APS Division of Plasma Physics Meeting, 11/16-20, 20

Shock compression of glow discharge polymer (GDP): density functional theory (DFT) simulations and experiments on Sandia's Z-machine

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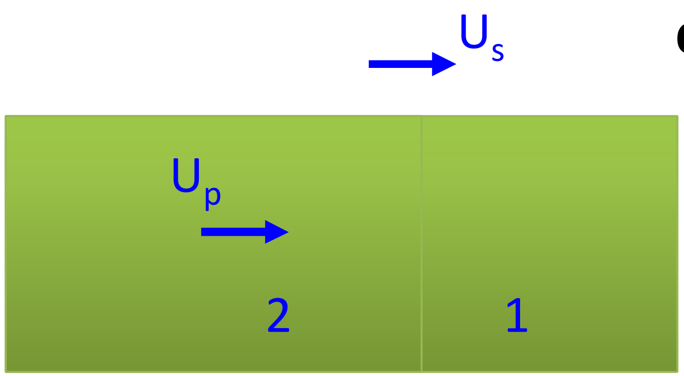
GDP fusion capsules

- **GDP is one of the possible ICF ablator materials**
  - Imploded using a series of convergent shocks
  - Initial shock at approximately 3 Mbar
    - *Just above complete dissociation*
    - *10% error bars too large for adequate EOS*
- **First-principles thermodynamics using Density Functional Theory (DFT)**
  - An excellent track record predicting hydrocarbon and other materials' shock properties
  - D_2, C(Diamond), Polymethylpentene (TPX), Polyethylene, Ethane
  - CO_2, H_2O, SiO_2, MgO
  - Neon, Argon, Krypton, Xenon, Tungsten, Copper, Platinum, Aluminum
- **GDP has a complex structure – we expected a challenge, and got one**

GDP fusion capsule for NIF

Shock compression is a way to investigate thermo-physical properties of matter at extreme pressures

- *Conservation of mass, energy, and momentum* lead to the **Rankine-Hugoniot condition** for the initial (1) and final state (2)

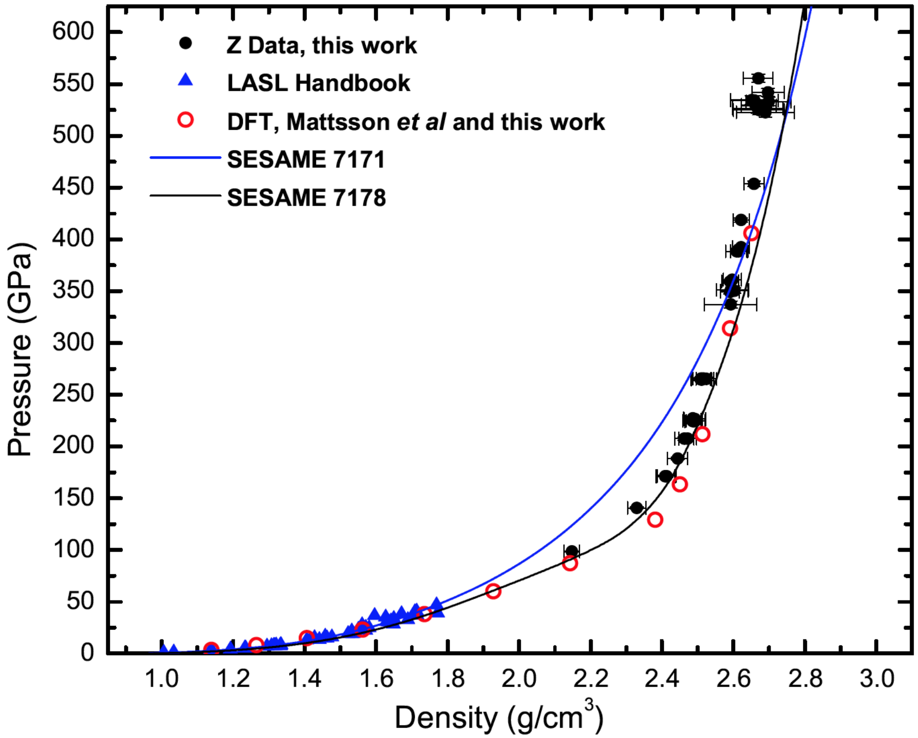


$$2\left(E_2 - E_1\right) = \left(P_2 + P_1\right)\left(\upsilon_1 - \upsilon_2\right)$$

- With high accuracy measure and/ or calculate thermo-physical properties
- *First Principles Thermodynamics*

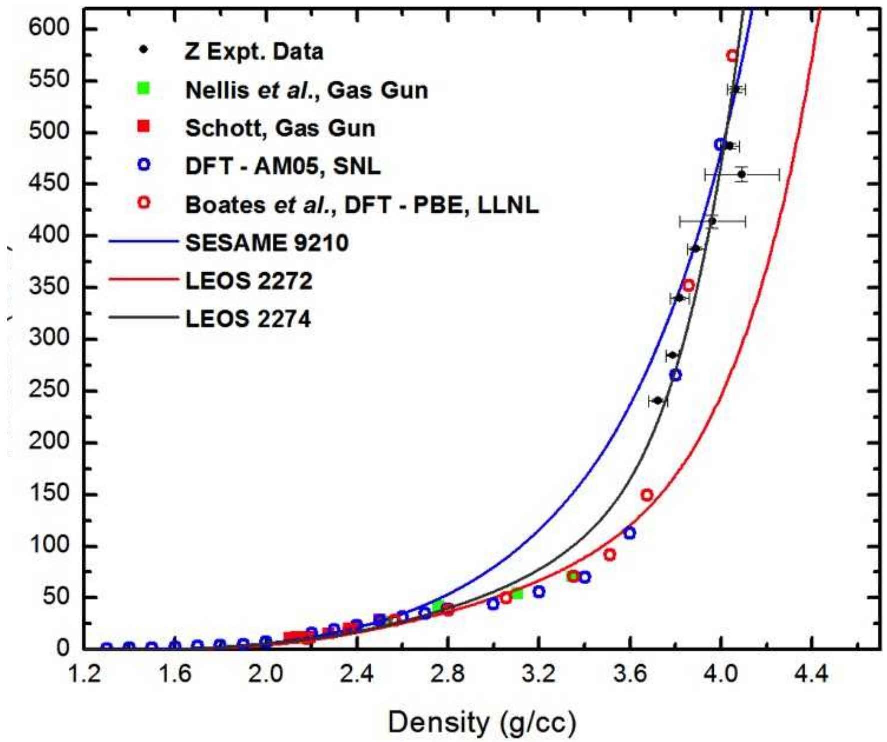
Examples of past DFT Hugoniot predictions

First shock in Polymethane-pentene (PMP/TPX)



Seth Root et al J. Applied Physics, accepted for publication (2015).

