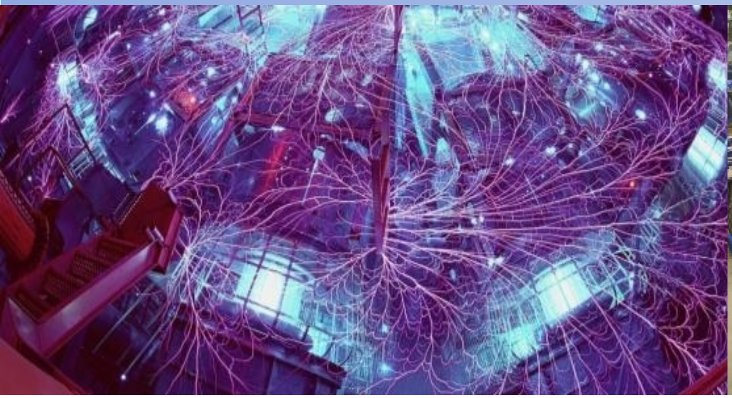
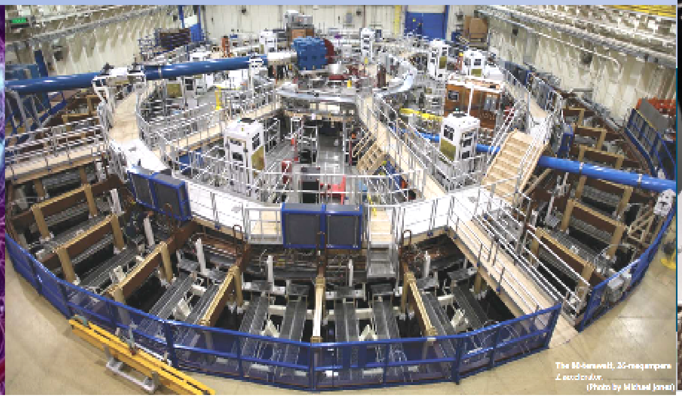
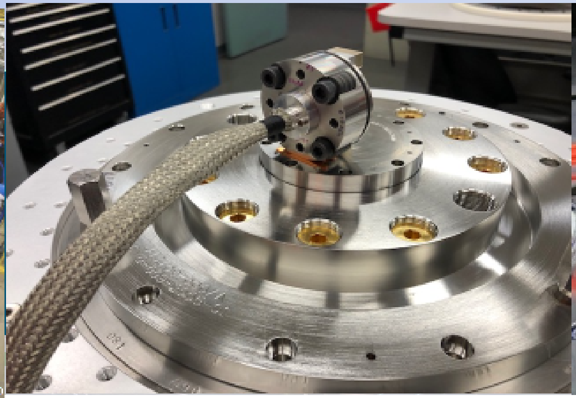
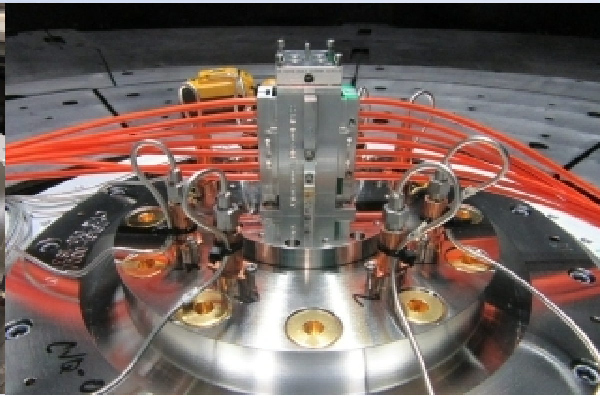
SAND2020-11768PE

Materials investigation at high pressure using dynamic compression

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

Sakun Duwal
Sandia National Laboratories
WSU Chemistry Departmental Seminar
sduwal@sandia.gov

11/02/2020

Sandia National Labs is managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a subsidiary of Honeywell International, Inc., for the U.S Dept. of Energy's National Nuclear Security Administration under contract DE-NA0003525.

SAND2020-XXXX

Abstract

Studying materials at extreme conditions is not only relevant to nuclear weapon conditions, but also to planetary interiors. Highly accurate equation of state data at extreme conditions provides a solid basis for models that enable predictive simulation of material properties. In this talk, I will cover the investigation of two different material categories at extreme conditions: pre-compressed H2-He gas-mixtures and solid TiO2 single crystals. Studying pre-compressed samples of H_2-He mixtures under dynamic loading allows us to access unique P-T states off of the principal Hugoniot. The understanding of material behavior at these P-T states is crucial to comprehend the origin and evolution of gas planets. Furthermore, the validation of mixture models has been a long-standing topic of interest with numerous conflicting theoretical results, particularly for non-ideal hydrogen systems. we present our experimental results on the pre-compressed samples of H_2-He mixtures using pulsed-power-driven flyer plates at the Z-facility, and impact studies on the two-stage light gas gun.

The high-pressure response of titanium dioxide (TiO2) is of interest because of its numerous industrial applications and its structural similarities to silica (SiO2). We used three platforms—Sandia's Z machine, Omega Laser Facility, and density-functional theory-based quantum molecular dynamics (QMD) simulations—to study the equation of state (EOS) of TiO2 at extreme conditions. We used magnetically accelerated flyer plates at Sandia to measure Hugoniot of TiO2 up to pressures of 855 GPa. We used a laser-driven shock wave at Omega to measure the shock temperature in TiO2. Our Z data show that rutile TiO2 reaches 2.2-fold compression at a pressure of 855 GPa and Omega data show that TiO2 is a reflecting liquid above 230 GPa.

The Group

Group 1640 - High Energy Density Material Physics

Thomas Mattson – Senior Manager

Department 1641

HEDP Theory

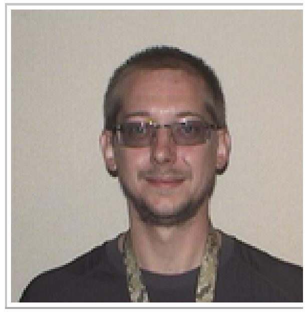
Luke Shulenburger - Manager

- Theory supporting ICF and DMP

Department 1646

Dynamic Material Properties

Chris Seagle - Manager



- Equation of States
- Phase boundaries
- Gas-guns
- Z-Machines

Department 1647

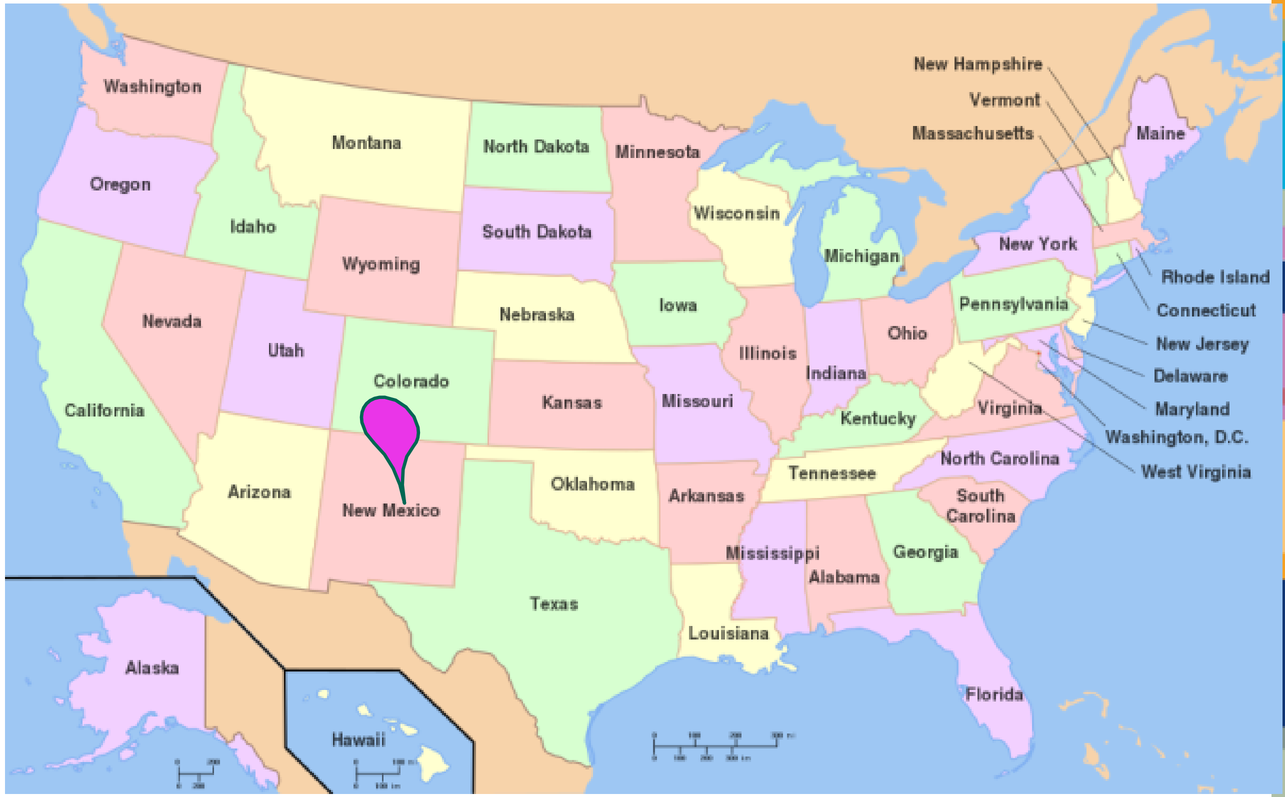
Solid Dynamics Experiments

Gordon Leifeste - Manager

- Gas-guns
- Thor pulsed power

Sandia National Laboratories

- One of three National Nuclear Security Administration (NNSA) lab
- Located in central New Mexico in Kirtland Air Force Base in Albuquerque
- Population: 11, 500
- Area: 8, 699 acres
- Budget: $ 3.6 billion



The Big Picture

- Sandia studies material dynamics as part of the Stockpile Stewardship Program (SSP)
- The science-based SSP aims to maintain confidence in the safety, security, and reliability of the U.S. nuclear stockpile without the use of nuclear testing
- The program has three main thrusts: inspection, manufacturing, and **science**
- Scientific areas of study are
  - Hydrodynamics
  - **Materials properties**
  - **High energy-density physics**
  - Micro-systems engineering
  - Computational capabilities
- Sandia works with other national labs to advance experimental techniques in material dynamics and apply these to materials of interest

www.energy.gov/nnsa/missions/maintaining-stockpile

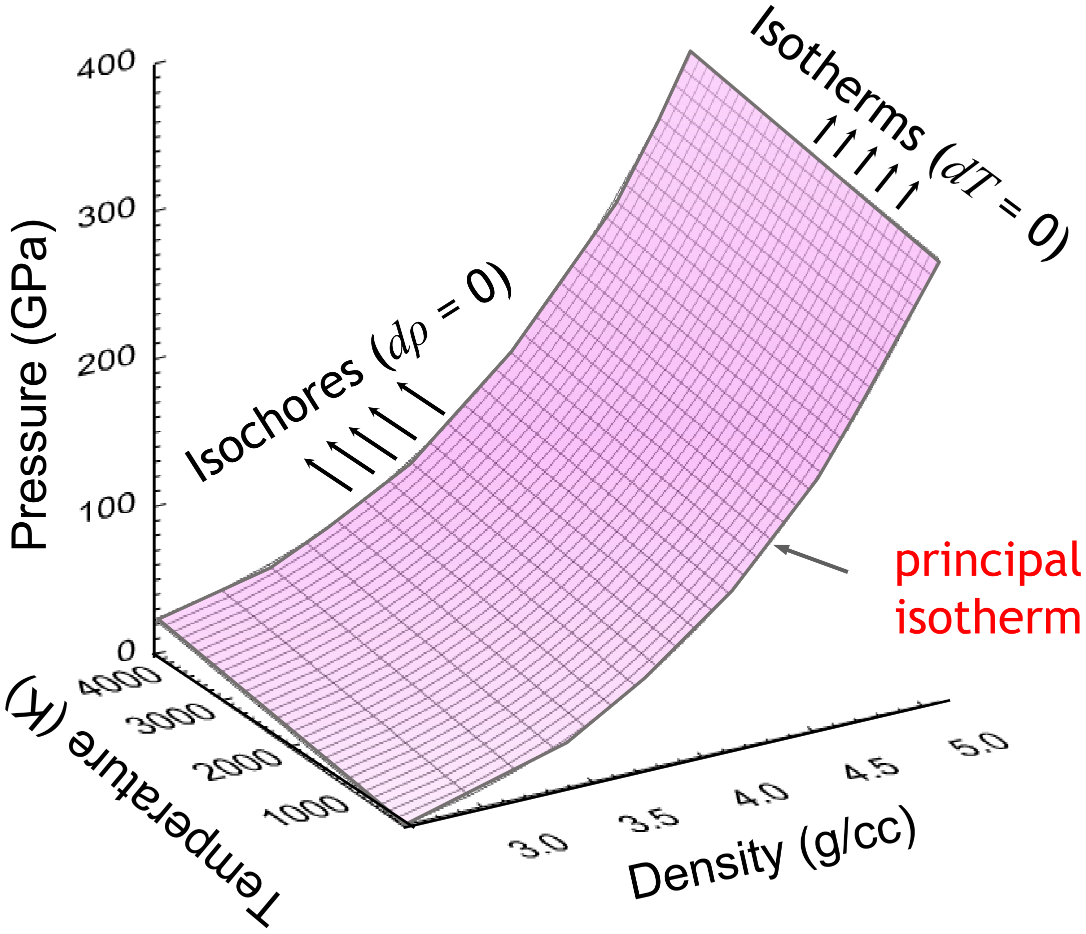
J.-P. Davis

Outline

- Equation of State Measurement → Why?
- Sandia's capabilities: Z, Gas gun and Thor to measure EOS
- STAR, Z pre-compression experiments
- Data: H_2-He and TiO_2
- Summary

Equation of State (EOS)

Equation of State: Relation between Temperature (T), Pressure (P), Volume (V)/Density, Internal Energy



J.-P. Davis

EOS data to predict material properties

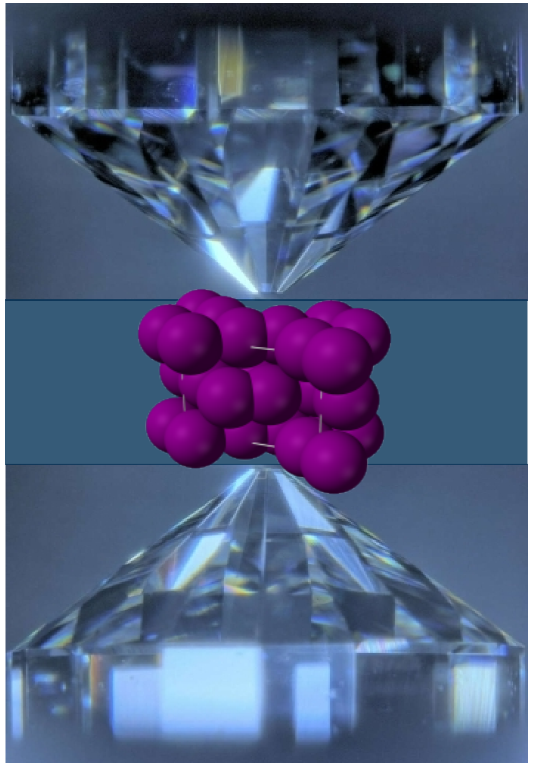
PV=*n*RT

Ideal gas law is not that ideal!

- At high pressure and at low temperature, real gases deviate from ideal gas;
- Ideal gas law and the EOS need to be modified:
  - Isothermal EOS: At constant T
  - Isochores EOS: At constant density

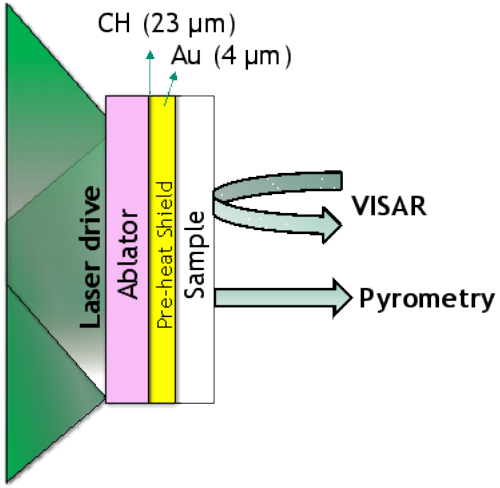
Ways to generate High Pressure

Static High Pressure

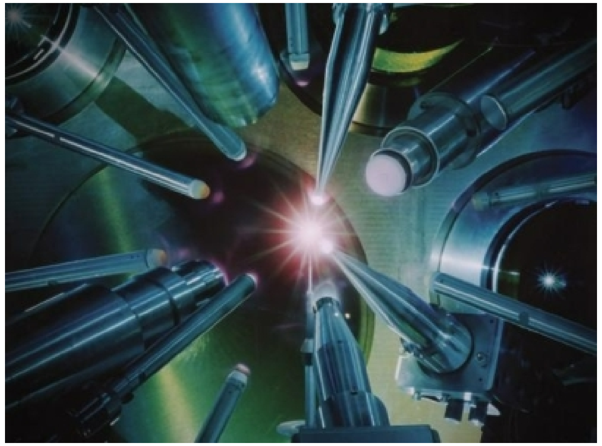


Diamond Anvil Cell
- P achieved 200 GPa
- Unlimited time

Dynamic Compression



Laser Drive:



Omega

NIF

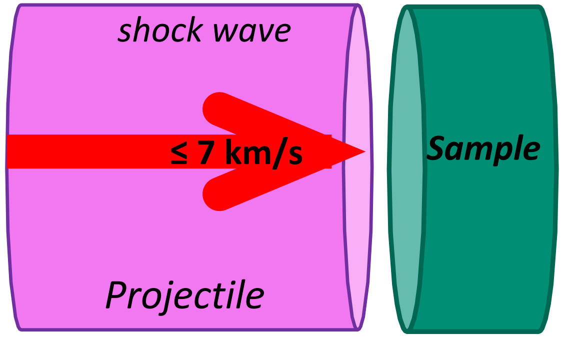
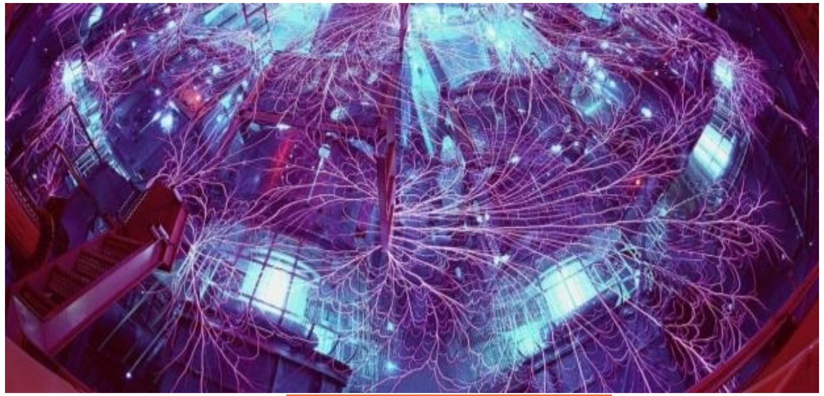


