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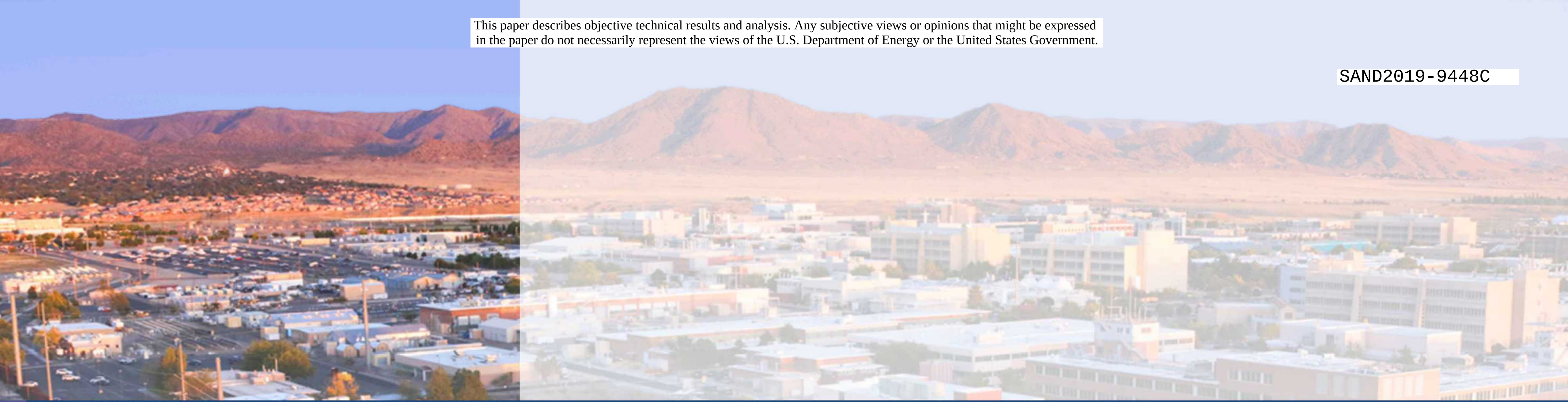

Extending *ab initio* Calculations with Machine Learned Potentials for Molecular Dynamics

SAND2019-####

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Z Fundamental Science Workshop (8/12/2019)

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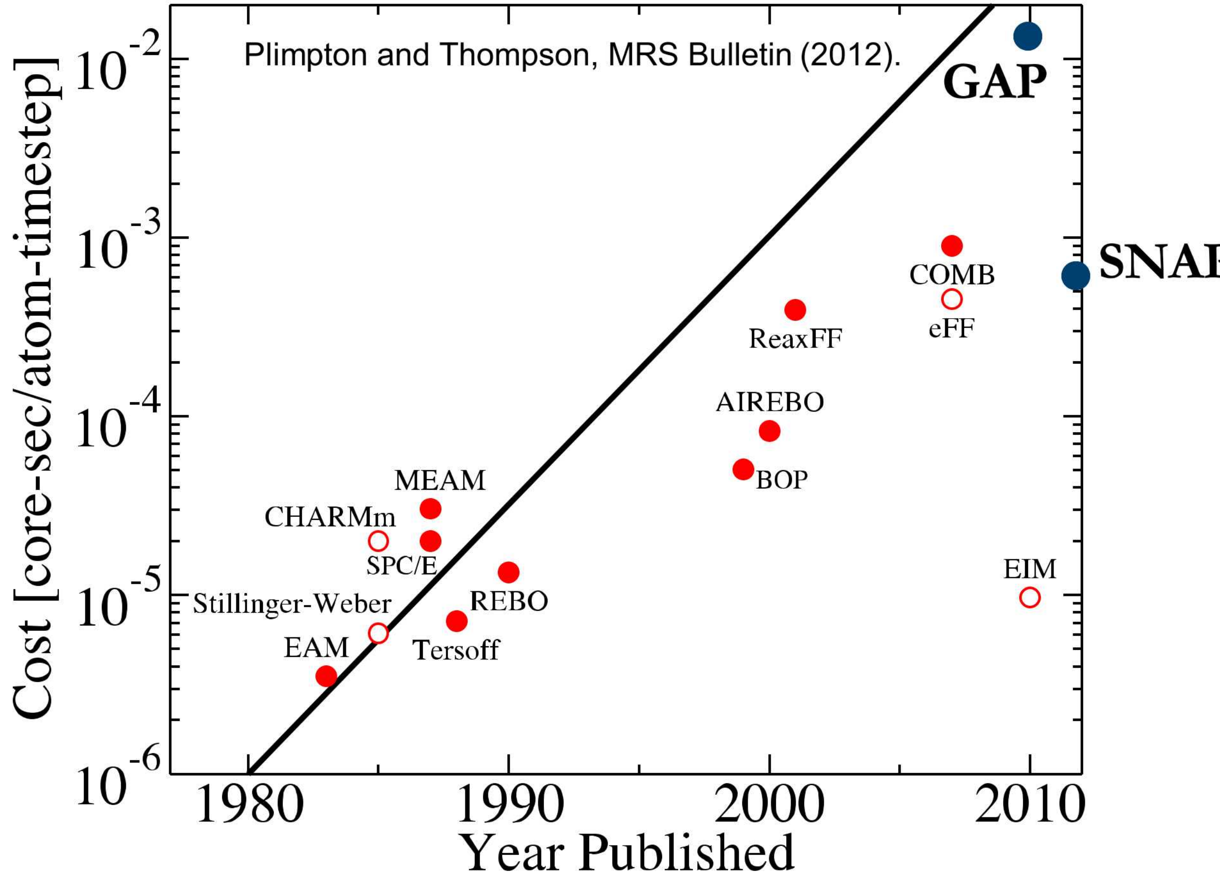
MD Approximations Change Over Time

