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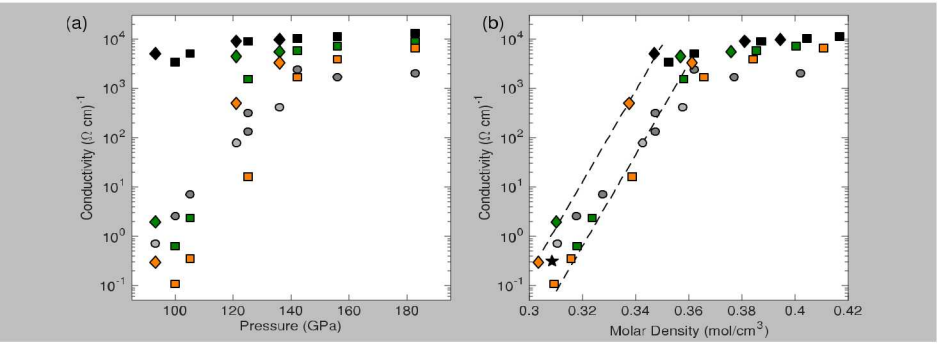

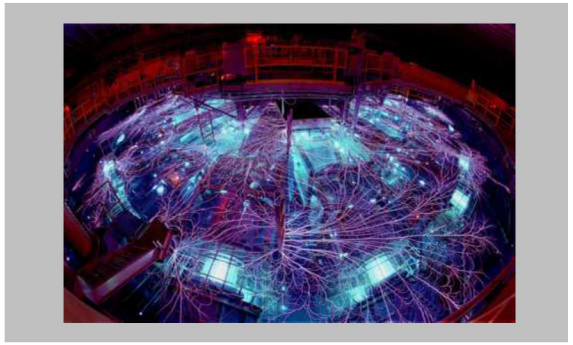

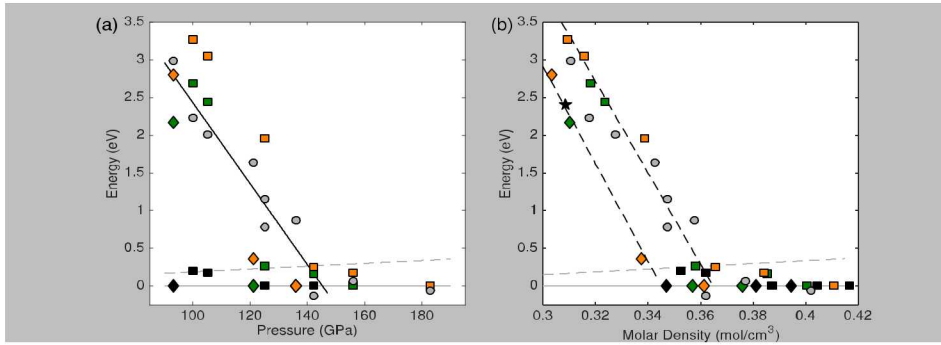

Dynamic compression experiments on deuterium and their implications for first-principles theory

M.D. Knudson and M.P. Desjarlais
Sandia National Laboratories

M. Preising and R. Redmer
University of Rostock

SAND2019 – XXXX

EPSC-DPS Joint Meeting 2019, Geneva, Switzerland

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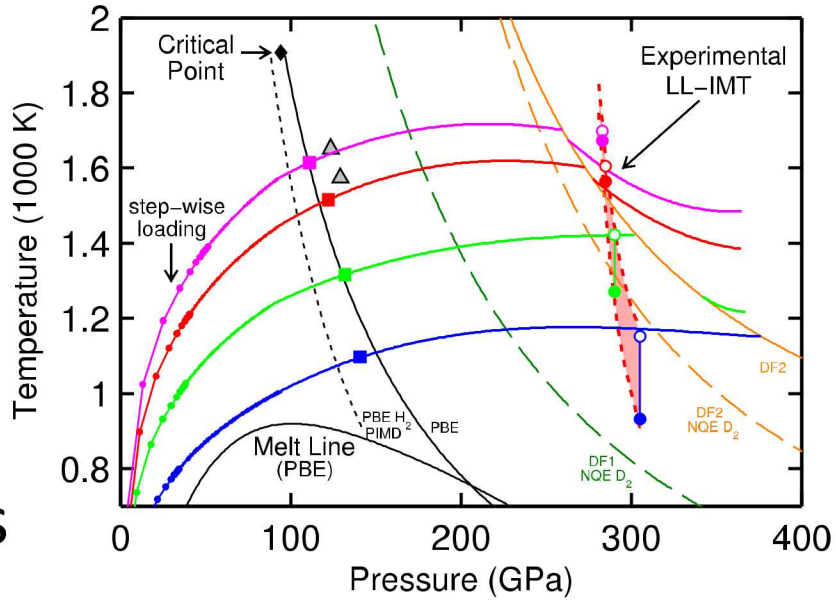
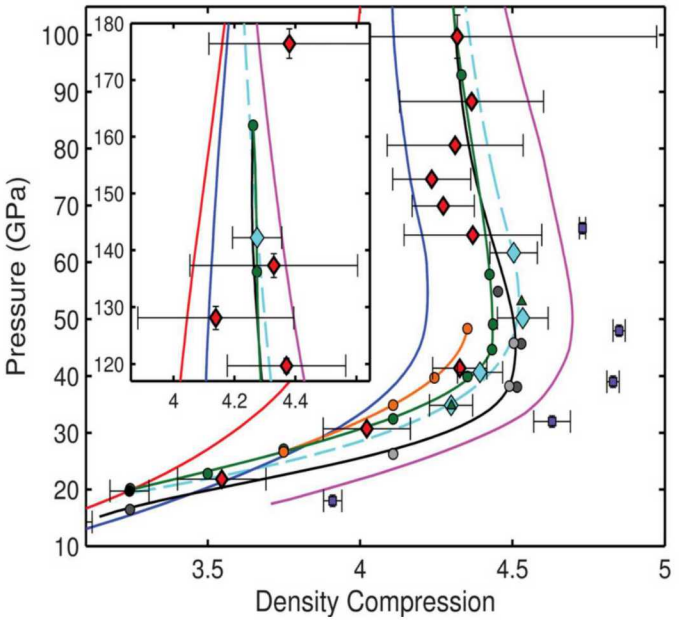
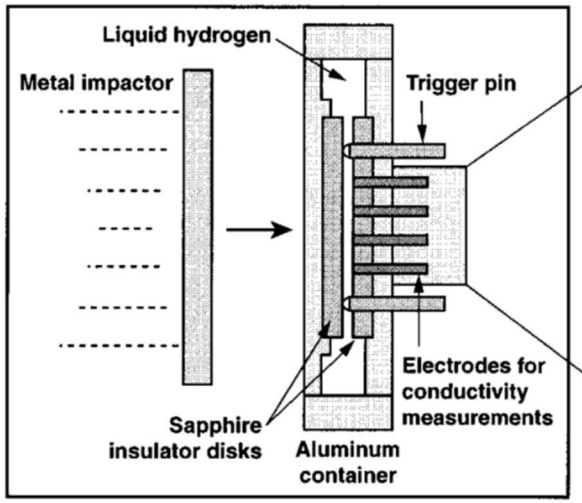
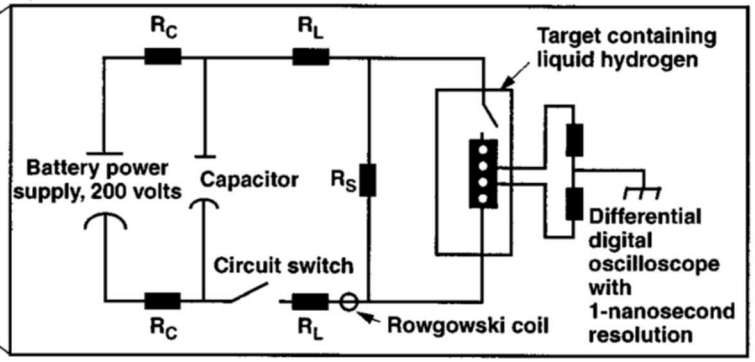
Outline

- Motivation
  - Insulator-metal transition at high-ρ and low-T
  - Insulator-metal transition at low-ρ and high-T
- Reanalysis of multiple-shock conductivity (σ) measurements
  - Inconsistencies in inferred T and semiconductor model used to interpret the measured σ
- Comparison of σ with various exchange-correlation (xc) functionals (PBE, DF1, DF2)
- Conclusions

