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Efficient prediction of equations of state and strength properties using new electronic structure methods

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LLNL-PRES-??????

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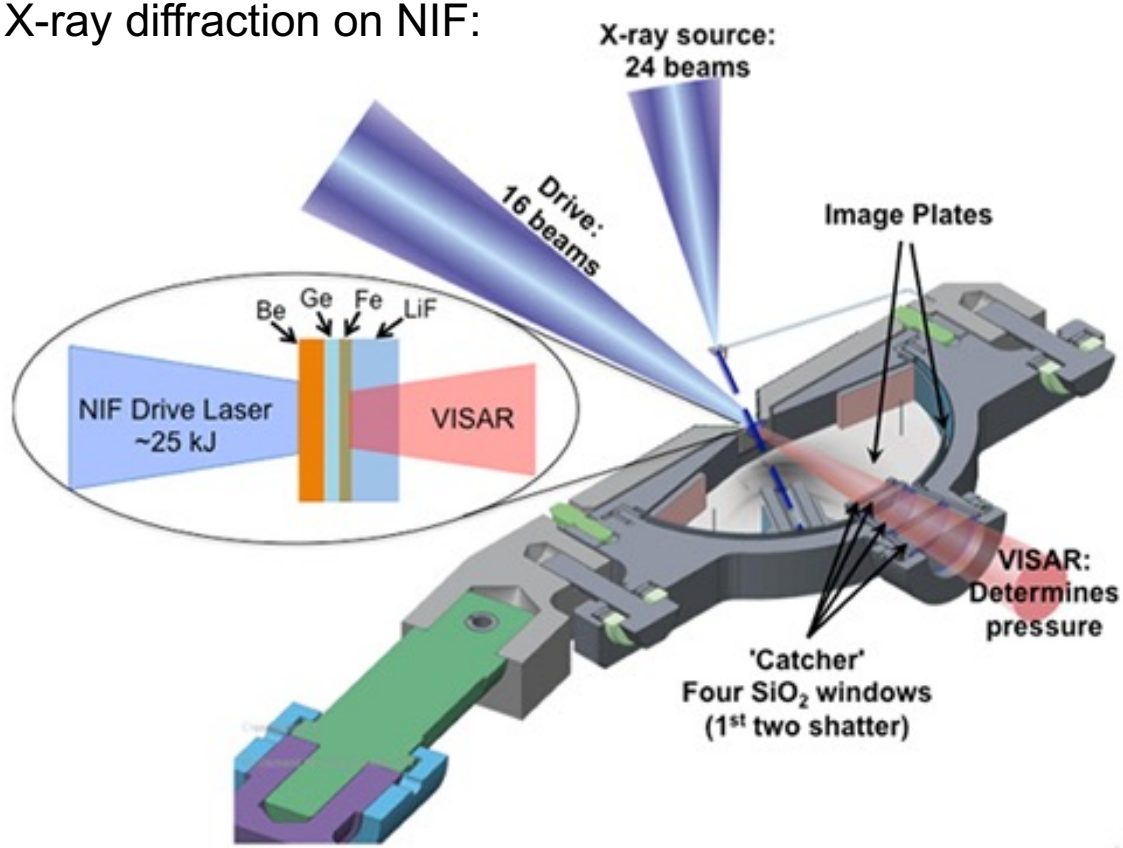
High energy density experiments: unusual materials and states

X-ray diffraction on NIF:



Ablators: Be, C_{dia}, Cu, …

X-ray shields: Au, Pb, Re, **Ge**, …

Tampers: C_{dia}, LiF, MgO

Glue

TPa states, solid or liquid

10 ns timescales

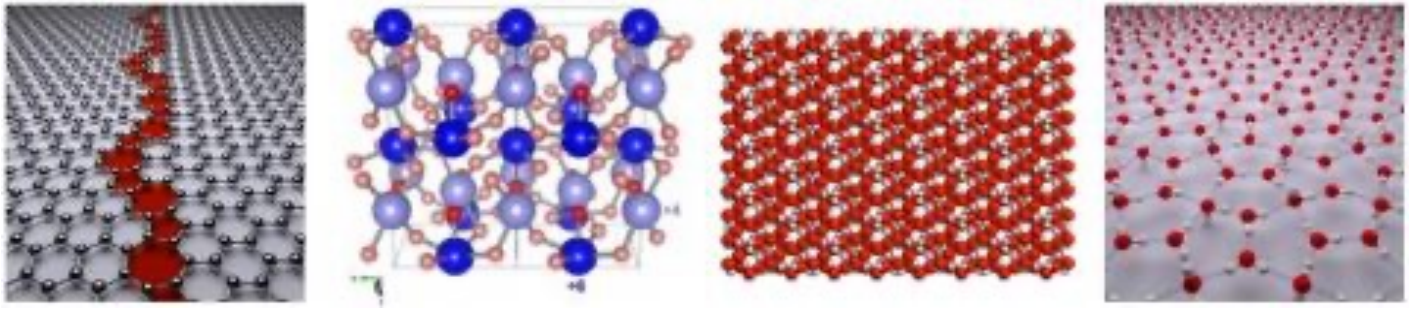
Design – Interpret – Relate to longer scales

Need: “adequate” hydrocode models, timely with experiments

No structure-searching!

Ambient, common, seen or suspected.

Minimal (Q)MD.



Model	Approach	Issues
EOS	DFT cold curve, ion-thermal	Accuracy, sensitivity to XC
Shear modulus	DFT elastic strain	Numerical noise
Flow stress	NxSG, dislocation model	Polymorphism, availability
Conductivity	Ambient or plasma	Range, Boltzmann

Improvements: speed and accuracy of DFT

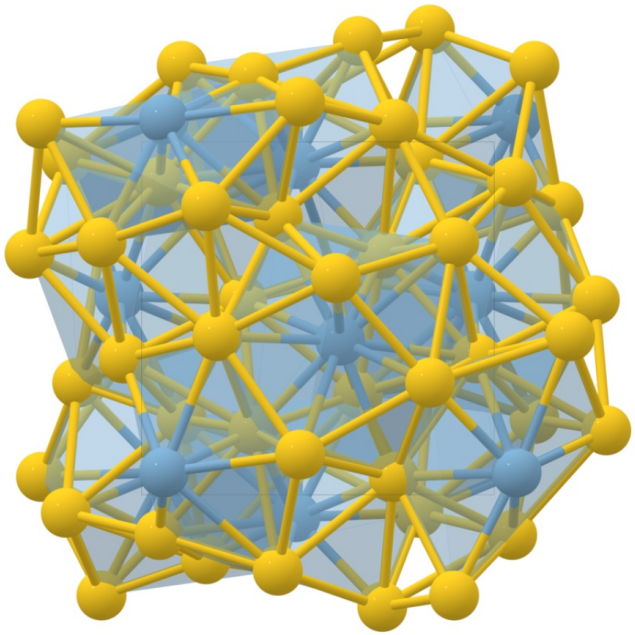
Electronic structure theory predicts EOS and (maybe) strength adequately – or better

FP-LMTO calculations:
3D multi-atom all-electron
(Per Söderlind)

SPARC-X calculations
(John Pask): fast

Less noise and uncertainty
in DFT predictions

High-pressure properties,
inc compounds and alloys



New data!

$$f(\rho,T) = f_c(\rho) + f_i(\rho,T) + f_e(\rho,T)$$

Ion-thermal EOS from elastic moduli

Boson energy for each phonon mode. Many similar; integrate to find ion-thermal EOS => details unimportant.
Can represent as a few effective Debye modes, or even just one.
Estimate Debye modes from elastic moduli, or bulk and shear moduli, or longitudinal and shear wave speeds.
Electronic structure: computationally easier than calculating phonons (symmetry, supercell, imaginary modes).

E. Madelung, Phys. Z. **11**, 898 (1910), A. Einstein, Ann. Phys. Leipzig **34**, 170 (1911).
O.L. Anderson, J. Phys. Chem. Solids **24**, 909 (1963).

