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Molecular Dynamics of High Pressure Tin Phases I:

Strength and deformation evaluations of empirical potentials

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Shock Compression of Condensed Matter Conference (SHOCK 2022)
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SAND 2022-XXXX

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Strength in materials, tri-lab effort

Strength is a measure of a material's ability to sustain an applied load without failure or irreversible deformation.

Strength response is "universal"
but mechanisms are unique to each system.

In the hydro code world,
EOS → controls volume compression
strength → controls deformability

In the real world,

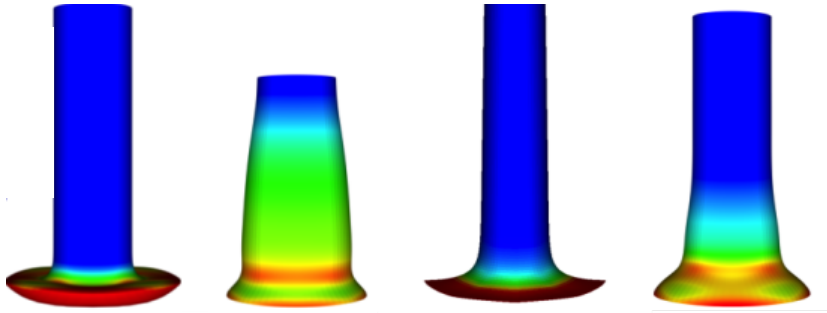
- Dislocation motion
- Dislocation generation
- Grain boundary motion
- Twinning and stacking faults
- Phase transition

History and path dependence

Microstructure

- Grain size
- Grain orientation
- Grain boundary orientation

Compressive plastic flow stress in textured polycrystalline metal (ignoring anisotropy)



Why tantalum?

Tantalum, as a high-Z **body-center-cubic (bcc) metal** with **no experimentally observed high-pressure phase transitions** up to 350 GPa, has potential use a standard for high-pressure studies. But, its properties depend on poorly understood elastic/plastic and dislocation dynamics. High melt temperature of 3290 K.

M. B. Prime, et al., "A broad study of tantalum strength from ambient to extreme conditions," *Acta Materialia*, 231, 117875 (2022)

Motivation: Multiphase multiplies the unknowns

Multi-phase problems have so many more unknowns, we'd like to have a tool to constrain some open questions related to microstructure and twin & dislocation behavior.

We want an atomistic scale perspective on aspects of strength.

DFT high accuracy, but expensive

Classical MD allows scaling to address larger length and time scale behavior

Continuum is broadly applicable to macroscopic experimental test scales

Some multi-scale questions accessible to atomistic study:

What lattice-specific behavior influences dislocation production/mobility and/or twinning?

Do the phase transformations wipe-out, modify or preserve grain size and orientation?

Does plastic strain reset at phase transition? If so under what conditions?

Tin is the material chosen for the effort

- Non-hazardous
- Multiple accessible solid phases at relatively low pressures

J. McNaney, J04.00001, "Overview and highlights of a tri-lab effort on multi-phase material strength," July 12th

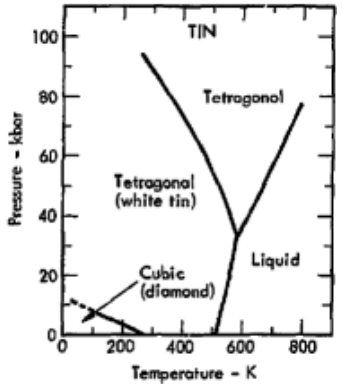
M. Prime, L05.0001, "Multiphase Strength Coupled with Phase Change Kinetics in RMI Experiments on Tin Across the β-γ Boundary," July 12th

W. Schill, O03.0001, "Simultaneous Bayesian calibration of strength, kinetics, and phase boundaries," July 13th

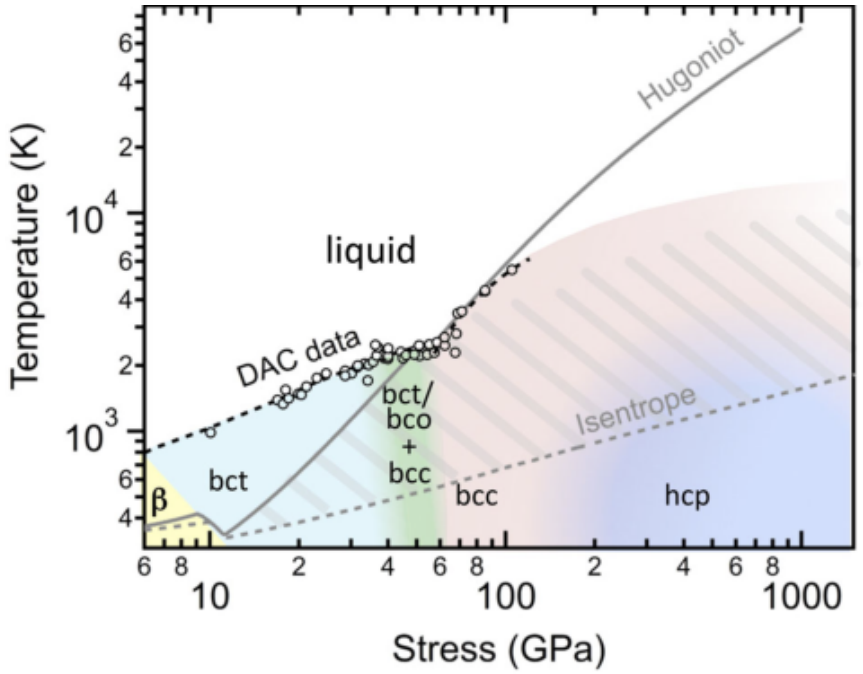
C. Battaile, O05.0004, "Measuring the Strength of Metals by Extending the Richtmyer-Meshkov Instability to Shockless Loading," July 13th

Tin phase diagrams from experiments and dft

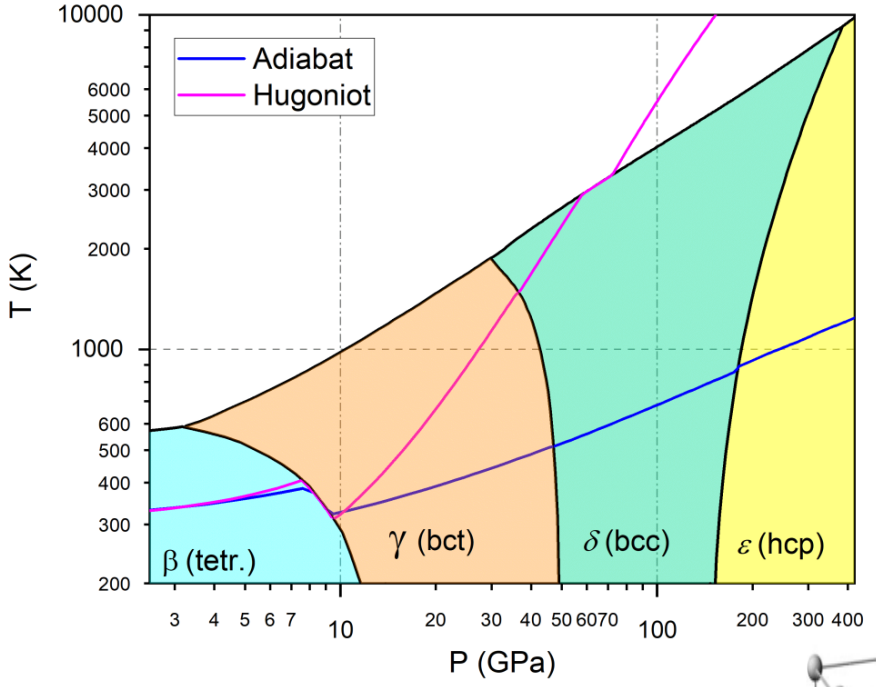
Young, LLNL (1975)

