SAND2012-10179P

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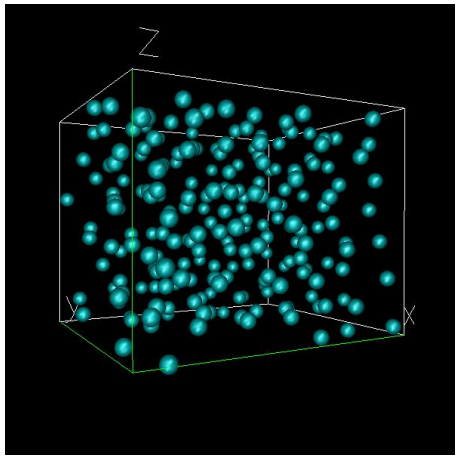

First-principles calculations and Z data for shocked beryllium

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Albuquerque, New Mexico

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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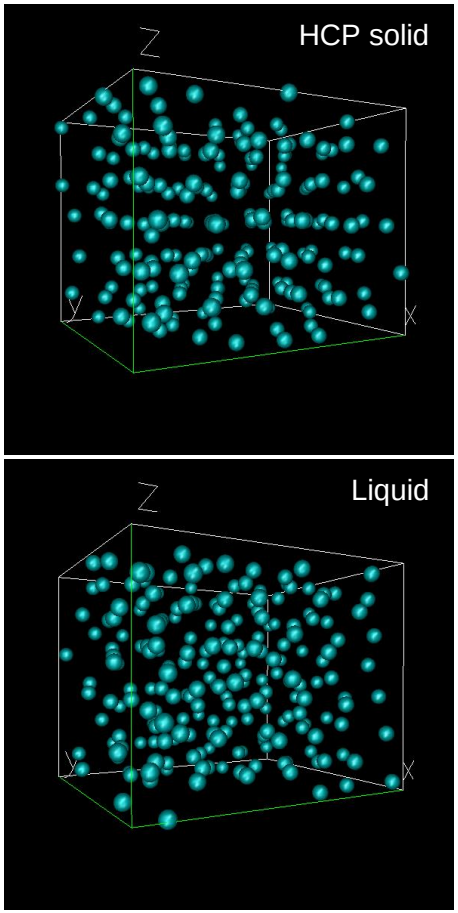
Beryllium modeling with DFT / QMD

- Solid and Liquid Hugoniot states
- Longitudinal and Bulk sounds speeds BCC and HCP
- Melt boundary, emphasis on Hugoniot
- Phase coexistence region
- Accurate pressure isotherms
- Solid/Liquid entropy differences



DFT QMD calculations with VASP

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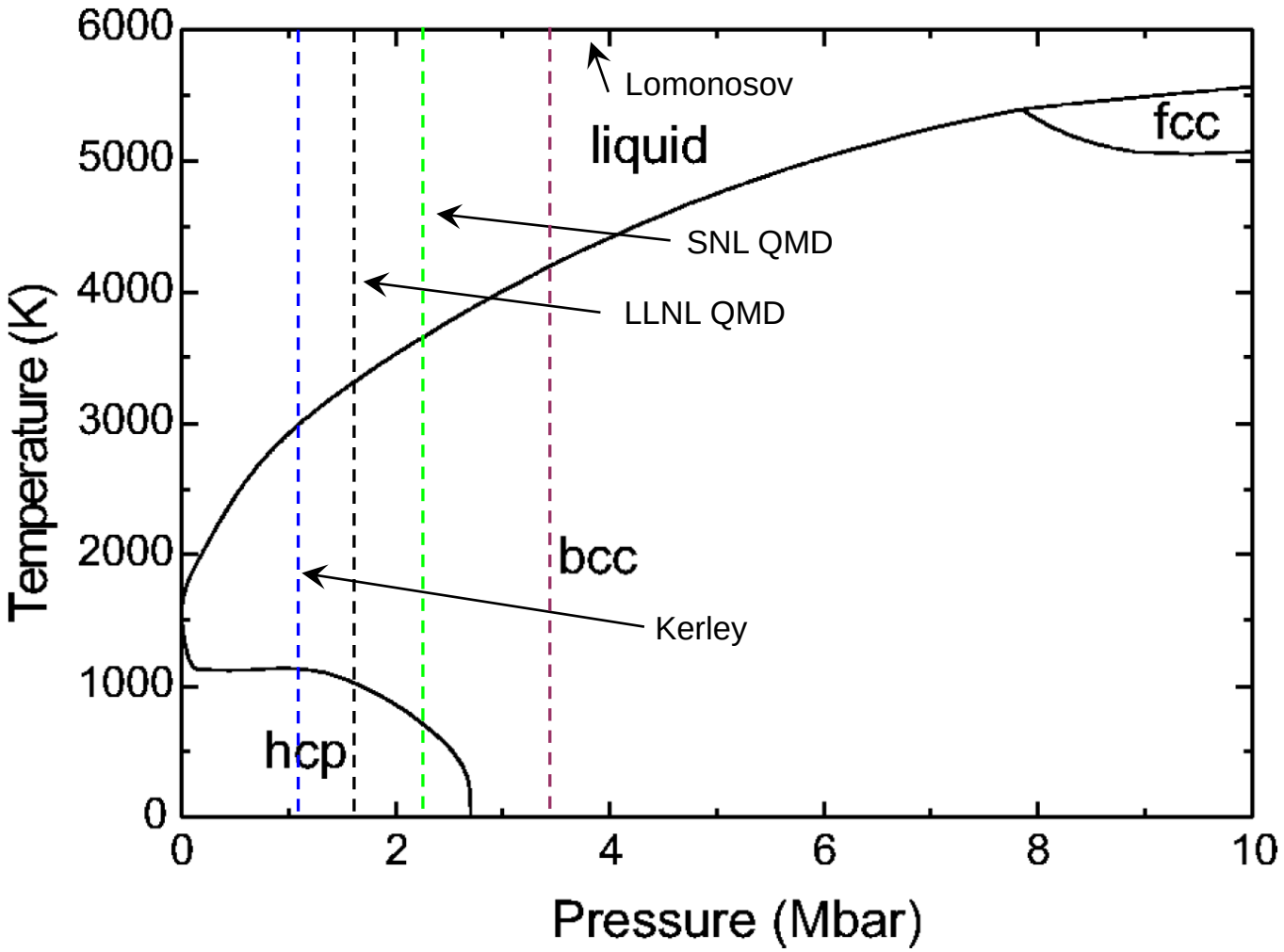
- The simulations are performed with VASP (Vienna Ab initio Simulation Program), a plane wave density functional code (with several in-house modifications for HEDP calculations)
- Exchange and Correlation functionals are Generalized Gradient Approximation (GGA/PBE)
- For Be we typically use 200+ atoms (BCC, HCP, and liquid); We use Projector Augmented Wave (PAW) all-electron, frozen core potentials for the atoms
- We generally perform our simulations in the Canonical Ensemble (N,V,T) using either velocity scaling or a Nosé-Hoover thermostat to regulate the temperature; Fermi statistics for the electrons
- Typical runs cover several picoseconds

Beryllium is generally believed to melt from the BCC phase on the Hugoniot …

… but melt predictions ranged from 1.2 Mbar to 3.5 Mbar



Sin'ko and Smirnov, Phys. Rev. B 71, 214108 (2005)

Details of the QMD beryllium melt calculations

- We use a hysteresis* method to compute an equilibrium melt temperature from a maximum metastable solid temperature and a minimum metastable liquid temperature at the same pressure.