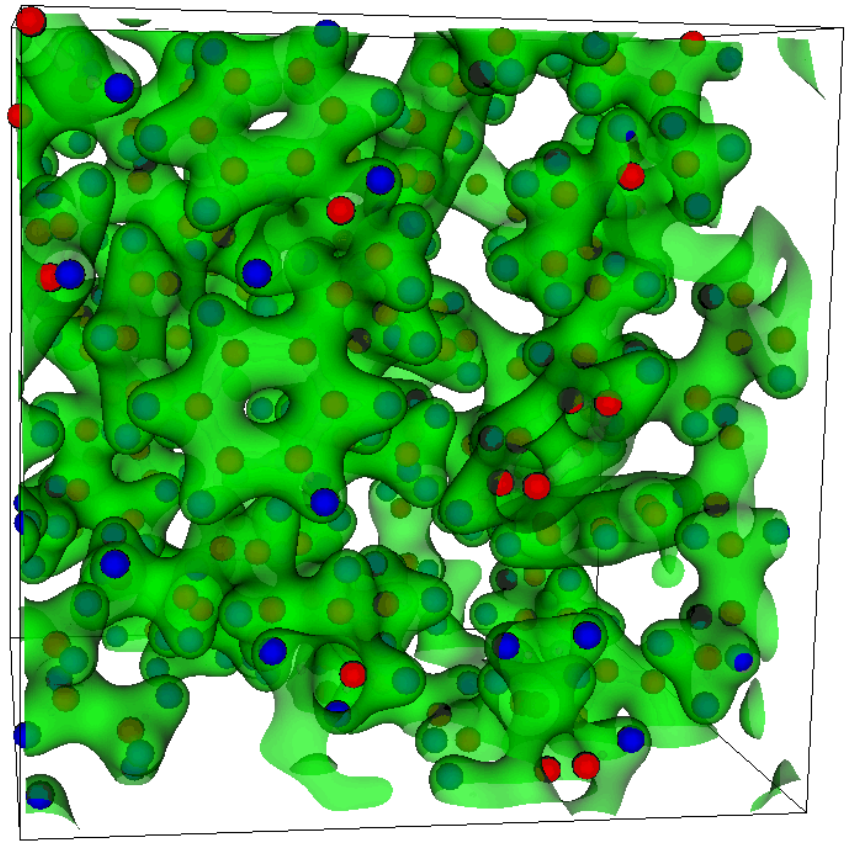
First shock in liquid CO2



Seth Root et al Phys. Rev. B. **87**, 224102 (2013).

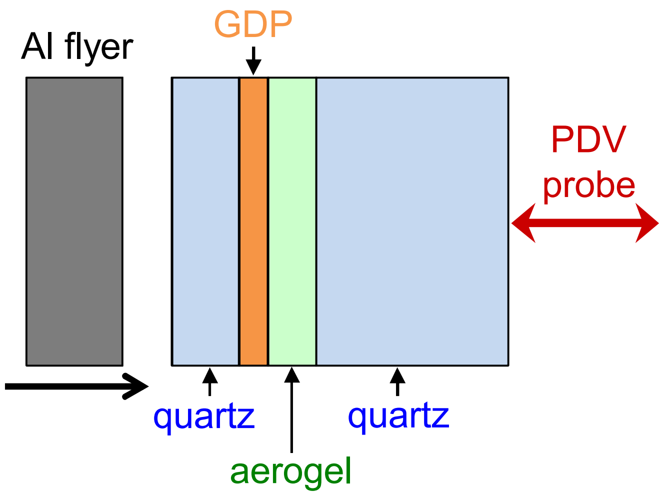
Use density functional theory (DFT) calculations to simulate glow discharge polymer.

