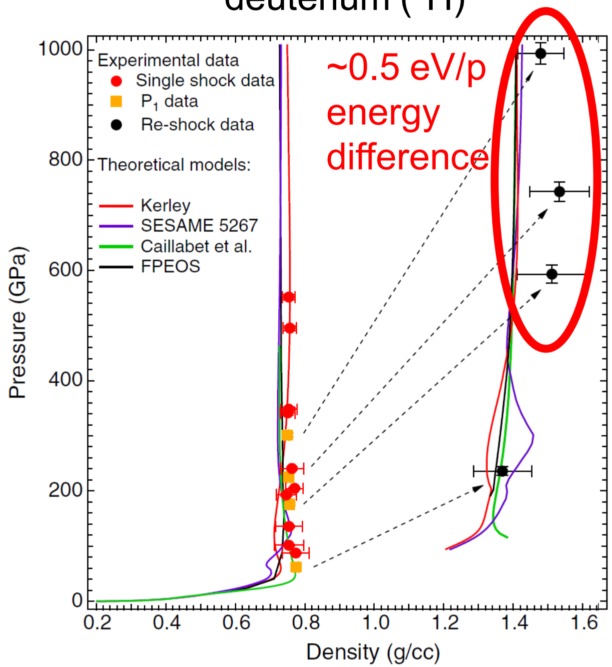
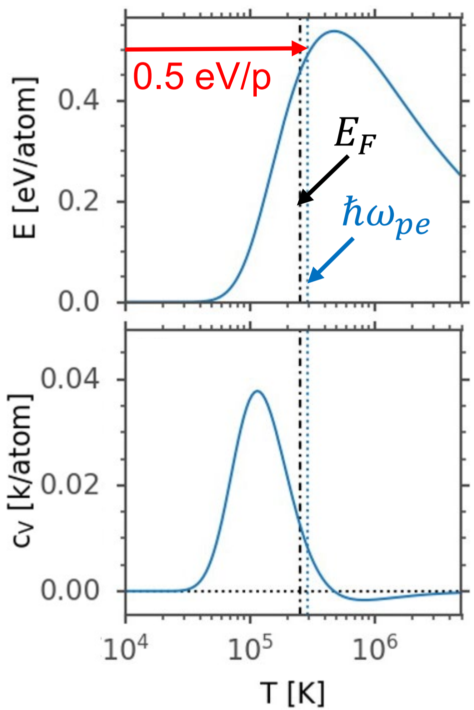
The Compressibility of Shocked Hydrogen to 1 TPa

Double-shock density
deuterium (^{2}H)



Fernandez-Pañella *et al* (2019)

Internal energy of plasma waves
deuterium at 1.5 g/cc



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2022-10-17

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Plasma wave energy influences shock compression in warm dense hydrogen near 1 TPa

- Recent shock, re-shock, and sound speed experiments[1,2] report higher compression of deuterium (^{2}H) than predicted by models at pressures between 0.4 and 1.0 TPa.

- The internal energy of electron plasma waves, not explicitly included in theoretical equation of state models, is sufficient to account for the missing compression energy in shocked hydrogen.

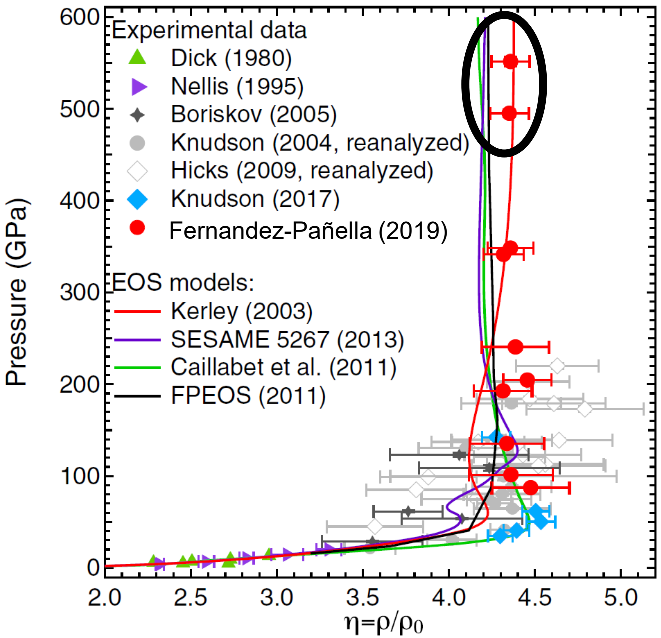
- The observed slowdown in sound speed near 0.2 TPa[2] matches the ion acoustic wave phase velocity; we conjecture the propagation of acoustic perturbations are impeded by charge buildup at an IAW wavefront

- Recent NIF experiments obtained D_2 shock data up to 1.2 and 2.6 TPa for single and double shocks, respectively. Compression analysis for these data is underway.

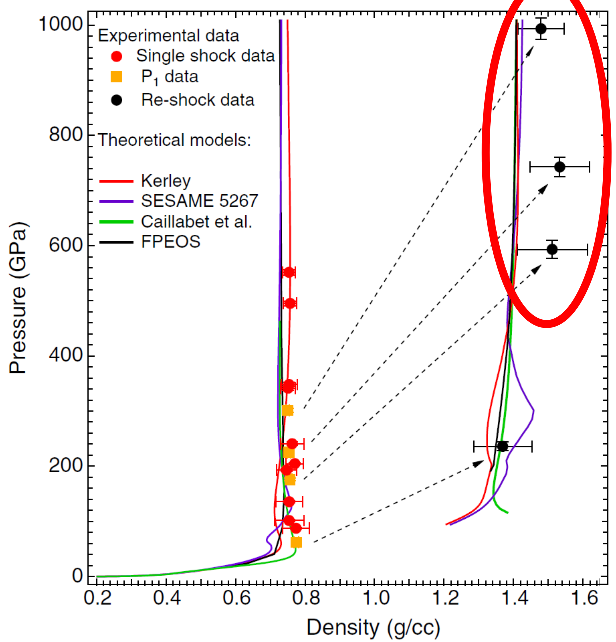
1. A. Fernandez-Pañella et al, Phys. Rev. Lett. 122, 255702 (2019).
2. D. E. Fratanduono et al, Phys. Plasmas 26, 012710 (2019).