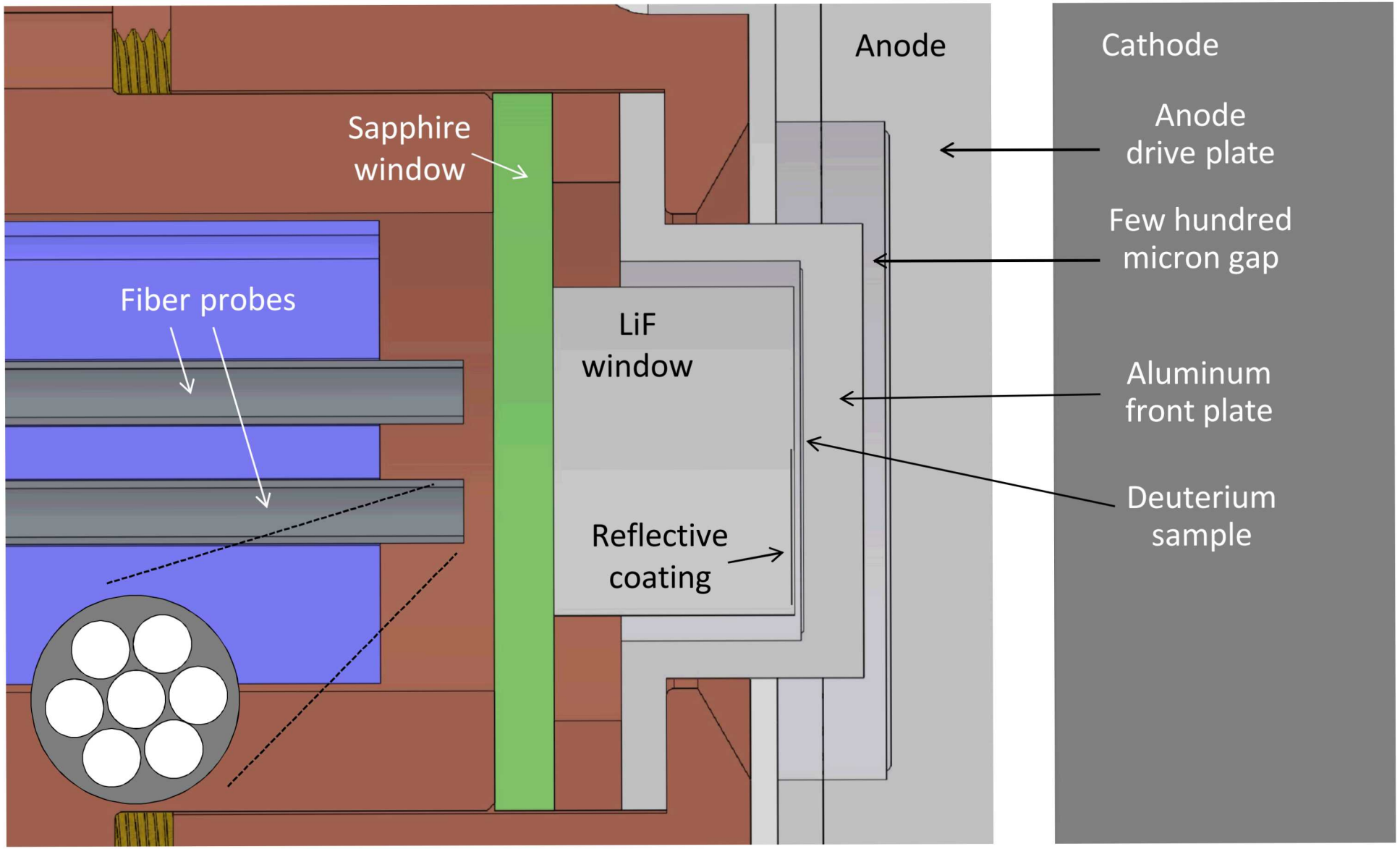




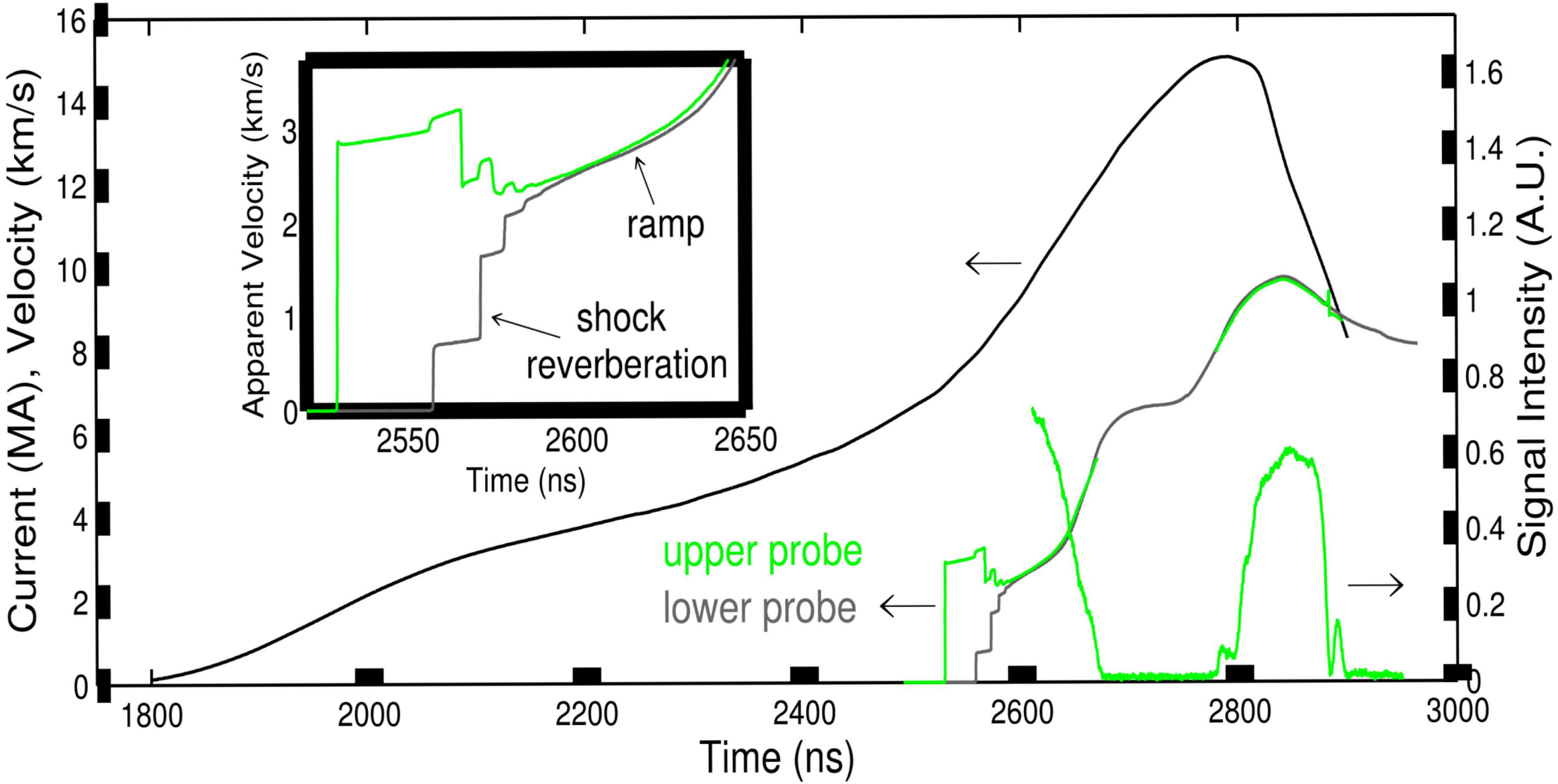




Experimental Configuration: high-ρ and low-T

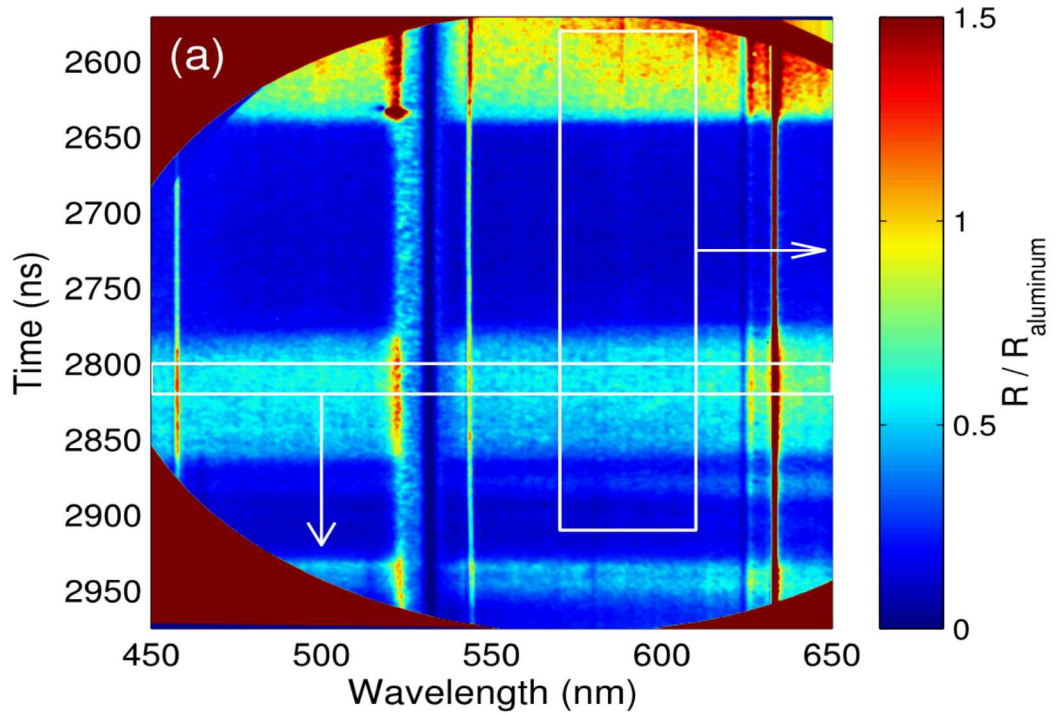


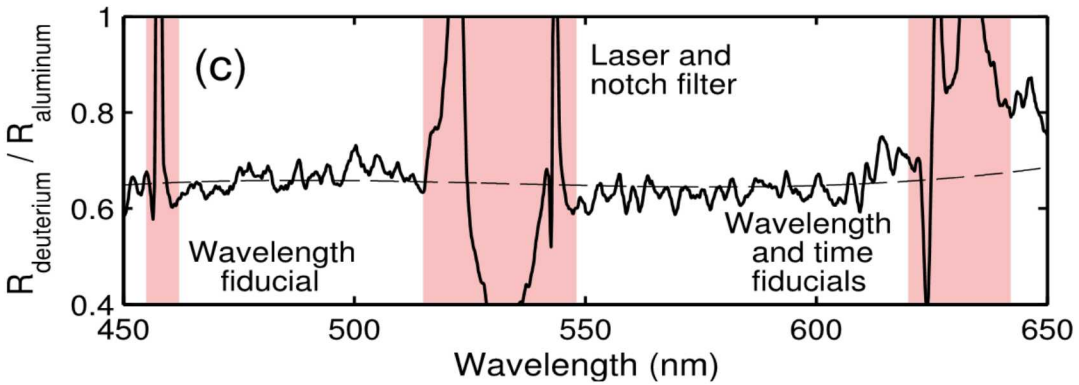
Representative data: high-ρ and low-T

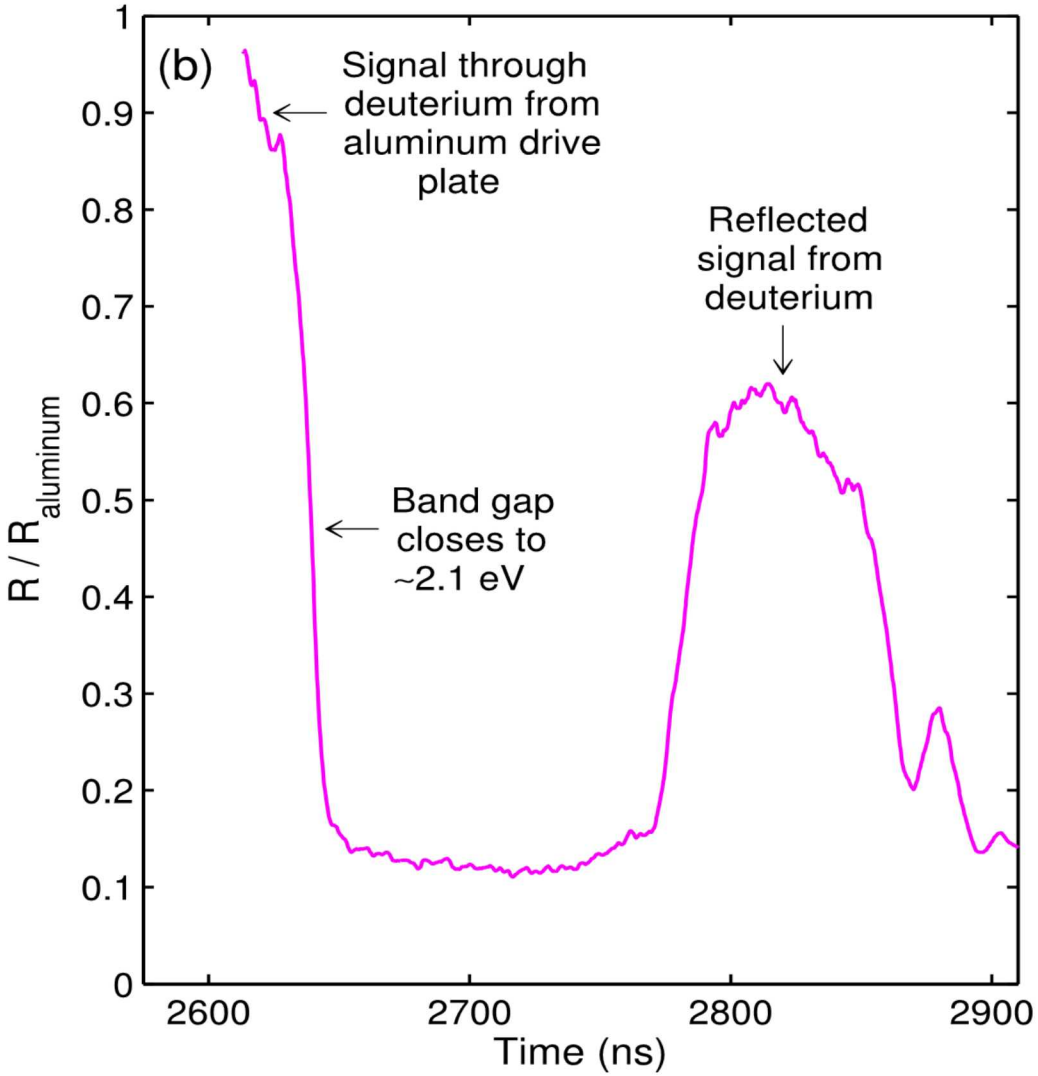


Knudson, Desjarlais et al, Science 348, 1455 (2015)

