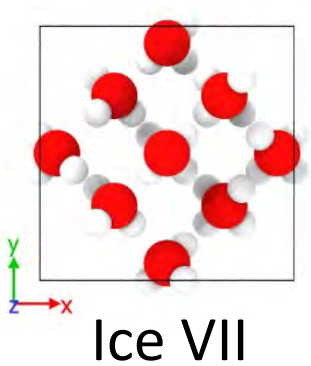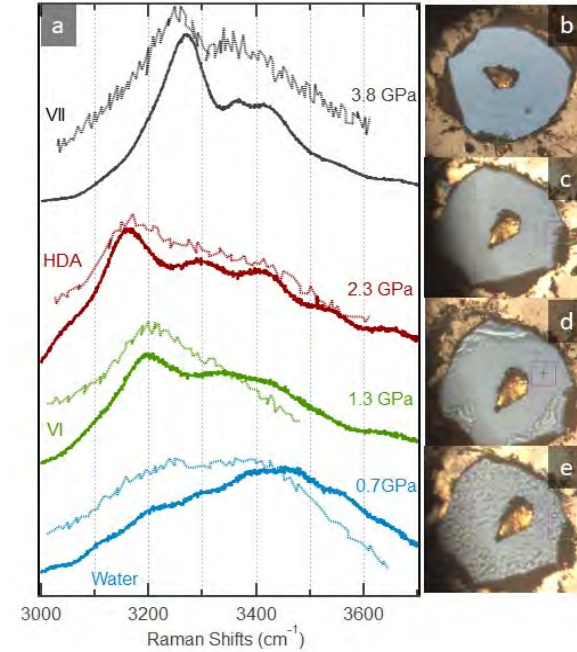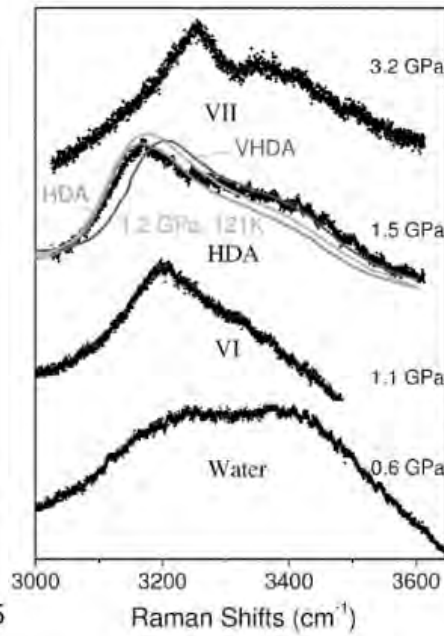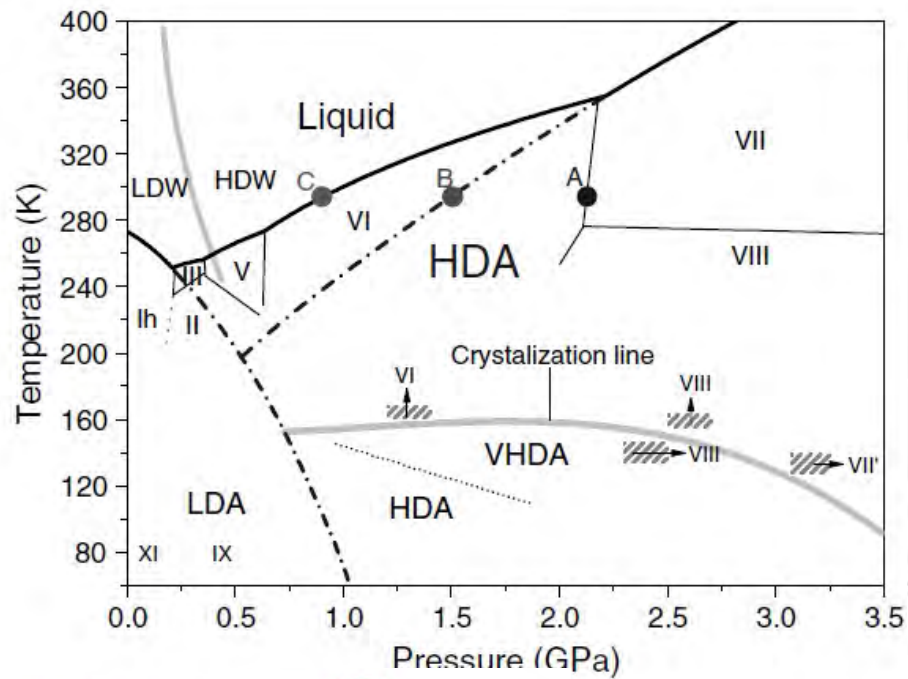


Ice VII  Ice VI  Ice Ih  Ice V  Ice IV

[1] Chen, J. Y., & Yoo, C. S. (2011). High density amorphous ice at room temperature. Proceedings of the National Academy of Sciences, 108(19), 7685-7688.

# Phase identification of water- Spectra comparison



| | ( IR spectra ) | ( Raman spectra ) | ( XRD spectra ) |
|---|---|---|---|
| ( HDA ) T=225K P=1.5GPa | | | $\lambda = 1.5418\text{Å}$ |
| ( VI ) T=300K P=1.5GPa | | | $\lambda = 0.4139\text{Å}$ |

[1] Chen, J. Y., & Yoo, C. S. (2011). High density amorphous ice at room temperature. Proceedings of the National Academy of Sciences, 108(19), 7685-7688.

# Phase identification of water at ambient temperature



Raman          XRD          Raman          XRD

# Phase identification of water at ambient temperature
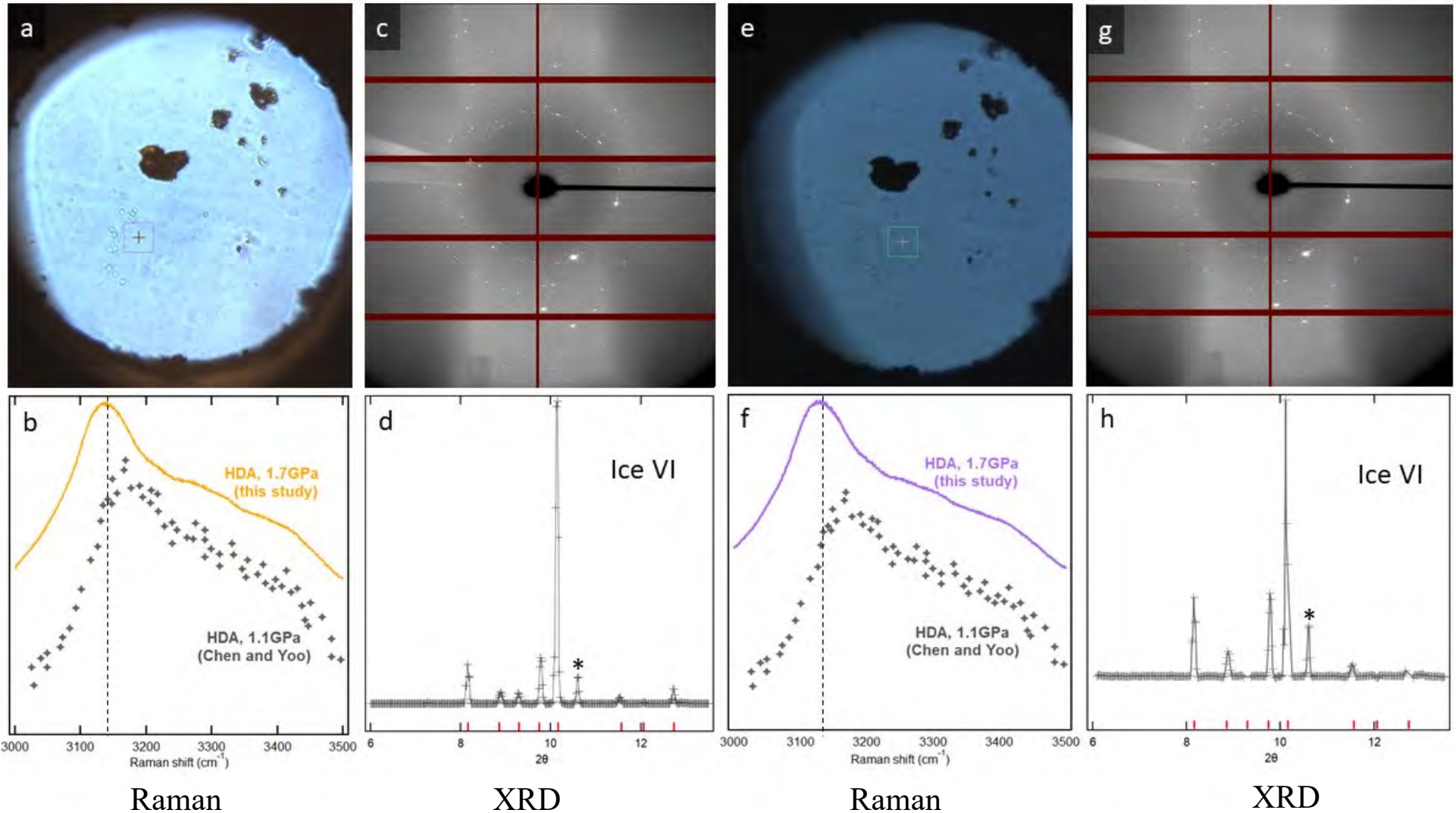


Ice VI-HDA coexistent system where the dominance of HDA or ice VI depends on phase fraction as well as interface !