Recent experimental data suggest model discrepancies in the high-pressure deuterium equation of state

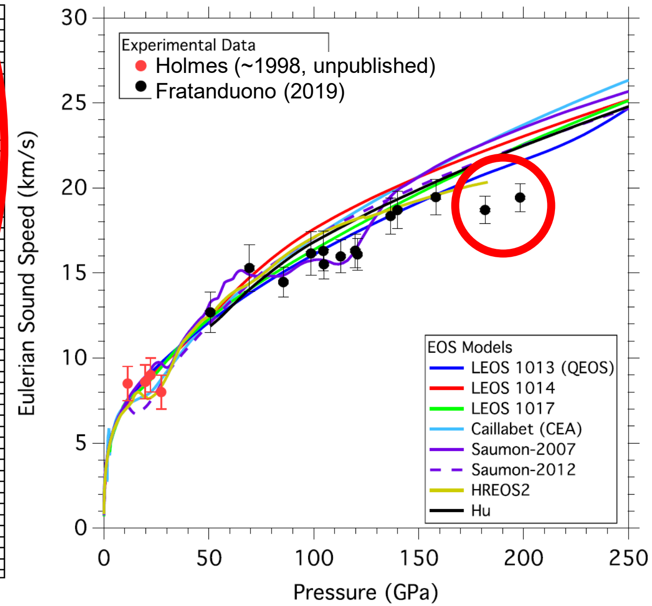
D$_2$ shock [1]



D$_2$ reshock [1]



D$_2$ shock sound speed [2]

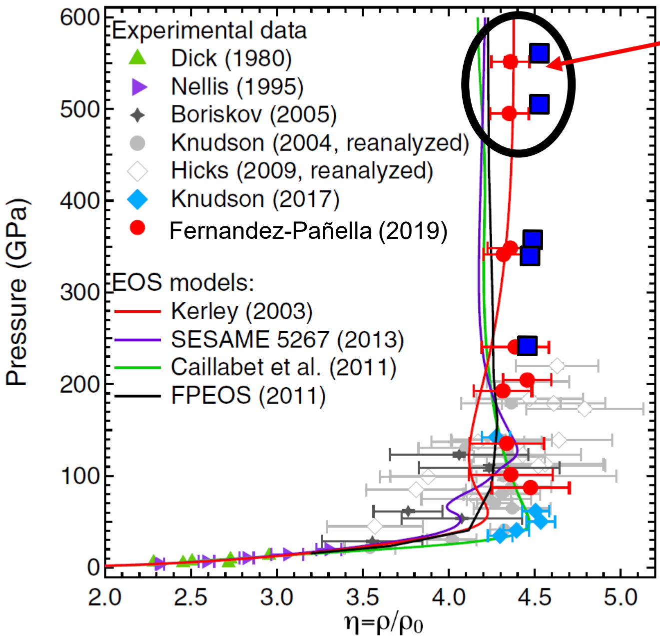


This is a systematic discrepancy consistent across 3 different experimental techniques: (1) shock, (2) reshock, (3) sound speed. Each technique has different systematic uncertainties.

1. A. Fernandez-Pañella et al, Phys. Rev. Lett. 122, 255702 (2019).
2. D. E. Fratanduono et al, Phys. Plasmas 26, 012710 (2019).

The shock compression difference can be explained by an additional internal energy of about 0.5 eV/atom

D_2 shock [1]



~5% (or 8%*) difference in the shock compression.

From the Rankine-Hugoniot energy relation:

$$E_1 - E_0 = \frac{1}{2}(P_1 + P_0)\left(\frac{1}{\rho_0} - \frac{1}{\rho_1}\right)$$

Rearrange terms (and define E_0=0):

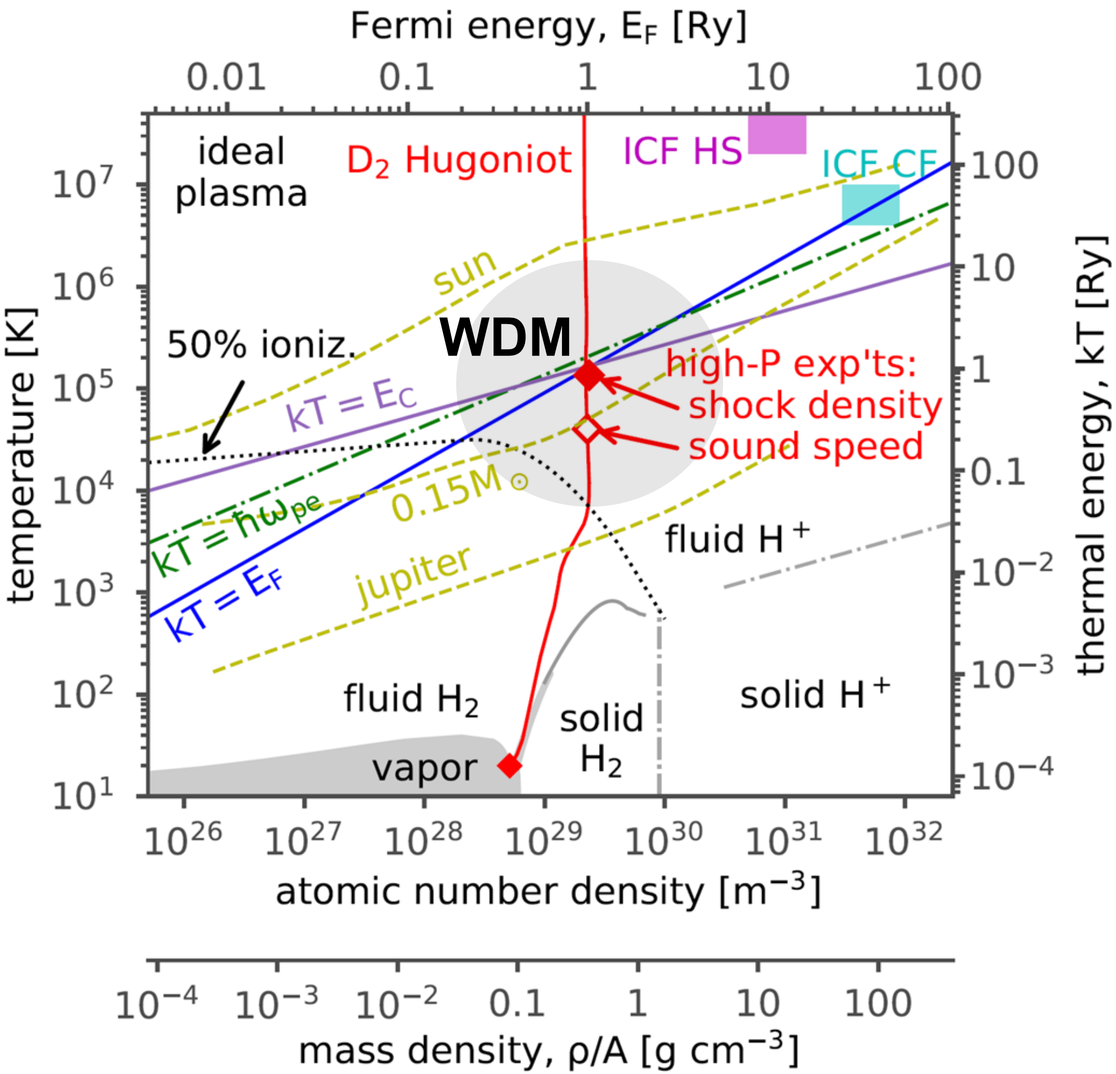
$$E_1 = \frac{P_1 + P_0}{2\rho_0}\left(1 - \frac{\rho_0}{\rho_1}\right)$$

The different final densities between theory and experiment gives a difference of order **0.5 eV/atom** in the final internal energy.

* 8% based on re-analysis using new measurements [2] of the shock standard.

1. A. Fernandez-Pañella et al, Phys. Rev. Lett. 122, 255702 (2019).
2. M. C. Marshall et al, Phys. Rev. B 99, 174101 (2019).

