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Probing Structural and Magnetic Phase Changes in the Shock Response of Iron with Molecular Dynamics

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Background & Motivation

Behavior of metals under HED conditions is difficult to predict

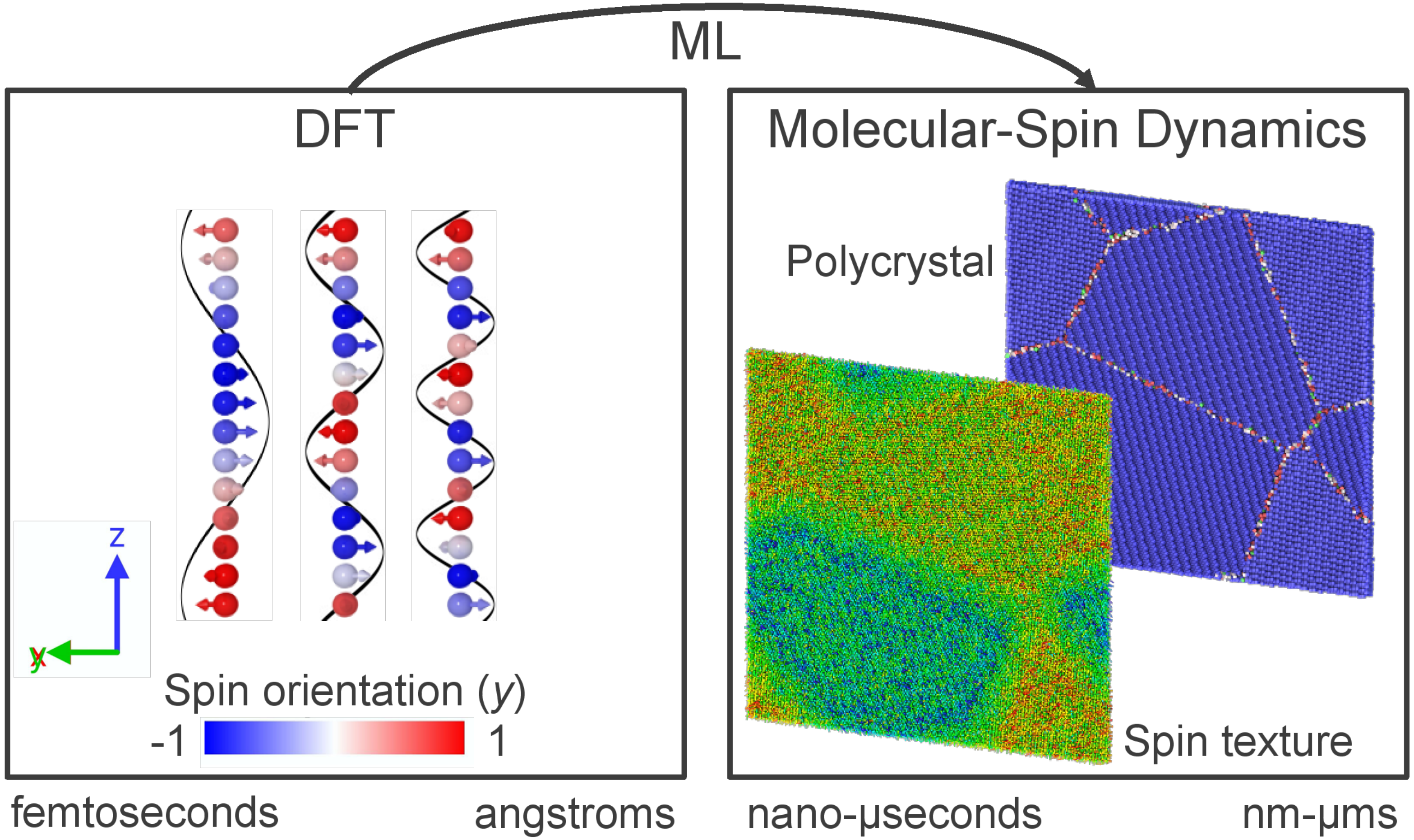
- Geophysics, fusion energy science, etc.
- Accounting for magnetic degrees of freedom is important