Still used in recent literature e.g. X. Liu & H.-Q. Fan, *R. Soc. open sci.* **5**, 171921 (2018),
D.C. Swift et al, Phys. Rev. B **105**, 024110 (2022).

$$v_s = \sqrt{\frac{G_x}{\rho}}; v_p = \sqrt{\frac{B_x + 4G_x/3}{\rho}}; v_m = \left(\frac{2/{v_s}^3 + 1/{v_p}^3}{3}\right)^{-1/3}$$

Use average wave speed instead of integrating over orientations and polarizations.

$$\Theta = \frac{h}{k}\left(\frac{3nN_A\rho}{4\pi M}\right)^{1/3} v_m,$$

Hierarchy of approximations, but at least likely to predict systematic variations. Accurate if isotropic?

Grueneisen parameter: logarithmic derivative of Debye temperature $\Gamma(\rho) = \frac{\rho}{\theta_D}\frac{d\theta_D}{d\rho} = \frac{\rho}{v_m}\frac{\partial v_m}{\partial \rho} + \frac{1}{3}$

Converse: estimate G from θ_D and B. Numerical inverse of $\theta_D(\rho, v_m(B,G))$ by bisection, bracket $=(\alpha_1, \alpha_2)B$.

Dislocation plasticity model for HED conditions

Stresses high enough to cause plastic flow; wide range of pressure, temperature, strain rate; polycrystal.

Plastic relaxation rate: Orowan equation for single dislocation density: $\dot{\epsilon}_p = \frac{\eta}{M}\rho_d b \bar{v}_d \qquad \bar{v}_d = bZ$

Redefine dislocation density as per atom rather than length/volume: $\phi_d = \rho_d \frac{m_a}{b\rho} \qquad \dot{\epsilon}_p = \frac{\eta}{M}\phi_d Z \gamma$

Structure-dependent Burgers length-scale: $\gamma \equiv \frac{6 f_v^3}{\pi}$

Dislocation hop rate: $Z = Z_0 \left[\exp\left(-N \frac{E_P - E_\tau}{k_\mathrm{B} T} \right) - \exp\left(-N \frac{E_P + E_\tau}{k_\mathrm{B} T} \right) \right]$

Enthalpy from applied shear stress: $\|\sigma\| v_\mathrm{WS} f_v$

Dislocation evolution rate: $\dot{\phi}_d = \frac{\pi}{2}\frac{\phi_d}{M} Z \left(1 - \frac{2}{\bar{L}}\right) - \frac{2\phi_d Z_a}{\bar{L}} = \frac{\dot{\epsilon}_p}{\gamma}\frac{\pi}{2}\left(1 - \frac{2}{\bar{L}}\right) - \frac{2\phi_d Z_a}{\bar{L}} \qquad \bar{L} = \sqrt{\frac{2M}{\phi_d}}$

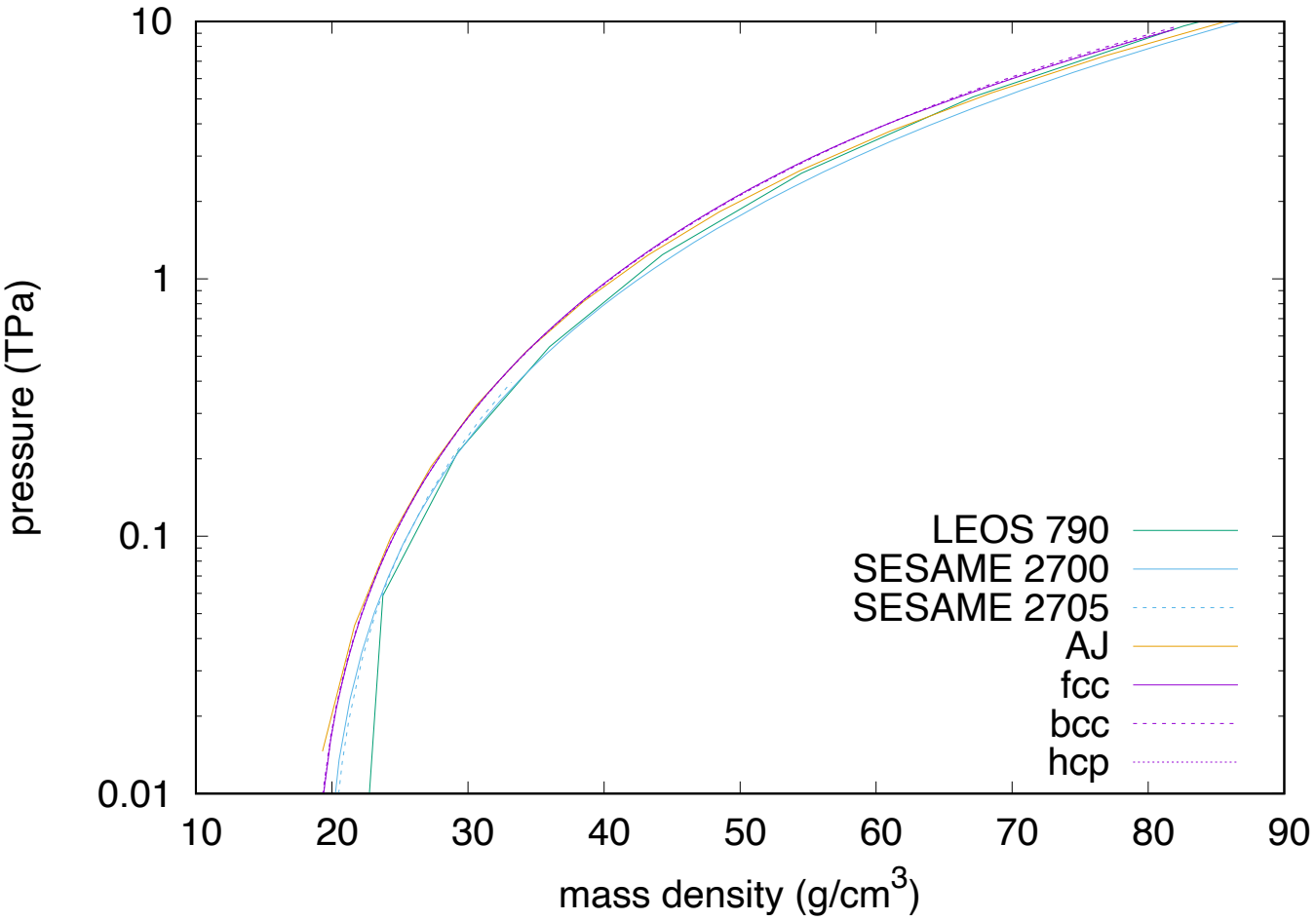
Annihilation from attraction: Z with $E_\tau \to E_m = \frac{G v_\mathrm{WS} f_v}{\bar{L}}$ (strain fields from opposite Burgers vectors)

Hardening: $Z \to Z f_h$: $f_h \equiv 1 - \frac{1}{\bar{L}'}$ mean distance to dissimilar dislocation: $\bar{L}' = \sqrt{\frac{M-1}{\phi_d}}$

Parameters (vary with state): Peierls barrier E_P, hop attempt rate Z_0, shear modulus G. Used AJ theory.

D.C. Swift et al, arXiv:2110.06345

FP-LMTO cold curves for Au: more rigorous than atom-in-jellium



Per Söderlind:

