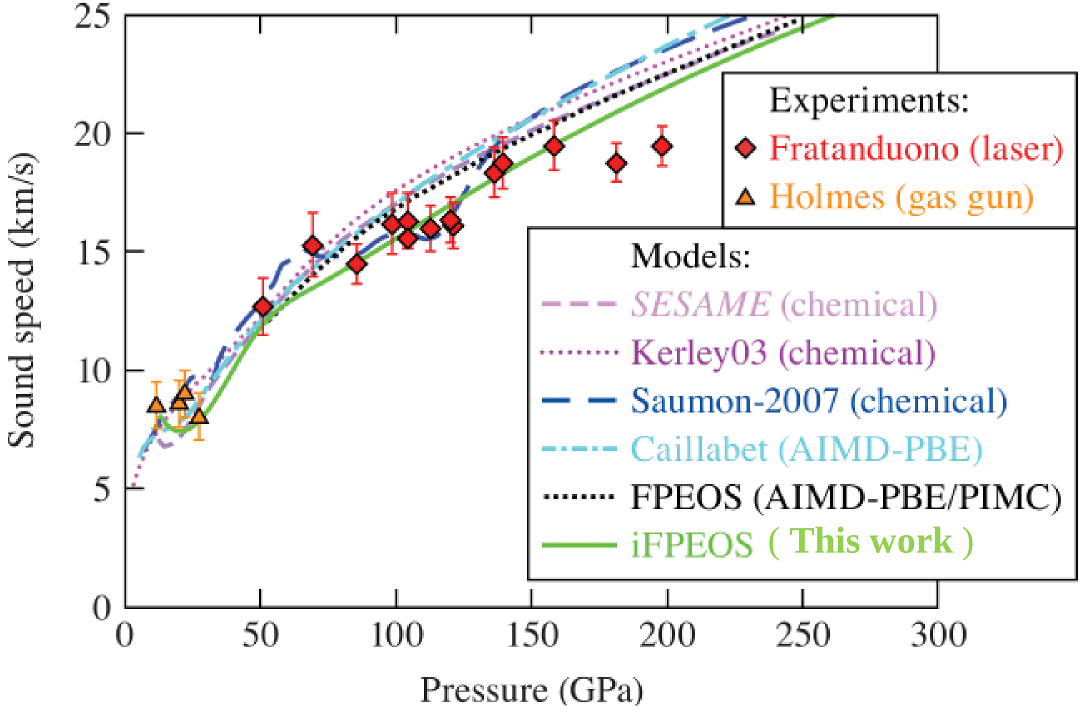
Improved first-principles equation-of-state table of deuterium for high-energy density science applications



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- We have applied MD driven by DFT using newly developed, *finite-temperature*, meta-GGA exchange-correlation functional T-SCAN-L** for consistent and accurate treatment of temperature effects across all conditions.

- Comparing iFPEOS to other EOS models, we conclude that iFPEOS does provide a better agreement with latest experimental measurements of properties such as the principal Hugoniot and sound speed for pressures up to ~200 GPa and temperatures up to ~60 000 K.

Even though we obtain better agreement with experiment at *P* < 200 GPa, the discrepancy between theory and experiment in the high-*P/T* regime is confirmed by iFPEOS.

EOS: Equation of State
MD: Molecular Dynamics
DFT: Density Functional Theory
GGA: Generalized Gradient Approximation
NQEs: Nuclear Quantum Effects

*D. I. Mihaylov, V. V. Karasiev, S. X. Hu, J. R. Rygg, V. N. Goncharov, and G. W. Collins, Phys. Rev. B 104, 144104 (2021).
**V. V. Karasiev, D. I. Mihaylov, and S. X. Hu, “Meta-gga exchange-correlation free-energy density functional to achieve unprecedented accuracy for warm-dense-matter simulations” (2021), submitted to Phys. Rev. Lett.

iFPEOS was motivated by the need for an accurate EOS table for D_2/DT and recent success of new developments in finite-*T* DFT

- Understanding EOS of D_2/DT/H_2 under extreme conditions is still challenging to both theory and experiment, which is evidenced by a systematic disagreement between latest experimental measurements of principal and reshock Hugoniot and sound speed and various EOS models in the high-*P*/*T* regime.