Ab-initio resolution of magnetic DOF is ideal

- Spatial and temporal restrictions

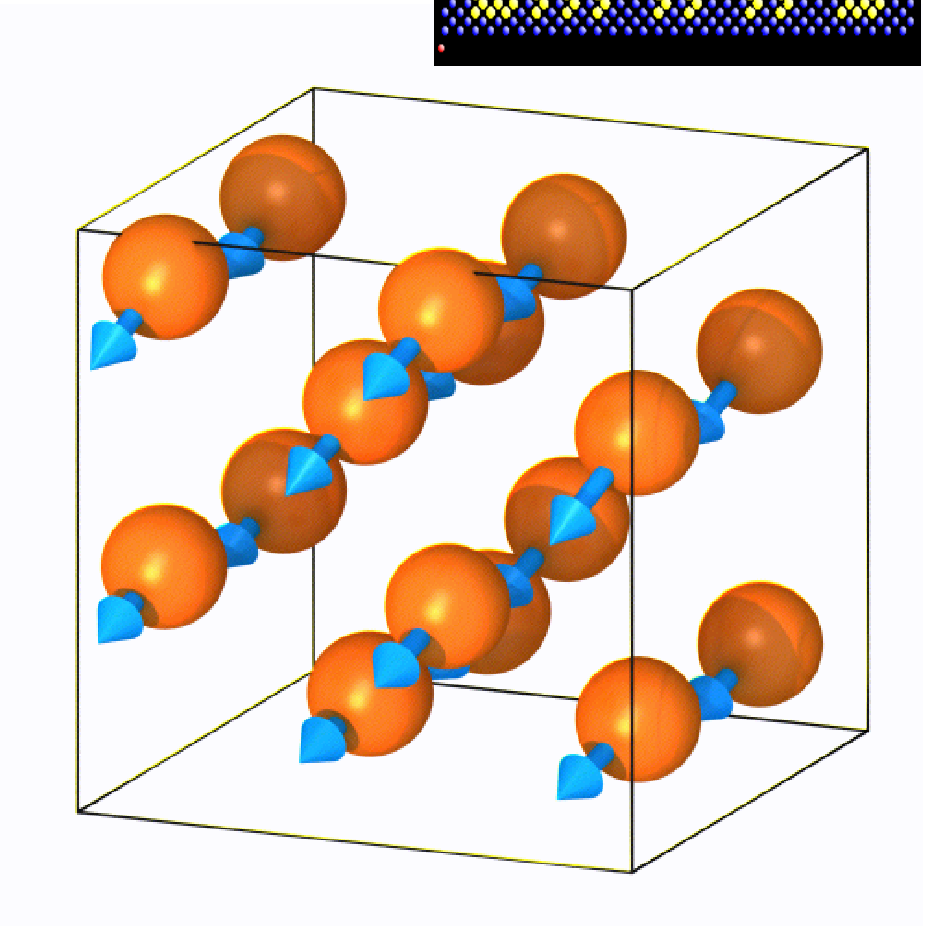
Molecular-Spin Dynamics models can be trained on DFT data

