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

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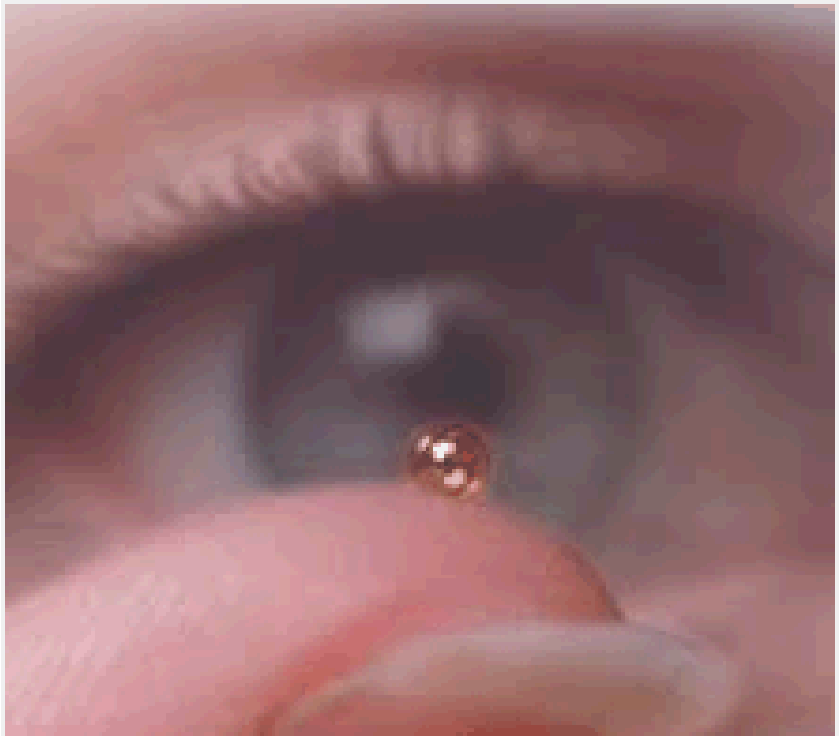
GDP fusion capsules

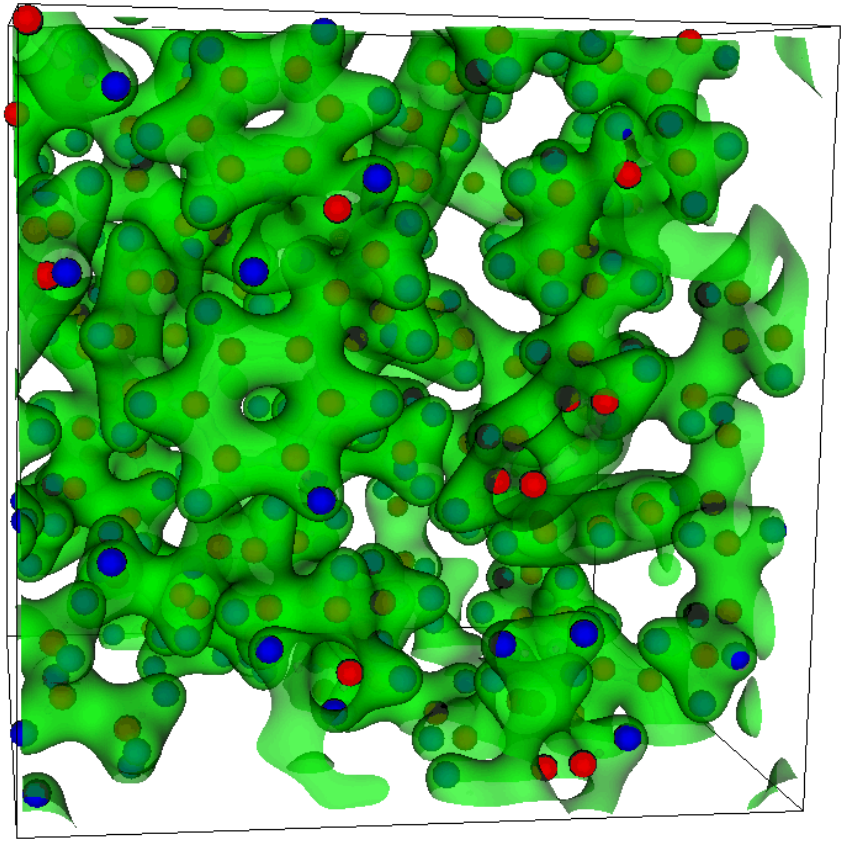
- **GDP is one of the possible ICF ablator materials**
- **Imploded using a series of convergent shocks**
- **Initial shock at approximately 3 Mbar**
  - **Just above complete dissociation**
  - **10% error bars too large**
- **VASP has a good track record with hydrocarbon material properties**