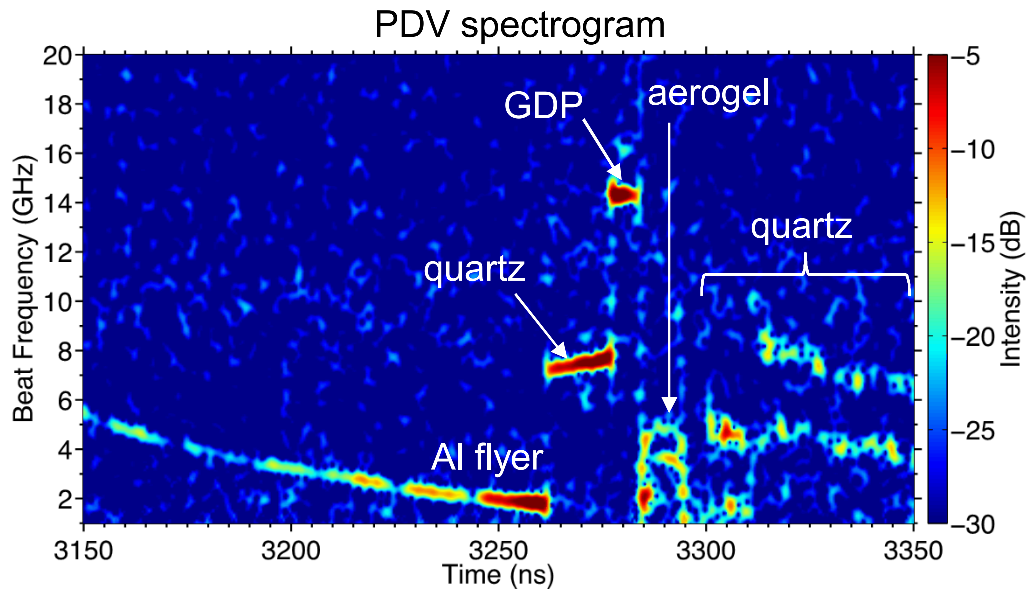
- **First-principles simulations DFT**
  - VASP – plane-wave code w PAW core-functions
  - Use of DFT codes simulating warm dense matter
    - *M. P. Desjarlais Phys. Rev. B **68**, 064204(2003)*
  - Great care in convergence
    - *A. E. Mattsson et. al. Modeling and Simulation in Material Science and Engineering **13**, R1 (2005)*
  - Mermin Finite Temperature DFT
  - Ions moved as classical point charges
- **Build a reference system**
  - 272 Hydrogen, 200 Carbon
  - Equilibrate at constant temperature and volume.
  - Equilibrated for 3000+fs at 0.1 to 0.5 fs
  - AM05 exchange-correlation functional
  - Standard deviation of energy and pressure <1%
  - Block averaging to reduce correlation
  - Initial atom positions courtesy of Sebastien Hamel (LLNL)
  - Additional reference states w varying H content



Quantum molecular dynamics (QMD) simulations give thermo-physical properties

Photonic Doppler Velocimetry (PDV) measurements of impact & shock velocities

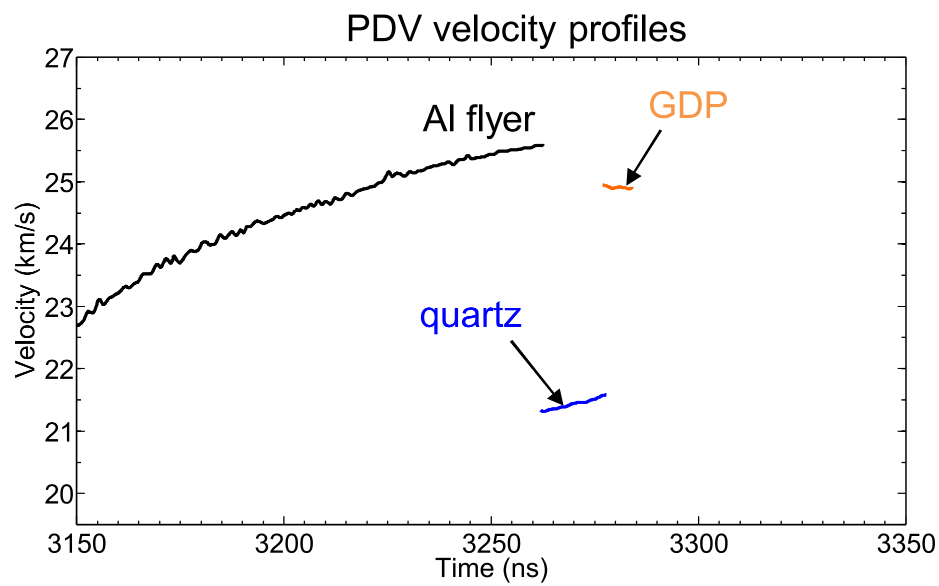




PDV mixes Doppler shifted target light with reference light

- Infrared light (1550 nm) transparent through GDP
- Velocity changes correspond to beat frequency shifts

$$f_b = \left| \frac{2v}{\lambda_T} + c_0 \left(\frac{1}{\lambda_T} - \frac{1}{\lambda_R} \right) \right|$$