# Phase identification of water at ambient temperature
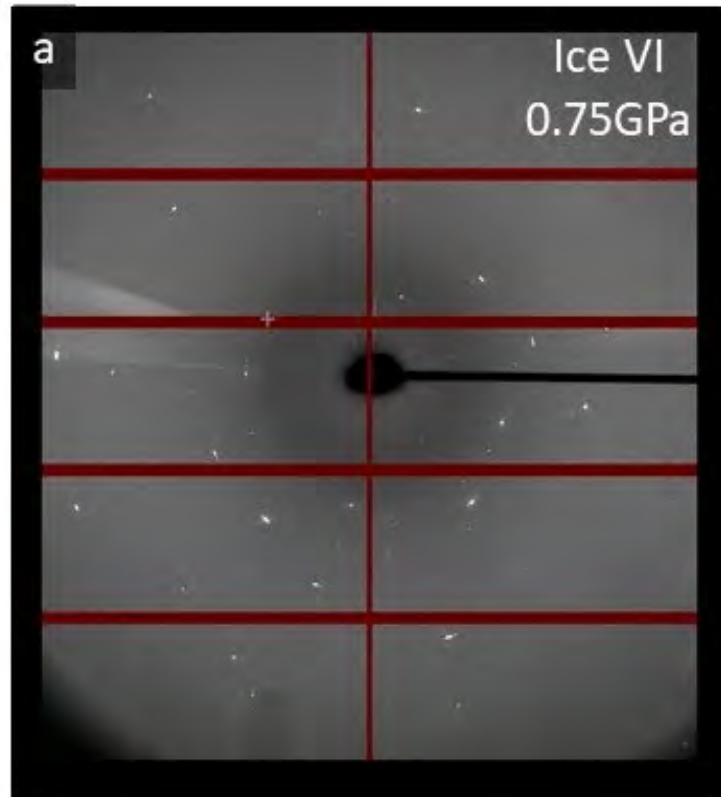


Ice VI grain size~2-3 μm        Ice VI grain size~0.5-1 μm
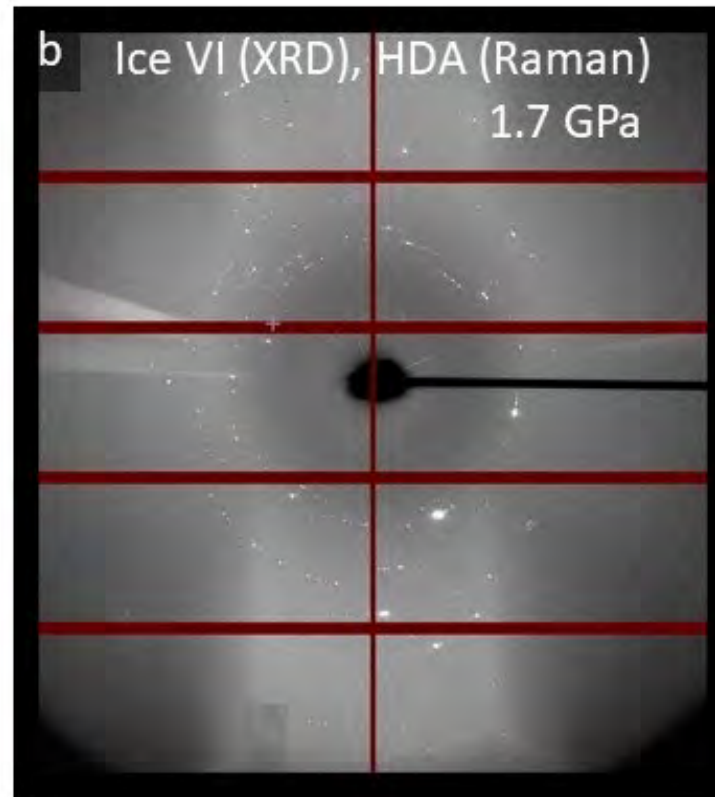
# Phase identification of water at ambient temperature



Ice VI grain size~2-3 µm          Ice VI grain size~0.5-1 µm

# Phase identification of water at ambient temperature

( Effects of phase fraction )

$$\frac{N_{VI}}{N_{HDA}} = \frac{1}{3} \qquad \frac{N_{VI}}{N_{HDA}} = \frac{1}{1} \qquad \frac{N_{VI}}{N_{HDA}} = \frac{3}{1}$$

( Effects of interface )
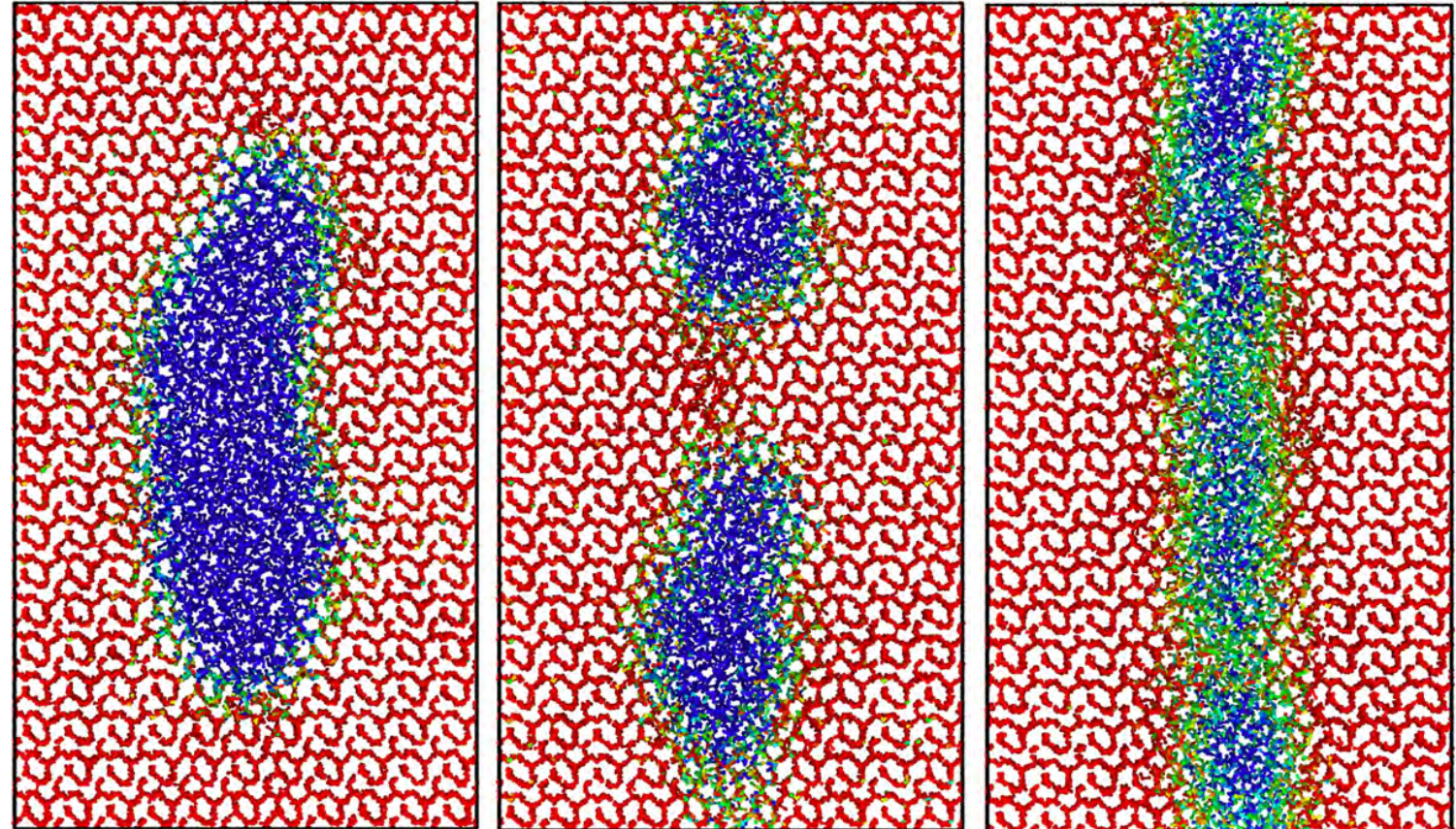
$$\frac{N_{VI}}{N_{HDA}} = 2.27 \qquad \frac{N_{VI}}{N_{HDA}} = 2.24 \qquad \frac{N_{VI}}{N_{HDA}} = 2.18$$