Plate Impact: Gas-guns

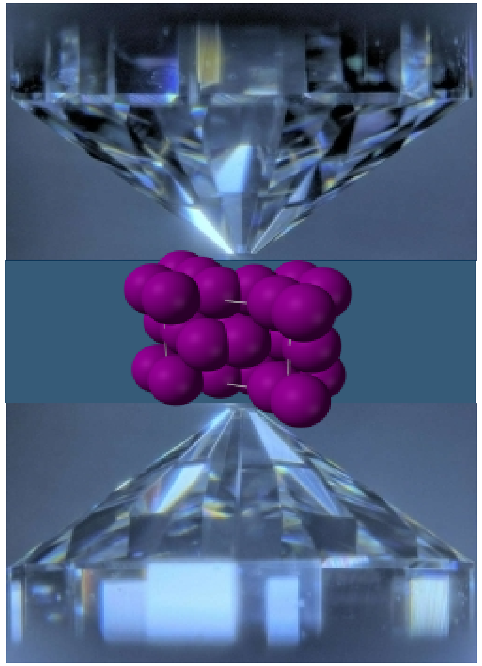


Z-Machine

- P achieved >1000 GPa
- Experiment time: several ns

Ways to generate High Pressure

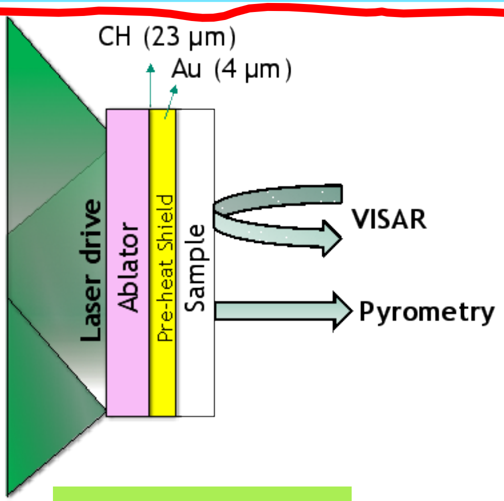
Static High Pressure



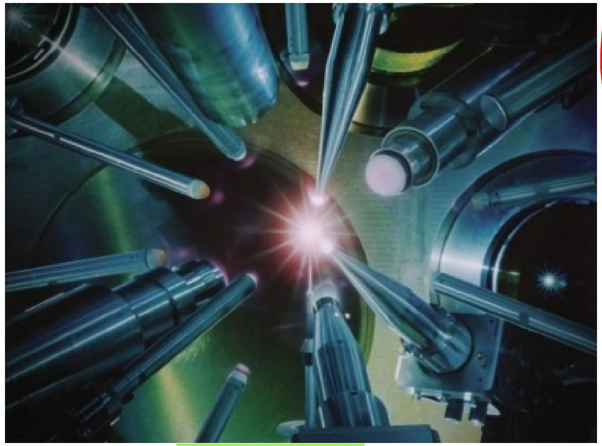
Diamond Anvil Cell

- P achieved 200 GPa
- Unlimited time

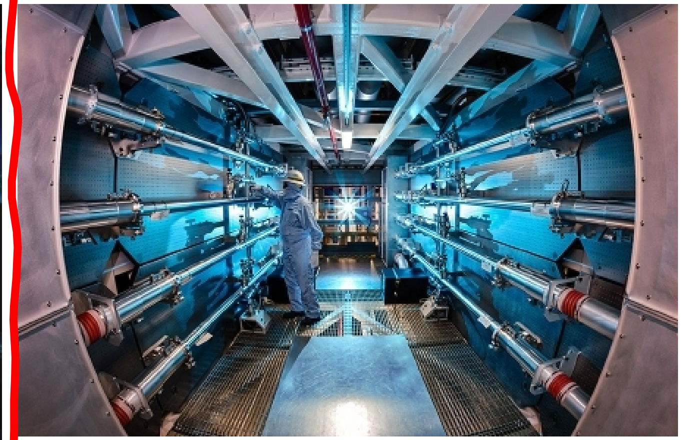
Dynamic Compression



Laser Drive:



Omega



NIF

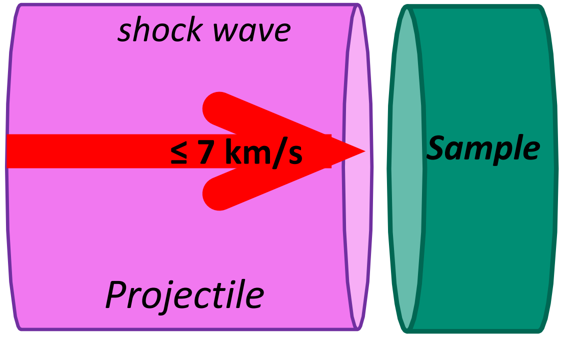
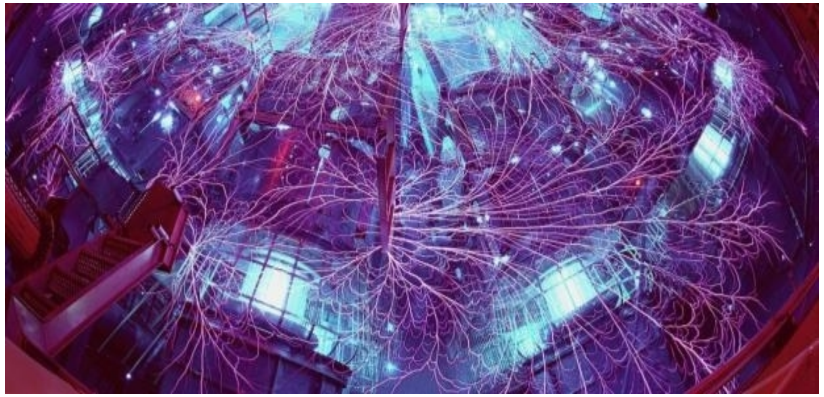


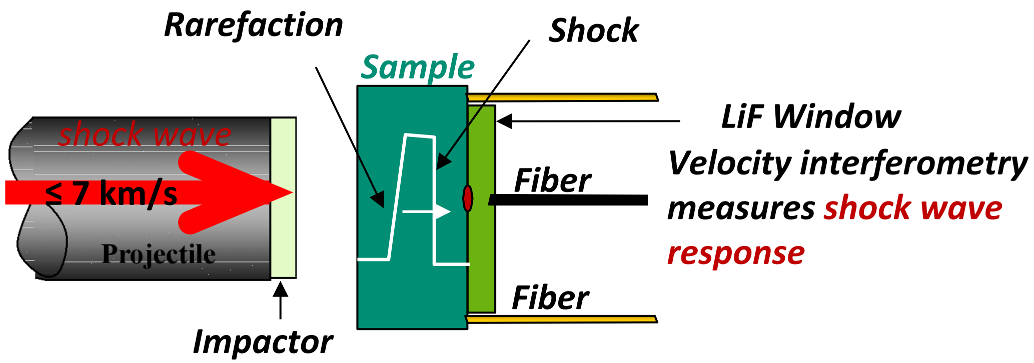
Plate Impact: Gas-guns



Z-Machine

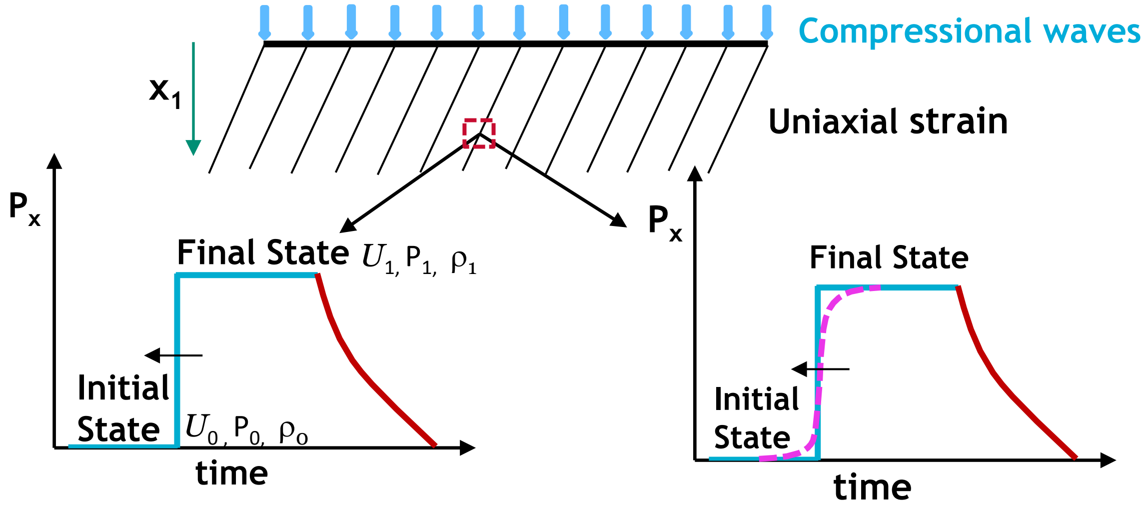
- P achieved >1000 GPa
- Experiment time: several ns

Impact experiments provide accurate EOS measurements



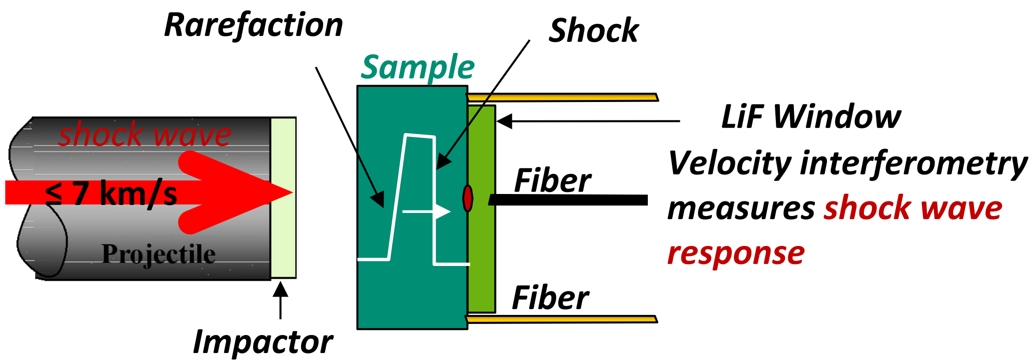
- Thin-plate impactors and targets produce uniaxial strain conditions for a short period of time (few µs) behind planar **shock wave**
- Shock speed from time-of-arrival gauges, particle velocity from velocity interferometry
- Using the measured shock wave response and jump conditions, EOS is determined

Impact experiments provide accurate EOS measurements



Shock Compression/ Release

Ramp Compression/ Release

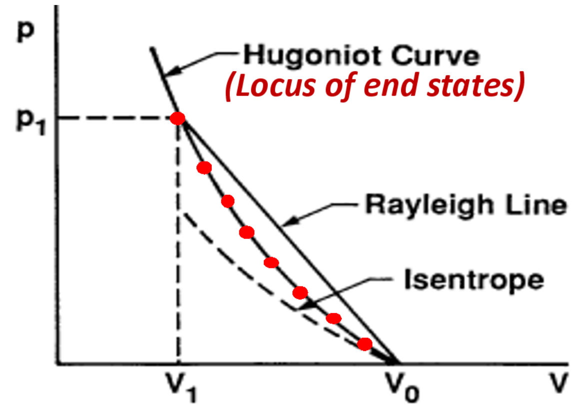


Rankine-Hugoniot Jump Conditions

$$\rho_0(U_S - U_0) = \rho_1(U_S - U_1)$$

$$P_1 - P_0 = \rho_0(U_S - U_0)(U_1 - U_0)$$

$$E_1 - E_0 = \frac{1}{2}(P_1 + P_0)(V_0 - V_1)$$

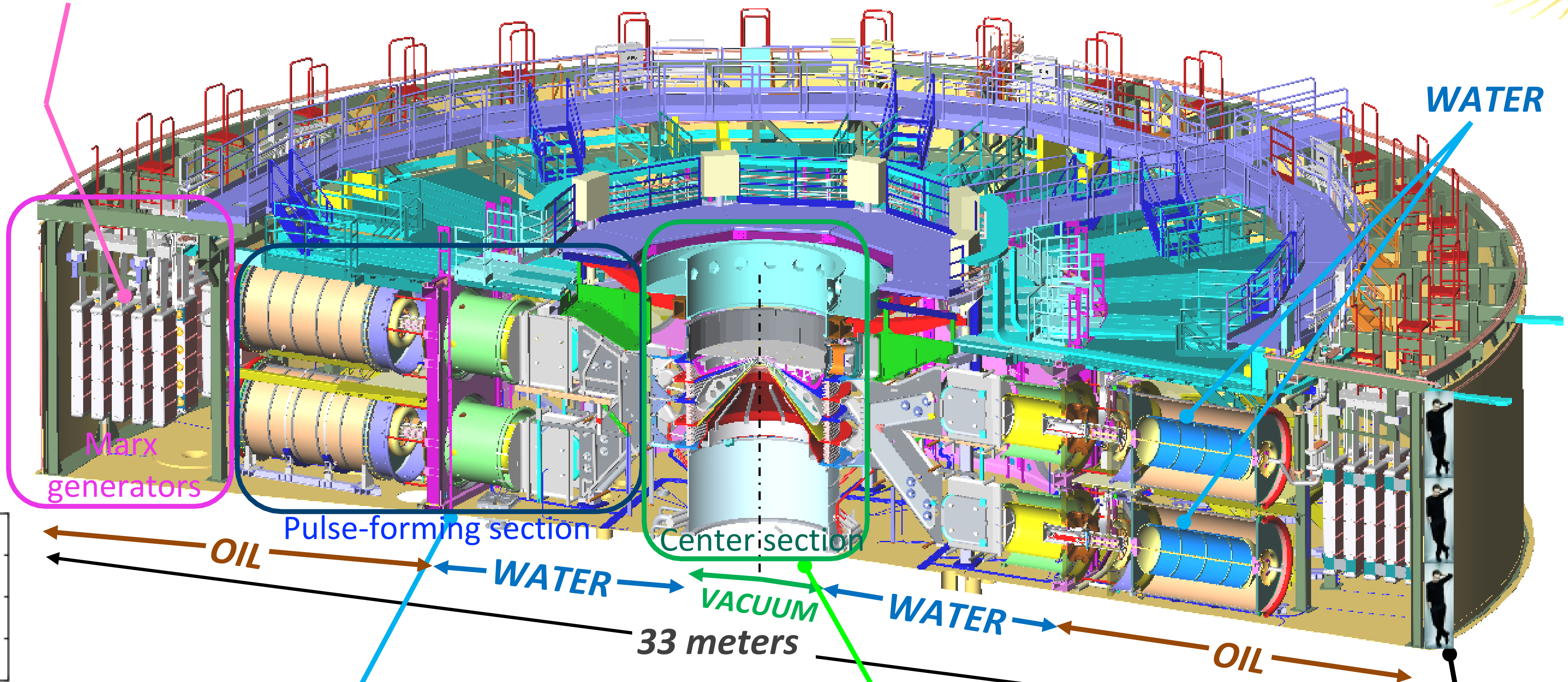


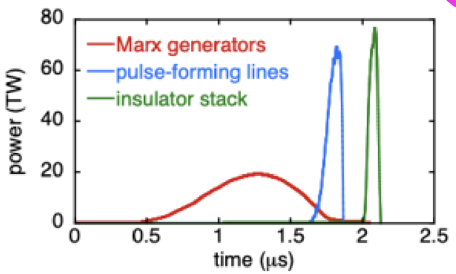
- Thin-plate impactors and targets produce uniaxial strain conditions for a short period of time (few µs) behind planar **shock wave**
- Shock speed from time-of-arrival gauges, particle velocity from velocity interferometry
- Using the measured shock wave response and jump conditions, EOS is determined

Z is a pulsed-power machine that delivers a powerful, short pulse of electrical energy

Energy storage section (850,000 gallons oil): stores 23 MJ in 36 banks of 60 capacitors (each 2.3 μF), charged in parallel (90 kV)

Energy stored in Marx generator discharged into the pulse forming section





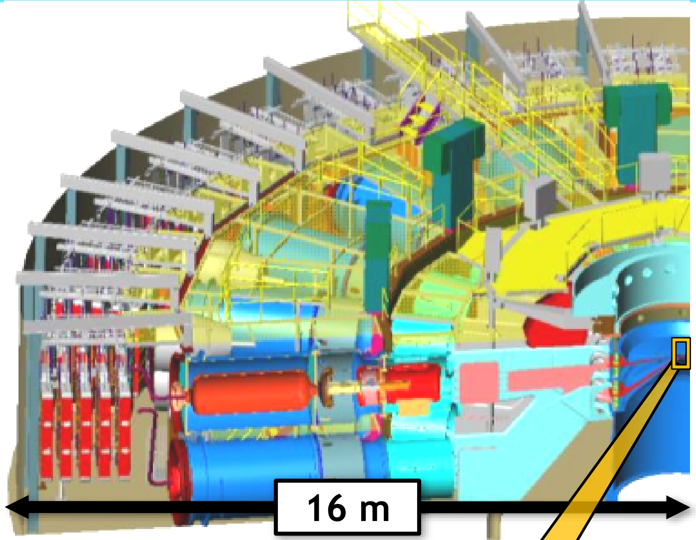
Pulse-forming section (250,000 gallons H_2O): laser-triggered SF_6 gas switches & transmission lines compress pulse to 100 ns rise time

Center section (10^{-5} torr vacuum): magnetically insulated transmission lines deliver up to 26 MA pulse to load, convolute reduces 4 feed gaps to 1

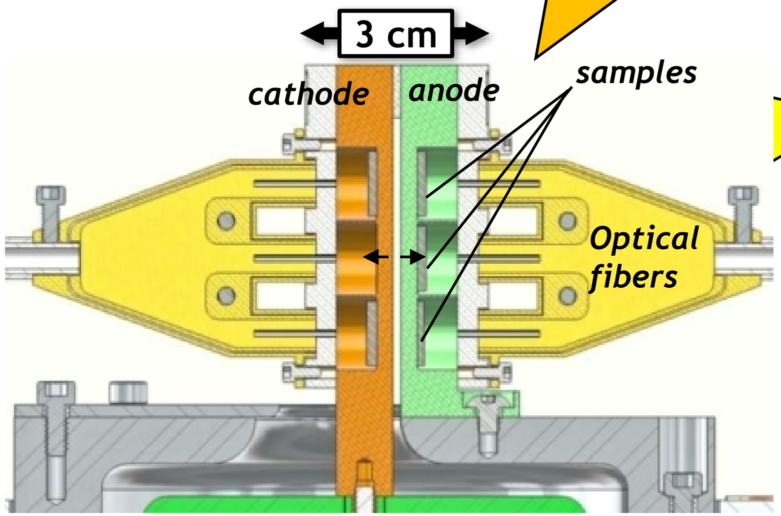
Cumberbatch for scale

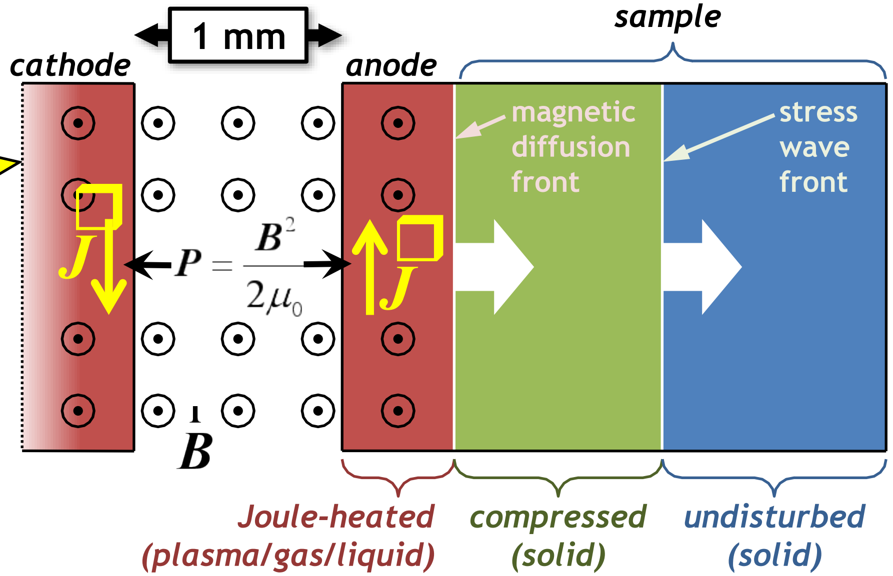
J.-P. Davis

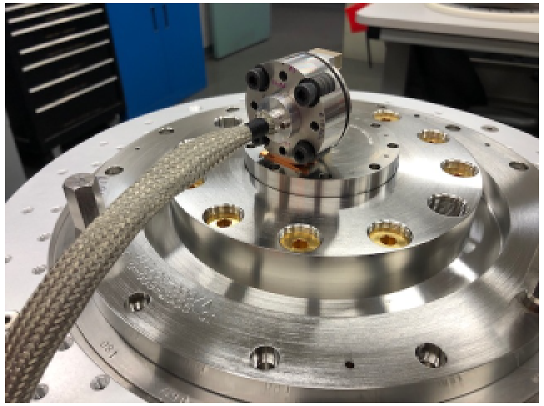
Shorted flat-plate electrodes convert current pulse to ramped magnetic loading



