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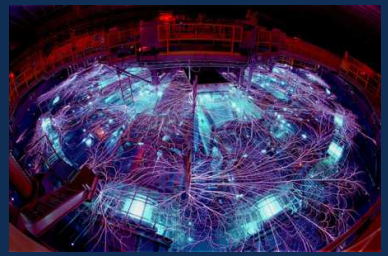

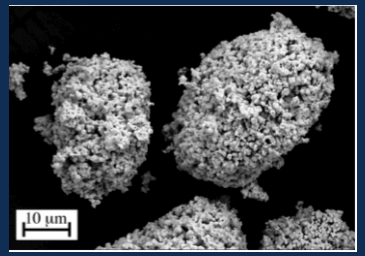

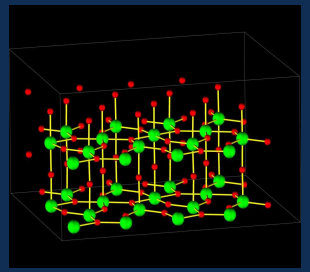

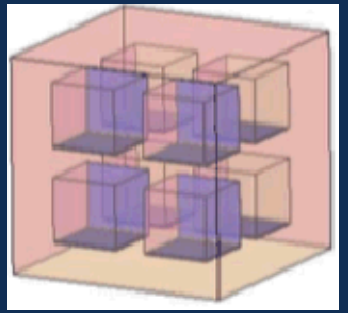

Modeling the Shock Hugoniot in Porous Materials

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SAND2017-2770 C

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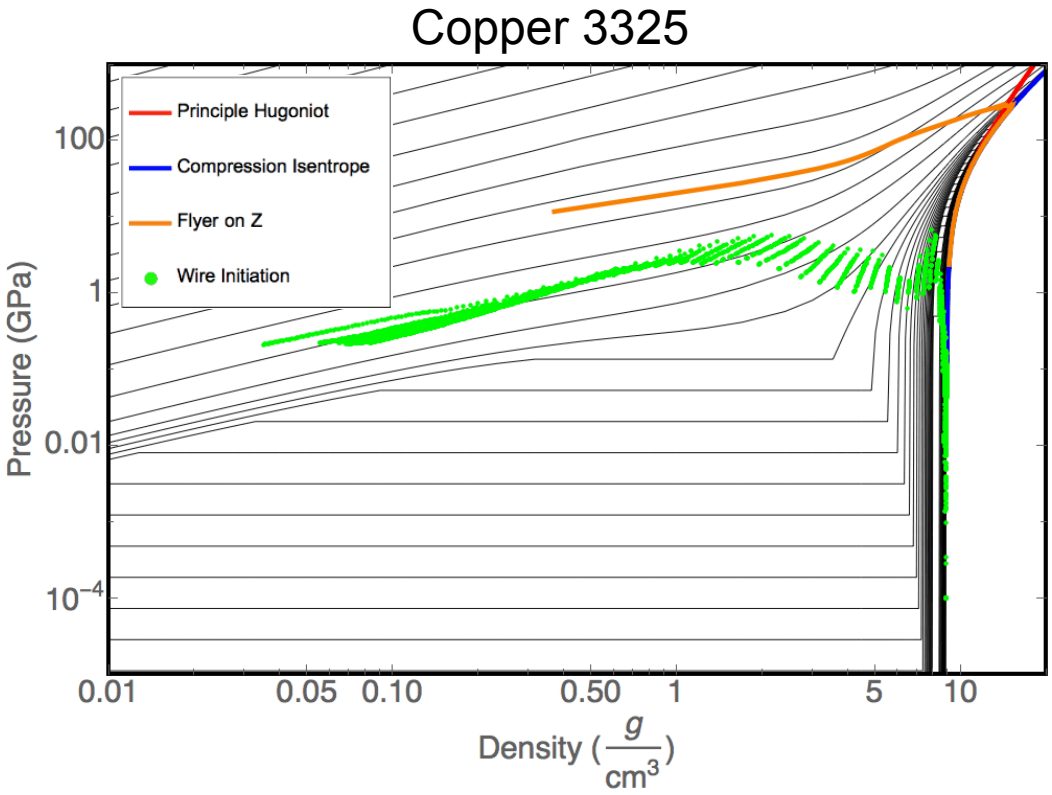
The warm dense matter expanded state is relevant and important