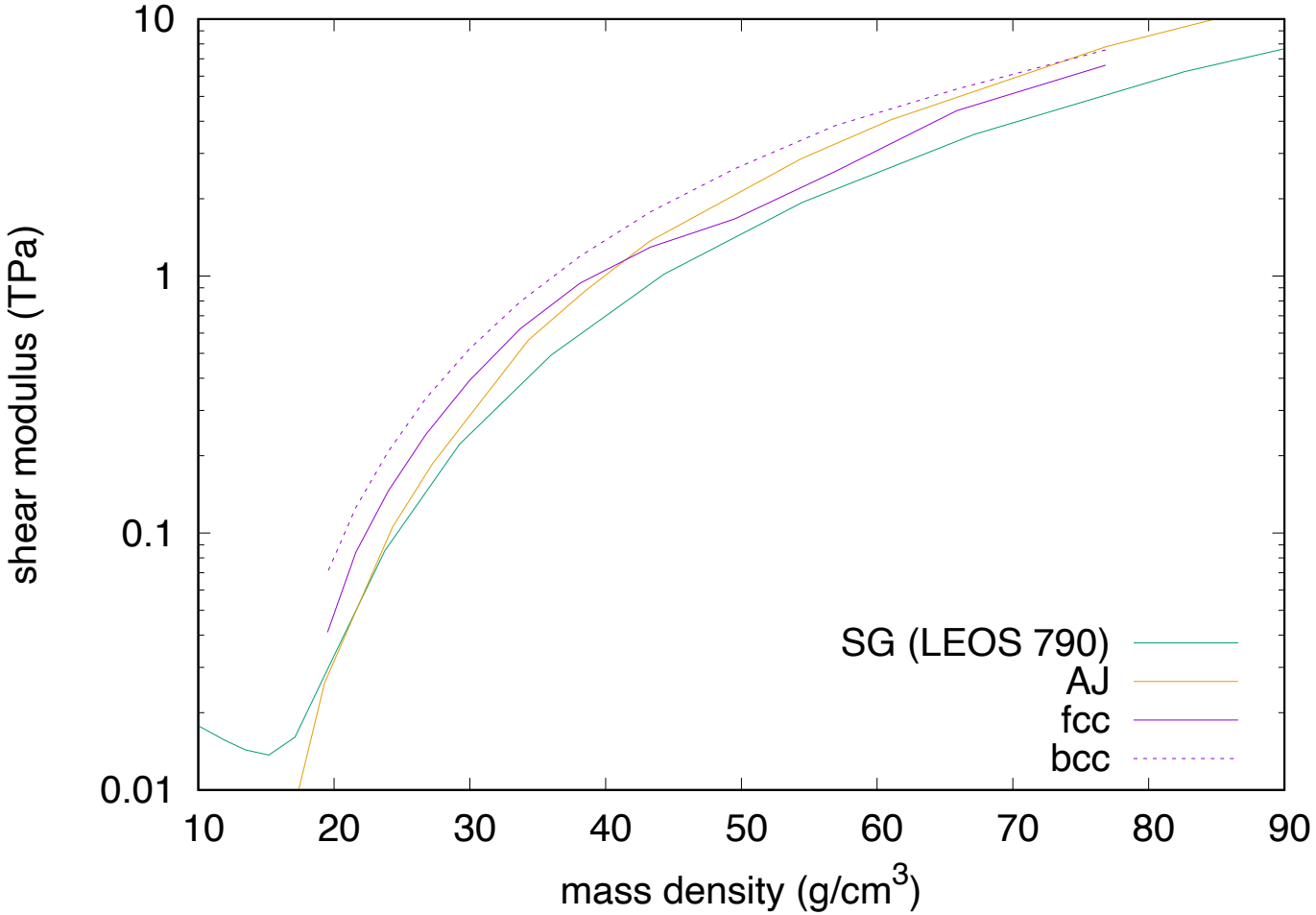
- All-electron, scalar-relativistic.
- 43 electrons/atom in valence.
- fcc, bcc, hcp (relaxed c/a)
- k-points:
  fcc, bcc ~3000
  hcp ~1200
- Cold curves: GGA.

Phase changes predicted:
fcc – hcp – bcc – hcp – fcc

TF models seem too soft at ~TPa.

Highest-quality theoretical prediction in this regime.

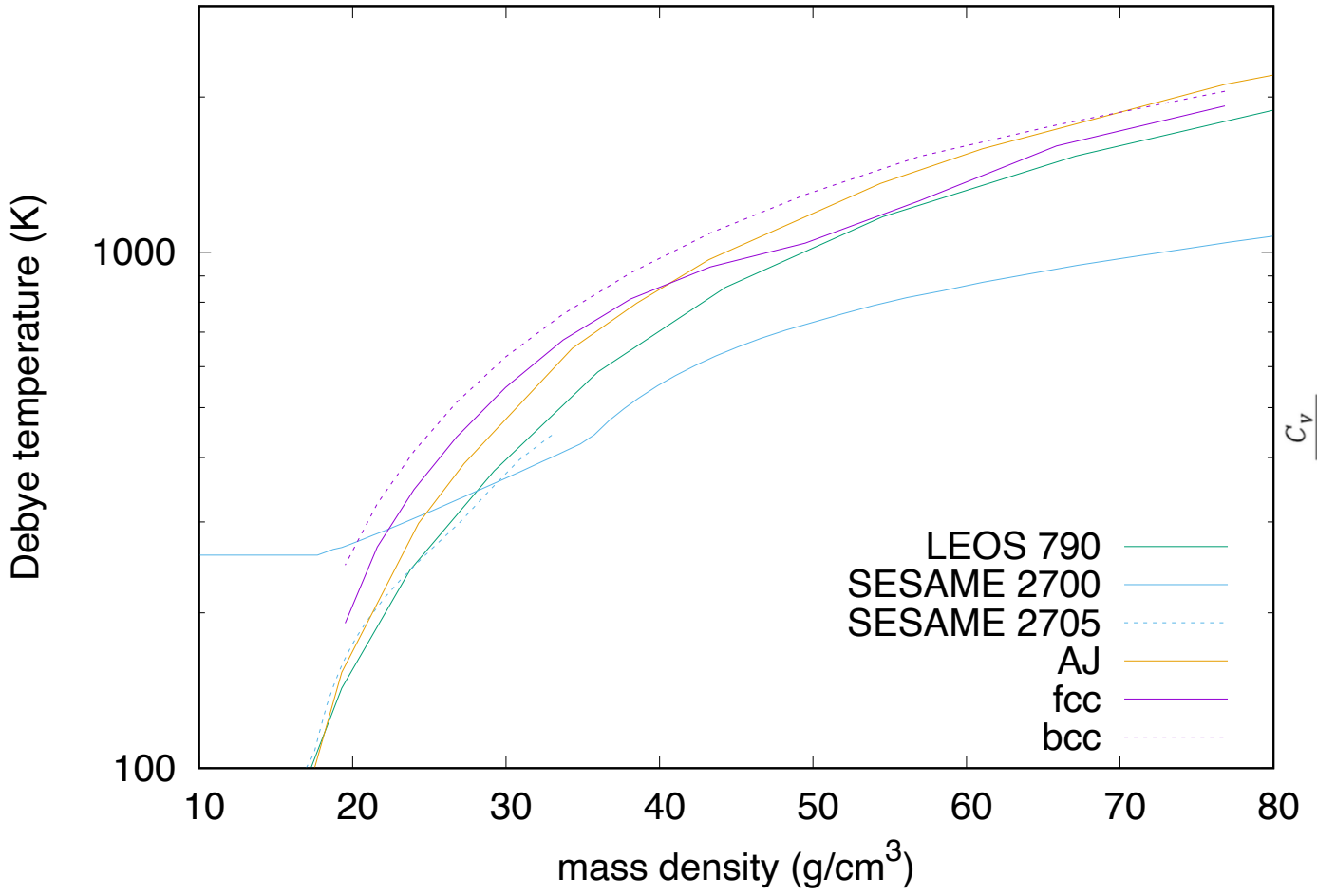
FP-LMTO shear modulus for Au: more rigorous, phase-aware



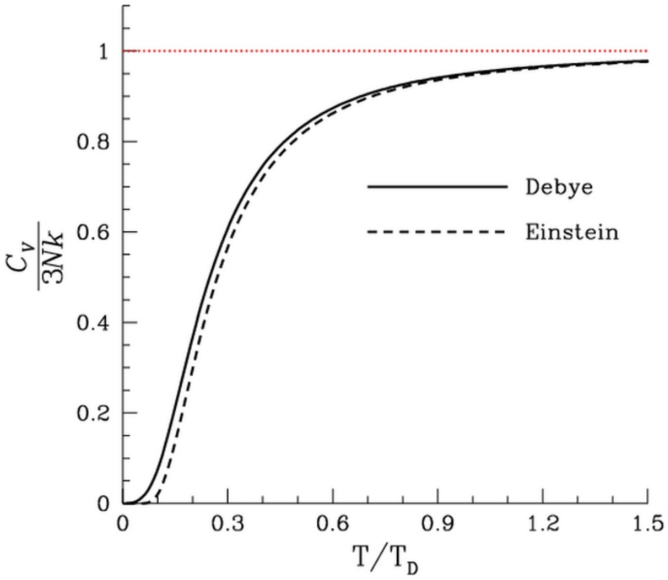
Per Söderlind:

- Elastic moduli: LDA.
- Shear modulus: Voigt average.