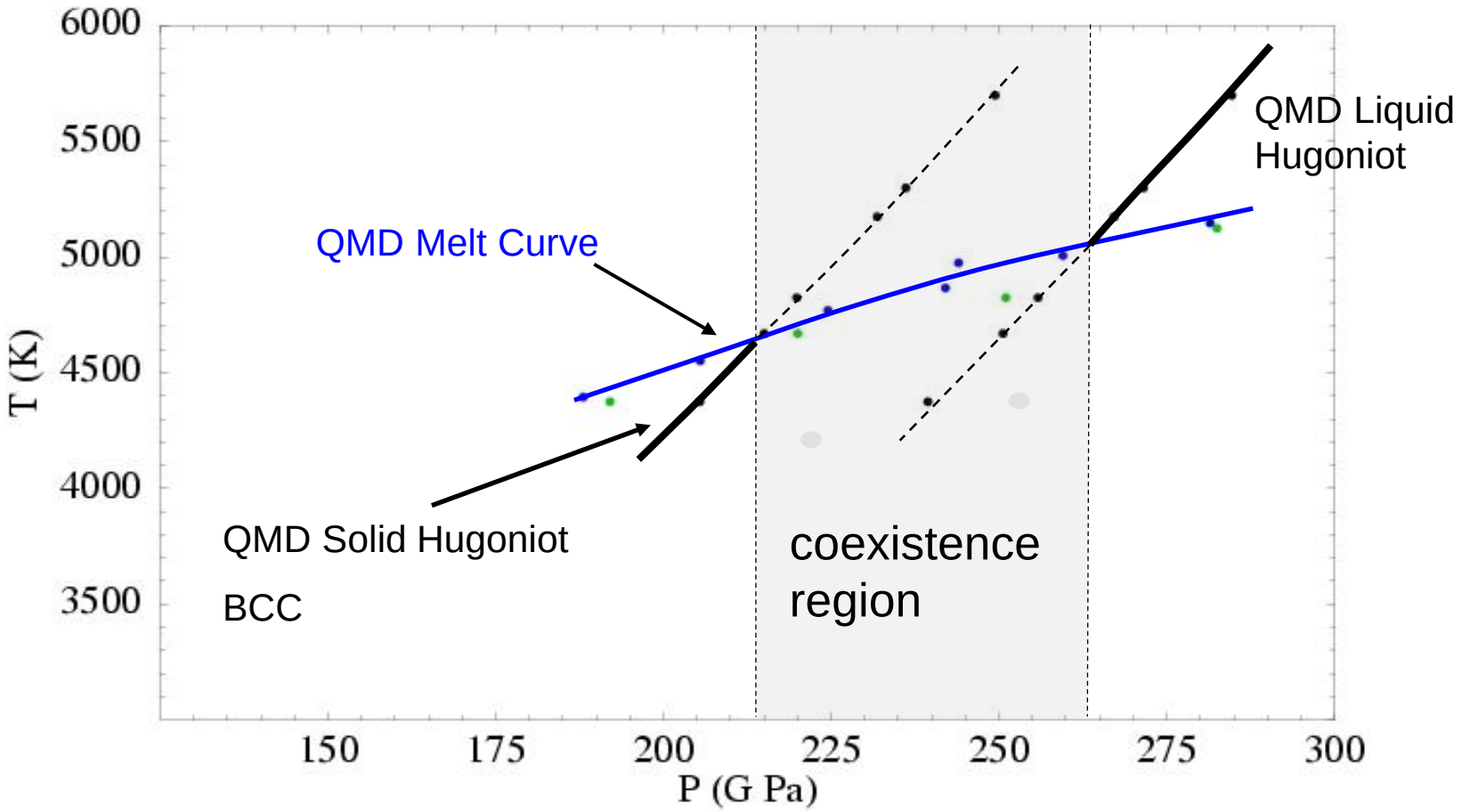
- Two-phase melting/freezing calculations are also used for the melt boundaries.

- The free energy difference $F_{liq}(V)$-$F_{sol}(V)$, combined with the calculated melt curve determines the coexistence region and the entropy change at melt.

- Poisson's ratio is determined from the longitudinal and bulk sound speeds, or by comparing the relative change of the longitudinal and transverse components of the stress tensor under strain.

* Luo, Strachan, and Swift, J. Chem. Phys. 120, 11640 (2004)

Our QMD calculations predicted that the shock melting of beryllium would begin around 213 GPa



The Hugoniot exits the coexistence region around 263 GPa

Comparison of Hugoniot and sound speed measurements with QMD calculation for BCC Be

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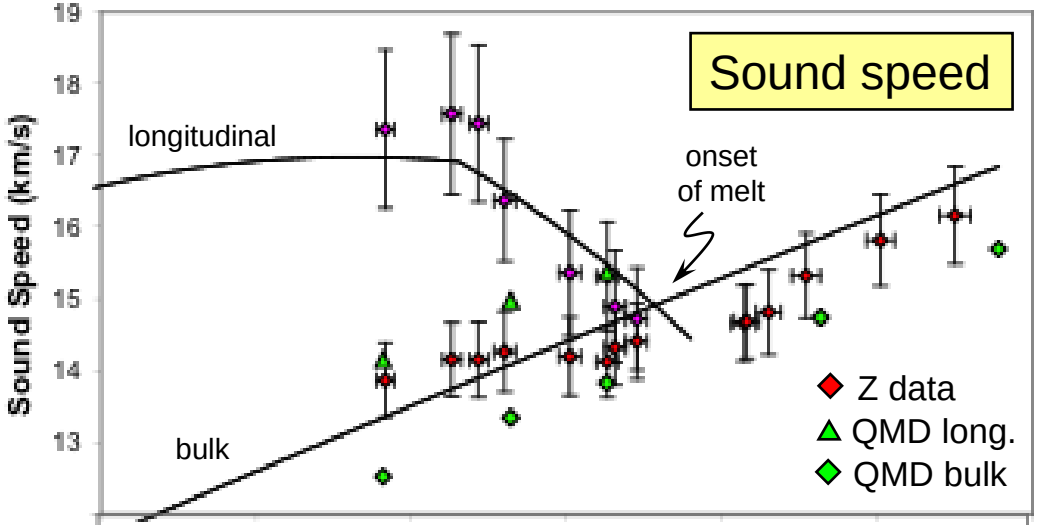
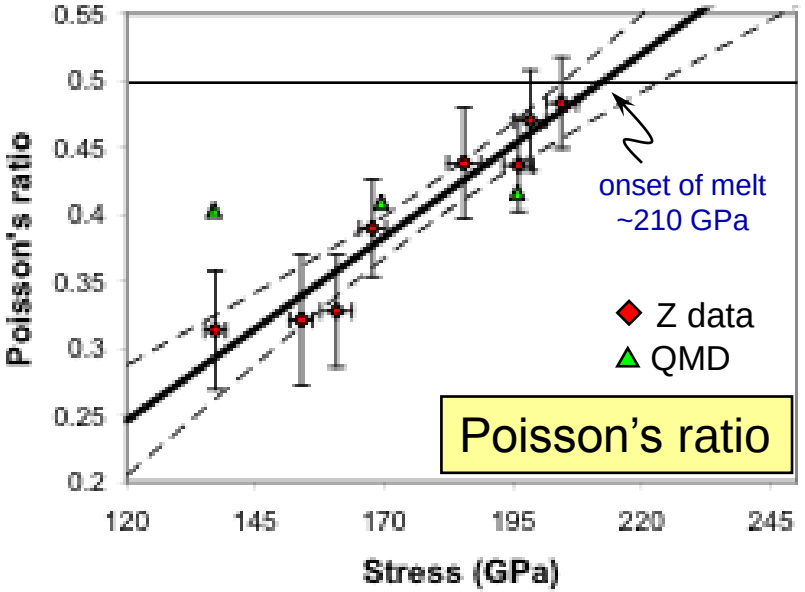
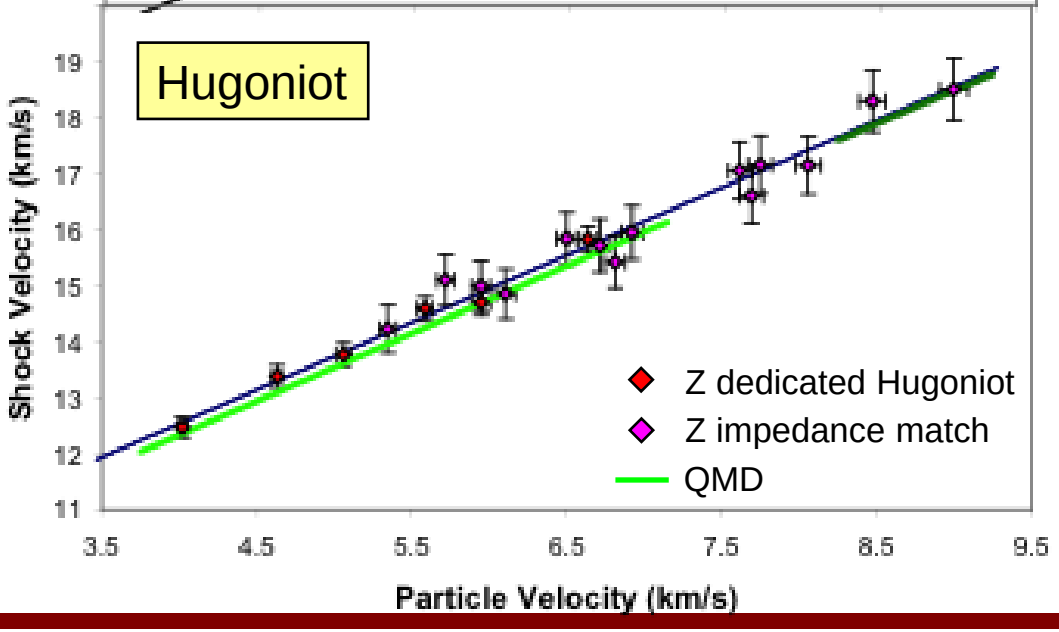
Onset of melt at 210 GPa in good agreement with QMD