Wavelength dependence indicative of a metal

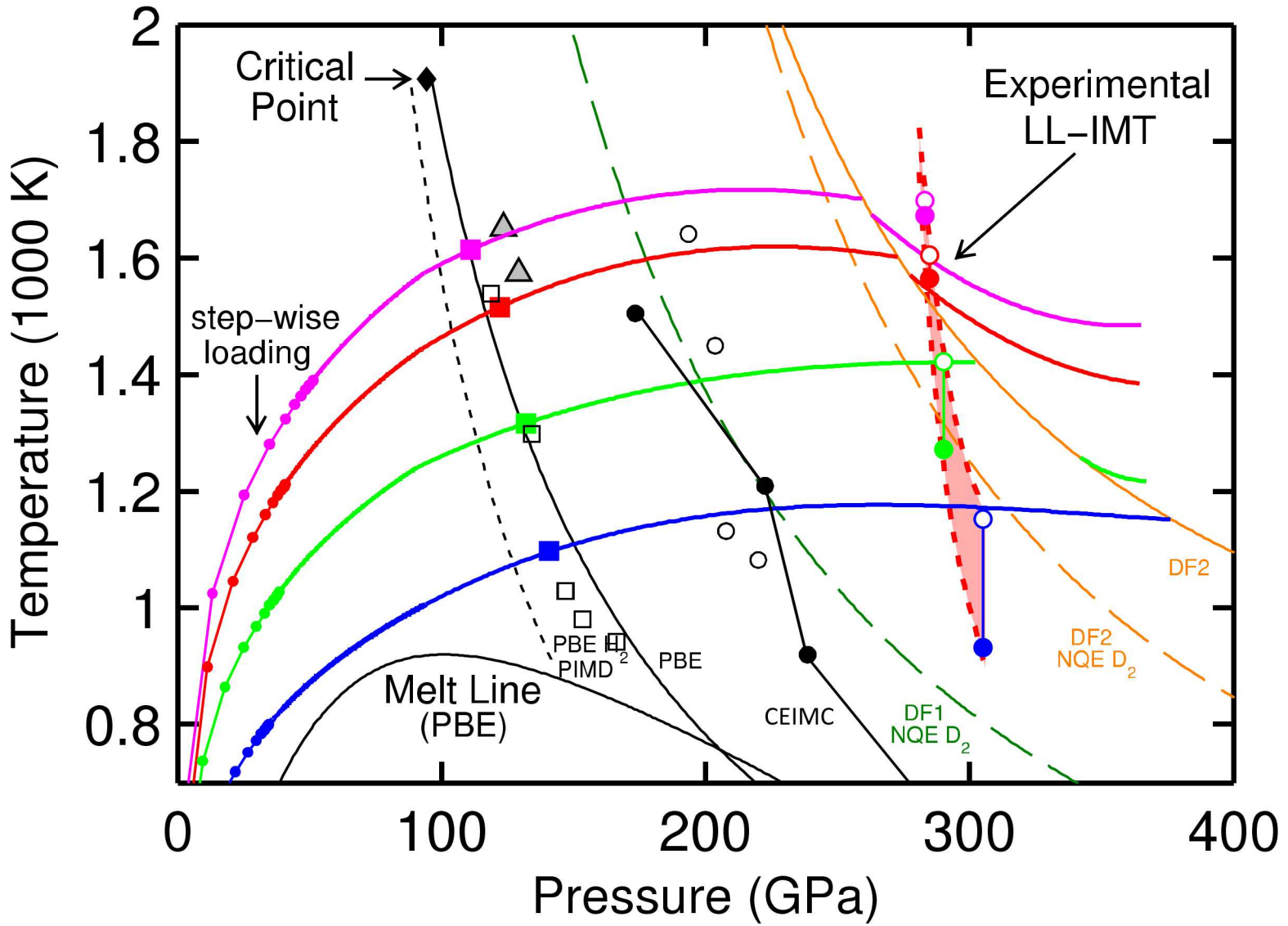




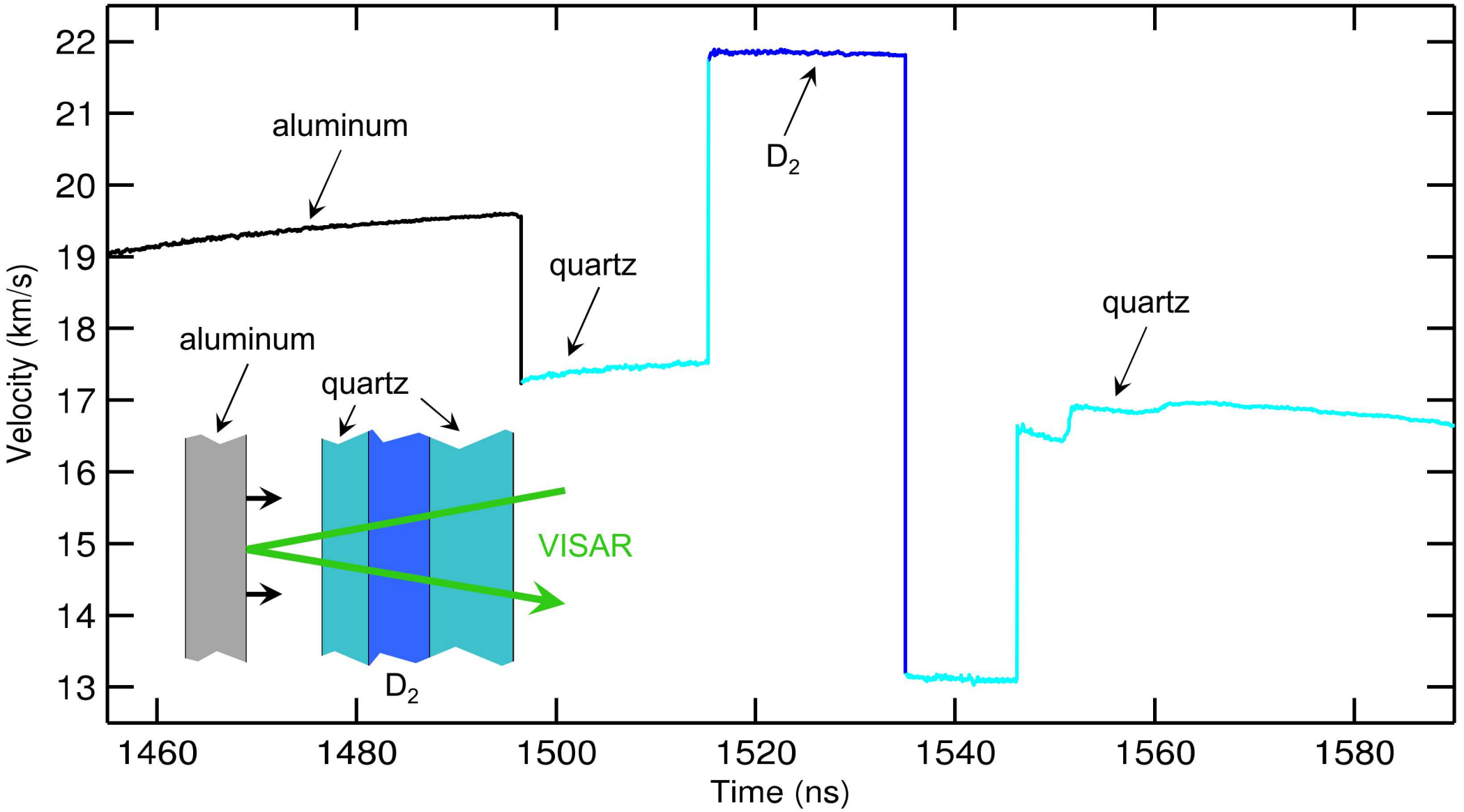


Insulator-metal transition at high-ρ and low-T

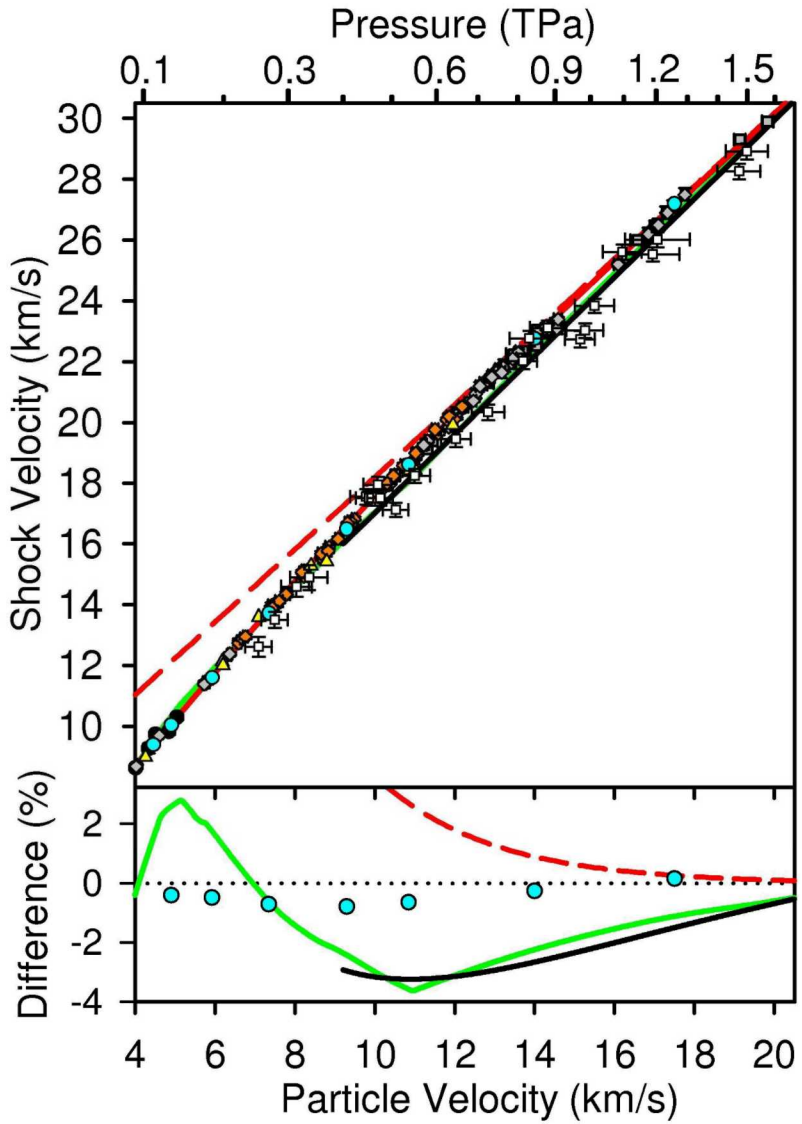
- Shock-ramp technique enabled experimental access to liquid-liquid, insulator-metal transition (LL-IMT)
- Experiments above ~250 GPa show clear evidence of metallization of deuterium
- Best agreement with nonlocal xc functionals
  - More complex T-dependence
- Not consistent with semi-local xc functional PBE
  - Dissociation occurs at much too low P