GDP fusion capsule for NIF

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

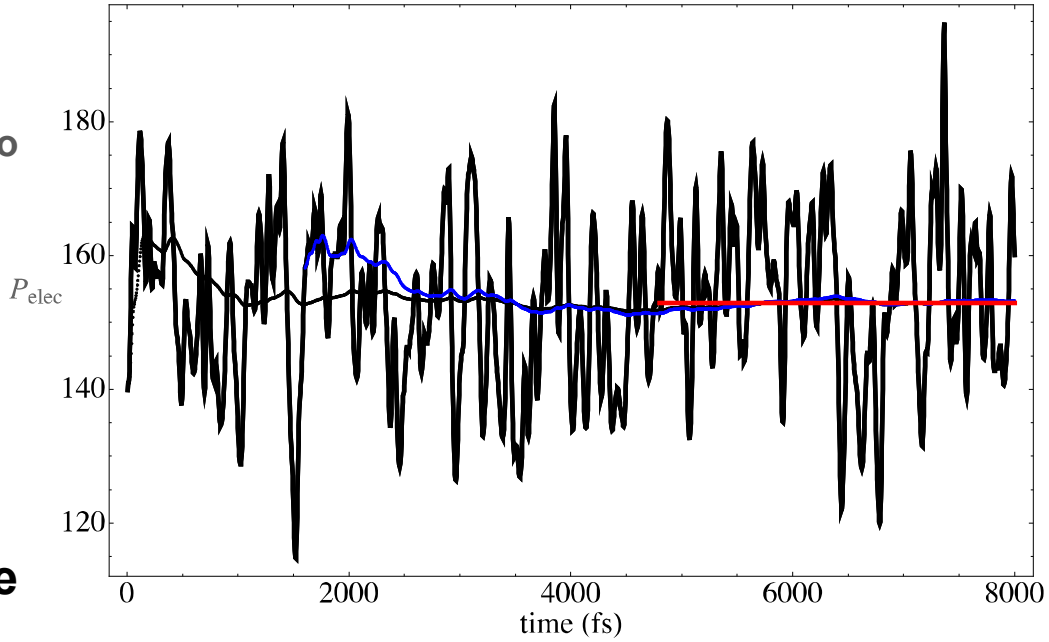
- **GDP is one of the possible ICF ablator materials**
- **First-principles simulations DFT**
  - **VASP – plane-wave code w PAW core-functions**
  - **Use of DFT codes simulating warm dense matter**
    - *M. P. Desjarlais Phys. Rev. B **68**, 064204(2003)*
  - **Great care in convergence**
    - *A. E. Mattsson et. al. Modeling and Simulation in Material Science and Engineering **13**, R1 (2005)*
- **Assemble reference system**
  - **272 Hydrogen, 200 Carbon**
  - **Equilibrate at constant temperature and volume.**
  - **Equilibrated for 3000+fs at 0.1 to 0.5 fs**
  - **AM05 potential**
  - **Standard deviation of energy and pressure <1%**
  - **Block averaging to reduce correlation**
  - **Atom positions courtesy of Sebastien Hamel (LLNL)**



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

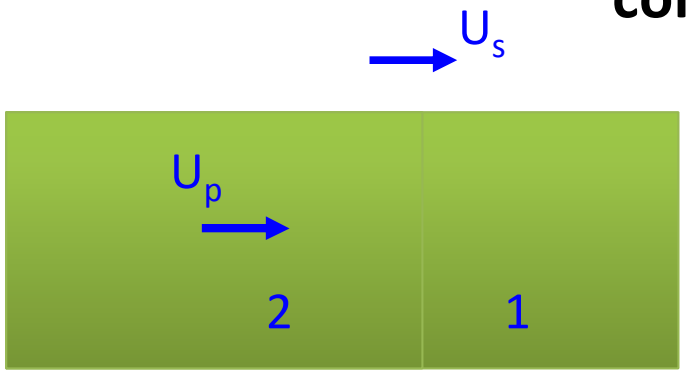
Molecular dynamics simulations to converge to a pressure and energy

- **Typically tens of ps**
- **Simulation Size**
  - **Larger number of atoms usually allow us to equilibrate in fewer time steps but take longer and more processors per time step**
  - **Higher probability of seeing stochastic events such as dissociation or melt at the correct density and temperature.**
  - **Smaller simulations become viable after melt.**
- **Large oscillations in pressure and energy but the mean will equilibrate to less than 1%. Often we try for <0.5% or less**
- **The mean pressure and energy are used to calculate the Hugoniot.**



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)



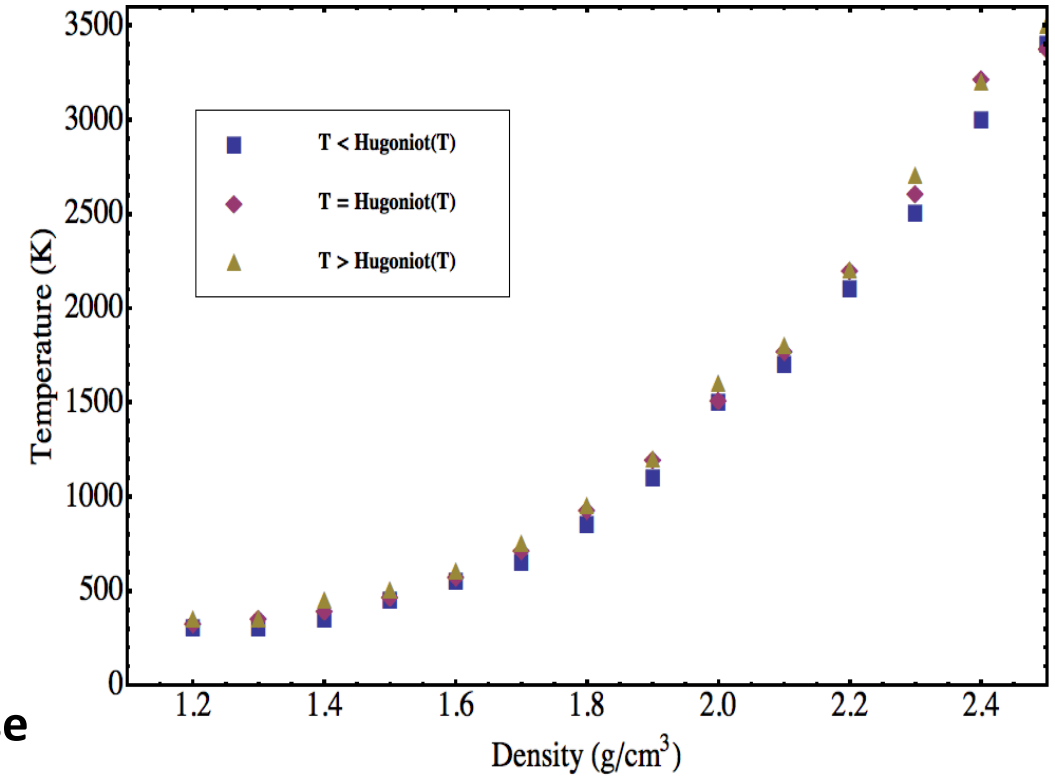
- E - internal energy
- P - pressure
- v – specific volume