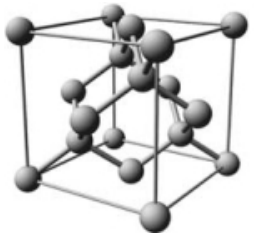


Lazicki, et. al., PRL 115 (2015) 075502



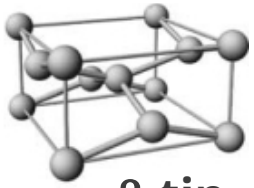
Rehn, et. al., PRB 103(2021) 184102





α-tin
Diamond
Below 300 K
Non-conducting

Diffusion-less

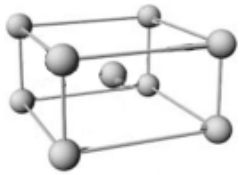


β-tin
Tetragonal
0 to ~8 GPa
Non-conducting
27% volume change

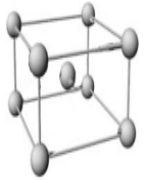
Diffusion driven

γ-tin
bct
~8 to ~40 GPa
Non-conducting
3-5% volume drop

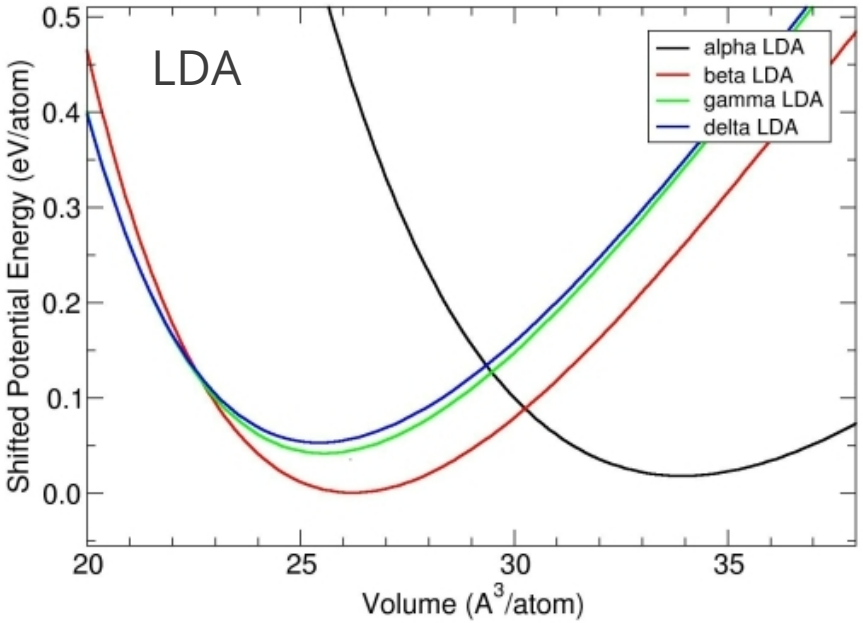
Diffusion-less

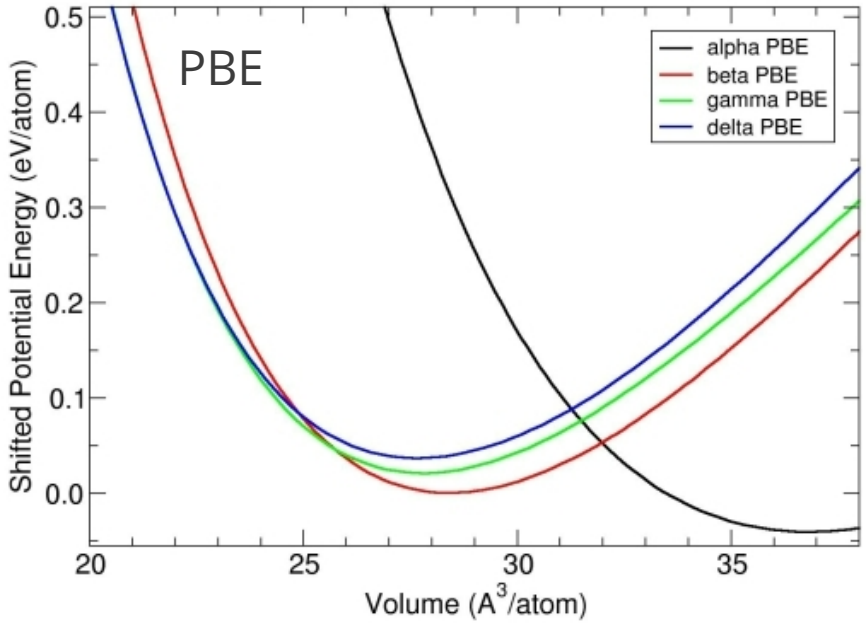


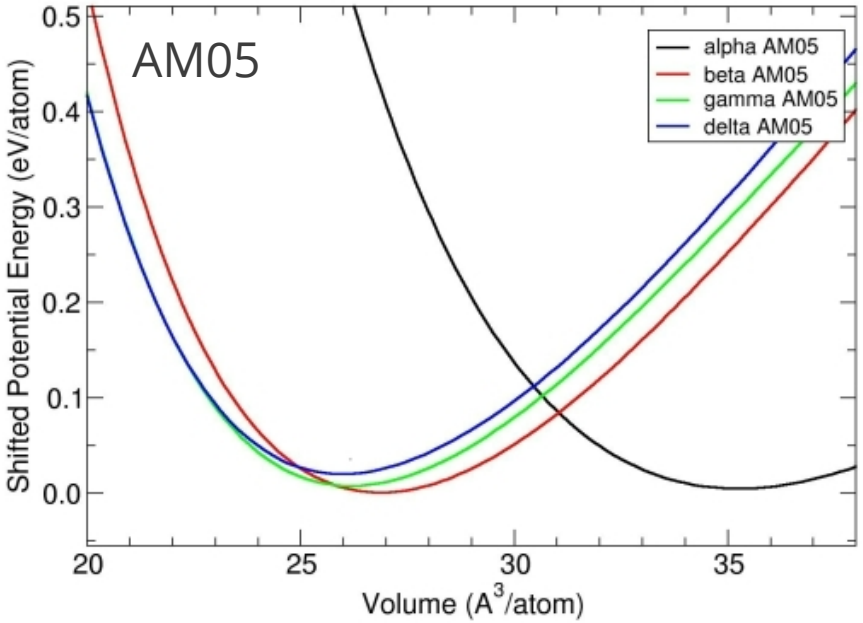
δ-tin
bcc
~40 to 150+ GPa
Non-conducting
No volume drop