- Applied to study emergent material behavior



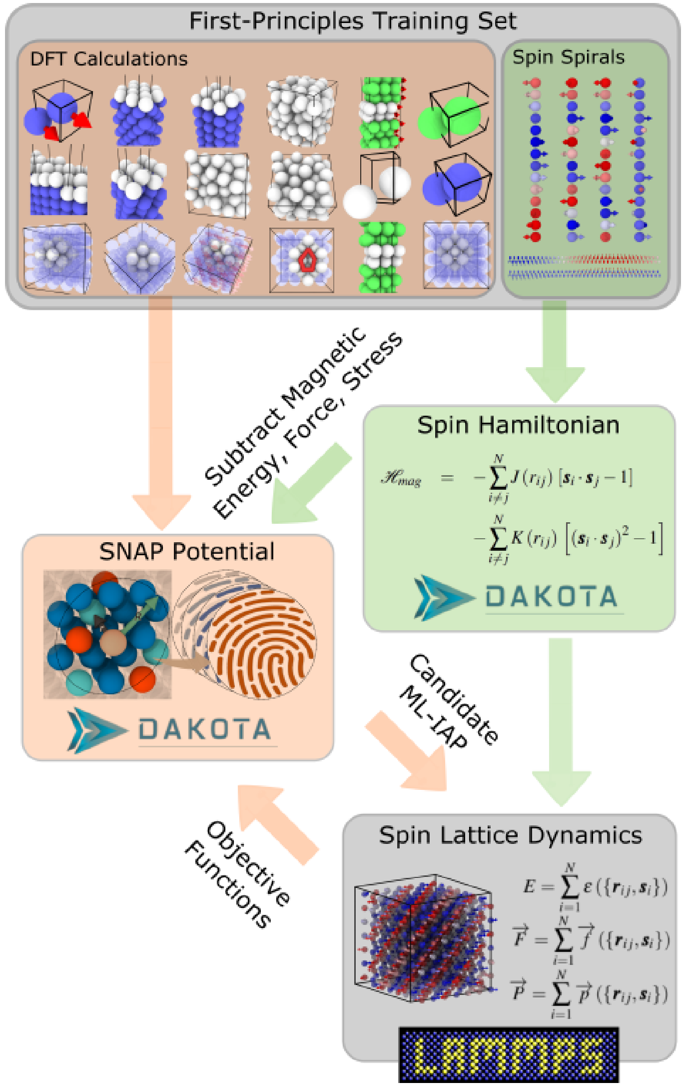
Molecular-Spin Dynamics

- Molecular Dynamics
  - Atoms interact via nonmagnetic interatomic potential, $U(\mathbf{R})$
  - $\mathcal{H}_{MD} = \sum_i \frac{\mathbf{p}_i{}^2}{2m} + U(\mathbf{R})$
- Spin Dynamics
  - Atoms interact via exchange function which conserves total angular momentum
  - $\mathcal{H}_s = -\sum_{i,j}^{N} J_{ij}(\mathbf{R})[\vec{s_i} \cdot \vec{s_j} - 1] - \sum_{i,j}^{N} K_{ij}(\mathbf{R})\left[(\vec{s_i} \cdot \vec{s_j})^2 - 1\right]$
  - Additional physics can be easily incorporated
    - Magnetocrystalline anisotropies, external magnetic fields, longitudinal fluctutations, etc.
- Molecular-Spin Dynamics
  - $\mathcal{H}_{MSD} = \mathcal{H}_l + \mathcal{H}_s = \sum_i \frac{\mathbf{p}_i{}^2}{2m} + U(\mathbf{R}) - \sum_{i,j}^{N} J_{ij}(\mathbf{R})[\vec{s_i} \cdot \vec{s_j} - 1] - \sum_{i,j}^{N} K_{ij}(\mathbf{R})\left[(\vec{s_i} \cdot \vec{s_j})^2 - 1\right]$ (Implemented in LAMMPS)



ML Framework for α-Fe

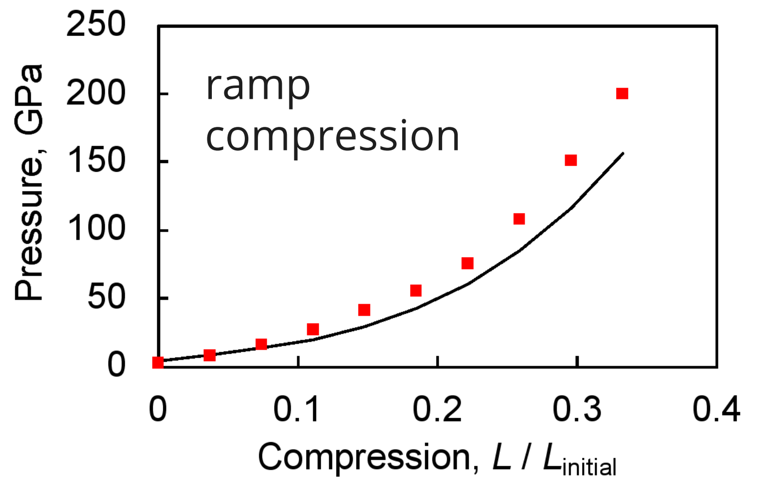
1) Generate ab-initio dataset for Fe (VASP)
   - Fe data generated for 0 - 1200K and 0 - 13 Gpa
   - Spin spiral data generated for different degrees of compression
2) Use genetic algorithm (GA) to parameterize spin Hamiltonian using spin spiral data
3) Subtract magnetic energies/forces/stress from DFT Fe data and train interatomic potential, $U(R)$, using GA
   - $U(R)$ built using spectral neighbor analysis potential (SNAP)
   - $E^i_{SNAP} = \beta_0 + \boldsymbol{\beta} \cdot \boldsymbol{B}^i$ and $F_j^{SNAP} = -\boldsymbol{\beta} \cdot \sum_{i=1}^{N} \frac{\partial \boldsymbol{B}_i}{\partial \boldsymbol{r}_j}$
4) Evaluate candidate on predetermined set of objective functions (OFs)
   - Using candidate SNAP potential and parameterized spin Hamiltonian
5) Continue GA until desired OFs accuracy is reached