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

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

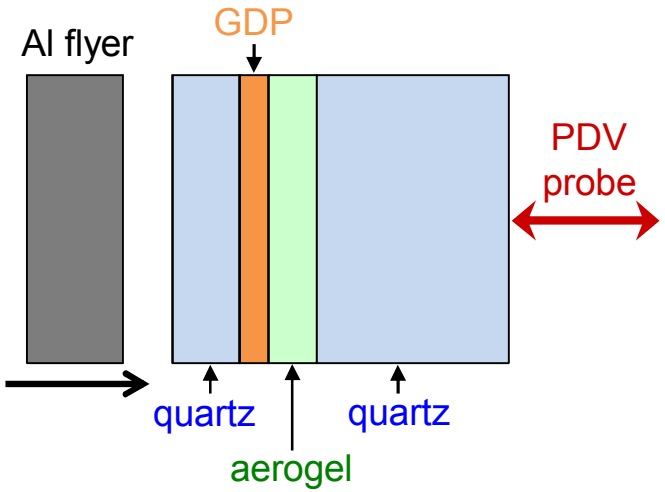
Use interpolation between two simulations to get actual Hugoniot value

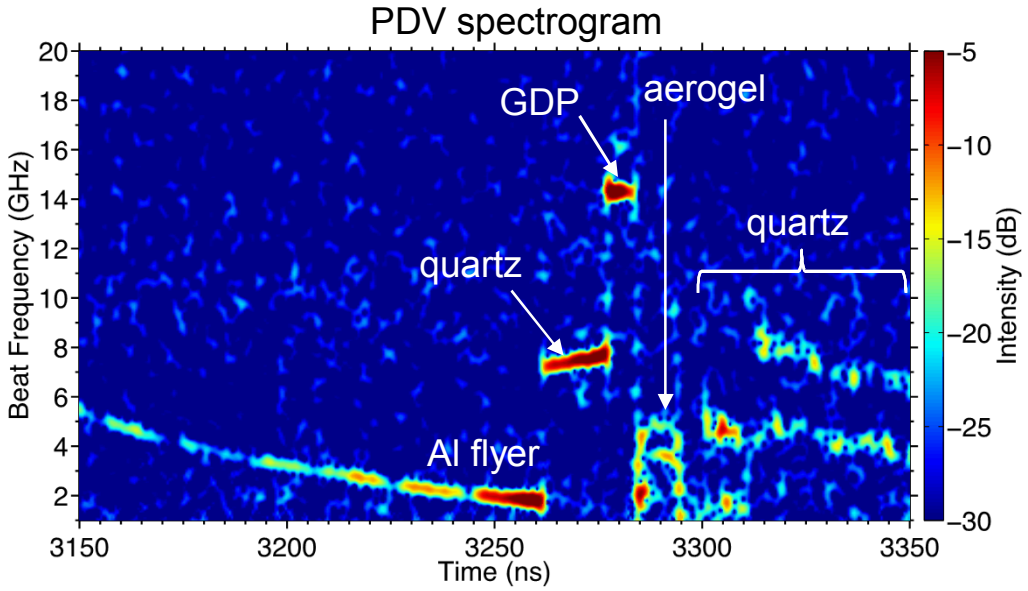
- Run 2 or more simulations at each volume/density.
- One with temperature such that energy/pressure are below the Hugoniot relation
- Second simulation with the temperature too high
- Interpolate between for actual Hugoniot point
- If close, extrapolation is is viable (must be checked on a case-by-case basis).
- Small density increases to avoid too much instantaneous compression.