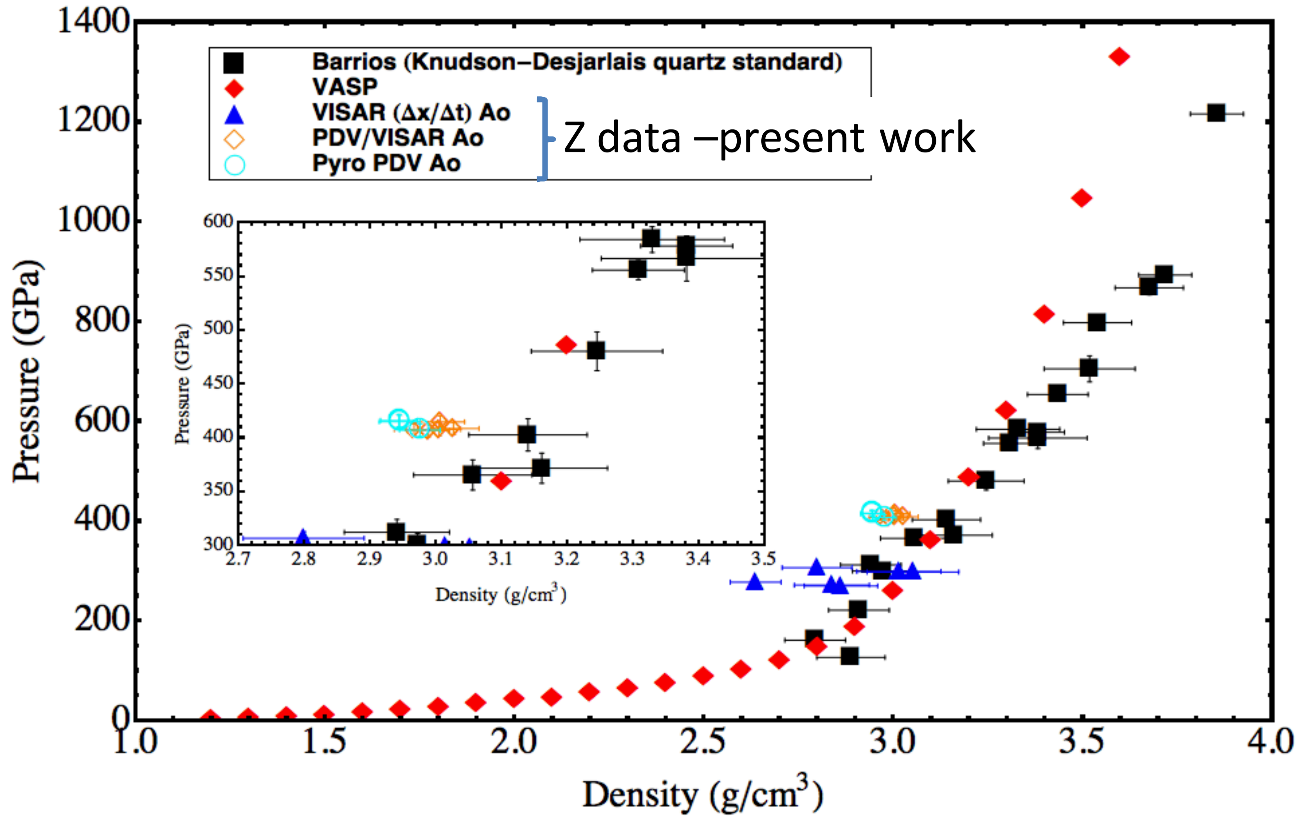


Courtesy of Tom Ao

DFT/MD (QMD) simulations compared to experimental data from Z and Omega

M. Barrios' data from Knudson and Desjarlais, PRB **88**, 184107 (2013).

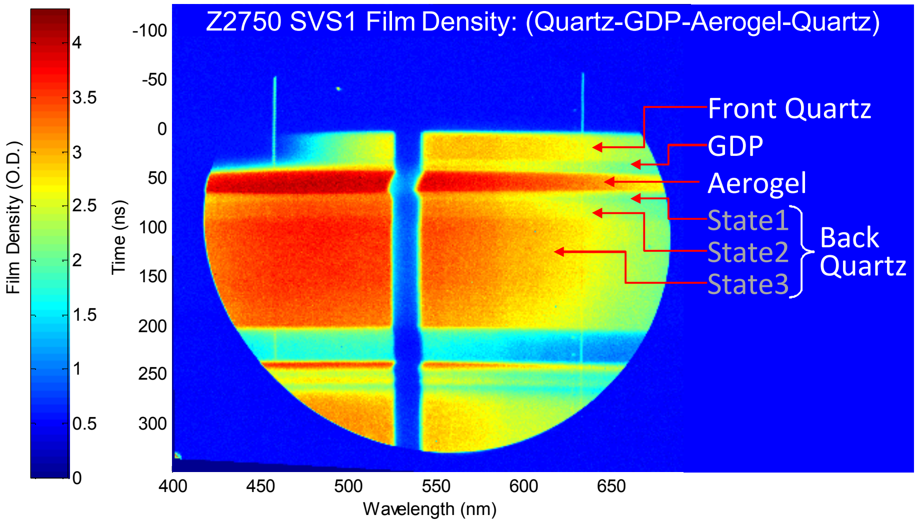
T. Ao's GDP data from Z, present work.



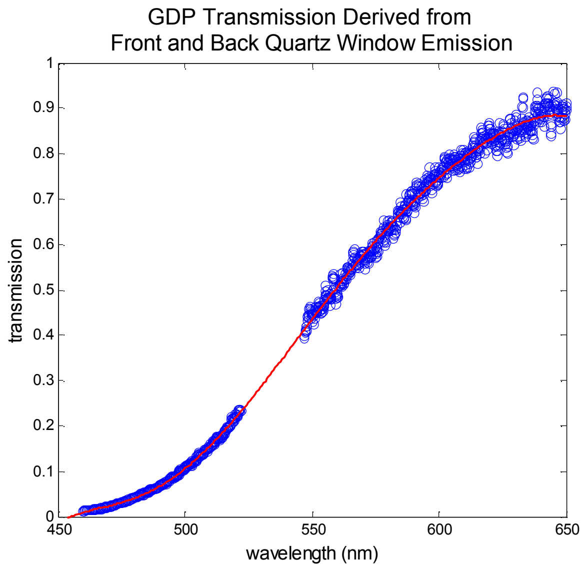
- **DFT agreement with Omega data (M. Barrios,LLNL) to 600 GPa**
- **Transit-time method on Z for thick samples (250 micron)**
- **Direct method on thin samples (180 micron): VISAR and PDV**
- **Improved manufacturing made the difference**

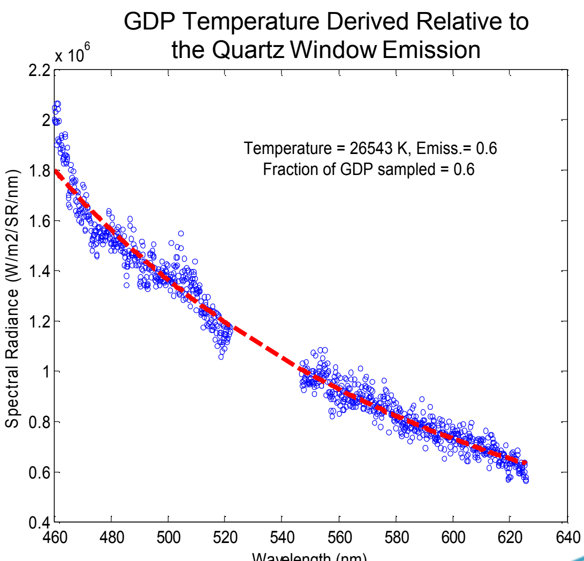
Streaked Pyrometry is an essential tool to measure the Temperature of GDP and other DMP samples and verify/support EOS calculations.

Glow Discharge Polymer (GDP) is used in the production of ICF capsules.