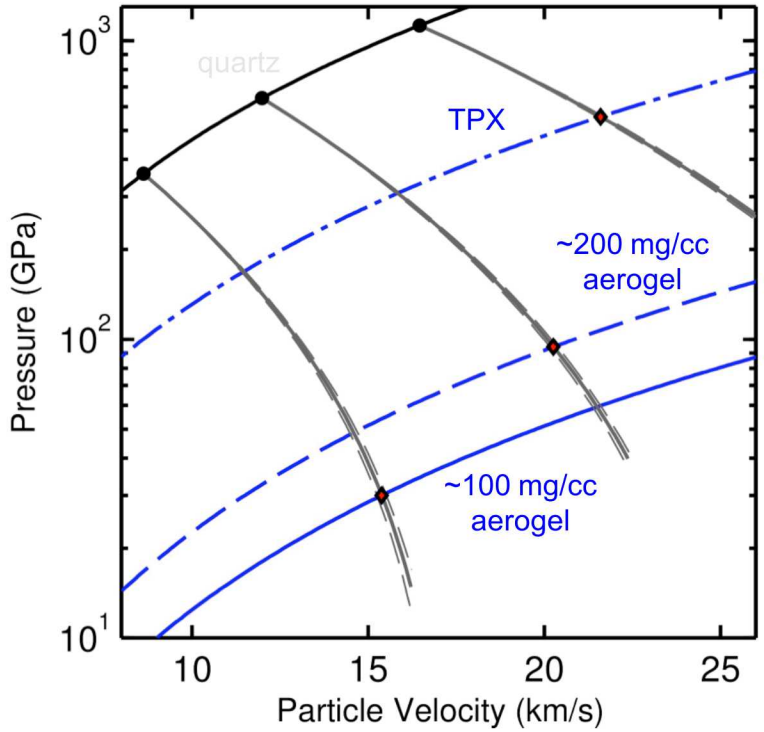
Experimental configuration and data: low-ρ and high-T

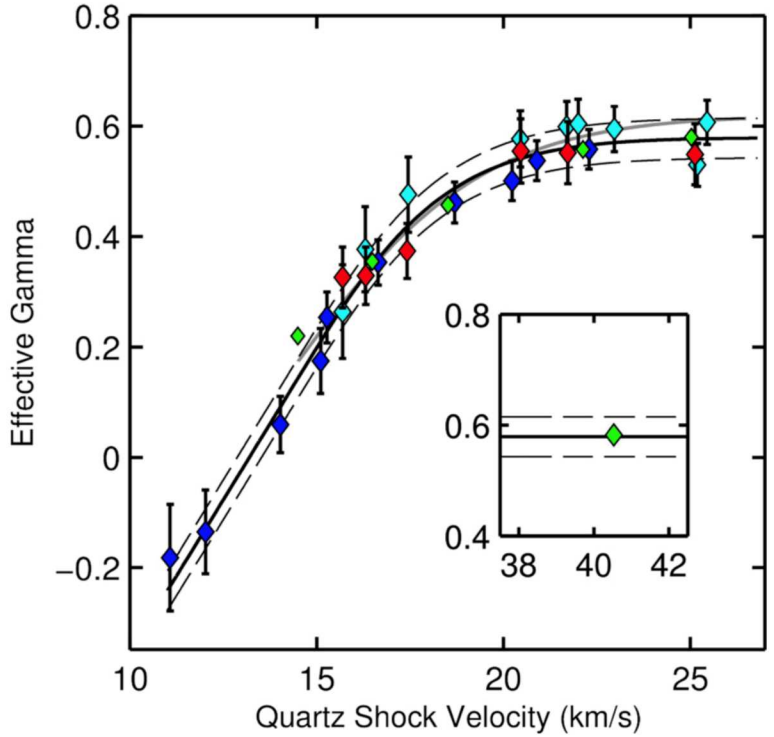


Quartz high precision standard



- Nearly 300 Hugoniot points for quartz have been obtained between 1 and 15 Mbar
- A release model was developed using release measurements obtained from TPX, and both ~200 mg/cc and ~100 mg/cc aerogel and based on insight gained from QMD calculations

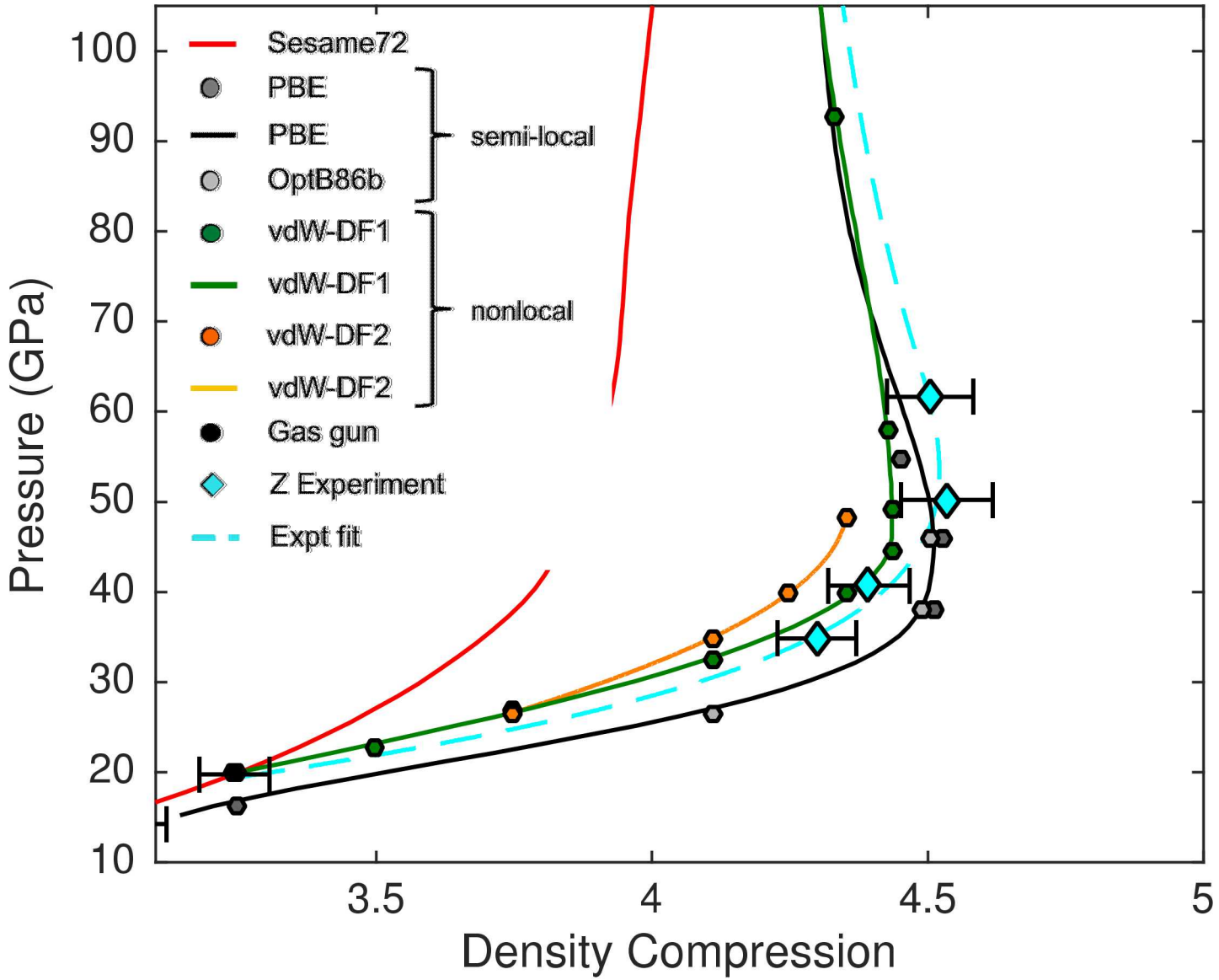




Knudson and Desjarlais, PRL 103, 225501 (2009); PRB 88, 184107 (2013); Desjarlais, Knudson, and Cochrane, JAP 122, 035901 (2017)

