Los Alamos NATIONAL LABORATORY

New Sesame Equation of State for Iridium

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Shock Compression of Condensed Matter – 2023
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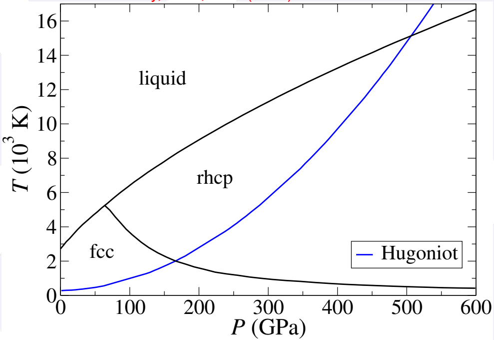
Why Ir?

- Mainly interested in use as high-impedance flyer for Z
  - Second-highest density among naturally-occurring metals, $\rho_0 = 22.56$ g/cm^3
  - High melting temperature, $T_{\mathrm{melt}} \approx 2720$ K (4435 °F)
  - Excellent corrosion resistance
- Known fcc and liquid phases
  - T_{melt} is an ITS-90 fixed point
- Tentative identification of a random-stacking hex closed packed (rhcp) phase
  - not studied here
- We have constructed a *preliminary* EOS table for Ir
  - Sesame 93550

wikipedia

Burakovsky, et al., PRB (2016)



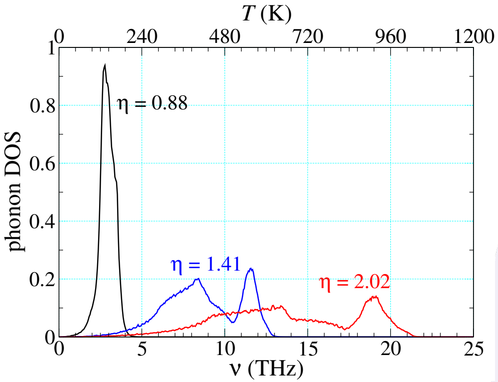
Sesame Model

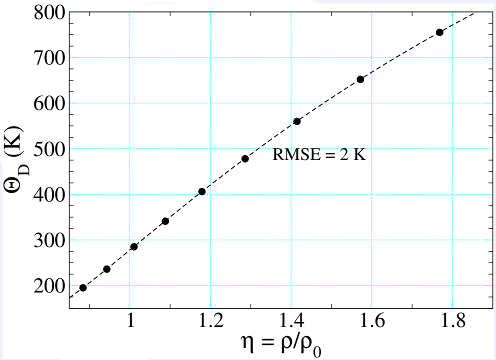
- 3-part decomposition of Helmholtz free energy

$$F(\rho, T) = \phi(\rho) + F_{\text{ion}}(\rho, T) + F_{\text{elec}}(\rho, T)$$

- Included only fcc and liquid phases
- Cold curve based on fit to shock data, then 0 K TFD
  - Assumes Mie-Grüneisen form
    - Accurate to within $\sim$10% over the domain of the shock data
- Ionic excitations
  - Quasiharmonic approximation: Θ and Γ are $f(\rho)$ only
  - Lindemann melt line
  - hightliq interpolation (liquid)
- Electronic excitations
  - Thomas-Fermi-Dirac

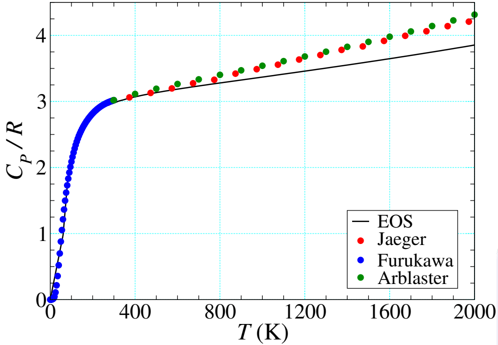
Ion Thermals

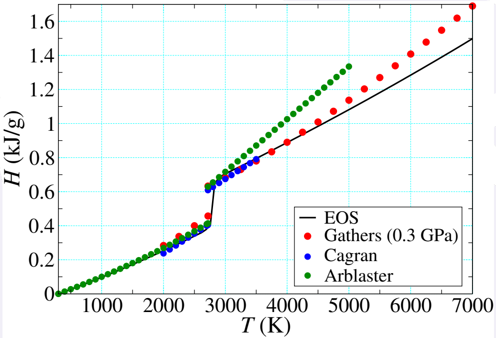




- Phonons by direct force method $\rightarrow$ density of states as $f(\eta)$ (left)
- Moments of phonon DOS $\rightarrow$ characteristic temperatures $\Theta(\eta)$ (right)
- $\Theta(\eta) \rightarrow \Gamma(\eta)$ through $\Gamma = \mathrm{d} \ln \Theta / \mathrm{d} \ln \eta$

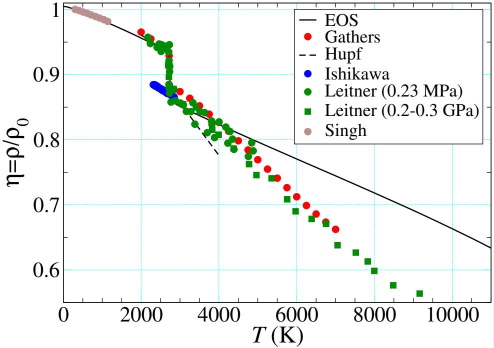
Isobaric Results

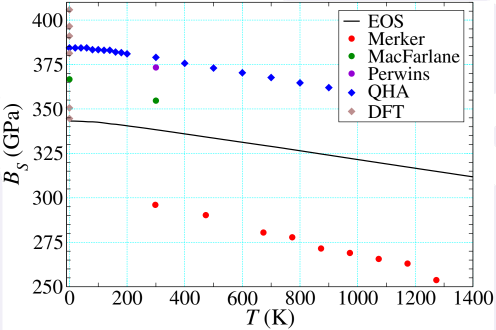




- Deviation in C_P due to some combination of strong anharmonicity (Moseley, et al., PRM 2020) and electronic contribution
- High $\rho \rightarrow$ low H for $T \gtrsim 5000$ K (next slide)