Twobody (B.C.)
Lennard-Jones, Hard Sphere, Coulomb, Bonded

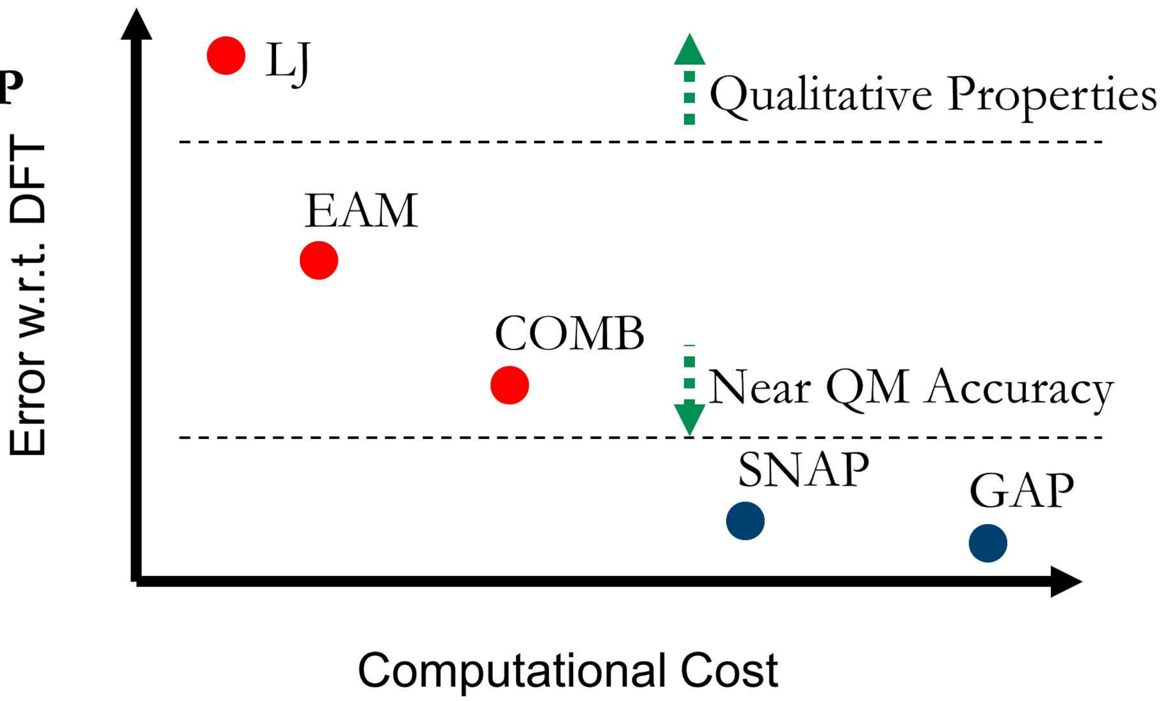
Manybody (1980s)
Stillinger-Weber, Tersoff, Embedded Atom Method

Advanced (90s-2000s)
REBO, BOP, COMB, ReaxFF

Big Data / Deep / Machine Learning (2010s)
GAP, SNAP, NN,…


Plimpton and Thompson, MRS Bulletin (2012).

Resources are limited, which is your best choice?



Machine Learning to Bridge Electronic-Atomic

Classical, Empirical Potentials

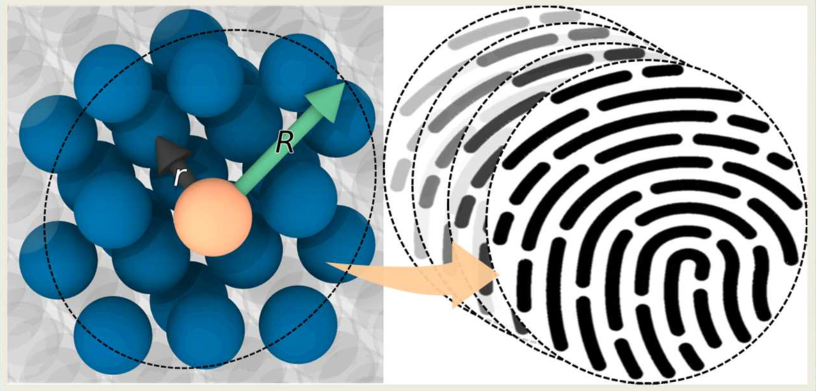
- Metals
    - EAM: Assume spherical electron density

$$E_i = F_\alpha\left(\sum_{j\neq i}\rho_\beta(r_{ij})\right) + \frac{1}{2}\sum_{j\neq i}\emptyset_{\alpha\beta}(r_{ij})$$

- Inorganic
    - Stillinger-Weber: Assume 2,3-body harmonic springs
- Organic
    - ReaxFF: Assume covalent bonding, smooth bond-orders between all interacting atoms

Machine Learned Potentials

- Metals, Inorganic, Organic, etc.
    - Assume energy and forces are some function of local atomic neighborhood descriptors
- Needs reference data to be properly trained to get the ‘right’ energies and forces



Spectral Neighborhood Analysis Potential (SNAP)

Model Form

- Energy of atom i expressed as a basis expansion over K components of the bispectrum (B^i_k)

$$E^i_{SNAP} = \alpha_0 + \sum_k \left[\alpha_k^{(1)}\left(B^i_k - B_{k_0}\right) \quad + \alpha_k^{(n)}(\dots)^n\right]$$

Regression Method

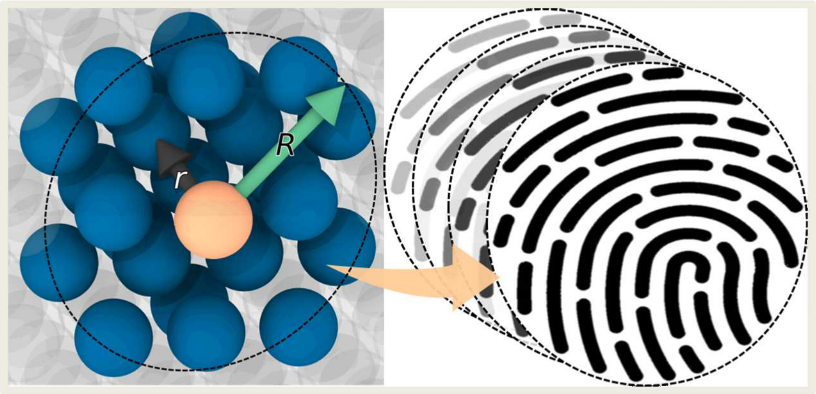
- α vector fully describes a SNAP potential
- Linear kernel decouples MD speed from training set size