Debye temperature deduced from DFT and EOS models

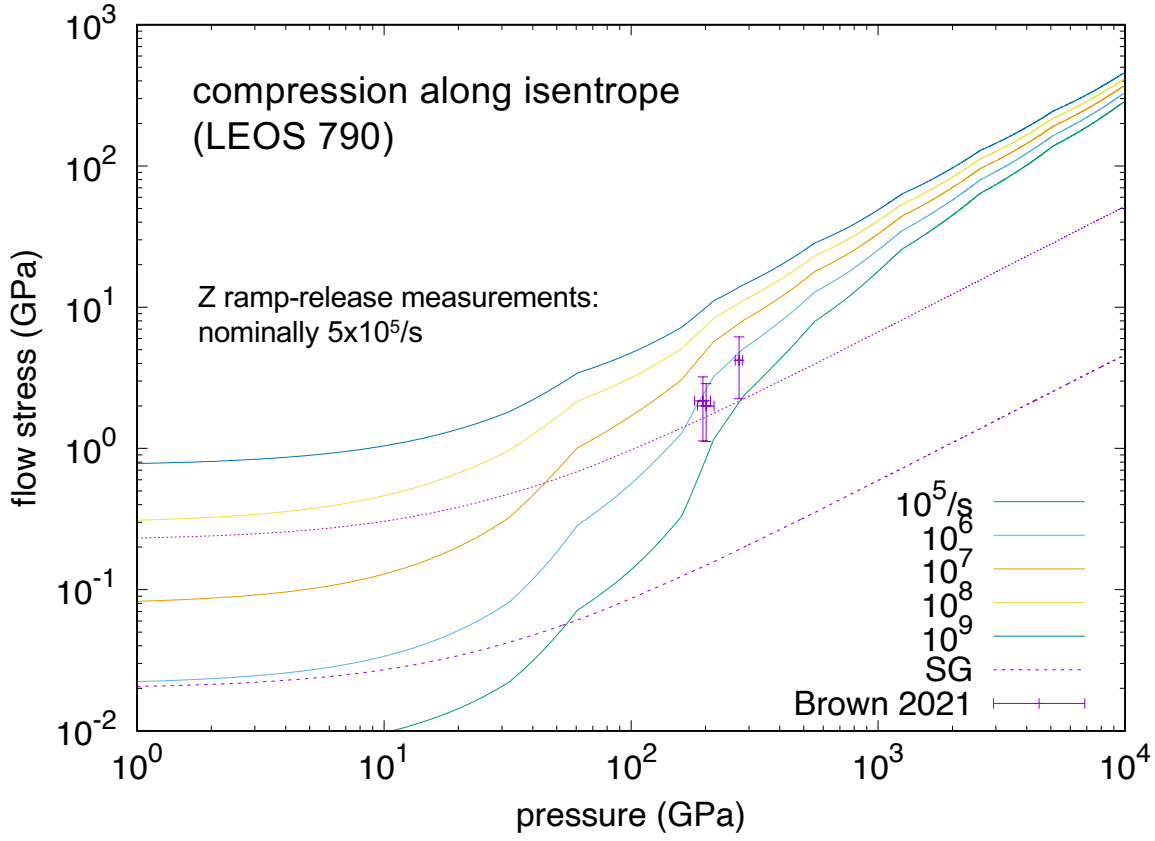


EOS models:
deduced θ_D from T
where $c_v = c_{DP}/2 \simeq \theta_D/2.9829$



Deduce $G(\rho)$ consistent with any EOS model

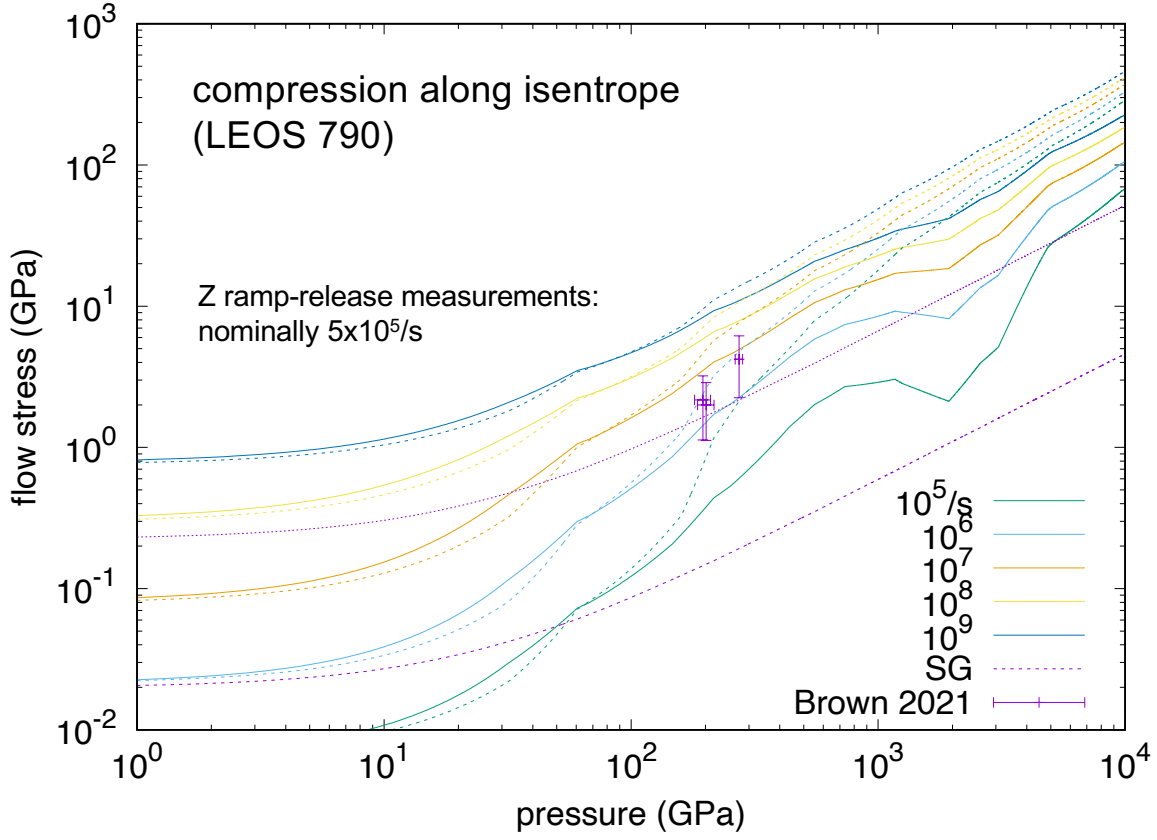
Performance of AJ dislocation model for Au



J.L. Brown et al, J. Dyn. Behavior Mat. ***7****, 196-206 (2021)*

D.C. Swift et al, arXiv:2110.06345

Performance of FP-LMTO dislocation model for Au



B, G → T_{Debye} → $T_{Einstein}$ → dislocation $\dot{\epsilon}_p$

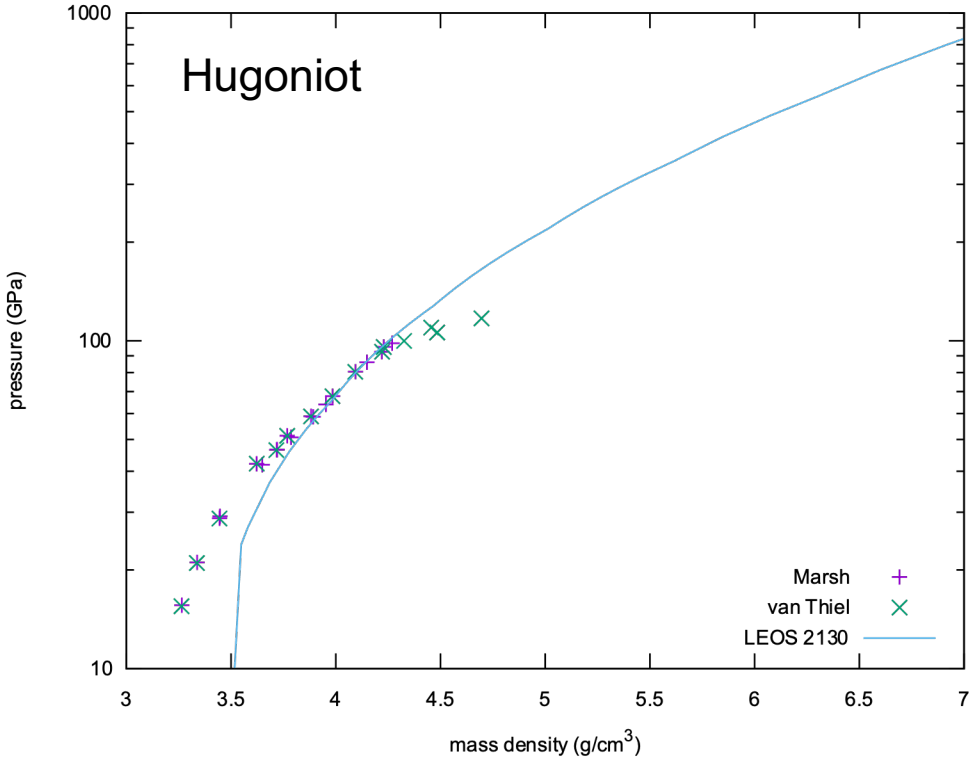
Calibrated f_b against SG Y_0 at 10^6/s.