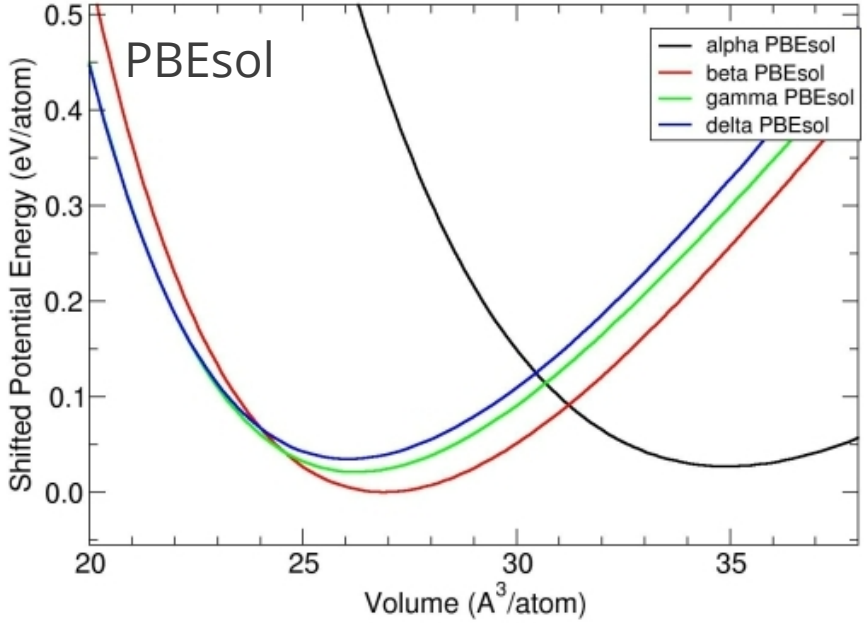


DFT cold curves and uncertainty between functionals



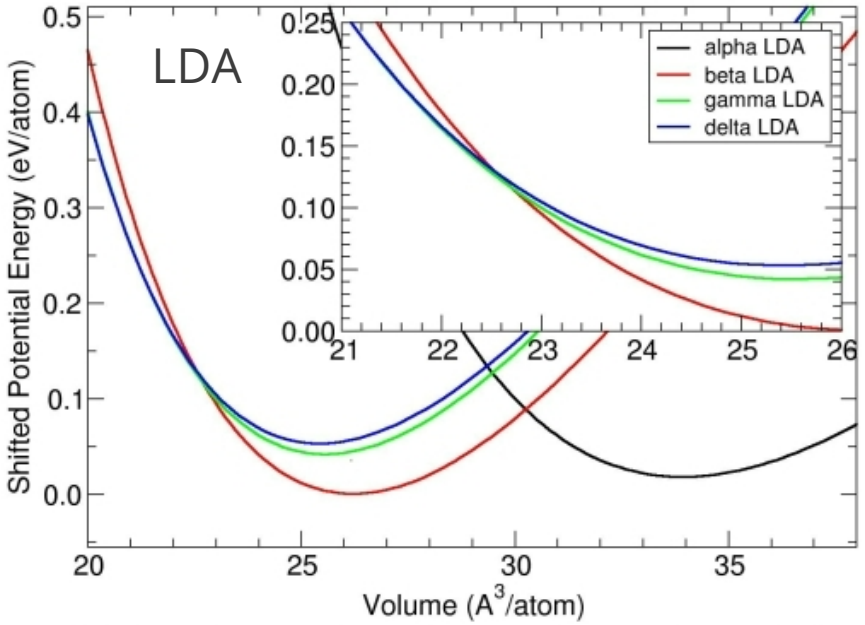


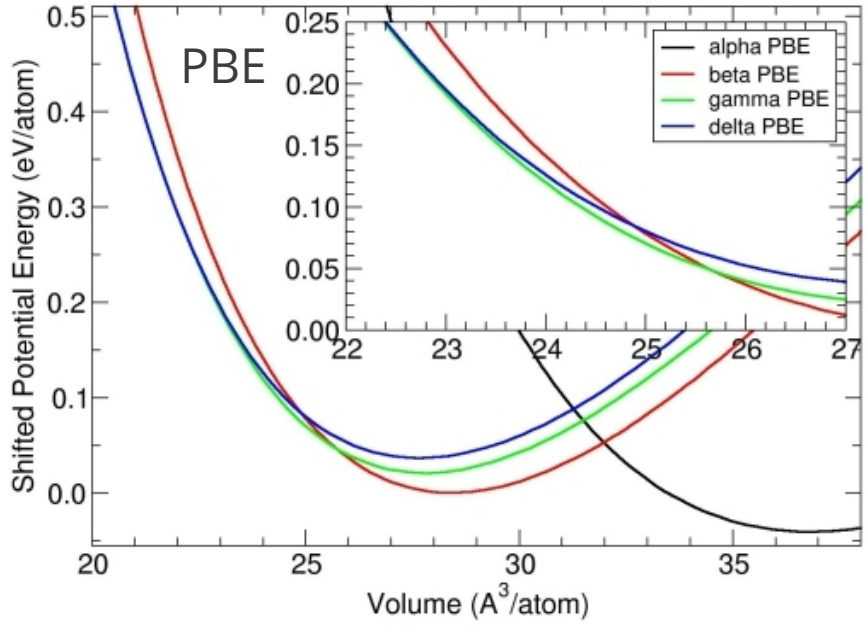


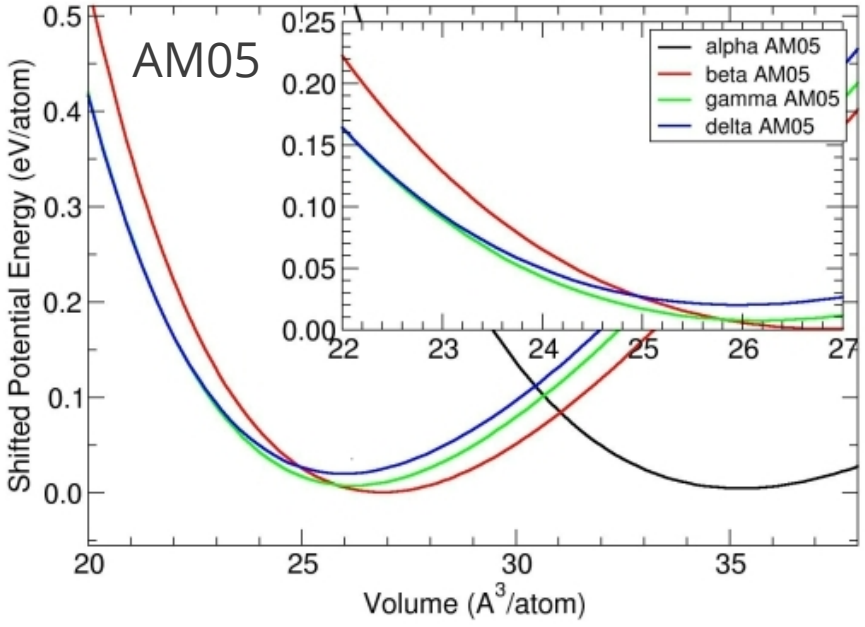


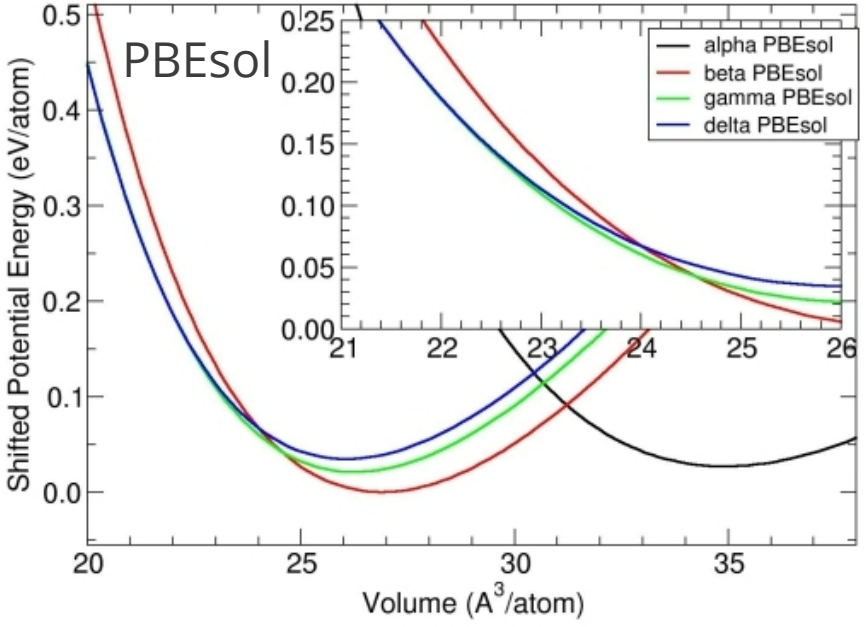
- Energy-volume curves are highly dependent on exchange-correlation functional used
- Only PBE predicts the alpha phase to be lowest in energy at 0 K
- Beta, gamma, and delta curves are also very similar to each other

DFT cold curves and uncertainty between functionals









Transition order depends on subtle energy differences

P (GPa)	AM05	LDA	PBE	PBE sol	Expt.
$\alpha > \beta$	1.5	-5.4	-2.9	-2.5	1
$\beta > \gamma$	-2.1	-3.6	-0.5	-0.3	14
$\gamma > \delta$	36.7	4.8	12.3	36.7	50

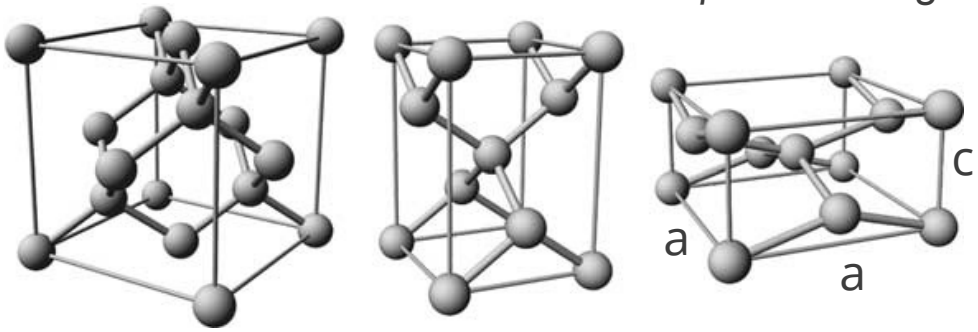
The issue is resolved with energy shifts in EOS models, but that's not an option here.