# Phase identification of water at ambient temperature

( Effects of phase fraction )

$$\frac{N_{VI}}{N_{HDA}} = \frac{1}{3} \qquad \frac{N_{VI}}{N_{HDA}} = \frac{1}{1} \qquad \frac{N_{VI}}{N_{HDA}} = \frac{3}{1}$$

$$\left( \frac{N_{VI}}{N_{HDA}} = \frac{1}{3} \right)$$

$$\left( \frac{N_{VI}}{N_{HDA}} = \frac{1}{1} \right)$$

$$\left( \frac{N_{VI}}{N_{HDA}} = \frac{3}{1} \right)$$

VI present!

VI dominant!

VI dominant!

(a)  (b)  (c)

HDA dominant!

HDA dominant!

VI dominant!
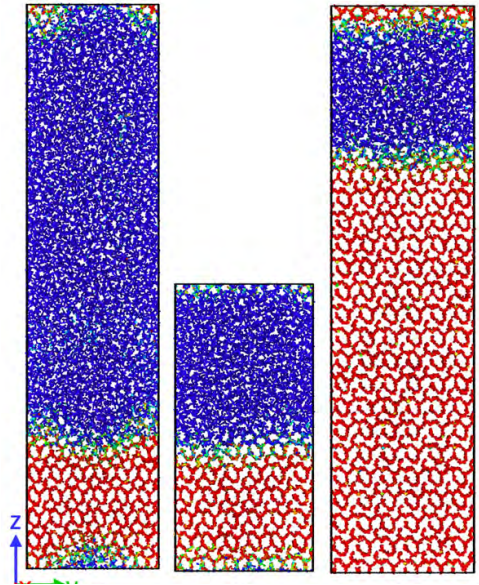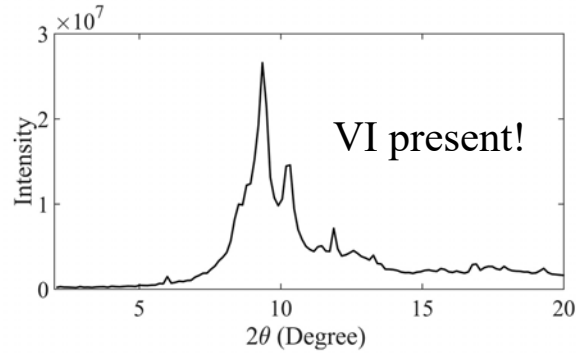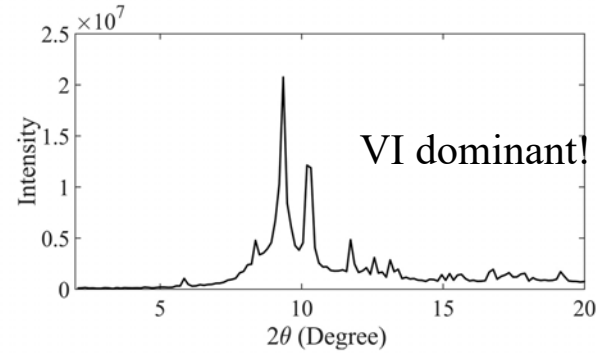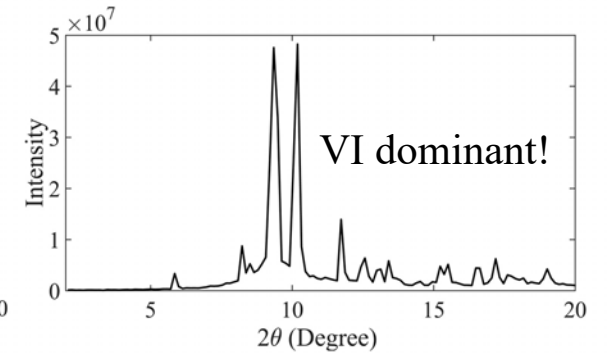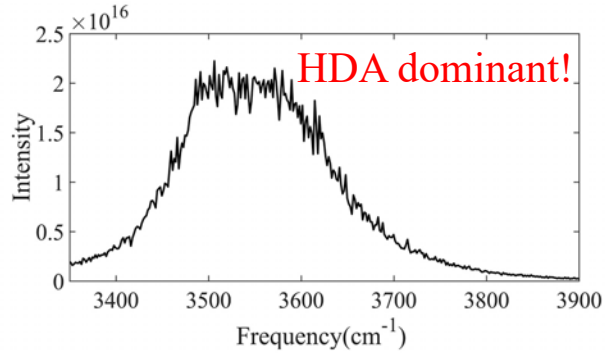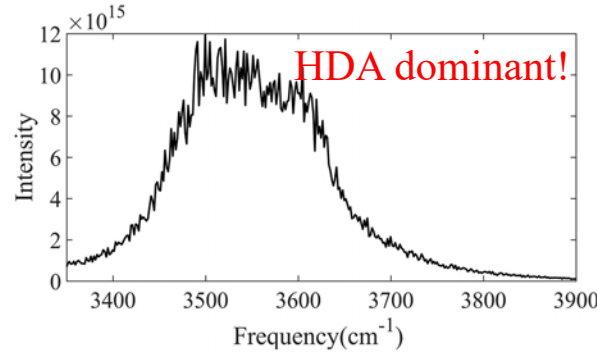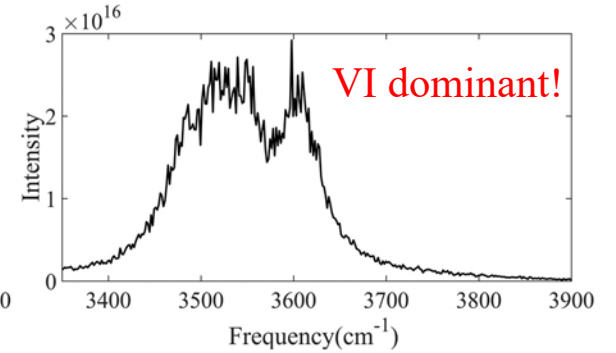
(d)  (e)  (f)

# Phase identification of water at ambient temperature

$\dfrac{N_{VI}}{N_{HDA}} = 2.27$
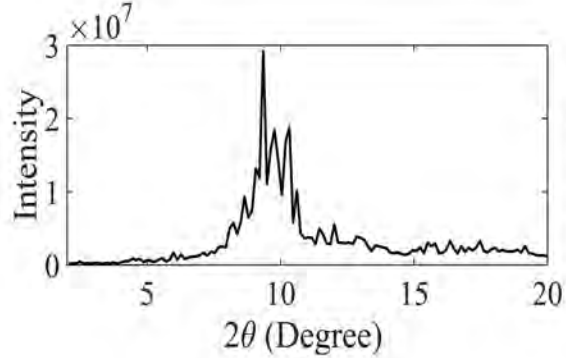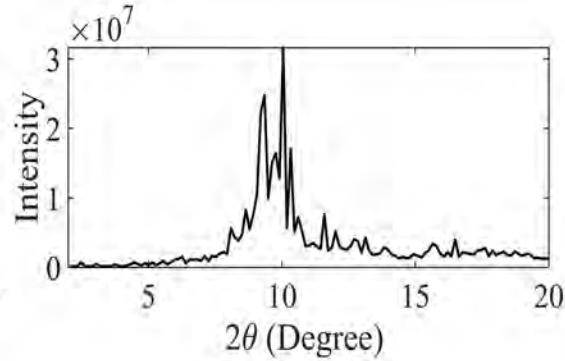
$\dfrac{N_{VI}}{N_{HDA}} = 2.24$

$\dfrac{N_{VI}}{N_{HDA}} = 2.18$

( Effects of interface )

$\dfrac{N_{VI}}{N_{HDA}} = 2.27$   $\dfrac{N_{VI}}{N_{HDA}} = 2.24$   $\dfrac{N_{VI}}{N_{HDA}} = 2.18$



(a)   (b)   (c)

(d)   (e)   (f)

VI dominant!