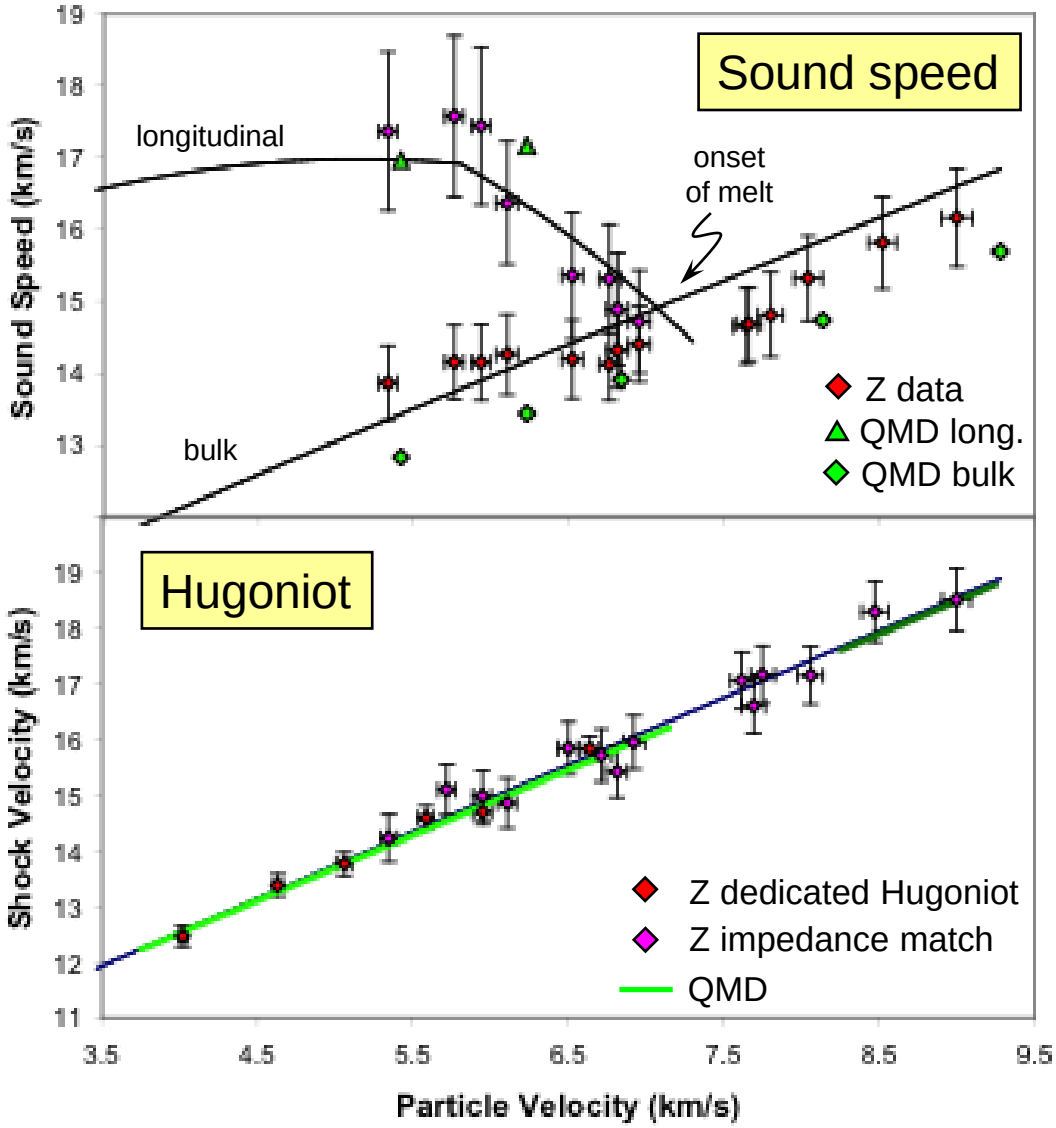


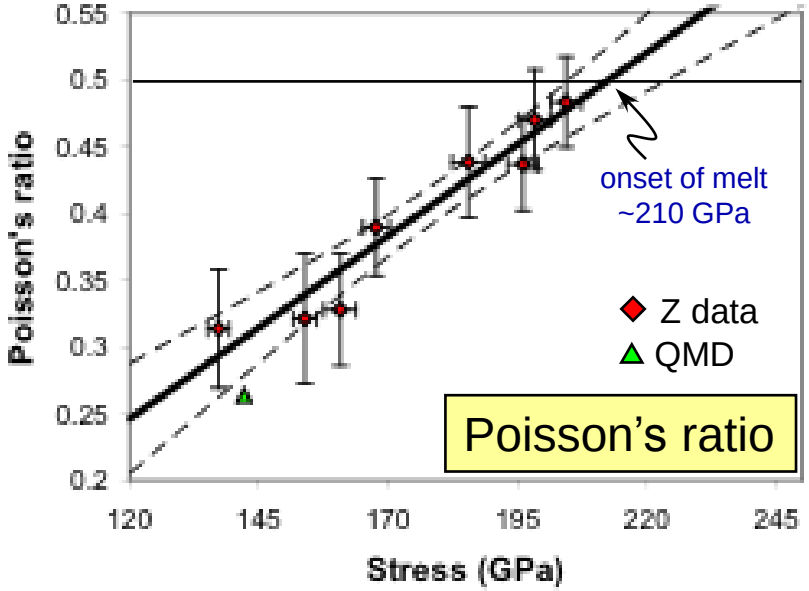




- QMD bcc Hugoniot appears systematically soft relative to experiment
- QMD bulk sound speed in decent agreement with experiment
- QMD longitudinal sound speed significantly low relative to experiment

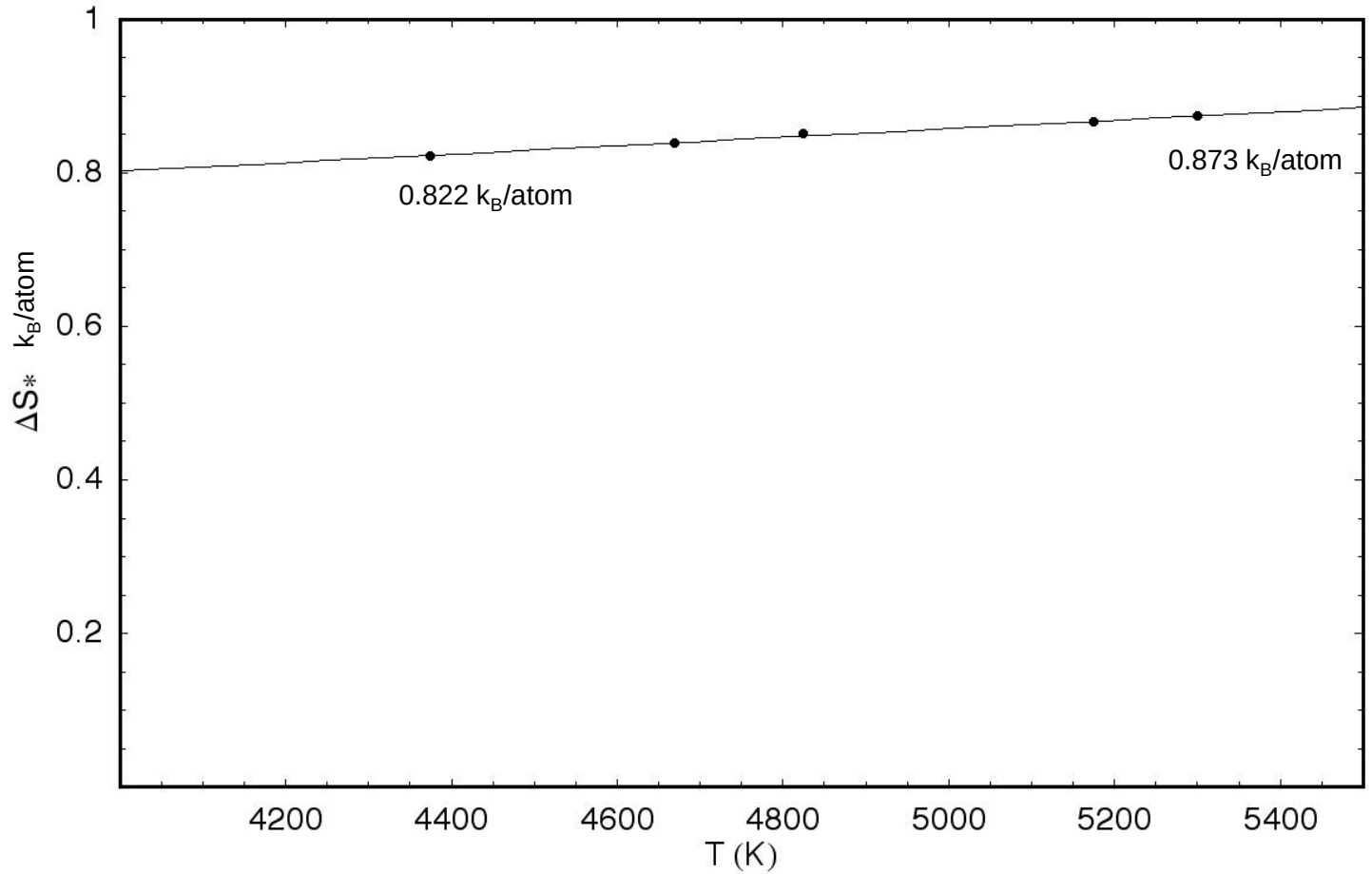
Comparison of Hugoniot and sound speed measurements with QMD calculation for HCP Be