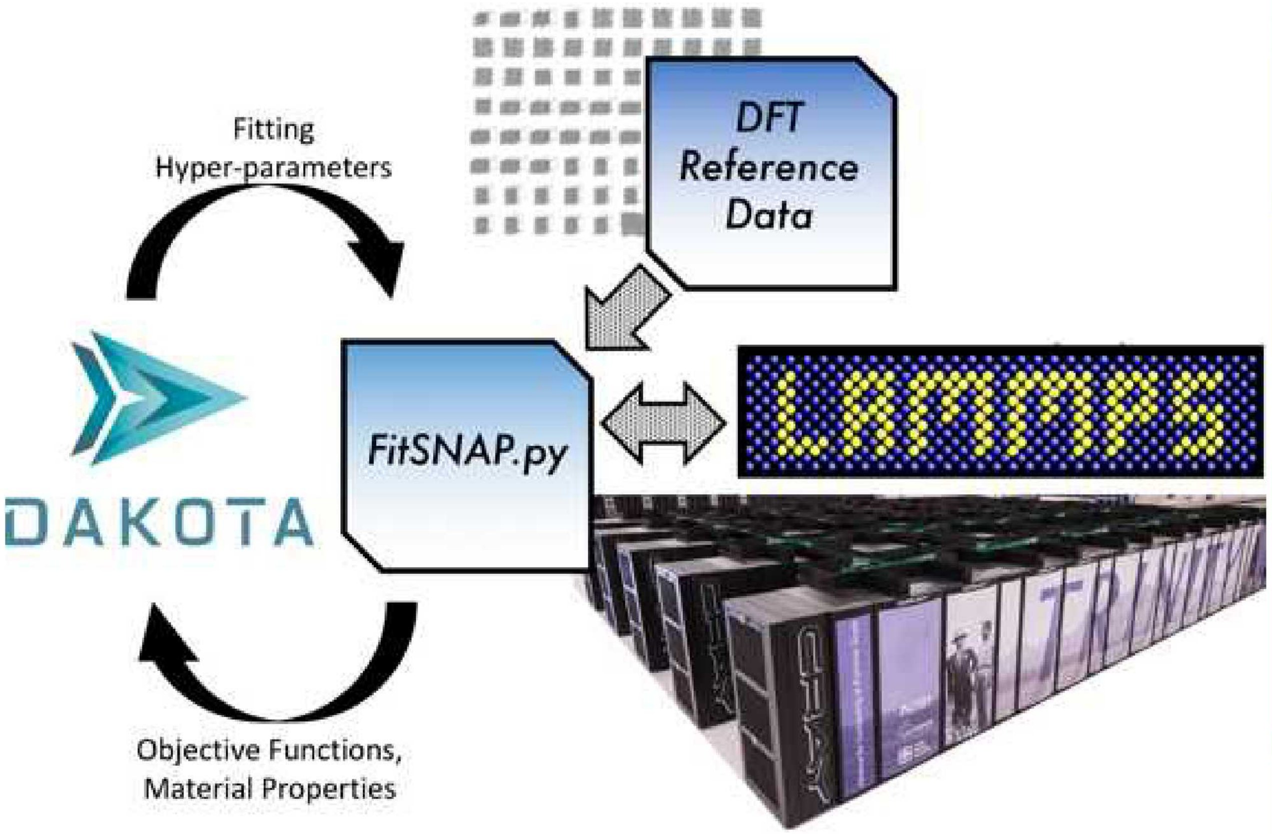
$$min(\|w \cdot D\alpha - T\|^2 - \gamma_n \|\alpha\|^n)$$