- Current pulse of 7-26 MA delivered to load
- Controllable pulse shape, rise time 100-1200 ns
- Stripline = parallel flat-plate electrodes shorted at one end, <u>identical loading of sample pairs</u>
- magnetic ($J \times B$) force induces ramped stress wave in electrode material
- stress wave propagates into ambient material
- mm sized samples → No edge waves
- 1 mm Cathode and anode plates become the flyer plates
- Magnetic pressure produced in A-K gap through Lorentz force





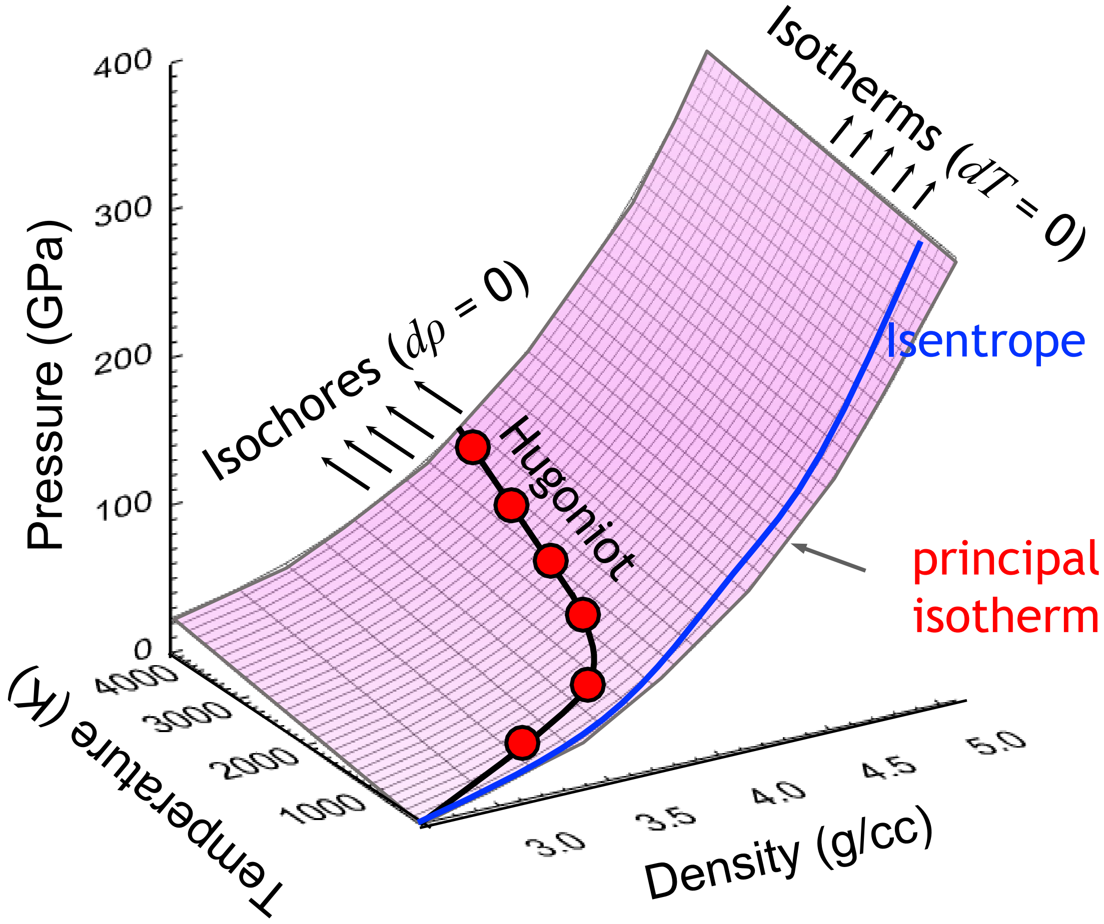


Magnetically launched flyer plates on Z produce highly accurate shock pressures to tens of Mbars with extreme temperatures.

J.-P. Davis

Equation of State (EOS)

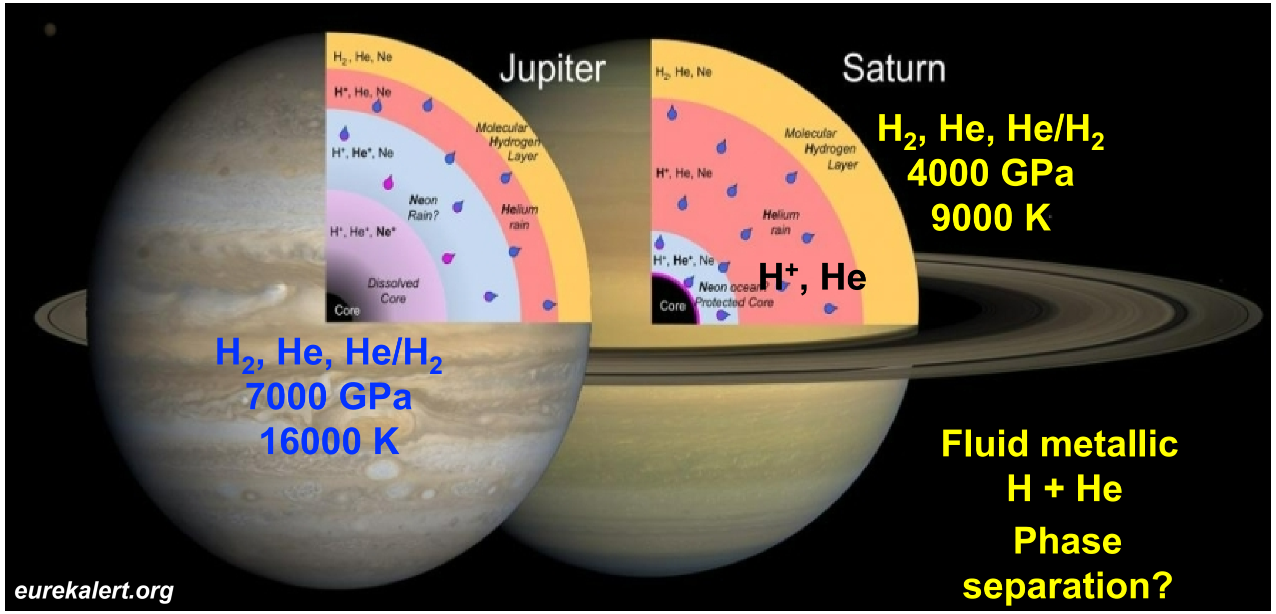
Equation of State: Relation between Temperature (T), Pressure (P), Volume (V)/Density, Internal Energy

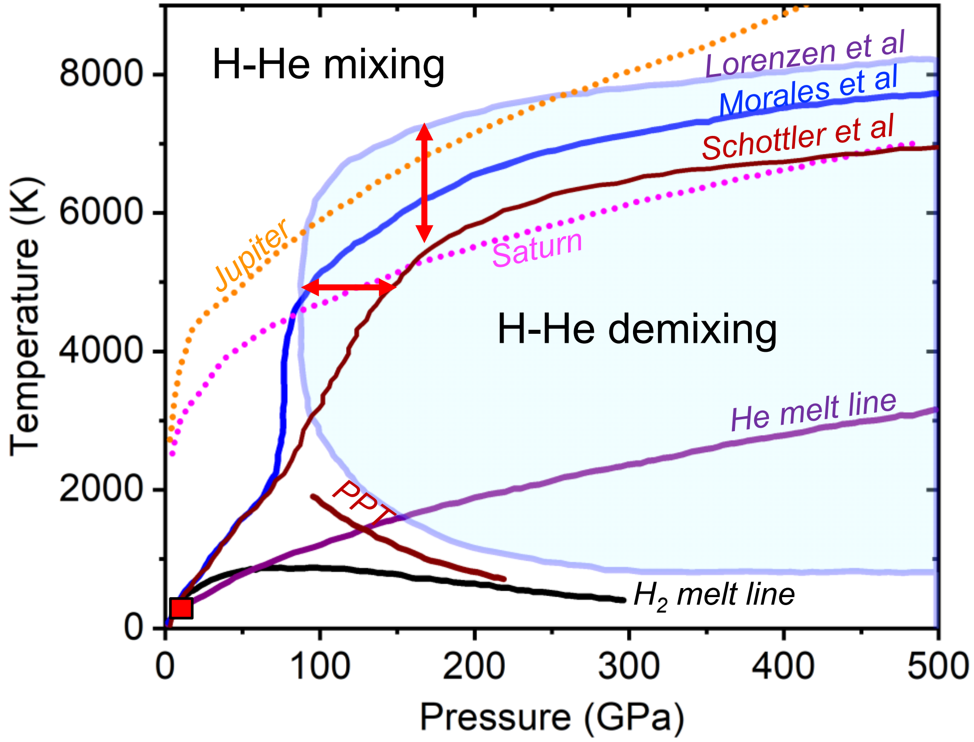


Outline

- Equation of State Measurement → Why?
- Sandia’s capabilities: Z, Gas gun to measure EOS
- STAR, Z pre-compression experiments on H_2-He mixtures
- Data: H_2-He and TiO_2
- Summary

Why Study Pre-Compressed H_2-He Mixtures?



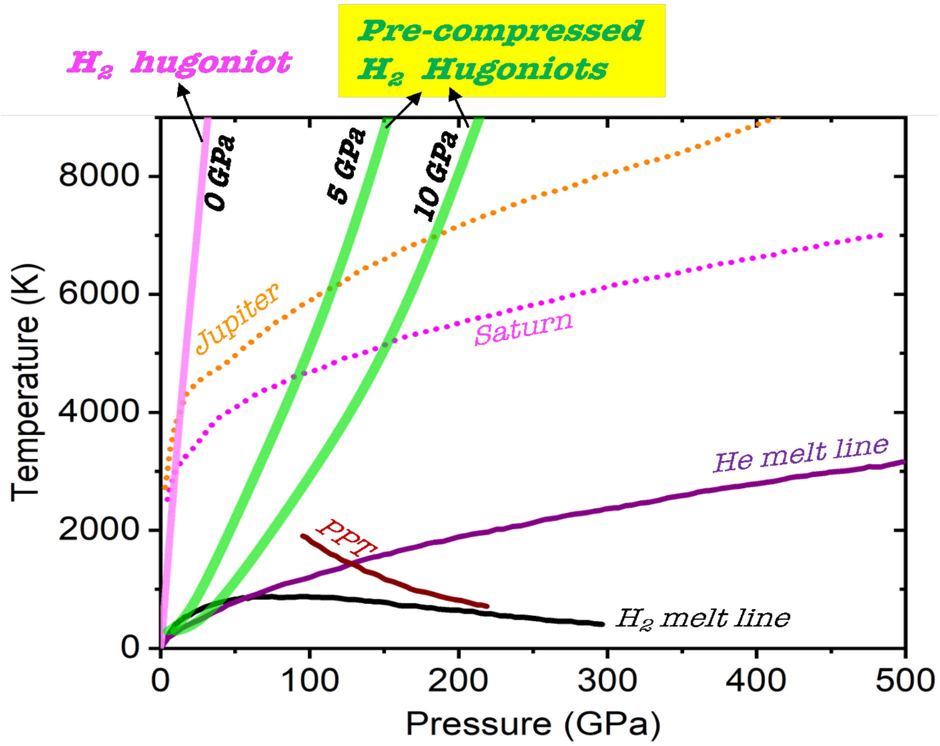


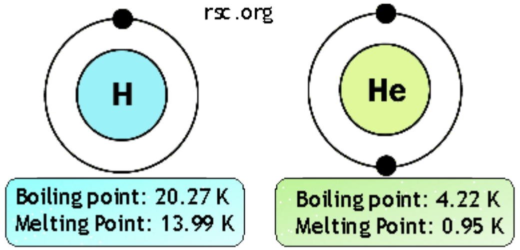
- Most abundant elements in the universe
- Studying EOS of H_2-He mixtures imperative for successful modeling of gas giant planets

- Huge variations between theoretical studies
- Dearth of experimental data
- Develop a capability to study H-He EOS using Z and STAR gas guns

Study equation of states of H_2-He mixtures and validate mixture models

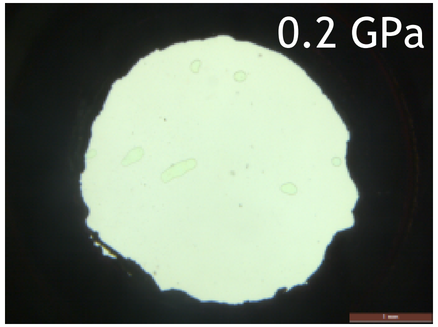
Pre-compression only method to achieve high density states





High-Pressure Gas Loader ~25,000 psi

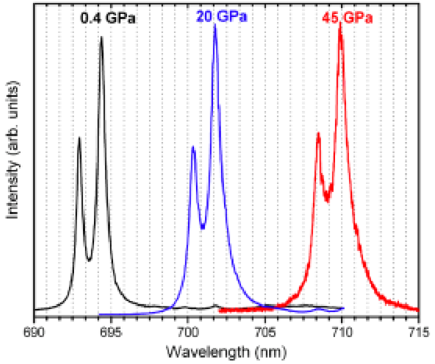
Diamond Anvil Cell (DAC)



Micro-photograph of fluid mixture of H_2-He through the anvil window

Sample thickness: 0.3728 mm

Sample diameter: 3.6 mm

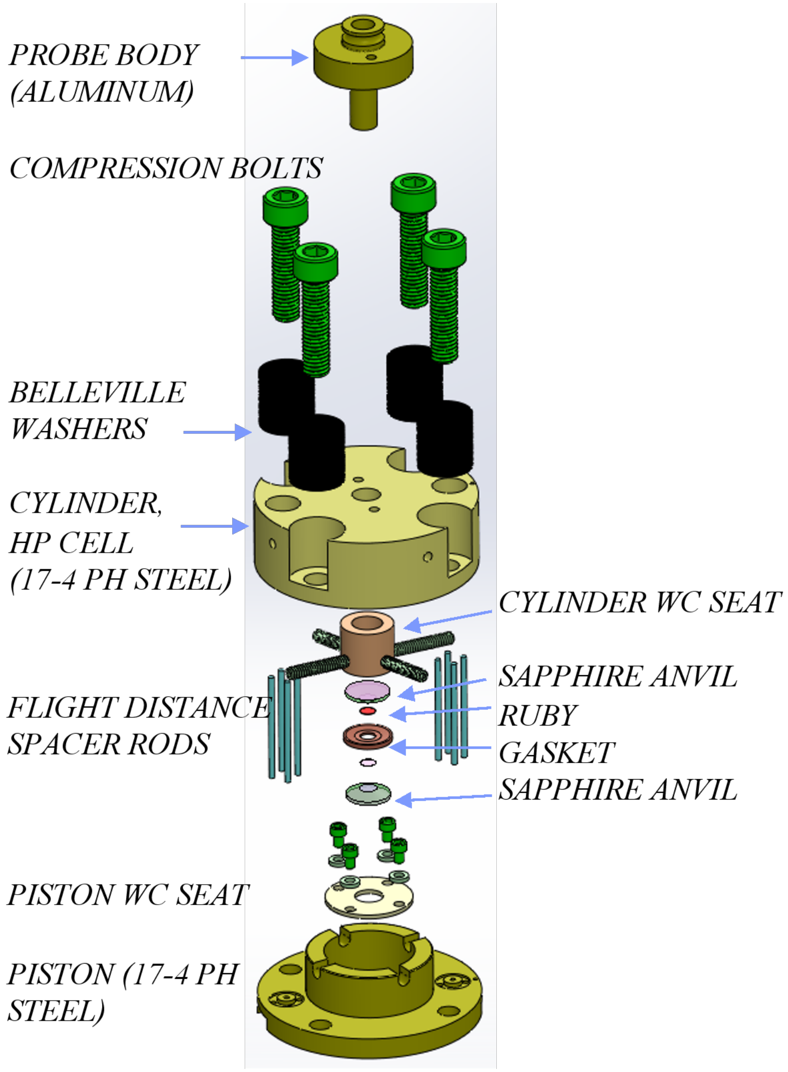


Ruby-shift with change in pressure

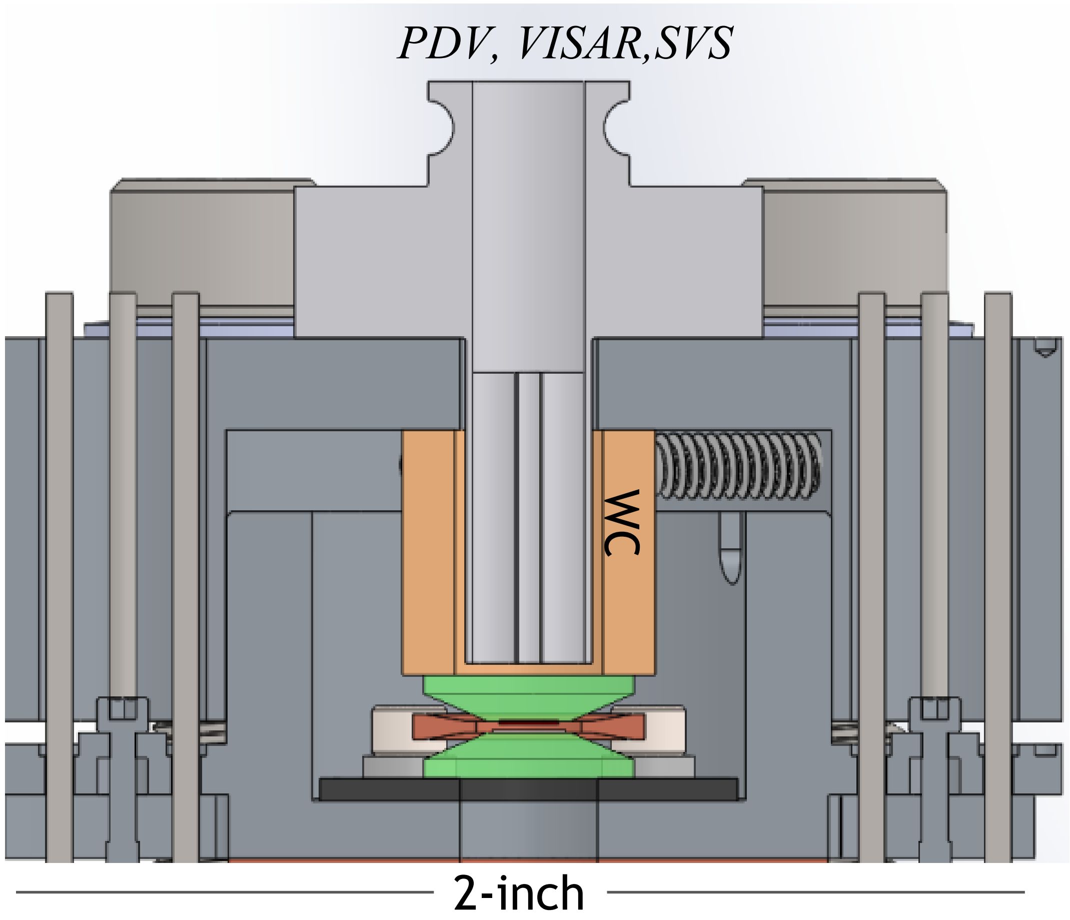
- **Pre-compression allows to access unique state off of the Hugoniot.**
- **Probe different planetary states of Jupiter and Saturn**
- **Only method to achieve high density states**