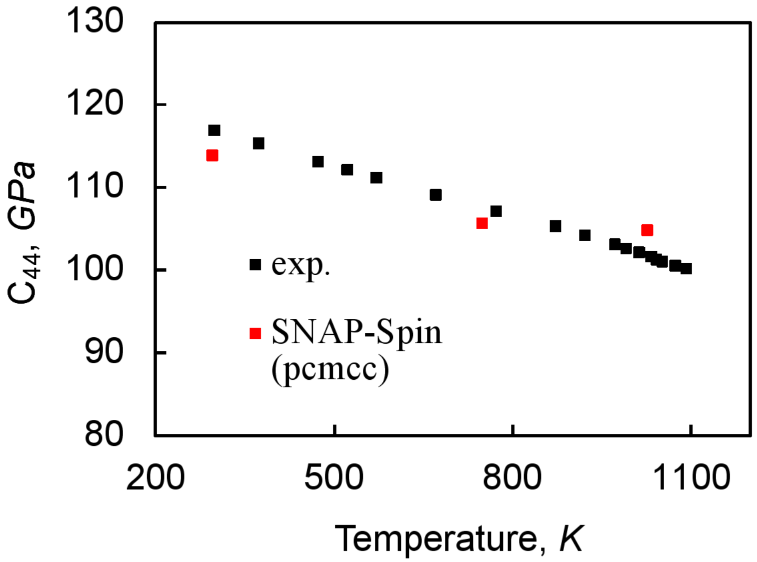


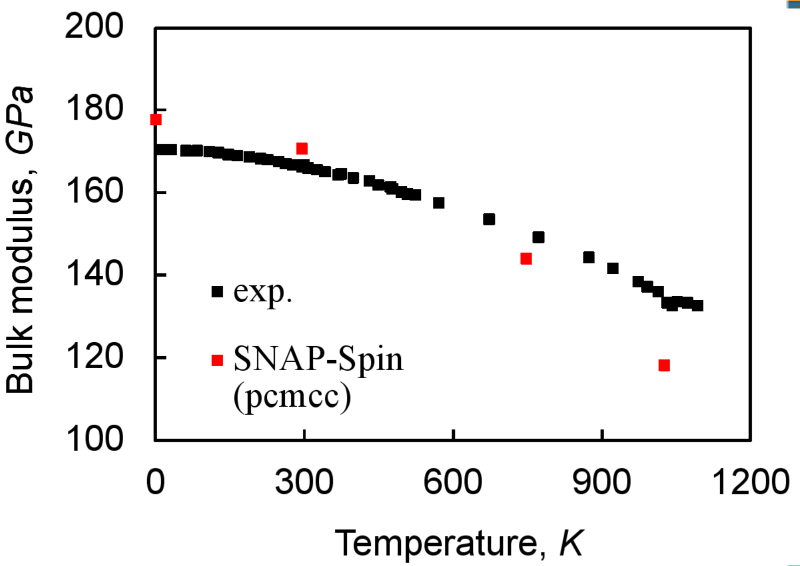
Shock Response of MSD Model

- Trained SNAP-Spin model on high pressure / temperature data
- Includes bcc / fcc / hcp collinear spin DFT (VASP) data
  - Up to 400 GPa and 6500K

0 K Objective Functions				
	MSD	Exp/DFT	Units	Error
C_{11}	243.3	239.6	GPa	1.5%
C_{44}	111.3	120.8	GPa	7.9%
Bulk modulus	177.8	169.6	GPa	4.8%
$(C_{11} - C_{12}) / 2$	42.7	51.9	GPa	17.7%
Poisson ratio	0.389	0.36	-	8.1%
FCC trans. press.	25.644	19	GPa	35%
HCP trans. press.	15.487	13	GPa	19%