Insulator-metal transition at low-ρ and high-T

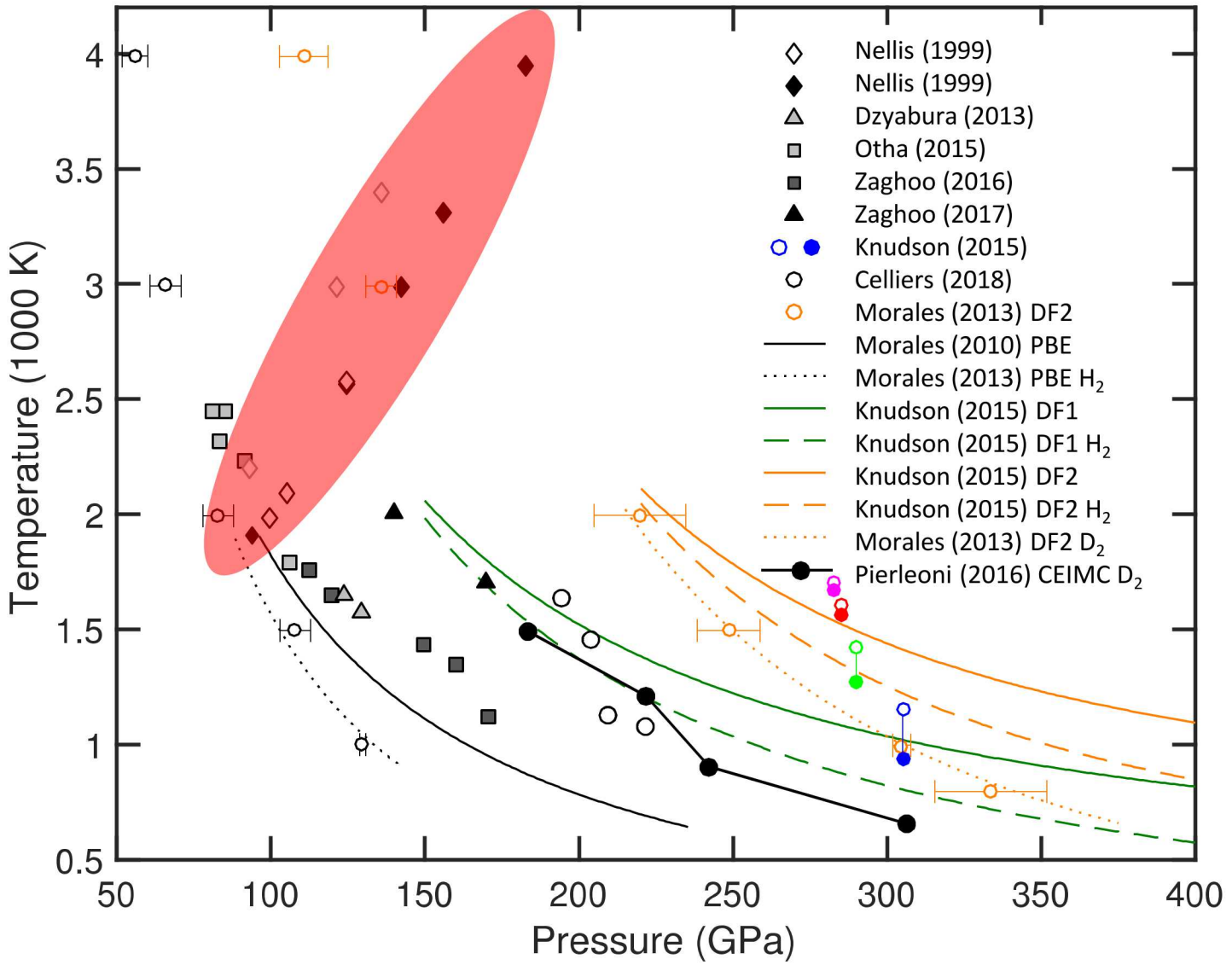
- Recent high-precision measurements of the deuterium Hugoniot near the IMT
  - Provides unique insight into the dissociation process
- Results compared with various xc functionals
  - Nonlocal functionals better describe the onset of dissociation
  - These same functionals exhibit a significantly wider P range for dissociation to complete
- Provide means for evaluation of future theoretical developments and new xc functionals



Knudson and Desjarlais, PRL 118, 035501 (2017)

Insulator-metal transition at intermediate ρ and T

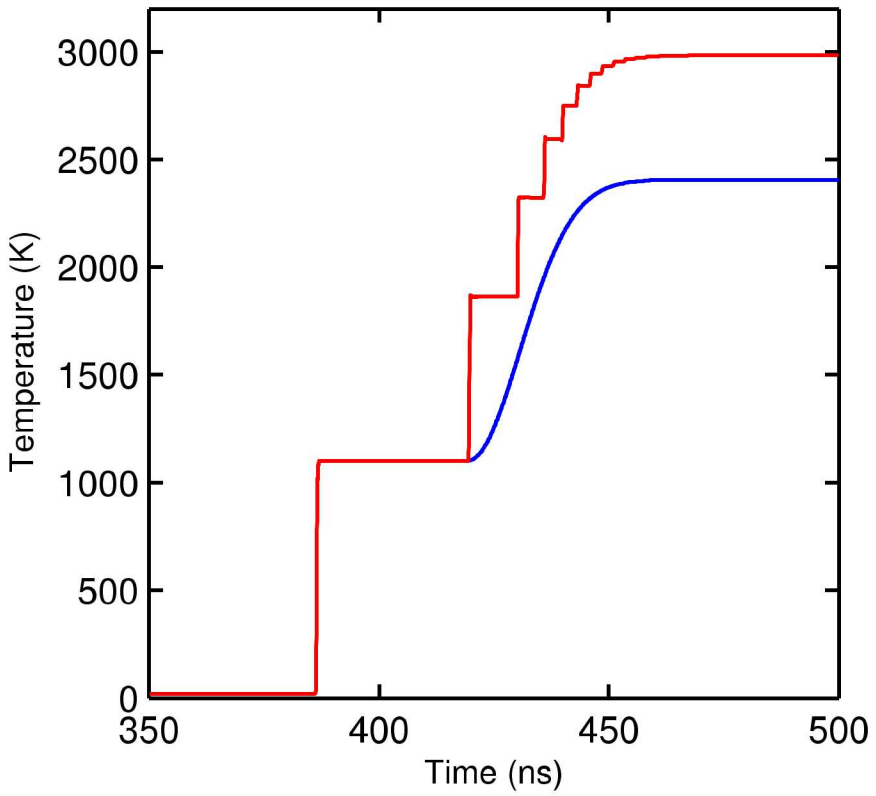
- The first experiments to address the IMT in liquid hydrogen and deuterium were performed by Nellis, Weir, and Mitchell in the 1990's
- Gas-gun technique
  - Multiple-shock compression at successively higher P and T states
  - Measured electrical conductivity
- Set of experiments provides a very good test of first-principles methods
  - Probes a regime in which both T and ρ (or P) play an important role



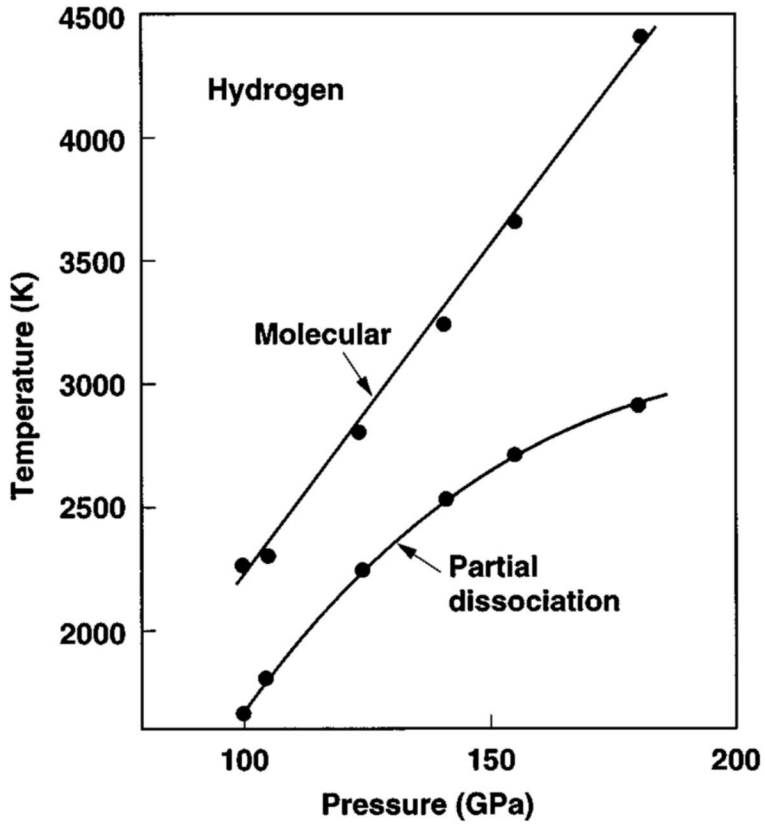
Reanalysis of inferred temperature states

- Nellis et al used two different methods to infer T and ρ
  - Method 1: Hydrocode simulation using Kerley EOS
  - Method 2: Isentrope calculation from first shock state for Ross EOS
- These methods described as being different but equivalent
  - This is incorrect
  - Method 2 fails to account for non-negligible increase in entropy from subsequent shocks during ring-up
- Reported T is too low and reported ρ is too high