HDA dominant!

-MD data
-Smoothed data
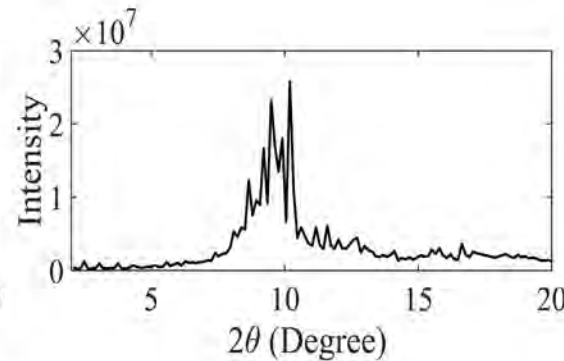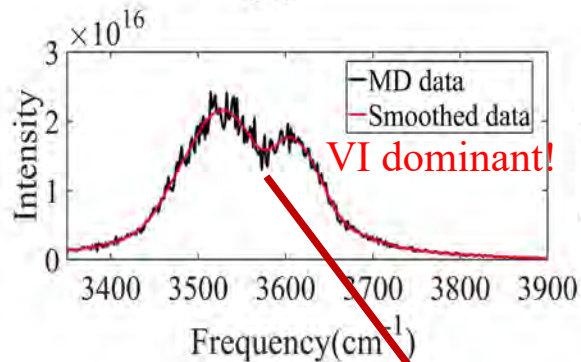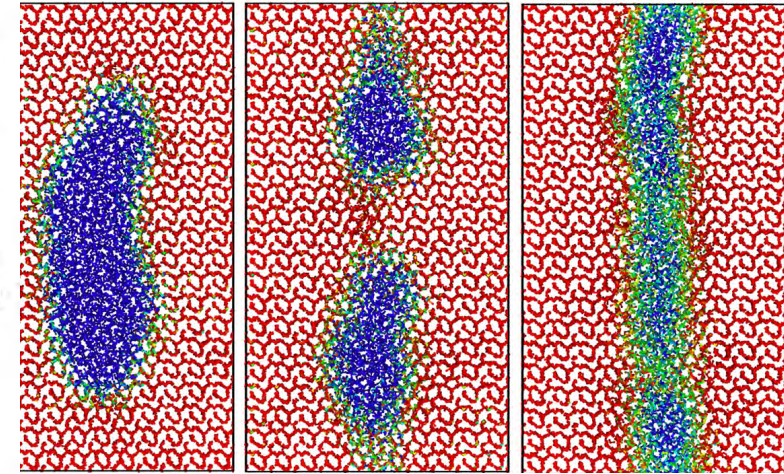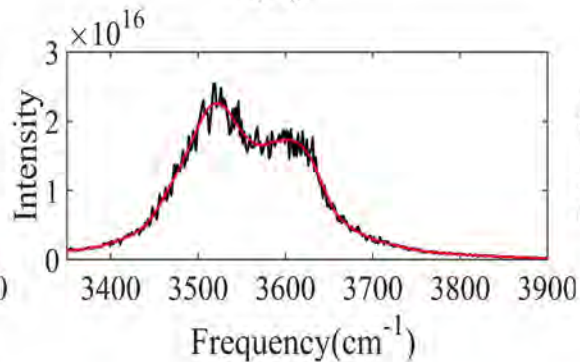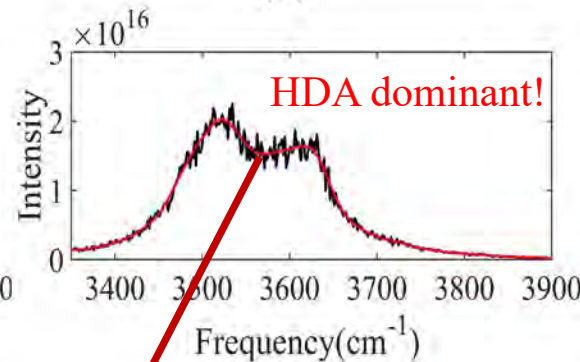
The minima becomes flat by increasing interface area turning into HDA spectra !

coexistent ice VI/HDA system with a large number of <u>small ice VI grains</u> with <u>large interface area </u>with ice HDA regions.

# Concluding marks from two studies:

- MD simulations captured the phase transformation mechanisms in Aluminum under high pressures

- Phase transformation and plastic deformation mechanisms are deduced from time-resolved synthetic XRD data

- Good agreement was observed between experimental and synthetic XRD analysis

- Raman and XRD diagnostics presented inconsistent understanding about the the phase of water at high pressure

- MD simulations showed that a water coexistent system explains the uncertainties within experimental Raman and XRD diagnostics

# 3) Developing AI-powered models for classification of large XRD data

- Time-resolved XRD Images
  - Data intensive
  - Include many uncertainties

- Analyze XRD
  - Human expert
  - Conventional indexing software are contentious

- Experimental XRD
  - Expensive
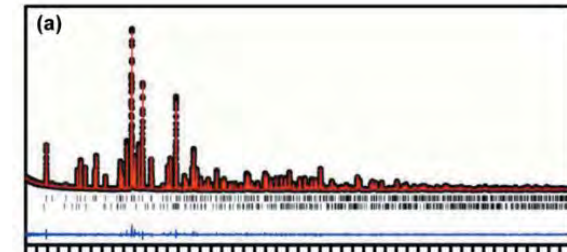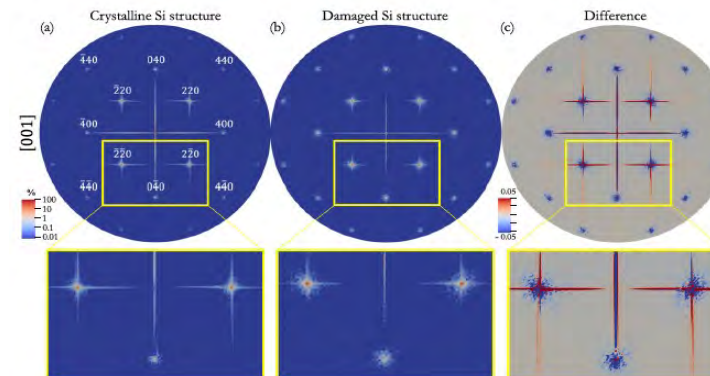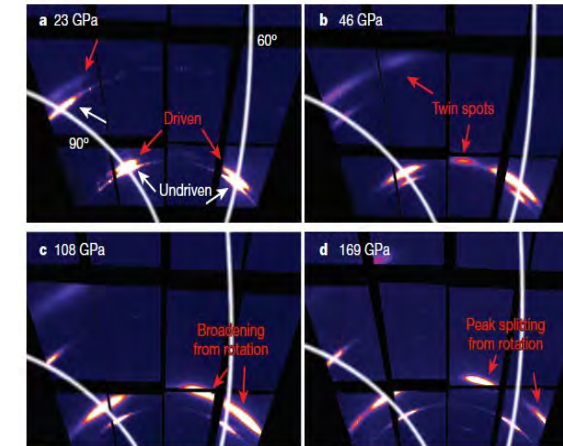
Synthetic
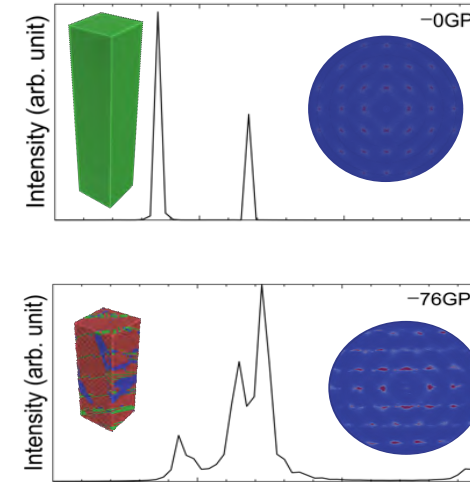
Experiments