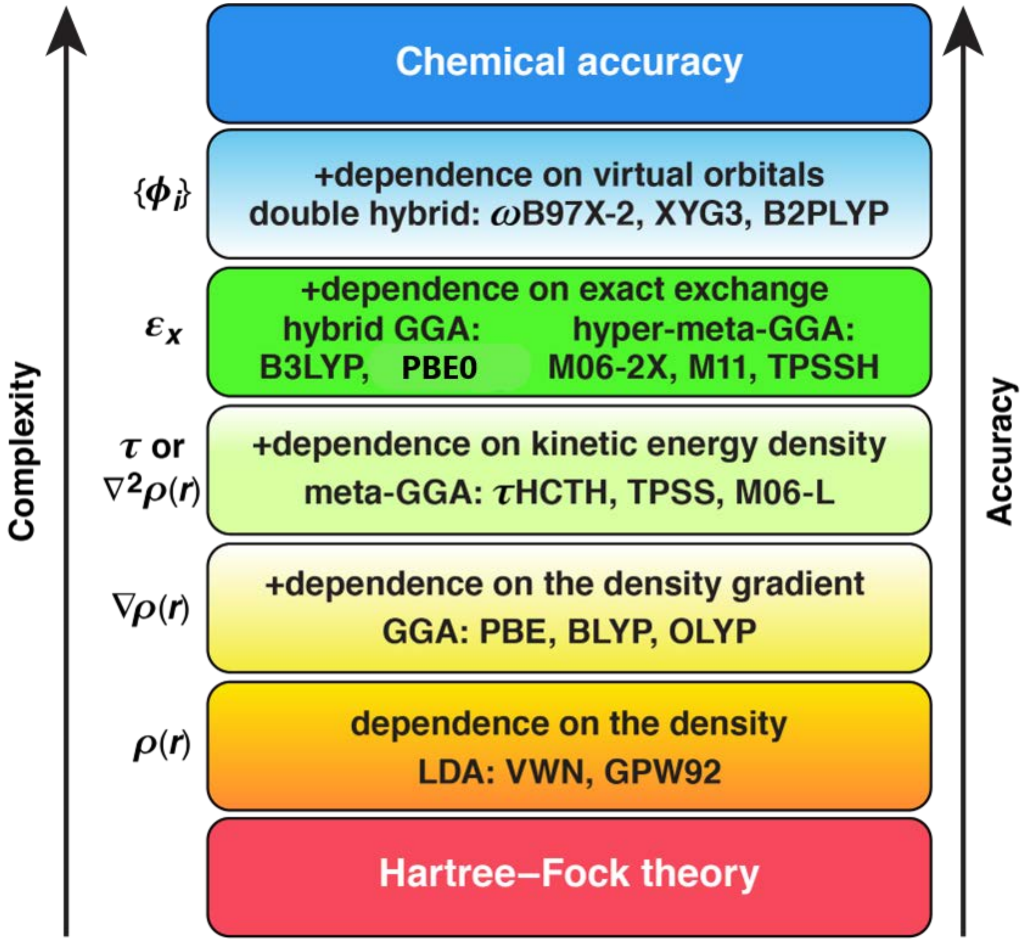
- Recently, it was shown that DFT+MD using the SCAN-L + rVV10 XC functional is remarkably successful, and much more accurate than PBE, in predicting the conditions for the dissociation of molecular to atomic fluid in dense deuterium/hydrogen*.

- Additionally, iFPEOS was motivated by the development of new finite-T version of the SCAN-L functional (T-SCAN-L) and the need to take into account nuclear quantum effects (NQEs).

* J. Hinz, V. V. Karasiev, S. X. Hu, M. Zaghoo, D. Mejía-Rodríguez, S. B. Trickey, and L. Calderín, Phys. Rev. Res. 2, 032065(R) (2020).

iFPEOS is based on a recently developed ***advanced***, ***thermal*** exchange-correlation functional T-SCAN-L, which is at the meta-GGA level of DFT.