T profiles using the two methods from Nellis et al



Comparison of inferred T from the two different methods



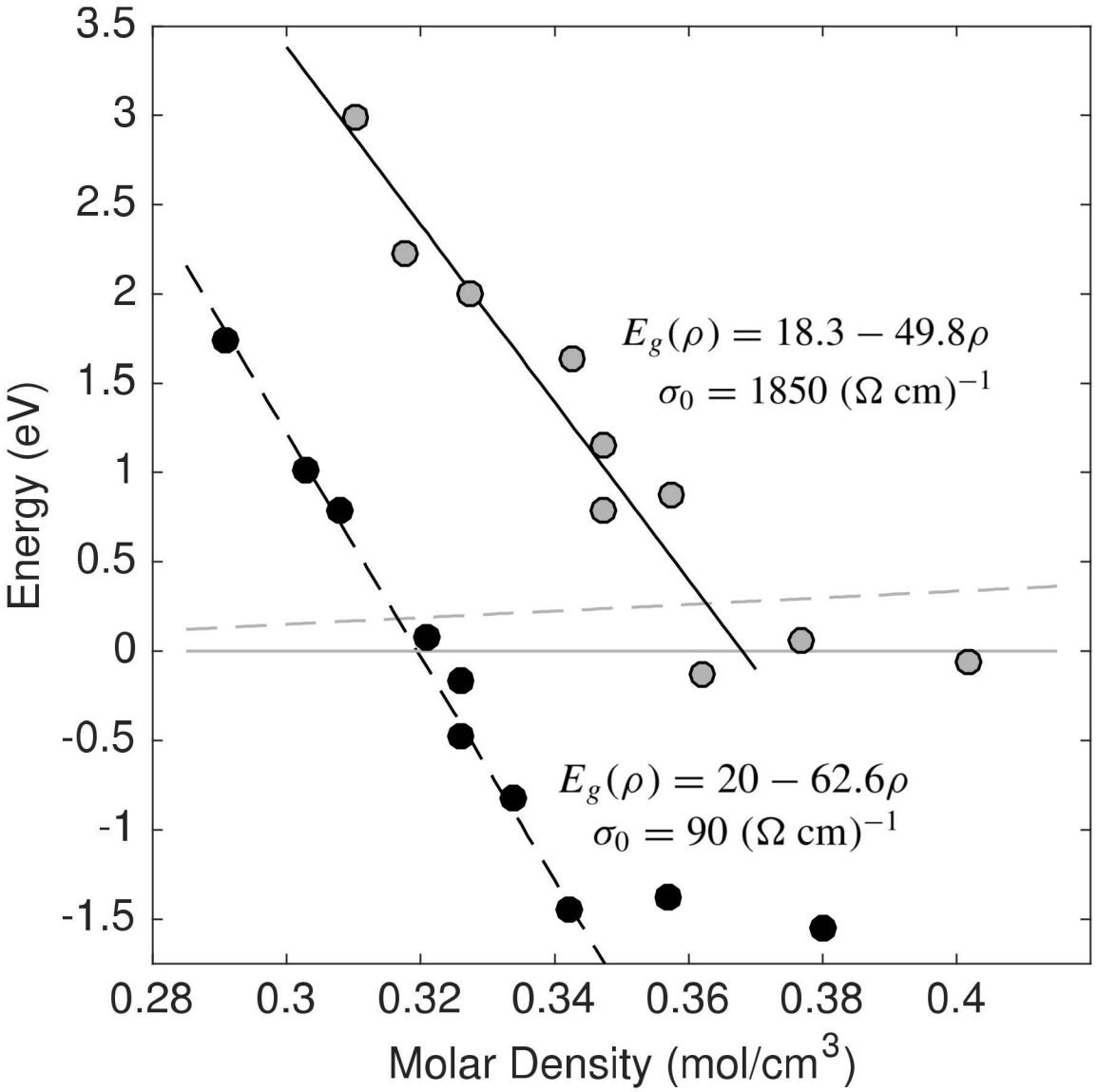
Reanalysis of semiconducting model

- Nellis et al appealed to a simplified semi-conducting model to infer an energy gap as a function of ρ

$$\sigma = \sigma_0 \exp[-E_g(\rho)/2k_B T]$$

- The limiting conductivity was taken to be a free parameter
  - This results in a negative energy gap for $\rho > 0.32$ mol/cm^3

- With the limiting conductivity taken from experiment the result is more physical
  - The gap closes at $\rho \sim 0.37$ mol/cm3

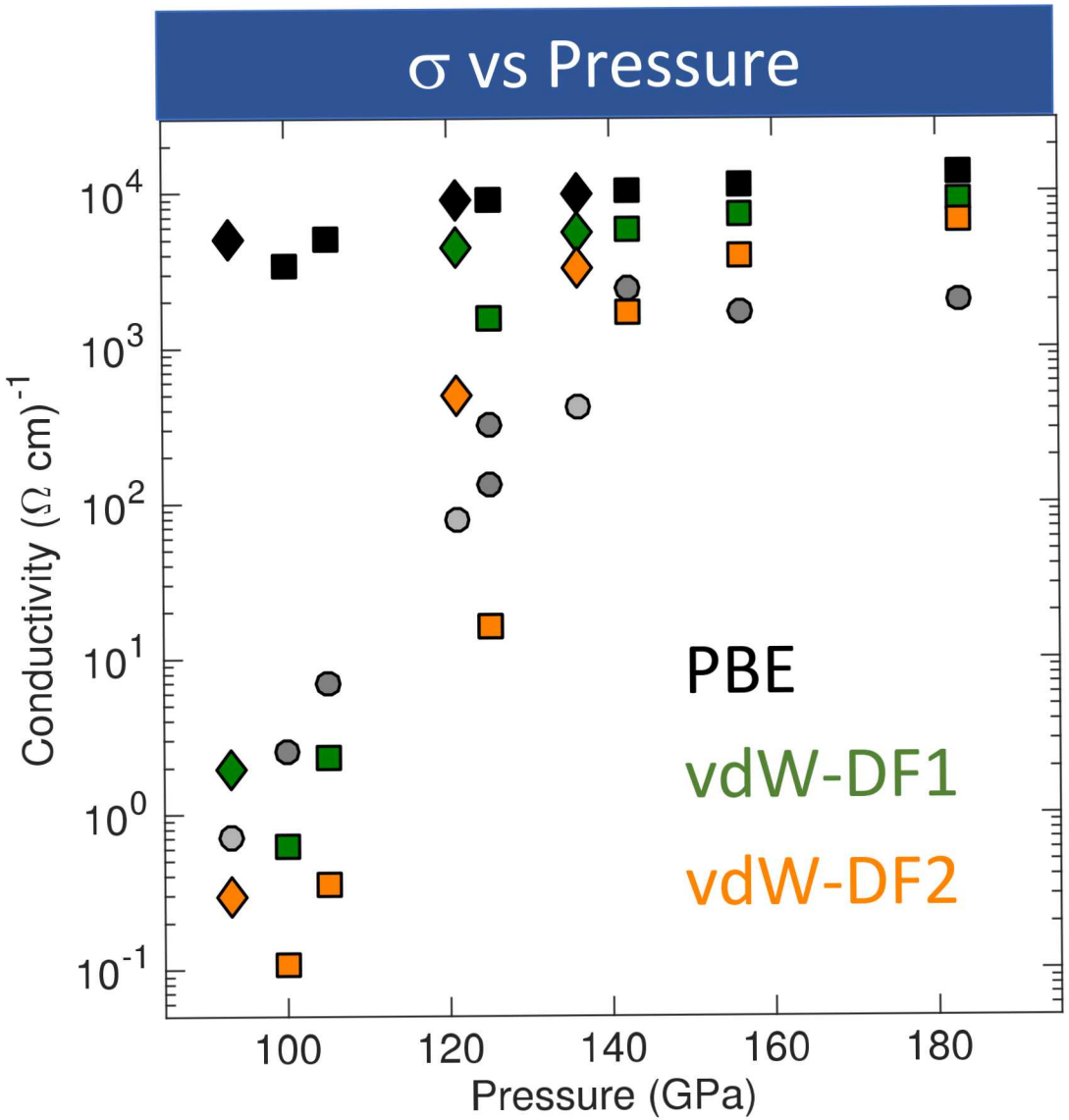


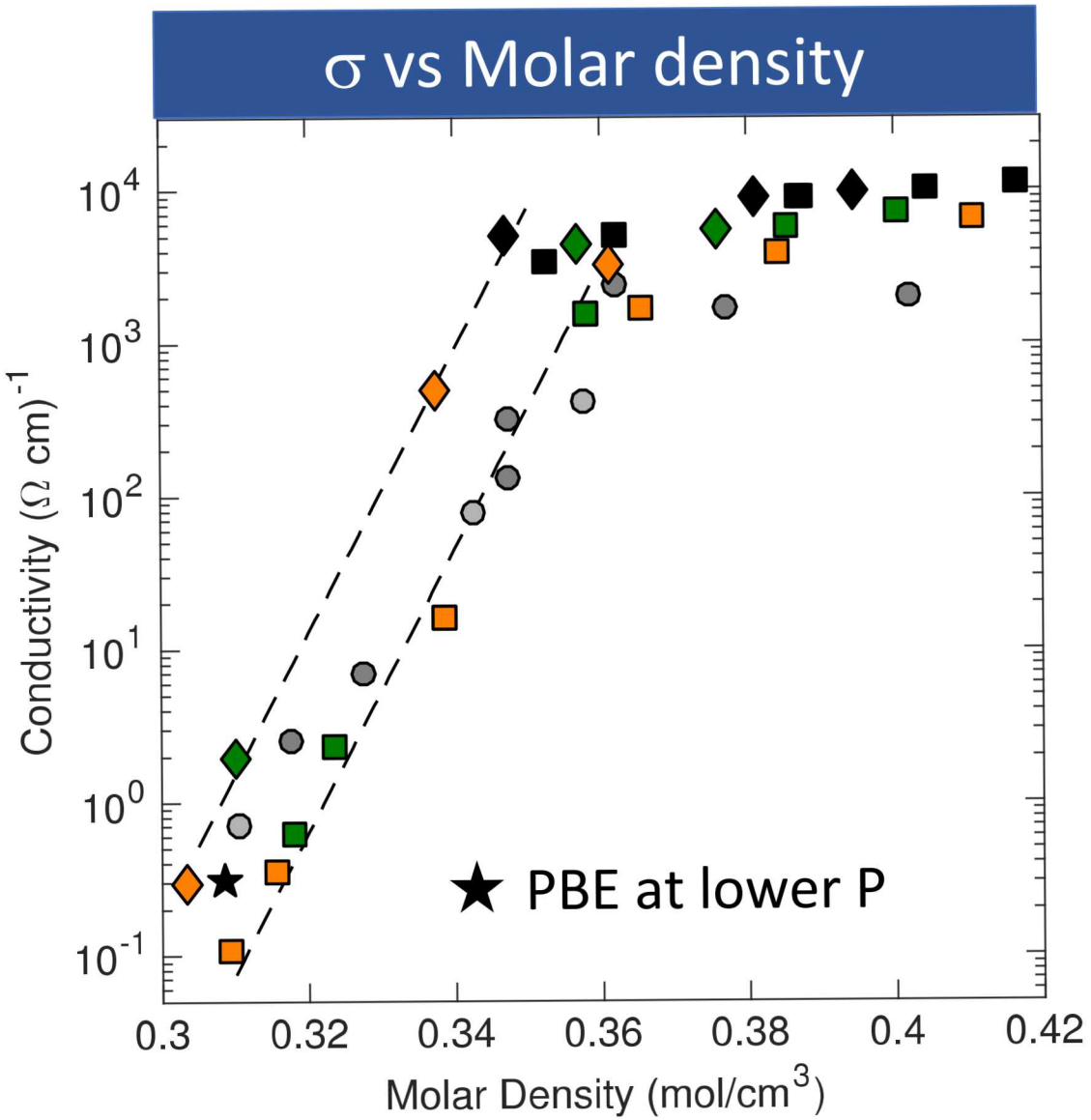
Determination of the peak state of the system