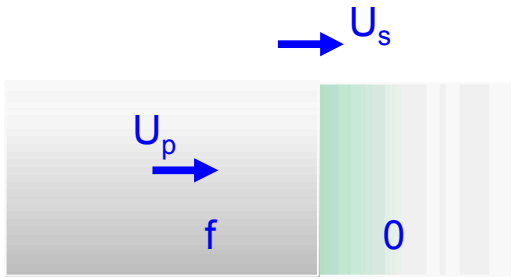
- Orange is a snapshot in time of a copper flyer on Z (ALEGRA)
  - Ambient through WDM simultaneously
- Green is copper wire initiation
- Experiments measure in the compression
- Developers extrapolate EOS in expansion
- Need to measure in expansion



Shock compression is a way to investigate thermo-physical properties of matter at extreme pressures

- *Conservation of mass, energy, and momentum* lead to the **Rankine-Hugoniot condition** for the initial (0) and final state (f)
- *Uniform final state for strong shocks*

  - E - internal energy
  - P - pressure
  - v – specific volume

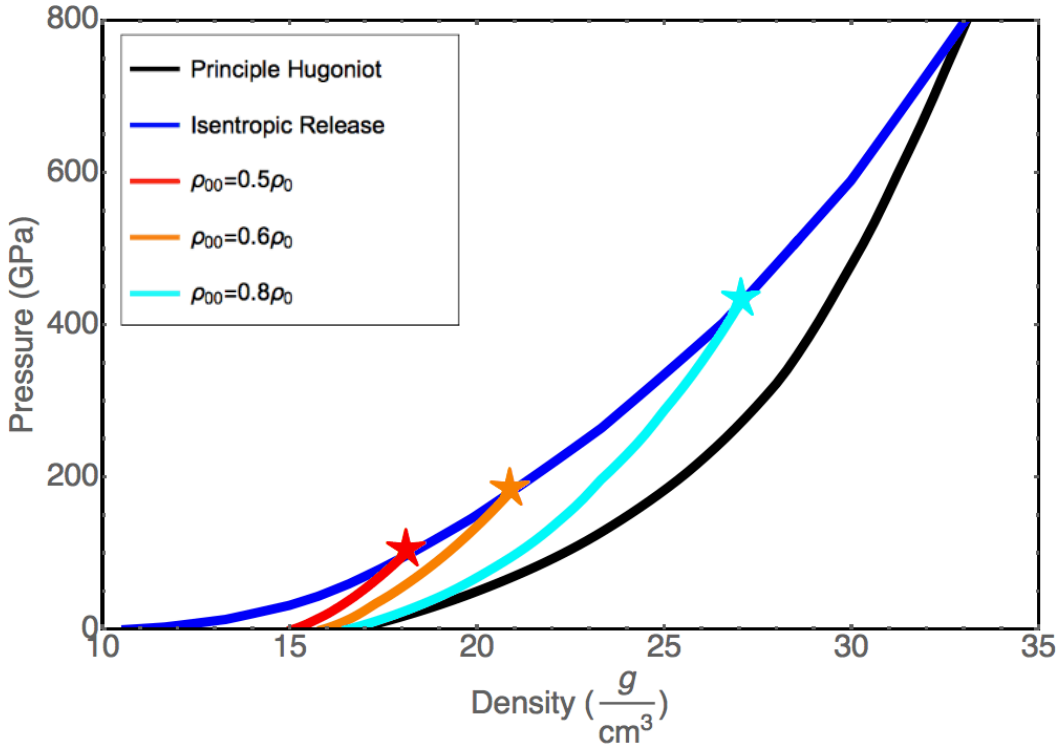


$$E_f - E_0 = \frac{1}{2}(P_f + P_0)(v_0 - v_f)$$

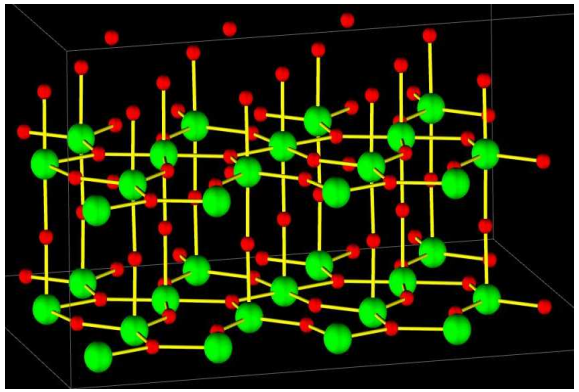
The properties of the initial state contribute critically to setting the thermodynamic state after the shock