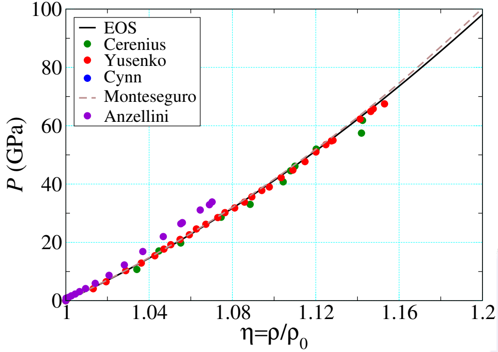
Isobaric Results

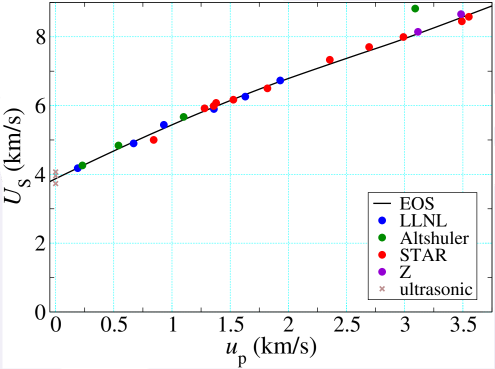




- Liquid needs to deviate strongly from solid $\rightarrow$ hard optimization problem
  - This is where automation (particle swarm, MCMC) could be really helpful
- Bulk modulus in middle of experimental data, low end of DFT
  - PW91 and PBE give a result very close ($<$ 2%) to the EOS

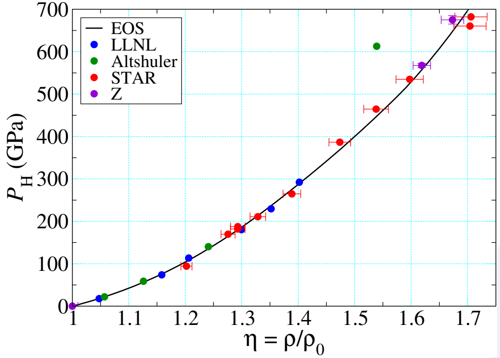
Static and Dynamic Compression

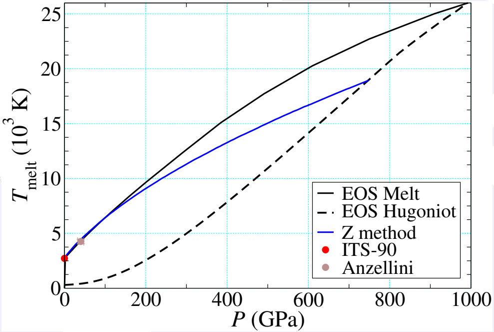




- New Z points consistent with previous shock data
- Fitting to shock data recovers most of the DAC data
  - Only functional that isn't too stiff *and* recovers ρ_0 is PW91

Shock and Melt

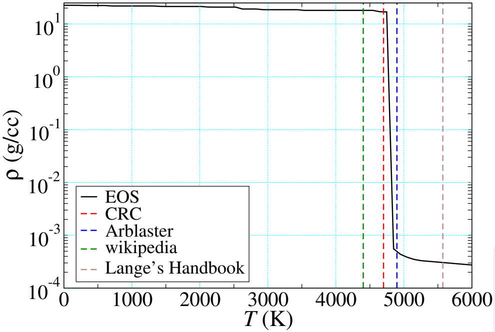


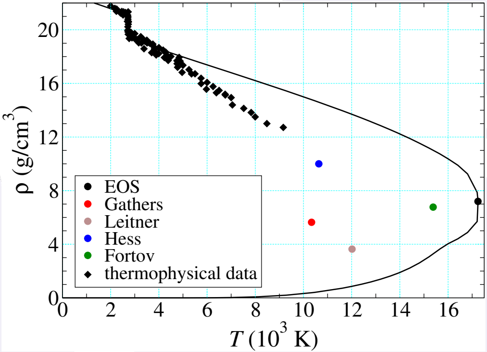


- Melt line agrees well with DFT (Z method) $P \leq$ 150 GPa, then stiffer
  - Both consistent with 2 experimental points
  - Important parameter (dΓ/dρ) poorly constrained by phonon calculations
- Z method predicts highest P_{H} points almost melted, EOS predicts shock melting at P_{H} $\sim$1000 GPa

Burakovsky, et al., PRB (2016)

Vapor Dome





- Use boiling temperature (T_b) to constrain cohesive energy
  - Reference values for T_b vary by over 1000 K!
  - Cohesive energy from Kittel agrees well with "best" T_b
- This still yields a critical temperature (T_c) that is much too high
  - Again, liquid ρ too high $\rightarrow$ T_c too high

Next Steps and Acknowledgements

- Would be nice to:
  - Fix the liquid density – move over to MCMC (uncertainty quantification!)
  - Nail down location of shock melting
  - Better constrain the critical point. DFT?
  - Replace TFD with QAA (Tartarus)
  - Coupled calibration with strength model (SNL)
- Thanks to:
  - Scott Crockett for funding
  - Travis Sjostrom and Danny Rehn for code development
  - Scott Crockett, Josh Townsend, and coauthors for lots of helpful conversations