- QMD hcp Hugoniot in better agreement with experiment
- QMD bulk sound speed in decent agreement with experiment
- QMD longitudinal sound speed in much better agreement with experiment

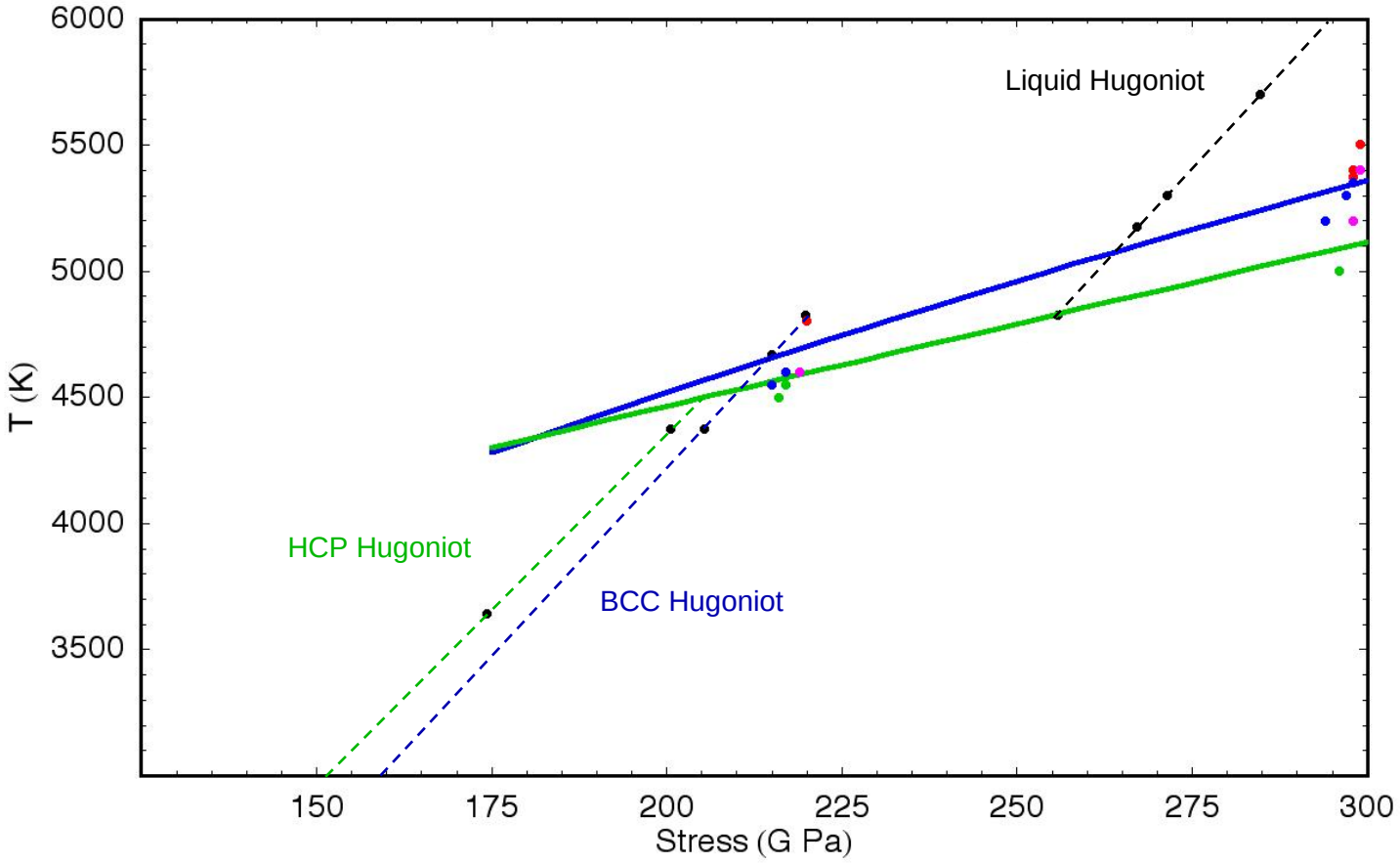
We extracted the constant-volume entropy change along the melt curve



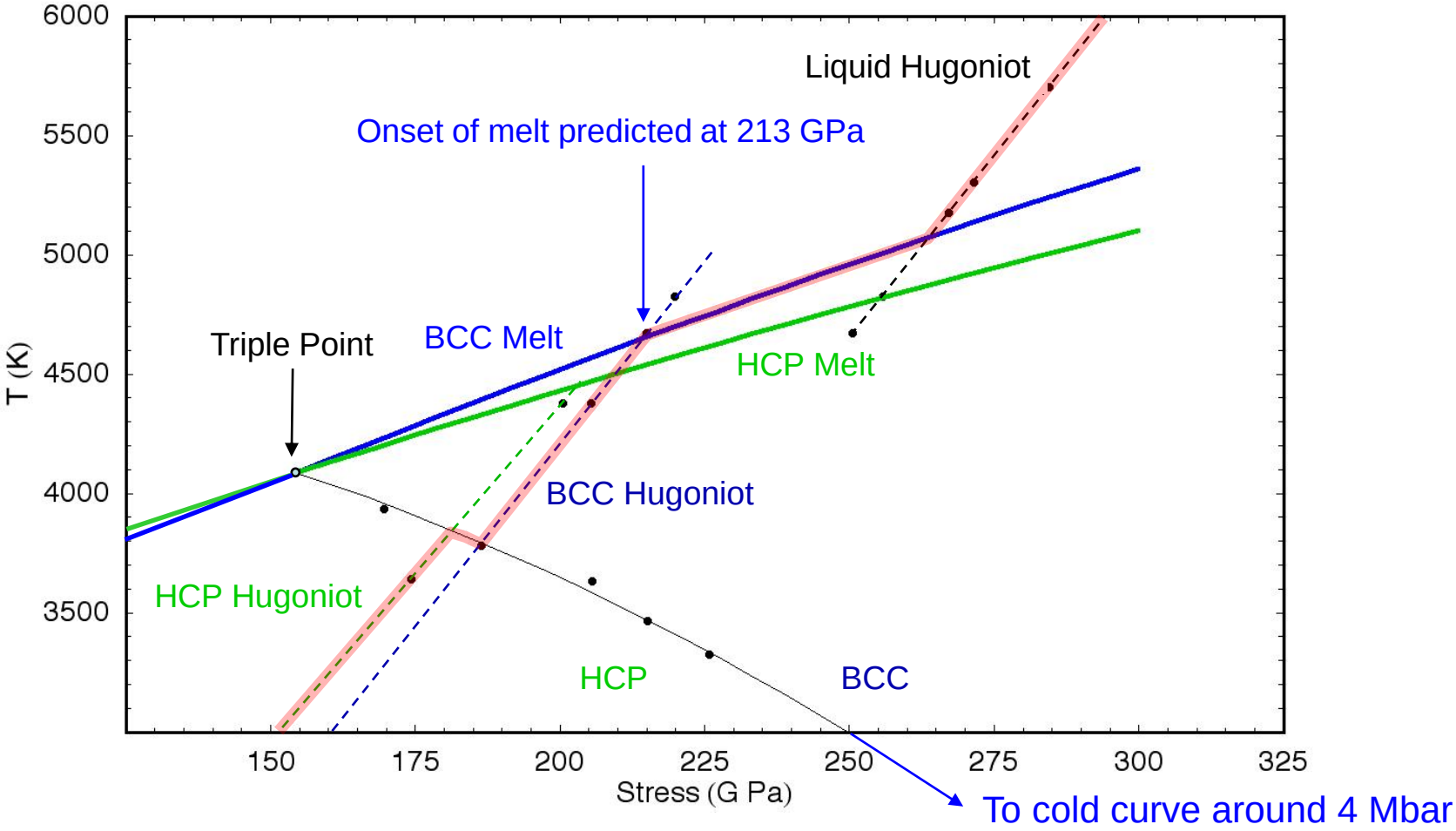
Our calculated entropy change at melt for constant volume (0.85 +/- 0.025 k_B/atom) is in very good agreement with the 'universal' range (~ 0.80 +/- 0.10 k_B/atom) for normal melting*