Using porous materials gives gas guns access to regions of EOS space normally reserved for Z, NIF, OMEGA…

- Hugoniot assumes 18km/s aluminum impacting tantalum
- Must know window material under extreme conditions to high degree of accuracy
- Cyan is release to LiF window
  - Can be reached by Ta flyer at 7 km/s hitting porous material
- Red is TPX window
  - Can be reached by a Ta flyer at 3 km/s
- Orange is an arbitrary plastic window:
  - $0.6\rho_0$
  - Ta Flyer at 5 km/s
- Using porous material, only need to know properties of the unshocked material



Density functional theory (DFT) based MD is an established approach for first-principles thermodynamics



J App Phys, Vol. 94, No. 9. (Nov 2003)

VASP code (Georg Kresse, Vienna, Austria)

- Finite-temperature DFT (Mermin)
- Plane-wave basis-set for controlled convergence and free electrons/ionization
- Projector augmented wave core functions (PAW)

DFT calculations are generally predictive for shock properties

- *Carbon, quartz, H_2O, xenon, krypton, argon, CO_2, CH_2, deuterium – simulations show excellent agreement with experiments (PRL, PRB, JCP, and SCIENCE)*

Model system for tantala (Ta_2O_5)

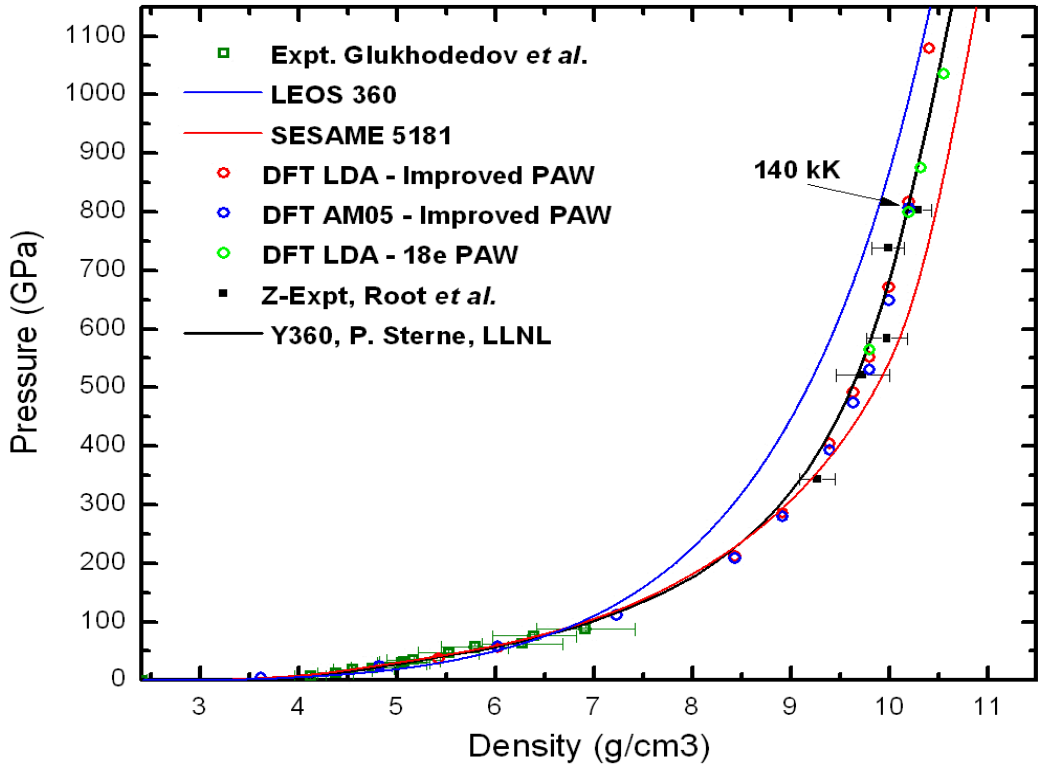
14 atom unit cell x (2x2x2)
80 oxygen 32 tantalum
LDA XC-potential

First-principles thermodynamics – long simulations yield $P(\rho,T)$, $E(\rho,T)$, structure, etc.