w : Weights

D : Set of descriptors

T : DFT training





Fitting Hyper-parameters

DFT Reference Data

DAKOTA

FitSNAP.py

LAMMPS

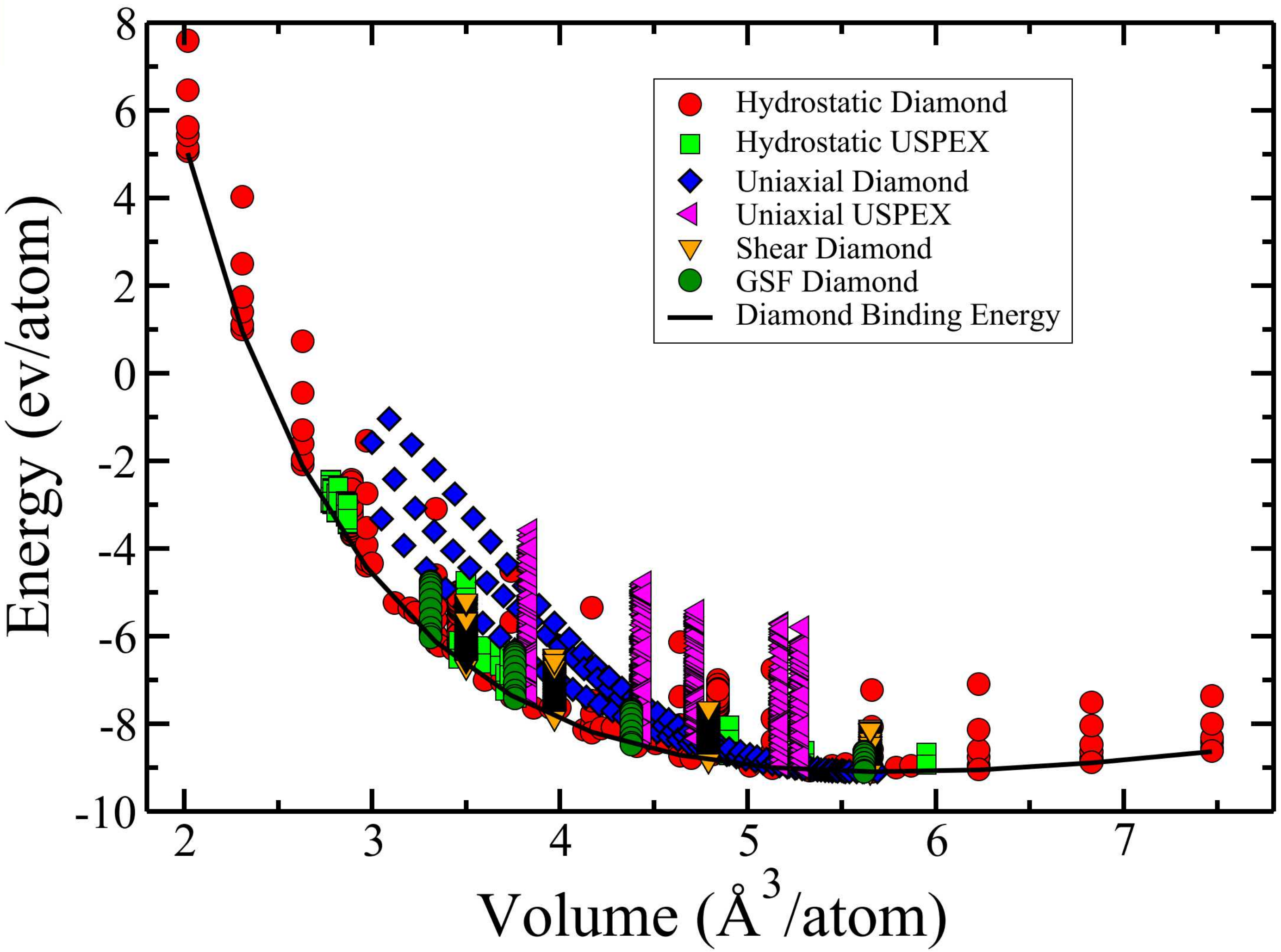
Objective Functions, Material Properties

ML-IAP for HED States of Carbon: Training

DFT Training Data

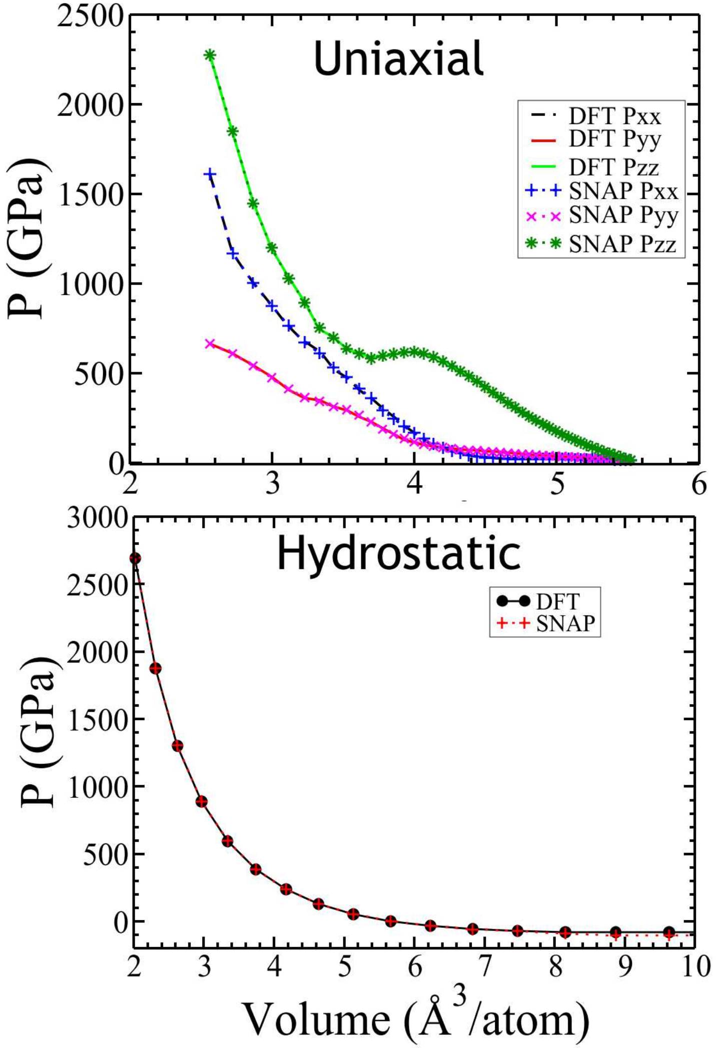
- Structures included (by-hand) were chosen for their relevance to HED experiments on Carbon
- Focus on high pressures, condensed phases (i.e. not fullerenes)
- Database details:
  - 7000+ configurations
  - Pressures between 0-1TPa
  - Temperatures between 0-20,000K
  - Diamond, Graphene, BC8 and thousands of crystal search results.