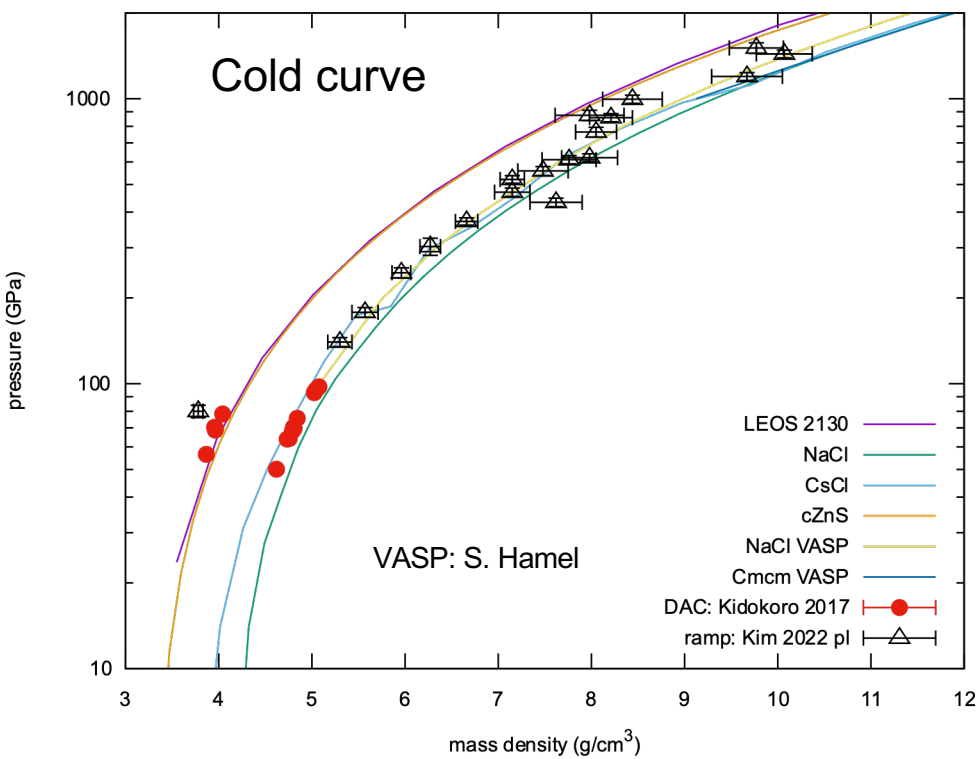
Calculated for fcc only.

Comparison with AJ calibration (dotted).

J.L. Brown et al, J. Dyn. Behavior Mat. ***7****, 196-206 (2021)*

Multiphase EOS predictions e.g. SiC for Z/NIF Discovery Science



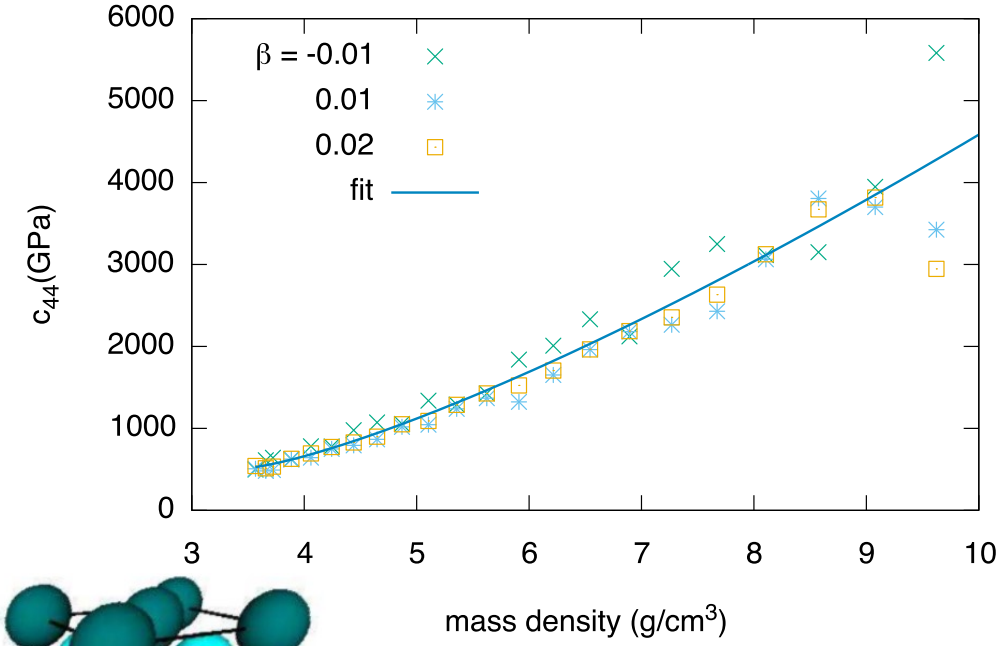


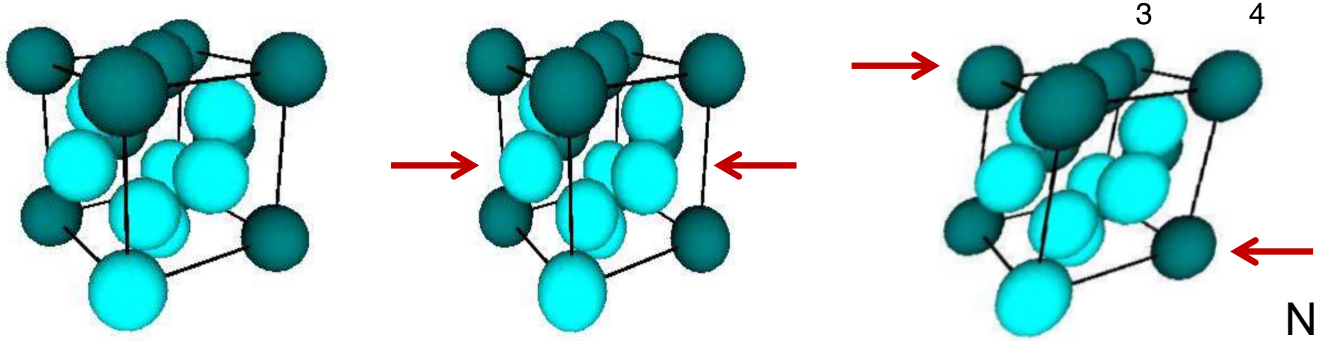
Constructing multiphase EOS models.
Working on associated models e.g. ceramic strength.

CASTEP EOS and elasticity calculations for diamond

- Local density approximation
- Pseudopotential
- Plane wave basis set
- Pulay corrections
- Monkhorst-Pack k-point symmetry reductions
- Lattice cell optimization
- Troullier-Martins potentials

*Swift et al, Phys. Rev. B **105**, 014109 (2022)*





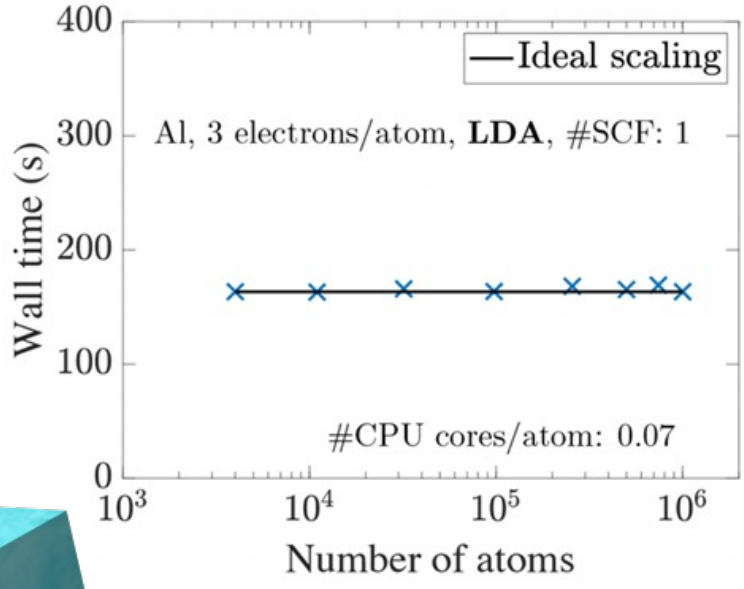
Numerical noise in elastic moduli, filter by hand.