None of the functionals predict the experimental transition pressures

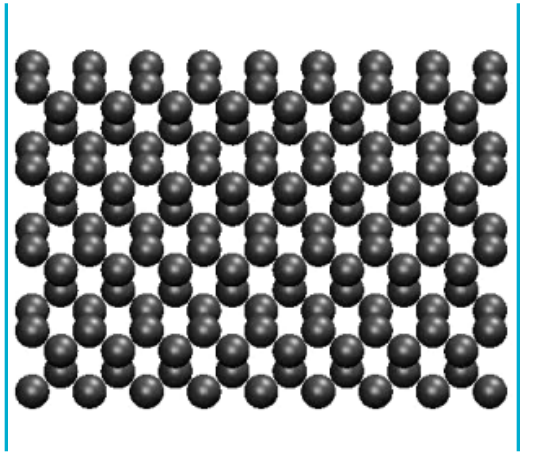
Low Pressure phases of tin

α = Diamond = cd

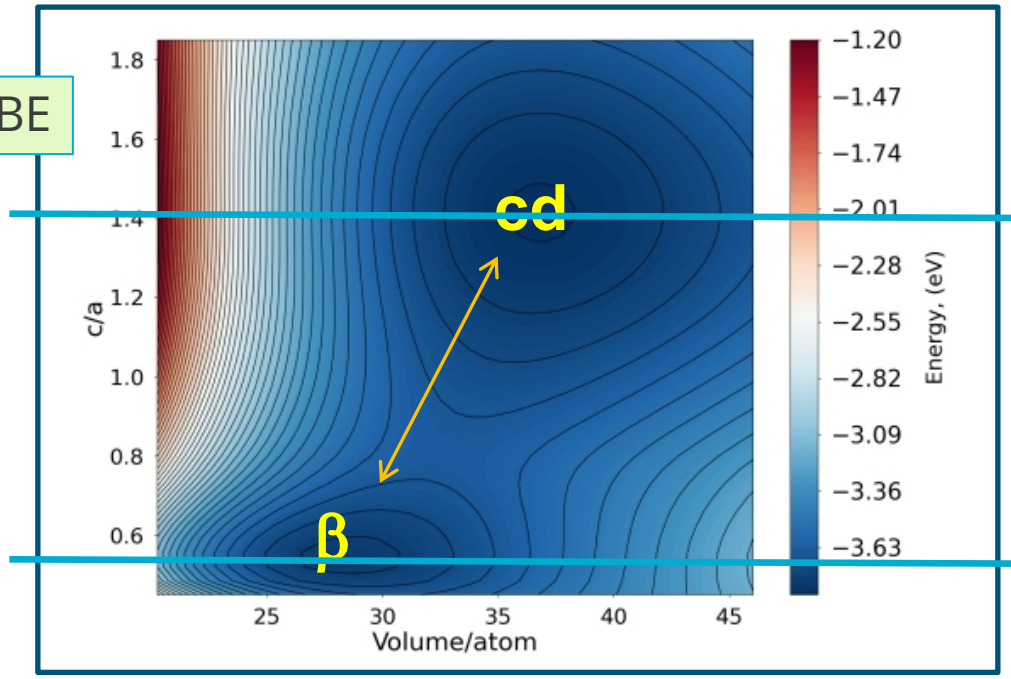
β-tin = tetragonal



Diffusion-less phase transition



DFT PBE



α – diam

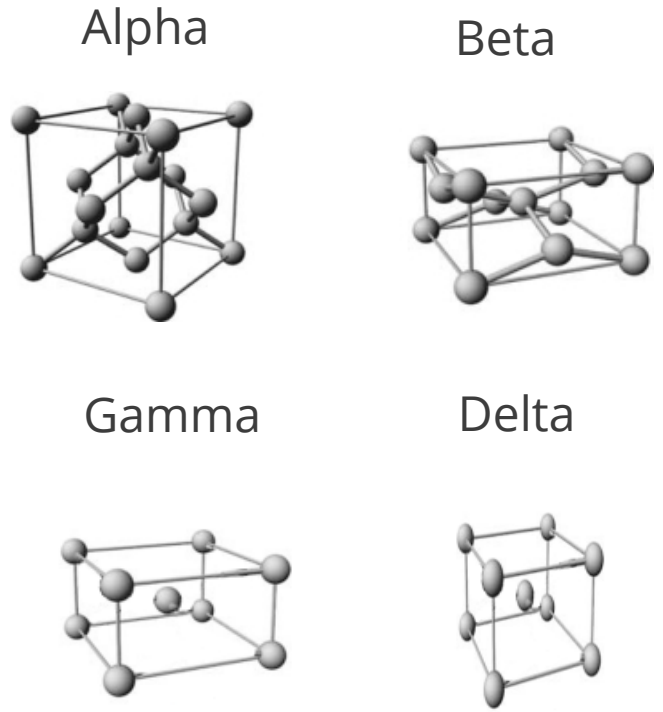
β - tetragonal

Tin is a ductile metal with signatures of ionic and covalently-bonded materials

MEAM, Tersoff, 3-body potentials

Low pressure states are open structures

Empirical potentials



EAM and MEAM potentials have been successful

1998 Ravelo/Baskes MEAM	alpha, beta and liquid
2017 Vella & Chen MEAM	liquid
2018 Etesami MEAM	alpha, beta, liquid
2018 Ko 2NN MEAM	alpha, beta
2006 Foiles EAM with Tersoff	liquid and gamma
2014 Sapzhnikov EAM	gamma, delta and liquid

Tin is a challenge because it exhibits open structures and close-packed structures.

As recently as 2020 and 2021, Ravelo MEAM potential has been the basis of numerous shock studies, which emphasize coarse Hugoniot response rather than structure details.

SNAP and ANI potentials could be quickly trained from DFT (See Talk II)

- Different DFT functionals give wide variety of transition pressures and even transition ordering
- The approach and any issues with fitting a multi-phase material like tin would generalize to other materials

LAVA tool to evaluate behaviors in LAMMPS & VASP

https://github.com/lanl/LAVA

The Lava Wrapper is a general-purpose calculator from LANL that provides a python interface to enable one-click calculation of the many common properties with lammps and vasp.

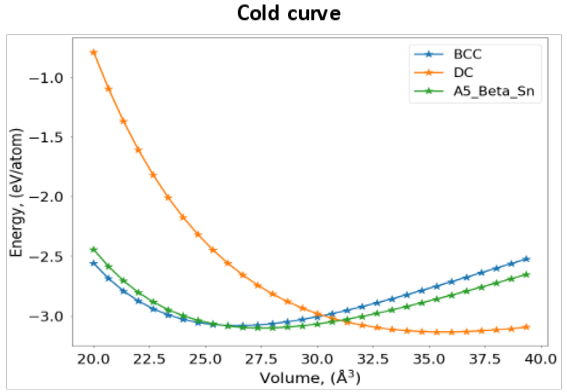
It provides a set of classes and functions to generate configs, run lammps/vasp calculation, retrieve the output, postprocess and plot the results.

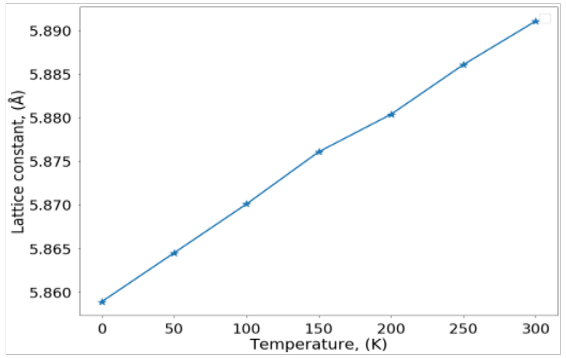
beta-Sn

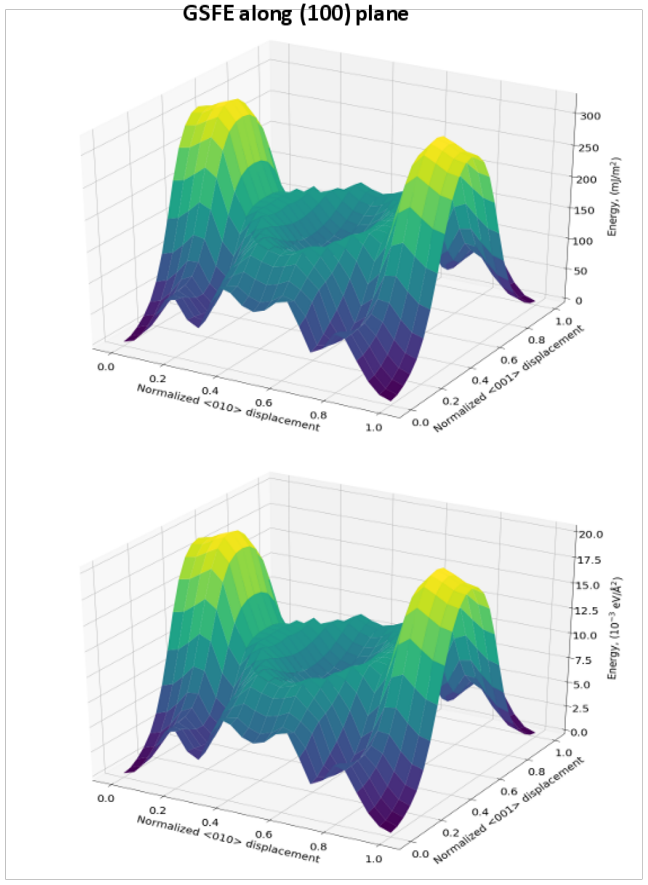
a (A)	5.8589
c/a	0.5472
Ecoh (eV/atom)	-3.1019
C11 (GPa)	119.795
C12 (GPa)	16.611
C13 (GPa)	36.911
C33 (GPa)	93.694
C44 (GPa)	9.485
B (GPa)	51.006
Esuf_100 (mJ/m2)	345.025
Esuf_101 (mJ/m2)	366.551
Evac (eV)	0.855
Eint (eV)	1.715

Thermal expansion

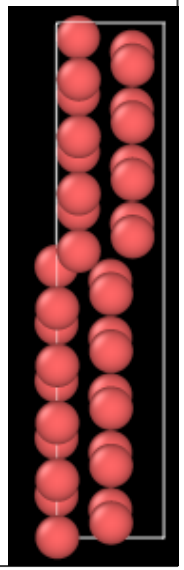
#Temp	average_lat	d_lat(%)
0	5.8589	0.0006
50	5.8645	0.0964
100	5.8701	0.1918
150	5.8761	0.2934
200	5.8804	0.3667
250	5.8861	0.4637
300	5.8911	0.55