Pre-compression is the only method to achieve high density states of a homogeneous mixture initial condition

Pre-Compression Capability at Z and STAR

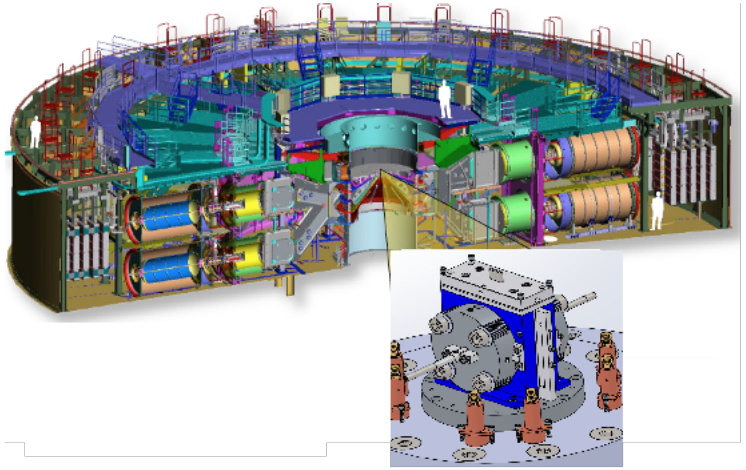


Components of pre-compression cell

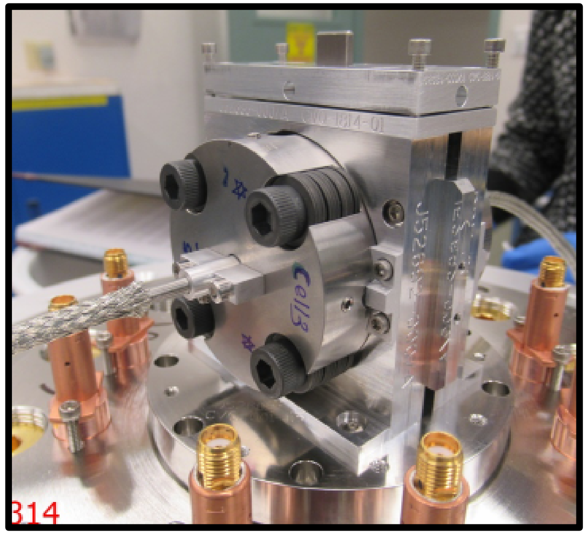


Side View of the closed DAC

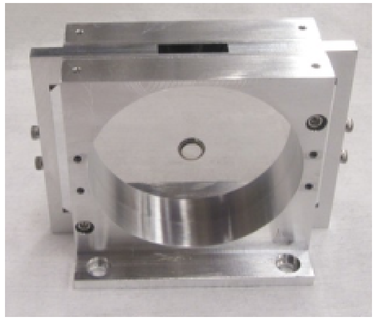
Pre-Compression Capabilities at Z and STAR



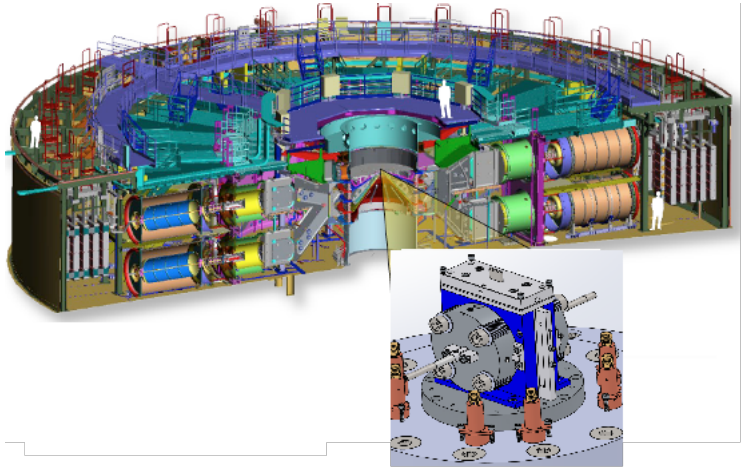
Target assembly for Z-experiment

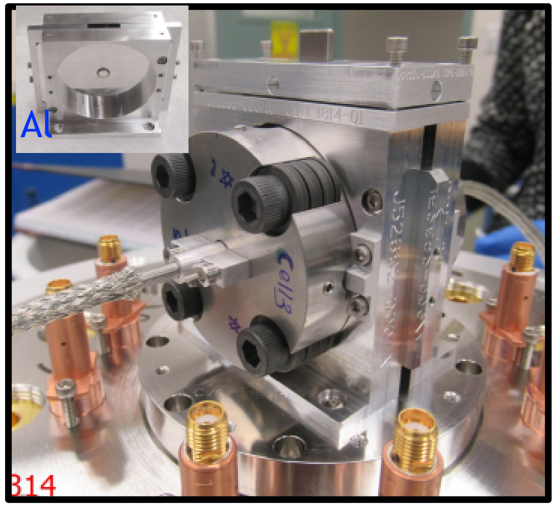


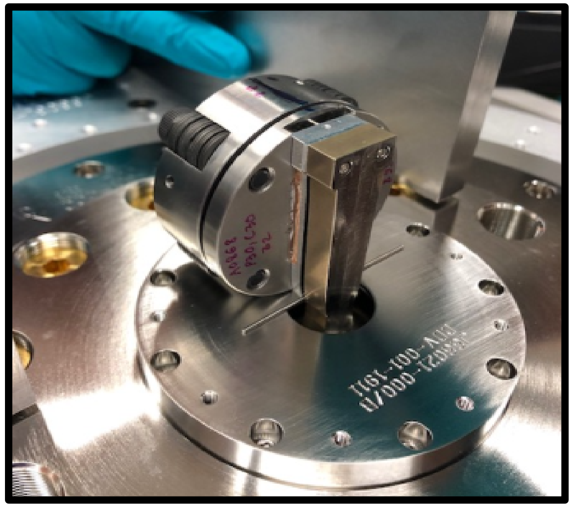
Al



Pre-Compression Capabilities at Z and STAR

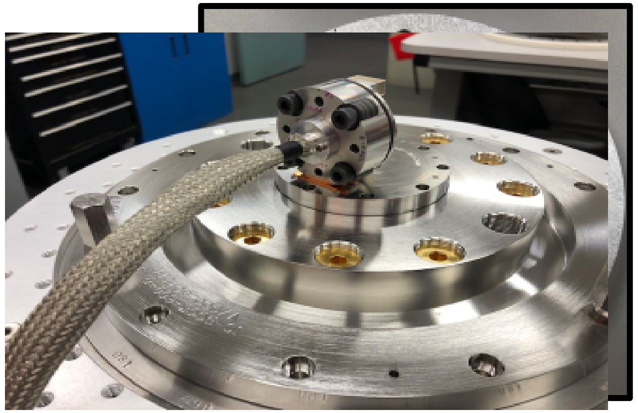
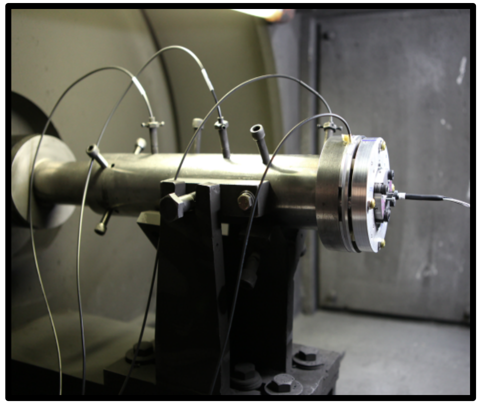
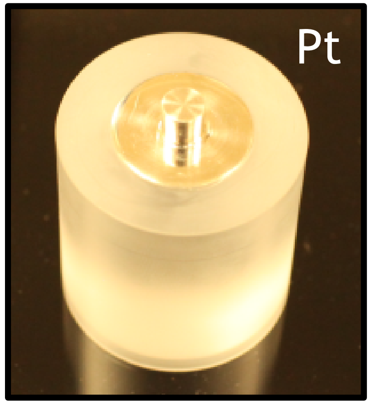


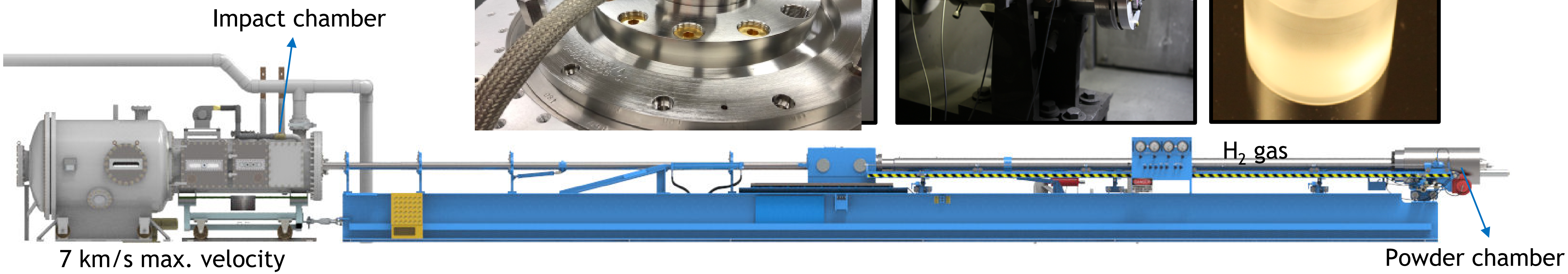




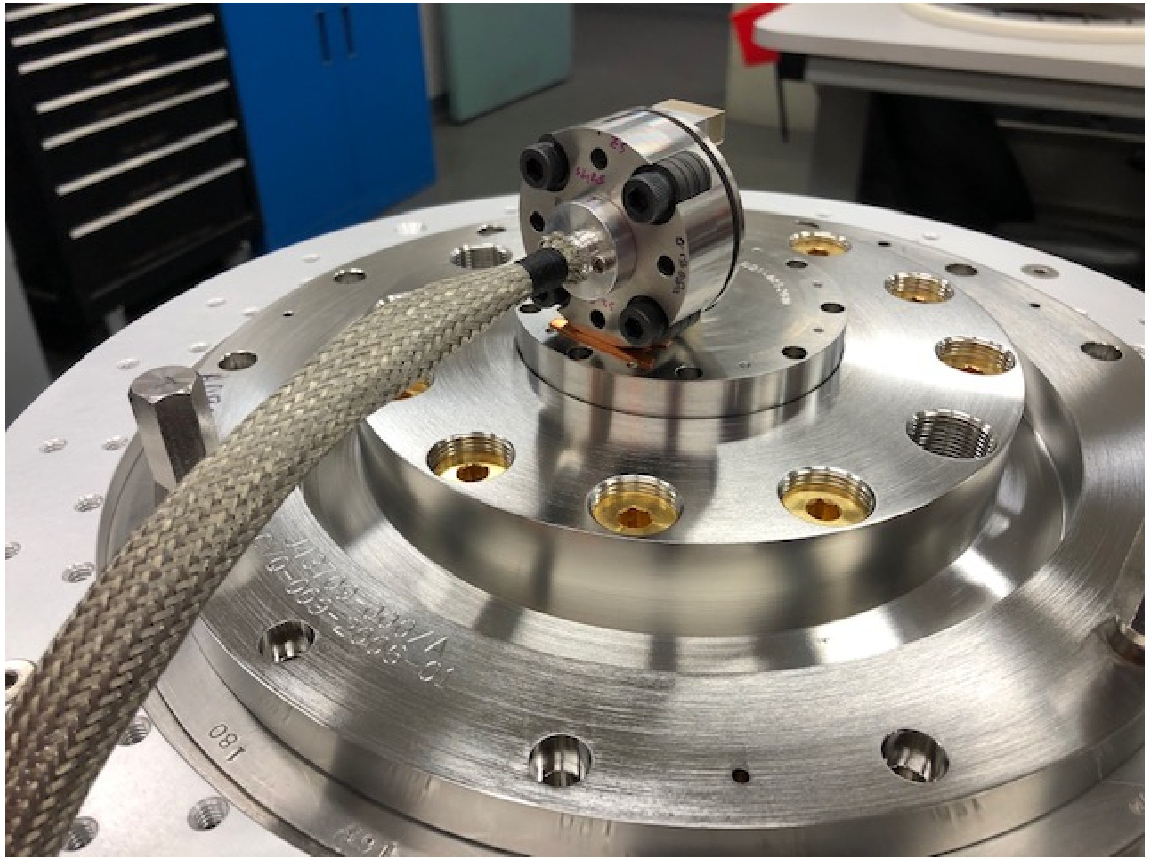
Target assembly for Z-experiment

Two stage light gas-gun at STAR

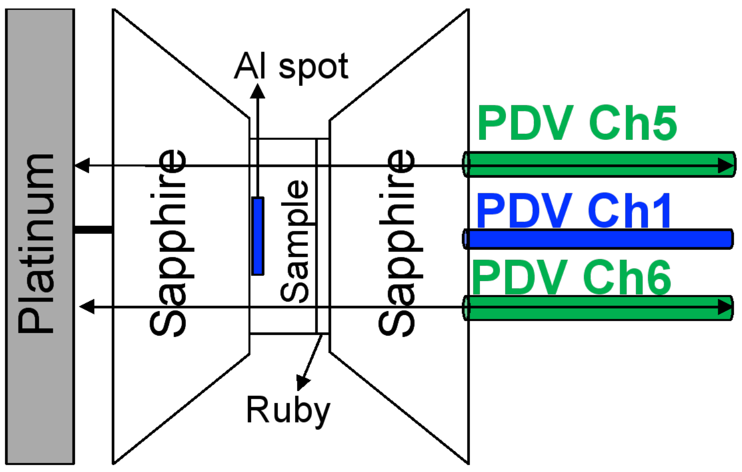






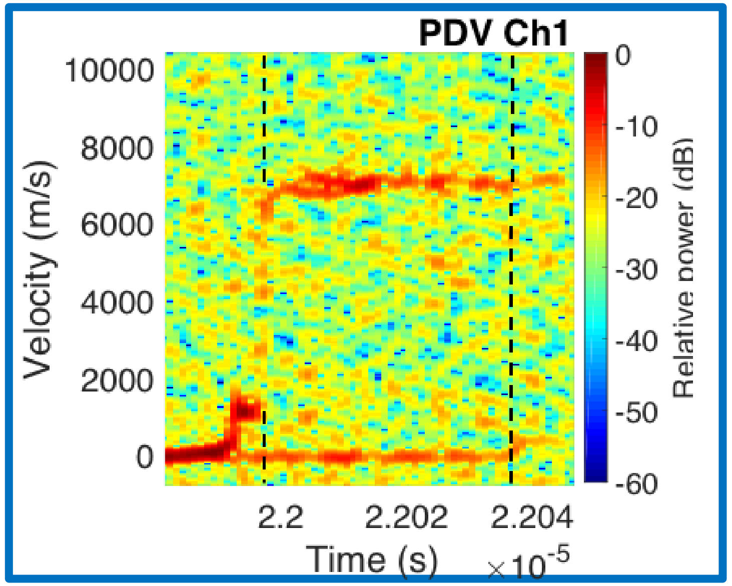
Pre- and Post-Shot

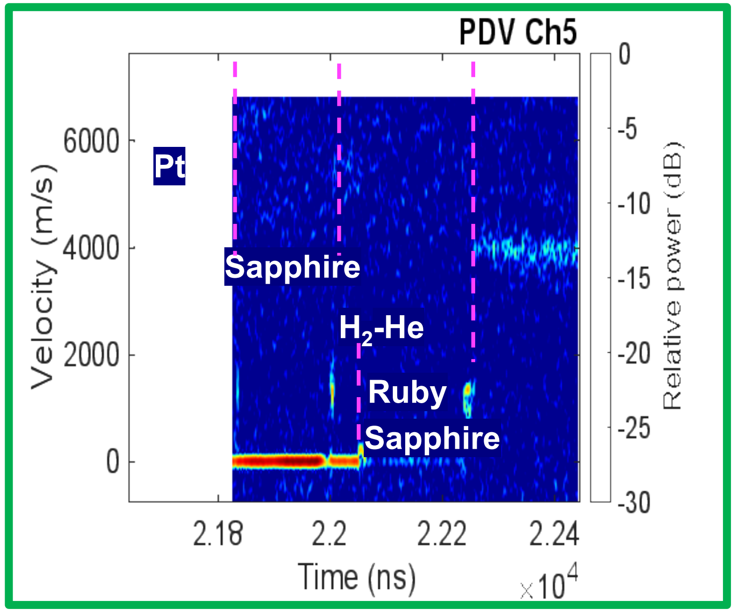


Velocimetry Showing Particle Velocity and Shock Transit Times



Schematic for shock loading pre-compressed H_2 -He mixture with a Pt flyer at STAR





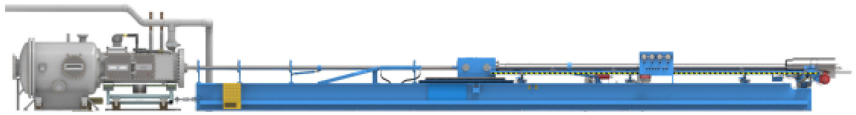
	Pt Velocity (km/s)	*U_p (km/s)*	*U_s (km/s)*	*P (GPa)*
PC_20	5.01	7.02±0.03	9.52±0.24	9.69±0.76

$$\rho_0(U_S - U_0) = \rho_1(U_S - U_1)$$

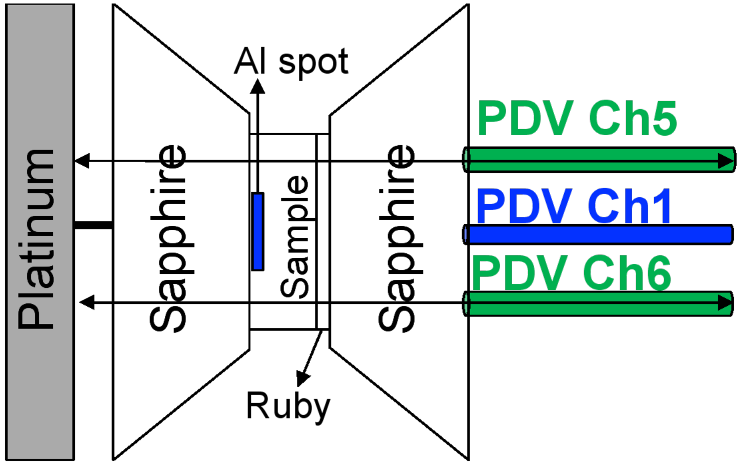
$$P_1 - P_0 = \rho_0(U_S - U_0)(U_1 - U_0)$$

$$E_1 - E_0 = \frac{1}{2}(P_1 + P_0)(V_0 - V_1)$$

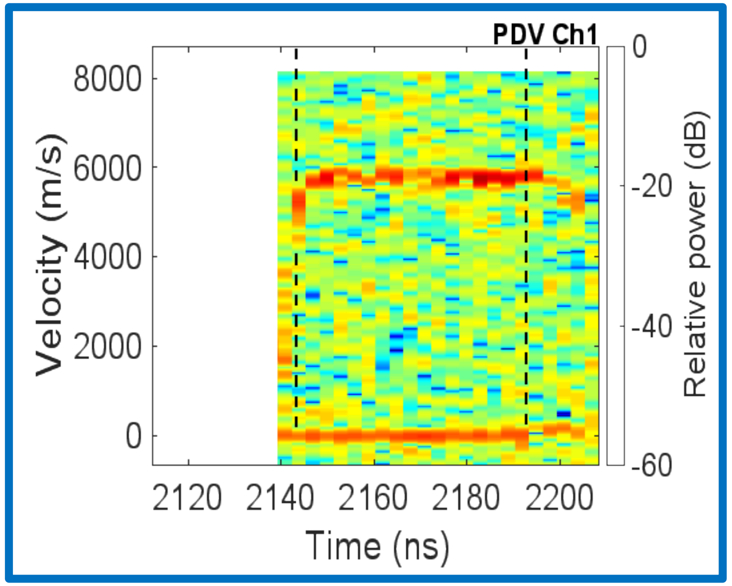
- Successfully measured the particle velocity, shock velocity, and transit times from PDV data.
- Uncertainties in shock velocity, initial density