In general DFT and experimental data in excellent agreement for shocked materials

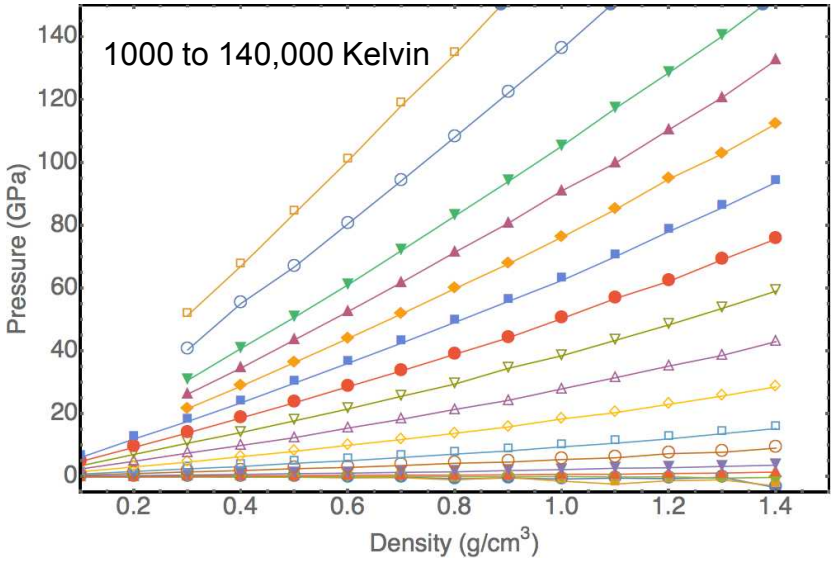
- Each Hugoniot point is a separate series of calculations
- Each locus of Hugoniot points would require a new series of calculations
- Don't have a trusted tantala or aerogel EOS table and don't want to do a series of calculations for each initial density
- Use DFT to build the tables

Krypton Hugoniot [Mattsson, PRB 2014]