In collaboration with Ivan Oleynik at USF



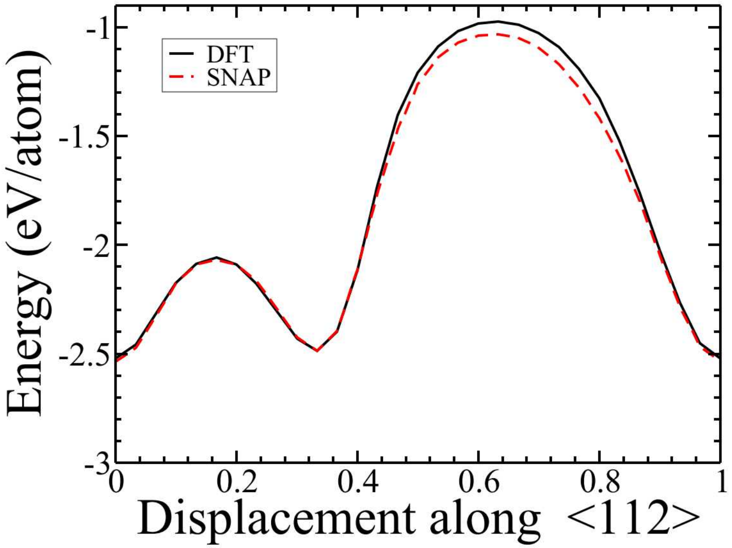
ML-IAP for HED States of Carbon: Material Properties

Elastic Properties: ✓

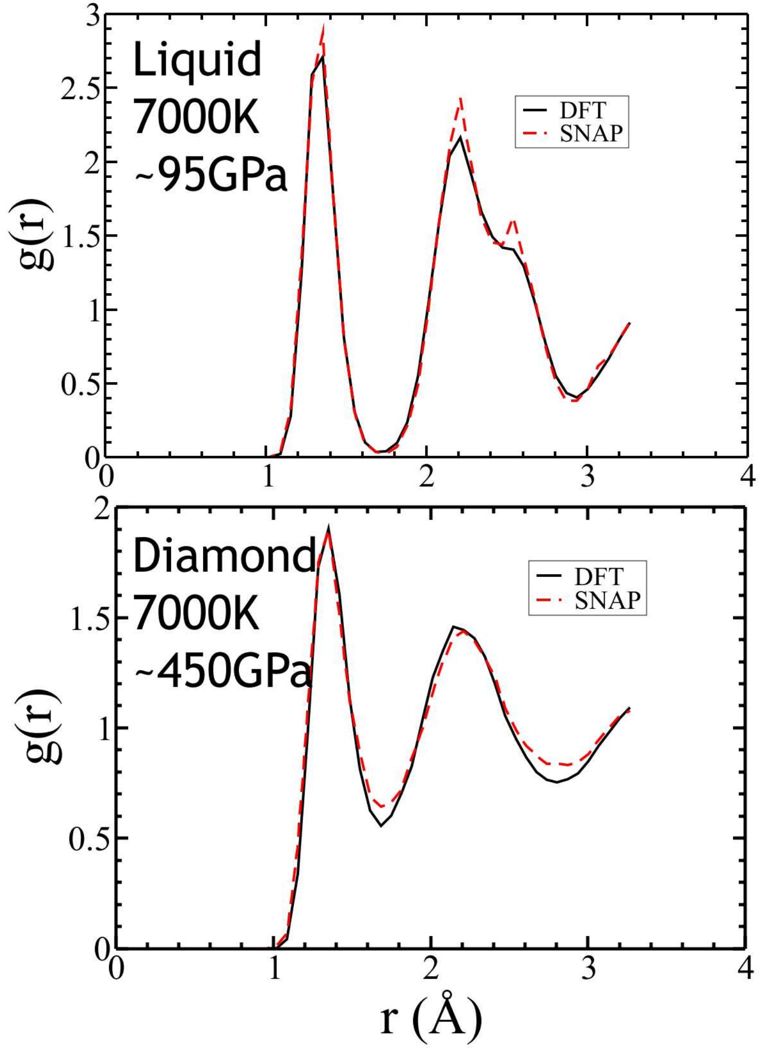


Defect Properties: ✓

Generalized Stacking Fault at 1TPa



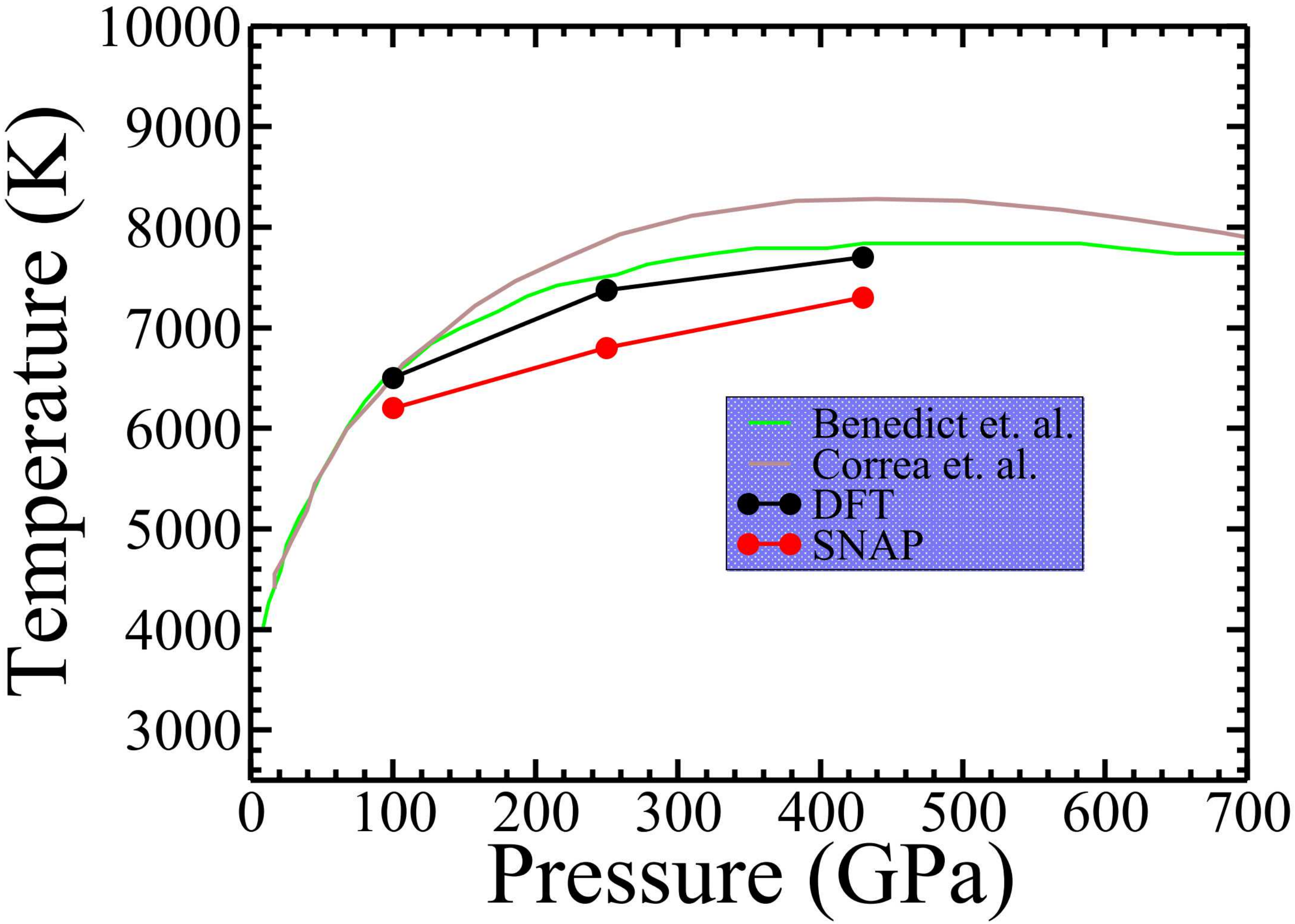
HED State Properties: ✓/?