[*Duane Wallace in *Statistical Physics of Crystals and Liquids*]

Two-phase calculations suggest an HCP melt curve that is lower than the BCC melt curve

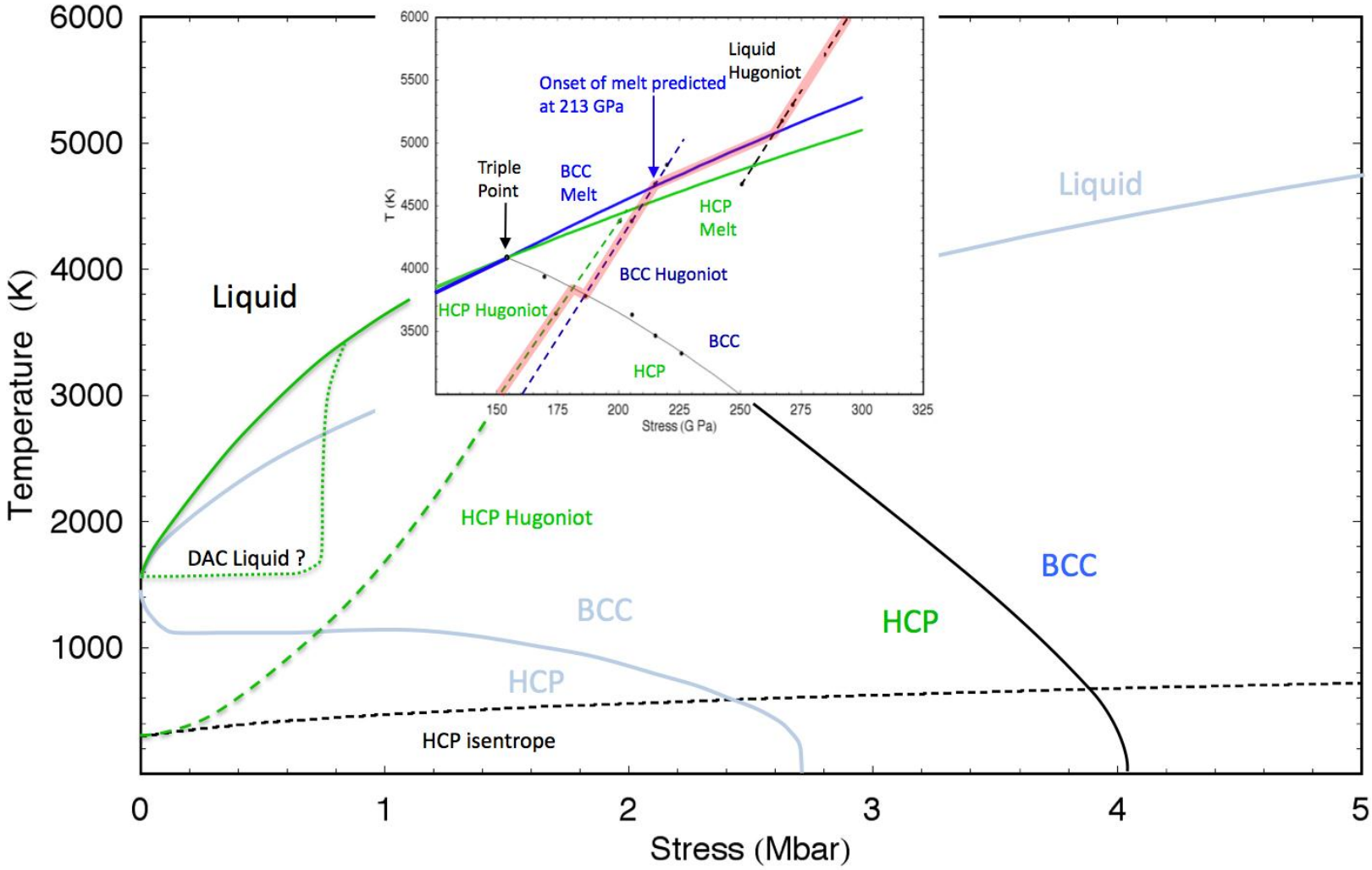


Our QMD calculations are consistent with the Z data and provide new insight into the Be phase diagram



The composite Hugoniot path is highlighted

Although much progress has been made, the beryllium phase diagram is still not complete



Summary

- Hugoniot data for solid in best agreement with HCP
- Longitudinal sound speeds strongly favor HCP
- Predicted onset of melt (213 GPa, BCC) in very good agreement with Z data
- Solid/liquid entropy differences in very good agreement with ‘universal’ range for normal melting
- Data and calculations suggest changes to the Be phase diagram -> HCP to BCC phase boundary is much closer to the high pressure melt curve than previously believed

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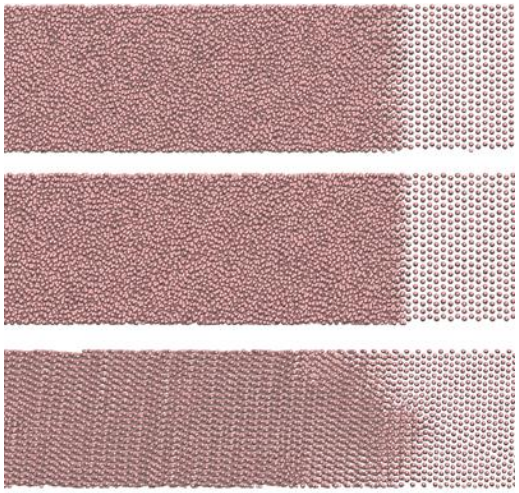

Modeling Dynamic Compression of Beryllium using Large-Scale Molecular Dynamics Simulation

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U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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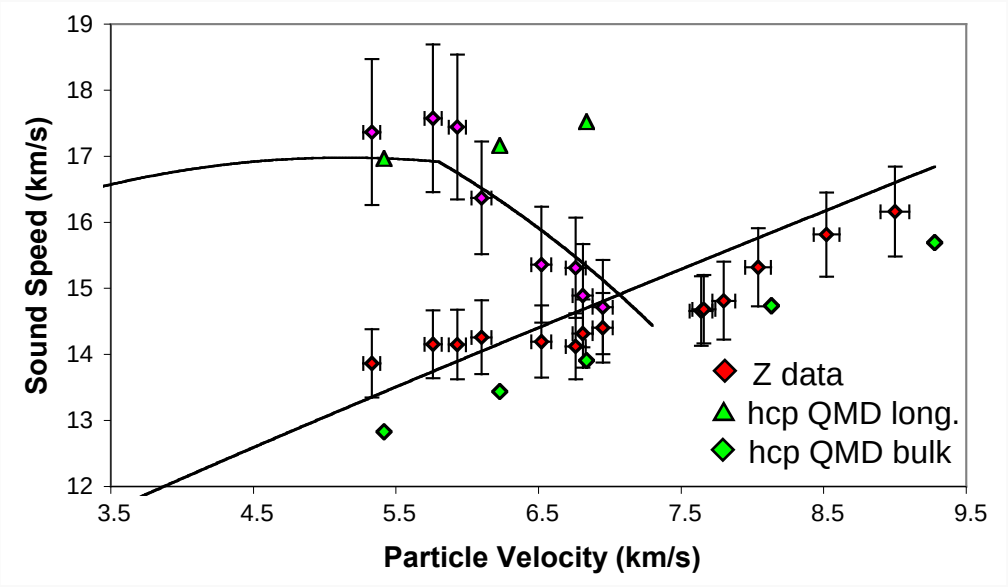
Modeling Ramp Compression Experiments using Large-Scale Molecular Dynamics Simulation