- The shock velocity in both the front and back quartz windows is measured by VISAR and PDV to be 21.5 km/s.
- Dividing the Front/Back quartz window emission yields the Transmission through the GDP & Aerogel layers and Fresnel interface reflections.
- The corresponding temperature is 41,000K.*
  **P.M. Celliers et.al., PRL 104, 184503 (2010)*
- The amplitude of the VISAR signal indicates that the GDP reflectivity and therefore the emissivity is similar to quartz .
- Correcting for the increased transmission as the shock wave propagates through the GDP yields the Spectral Radiance of the GDP which can be fit to a Planck Grey Body function yielding temperature: T = 26.5 $\pm$3 kK in this case.

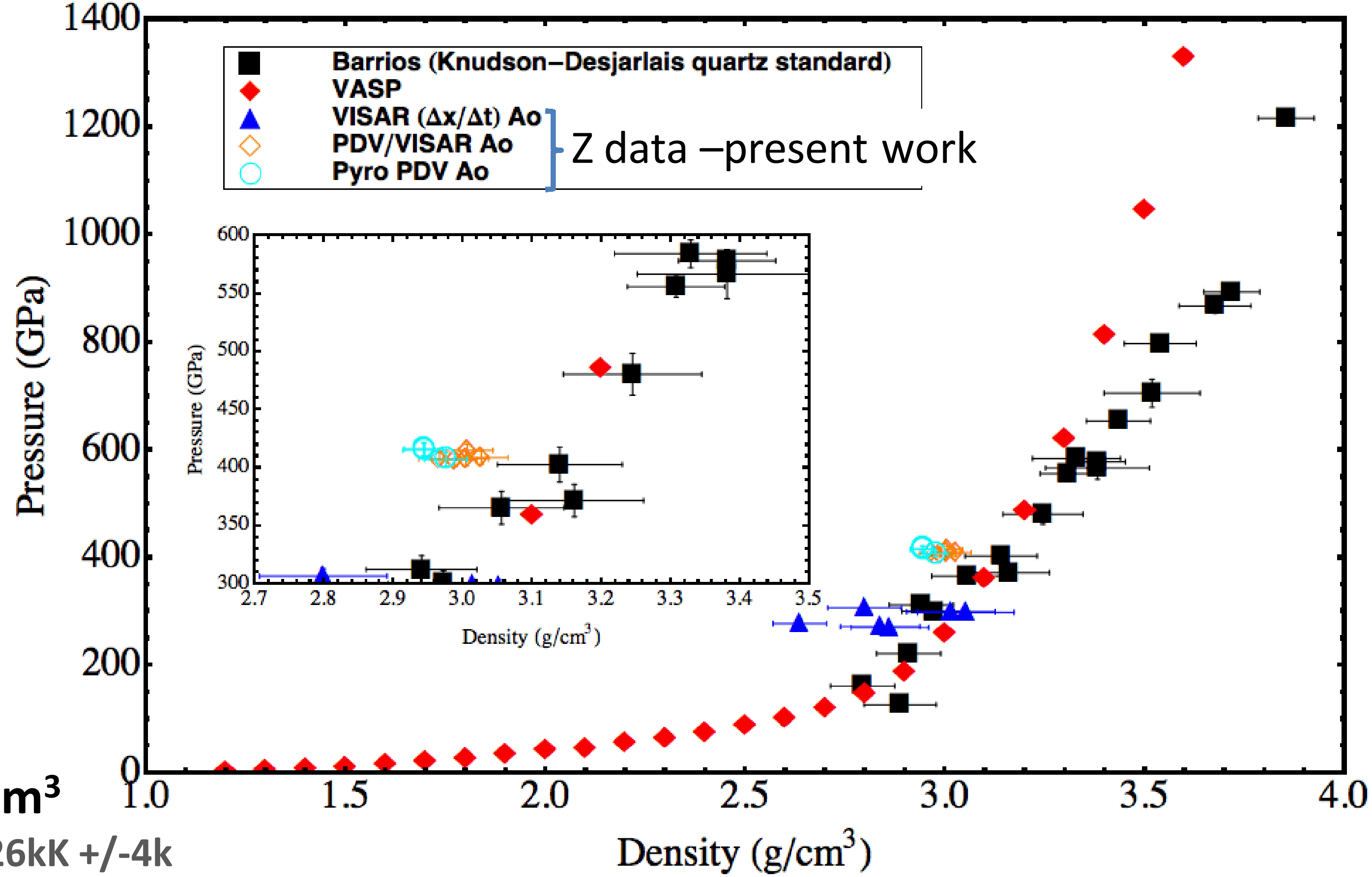




Dave Bliss

Intriguing differences between DFT/QMD simulations and new Z data in P & T

- **Experimental uncertainties**
  - The initial density (1.03 +/- 0.02 g/cm^3) is the leading density uncertainty
  - Pressure accuracy is about 1.5%
- **New Z data differ by 6% in density or 30% in pressure from LLNL/Omega and DFT**

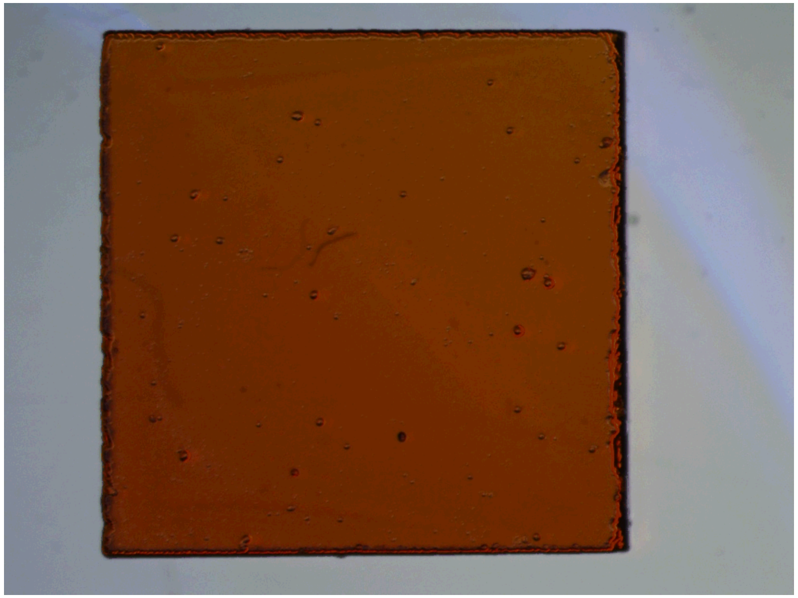


- **Shock state at 3 g/cm^3**
  - New Z: 400 GPa and 26kK +/-4k
  - DFT: 270 GPa and 15kK
- **DFT shows that a composition $CH_{1.5}$ would yield 26 kK/ 400 GPa**
- ***Is there uncertainty in chemical composition of GDP as shot?***