[2]Coppari, F. et al., 2019. Optimized x-ray sources for x-ray diffraction measurements at the Omega Laser Facility. Review of Scientific Instruments 90, 125113
[3]Park, W.B. et al., 2017. Classification of crystal structure using a convolutional neural network. IUCrJ 4, 486-494.. doi:10.1107/s205225251700714x

UNIVERSITY of ROCHESTER

# 3) Developing AI-powered models for classification of large XRD data

- Time-resolved XRD Images
  - Data intensive
  - Include many uncertainties

- Analyze XRD
  - Human expert
  - Conventional indexing software are contentious

Synthetic

Experiments



**Develop automated deep learning computer vision techniques to mine such information-rich data to classify crystal structures and filter and detect lattice-level mechanisms responsible for phase transformation and plastic deformation under extreme conditions**

- Experimental XRD
  - Expensive

[2]Coppari, F. et al., 2019. Optimized x-ray sources for x-ray diffraction measurements at the Omega Laser Facility. Review of Scientific Instruments 90, 125113
[3]Park, W.B. et al., 2017. Classification of crystal structure using a convolutional neural network. IUCrJ 4, 486-494.. doi:10.1107/s205225251700714x

UNIVERSITY of ROCHESTER

# Developing AI-powered models for classification of large XRD data

- Time-resolved XRD Images
  - Data intensive
  - Include many uncertainties

- <span style="color:red">Generation of static and time-resolved synthetic 1D and 2D XRD images</span>

- Analyze XRD
  - Human expert
  - Conventional indexing software are contentious

- <span style="color:red">Supervised deep learning for lattice structure classification and temporal lattice dynamics identification</span>
- <span style="color:red">Interpretation of deep learning models and predictions using explainable deep learning tools</span>

- Experimental XRD
  - Expensive

- <span style="color:red">Domain adaptation to scarce experimental data</span>

[2]Coppari, F. et al., 2019. Optimized x-ray sources for x-ray diffraction measurements at the Omega Laser Facility. Review of Scientific Instruments 90, 125113
[3]Park, W.B. et al., 2017. Classification of crystal structure using a convolutional neural network. IUCrJ 4, 486-494., doi:10.1107/s205225251700714x

Abbreviation: Classification, Regression, Preprocess, Database, Augmentation, Function, True, False, Experimental, Simulated, ICSD, Cambridge Crystallographic Data Centre,