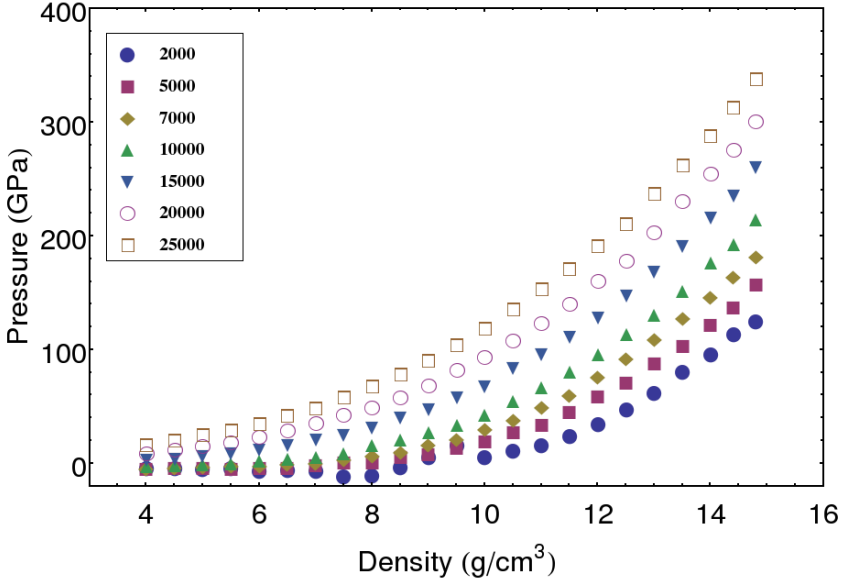


DFT created Aerogel and Tantala EOS tables

Isotherms of DFT created Aerogel EOS



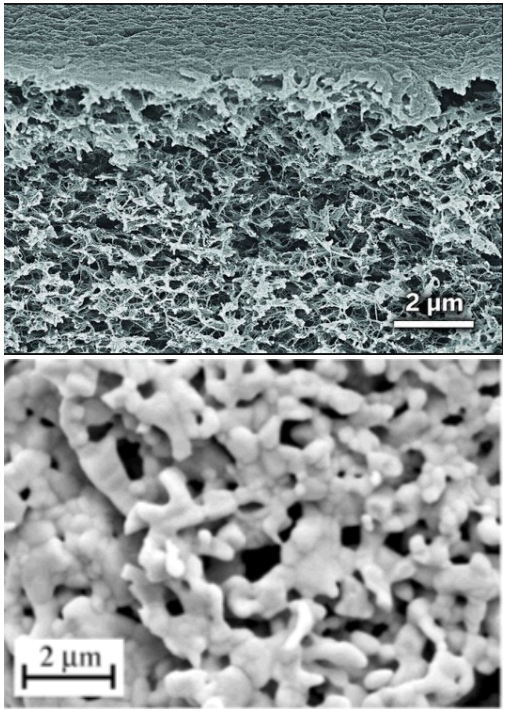
Isotherms of DFT created Tantala EOS



- Require a thermodynamically consistent EOS
- Each point is a DFT calculation
  - Lines to aid eye along isotherms
- Past DFT success prompted this method for a viable EOS

A shocked porous material reaches higher temperature at a given density – gives flexibility in accessing phase space

- Porous materials have added complexity compared to full-density systems
- The behavior of porous materials is important for many applications – and challenging to model
  - Iron oxide is studied in the context of planetary formation/impact
  - silica aerogel is used to mimic liquid deuterium and impedance match target materials
    - Plethora of experimental data
    - Journal of Applied Physics 114, 053510 (2013)
- Tantala is a transition metal and high mass density without magnetic strong electronic correlation
  - Have data over extensive range [Cochrane SCCM 2015]



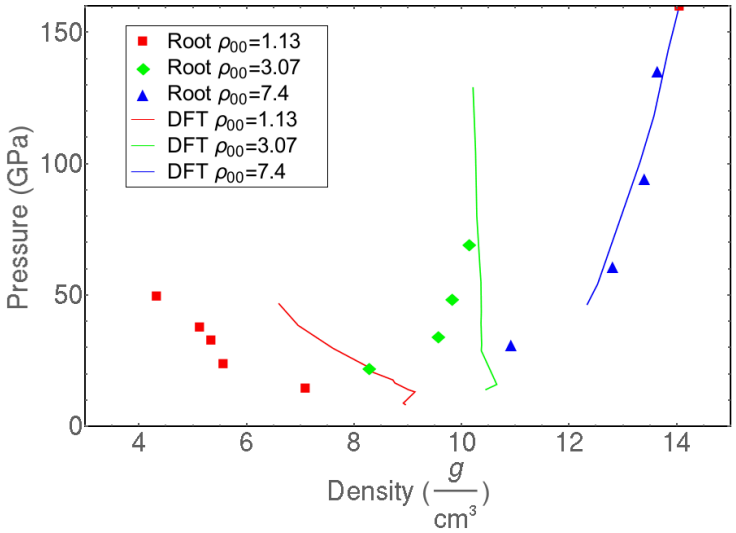
Micrographs of aerogel and porous tantala Ta_2O_5

The calculated tantala Hugoniots for varying initial densities are not in good agreement with data

- Using the Rankine Hugoniot relation on the DFT created table, calculate the locus of points for each initial density
- Table matches the highest density Hugoniot well
- As initial density decreases, the tables approximation to experimental data deteriorates
- Most common porous approximation is to change from v_0 to v_{00} in RH relation
- Inconsistent initial energy and density

$$E_f - E_0 = \frac{1}{2}(P_f + P_0)(v_{00} - v_f)$$

Tantala Hugoniot for different initial densities (nominal density 8.50 g/cm^3)



The ability to represent the experimental data deteriorates as the initial density (ρ_{00}) decreases,

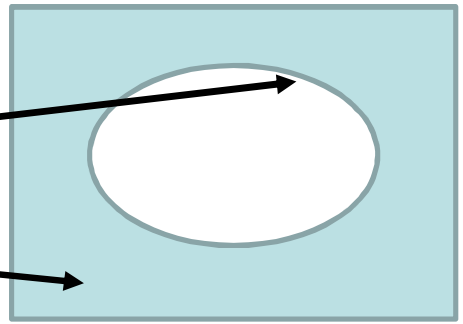
Porous materials pose a challenge for the reference state.