Ultra-high velocity flyer plate experiments on Sandia's Z machine, from Desjarlais, Knudson and Lemke, SCCM 2007

Motivation

- Continuum models require underlying models of the materials behavior
- Quantum methods can provide very complete description for 100s of atoms
- MD can act as a bridge, incorporating the dominant parts of the quantum description, extending it out to very large numbers of atoms

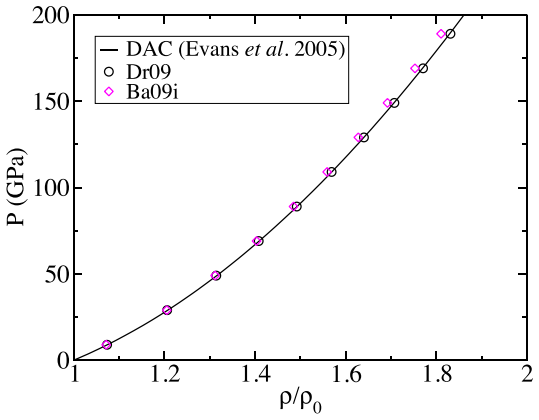
Objectives

- Develop an interatomic potential for Beryllium that describes mechanical and thermodynamic properties up to shock melting (5000 K, 250 MPa)
- Develop new methods for estimating sound speed in NEMD simulations of shock and ramp loading

MEAM Potential for Beryllium

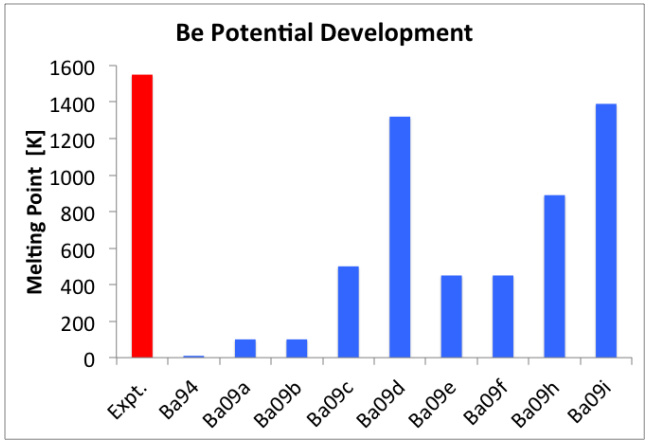
- Collaborated with Mike Baskes to find good beryllium parameters for MEAM potential (Ba09i)
- Accurately predicted elastic constants, static compression and melting pressure

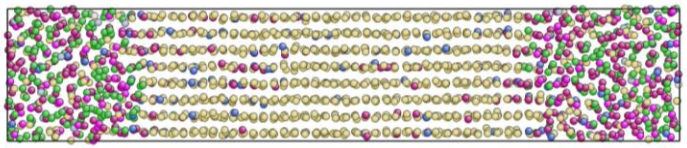
A. P. Thompson, J. M. D. Lane, and M. Desjarlais, "Molecular Dynamics Simulation of Dynamic Response of Beryllium," Shocked Compression of Condensed Matter, AIP Conf. Proc. 1426 1311 (2012).



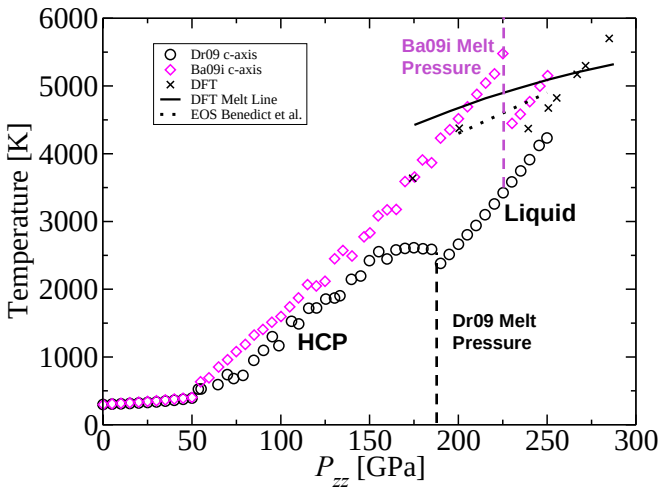
Static compression

Meltin g point



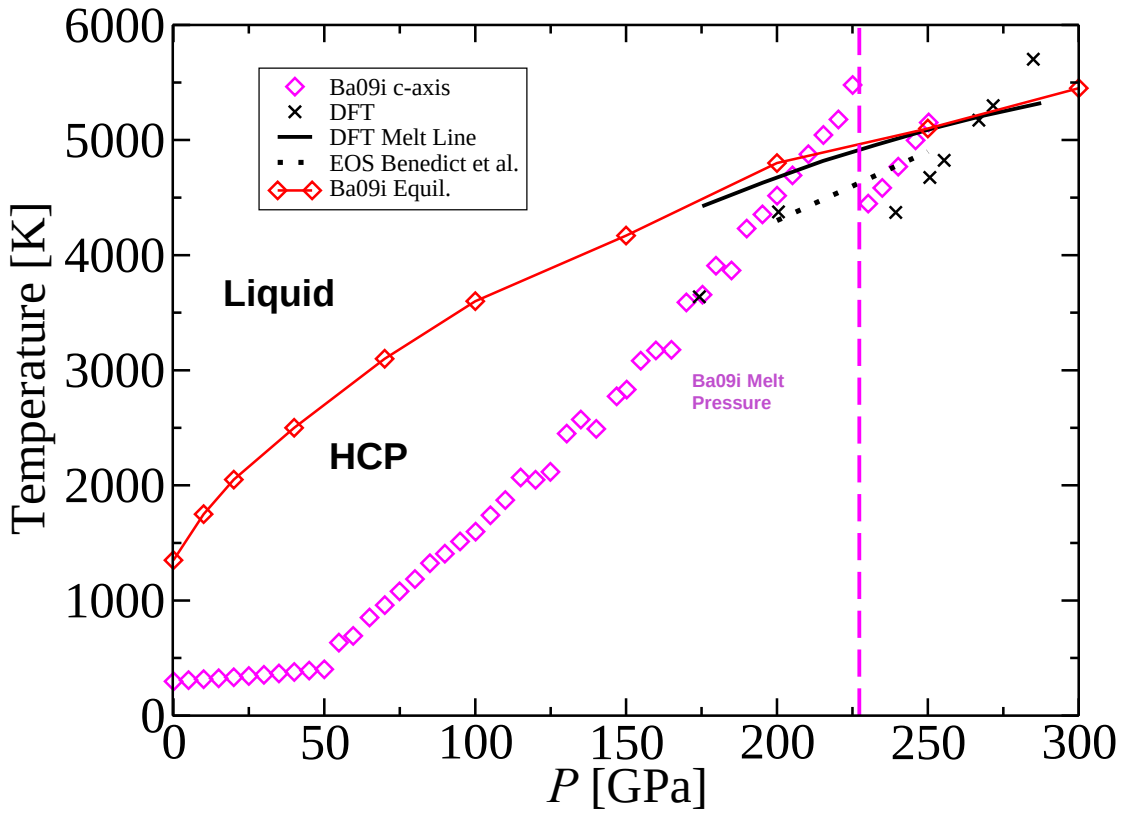


Dynamic compression



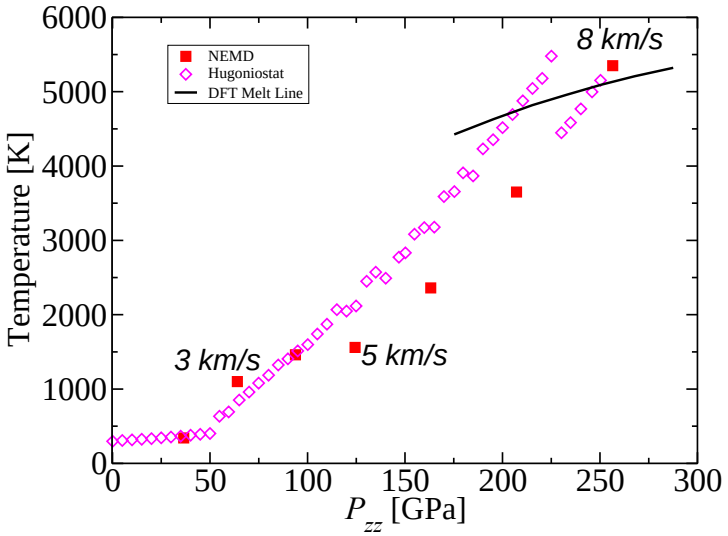
Comparison of Equilibrium and Hugoniot Melt Pressures