Jacob's ladder of **zero-temperature** XC functionals:



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Finite-temperature XC density functionals at corresponding level of theory:

← **KDT0† : finite-*T* extension to PBE0**

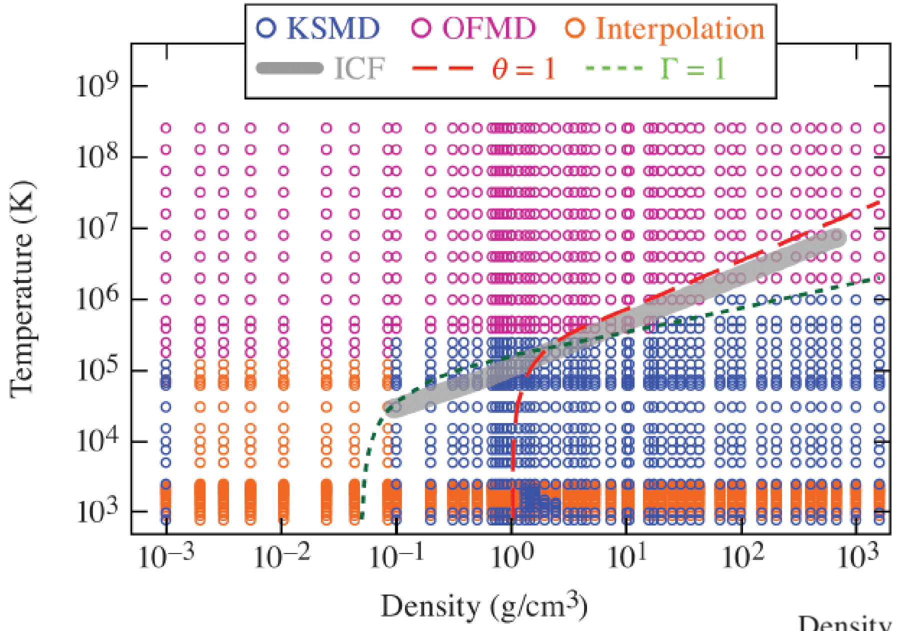
← ***T*-SCAN-L †† : finite-*T* extension to SCAN-L, the de-orbitalized version of SCAN**

← **Finite-temperature GGA (KDT16)***

← **Finite-temperature LDA (KSDT)**, (corrKSDT)***

* V. V. Karasiev, J. W. Dufty, and S. B. Trickey, Phys. Rev. Lett. 120, 076401 (2018).
** V. V. Karasiev *et al.*, Phys. Rev. Lett. 112, 076403 (2014).
† D. I. Mihaylov, V. V. Karasiev, and S. X. Hu, Physical Review B 101 (24), 245141.
†† V. V. Karasiev, D. I. Mihaylov, and S. X. Hu, Meta-gg aexchange-correlation free-energy density functional to achieve unprecedented accuracy for warm-dense-matter simulations (2021), submitted to Phys. Rev. Lett.

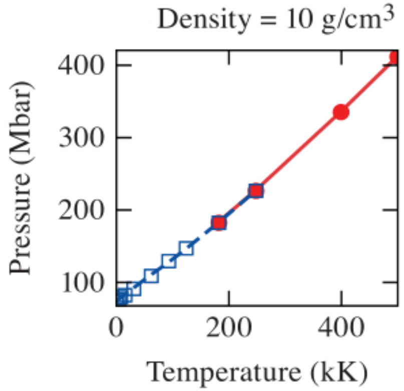
We use orbital free DFT with T-SCAN-L for XC to cover temperatures up to 256 MK