Warm dense matter occurs is at confluence of energy scales



Atomic Energy (Rydberg):

$$E_R = 13.6\ \text{eV} = 1\ \text{Ry}$$

Thermal Energy:

$$E_T = k_B T$$

Fermi Energy:

$$E_F = (3\pi^2)^{2/3}\frac{\hbar^2}{2m_e}n_e^{2/3}$$

Coulomb Potential Energy:

$$E_C = \frac{e^2}{4\pi\varepsilon_0}\left(\frac{4}{3}\pi n_e\right)^{1/3}$$

Plasmon energy:

$$\hbar\omega_{pe} = \hbar\left(\frac{n_e e^2}{\varepsilon_0 m_e}\right)^{1/2}$$

*many contours assume full ionization

Plasma wave excitations offer an energy reservoir

Electron-plasma waves (EPW). In this high-frequency oscillation mode, ions are a “fixed” neutralizing fluid, and the electron density fluctuates near the e⁻ plasma frequency.

$$\omega_{epw}^2 = \omega_{pe}^2(1 + 3q^2\lambda_{De}^2)$$

$\omega_{pe} = \sqrt{\frac{n_e e^2}{\epsilon_0 m_e}}$: electron plasma frequency

$\frac{1}{\lambda_{De}^2} = \frac{e^2 n_e}{\epsilon_0 k_B T}$: e⁻ Debye length

Ion-acoustic waves (IAW). At lower frequencies, the ions can also respond, and the ions and electrons oscillate in phase, with adjustments due to plasma screening.

$$\omega_{iaw}^2 = \omega_{pi}^2 \frac{q^2\lambda_{Di}^2}{1 + q^2\lambda_{Di}^2}$$

$\omega_{pi} = \omega_{pe}\sqrt{\frac{m_e}{m_i}}$: ion plasma frequency

$\frac{1}{\lambda_{Di}^2} = \frac{e^2}{\epsilon_0 k_B T}\left(n_e + \sum_i Z_i^2 n_i\right)$: Debye length

Plasma oscillations are not explicitly included in the theoretical models

Plasma wave excitations offer an energy reservoir that may play a contributing role in warm dense hydrogen

Plasma wave dispersion relations have similar form for degenerate electrons[1,2]

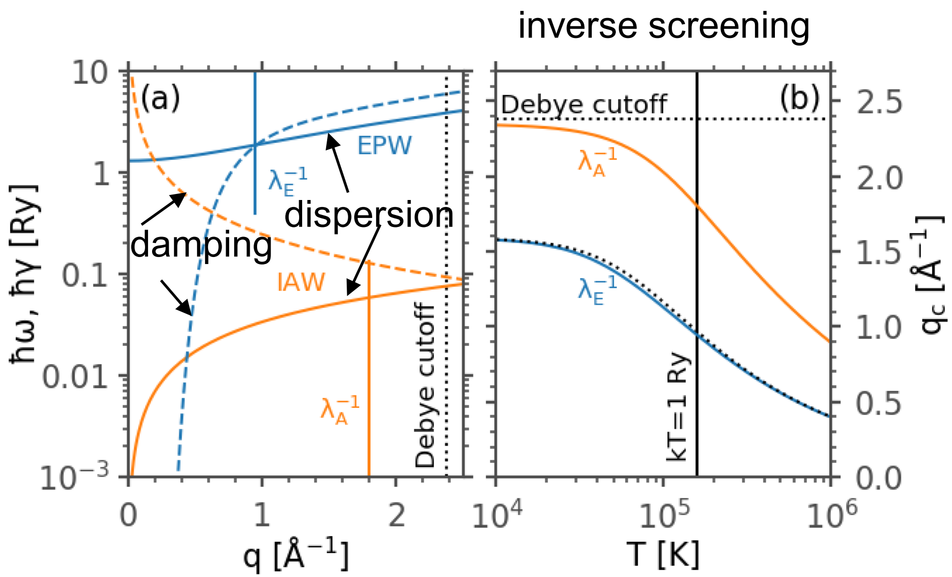
nondegenerate

$$\frac{\omega_{epw}^2}{\omega_{pe}^2} = 1 + 3k^2\lambda_{De}^2 \qquad \frac{\omega_{iaw}^2}{\omega_{pi}^2} = \frac{k^2\lambda_{Di}^2}{1+k^2\lambda_{Di}^2}$$

degenerate

$$\frac{\omega_{epw}^2}{\omega_{pe}^2} = 1 + k^2\lambda_{Q,e}^2 \qquad \frac{\omega_{iaw}^2}{\omega_{pi}^2} = \frac{k^2\lambda_{Q,i}^2}{1+k^2\lambda_{Q,i}^2}$$

Solutions for arbitrary degeneracy have been published previously[1,2]

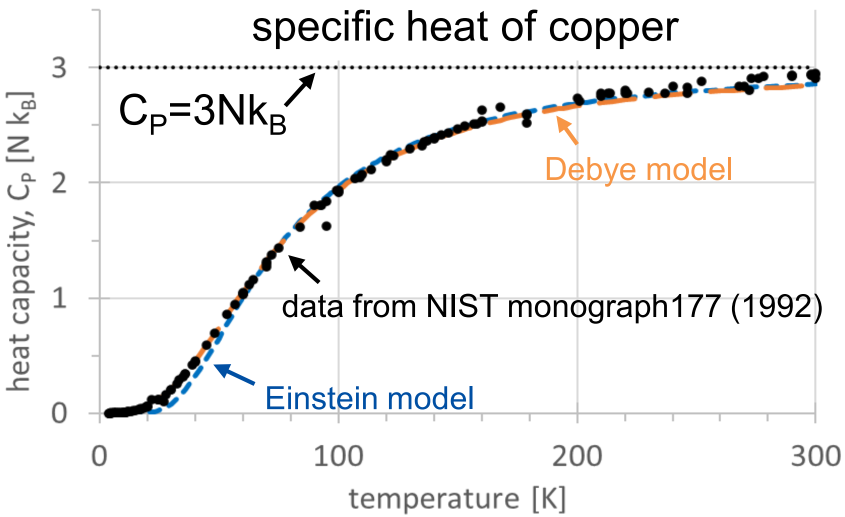


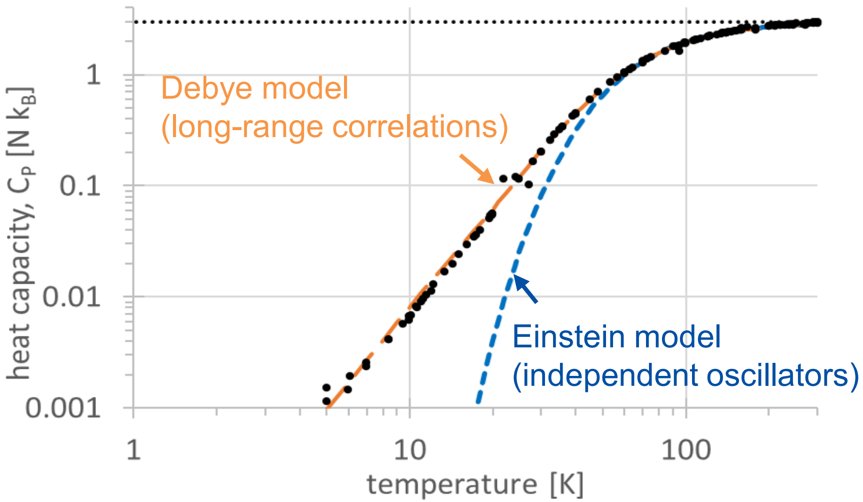
for E_F=1 Ry (n_e=2×10^{29} m^{-3})

1. D. Pines and J. R. Schrieffer, Phys Rev 125, 804 (1962).
2. D. Melrose and A. Mushtaq, Phys. Rev. E 82, 056402 (2010).
…and many others

Ion acoustic waves (IAW) are critically damped (damping rate faster than the oscillation frequency) by the velocity match to the ion distribution function, so we will focus on the electron plasma waves (EPW)

Review: Long-range correlations in solids are well-represented using the Debye model for phonons.