Example GSFE structure



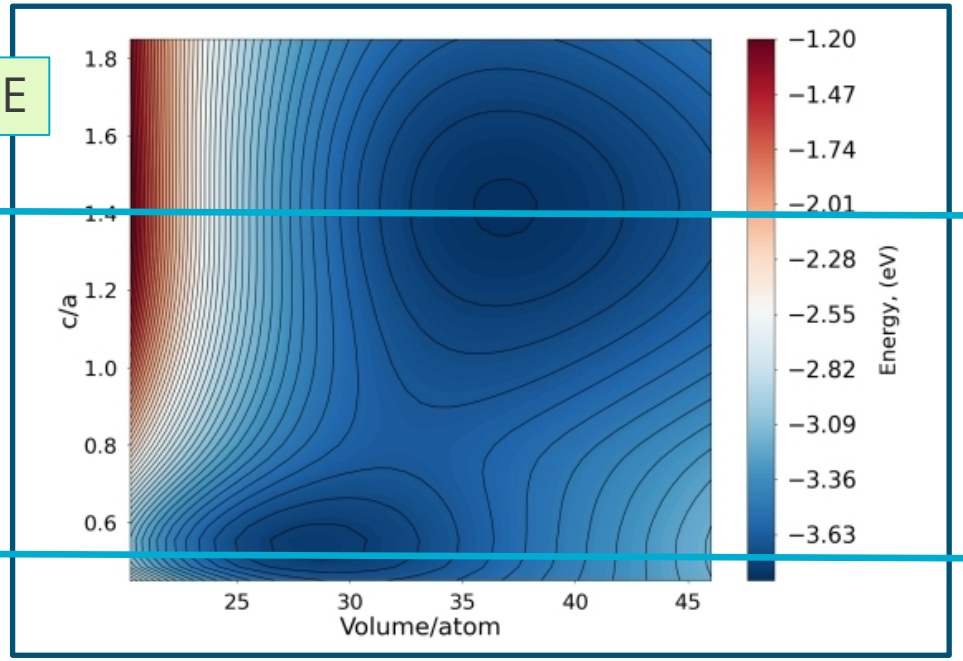
<u>Automatic Property Calculations:</u>

- Minimum energy for lattice parameters a and c/a
- Cold curves (energy vs volume)
- Thermal expansion
- Elastic constants
- Cohesive energies
- Melt temperature
- Generalized stacking fault energies
- 2D Bain and Trigonal Paths