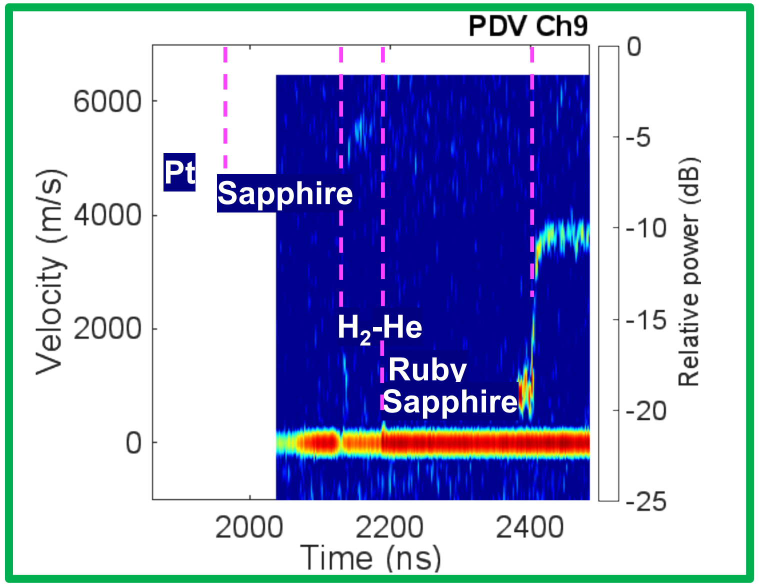


Velocimetry Showing Particle Velocity and Shock Transit Times



Schematic for shock loading pre-compressed H_2 -He mixture with a Pt flyer at STAR.





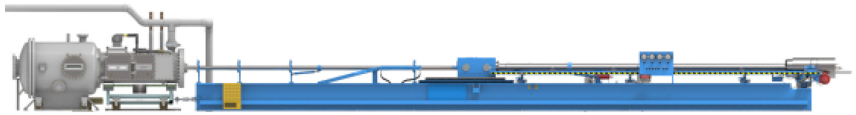
	Pt Velocity (km/s)	*U_p (km/s)*	*U_s (km/s)*	*P (GPa)*
PC_20	5.01	7.02±0.03	9.52±0.24	9.69±0.76
PC_24	4.25	5.76±0.05	7.86±0.31	6.64±0.49

$$\rho_0(U_S - U_0) = \rho_1(U_S - U_1)$$

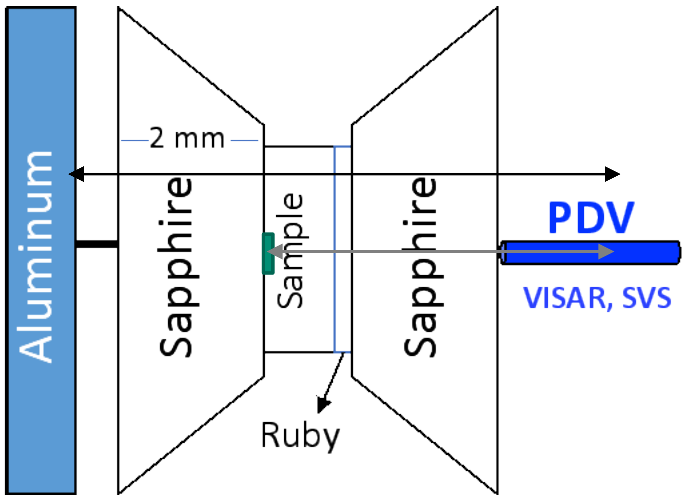
$$P_1 - P_0 = \rho_0(U_S - U_0)(U_1 - U_0)$$

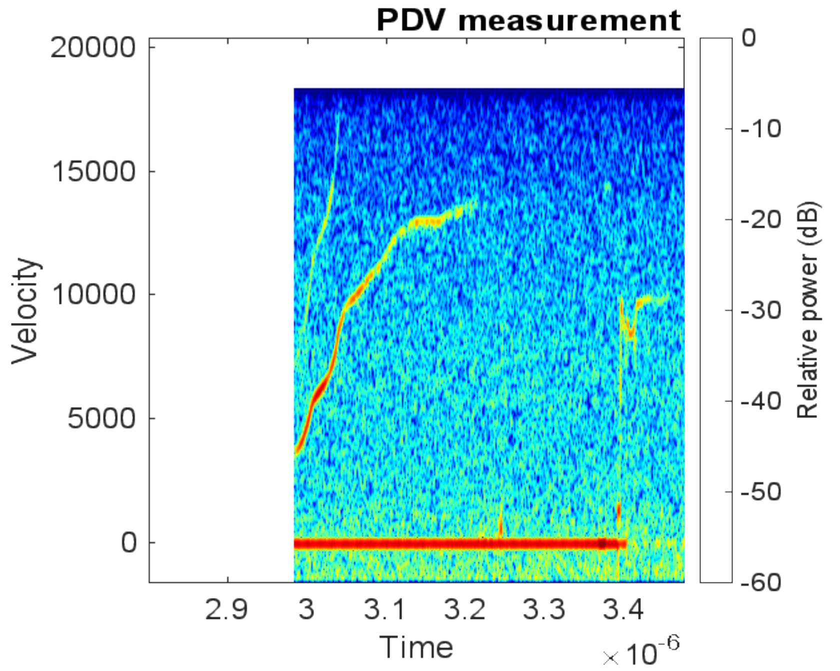
$$E_1 - E_0 = \frac{1}{2}(P_1 + P_0)(V_0 - V_1)$$

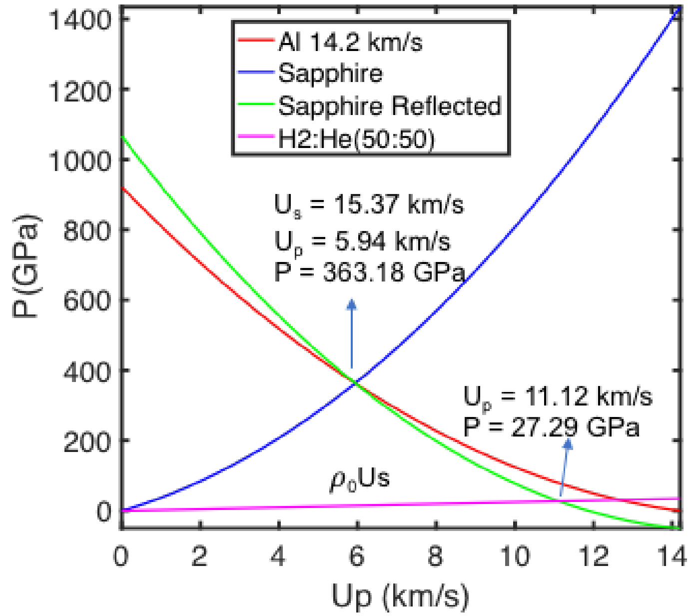
- Measured the particle velocity, shock velocity, and transit times from PDV data.
- Uncertainties in initial density, shock velocity, break-out times



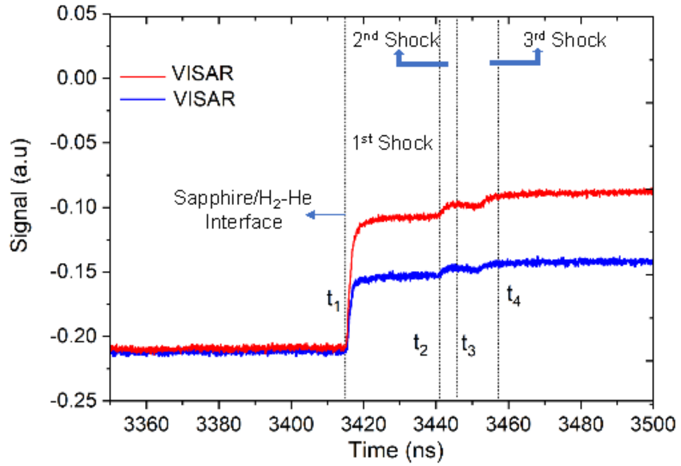
Z- Velocimetry Measurements of H_2-He (50:50)





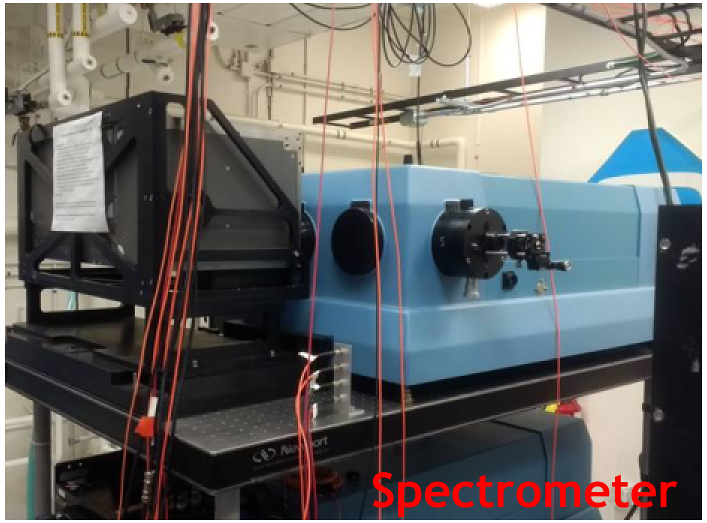


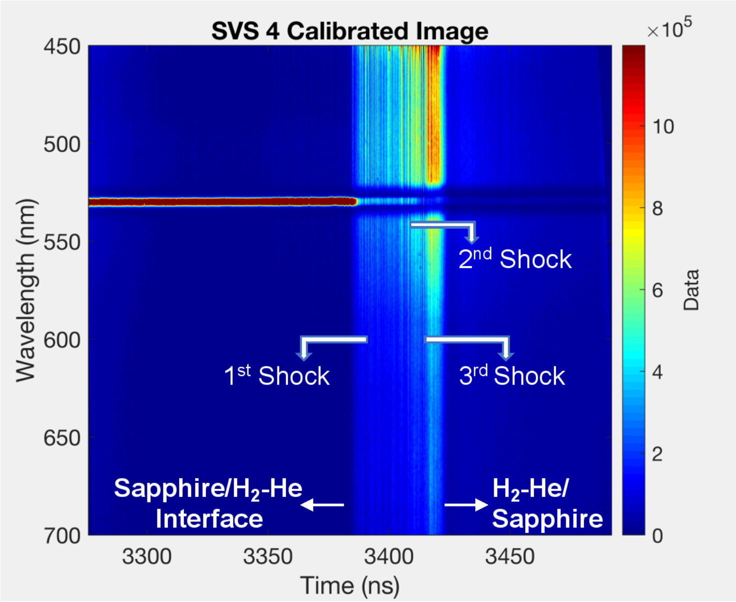
Impedance matching in the P-U_p plane.

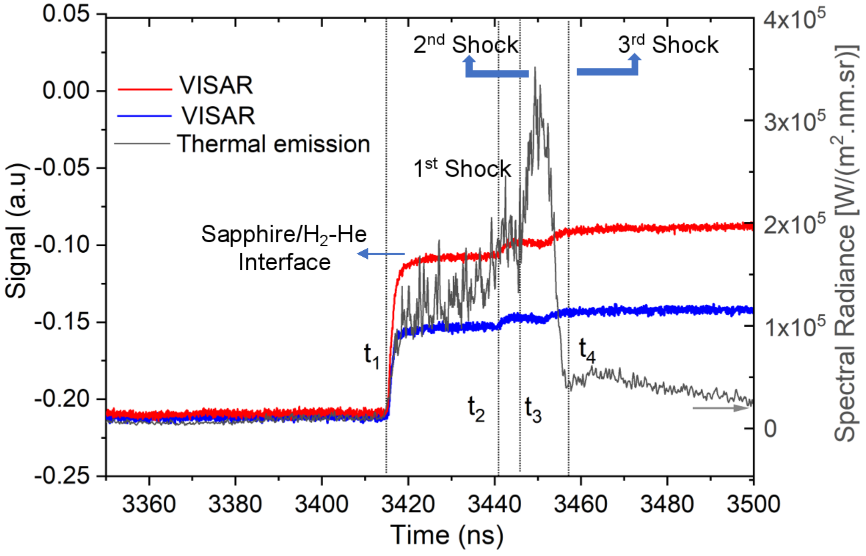


- Al flyer velocity of 14.2 km/s was obtained
- Shock front not reflective
- VISAR measurements
- Transit time measurement → Us obtained: 17.79 ± 0.38 km/s

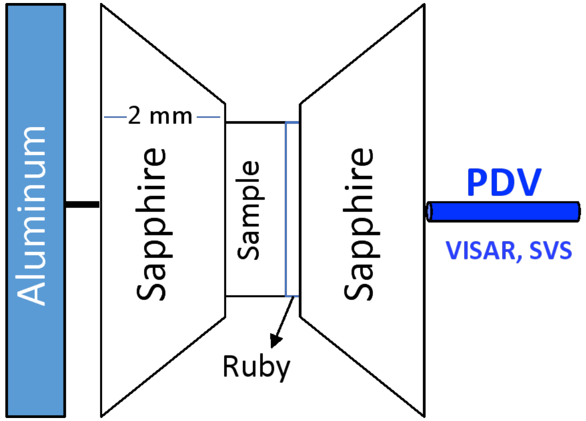
VISAR and Temperature Measurements of Gas Mixtures (H_2:He)

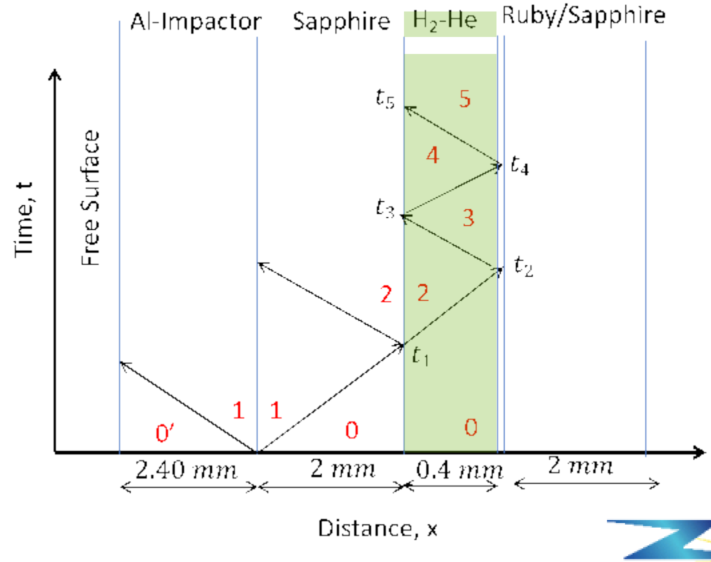




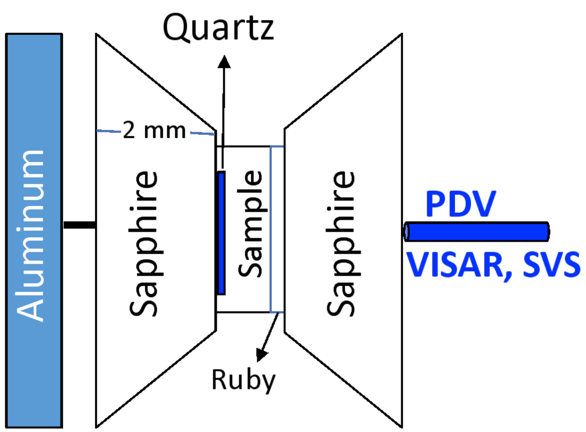


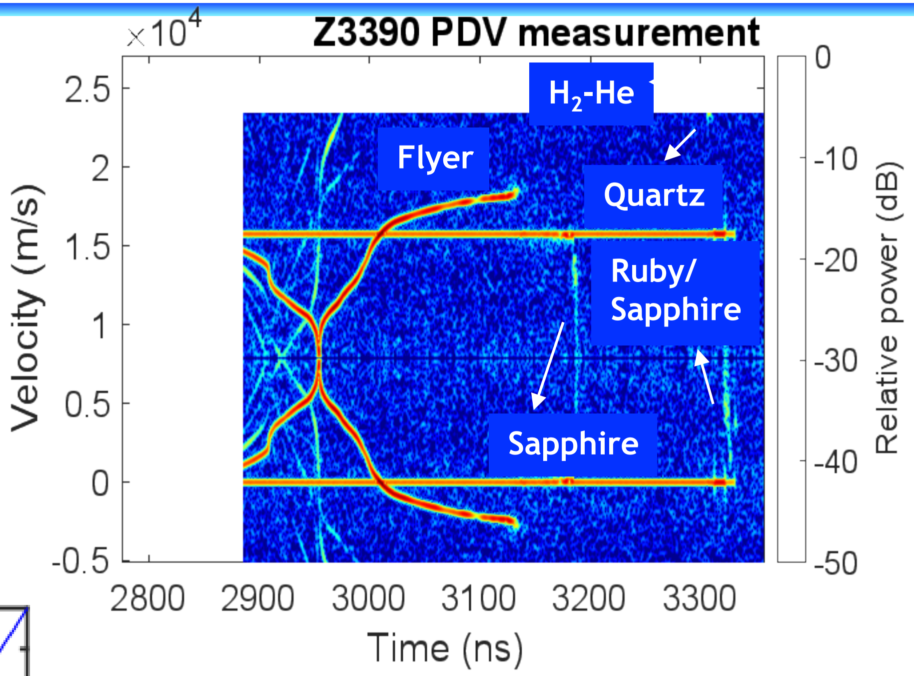
- Streaked Visible Spectroscopy (SVS)
- Simultaneous VISAR and temperature measurements were performed
- Thermal emission data correlates well with the VISAR data.