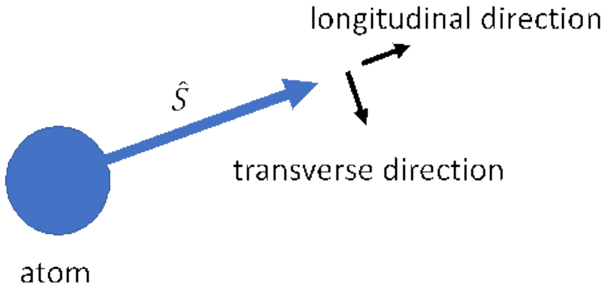


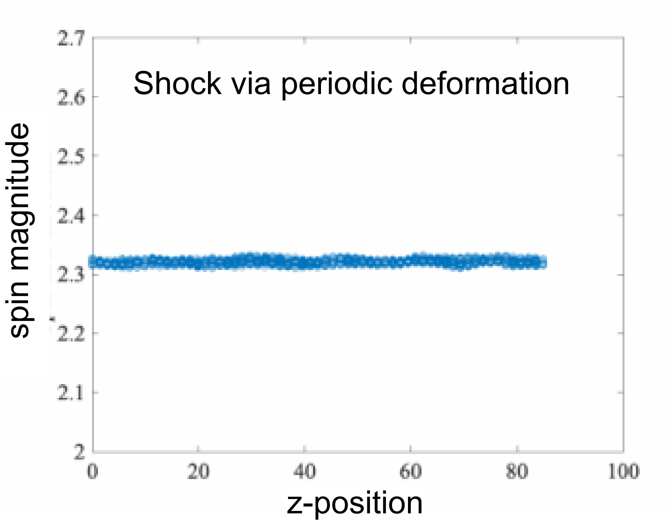


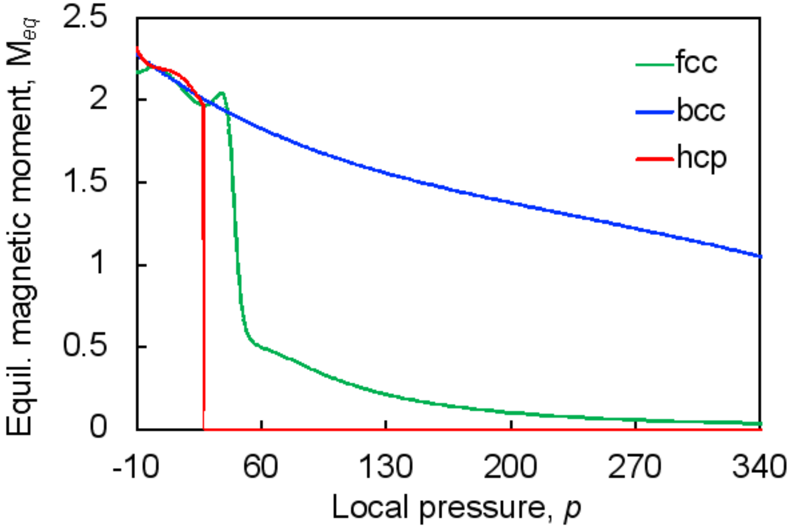


Shock Response of MSD Model

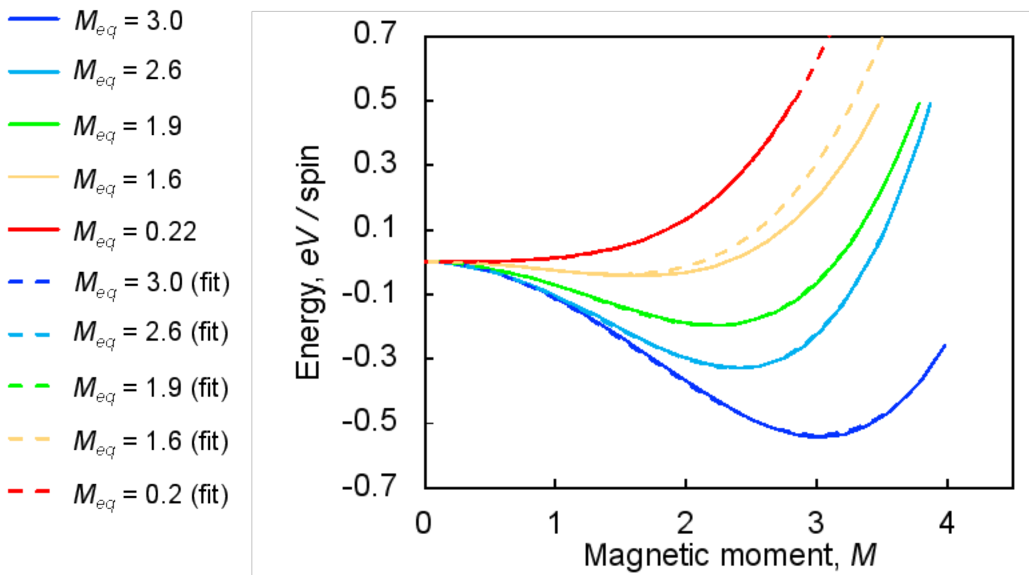
- Longitudinal changes in magnetic vector
  - $\mathcal{H}_{Landau} = \sum_i (AS_i^{\,2} + BS_i^{\,4} + CS_i^{\,6})$
  - $\hat{S} \rightarrow$ spin vector of atom
  - $\hat{S} = \frac{-\widehat{M}}{g\mu_B} = S_x\hat{\imath} + S_y\hat{\jmath} + S_z\hat{k}$
  - $S_i = \|\hat{S}\| = \sqrt{(S_x)^2 + (S_y)^2 + (S_z)^2}$
  - $A(p), B(p)$, and $C(p)$ are fitted to noncollinear spin data for iron†