ML-IAP for HED States of Carbon: Extrapolation

Beyond Training

- Diamond melt curve using 2-phase, stable interface, MD simulations
- Slopes look good w.r.t. DFT, max error is 7% at P=250GPa
- Still some discrepancy, should new training data be added?
  - ○ Improve agreement at these state points
  - ○ Temper the system from bad dynamics at very high P,T
  - ○ Sacrifice accuracy elsewhere in the fitted properties

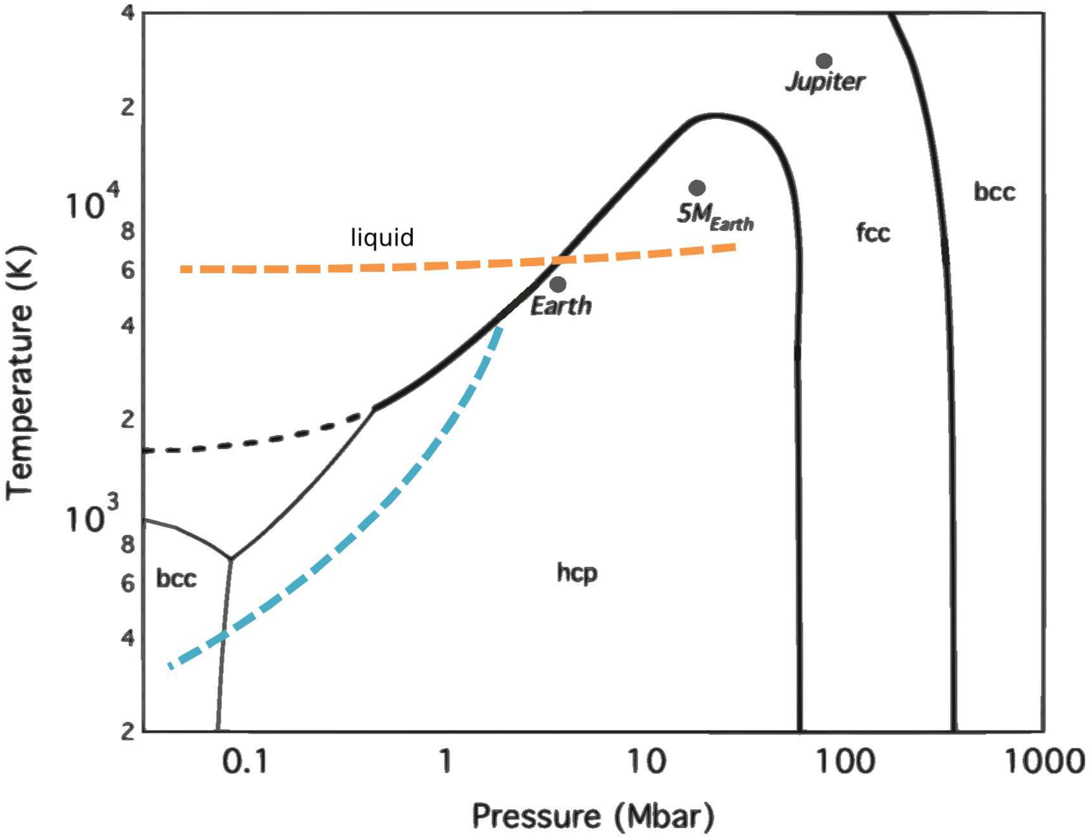


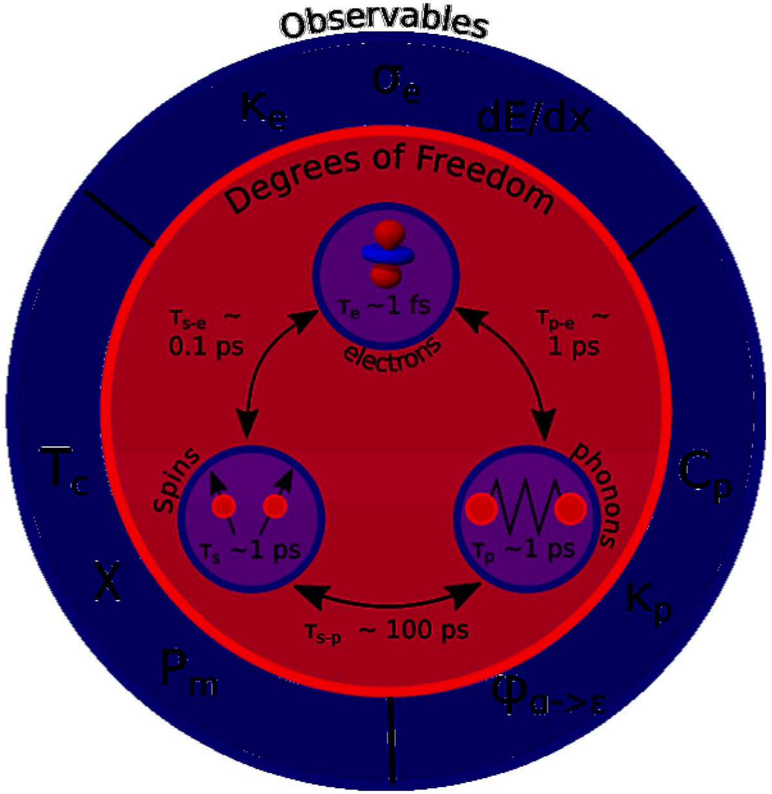
Correa et. al., PNAS 103, 0510489103 (2005) Benedict et. al., Phys Rev 89, 224109 (2014)