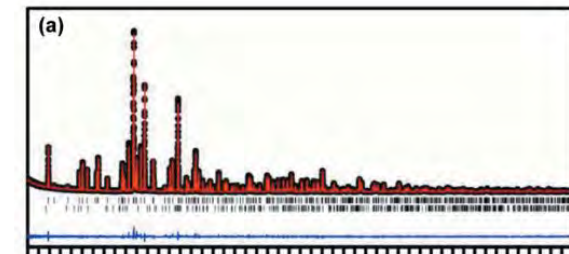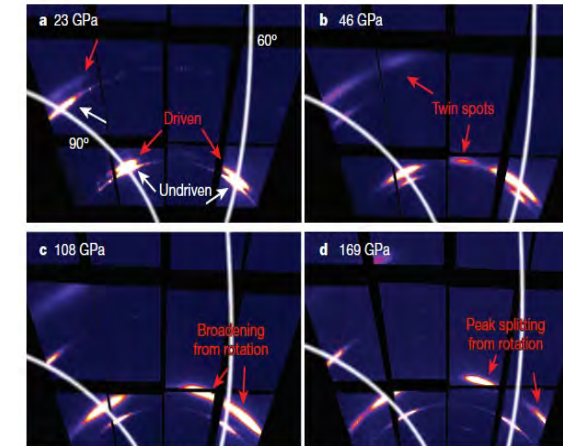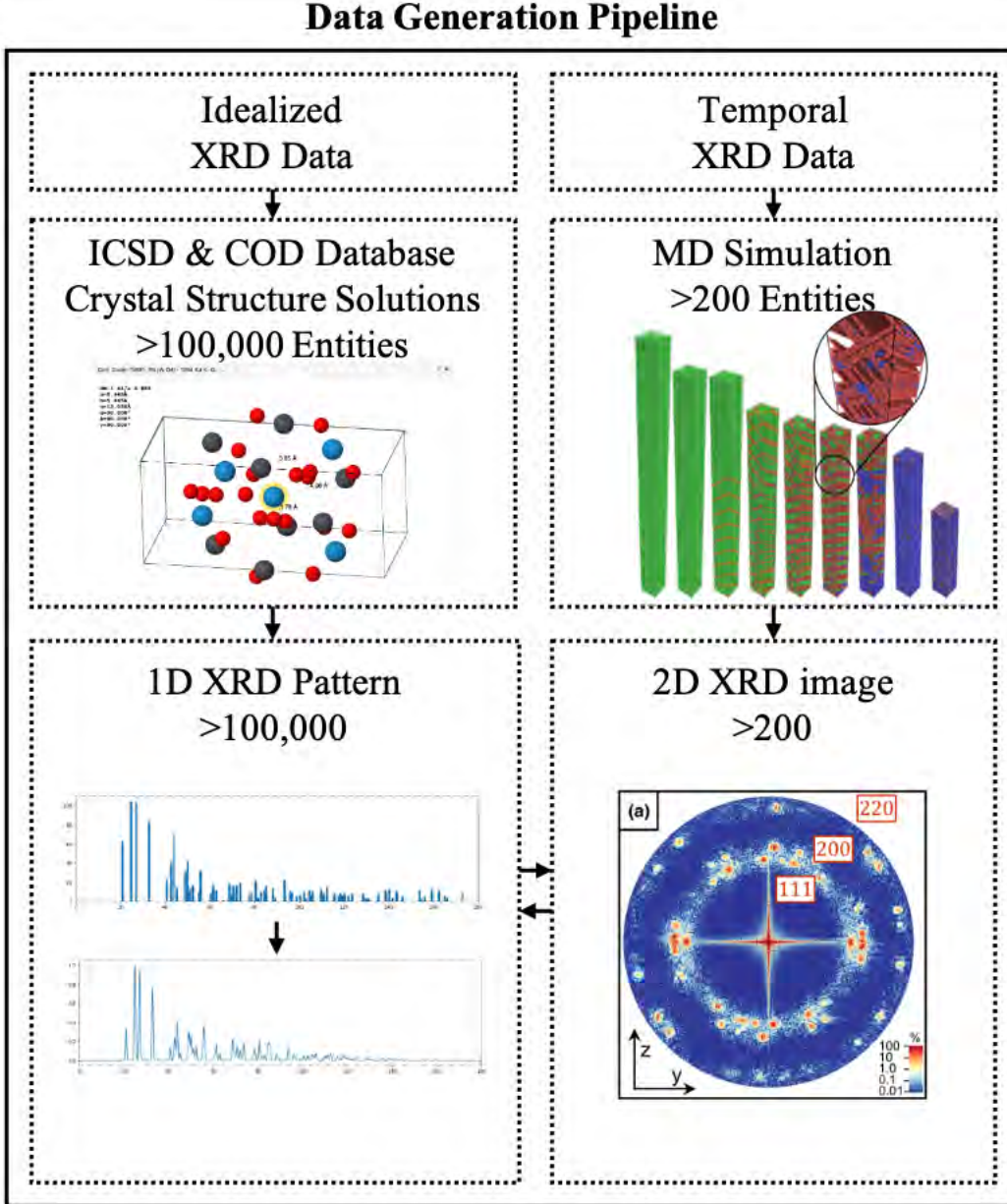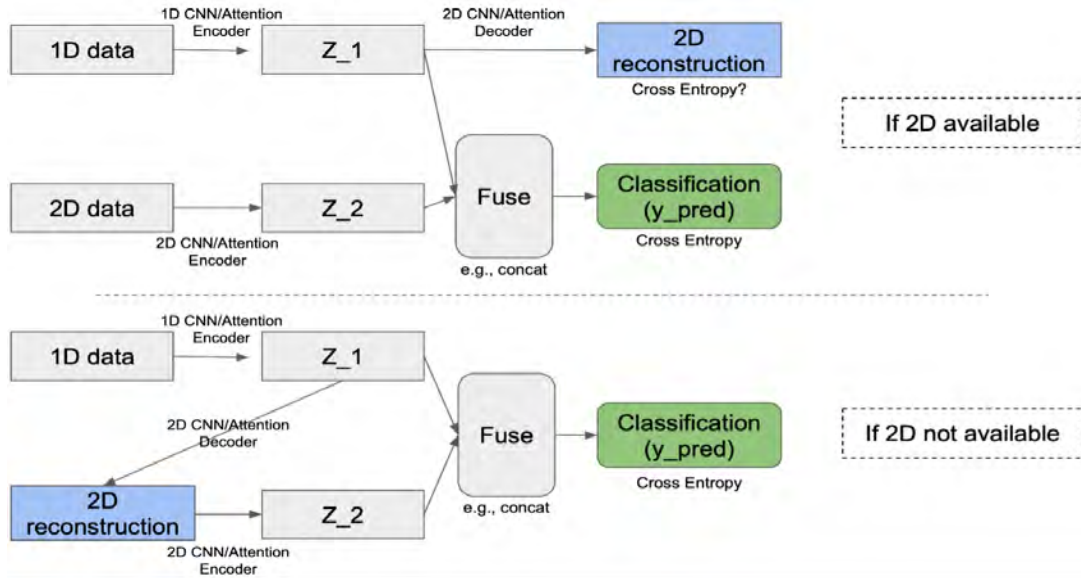| LITERATURE TITLE | YEAR | TYPE | PREDICTION GOAL TYPE | INDEX | NUM | EXP. ORIGIN | PRE | AUG | NUM | SIM. DB | ORIGIN | PRE | AUG | NUM | MODEL TYPE | FUNC | TRAINING SET | TESTING SET | ACCURACY | MATERIAL |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| A data-driven XRD analysis protocol for phase identification… | 2021 | XRD | C | Phase | 21 | 36, from 3 binary oxides | T | F | 36 | ICSD | 218 | T | T | 89,943 | CNN | | 80% sim. | 20% sim. | 94.36 | Li-La-Zr-O quaternary compositional |
| | | | | | | | | | | | | | | | | | | exp. | 88.88 | |
| | | | R | Phase Fraction | R | | | | | | | | | 13,930,000 | | | | 20% sim. | 0.004612 | |
| | | | | | | | | | | | | | | | | | | exp. | 0.008260 | |
| A deep-learning technique for phase identification… | 2020 | XRD | C | Phase | 38 | 50+50 | F | F | 50+50 | ICSD | 170 | T | F | 800,942 | CNN | ReLu | 600,942+100,000 sim. | 100,000 sim. | 100 | Sr-Li-Al-O quaternary compositional |
| | | | | | | | | | | | | | | | | | | 50 exp. Li2O… | 100 | |
| | | | | | | | | | | | | | | | | | | 50 exp. SrAl2O4… | 98.67 | |
| | | | | Three step phase fraction | 3 | | | | 50 | | | | | | | | | 100,000 sim. | 98 | |
| | | | | | | | | | | | | | | | | | | 50 exp. Li2O… | 86 | |
| Crystal symmetry determination… | 2020 | EBSD | C | Bravais lattices | 14 | ? | | | | ? | | | | | CNN | ResNet50 Xception | ? | 50000 | 93.5 / 91.2 | Mo3Si, Al, Ta, Ti, Ilmenite, Sn, Anatase, +7… |
| | | | | | | | | | | | | | | | CNN | ResNet50 Xception | | 300000 | >90 / >90 | |
| Rapid Identification of X-ray Diffraction Patterns Based on Very Limited… | 2020 | XRD | C | material one-by-one | 1012 | 10 | T | T | 10*3=30 | CCDC | 1012 | T | T | 1012*72=72864 | CNN | ReLu Adam Keras TensoeFlow | 58,292 (80% sim.) | 14,572 (20% sim.) | Unknown | 1012 patterns sim. 10 MOFs exp. |
| | | | | | | | | | | | | | | | | | | 30 exp. | 96.7 | |
| Fast and interpretable classification… | 2019 | XRD | C | Dimensionality | 3 | 75/115 | F | T | 2000 | ICSD | 164 | T | T | 2000 | a-CNN | | all sim.+80% exp. | 20% exp. | 92.9 | Perovskites-inspired 3D, 2D, 0D |
| | | | | Space group | 7 | 88/115 | | | 2000 | | | | | 2000 | | | | | 89.3 | |
| Insightful classification of crystal structures using deep learning | 2018 | Diffraction Fingerprints | C | Crystal Structure | 8 | None | | | | AFLOW LIB | 10,517 | T | F | 10,517 | CNN | Adam TensorFlow Keras | 90% sim. | 10% sim. | 100 | 83 chemical species + manually mapped atomic num. |
| | | | | | | | | | | | | | | | | | | 10,517*10 defected | >97 | |
| Classification of crystal structure… | 2017 | XRD | C | Crystal system | 7 | 1+1 | F | F | 2 | ICSD | 150,000 | T | F | 150,000 | CNN | ReLu | 80% sim. | 20% sim. | 94.99 | All ICSD database |
| | | | | | | | | | | | | | | | | | | 1+1 exp. | 100 | |
| | | | | Extinction Group | 101 | | | | | | | | | | | | | 20% sim. | 83.830 | |
| | | | | | | | | | | | | | | | | | | 1+1 exp. | 0 | |
| | | | | Space Group | 230 | | | | | | | | | | | | | 20% sim. | 81.140 | |
| | | | | | | | | | | | | | | | | | | 1+1 exp. | 0 | |
| Generalized machine learning technique for… | 2015 | XRD, Raman Fluore… | C | Phase | 4 | 64 | T | T | 64 | NONE | | | | | ML | | | 2/3 exp. | 1/3 exp. | 99.22 | alpha-Al2O3, gamma, theta-Al2O3, NiO, Inconel Oxide |
| | | | | | | 144 | T | T | 144 | | | | | | | | | 2/3 exp. | 1/3 exp. | 97.6 | |
| | | | | | | 144 | T | T | 144 | | | | | | | | | 2/3 exp. | 1/3 exp. | 95.2 | |

| LITERATURE | | | PREDICTION GOAL | | EXPERIMENTAL DATASET | SIMULATED DATASET | MODEL | | TRAINING SET | TESTING SET | ACCURACY | MATERIAL |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| TITLE | YEAR | TYPE | TYPE | IN… | | | | UNC | | | | |
| A data-driven XRD analysis protocol for phase identification… | 2021 | XRD | C | Ph… | | | | | 80% sim. | 20% sim. | 94.36 | Li-La-Zr-O quaternary compositional |
| | | | | | | | | | | exp. | 88.88 | |
| | | | R | Ph… Fra… | | | | | | 20% sim. | 0.004612 | |
| | | | | | | | | | | exp. | 0.008260 | |
| A deep-learning technique for phase identification… | 2020 | XRD | C | Ph… | | | | …eLu | 600,942+100,000 sim. | 100,000 sim. | 100 | Sr-Li-Al-O quaternary compositional |
| | | | | | | | | | | 50 exp. Li2O… | 100 | |
| | | | | | | | | | | 50 exp. SrAl2O4… | 98.67 | |
| | | | | Thre… pl… fra… | | | | | | 100,000 sim. | 98 | |
| | | | | | | | | | | 50 exp. Li2O… | 86 | |
| Crystal symmetry determination… | 2020 | EBSD | C | Br… lat… | | | | …Net50 …ption …Net50 …ption | ? | 50000 | 93.5 | Mo3Si, Al, Ta, Ti, Ilmenite, Sn, Anatase, +7… |
| | | | | | | | | | | | 91.2 | |
| | | | | | | | | | | 300000 | >90 | |
| | | | | | | | | | | | >90 | |
| Rapid Identification of X-ray Diffraction Patterns Based on Very Limited… | 2020 | XRD | C | ma… one-… | | | | …eLu …dam …eras …peFlow | 58,292 (80% sim.) | 14,572 (20% sim.) | Unknown | 1012 patterns sim. 10 MOFs exp. |
| | | | | | | | | | | 30 exp. | 96.7 | |
| Fast and interpretable classification… | 2019 | XRD | C | Dime… | | | | | all sim.+80% exp. | 20% exp. | 92.9 | Perovskites-inspired 3D, 2D, 0D |
| | | | | Spac… | | | | | | | 89.3 | |
| Insightful classification of crystal structures using deep learning | 2018 | Diffraction Fingerprints | C | Cr… Stru… | | | | …dam …orFlow …eras | 90% sim. | 10% sim. | 100 | 83 chemical species + manually mapped atomic num. |
| | | | | | | | | | | 10,517*10 defected | >97 | |
| Classification of crystal structure… | 2017 | XRD | C | Cr… sy… | | | | …eLu | 80% sim. | 20% sim. | 94.99 | All ICSD database |
| | | | | | | | | | | 1+1 exp. | 100 | |
| | | | | Exti… G… | | | | | | 20% sim. | 83.83 | |
| | | | | | | | | | | 1+1 exp. | 0 | |
| | | | | Spac… | | | | | | 20% sim. | 81.14 | |
| | | | | | | | | | | 1+1 exp. | 0 | |
| Generalized machine learning technique for… | 2015 | XRD Raman Fluore… | C | Phase | 4 | 144 T | T | NONE | 144 | ML | | alpha-Al2O3, gamma, theta-Al2O3, NiO, Inconel Oxide |
| | | | | | | | | | 2/3 exp. | 1/3 exp. | 99.22 | |
| | | | | | | | | | 2/3 exp. | 1/3 exp. | 97.6 | |
| | | | | | | | | | 2/3 exp. | 1/3 exp. | 95.2 | |