>> Dislocation core energies and slip systems

MEAM potentials at low pressures: 2D Bain Paths

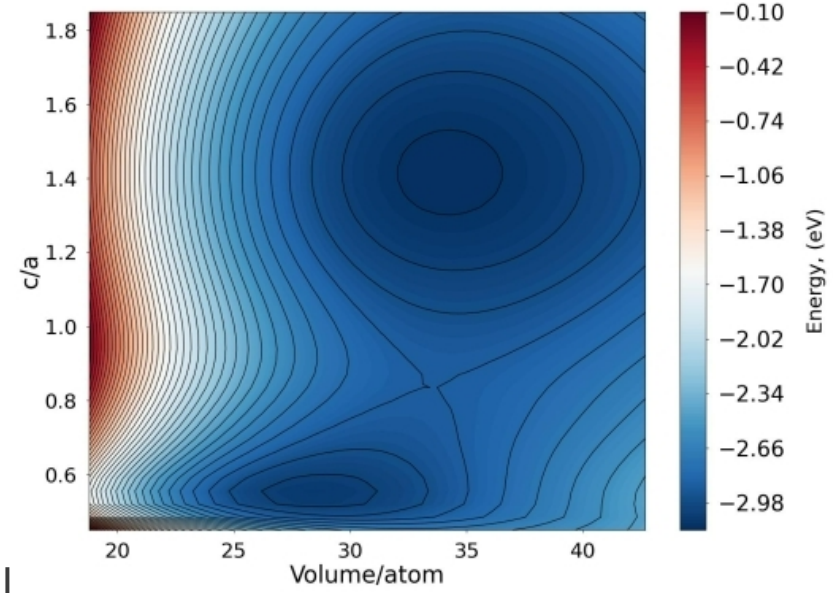
DFT PBE



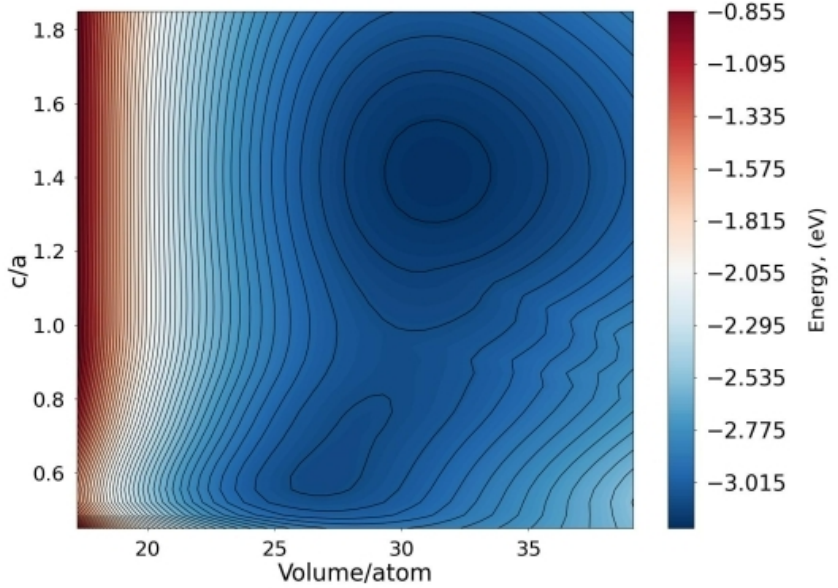
diam

β-tin tetragonal

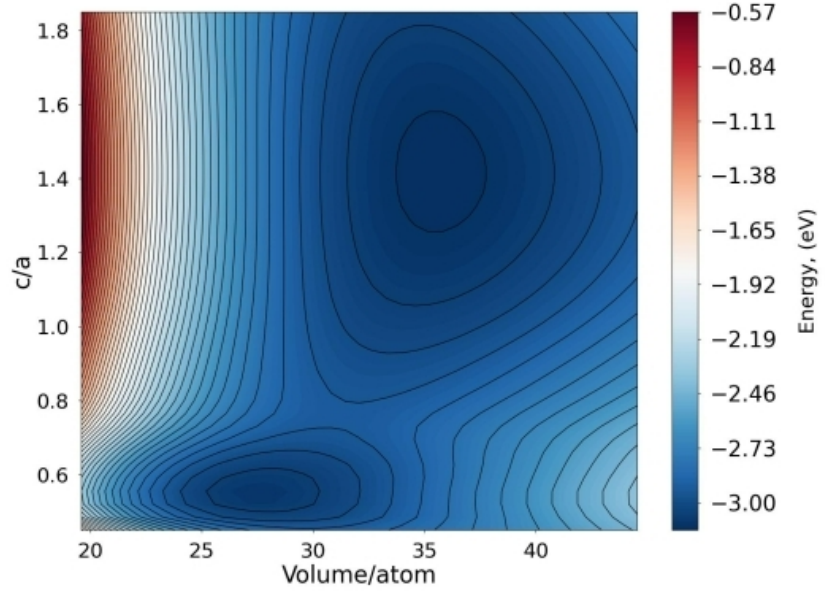
Ravelo



VellaChen

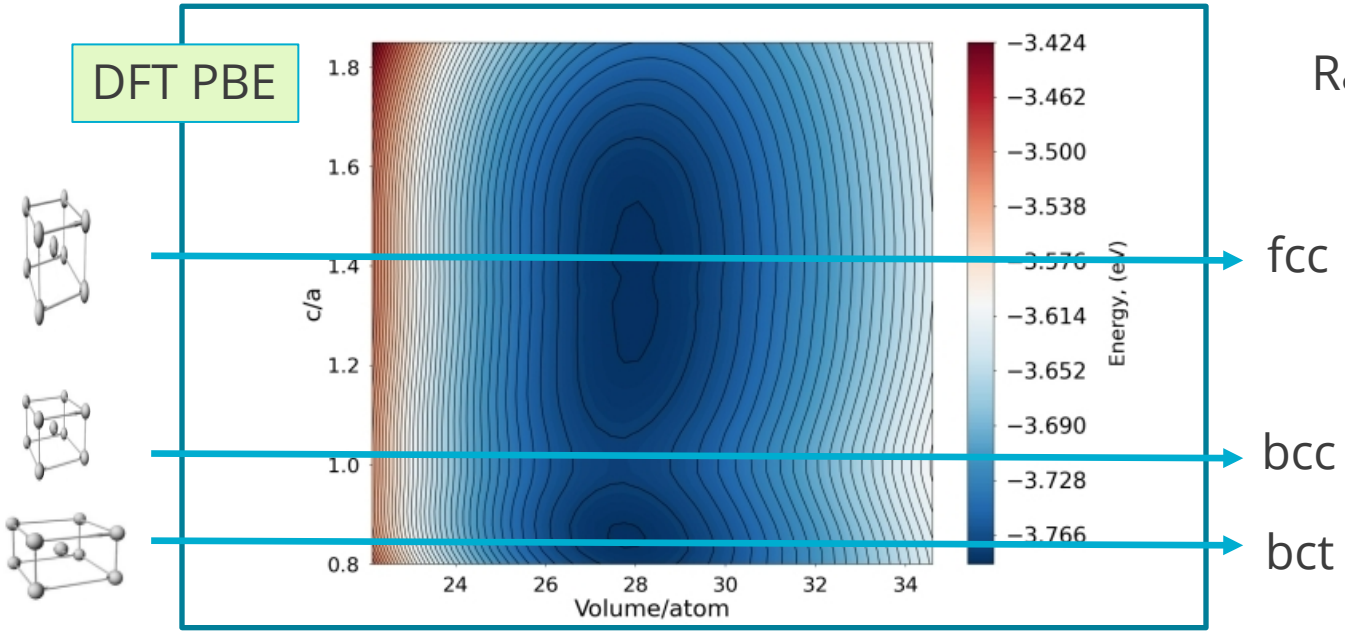