- Classical solid[1]: N particles in 3D (with 1 kinetic and 1 potential energy degree of freedom per D):

$$\frac{C_{\text{Dulong-Petit}}}{Nk_B} = 3$$

- Einstein model[2]: particles are quantum mechanical oscillators with characteristic temperature $T_E = \theta_E T$ and Bose-Einstein statistics:

$$\frac{C_{\text{Einstein}}}{Nk_B} = 3\theta_E^2 \frac{e^{\theta_E}}{(e^{\theta_E} - 1)^2}$$

- Debye model[3]: integrate long-range correlations in the lattice (i.e., *phonons*) up to a cutoff $T_D = \theta_D T$ related to sound speed and density.

$$\frac{C_{\text{Debye}}}{Nk_B} = 3\theta_D^{-3} \int_0^{\theta_D} \frac{x^4 e^x}{(e^x - 1)^2} dx$$

1. A. Petit and P. Dulong, Ann. Chem. Phys. 10, 395 (1819).
2. A. Einstein, Ann. Physik 22, 180 (1907).
3. P. Debye, Ann. Physik 39, 789 (1912).

A Debye-type model for plasma oscillations: setup

The Debye model for phonon internal energy is:

$$E = \int_0^{q_c} \epsilon(q) g(q) n_{BE}(q)\, dq$$

$\epsilon(q) = \hbar\omega(q)$: dispersion relation
$g(q)$: density of states
$n_{BE}(q)$: occupancy from Bose-Einstein distribution
q_c: cutoff wavenumber (highest supported wavenumber)

Dispersion relation

$$\epsilon(q) = \hbar\omega(q)$$

Modify the dispersion relation to that of plasma oscillations

Density of states

$$g(q) = \frac{Vq^2}{2\pi^2}$$

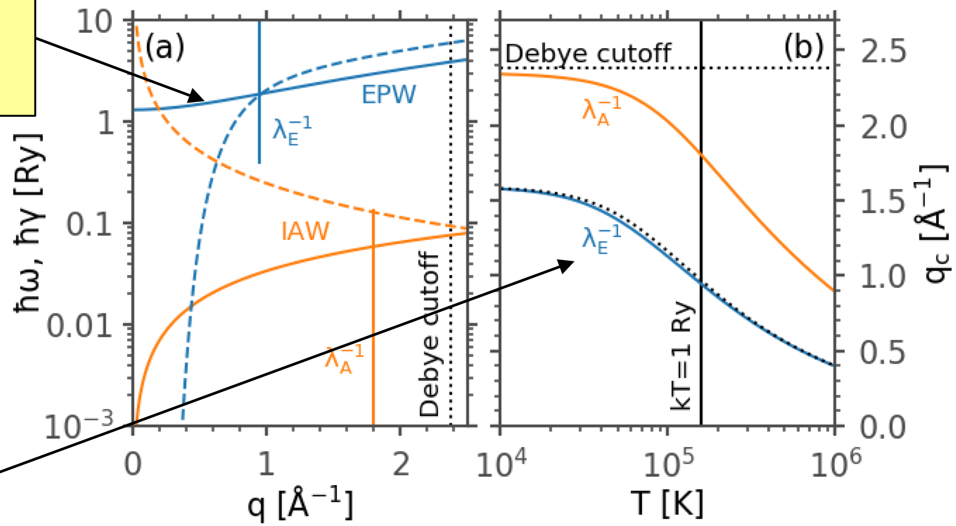
Bose-Einstein distribution function

$$n_{BE}(q) = \frac{1}{e^{\epsilon(q)/k_B T} - 1}$$

Cutoff wavenumber

$$q_c \propto \frac{1}{\lambda_{screen}}$$

Modify the cutoff wavenumber for screening



Putting it all together

$$E = \frac{V}{2\pi^2} \int_0^{q_c} \frac{\hbar\omega(q) q^2 dq}{e^{\hbar\omega(q)/k_B T} - 1} \qquad C_V = \left(\frac{\partial E}{\partial T}\right)_V$$

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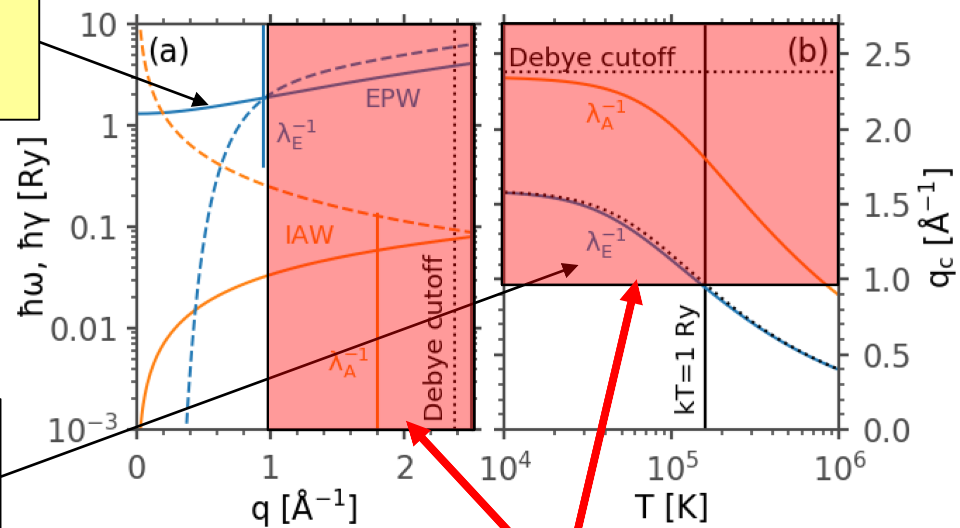