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

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

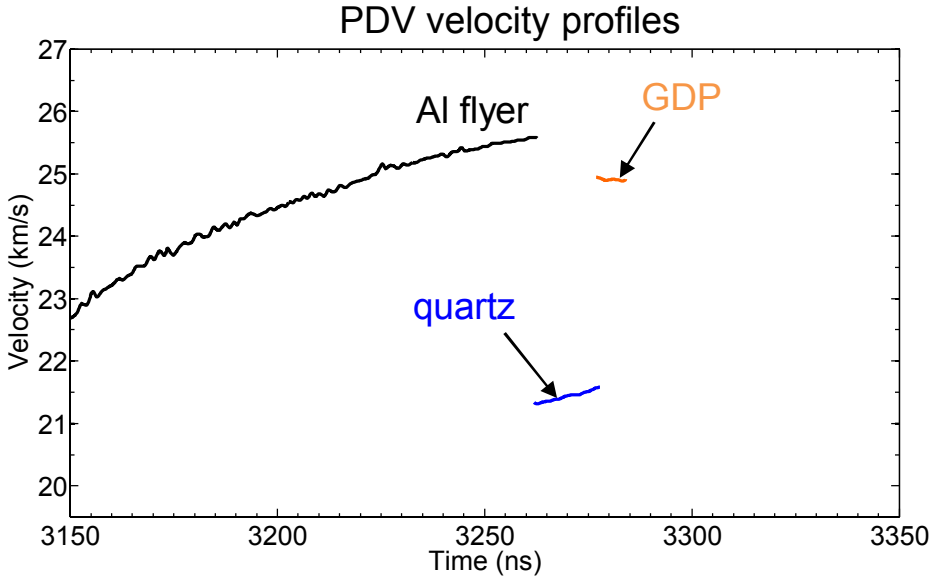




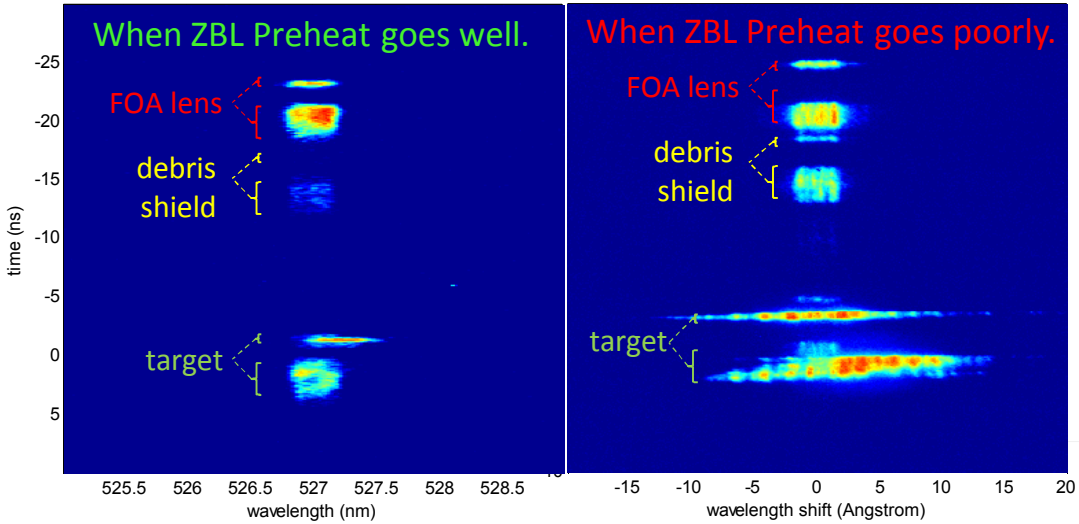
PDV mixes Doppler shifted target light with reference light

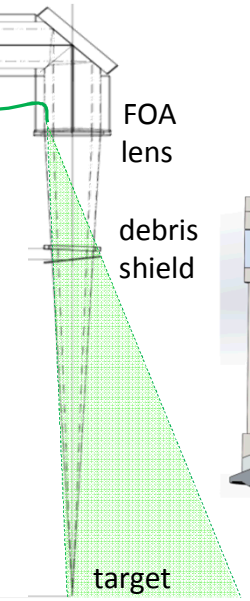
- Infrared light (1550 nm) transparent through GDP
- Velocity changes correspond to beat frequency shifts

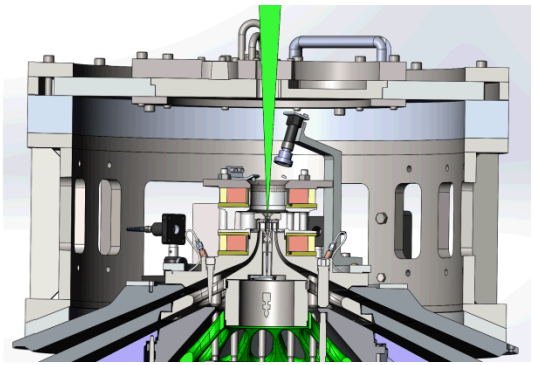
$$f_b = \left| \frac{2v}{\lambda_T} + c_0 \left(\frac{1}{\lambda_T} - \frac{1}{\lambda_R} \right) \right|$$

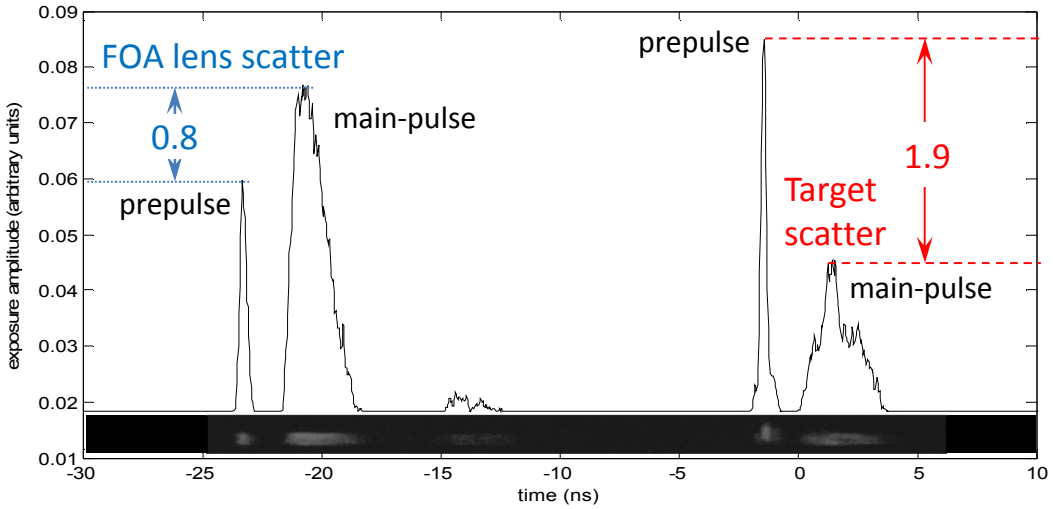


The SVS based ZBL backscatter diagnostic is a simple yet effective measurement of laser preheat conditions for MagLIF related shots.





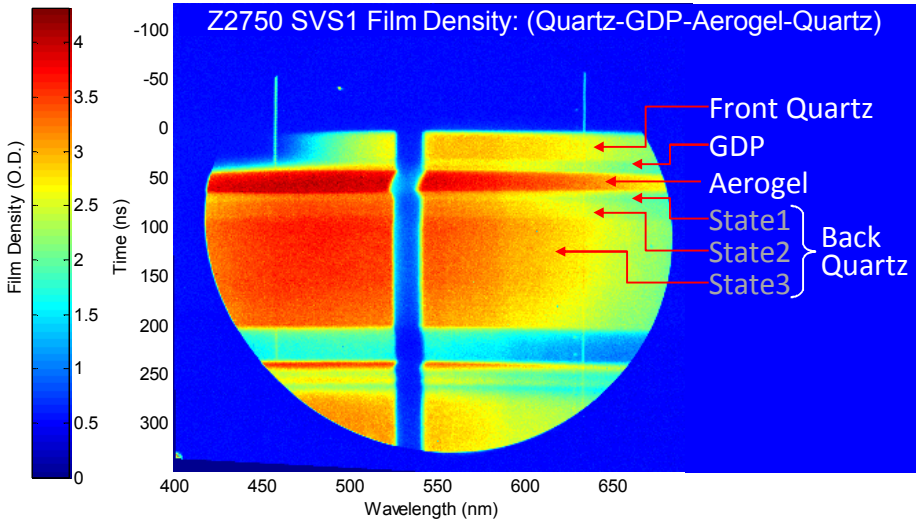




For a successful preheat, the scattered laser light at the target is preferentially scattered from the prepulse vs. the main-pulse indicating that the laser entry window is ablated away by the prepulse, thereby reducing the scatter of the main-pulse by the window. The design of the preheat laser pulse-shape appears to be successful.