- P is experimentally constrained and does not depend upon the hydrogen/deuterium EOS
- In this regime P depends much more strongly on ρ than T
- Chose to fix T, then varied ρ to match experimentally measured P

		Kerley03			PBE		DF1		DF2	
Experiment	σ $(\Omega\ \mathrm{cm})^{-1}$	P (GPa)	T (K)	ρ (g/cm^3)	σ $(\Omega\ \mathrm{cm})^{-1}$	ρ (g/cm^3)	σ $(\Omega\ \mathrm{cm})^{-1}$	ρ (g/cm^3)	σ $(\Omega\ \mathrm{cm})^{-1}$	ρ (g/cm^3)
SLDMS4-D_2	0.71	93	2204	1.25	4989	1.397	1.99	1.249	0.30	1.222
SLDMS5-D_2	77	121	2987	1.38	8919	1.534	4518	1.437	493	1.359
SLDMS8-D_2	417	136	3397	1.44	9970	1.589	5461	1.514	3234	1.455
SLDMS6-H_2	2.6	100	1978	0.64	3456	0.710	0.62	0.641	0.11	0.624
SLDMS13-H_2	7.1	105	2093	0.66	4990	0.729	2.32	0.652	0.35	0.636
SLDMS7-H_2	135	125	2567	0.70	9034	0.780	1546	0.722	16.2	0.683
SLDMS9-H_2	313	125	2573	0.70	9034	0.781	1546	0.722	16.2	0.683
SLDMS12-H_2	2380	142	2984	0.73	10410	0.816	5671	0.777	1693	0.737
SLDMS10-H_2	1670	156	3310	0.76	11290	0.840	7061	0.807	3880	0.774
SLDMS11-H_2	2000	183	3951	0.81	13180	0.881	8907	0.854	6551	0.827

Predicted conductivity at the peak states

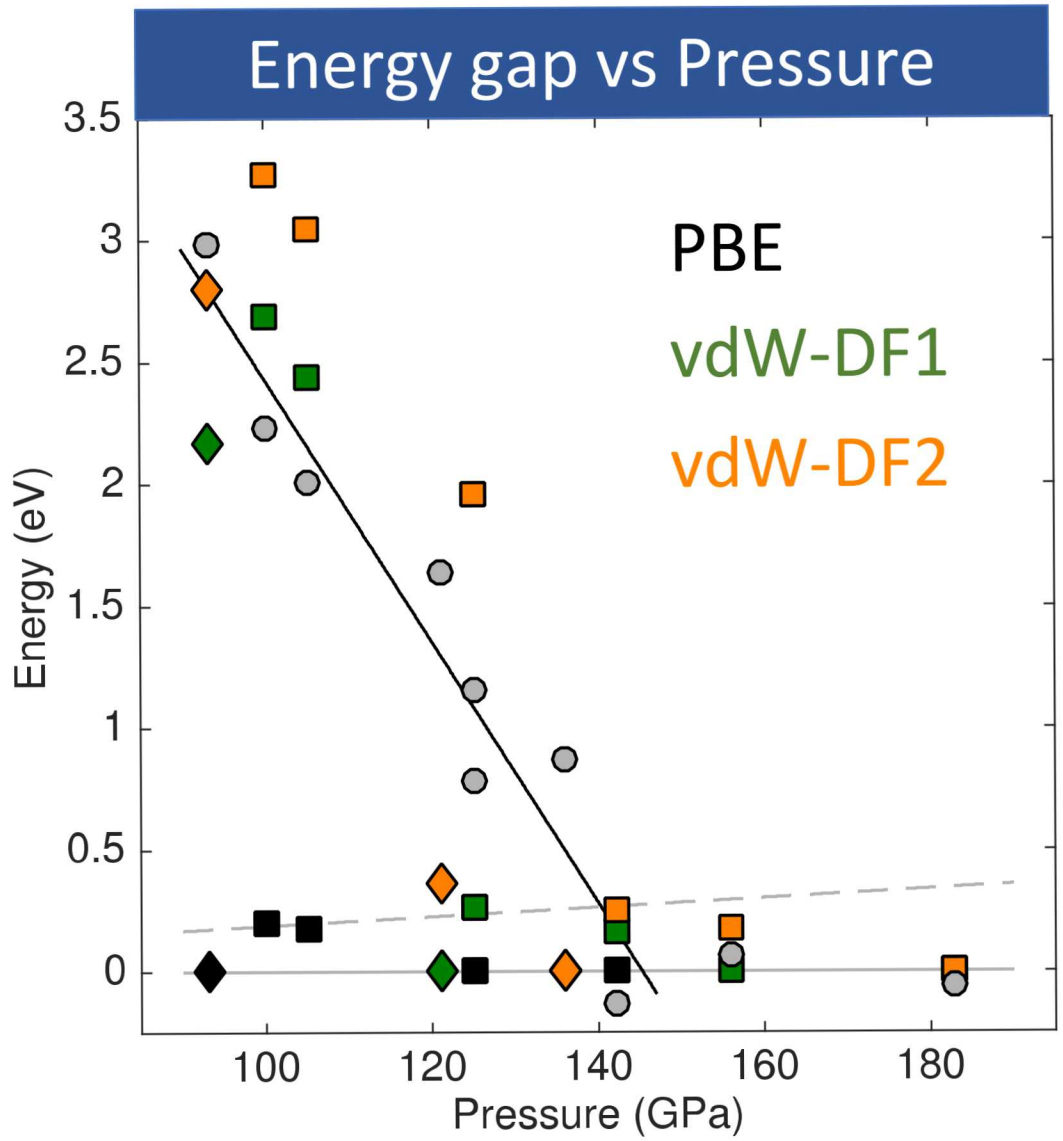


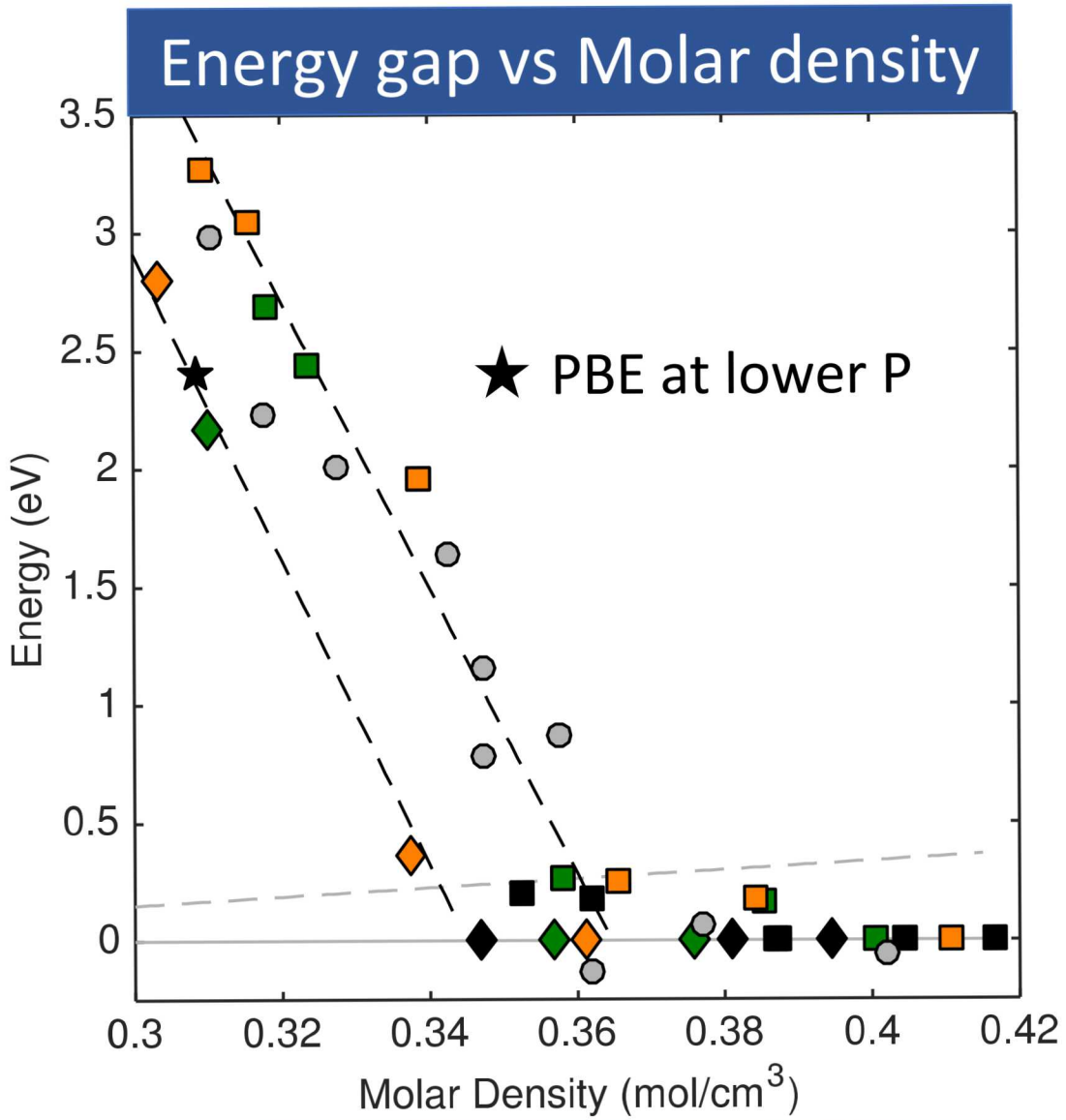


Nellis et al, PRB 59, 3434 (1999)

Knudson, Desjarlais et al, PRB 98, 174110 (2018)

Predicted energy gap at the peak states





Nellis et al, PRB 59, 3434 (1999)

Knudson, Desjarlais et al, PRB 98, 174110 (2018)