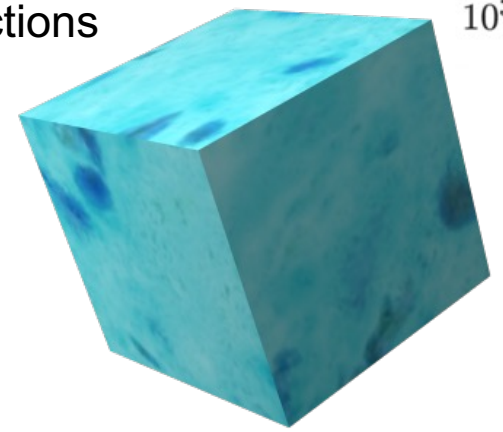
Electronic structure calculations in SPARC-X

- Pseudopotential
- Finite element, 12th order polynomials
- Advanced optimization and convergence
- Parallel, GPU

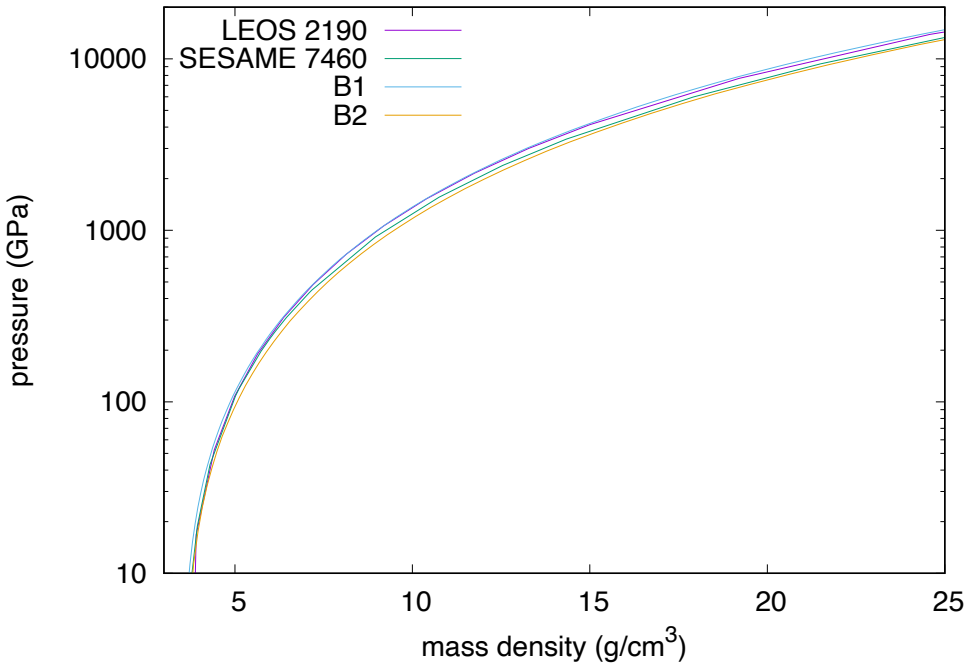
- Hamann ONCV LDA or Pask SMPS soft PBE potentials

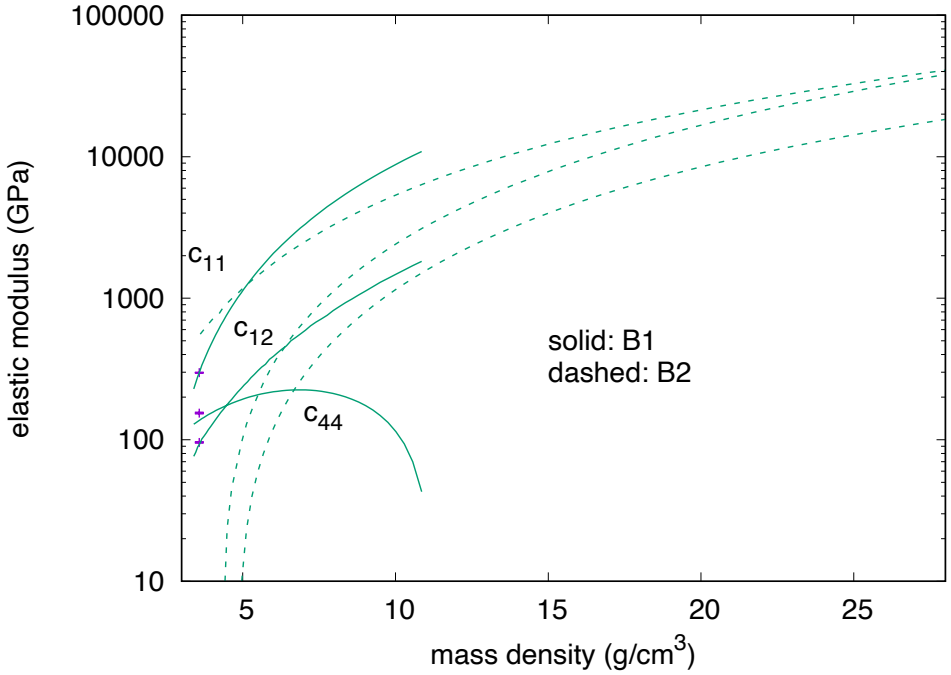
Not currently implemented:
- Pulay corrections
- Monkhorst-Pack k-point symmetry reductions
- Lattice cell optimization





SPARC-X EOS and elasticity calculations for MgO (B1, B2)