Ba09i equilibrium melt curve in very good agreement with DFT estimates



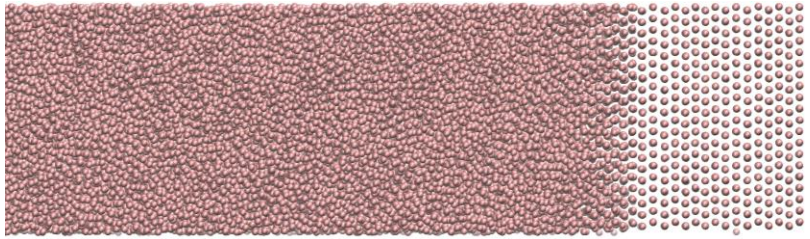
Shock and Ramp Compression of Beryllium

- An unexpected amorphous solid state is observed in beryllium between the elastic/plastic response and the liquid response
- Ramped impact showed similar behavior

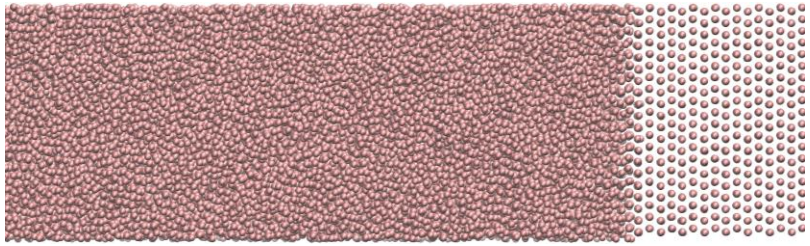


Large-scale (2.3 x 3.9 x 370 nm) nonequilibrium ramp simulations were run with 400,000 atoms on 1600 processors of Red Sky.

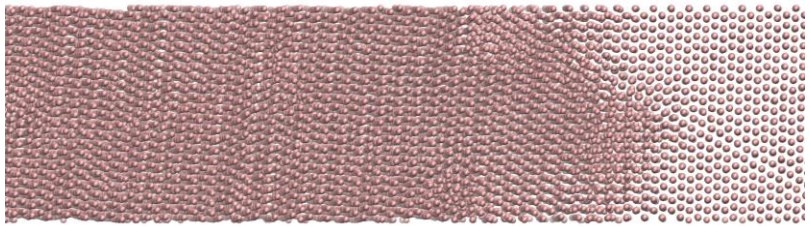
8 km/s



5 km/s



3 km/s

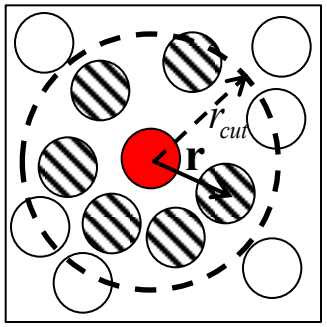


Examples of shock response of beryllium for three different shock piston velocities. At 3 km/s the material deforms elastically and plastically, at 5 km/s an amorphous solid is produced, at 8 km/s the material is fully melted.

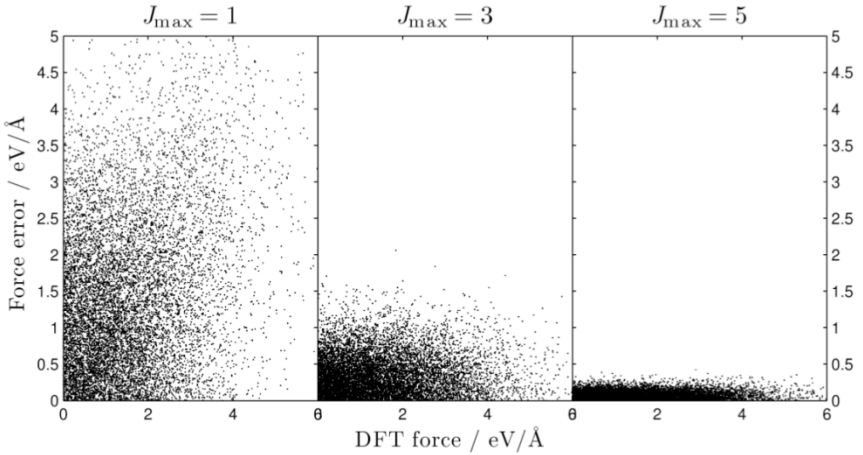
GAP Approach for Interatomic Potentials

GAP: A systematic, informatics approach

- Based on QM and mathematics rather than empiricism.
- Local density around each atom expanded in 4D hyperspherical harmonics, analogous to Fourier series
- Atomic configurations described by bispectrum of lowest-order coefficients in series
- Preserves universal physical symmetries: invariance w.r.t. rotation, translation, permutation
- Gaussian process (GP) regression used to interpolate energy of QM configurations
- 100-1000x more expensive than MEAM
- Far cheaper than QM, linear scaling
- Can trade performance and accuracy



$$r(\mathbf{r}) = \sum_{j=0}^{\infty} \sum_{m=-j}^{j} c^{j}_{m} Y^{j}_{m}(q, f, w)$$

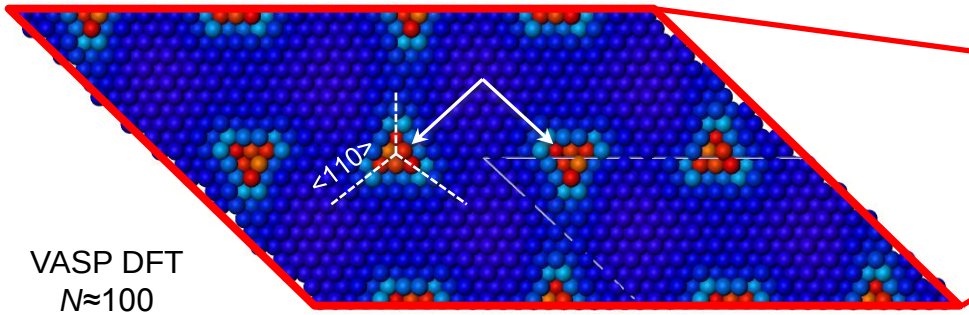


Diamond: Force errors for GAP fitted to DFT. Adding higher-order GAP coefficients systematically increases accuracy
Bartok et al., *PRL* **104** 136403 (2010)

SNAP: Predictive Model for Tantalum

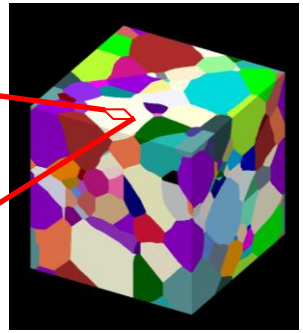
Objective: model the interactions of dislocation cores with grain boundaries to understand microscopic failure mechanisms. Existing tantalum potentials do not match key results from DFT calculations.