Looking for sources of the discrepancy with new Z data

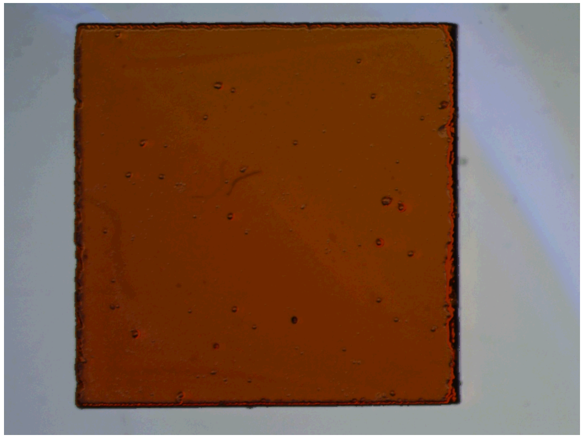
- **Initial state is a complex polymer with little known about the structure**
  - Significant residual strain
  - UV light can crosslink the polymer
  - UV cured epoxy is used in target assembly…

- **New samples will be used for a UV sensitivity study**
  - 10 pairs of GDP samples
  - Each pair of samples will be given a timed UV exposure
    - *One sample in a pair analyzed using Raman spectroscopy and mass spectroscopy*
    - *The other shot on Z*
    - *Objective is to quantify effect of UV exposure*



A mass spectroscopy analysis of remaining samples reveal variations in chemical content

	Date	Time	Carbon	Hydrogen
baked	5/22/15	9:48:05 AM	84.69%	10.06%
baked	5/22/15	10:29:38 AM	85.38%	10.14%
baked	5/22/15	11:10:02 AM	78.51%	9.11%
baked	5/22/15	11:21:40 AM	85.65%	10.03%
Average			**85.24%**	**10.08%**
unbaked	5/22/15	9:43:18 AM	64.56%	7.69%
unbaked	5/22/15	10:24:50 AM	69.67%	8.22%
unbaked	5/22/15	11:05:14 AM	68.78%	8.12%
unbaked	5/22/15	11:16:53 AM	67.32%	8.07%
unbaked	5/22/15	11:35:27 AM	67.94%	7.95%
unbaked	5/22/15	11:46:18 AM	61.73%	7.32%
unbaked	5/22/15	12:00:29 PM	65.60%	7.84%
Average			**66.51%**	**7.89%**



Translates to different composition range

- $CH_{1.41}$
- $CH_{1.36}$

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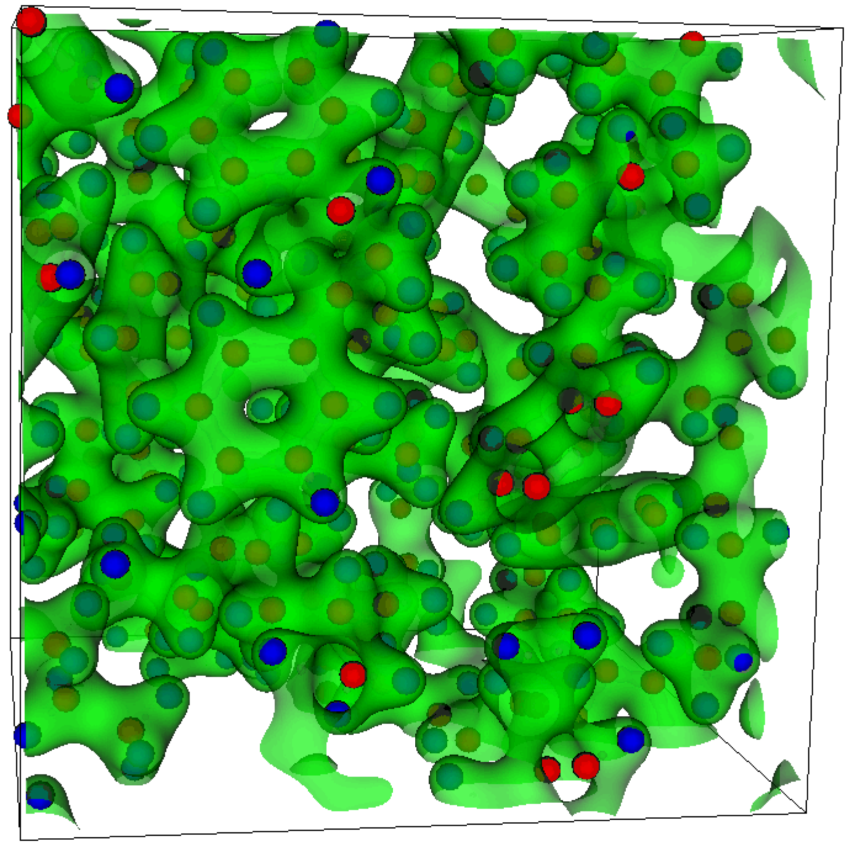
Is the calculated initial state the source of the discrepancy?