- The initial and final states of a steady shock satisfy

$$E_f - E_0 = \frac{1}{2}(P_f + P_0)(v_0 - v_f)$$

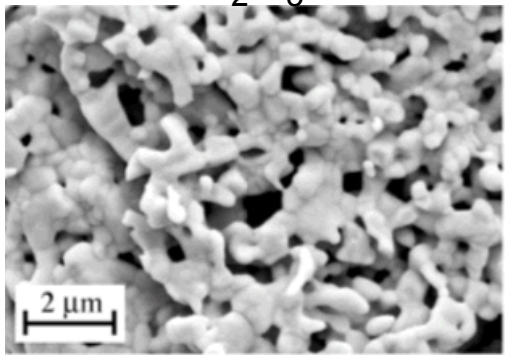
- Assume RH can be applied to porous materials
- For an initially porous material at v_{00}, E_0 will not be correctly determined from an EOS table at the initial ρ_0,T
- A common approximation is to use E_0 from the ambient state (ρ_0) but use v_{00} to satisfy the initial (porous) density
  - *An assumption that there is no energy cost to create a surface*
- This is incomplete and the additional information of surface energy and – area must be included

$$E_0 = E_{Bulk} + E_{Surface}$$

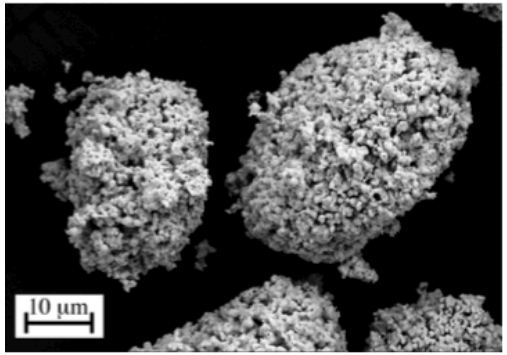


Creating porous samples

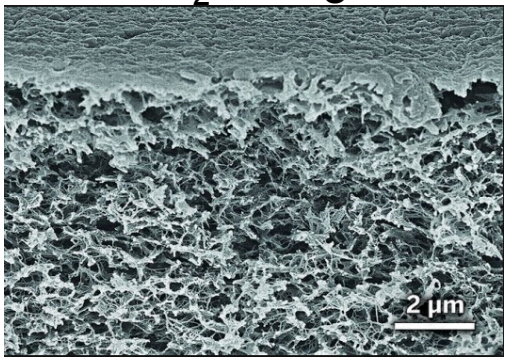
Ta_2O_5



Ta_2O_5



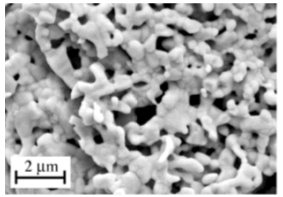
SiO_2 Aerogel

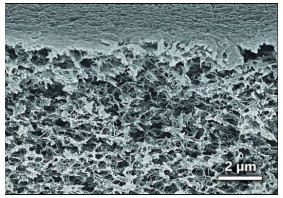


- Tantala flakes:
  - placed in a container and tamped or pressed
  - Vibration used to help create uniform distribution
  - Assume pure Ta_2O_5
- Aerogel:
  - Chemically dissolved
  - Another chemical rinse to stabilize
  - Assume pure SiO_2, no residual byproducts