Ko

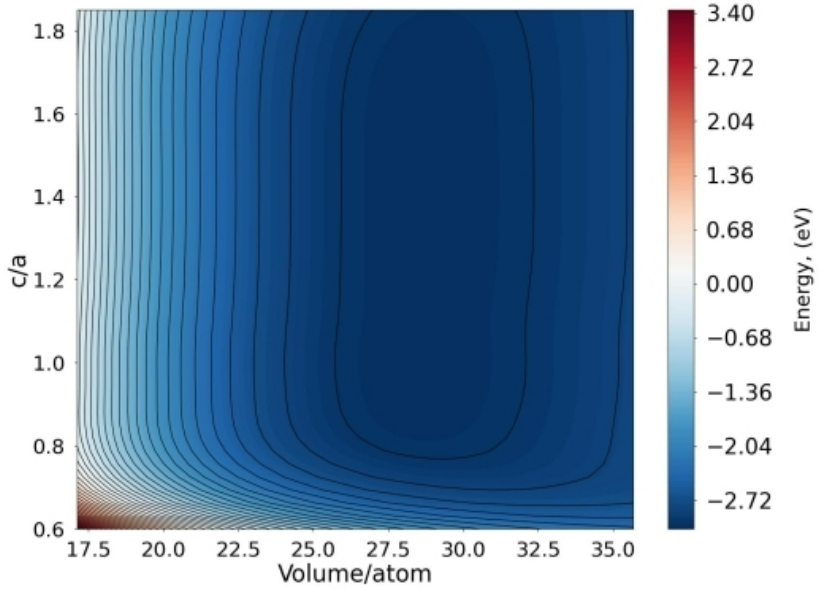


MEAM potentials at high pressures : 2D Bain Paths

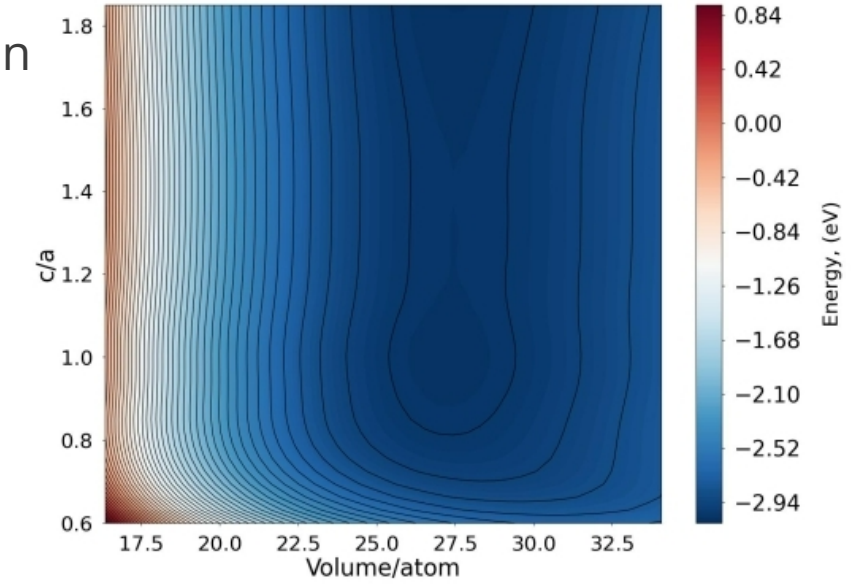
DFT PBE



Ravelo



VellaChen



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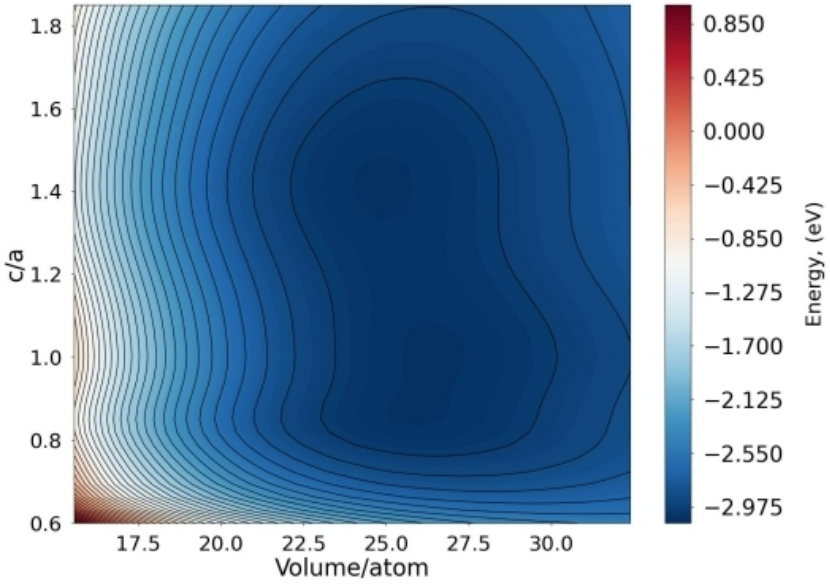
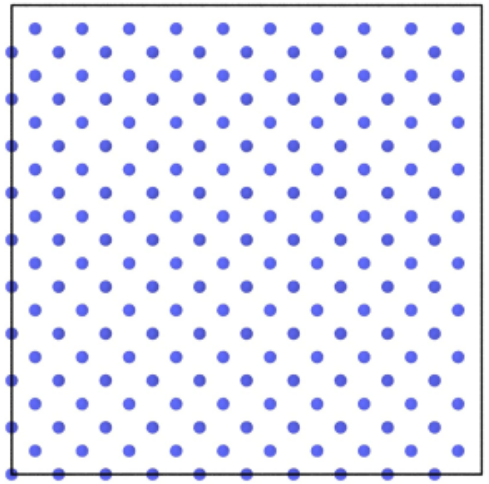