accessible wavenumbers for *ab initio* simulations with 64 atoms

A Debye-type model for plasma oscillations: results

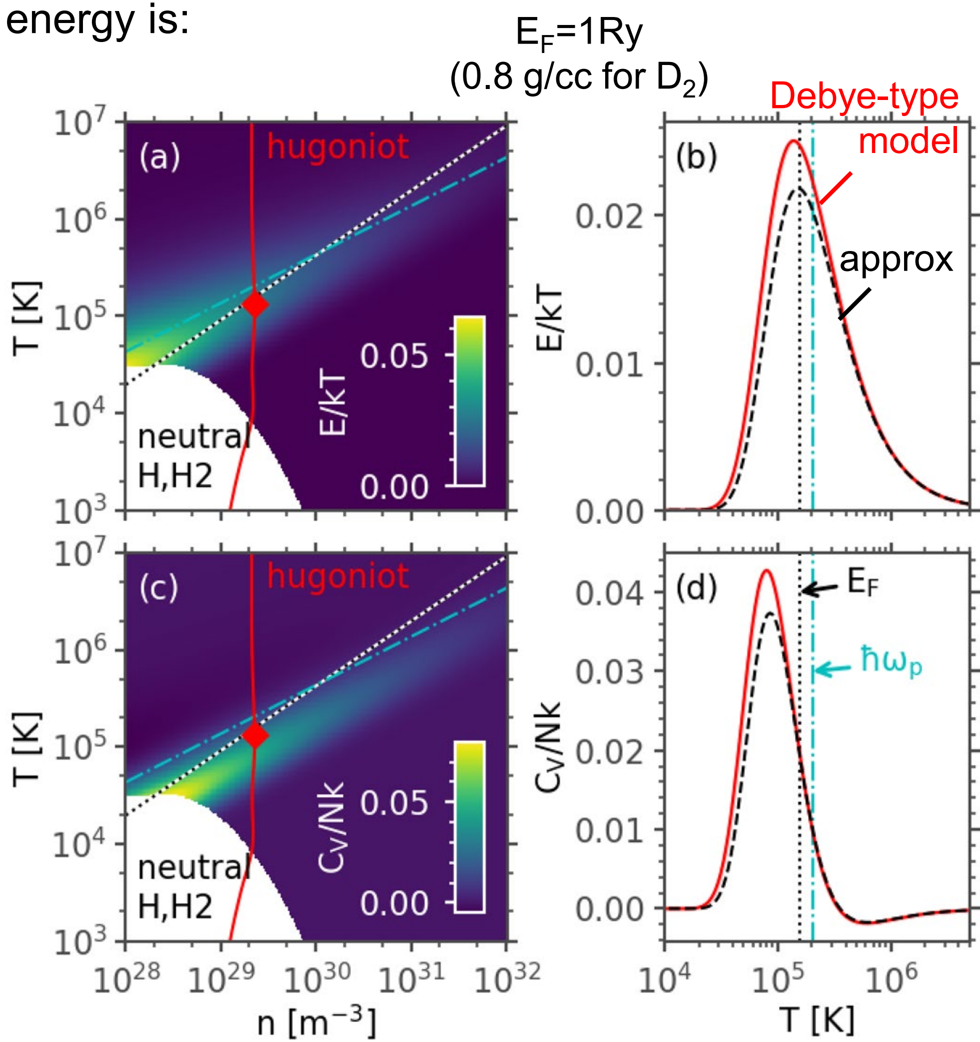
The Debye-type model for plasmon internal energy is:

$$E = \frac{V}{2\pi^2}\int_0^{1/\lambda_E} \frac{\hbar\omega_q q^2 dq}{e^{\hbar\omega_q/k_BT} - 1}$$

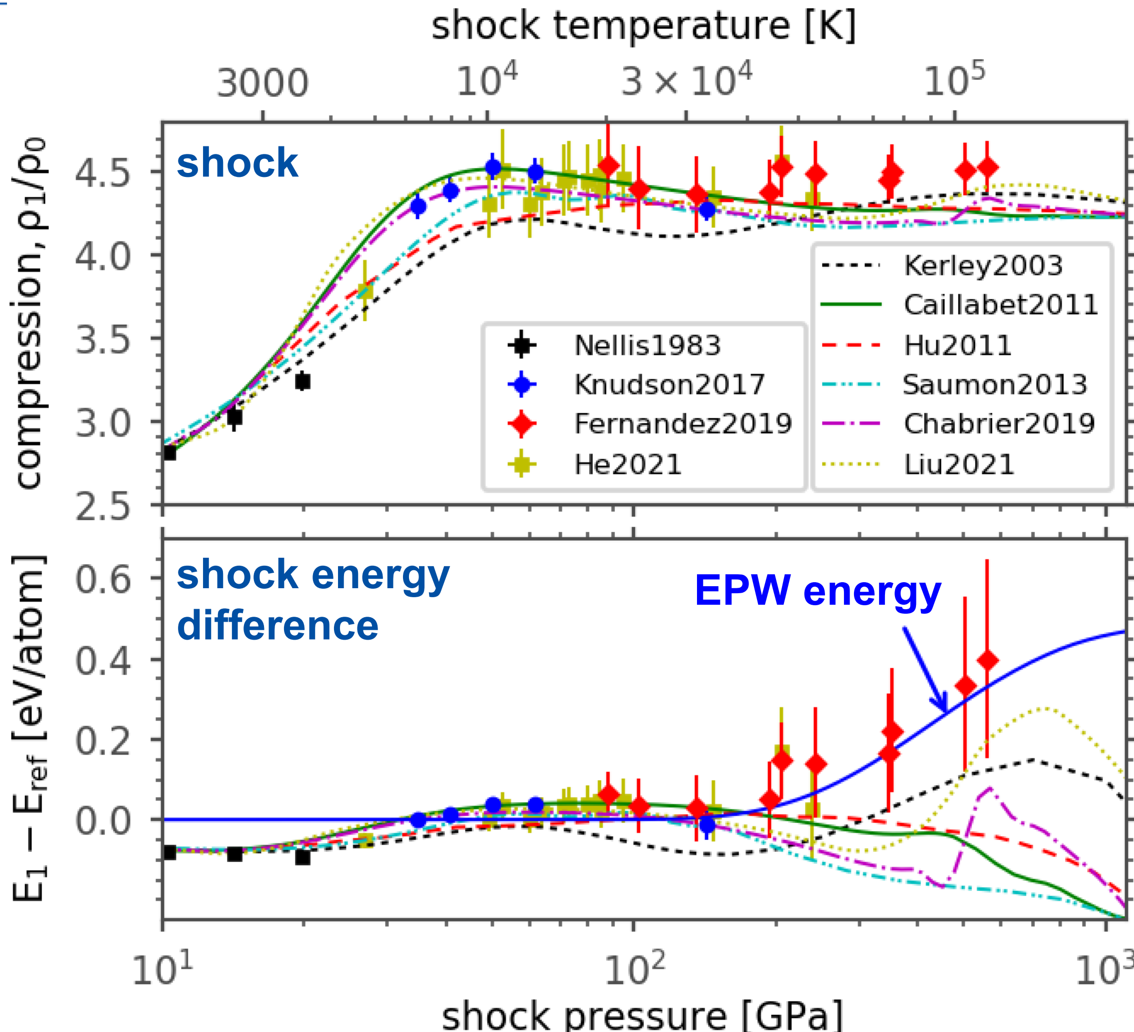
$$C_V = \left(\frac{\partial E}{\partial T}\right)_V$$

A non-integral approximation is obtained by assuming the oscillation energy is constant with the value at the cutoff length:

$$\frac{E}{NkT} \approx \frac{V}{6\pi^2\lambda_E^3}\frac{\sqrt{2}\hbar\omega_{pe}}{e^{\sqrt{2}\hbar\omega_{pe}/k_BT} - 1}$$