**Data Generation Pipeline**

Idealized XRD Data → ICSD & COD Database Crystal Structure Solutions >100,000 Entities → 1D XRD Pattern >100,000

Temporal XRD Data → MD Simulation >200 Entities → 2D XRD image >200
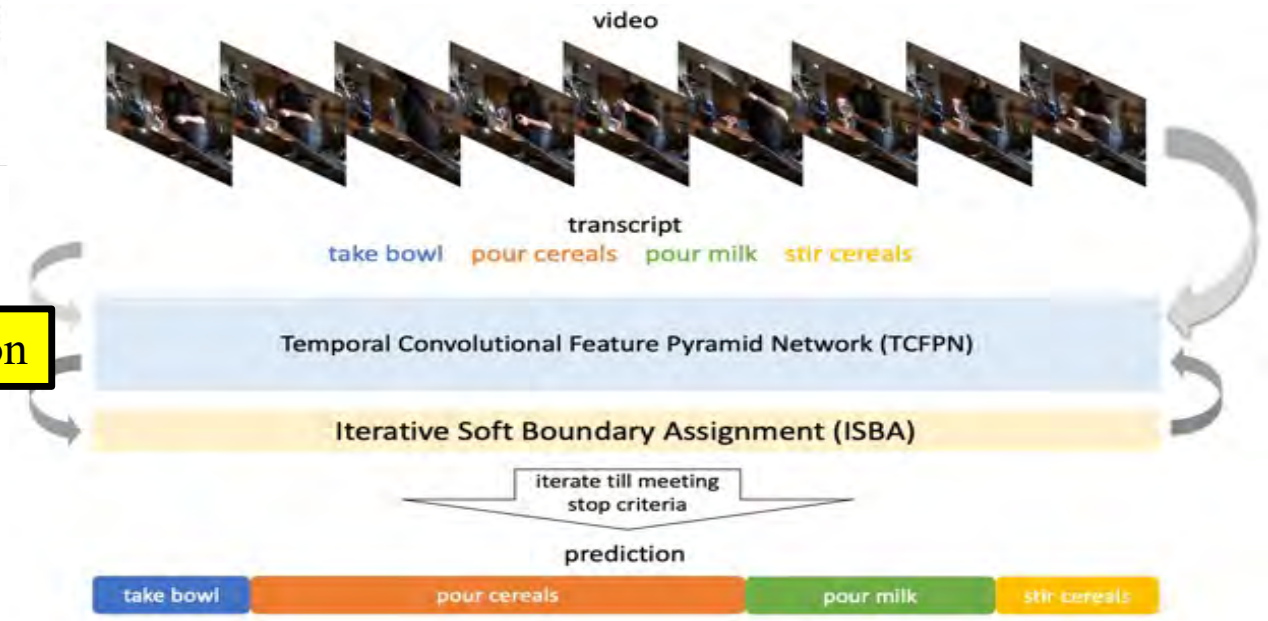
# Developing AI-powered models for classification of large XRD data



Static classification

Temporal classification

# Current Progress

Total Data Availability:
COD: 476,830 crystal structures
ICSD: 250,343 crystal structures

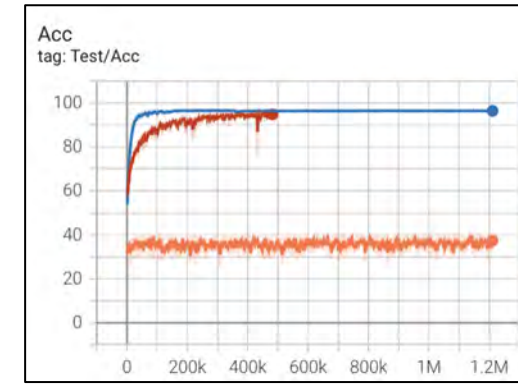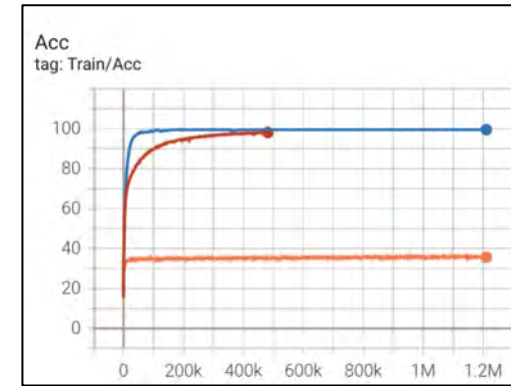For Dataset size of 47,049(COD)
Shuffling the data
- dividing train/test sets randomly

Accuracy comparison between
- Logistic Regression
- Deep Neural Networks
- Convolutional Neural Network



## Classification of 7 crystal systems



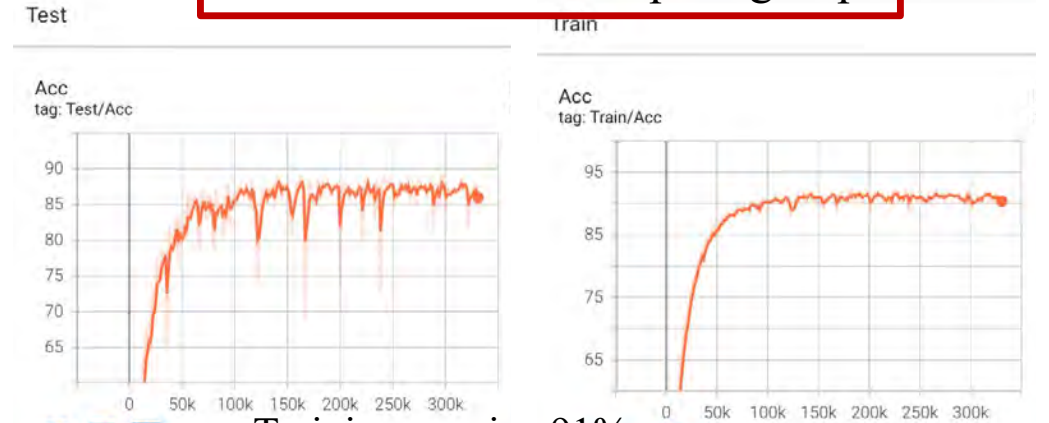For DNN:
While training acc. is 100%
Testing acc. reach 96.42%