Inconsistency of the PBE xc functional

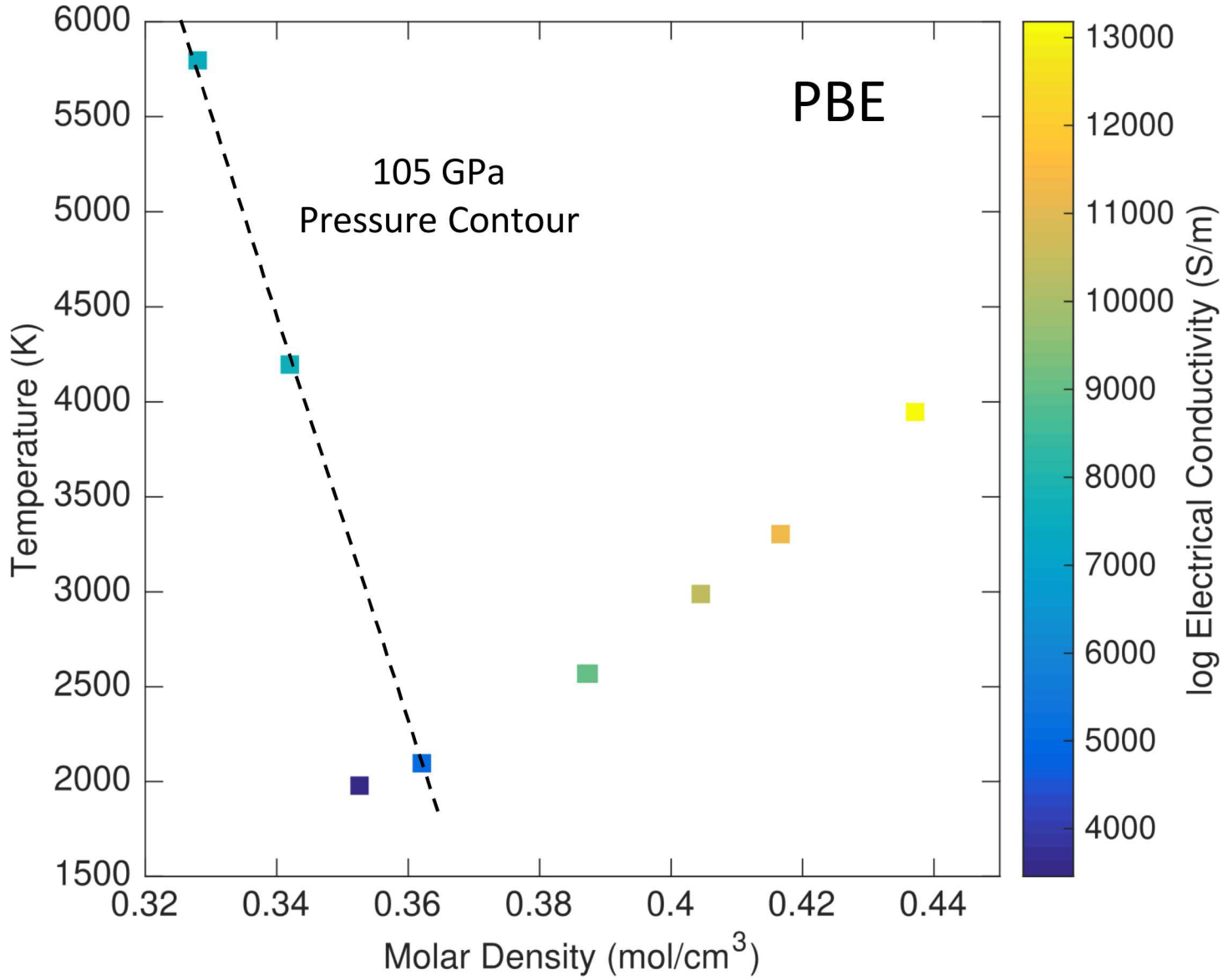
- What if we fix P and ρ instead of P and T?
- SLDMS13-H_2 (105 GPa)
  - Kerley EOS predicts

    T = 2093 K ρ = 0.66 g/cm^3
  - Fix T and P

    ρ = 0.729 g/cm^3 σ = 4990 (Ω cm)$^{-1}$
  - Fix P and ρ

    T = 5800 K σ = 7500 (Ω cm)$^{-1}$
  - Fix P and ρ = 0.689 g/cm3

    T = 4200 K σ = 7600 (Ω cm)$^{-1}$
- Regardless of how one tries to equate the experimental P, the predicted σ is inconsistent with the measured value



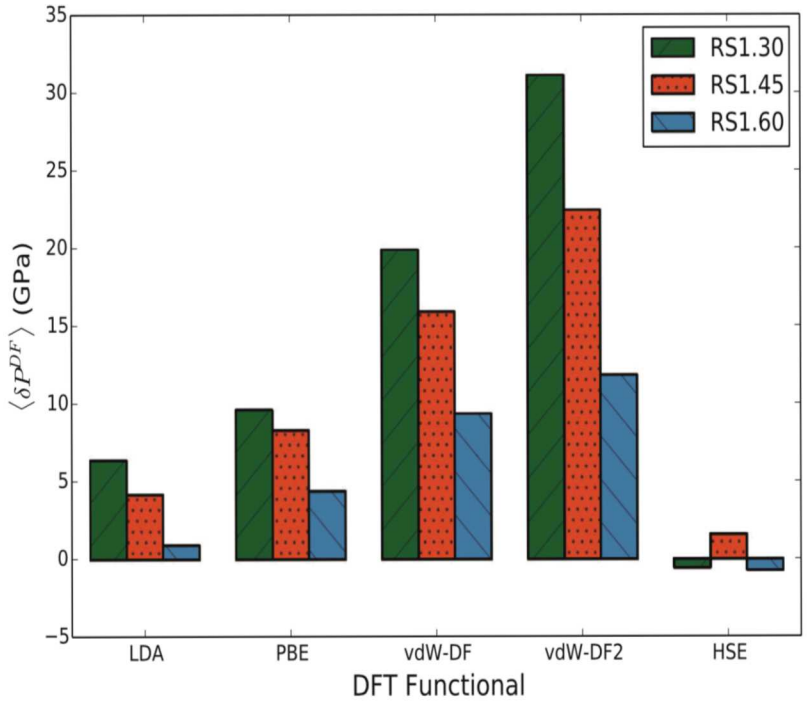
There appears to be a systematic trend in P

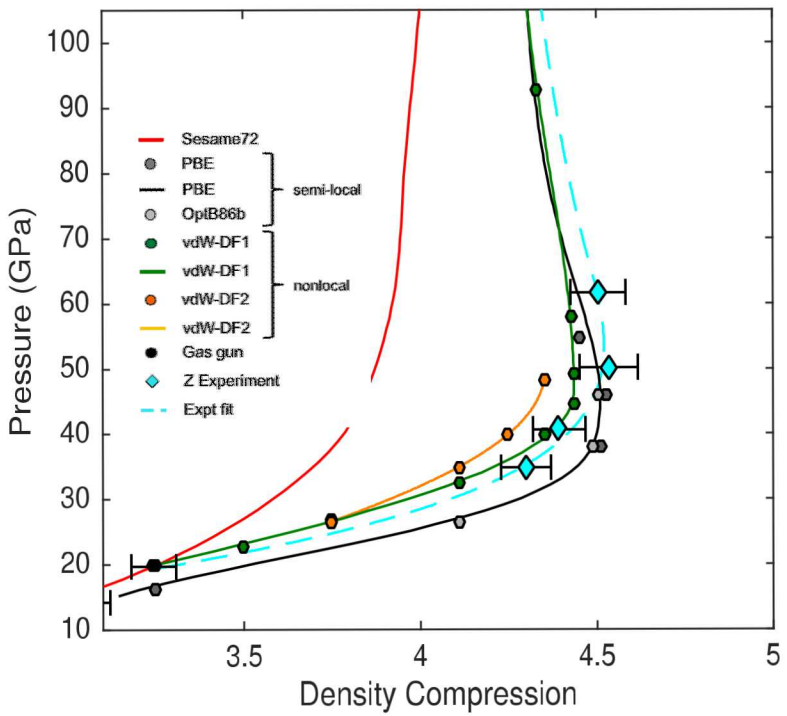
$$P_{PBE} < P_{DF1} < P_{DF2}$$

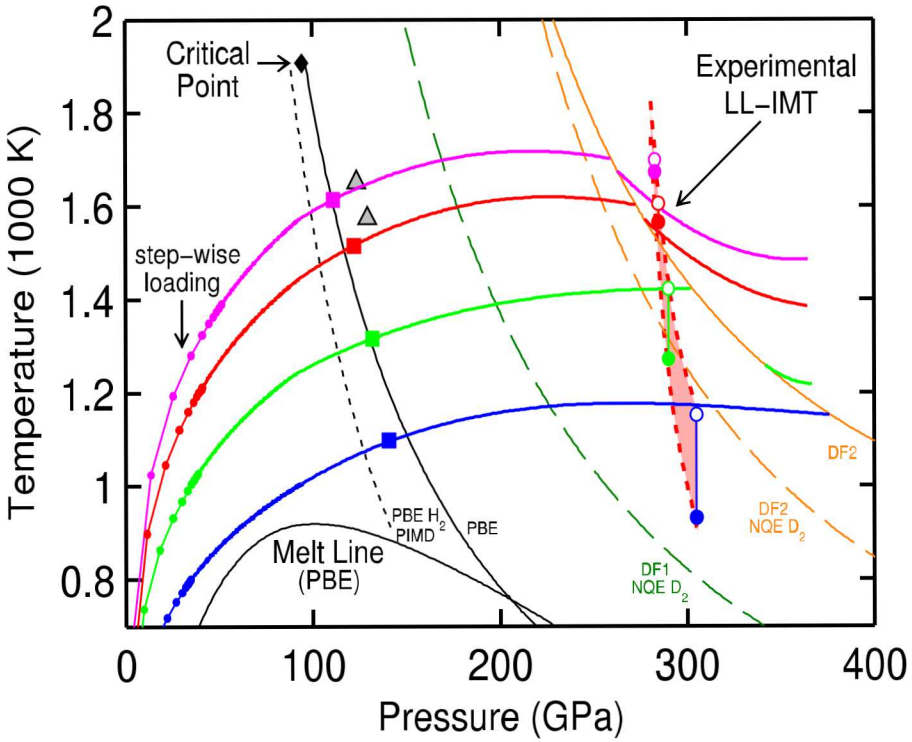
Pressure errors with respect to Quantum Monte Carlo

High-T, low-ρ dissociation along the Principal Hugoniot

Low-T, high-ρ dissociation above the melt boundary







Clay et al, PRB 89, 184106 (2014)

Knudson and Desjarlais, PRL 118, 035501 (2017)

Knudson, Desjarlais et al, Science 348, 1455 (2015)

Conclusions

- Performed a reanalysis of the Nellis, Weir, and Mitchell experiments
  - Corrected an inconsistency in the inferred T and ρ states and semiconducting model
- Performed a detailed comparison of the measured σ with first-principles density functional theory using various xc functionals
- Results found to be inconsistent with the semi-local xc functional PBE
  - Inconsistency likely stems from P errors associated with the PBE xc functional that result from premature dissociation
  - Calculated P are too low at these T and ρ conditions
- Results found to be in better agreement with non-local vdW functionals
- Together with previous comparisons at high-T, low-ρ and low-T, high-ρ, these results provide a consistent picture for the IMT over a wide T and ρ range