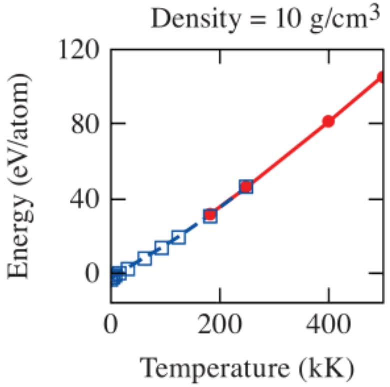


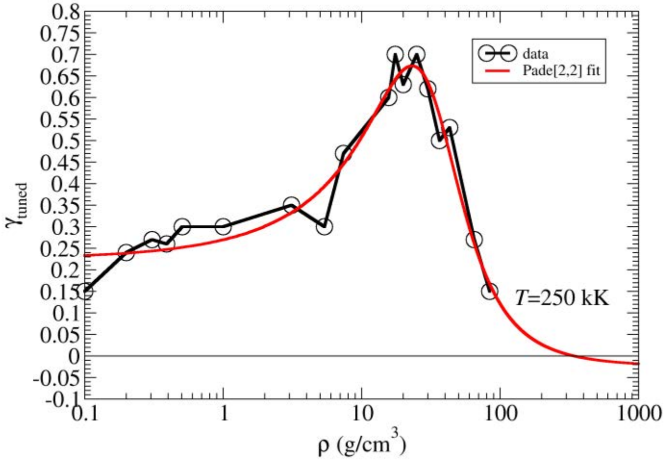
Orbital-free (OF) calculations are carried out with newly-developed, orbital-free, free-energy, non-interacting, tunable density functional LKTFγTF (unpublished).

$$F_s^{LKTF\gamma TF}[n,T] = \gamma F_s^{LKTF}[n,T] + (1-\gamma) F_s^{TF}[n,T]$$

LKTFγTF is a one-parameter convex combination of LKTF* (*Luo, Karasiev, Trickey Free-energy)* and TF (Thomas-Fermi). γ is a free parameter and is tuned based on a reference Kohn-Sham calculation.