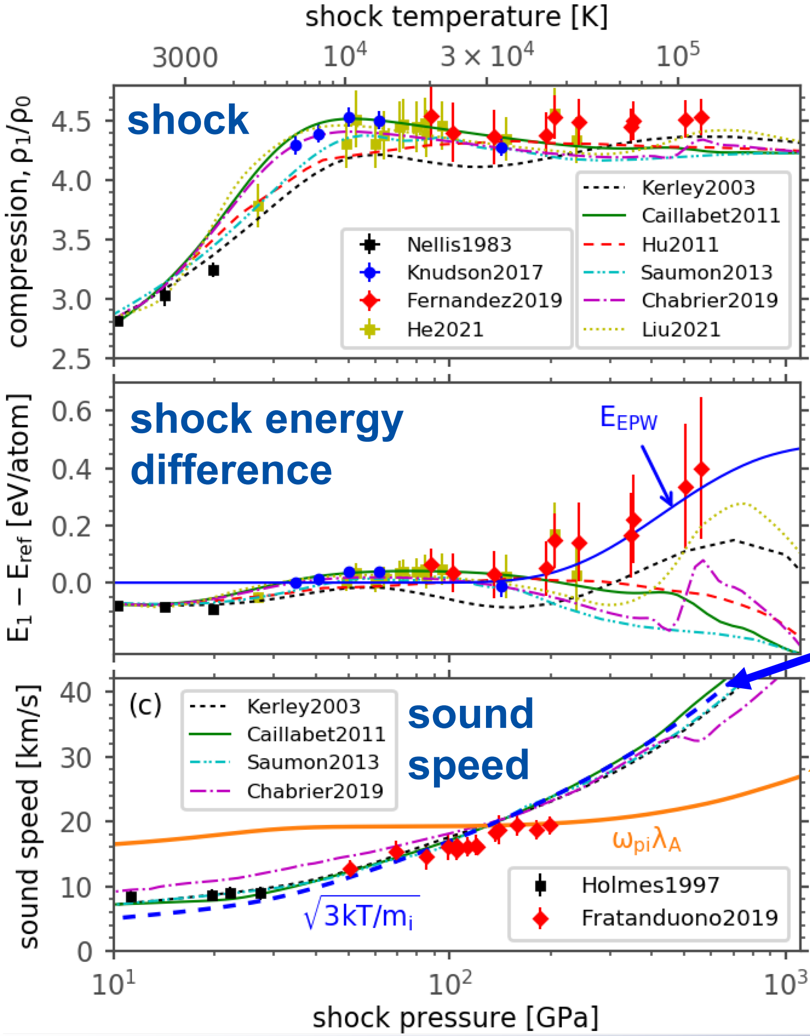
Electron plasma wave energy tracks difference between experiments and models above 200 GPa



4.3x “reference” compression

ΔE vs 4.3x reference compression

Low observed sound speed near 200 GPa is coincident with velocity match to ion acoustic wave (IAW)



Simple $(3T/m)^{1/2}$ dependence of sound speed matches theoretical models at high pressure

Observed slowdown at ~200 GPa matches IAW phase velocity

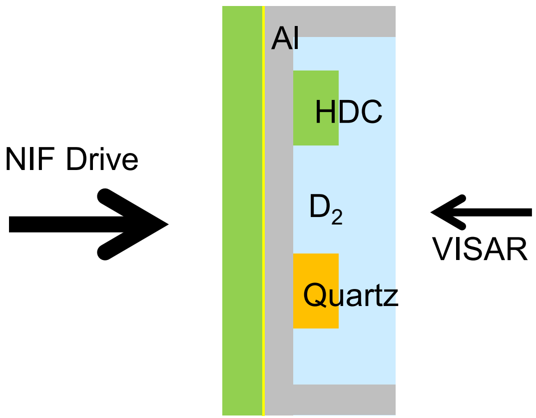
Conjecture: charge density fluctuations restrict speed of pressure perturbations when the velocities are closely matched.

What next?

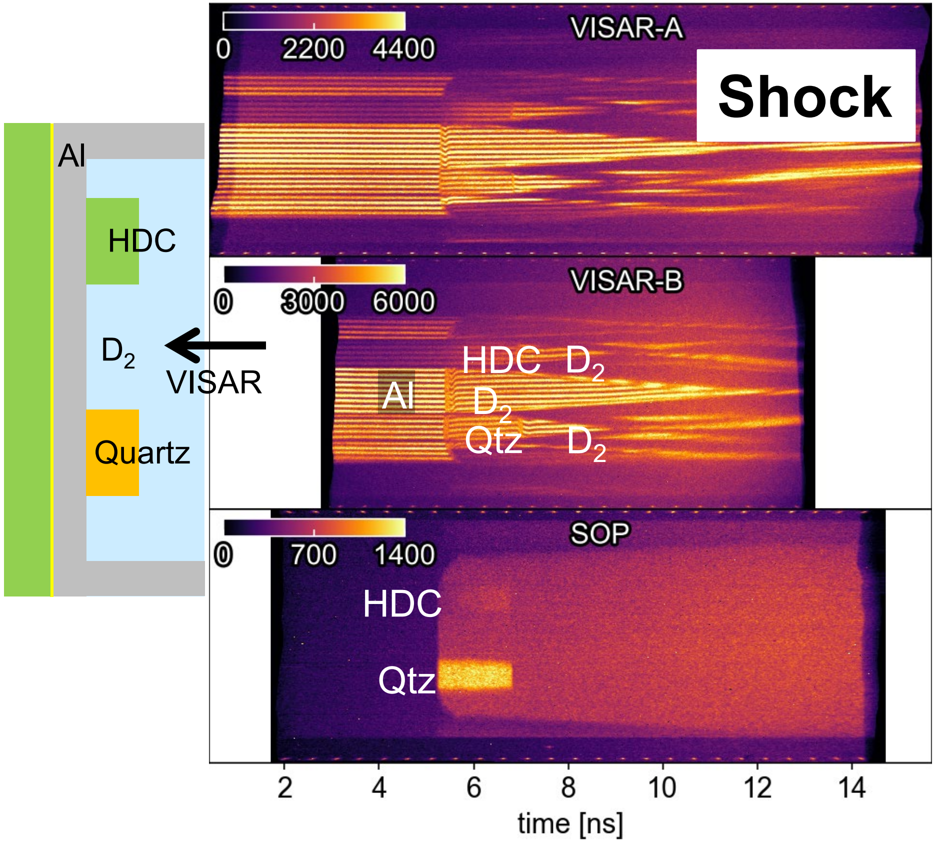
1. OMEGA: New D_2 double shock experiments (Z. Sprowal) will reach same conditions as Fernandez-Panella re-shock by different compression path; experimental configuration will enable temperature and reflectance measurements