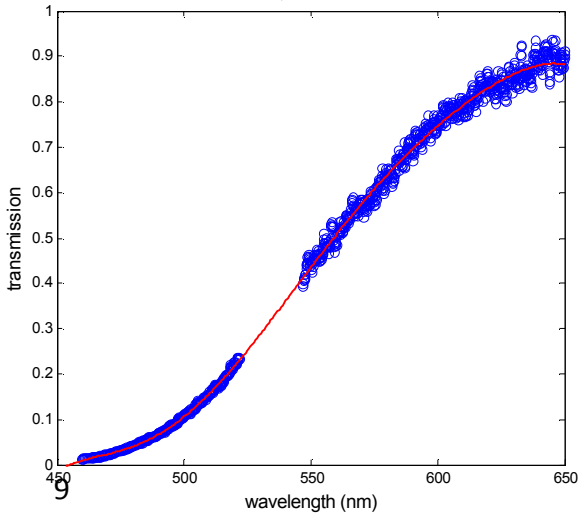
Streaked Pyrometry is an essential tool to measure the Temperature of GDP and other DMP samples and verify/support EOS calculations.

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

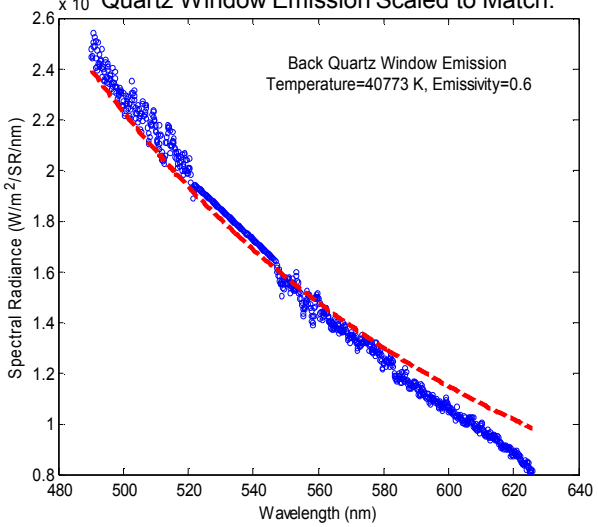


- The shock velocity in both the front and back quartz windows is measured by VISAR and PDV to be 21.5 km/s.
- Dividing the Front/Back quartz window emission yields the Transmission through the GDP & Aerogel layers and Fresnel interface reflections.
- The corresponding temperature is 41,000K.*
  **P.M. 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 ±3 kK in this case.

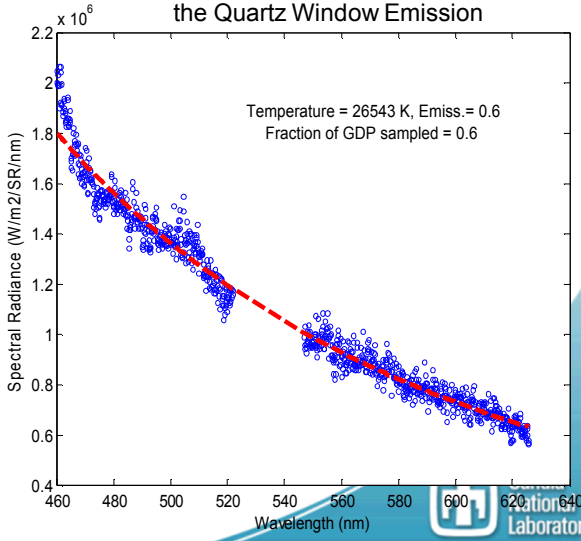
GDP Transmission Derived from Front and Back Quartz Window Emission



Quartz Temperature Derived from Shock Velocity.