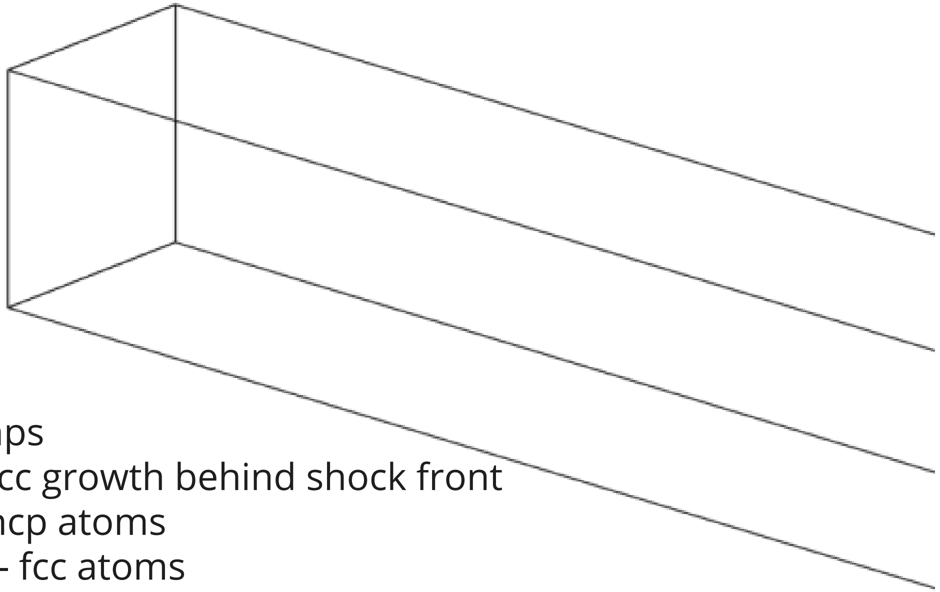
- Magnetic moment adjusted on the fly
  - Based on local pressure & phase



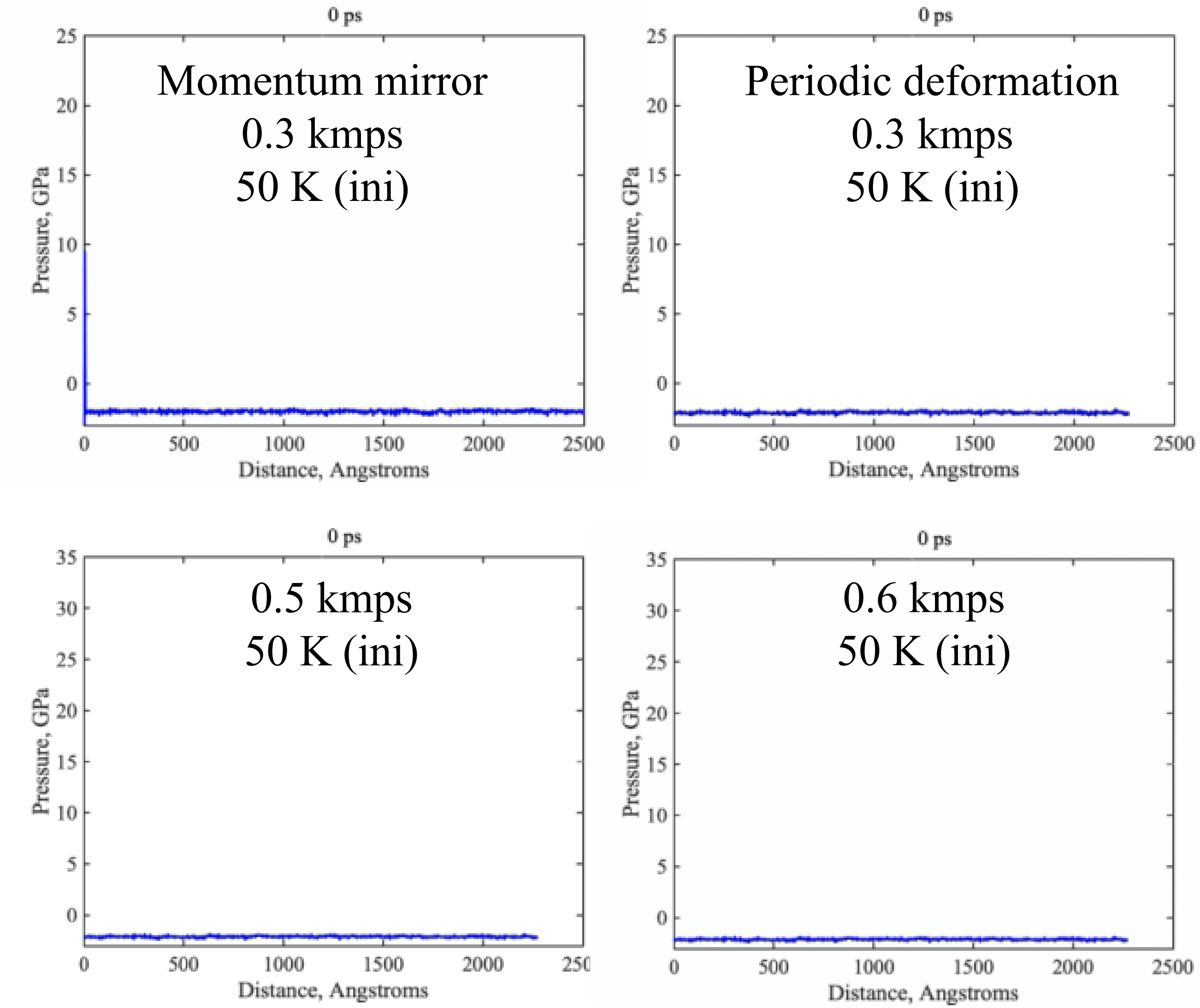
†Gambino, Davide, et al. "Longitudinal spin fluctuations in bcc and liquid Fe at high temperature and pressure calculated with a supercell approach." *Physical Review B* 102.1 (2020): 014402.