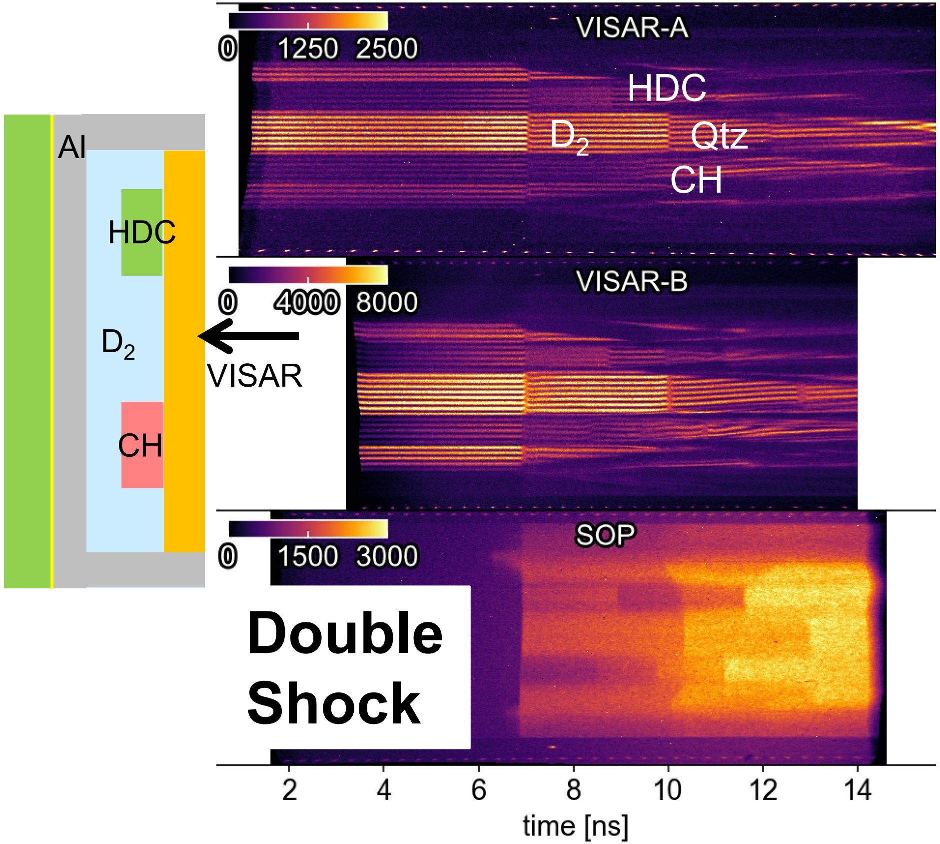
See talk by Z. Sprowal, YO05.00007

2. NIF: D_2 shock and re-shock measurements to pressures above 1 TPa to investigate if compression continues to exceed models at higher temperatures. Experimental configuration uses multiple impedance standards to check systematics of release.



D_2 shock and double shock data up to 1.2 and 2.6 TPa were obtained at the NIF; analysis is underway





summary

The plasma wave energy can account for shock compression discrepancies in warm dense hydrogen

- Recent shock, re-shock, and sound speed experiments[1,2] report higher compression of deuterium (^{2}H) at between 0.4 and 1.0 TPa than predicted by models.

- The internal energy of electron plasma waves, not explicitly included in theoretical equation of state models, is sufficient to account for the missing compression energy in shocked hydrogen.

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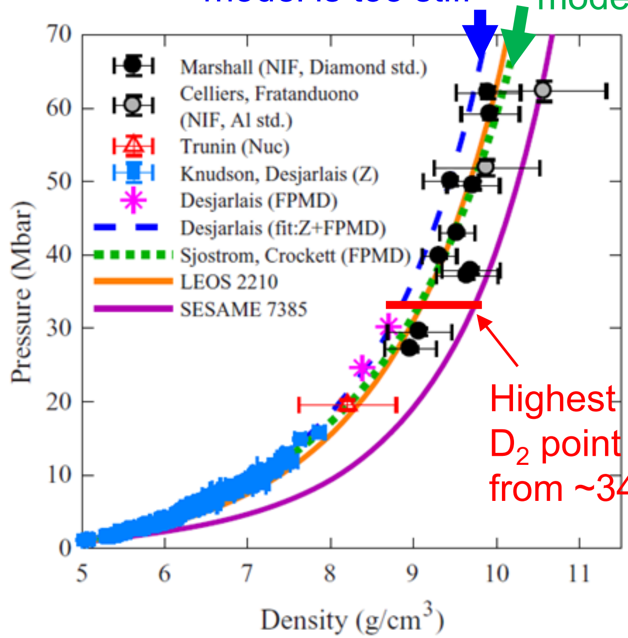
Extras

Compression discrepancy is even larger than reported! M. Marshall quartz data is softer than Desjarlais extrap.