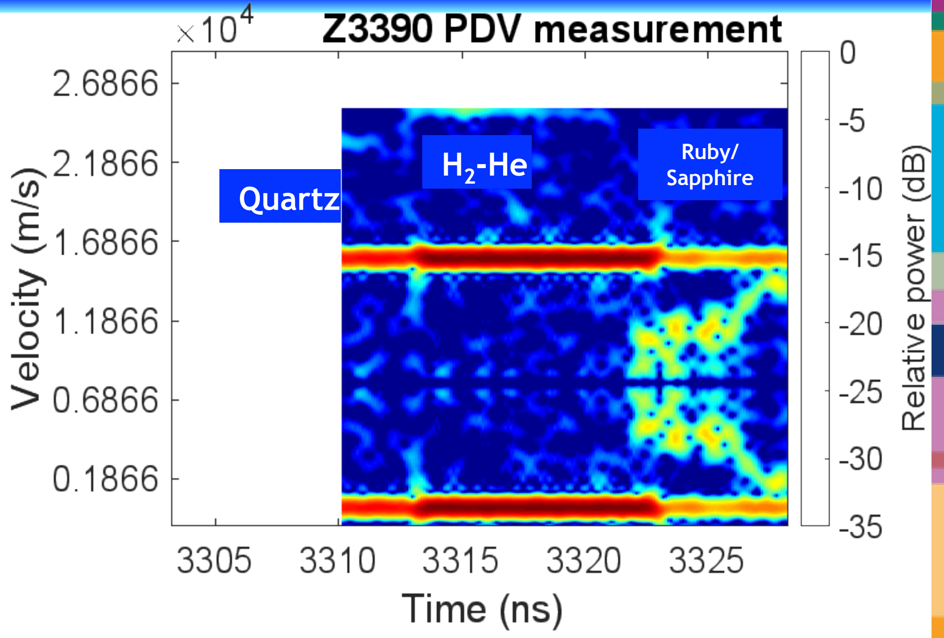


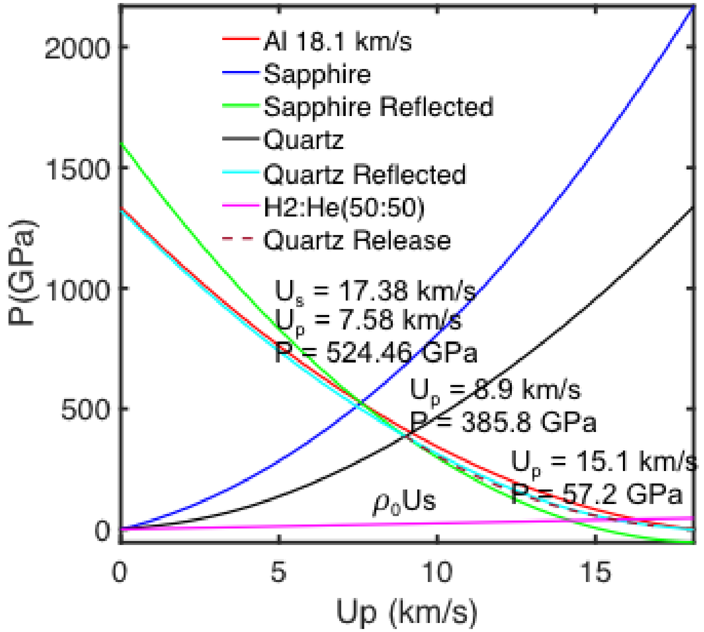


Directly Measured Shock Velocity from the Mixture at 57 GPa



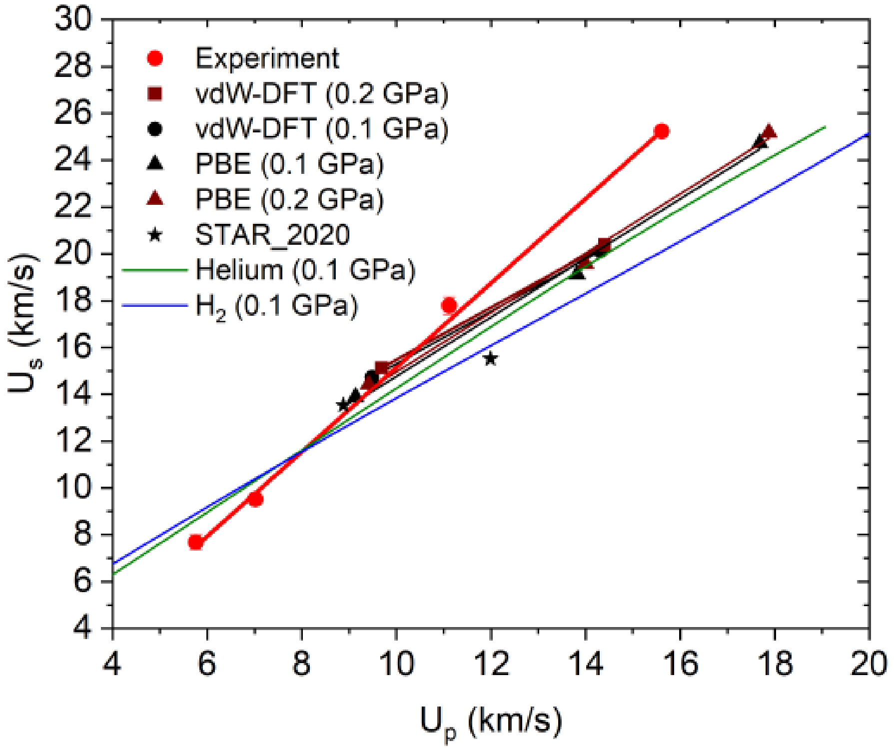






- Flyer velocity: 18.1 km/s
- Used the well established quartz release model
- Shock velocity directly measured from the reflective shock front in quartz and sample
- Pressure and Particle velocity obtained from Impedance Matching

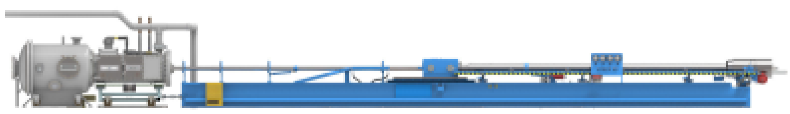
Hugoniot of Hydrogen-Helium Mixture

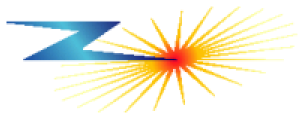


- Equation of state less compressible than DFT
- U_p affected by initial density and refractive index
  - Uncertainty in initial Density
  - Uncertainty in refractive Index for Window correction

$P=\rho_0 UsUp$

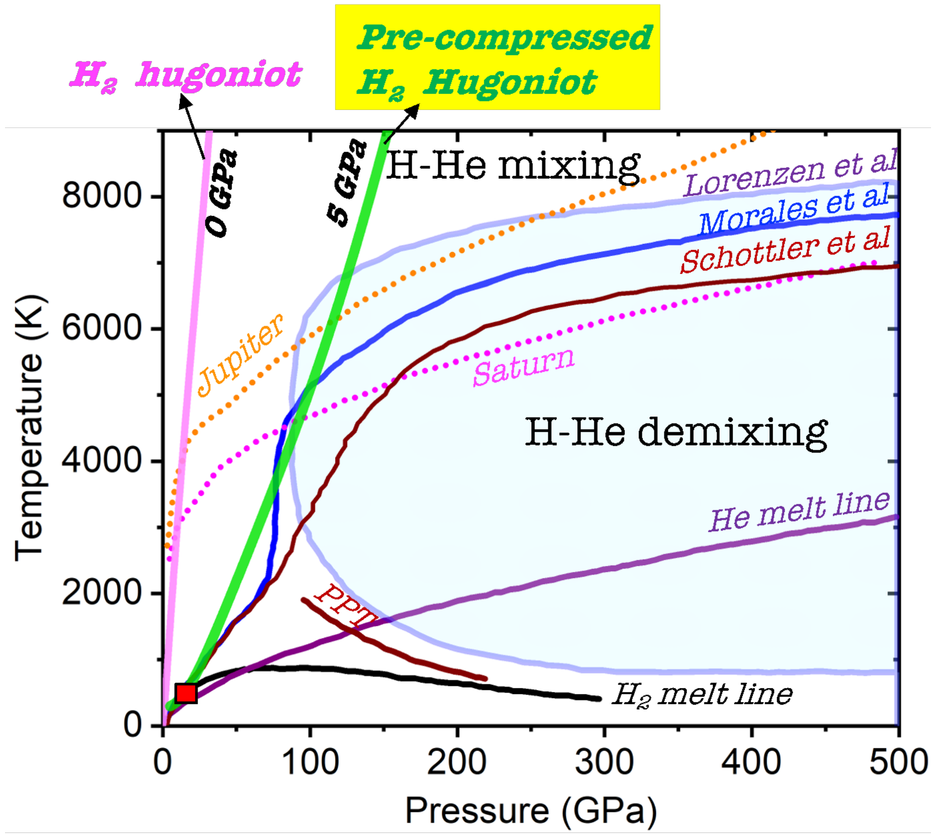
Expt. No.	*Initial P (GPa)*	*Sample thickness (μm)*	*Transit time (μs)*	*Flyer*	*Flyer Velocity (km/s)*	*U_p (km/s)*	*U_s (km/s)*	*P (GPa)*	*Rho_0 (g/cm^3) (vdw-DFT)*	*Alpha-quartz U_s (km/s)*
STAR_PC_20[a]	0.134	381.0 ± 2.4	0.040±0.001	Pt	5.01	7.02 ± 0.03	9.52 ± 0.24	11.69 ± 0.92	0.173	
STAR_PC_24[a]	0.16	399.5 ± 2.2	0.052±0.002	Pt	4.25	5.76 ± 0.05	7.68 ± 0.31	7.94 ± 0.59	0.176	
STAR_PC_30[a]	0.15	213.3 ± 2.5	0.0168±0.0004	Pt	5.78	8.78 ± 0.12	12.65 ± 0.15	19.47+1.34	0.174	
STAR_PC_31[a]	0.15	251.2 ± 2.3	0.01533±0.0008	Pt	6.23	11.99 ± 0.09	16.39 ± 0.50	34.34 + 2.53	0.174	
Z3326[b]	0.22	455.0 ± 2.0	0.0256±0.00054	Al	14.2	*11.18± 0.09*	17.80 ± 0.38	36.44 ± 1.85	0.182	
Z3390[c]	0.150	221.2 ± 2.3	0.0087±0.00056	Al	18.1	*15.09± 0.08*	25.24 ± 0.09	66.42 ± 4.45	0.174	16.27 ± 0.01





Summary and Path Forward

- **Developing pre-compression capability at Z and STAR to study H_2-He mixtures**
- **Challenging Experiments → Ongoing Learning Process**
- **Constrain Initial Density and Refractive Index**
- **More experiments scheduled at Z and STAR to build an EOS for mixtures with quartz standard to differentiate between the models**



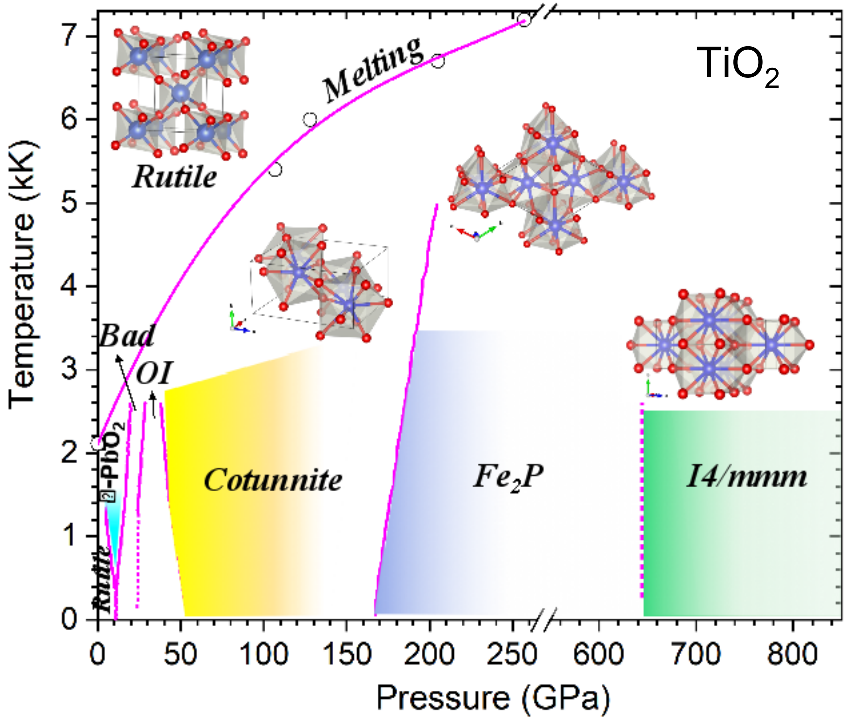
- Equation of State Measurement → Why?
- Sandia's capabilities: Z, Gas gun and Thor
- STAR, Z pre-compression experiments
- Data: H_2-He and **TiO_2**
- Summary

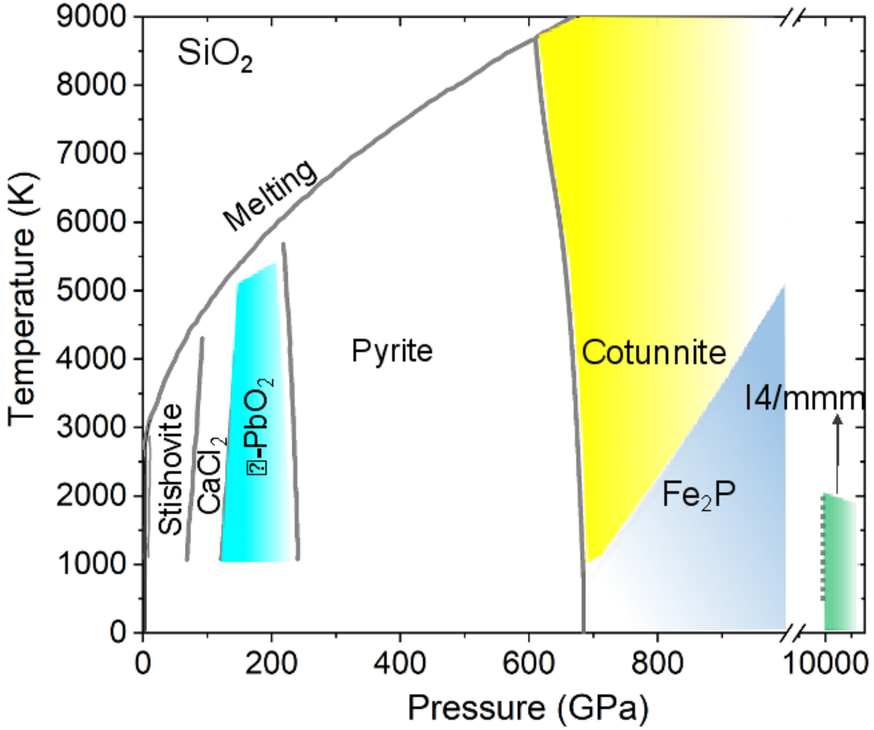
TiO_2: Applications/Motivation

Industrial applications of TiO_2

TiO_2: Applications/Motivation

Industrial applications of TiO_2



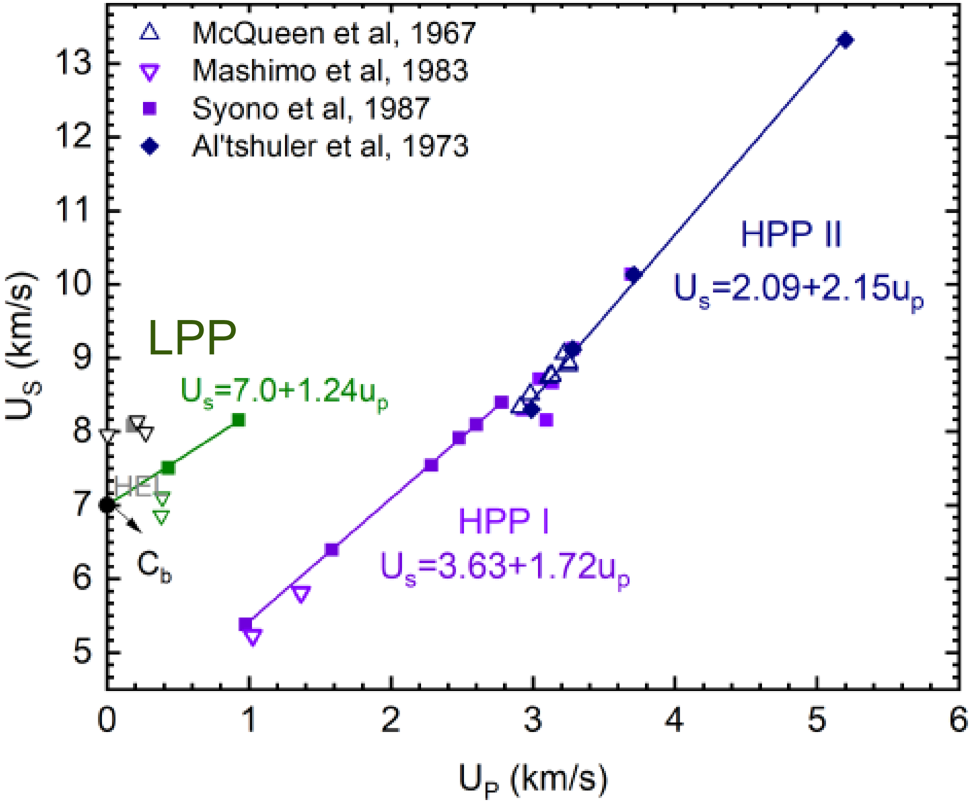


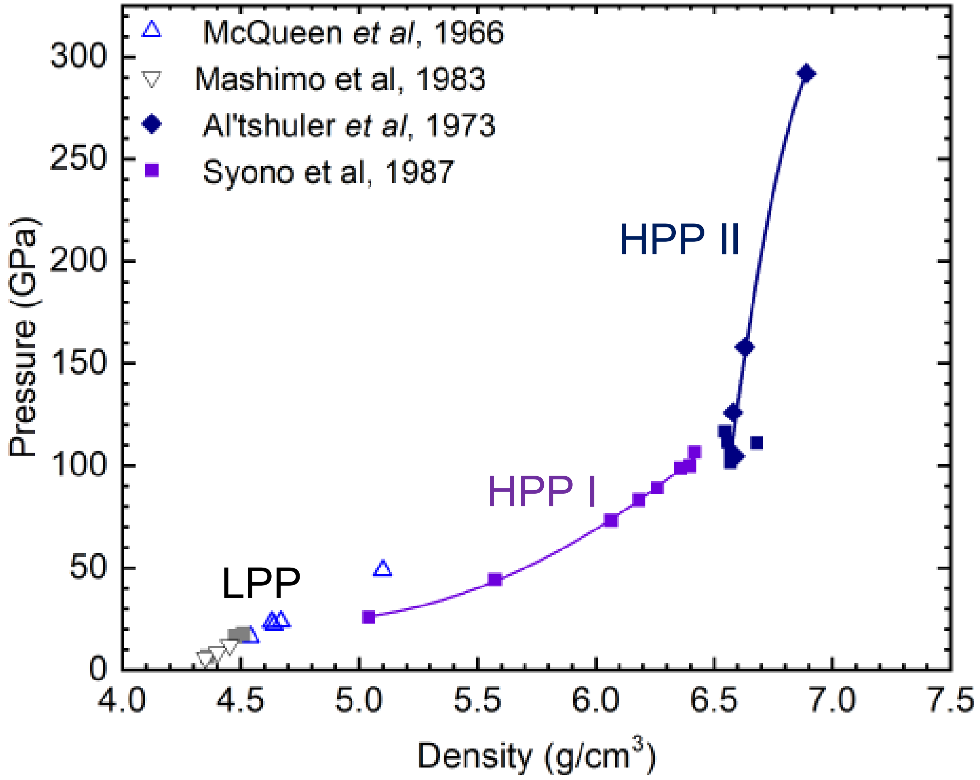
- Structural analogs to earth's mantle most abundant material, SiO_2
- Predicted phase transitions in SiO_2 at 10 TPa could happen at pressures as low at 640 Gpa
- Polymorphs: Rutile, Anatase and Brookite → Rutile is most stable