Screw Dislocation Dipole in Tantalum



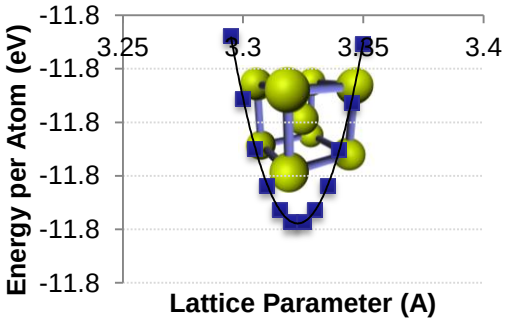
VASP DFT
$N \approx 100$

Polycrystalline Tantalum Sample



LAMMPS MD
$N \approx 10^8$

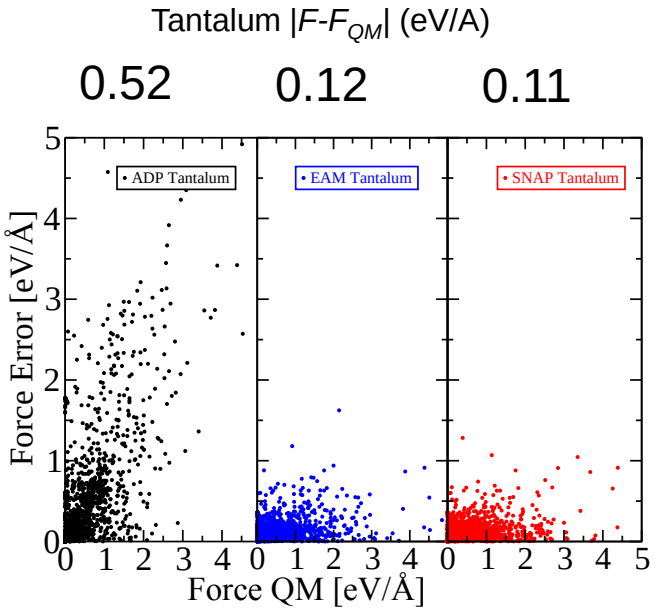
VASP DFT Training Data



- 78 DFT configurations
- ~100-atom supercells with perturbed atoms: BCC, FCC, A15, Liquid
- Relaxed Surfaces
- Generalized stacking faults, relaxed and unrelaxed

SNAP Potential for Tantalum

- The SNAP potential accuracy is comparable to the best existing potentials
- Efforts to further improve accuracy are underway
- Given suitable DFT training data, a SNAP potential for Be is within reach



BCC Lattice and Elastic Constants

	a [A]	C11 [Gpa]	C12 [Gpa]	C44 [Gpa]
Expt	3.303	266	158	87
ADP	3.305	265	163	85
EAM	3.303	247	144	87
DFT	3.32	260	160	70
SNAP(E,F)	3.309	379	308	34
SNAP(E,F,V)	3.316	287	155	57

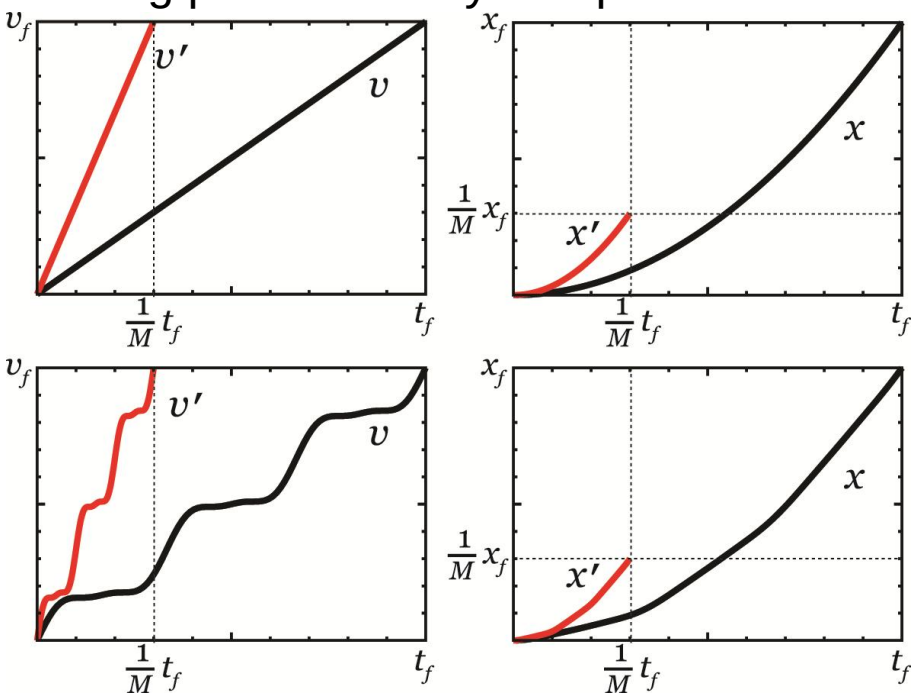
MD study of high-pressure high-density Be

- Shock and quasi-isentropic loading
  - Atomistic techniques recently developed in LDRD
  - Quantitative material properties and mechanisms
    - Characteristics extraction, wave speed & plasticity analysis

- Measurement of bulk and longitudinal wave speeds for ramp and shock wave studies.
  - Stress tensor approach
  - Time-of-flight analysis
    - Pulse propagation
    - Ramp wave evolution similar to Charice analysis to determine wave-speed from characteristic curves.

Scaling and dynamic similarity theory

Driving piston velocity and position:



Dynamic similarity:

$$\frac{F_{\text{model}}}{F_{\text{actual}}} = \frac{M_{\text{m}}\frac{L_{\text{m}}}{T_{\text{m}}^2}}{M_{\text{a}}\frac{L_{\text{a}}}{T_{\text{a}}^2}} = \frac{\rho_{\text{m}} A \frac{L_{\text{m}}^2}{T_{\text{m}}^2}}{\rho_{\text{a}} A \frac{L_{\text{a}}^2}{T_{\text{a}}^2}}$$

$$= \lambda_\rho \left(\frac{\lambda_L}{\lambda_T}\right)^2 = 1$$

Invariant to scaling:

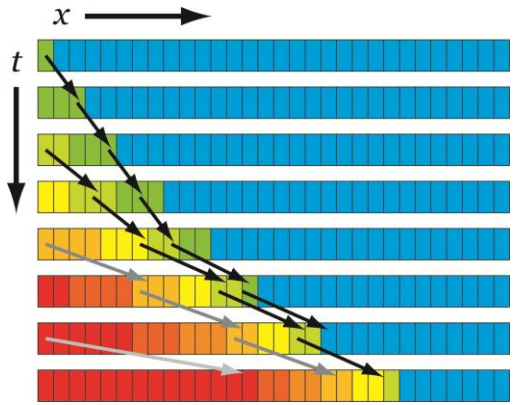
Velocity	Stress
Strain	Temperature*
Forces	Density

Subject to scaling:

Strain rates
Accelerations
Times and distances
and any extensive variable…

Characteristic curves from wave speed

- Construction from slope definition
  - Run simulation extracting continuum state variables
  - Calculate wave speeds over the range of states for load path
  - Characteristics have inverse slope, dx/dt = v ± c, where c is the wave speed
  - Starting at the piston (x_1, t_1), integrate to find (x_2, t_2), etc.



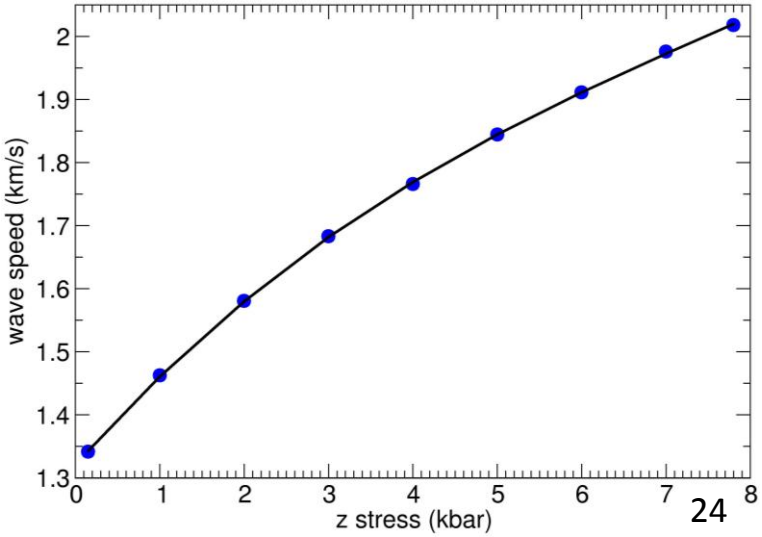
- Requires wave-speed as function of local state
  - Method 1: Gao (J. Mech. Phys Solids, 44, 1452) still in process

$$V_1 = \sqrt{B_{1111}/\rho}$$

$$B_{1111} = S_{11} + F_{1I}C_{I1K1}F_{1K}$$

    - S is the second Piola-Kirchoff stress tensor
    - F is the deformation gradient tensor
    - C is the material tangent modulus

  - Method 2: Time-of-flight was used, instead



Conclusions

- Large-scale NEMD simulations of the melt transition are within reach
- A potential that can reproduce QMD is needed
- The SNAP method may be the best approach
- Need better methods for calculating sound speed
  - Wavespeed from stress/strain sampling
  - Wavespeed from time-of-flight