M. Marshall et al, PRB 2019
Quartz Hugoniot measurements

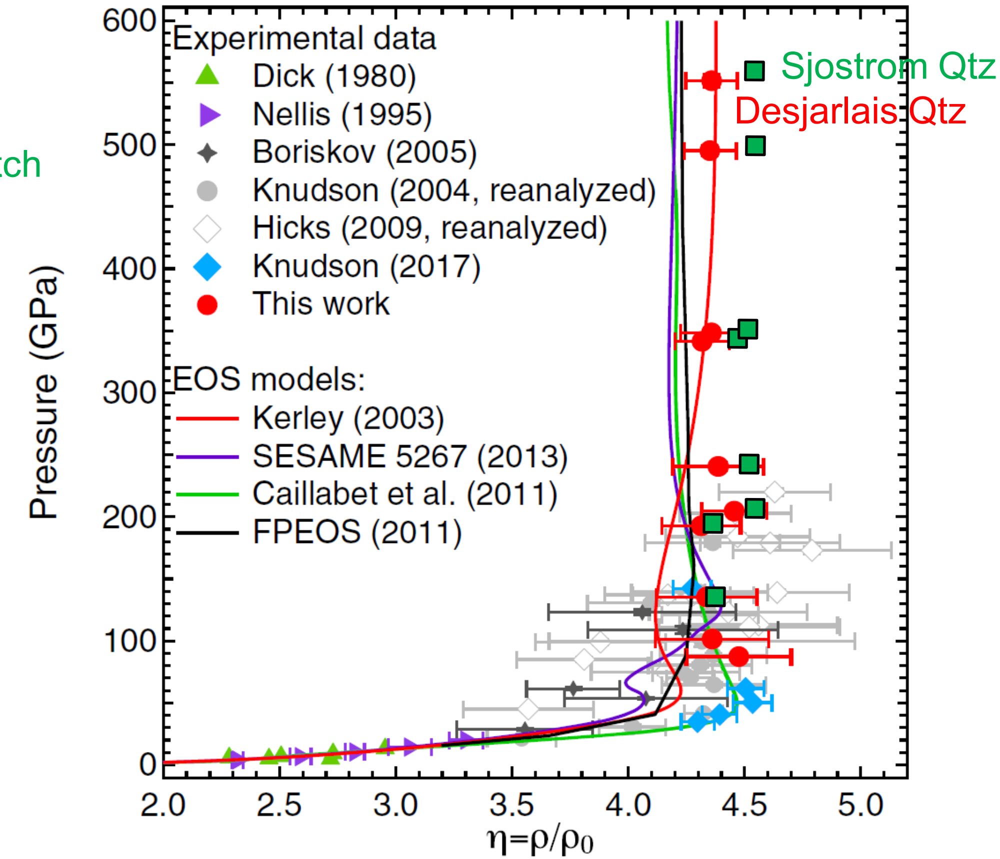
Desjarlais quartz model is too stiff

Sjostrom quartz model is better match

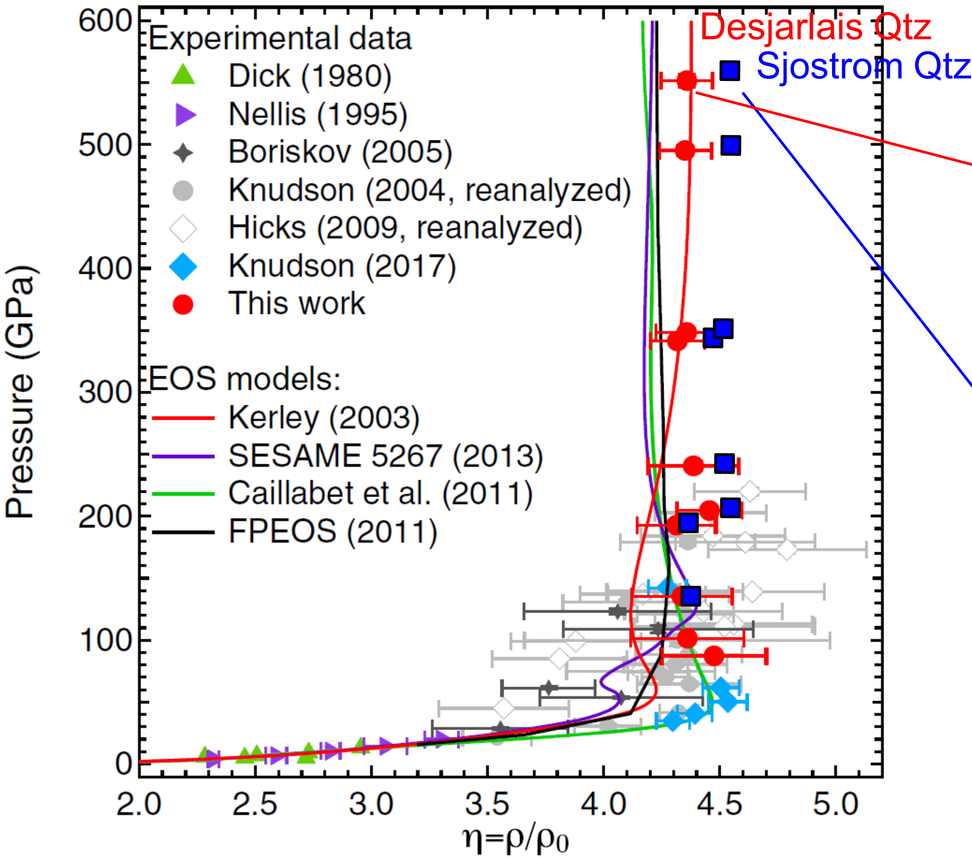


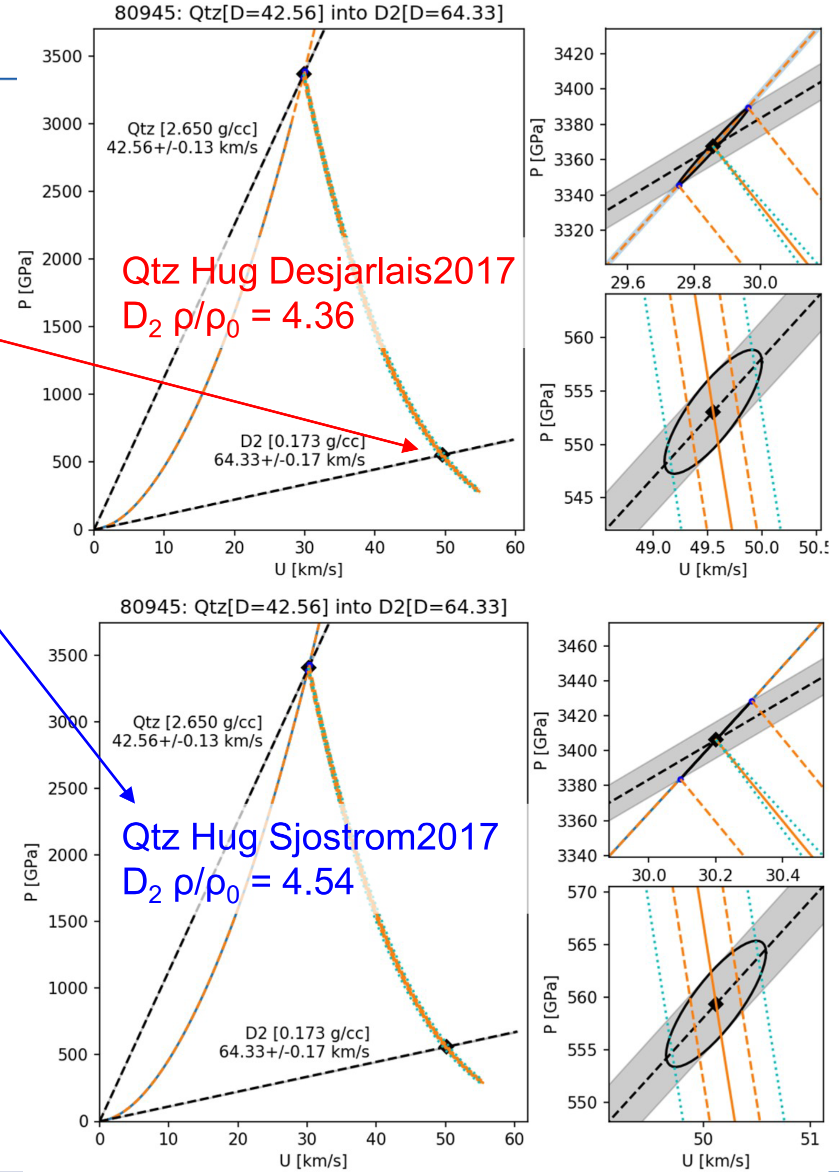
Highest Fernandez D_2 point releases from ~34 Mbar Qtz

Fernandez-Pañella 2019, with select points re-analyzed using Sjostrom Qtz Hugoniot



D_2 Hugoniot w/ Sjostrom Qtz





Fernandez-Panella 2019, select points re-analyzed with Sjostrom Hugoniot

Some additional considerations

There is, though, considerable uncertainty in the specifics, for example:

1. Choice of cutoff wavenumber, and whether to use a sharp cutoff
2. Density of states near the cutoff wavenumber probably deviates from the simple Debye DOS
3. Should the screening wavelength in the dispersion relation be modified from the cutoff wavelength?
4. Scattering/damping: the imaginary component of the dispersion relation was neglected in this analysis. How does the imaginary component affect the equilibrium population distribution? i.e. how does individual plasmon lifetime relate to the energy content?
5. At lower temperatures, the ionization model feeds directly into the plasmon energy content
6. Is the effective mass of the electrons equal to the rest mass?
7. How do “holes” in a semiconductor model or Fermi sea contribute?