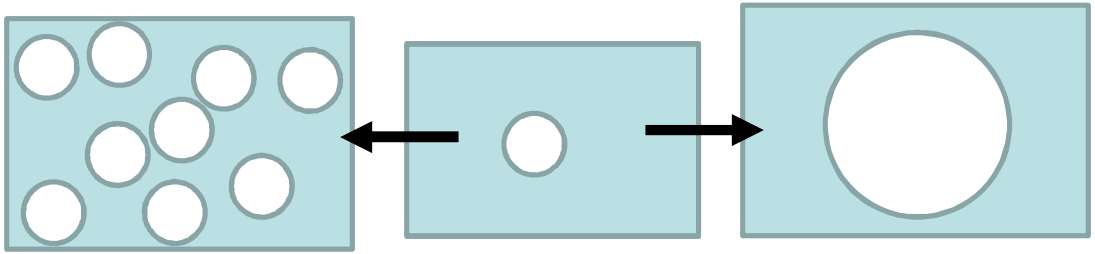
The surface energy plays an important role in porous materials by determining the internal energy of the initial state

- It is not possible to model the full macroscopic geometry with DFT methods
- σ_s is often known – or it can be determined via DFT calculations of surfaces
- Chose the form: $E_{surface} = \sigma_s * A_s$
- A_s (surface area) depends on morphology and length-scale – a challenging parameter to set – *for now a fitting parameter*
- N is constant ($(Nv_0)^{\frac{1}{3}}$ is feature size), v_{00} is initial volume, v_0 is dense volume
- *Feature size about 10 nm for tantala – smallest scale are relevant*





$$A_s = \frac{6v_0^{\frac{2}{3}}}{N^{\frac{1}{3}}}\left(\frac{v_{00}}{v_0} - 1\right)$$



$$A_s = \frac{6}{N^{\frac{1}{3}}}(v_{00} - v_0)^{\frac{2}{3}}$$

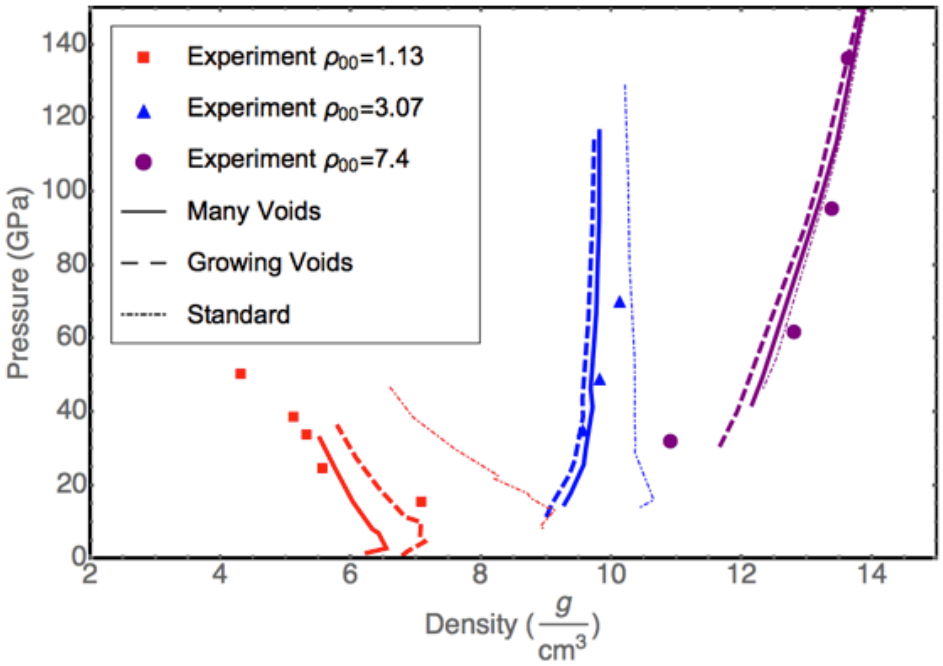
Increasing porosity growing the number of voids – or with a growing void

DFT based Tantala EOS Hugoniots with surface energy modifications are in better agreement with data

- Zero D (spherical cow) approximation to porous shock data
  - No surface roughness
  - Nonplanar surface energy?