Missing Physics/Chemistry in Current Comp. Tools

Approaching HED states via different physical processes:

1. Shock compression, coupled Pressure & Temperatur
    - Final HED state is determined by initial condit
2. Ramp compression, nearly isothermal Pressure rise





Phase transitions occur along these paths:

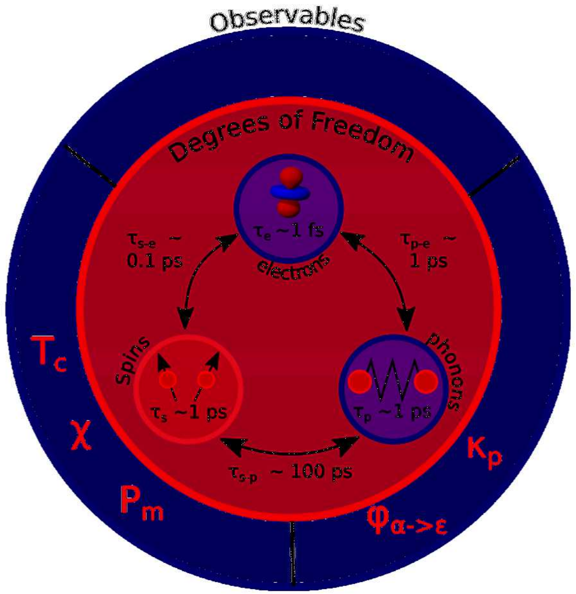
Structural : BCC to HCP, melting & solidification

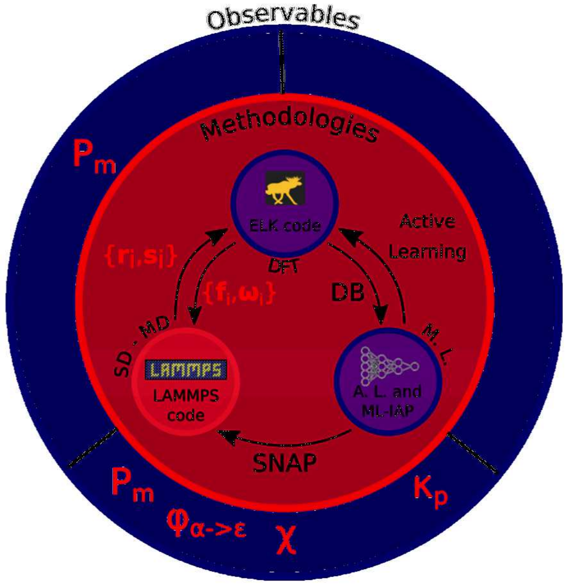
Magnetic : Ferro- to Non-magnetic

Electronic & Thermal : Discontinuous transport properties

LDRD-20-0014: A. Cangi, J. Tranchida, M. Wood, M. Desjarlais

Molecular Dynamics with Extra Degrees of Freedom





Classical spin dynamics

- Ruled by a per-atom resolution of the Landau-Lifschitz-Gilbert (LLG) equation:

$$\frac{d\boldsymbol{s}_i}{dt} = \frac{1}{(1+\lambda^2)}\left[\boldsymbol{\omega}_i \times \boldsymbol{s}_i + \lambda\, \boldsymbol{s}_i \times (\boldsymbol{\omega}_i \times \boldsymbol{s}_i)\right]$$

With $\mathbf{s}_i$ classical magnetic spins, lambda a damping coefficient and $\boldsymbol{\omega}_i$ precession vectors derived from a spin Hamiltonian.