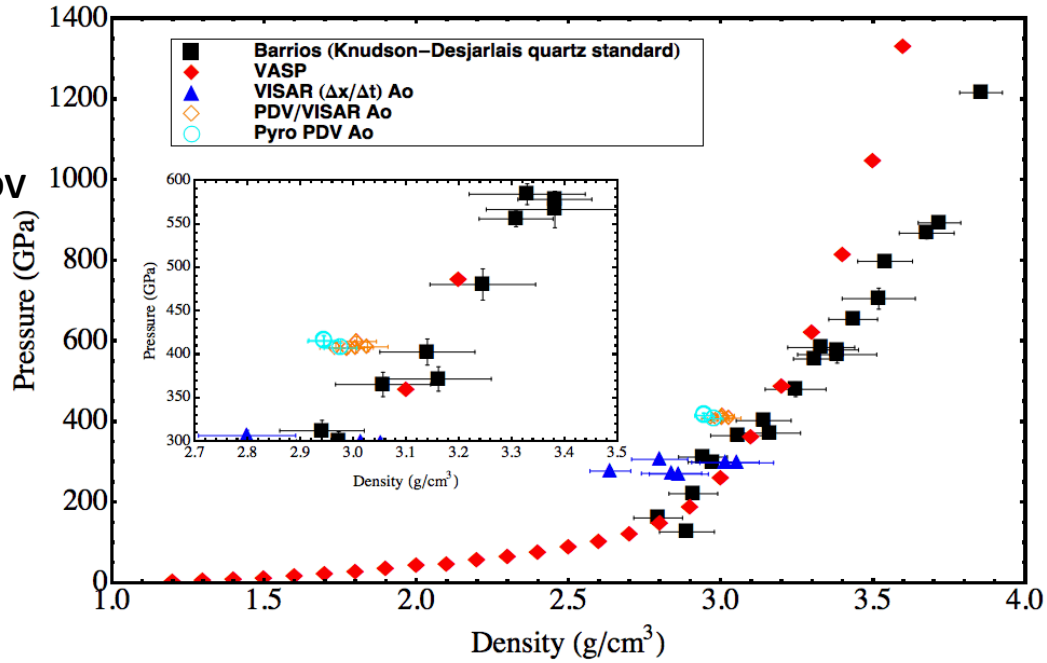


GDP Temperature Derived Relative to the Quartz Window Emission



VASP QMD simulations compared to experimental data

- VASP has good agreement with LLNL data at relevant pressure
- The large error bars on the VISAR data are from samples too thick to see through (250 micron)
- Thinner samples (180 micron) both VISAR and PDV can see through
- LLNL/VASP differ from Z data by 6% in density or 25% in pressure
- Streaked pyrometry indicates 26kK while VASP shows 15kK at 3Mbar
- Analysis of remaining GDP samples (by weight)
  - Baked/Pyro=C 85%, H 10%, N 0.9%, O_2 4%
  - UnBaked=C 69%, H 8%, N 0.25%, O_2 24%
  - VASP=C 89.7%, H 10.3%
- Using the above stoichiometry at 26kK, pressures are between 2.7 to 3.3 Mbar
- Still investigating possible reasons for discrepancy



Barrios data taken from Knudson and Desjarlais, PRB 88, 184107 (2013)

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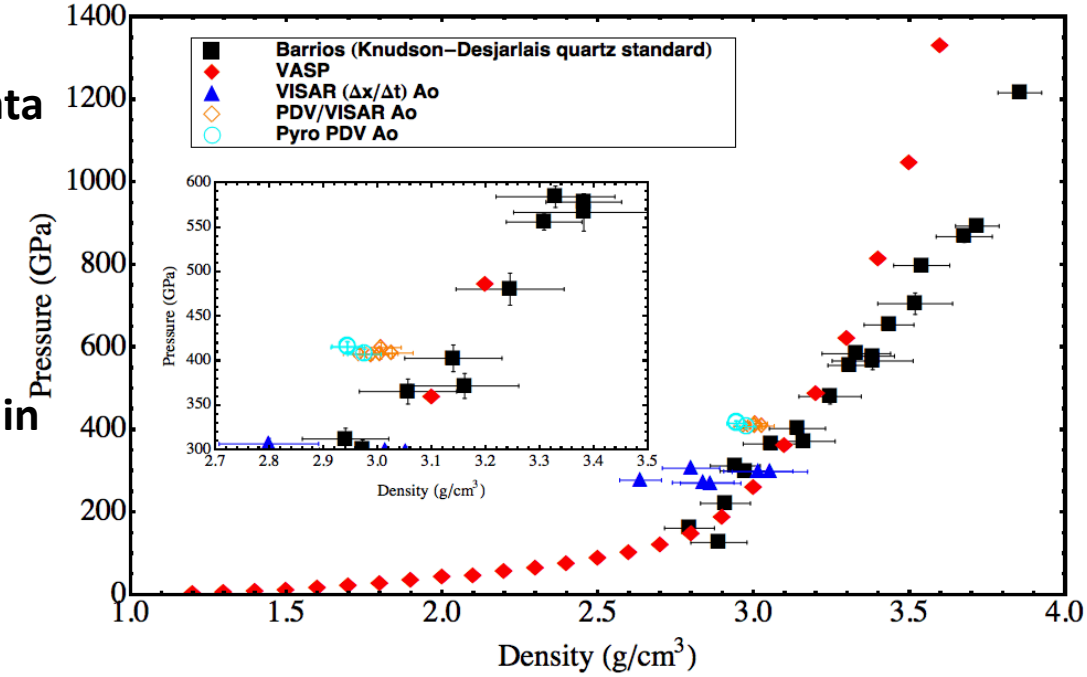
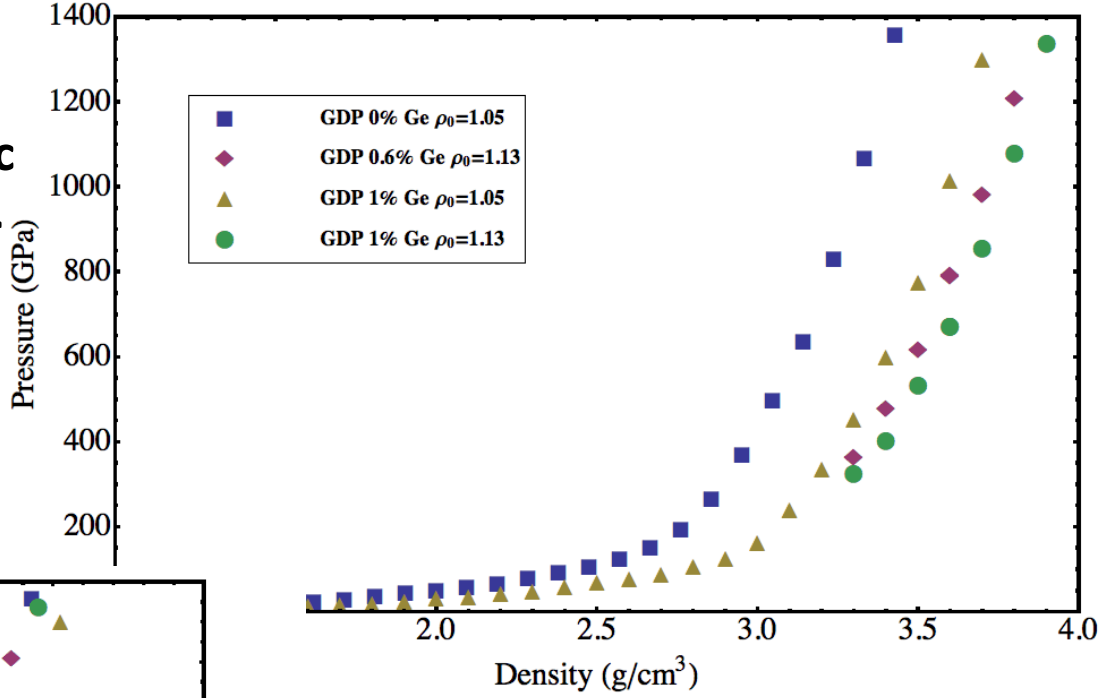
- VASP has good agreement with LLNL data at relevant pressure
- The large error bars on the VISAR data are from samples too thick to see through (250 micron)
- Thinner samples (180 micron) both VISAR and PDV can see through
- LLNL/VASP differ from Z data by 6% in density and 25% in pressure
- Z data has 2% error bars in density (primarily from reference density of 1.03 +/-0.02)
- Incorrect reference structure
- Adsorption of other elements (water)
- Both causing initial density to be very different.