$$E_f - (E_0 + \sigma_s A_s) = \frac{1}{2}(P_f + P_0)(v_{00} - v_f)$$

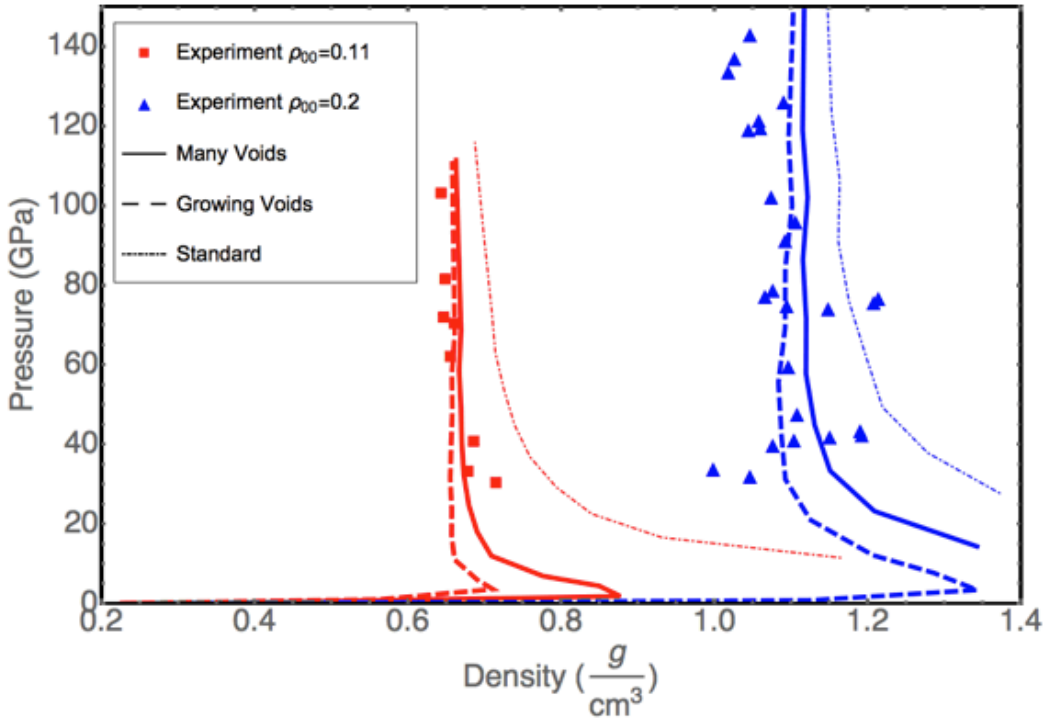
- The modification to the Rankine-Hugoniot relation shifts the calculated Hugoniot to be in line with experimental data
- The lowest density Hugoniot shows the crush-up followed by the turn-over as heating over rides compression
- *Correction term improves comparison to experiment in materials tested so far (tantala, silica aerogel, copper)*



Tantala Hugoniot for different initial densities (nominal density 8.50 g/cm^3)

The correction works very well also for silica (SiO_2) Aerogel – dramatically improving the agreement with data

- Rectilinear aerogel EOS table from DFT – nominal density 2.65 g/cm^3
- Hugoniots calculated from table do not match porous data (thin dotted lines)
- Surface correction recovers behavior for 0.11 and 0.2 g/cm^3 aerogels: many voids (solid) and growing (dashed)
- Many void approximation
  - σ_s = 0.25 eV/Å^2 and N=50,000
  - v_0 = 15.09 Angstoms/atom
  - $(N*v_0)^{1/3}$~10 nm
- Growing void approximation
  - σ_s = 0.25 eV/Å^2 and N=1500
  - $(N*v_0)^{1/3}$~3 nm
- *One tuning for both porosities*



There is a way to incorporate porosity into first-principles simulations of shocked materials

- Summary
  - The standard $E(V_0)$ Rankine-Hugoniot relation does not represent the behavior of a porous material Hugoniot well
  - Calculate or find the surface energy and tune a feature size
  - For tested materials, σ_s, A_s, and N are constant
  - *The correction (added energy term) is scaled by the ratio of the porous initial density (ρ_{00}) to the reference density (ρ_0)*
- Future Work
  - Can we determine the surface area?
    - *Experiments/simulations?*
  - Directly investigate the dynamics of porous shocks
    - *DCS – imaging at a small scale?*
    - *Is the shock planar and sharp or an average of many different particle and shock velocities?*