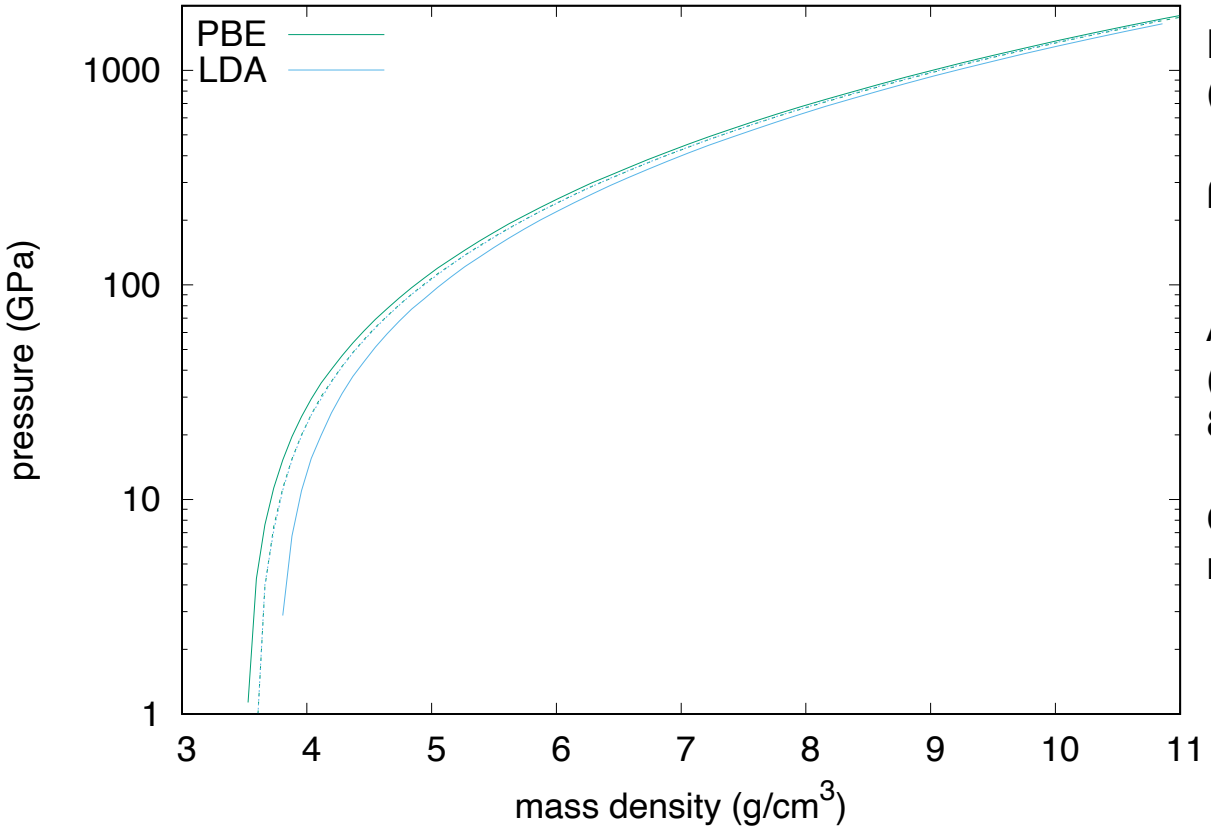




Predicts B1→B2 ~0.5 TPa.

Elastic moduli smooth enough to use directly.

Correction of DFT EOS models to match STP state: insensitive to exchange-correlation functional



DFT: ρ_0 close to STP value, typical accuracy (lattice parameter deviation ~1%).

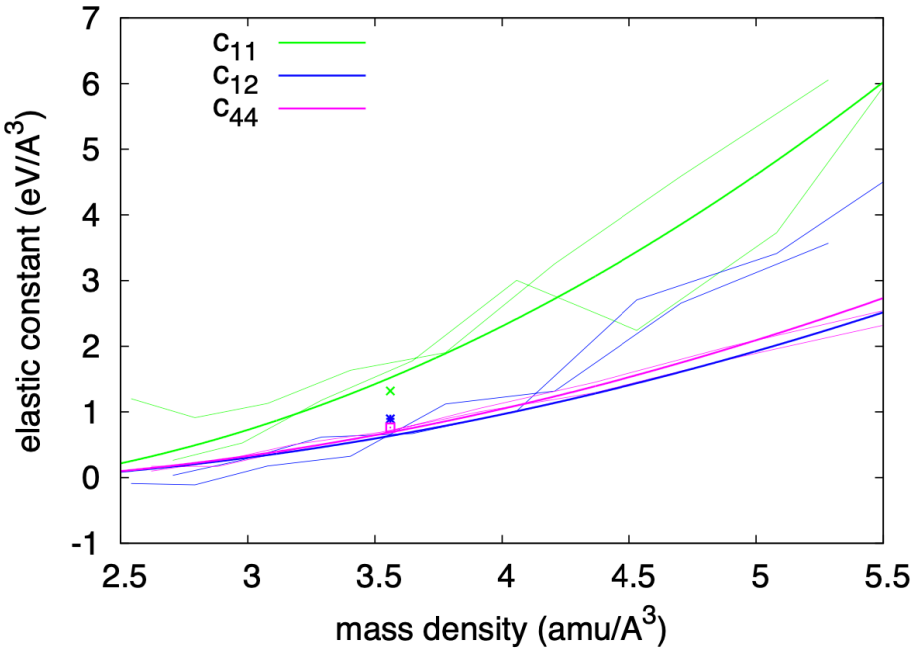
ρ_0 = 3.58 g/cm^3 ⇒ +3.530 GPa (PBE)
or -7.23 GPa (LDA)

Adjustment $\Delta e=\alpha\rho^\beta$ to match STP ρ. (Following H. Akbarzadeh et al, JPCM **5**, 8065, 1993, but consistent as ρ -> 0.)

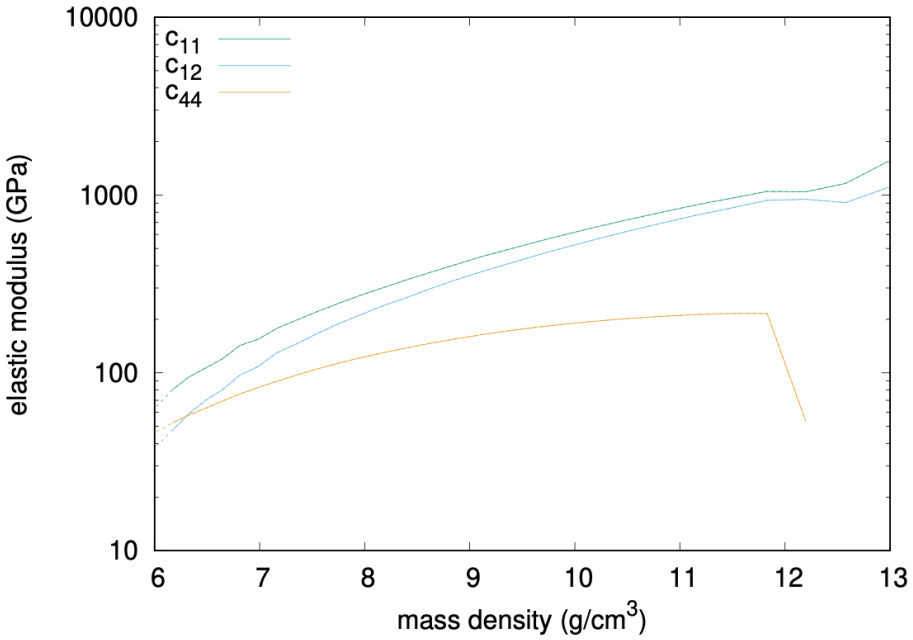
Corrected EOS from LDA and PBE remains bracketed, difference ~10x less.

DFT technology and capability are evolving

NiAl: CASTEP ~2005, several months

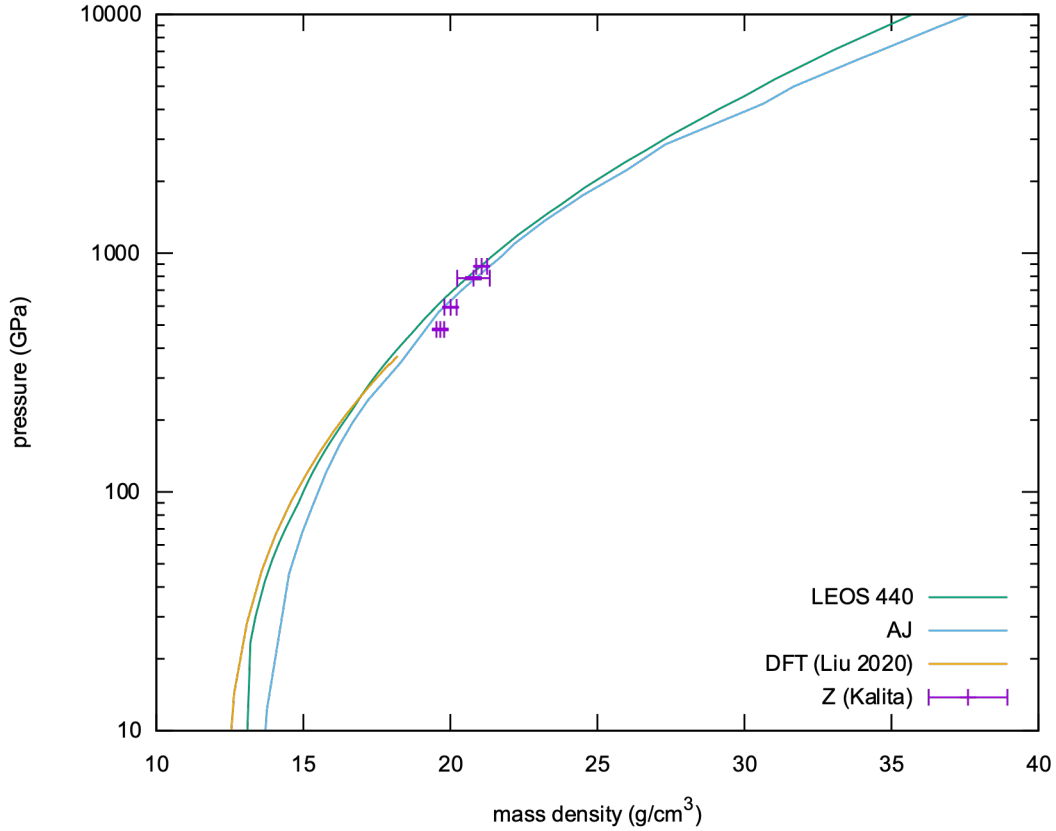


RuAl: SPARC 2022, 73 mins



- practical to predict properties of more complicated materials: compounds, alloys

First Hugoniot data for Ru !