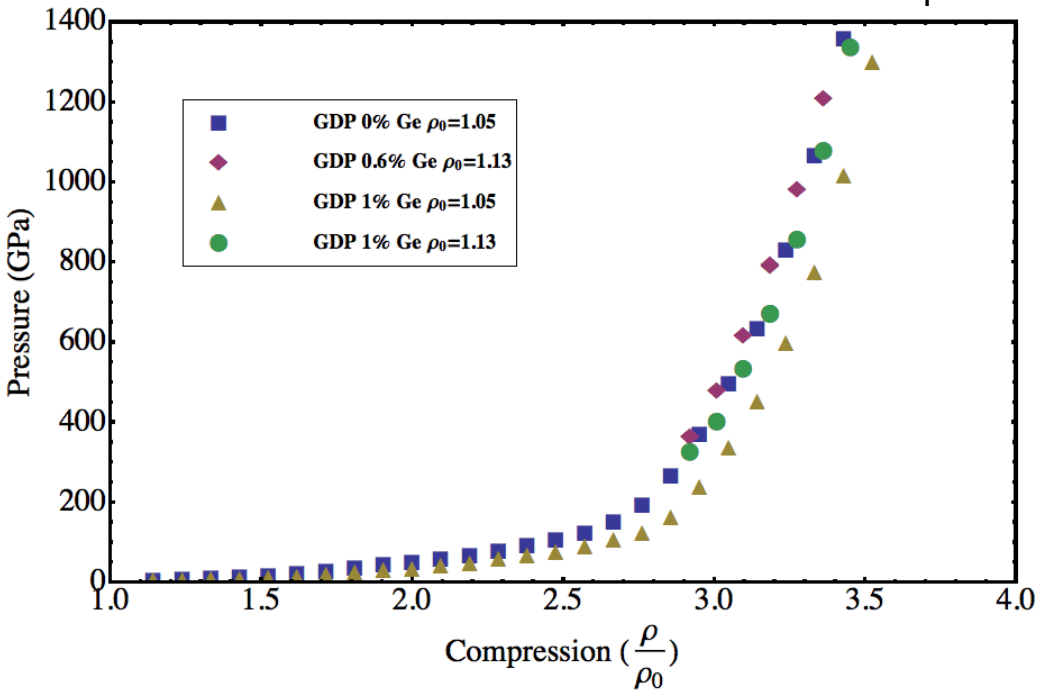


Barrios data taken from Knudson and Desjarlais, PRB 88, 184107 (2013)

GDP with germanium dopant

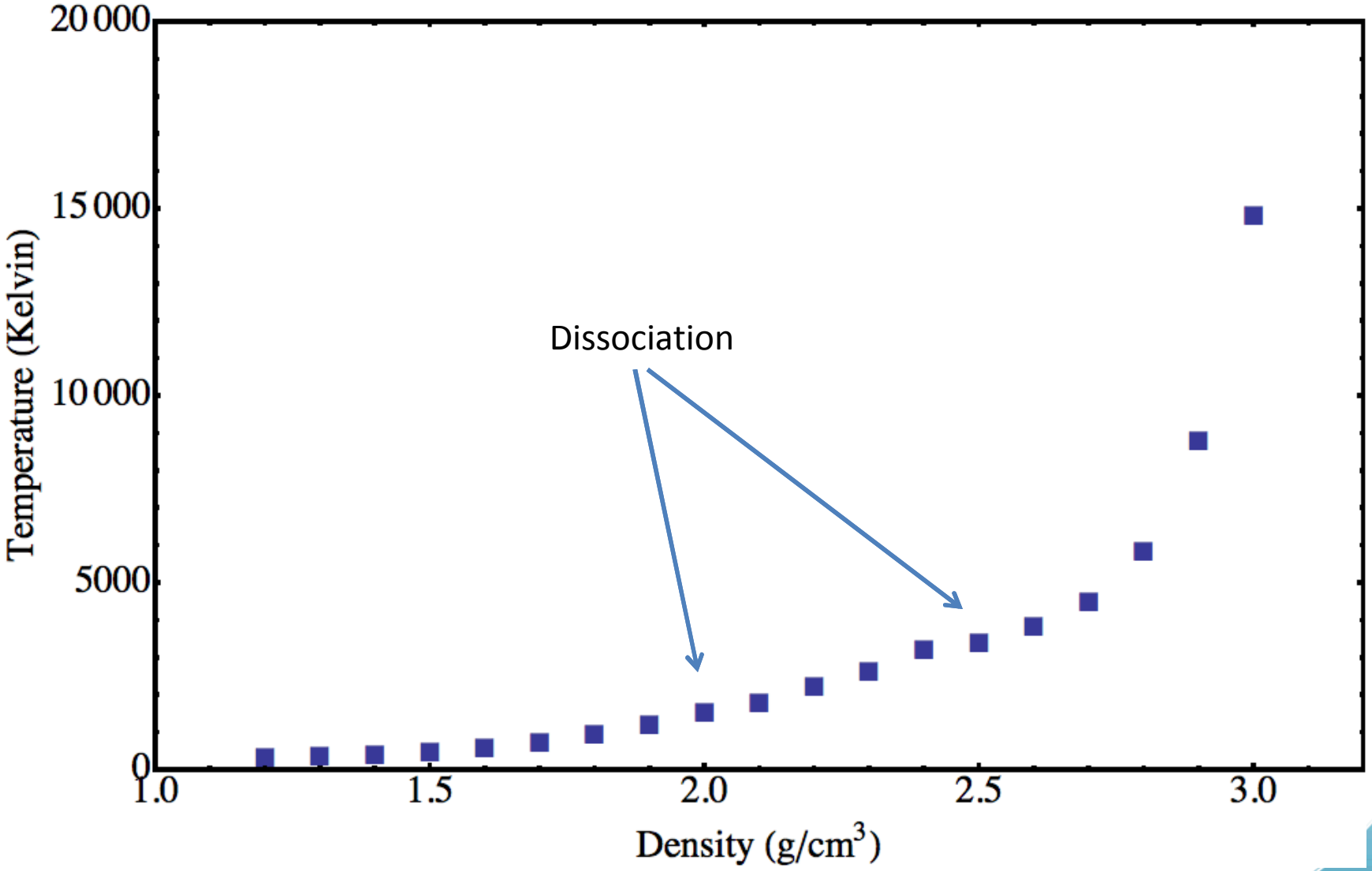
- Ge dopant at 0%, 0.6%, and 1%
- Density at 1.05 g/cc and 1.13 g/cc
- In some cases, substituted Ge for carbon and others just randomly placed Ge in the simulations
- Method of Ge placement made minor difference