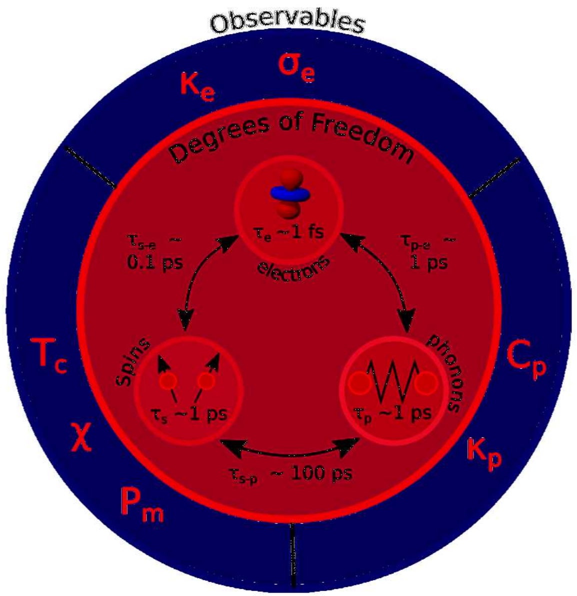
Spin Hamiltonian for HED conditions

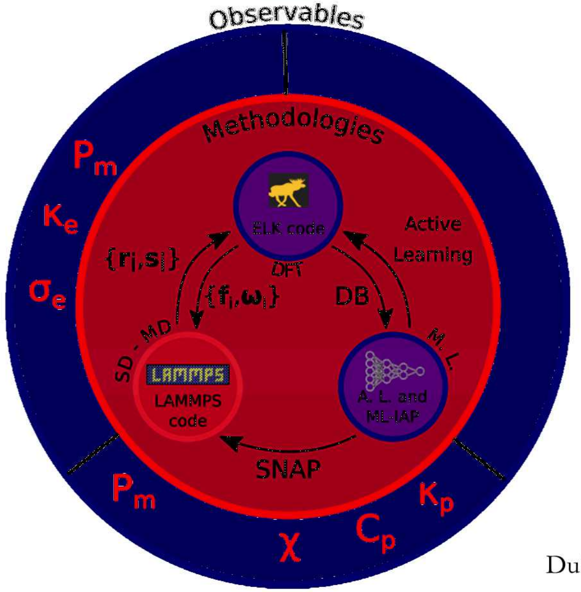
$$\boldsymbol{H}_{ex} = -\sum_{i}^{N}\sum_{n=1}^{4} I_i^n \left|\boldsymbol{s}_i\right|^{2n} - \sum_{i,j,i\neq j}^{N} \boldsymbol{s}_i \boldsymbol{s}_j \left(J_{ij} + K_{ij}\left|\boldsymbol{s}_i\right|\left|\boldsymbol{s}_j\right| + L_{ij}\boldsymbol{s}_i\boldsymbol{s}_j\right)$$

- Contains transverse and longitudinal spin fluctuations ‘necessary for high P/T conditions.
- Will be implemented in LAMMPS.

Surh, M. P. et al. (2016). Magnetostructural transition kinetics in shocked iron. Phys. Rev. Lett., 117(8), 085701.

Molecular Dynamics with Extra Degrees of Freedom



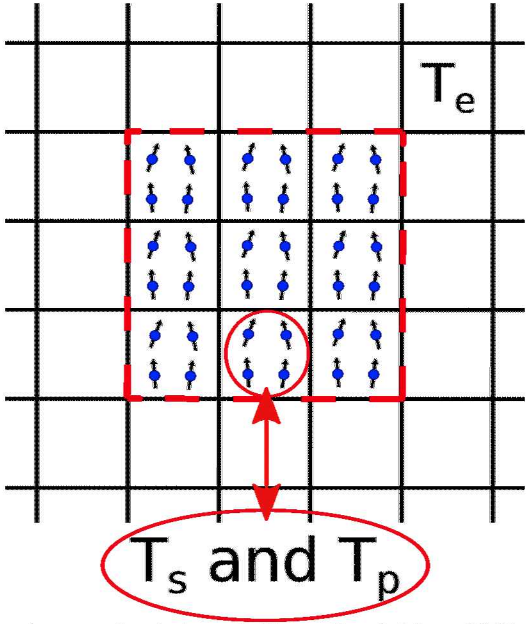


Diffusion equation for the electronic temperature

$$C_e \frac{\partial T_e}{\partial t} = \nabla \left(\kappa_e \nabla T_e \right) - g_{e-s}(T_e - T_s) - g_{e-p}(T_e - T_p)$$

with T_e, T_s and T_p the electronic, spin and lattice temperatures, C_e and $\varkappa_e$ the electronic heat-capacity and heat-conductivity, and g the electron-spin and electron-lattice relaxation rates.

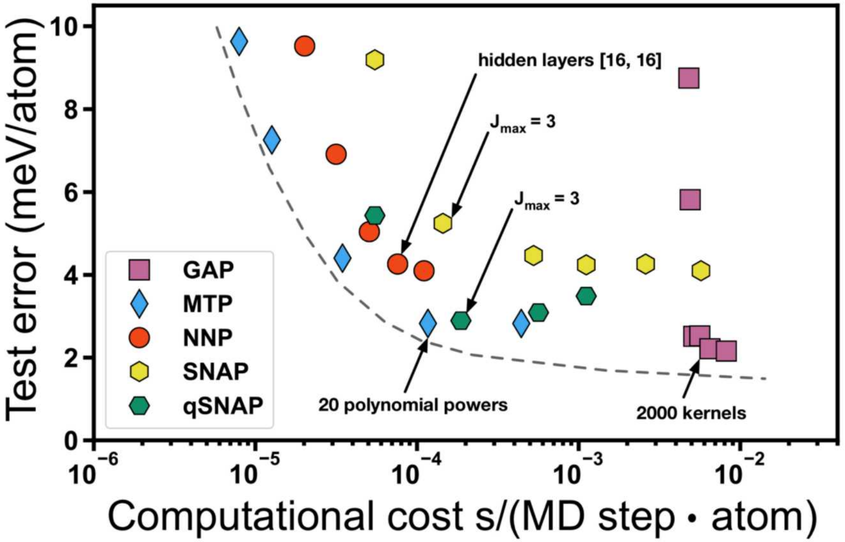
- Resolution on a grid of the diffusion equation for Te (in black on the right).
- This grid is overlay on top of a spin-lattice MD simulation cell (in red).
- Langevin thermostats, exchange coefficients and spin-lattice Hamiltonian are coupling the evolution of the three temperature.
- Model parametrized from ab initio calculations.



Duffy, D. M., & Rutherford, A. M. (2006). Including the effects of electronic stopping and electron–ion interactions in radiation damage simulations. J. Phys.: Cond. Matt., 19(1), 016207.

Conclusion + Ongoing SNAP Development Areas

- SNAP development is targeting a space where few other potentials exist, or where traditional potentials lack quantitative accuracy.
- Biggest worry about generated potentials is how ‘robust’ they are.
- Exploring accuracy/cost tradeoffs with other ML-IAP



ECP – EXAALT : Extending MD Time & Accuracy

- InP : Explicit multi-element bispectrum
- Neural Nets using bispectrum descriptors
- Learn-on-the-Fly methods for training curation

SciDAC-4: Plasma-Surface Interactions

- W-Be-H-He