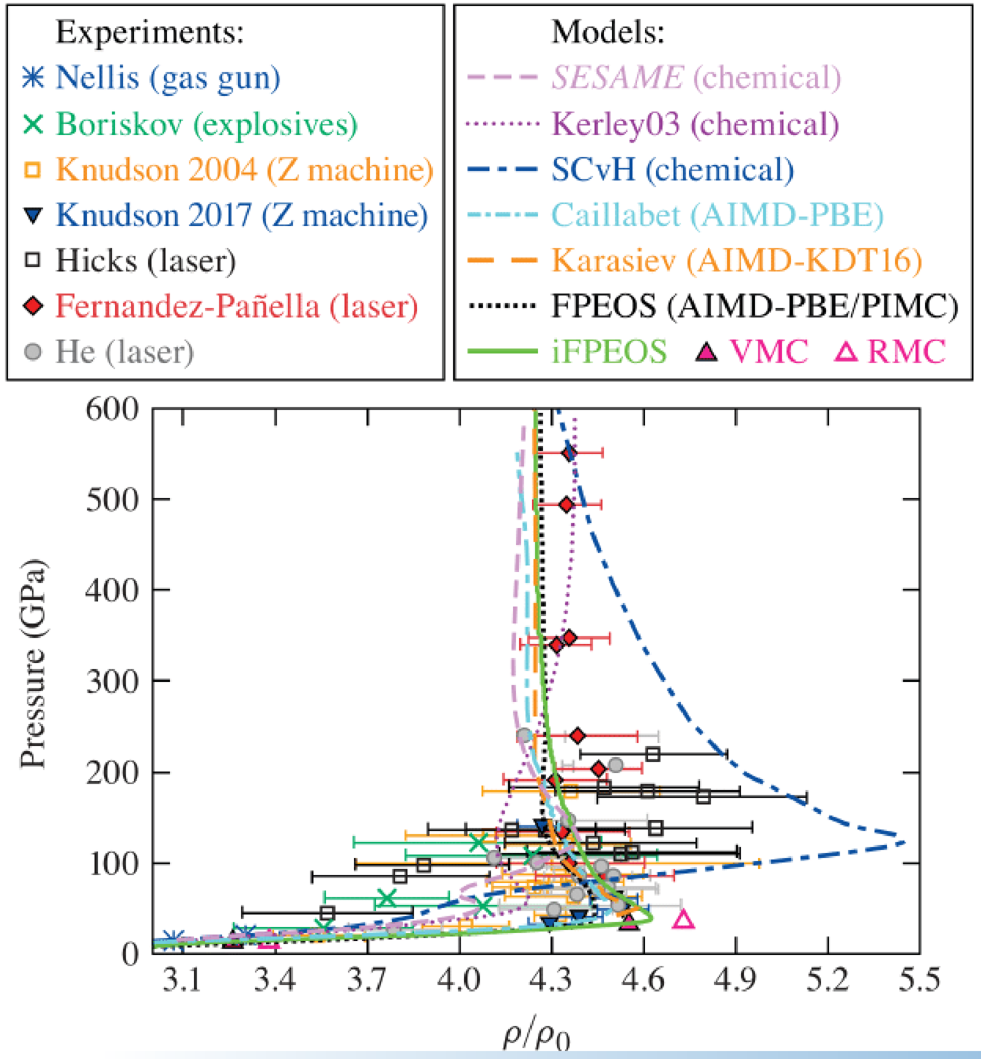




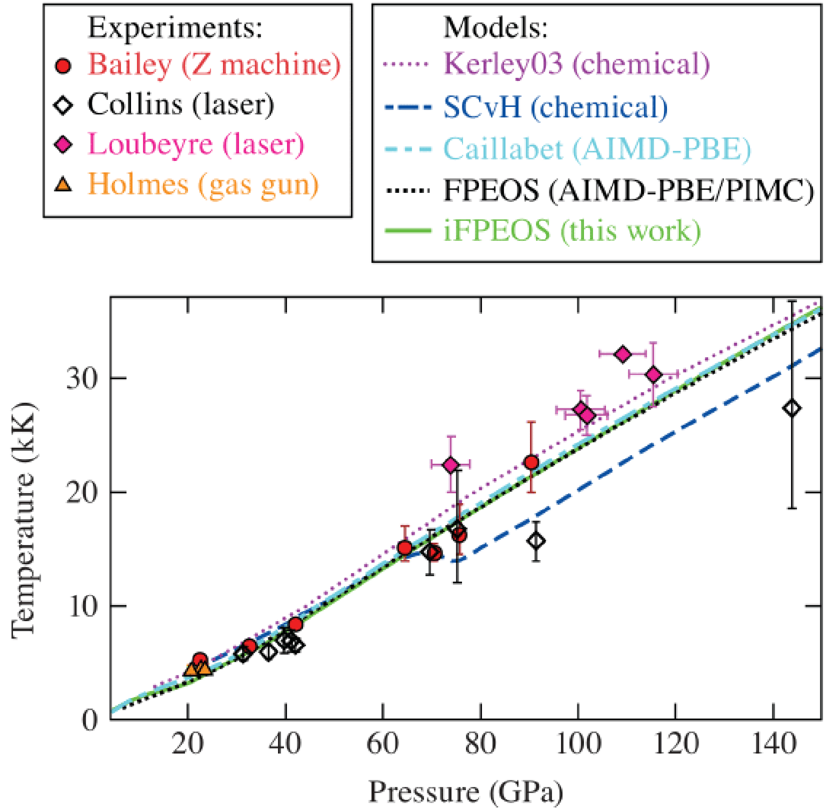


* K. Luo, V. V. Karasiev, and S. B. Trickey. "A simple generalized gradient approximation for the noninteracting kinetic energy density functional." Phys. Rev. B 98 (2018):