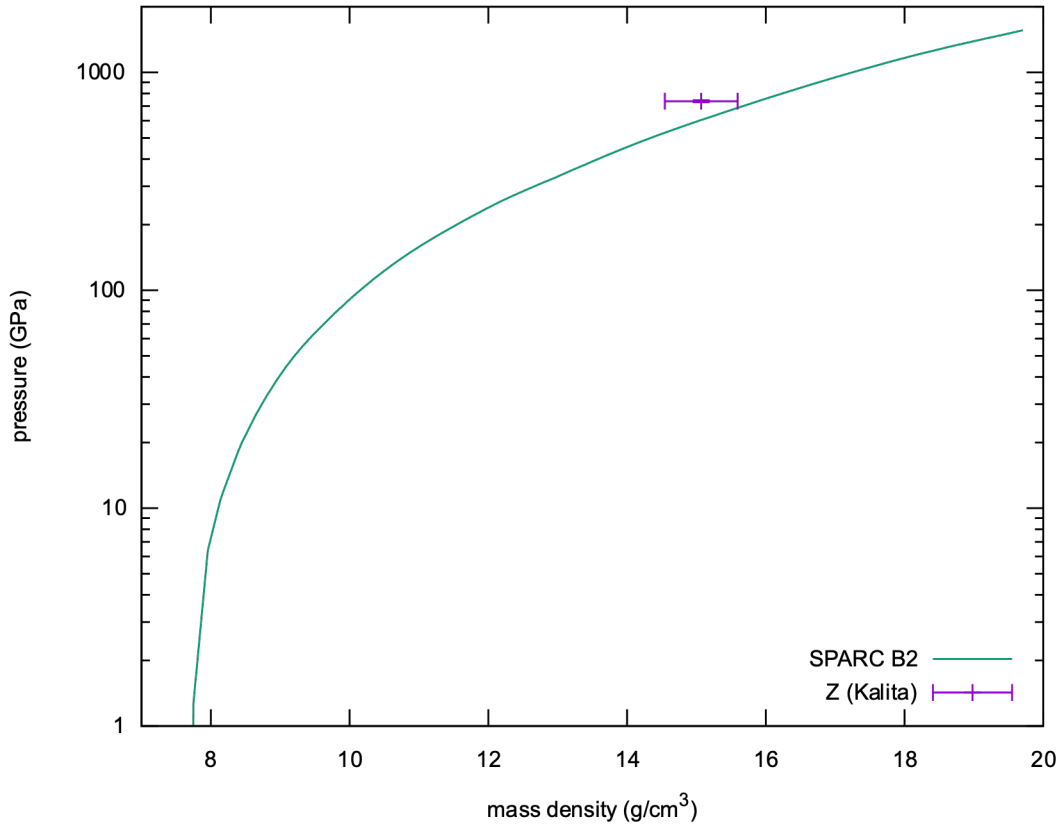
Ru

One of very few elements with no Hugoniot data reported.

QEOS, pseudopotential and atom-in-jellium EOS models constructed
Swift et al, arXiv:1909:05391

DFT study inc Hugoniot to 380 GPa
*Liu et al, Physica B **598**, 412434 (2020)*

First Hugoniot data for Ru and RuAl !



RuAl

Trident data on elastic-plastic transition, spall.
CASTEP cold curves ~2003.
SPARC EOS model constructed.
New samples made (McClellan & Byler).
Z Hugoniot data obtained (Kalita).

YZn

SPARC EOS model constructed <24 hours.
“Good match” to ~0.3 TPa shocks at DCS (Loomis & Peralta).

Minimally-adjusted electronic structure calculations predict EOS; basis for few-parameter plasticity model

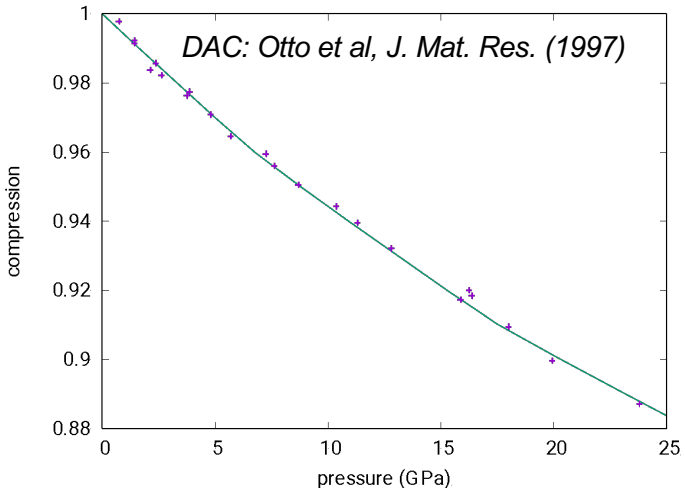
Highest-quality DFT used in place of AJ, older plane-wave codes for HED EOS and strength.

Minimally-corrected DFT EOS nearly independent of XC functional.

EOS models for more components of HED experiments in preparation: MgO, SiC, …

NiAl, B2 structure:

*D.C. Swift et al, Phys. Rev. B, **76**, 134111 (2007)*



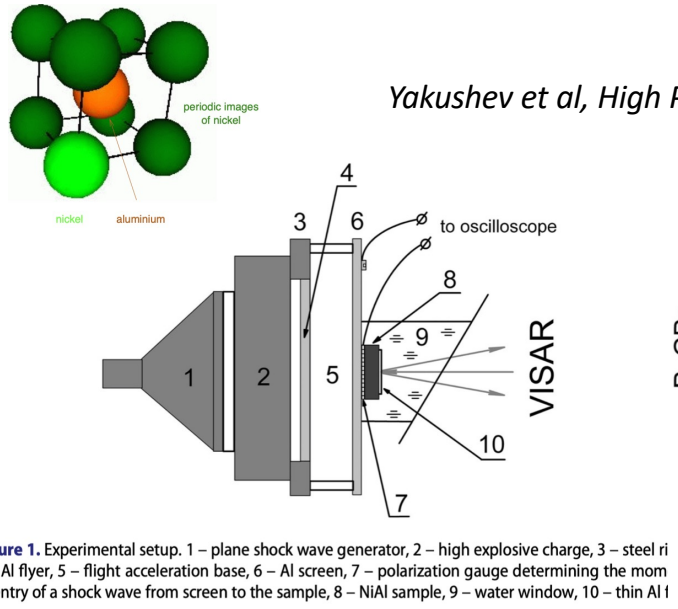
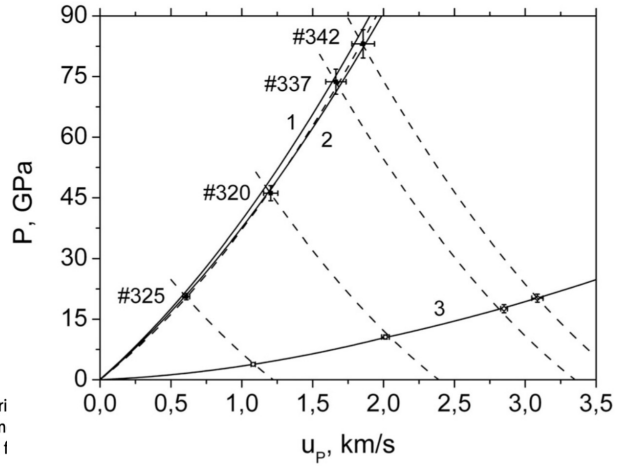


Figure 1. Experimental setup. 1 – plane shock wave generator, 2 – high explosive charge, 3 – steel ri
4 – Al flyer, 5 – flight acceleration base, 6 – Al screen, 7 – polarization gauge determining the mom
of entry of a shock wave from screen to the sample, 8 – NiAl sample, 9 – water window, 10 – thin Al f

*Yakushev et al, High Press. Res. **39**, 3, 471 (2019)*

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