TiO_2 as an analogue material to study phase transitions in SiO_2

Duwal et al. PRB.102.**2020**

Prior Shock Work

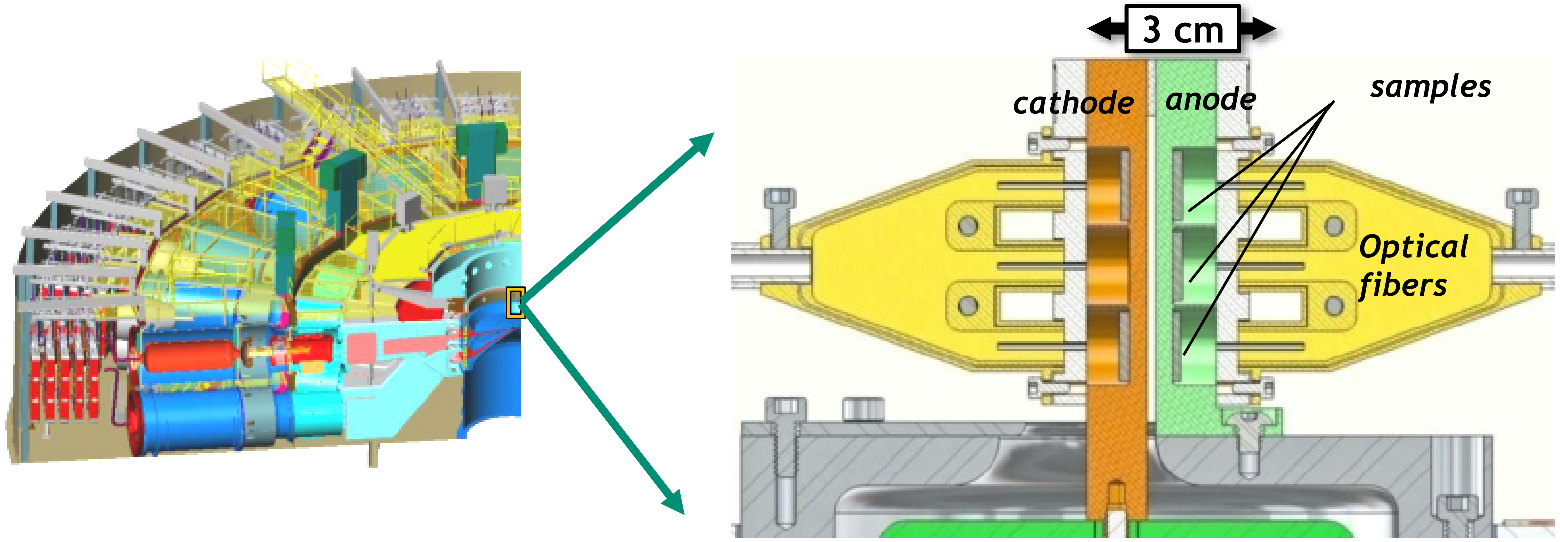




- Rutile → Low pressure phase, LPP at 30 GPa →High pressure phase, HPP I → HPP II at 100 GPa
- Hugoniot data of TiO_2 limited to ~292 GPa
- TiO_2 concluded to have 'virtually incompressible' Hugoniot by Al'tshuler et al

Obtain a complete equation of state for Rutile TiO_2 using three platforms: Z, Omega, DFT calculations

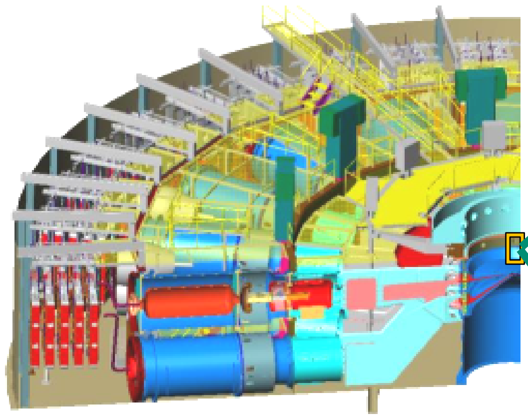
Z Experiments on TiO_2

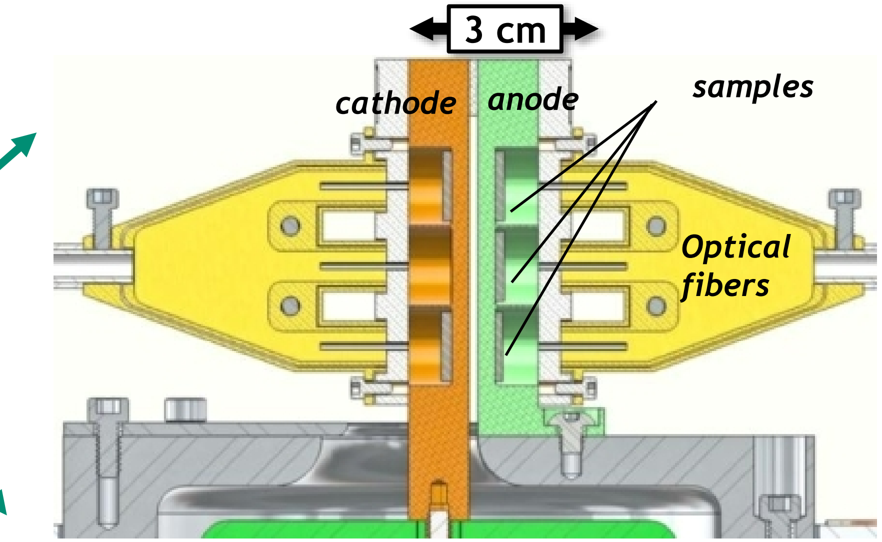


Target assembly for Z-experiment

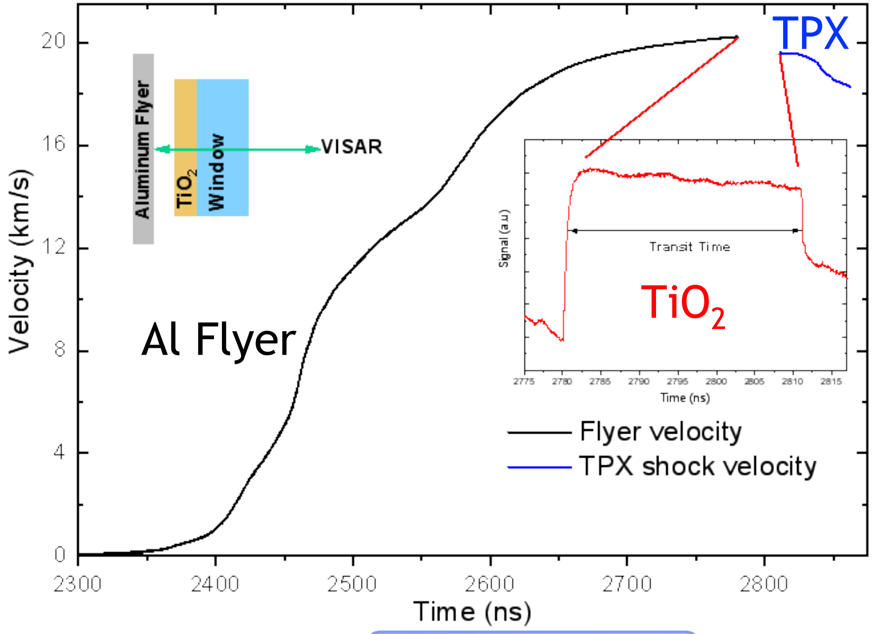
- Single crystal [110], [111], [001] TiO_2 investigated
- TiO_2 backed by TPX/Quartz window for release measurements
- Flyer Velocities: 12 to 22.5 km/s
- At impact velocities < 20 km/s the shock front is not reflective
- VISAR measures impact velocity, shock velocities and transit times

Z Experiments on TiO_2





Target assembly for Z-experiment



Z-VISAR trace

- Single crystal [110], [111], [001] TiO_2 investigated
- TiO_2 backed by TPX/Quartz window for release measurements
- Flyer Velocities: 12 to 22.5 km/s
- At impact velocities < 20 km/s the shock front is not reflective
- VISAR measures impact velocity, shock velocities and transit times

$$\rho_0(U_S - U_0) = \rho_1(U_S - U_1)$$

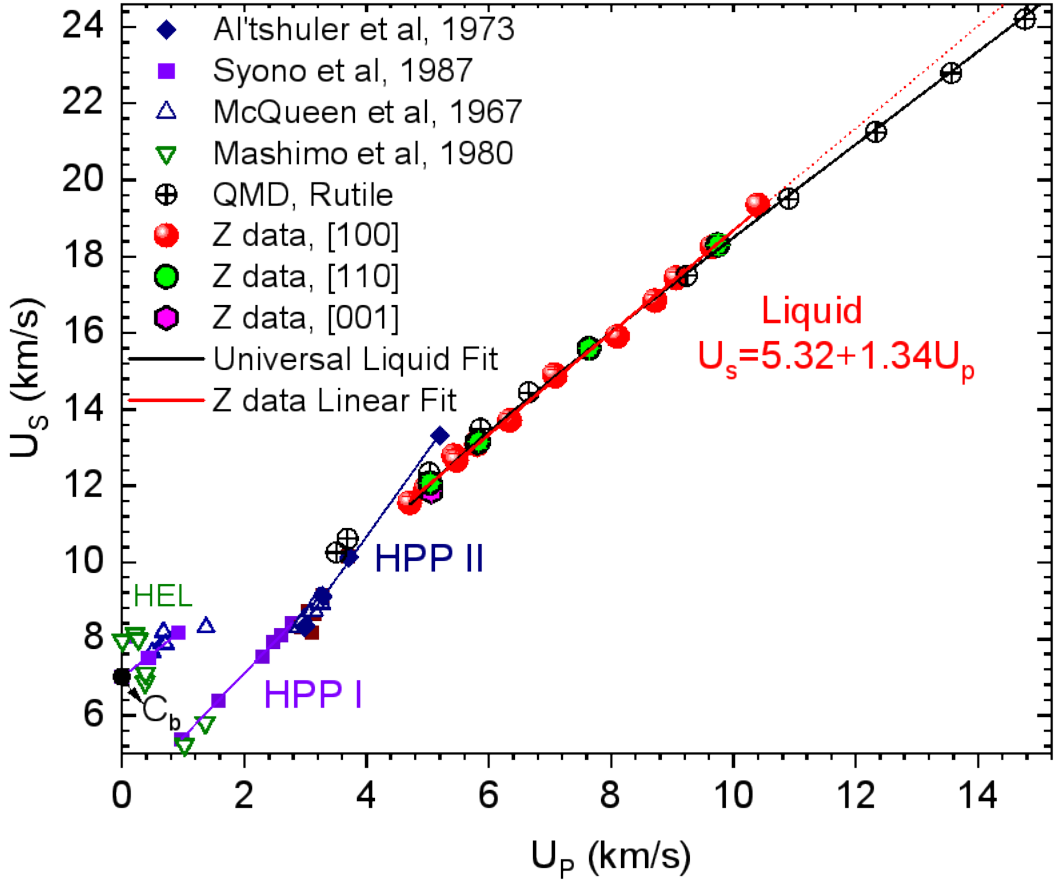
$$P_1 - P_0 = \rho_0(U_S - U_0)(U_1 - U_0)$$

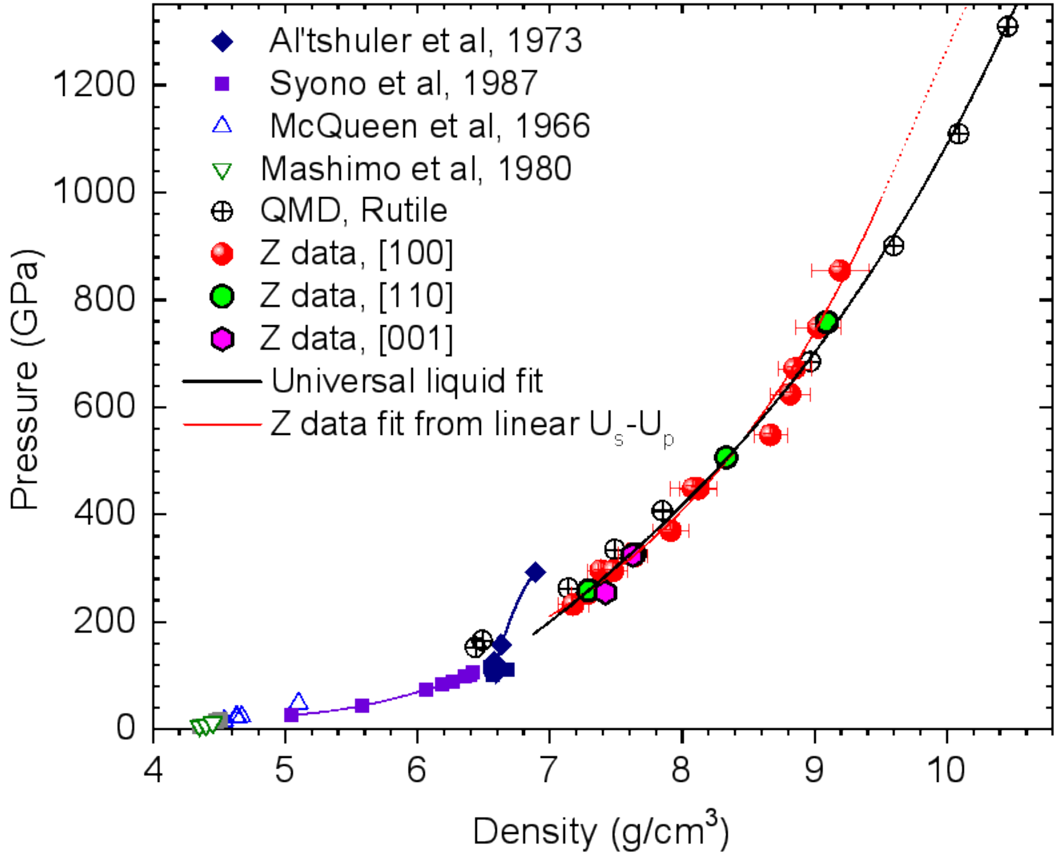
$$E_1 - E_0 = \frac{1}{2}(P_1 + P_0)(V_0 - V_1)$$

Twenty Z shots at different flyer velocities

Shot	Orientation	Window	V_F (km/s)	U_P (km/s)	U_S (km/s)	ρ (g/cm^3)	P (GPa)
Z3297S6[c]	[100]	TPX	22.49 ± 0.09	9.66 ± 0.07	18.24 ± 0.20	9.03 ± 0.11	748.8 ± 6.3
Z3297N6[c]	[100]	TPX	21.10 ± 0.06	9.06 ± 0.05	17.43 ± 0.15	8.85 ± 0.13	671.3 ± 4.4
Z3267S5[c]	[100]	TPX	20.24 ± 0.06	8.72 ± 0.06	16.83 ± 0.18	8.82 ± 0.15	623.5 ± 4.7
Z3267N5[c]	[100]	TPX	18.77 ± 0.06	8.10 ± 0.05	15.90 ± 0.15	8.67 ± 0.13	547.7 ± 3.9
Z3343N6	[100]	TPX	16.52 ± 0.09	7.08 ± 0.06	14.86 ± 0.18	8.12 ± 0.14	447.3 ± 4.5
Z3274S5	[100]	TPX	14.74 ± 0.06	6.34 ± 0.05	13.71 ± 0.18	7.91 ± 0.14	369.8 ± 3.5
Z3274N5	[100]	TPX	13.59 ± 0.06	6.35 ± 0.05	13.18 ± 0.14	7.62 ± 0.10	326.3 ± 2.8
Z3325N1[a]	[100]	TPX	12.83 ± 0.09	5.44 ± 0.03	12.79 ± 0.15	7.39 ± 0.11	295.2 ± 3.4
Z3427N5	[100]	TPX	10.94 ± 0.05	4.71 ± 0.05	11.55 ± 0.18	7.18 ± 0.12	231.2 ± 2.5
Z3441S7	[100]	Quartz	24.27 ± 0.09	10.40 ± 0.08	19.34 ± 0.26	9.19 ± 0.22	854.9 ± 8.0
Z3343N7	[100]	Quartz	16.52 ± 0.14	7.07 ± 0.09	14.91 ± 0.26	8.08 ± 0.18	448.2 ± 6.2
Z3391S2	[100]	Quartz	13.51 ± 0.06	5.80 ± 0.05	13.08 ± 0.14	7.64 ± 0.10	322.5 ± 2.8
Z3325N5[b]	[100]	Quartz	12.77 ± 0.07	5.47 ± 0.05	12.67 ± 0.06	7.48 ± 0.10	294.5 ± 3.0
Z3396S3	[100]	Quartz	11.56 ± 0.05	4.97 ± 0.05	11.93 ± 0.19	7.29 ± 0.13	252.0 ± 2.7
Z3441N7	[110]	TPX	22.68 ± 0.07	9.75 ± 0.07	18.30 ± 0.25	9.09 ± 0.21	758.2 ± 6.9
Z3343S3	[110]	TPX	17.81 ± 0.09	7.64 ± 0.06	15.59 ± 0.14	8.33 ± 0.12	506.1 ± 4.3
Z3391N5	[110]	TPX	13.15 ± 0.15	5.84 ± 0.05	13.15 ± 0.15	7.64 ± 0.11	326.2 ± 2.9
Z3396N5	[110]	TPX	11.74 ± 0.08	5.04 ± 0.05	12.08 ± 0.15	7.29 ± 0.11	258.7 ± 3.0
Z3391N2	[001]	TPX	13.56 ± 0.08	5.82 ± 0.06	13.14 ± 0.19	7.63 ± 0.14	325.0 ± 3.7
Z3396N2	[001]	TPX	11.70 ± 0.07	5.06 ± 0.05	11.85 ± 0.15	7.42 ± 0.11	254.9 ± 2.8