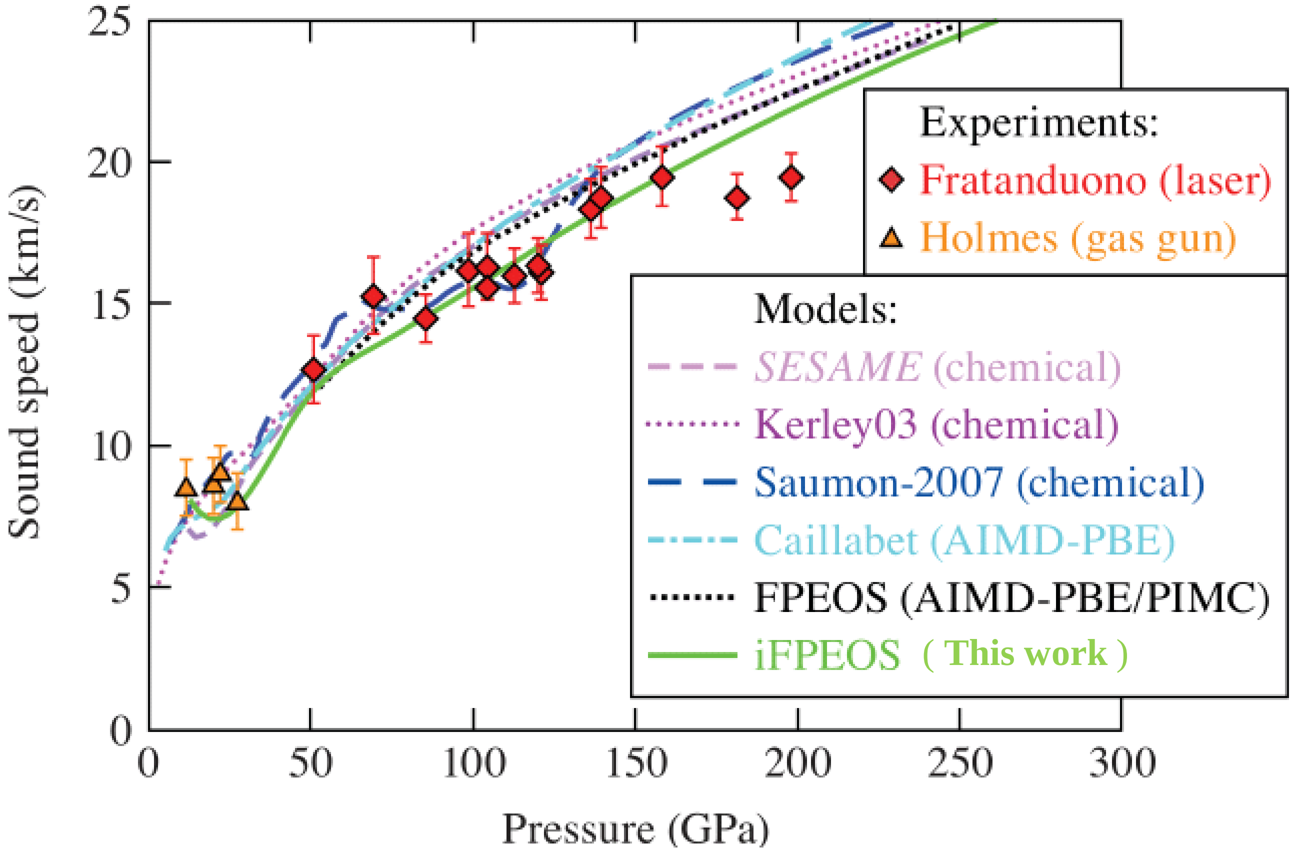
iFPEOS predicts a higher compressibility, but in the high-pressure regime, it is in agreement with PBE-based models which predict smaller compressibility compared to experiment.