Shock Response of MSD Model

- Tested both two methods to generate shocks
  - Momentum-mirror
  - Uniaxial shrinking of periodic domain
- Response for shocks < 1 kmps
  - Below 0.6 kmps no phase transformations detected
  - Dual-wave profile observed for shocks above 0.6 kmps

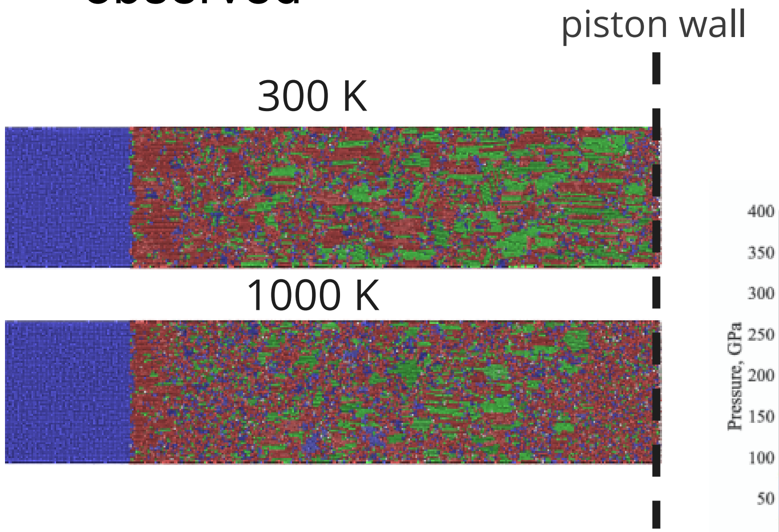


0.6 kmps
hcp / fcc growth behind shock front
red – hcp atoms
green - fcc atoms

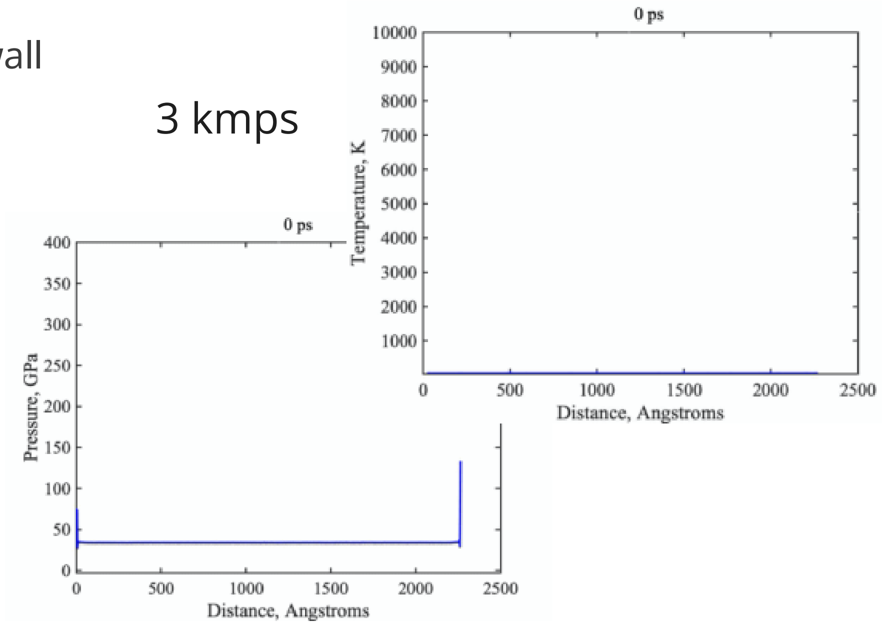


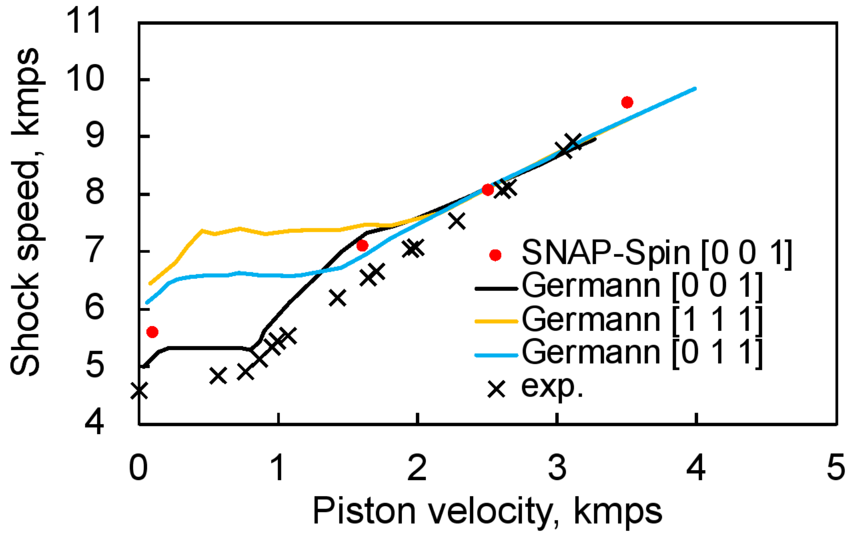
Shock Response of MSD Model

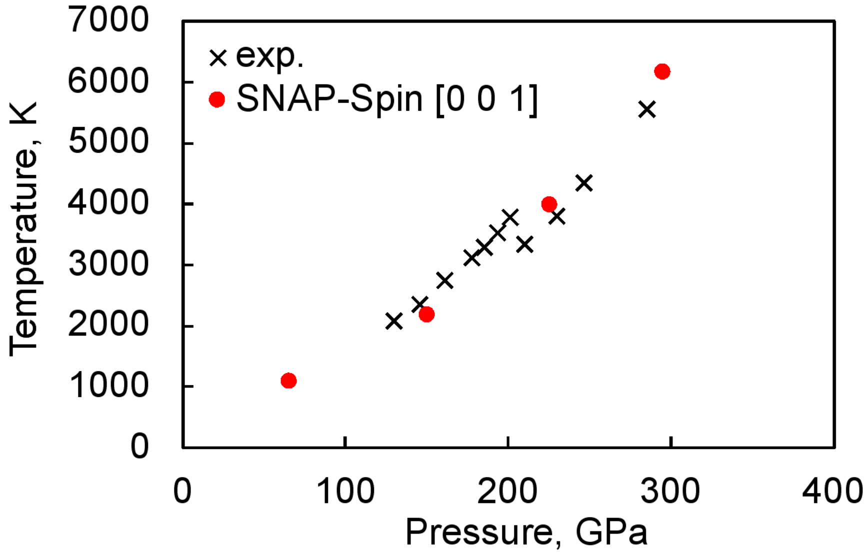
- Hugoniot curves for single crystal in good agreement with previous computational results and experiments
- Temperature / pressures in good agreement with experiments
- Potential was tested on 3-3.5 kmps and good stability was observed



blue – bcc atoms
red – hcp atoms
green - fcc atoms







Conclusion

- Illustrated framework for building α-Fe molecular-spin dynamics model from ab-initio data
- Showed impact of different equilibration methods on mechanical response of α-Fe
- Analyzed phonon/magnon spectra and thermal conductivity at corresponding temperatures
  - Good agreement with experiments
  - Able to reproduce acoustic peak shift up to ~700K
- Retrained potential for high temperature / pressure data
  - Good agreement for elastic properties / transition pressures / ramp curve
  - Dual-wave profile observed once bcc begins transforming into hcp phase