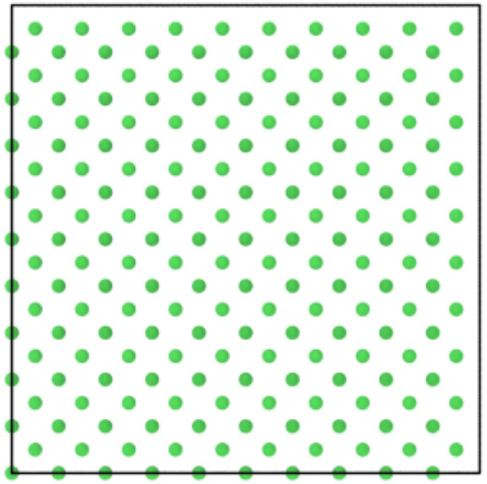
Finite temperature results promising but ultimately fail

Ravelo MEAM

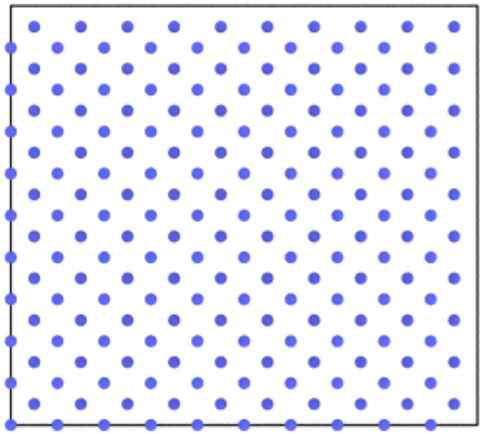
BCC stable



FCC unstable → BCC



β-tin unstable → BCC

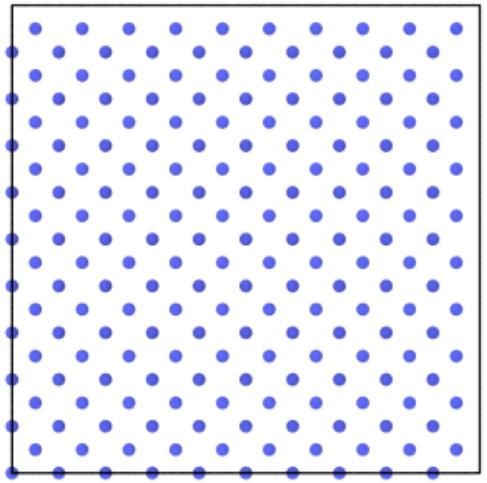


10 GPa
300 K
β-tin state

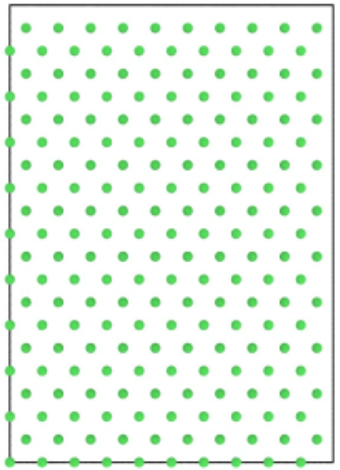
BCC / BCT
HCP
FCC

Ko MEAM

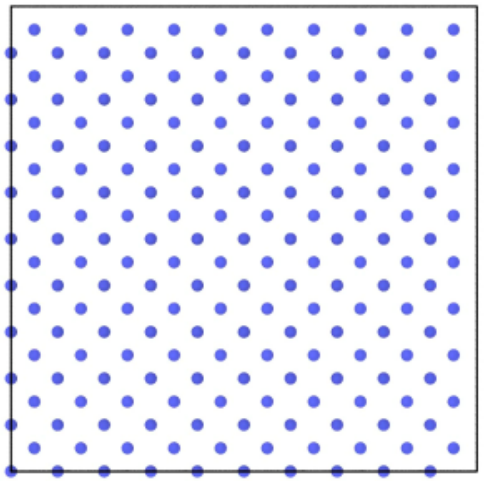
BCC stable



FCC stable



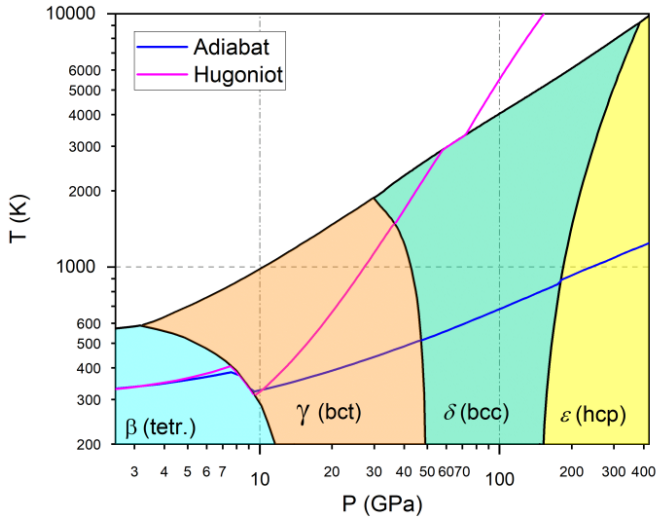
β-tin unstable → FCC



Conclusions: The good, the bad and the ugly

- The MEAM based Ravelo and the newer 2NN Ko interatomic potentials are very good for the covalent (open) structures of the alpha, beta phases and liquid.
  - Zero temperature calculations imply they may be capable of stabilizing the gamma and delta phases as well. They also do reasonably well with density pressure Hugoniot response
  - Unfortunately, they do not do well with predicted high pressure structure stability.
    - Ravelo offers a qualitative phenomenological tool for study of ramp into the delta (bcc) phase, only.
    - Ko predicts unphysical stability for the fcc phase

Challenges for empirical models, ML potential



Two paths forward for atomistic modeling of high pressure tin:

- There may be hope for a potential which focuses exclusively on melt and the higher pressure phases. This approach has been attempted at Sandia by Steven Foiles and Jean-Paul Davis c. 2006.
- Clearly a machine learning based potential will be much less constrained by functional forms which limit the classical empirical potentials like EAM, MEAM and Tersoff.

Mary Alice Cusentino will explore this second approach next.