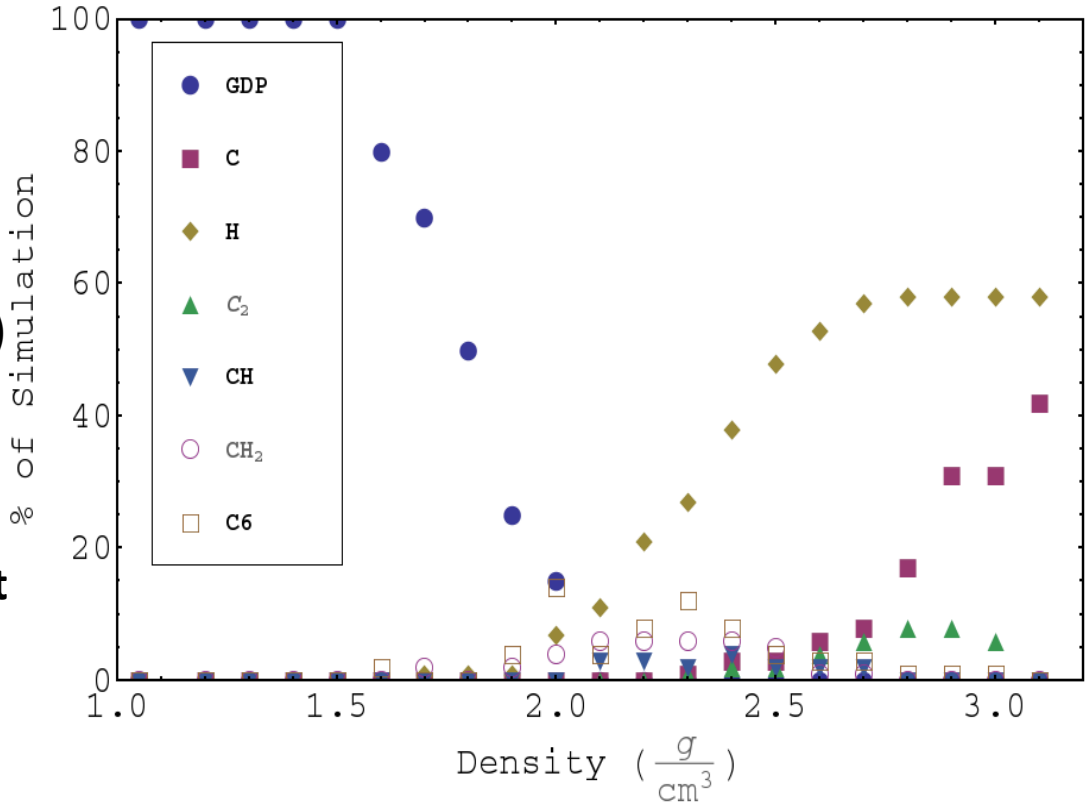
- At 1.05 g/cc, Ge significantly softens Hugoniot
- Keeping initial volume and adding germanium such that density increased to 1.13 g/cc shows compression ratio is similar but different enough to warrant a new EOS

Inflection in the Hugoniot is dissociation



Confirm shoulder in Hugoniot is from dissociation or melt by tracking "bonds"

- **Set bond lengths**
  - Found in reference simulation
  - If using experimental data, add 5% to 10% for atom vibration
- **Atoms stay within bond distance for Δt (5 carbon vibrations ~90fs)**
- **Simulation long enough for atoms to move apart if not bonded**
- **Run for 10s of picoseconds to get good statistics in transient regime**
- **Because of the variability of transient species, tend to only plot marker species**



Conclusions

- **Summary**
  - **Equilibrate simulation to steady state**
  - **Methods to calculate Hugoniot**
  - **Compare to experimental data**
    - *Good match to lower pressure data*
    - *Z error bars much too large*
  - **Compare to other simulations with germanium dopant**
    - *Germanium softens the Hugoniot*
    - *Need a new EOS for each dopant level*
  - **The inflection in the Hugoniot from dissociation**
  - **Did not analyze dissociation for doped simulations**
- **Future Work**
  - **Finish analyzing current Z experimental data**
  - **Improve experimental techniques (PdV at 1532nm)**
- **Acknowledgements**
  - **Sebastien Hamel and Loren Benedict for the GDP atom positions and insights**
  - **Sandia National Labs computing**
  - **Los Alamos National Labs computing**