- **Final state is a dense fluid – no reason to doubt DFT/QMD – method validated for a range of systems**

- **We explore initial states in DFT/QMD simulations to bound energy differences and structures**
  – Varied stoichiometry from $CH_{1.39}$ to $CH_{1.51}$ (assembled by Keith Jones, Arizona State)
  – Branched, unbranched, aromatics, no aromatics

- **Calculated the Hugoniot at 300 GPa**
  – The difference in the Hugoniot based on C to H ratio is not enough to account for the difference between Z and Omega/VASP



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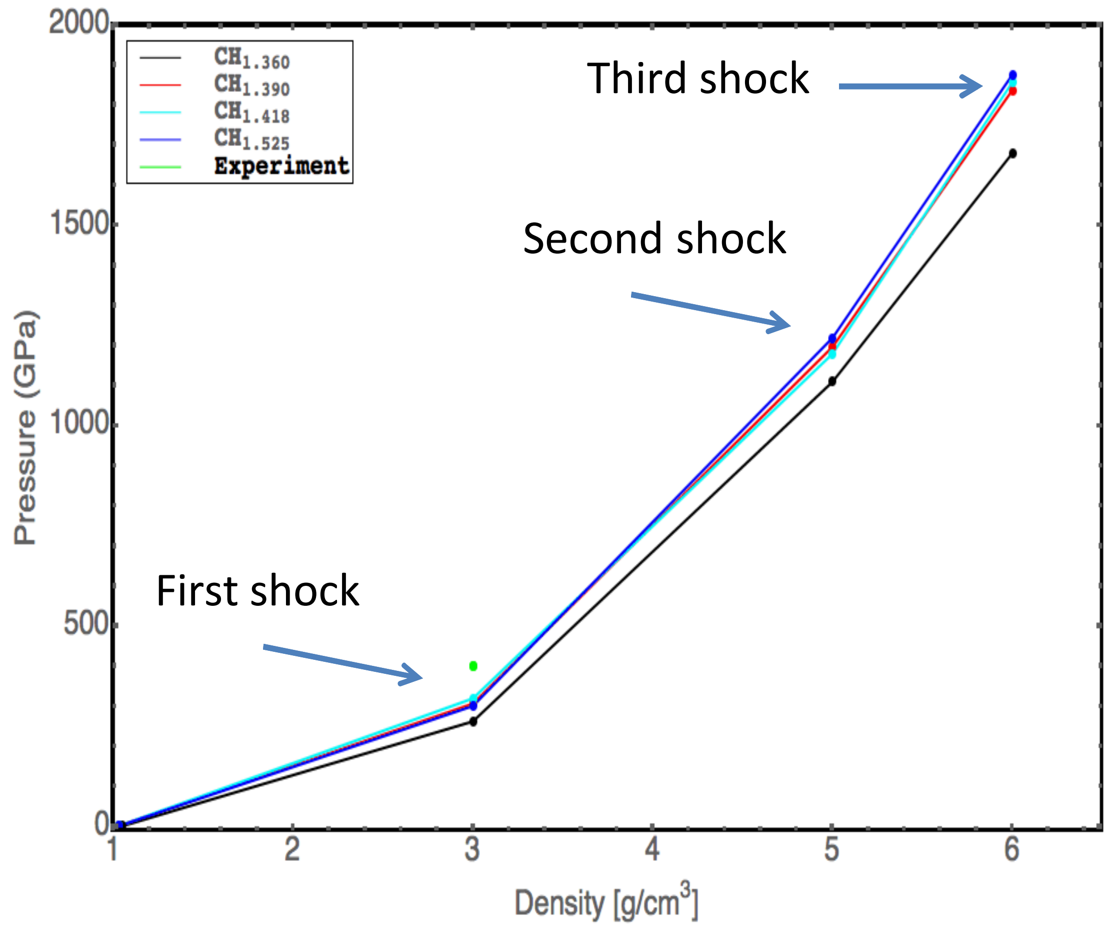
The chemical variations show systematic shifts in shock pressure – but does not recover the new Z data

- **The measured differences in composition cannot explain the difference to the new Z data**
- **The structural differences in double bonds and cross-linking shifts some, but not enough**
- **Even these small differences in composition lead to successive deviations over multiple shocks**



GDP is a challenging material for experiments and theory/simulations

- **Summary**
  - VASP and Omega (Barrios) data agree reasonably well up to 400 GPa
  - New Z data does NOT agree with with VASP/Omega
    - *For shock pressure and shock temperature*
  - Likely variations in stoichiometry and initial structure do not account for the differences
  - Using new Van der Waals exchange-correlation functional did not shift the Hugoniot enough to make a difference
- **Future Work**
  - Review/re-analyze Z experiments for systematic errors
  - Further explore if alternate initial states can be consistent with the experimental Hugoniot
  - UV effects on GDP and GDP Hugoniot
- **Acknowledgements**

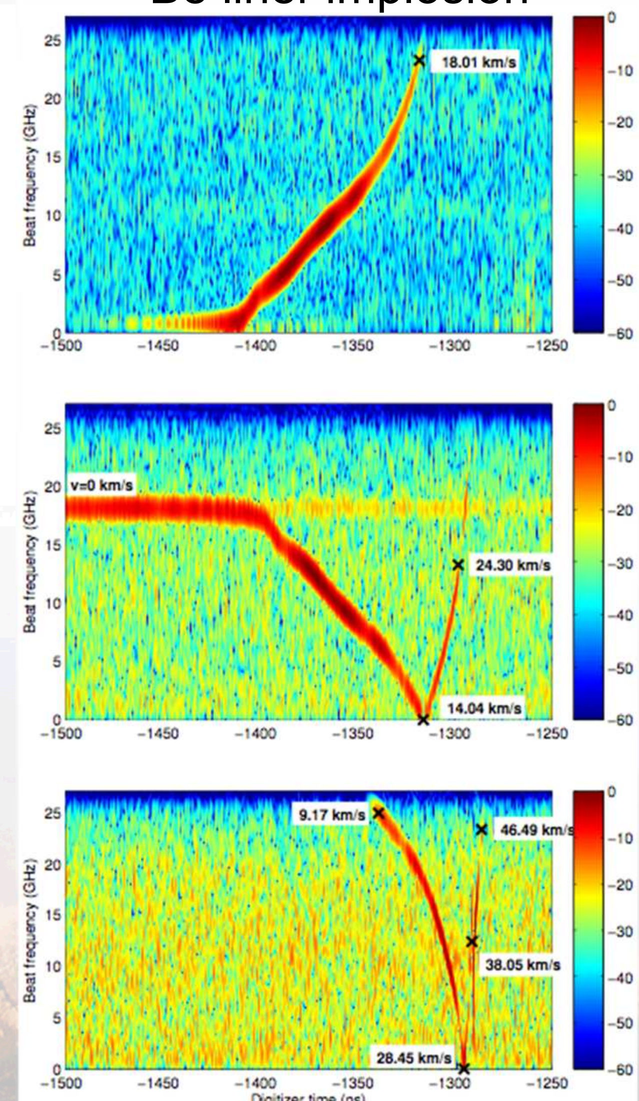
  - Sebastien Hamel and Loren Benedict for GDP structure and insights
  - Keith Jones for insights into GDP manufacture and stoichiometry
  - Sandia National Laboratories computing
  - Los Alamos National Laboratory computing