$$E - E_0 = \frac{1}{2}(P + P_0)\left(\frac{1}{\rho_0} - \frac{1}{\rho}\right)$$



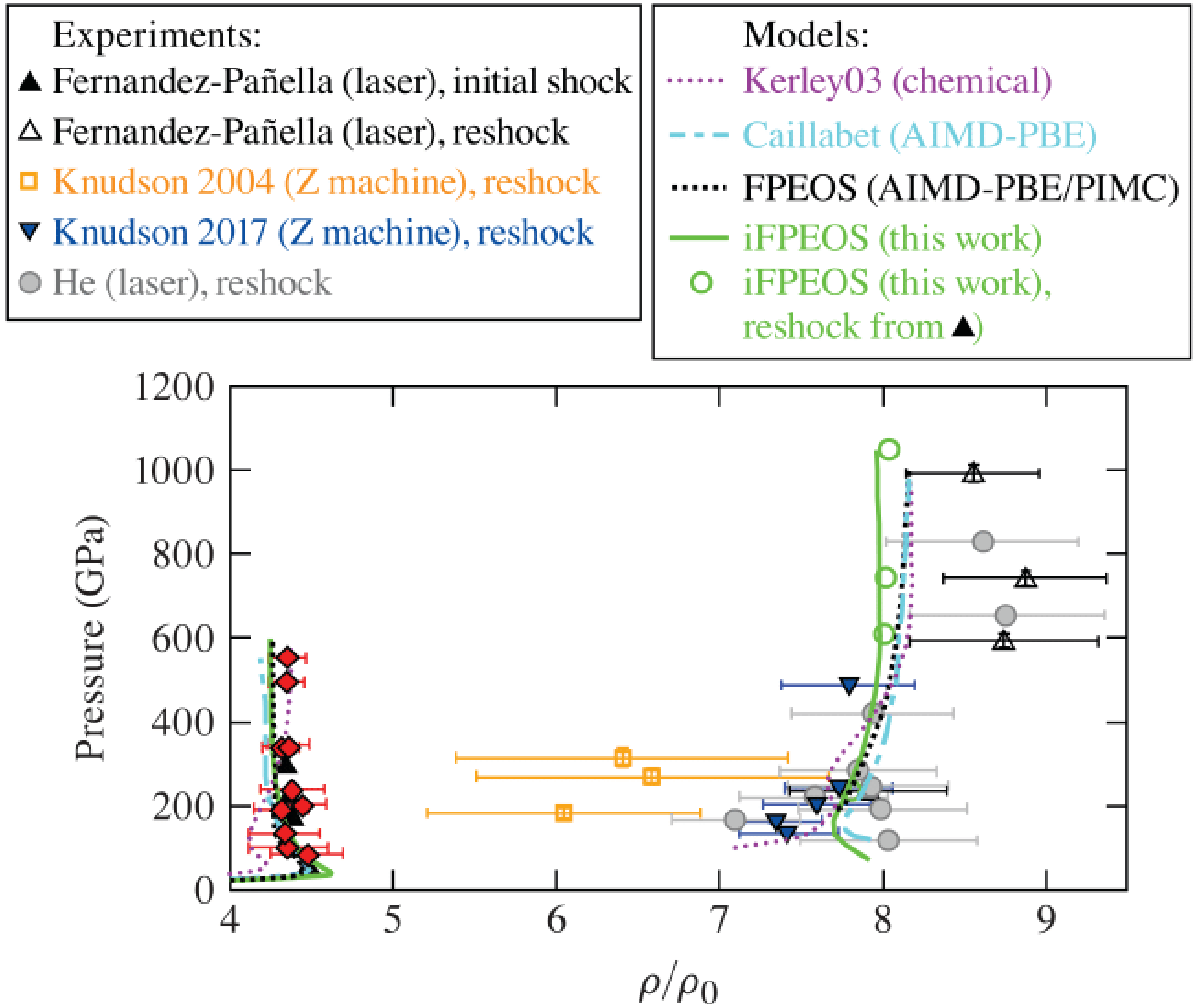
iFPEOS-predicted sound speed shows improved agreement with experiment in the 80-150 GPa pressure regime – corresponding to 15 – 40 kK, where XC thermal effects are most important.



Eulerian sound speed:

$$c = \sqrt{\left(\frac{\partial p}{\partial \rho}\right)_s}.$$

In the low-pressure regime iFPEOS agrees with other models and latest experimental measurements of double-shocked deuterium, but in the high-pressure regime iFPEOS predicts a significantly softer reshock Hugoniot.



$$E - E_0 = \frac{1}{2}(P + P_0)\left(\frac{1}{\rho_0} - \frac{1}{\rho}\right)$$

$$\rho = \frac{\rho_0(u_s - u_{p_0})}{(u_s - u_p)}$$

$$P = P_0 + \rho_0(u_s - u_{p_0})(u_p - u_p)$$

- **Reshock states (green circles) were determined by Impedance Matching (IM) with α-quartz*.**

- **iFPEOS reshock Hugoniot (green curve) is launched off of iFPEOS principal Hugoniot.**

* Supplemental Material - A. Fernandez-Panella, M. Millot, D. Fratanduono, M. Desjarlais, S. Hamel, M. Marshall, D. Erskine, P. Sterne, S. Haan, T. Boehly, et al., Physical review letters 122, 255702 (2019).

Summary

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