## Classification of 230 space groups



Training acc. is ~91%
Testing acc. Reach ~88%

```
-------------------------------------------------
Layer (type)          Output Shape        Param #
=================================================
Conv1d-1              [-1, 16, 1800]           96
BatchNorm1d-2         [-1, 16, 1800]           32
ReLU-3                [-1, 16, 1800]            0
MaxPool1d-4           [-1, 16, 900]            0
Conv1d-5              [-1, 32, 900]         2,592
BatchNorm1d-6         [-1, 32, 900]            64
ReLU-7                [-1, 32, 900]            0
MaxPool1d-8           [-1, 32, 450]            0
Flatten-9             [-1, 14400]             0
Linear-10             [-1, 7]            100,807
=================================================
Total params: 103,591
Trainable params: 103,591
Non-trainable params: 0
-------------------------------------------------
```

DNN Intermediary architecture



For DNN: Confusion matrix diagonals shows validity

# Developing AI-powered models for classification of large XRD data

*Interpretation of deep learning models and predictions using explainable deep learning tools, Class Activation Maps (CAMs)*

**Potential sources of misclassification**

1) The mixture of phases in the sample
2) Lack of XRD patterns in the specific class of training data
3) Missing peaks, or too few peaks, present in the XRD pattern
4) The peak shifting, broadening, or splitting on time-resolved XRD patterns may be due to various reasons (e.g. multiple plastic deformation mechanisms)
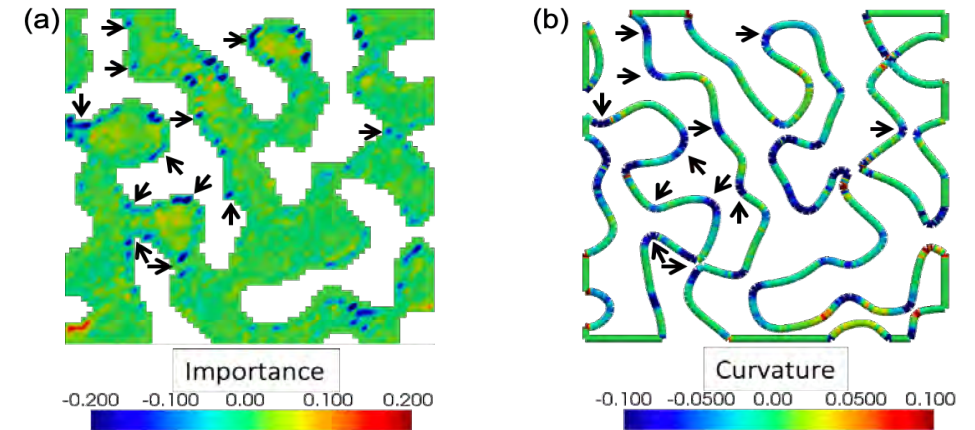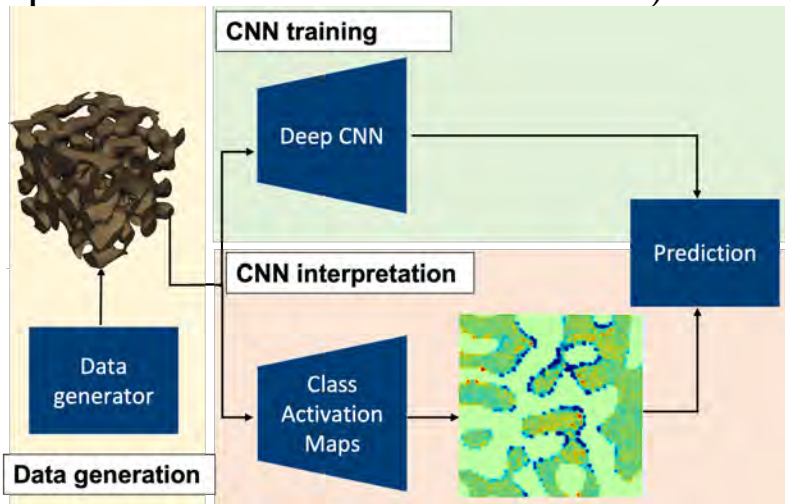


Cross-section of random structure (a) Neuron map of the neuron 38. (b) Gaussian curvature map. Saddle-shaped region is blue. Flat region is green

Mining structure-property linkage in nanoporous materials using an interpretative deep learning approach, Materialia, 2022

# Developing AI-powered models for classification of large XRD data

*Domain adaptation to scarce experimental data*
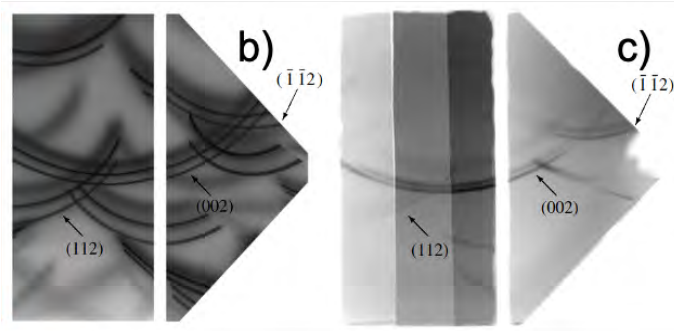


$$min_{\phi_S, c_S} \mathcal{L}_S\left(c_S\left(\phi_S(X_S)\right), Y_S\right),$$

$$\phi_T = \mathcal{A}(\phi_S, D_T),$$

$$min_{c_T} \mathcal{L}_T\left(c_T\left(\phi_T(X_T)\right), Y_T\right).$$

b) Simulated and (c) experimental film recordings of x-rays diffracted from shock compressed iron

As the NEMD simulations provide direct physical insight into the shock-deformation of materials at the lattice level, and the experimental and simulated time and length scales are converging, it is appropriate to make direct comparisons between the experimentally observed x-ray diffraction signals, and those predicted by the NEMD simulations

G. Kimminau, et al, Simulating picosecond x-ray diffraction from shocked crystals using post-processing molecular dynamics calculations, J. Phys. Condens. Matter. 20 (2008).

**UNIVERSITY** *of* **ROCHESTER**

# Collaborators and acknowledgements

Collaborators:
- Gillbert Collins, UR, LLE
- Russel Hemley, UIC
- Danae Polsin, UR, LLE
- Shuai Zhang, LLE
- Shanti Deemyad, University of Utah
- Saveez Saffarian, University of Utah
- Chenliang Xu, UR
- Hesam Askari, UR

Acknowledge support from:
- DOE
- NSF
- URA
- CIRC

## Questions and comments?

niaz@rochester.edu

supplementary

Next:

- Machine learning interatomic potentials for complex dynamic processes at high pressures.

- What materials? Na, Mg, Li, Fe

- DFT ⟶ MLIP ⟶ MD ⟶ XRD

| Method | Refs. | | Regressor | Implementation |
|---|---|---|---|---|
| Artificial neural networks (Behler) | [40] | (2007) | NN | Standalone ("RuNNer"); LAMMPS interface[67] |
| Gaussian approximation potentials (Bartók and Csányi) | [68] | (2010) | GPR | GAP code (custom); LAMMPS interface |
| Spectral neighbor analysis potential (SNAP) (Thompson) | [55,69] | (2015) | Linear fit | LAMMPS interface |
| Adaptive, generalizable, and neighborhood informed (AGNI) force fields (Ramprasad) | [70–72] | (2015) | KRR | LAMMPS interface |
| aenet (Artrith) | [73] | (2016) | NN | Standalone ("aenet") |
| Amp (Korshidi and Peterson) | [74] | (2016) | NN | Standalone ("amp"); LAMMPS interface |
| Moment tensor potentials (Shapeev) | [54,75] | (2016) | Linear fit | LAMMPS interface |
| DeePMD (E) | [76] | (2018) | NN | Standalone ("DeePMD-kit"); LAMMPS interface |



Ref: Transforming simple metals to topological insulators: Sodium to 18 Mbars

Structural Diversity of Sodium

Eugene Gregoryanz,[1]* Lars F. Lundegaard,[1] Malcolm I. McMahon,[1] Christophe Guillaume,[1] Richard J. Nelmes,[1] Mohamed Mezouar[2]

# Texturized nanocrystalline (NC) Al ramp loading

- The interatomic potential we used (developed by Winey et al.*) has also been tested and compared with experiment in terms of Hugoniot curve $P_H$, Gruneisen coefficient $\gamma$ and melting temperature $T_m$ at high pressure by Yang et al.** Specifically, they found the linear relation and the Hugoniot curve are almost identical for SC and NC Al and attributed it to because the fact that the grain size cannot nearly affect the dynamic properties of Al at high pressure.