Photonic Doppler Velocimetry overview

- **Frequency-shifted PDV**
  - What is PDV and how is it different from VISAR?
  - How can be PDV be used to measure extreme velocities?
    - Dealing with finite electrical bandwidth

- **Examples**
  - Cylindrical implosion
    - Hollow and liquid-filled liners
  - Plate impact experiments

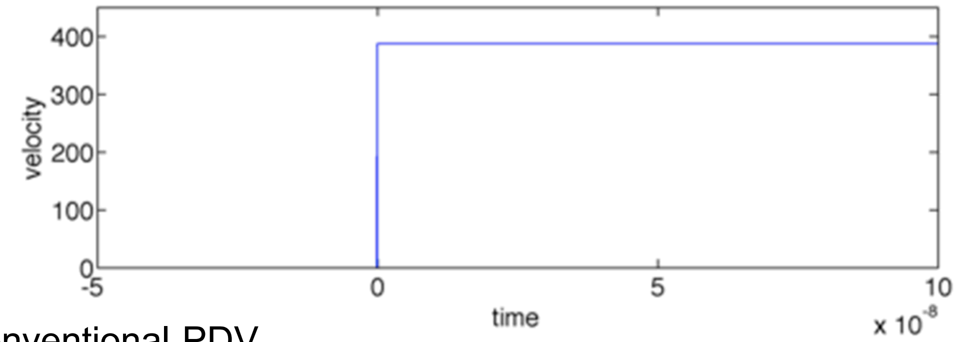
Be liner implosion



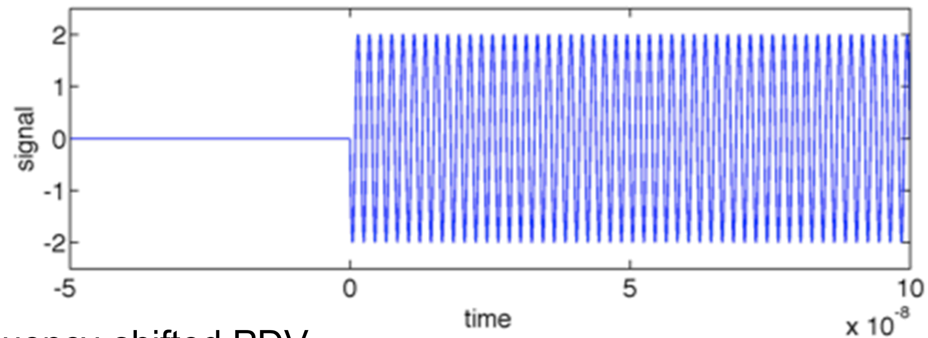
Differences between VISAR and PDV

- **VISAR mixes Doppler shifted light with a time-delayed version of itself**
  - Velocity changes correspond to fringe shifts
  - Typically visible light (532 nm)
- **PDV mixes Doppler shifted light with a reference source**
  - Conventional: single laser
  - Frequency-shifted: multiple lasers
  - Velocity changes correspond to frequency shifts
  - Infrared light (1550 nm)

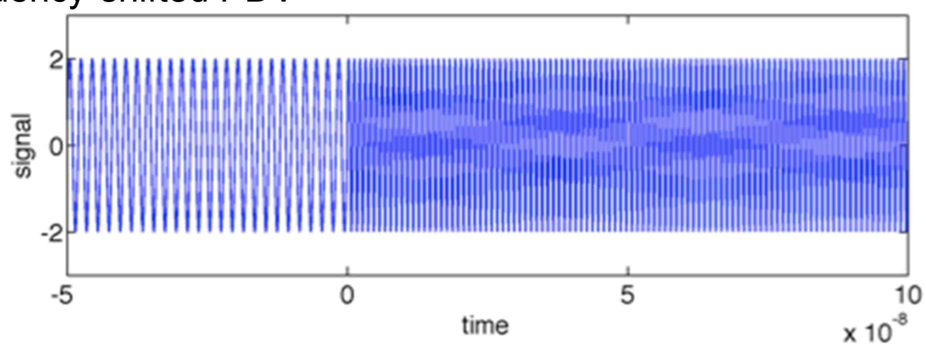
Velocity step



Conventional PDV



Frequency shifted PDV



Why bother with PDV?

- **Very simple to field**
  - Fiber-based, commercial components
- **Extremely compact**
- **Relatively low power requirements**
  - ~100 mW target power
  - ~1 mW reference power
  - Mostly class I hazard
- **Very robust to light variation**
  - >50 dB return variations are acceptable
- **No hardware time scale (etalon)**
  - Time resolution defined in the analysis
  - Can be optimized for different purposes (arrival time, etc.)
- **Tolerates multiple velocities (where VISAR loses contrast)**