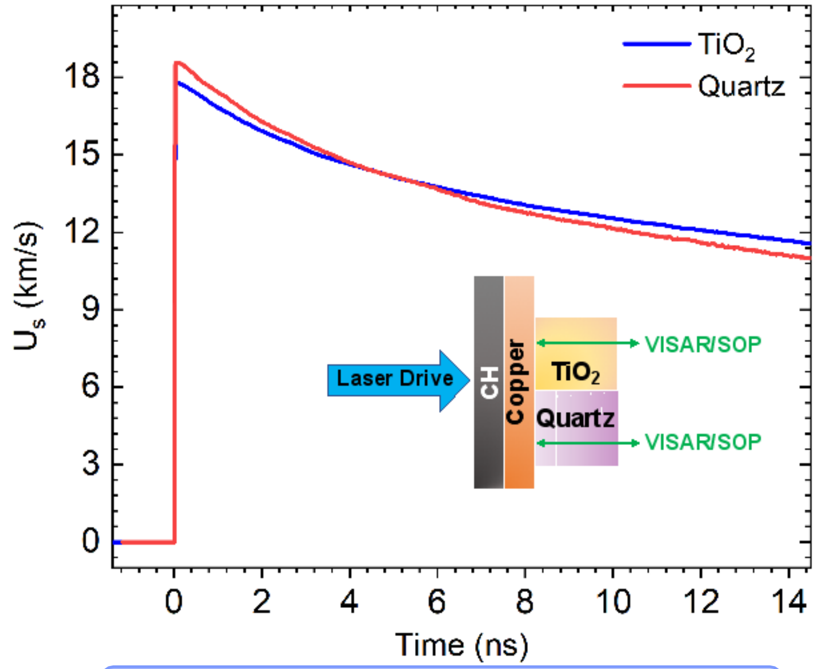
Z experimental Results



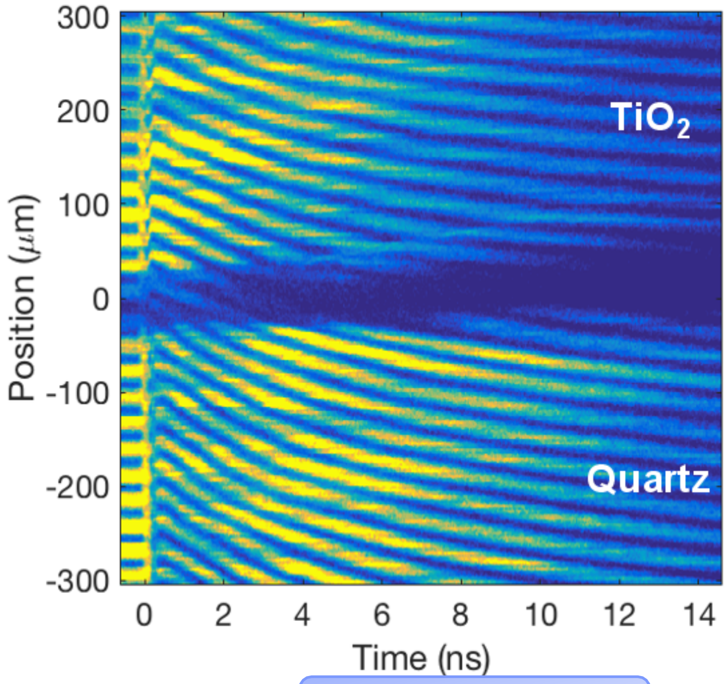


- More compressible Hugoniot data obtained from Z → strikingly different than what Al’tshuler reported
- Not incompressible at least up to 855 GPa
- Z-data are in excellent agreement with the QMD data
- No-orientation dependence observed indicating possible transformation to a liquid phase

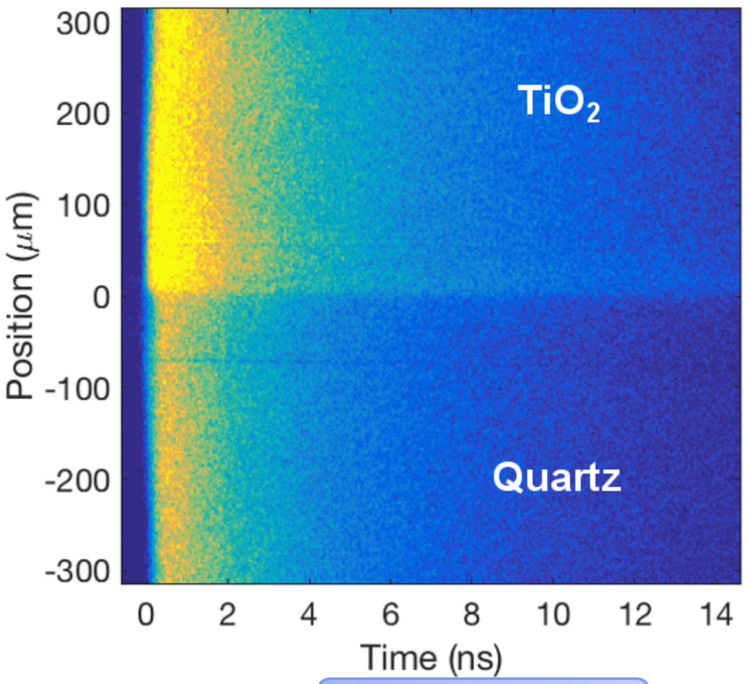
Omega decaying shock experiments to measure temperature



Decaying shock in TiO$_2$ and Quartz



Raw VISAR data

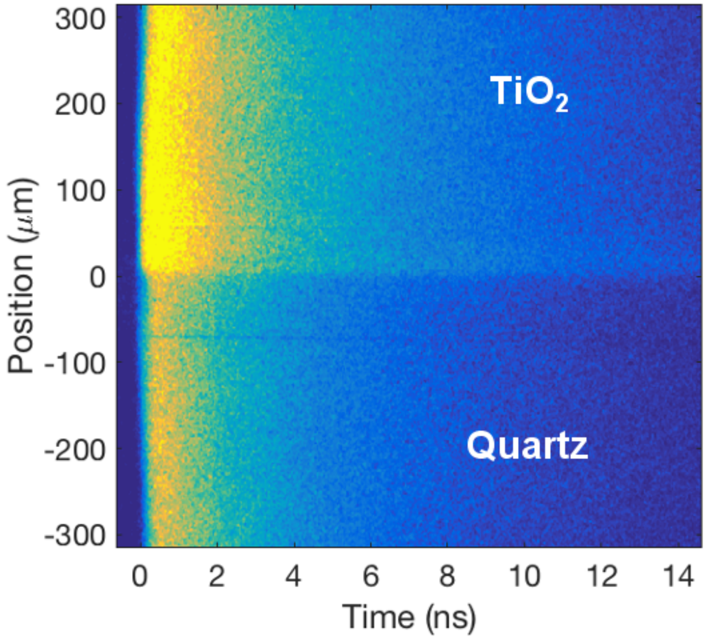


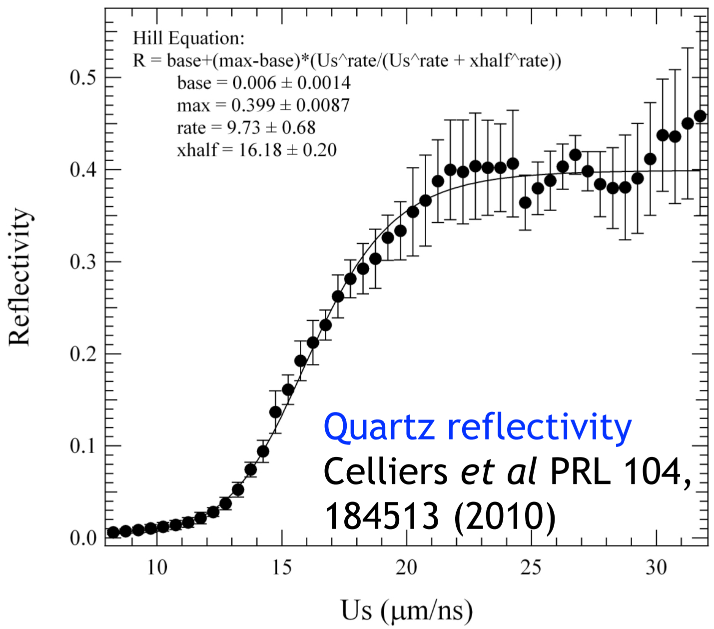
Raw SOP data

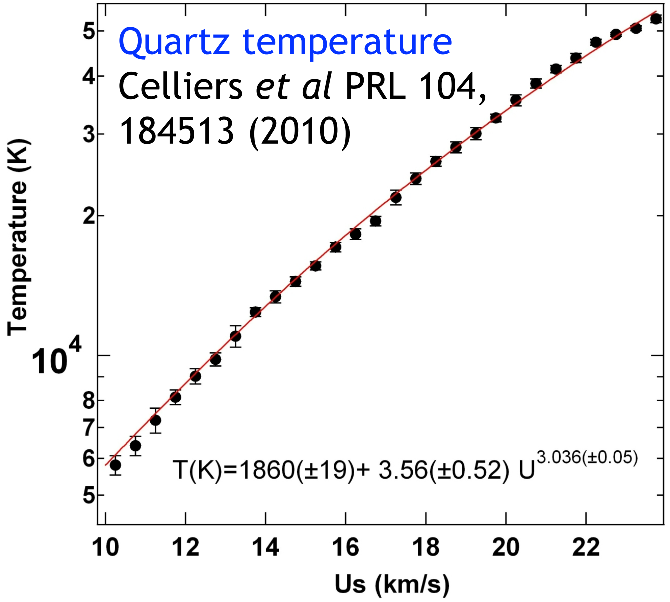
- Z-data lacks information on temperature
- Laser driven shock decay experiments to measure shock velocity as a function of temperature
- CH→ ablator; Copper→ preheat shield/reflectivity standard
- Temperature determined from emission and reflectivity
- SOP collects emission between 590-850 nm → VISAR measures shock reflectivity (532 nm) (corrected for window transmission)→ Temperature

Chad McCoy

Omega decaying shock experiments to measure temperature

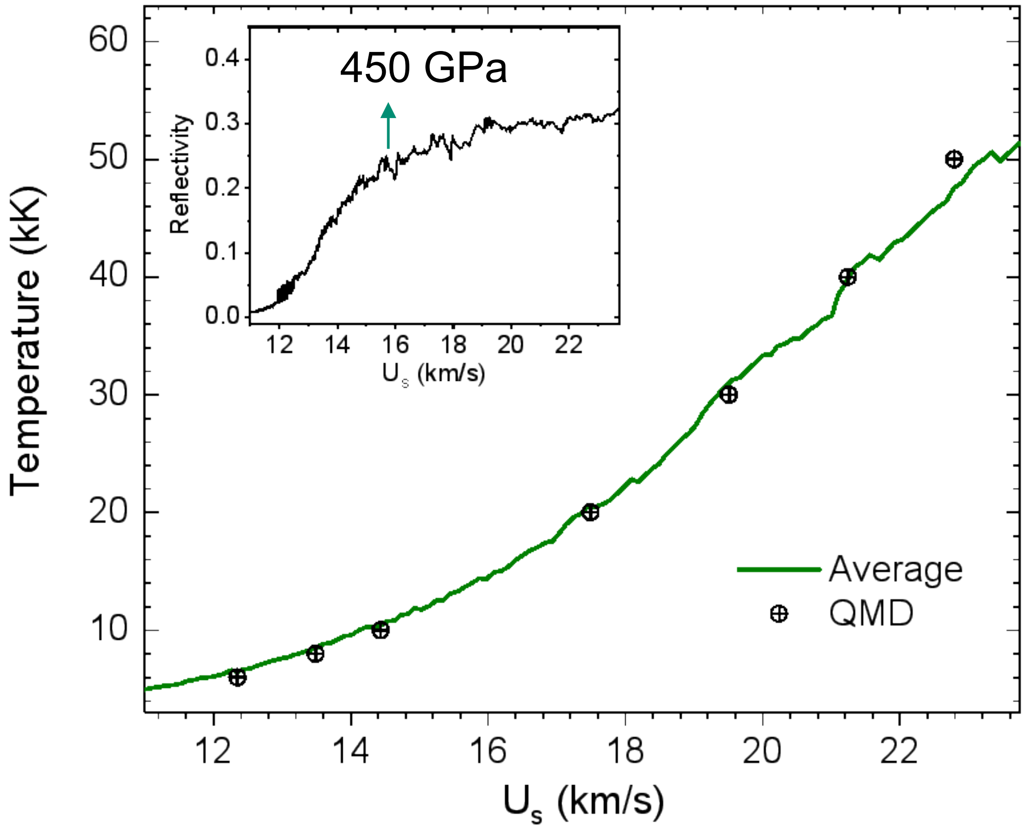


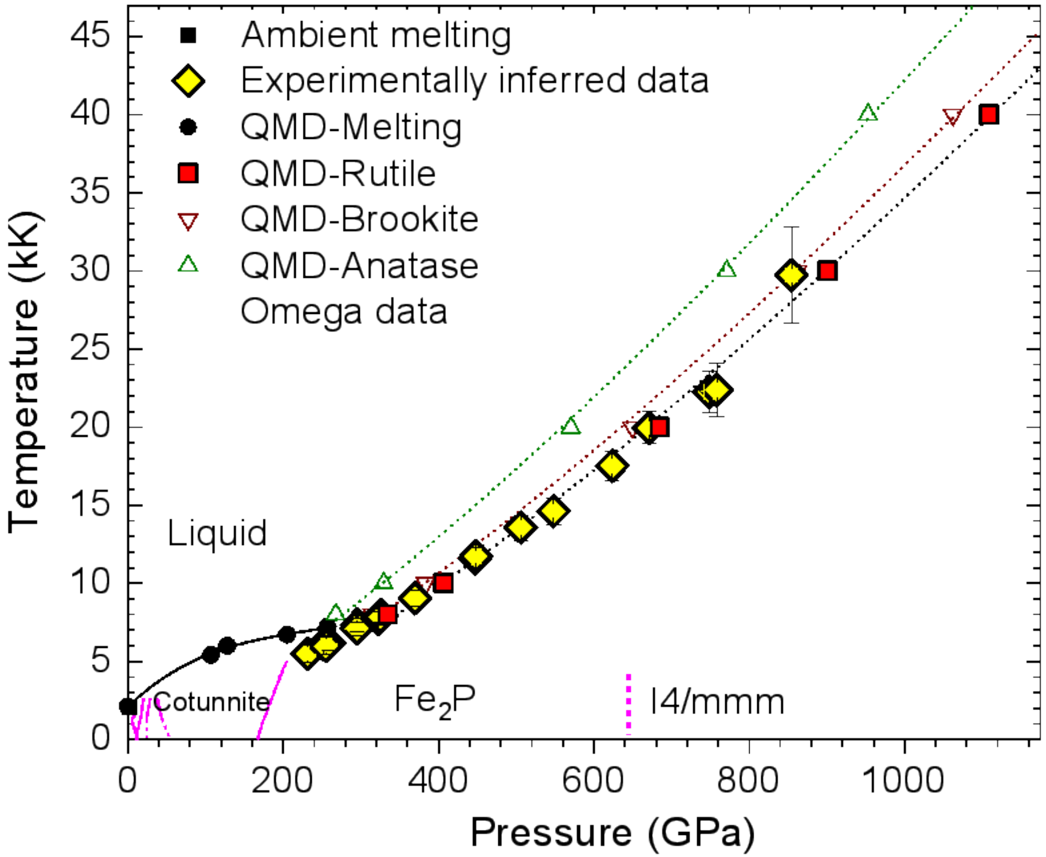




- Data calibrated to shock temperature in quartz
- SOP temperature determined from intensity, I, using gray-body equation: $T = \frac{T_0}{\ln\left(1+\frac{A}{I}\right)}$
- T_o and A, are calibration coefficients determined from quartz emission and known temperature as a function of shock velocity

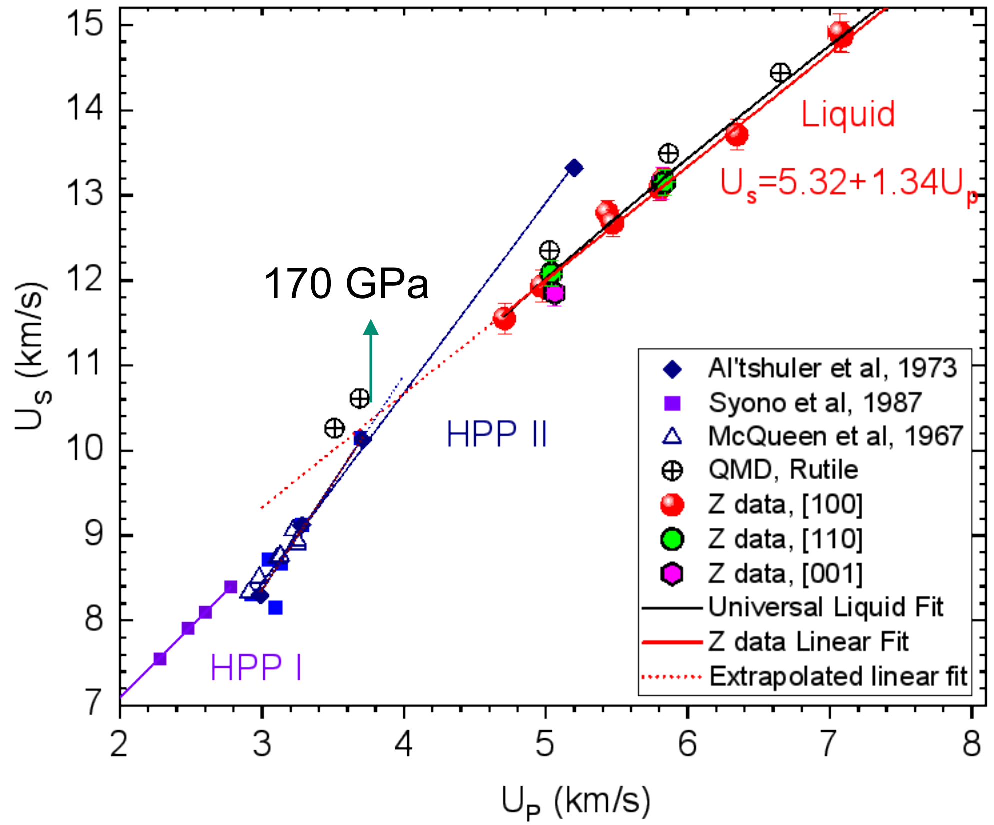
Omega data suggest liquid TiO_2 above 240 GPa

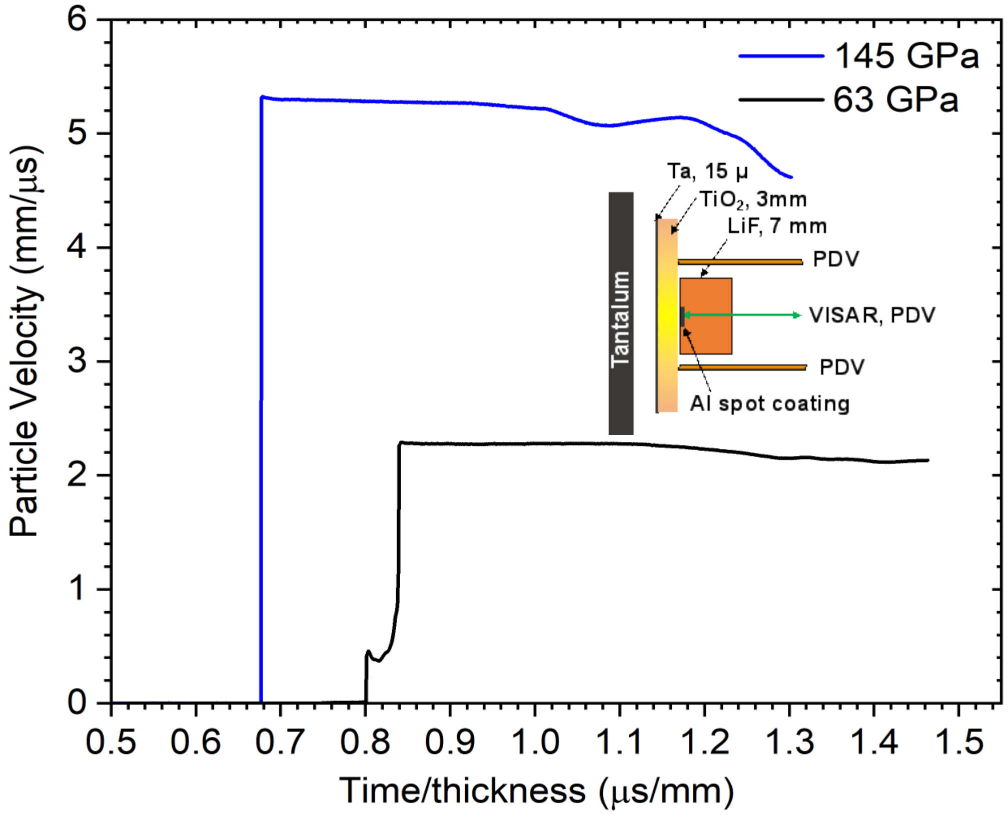


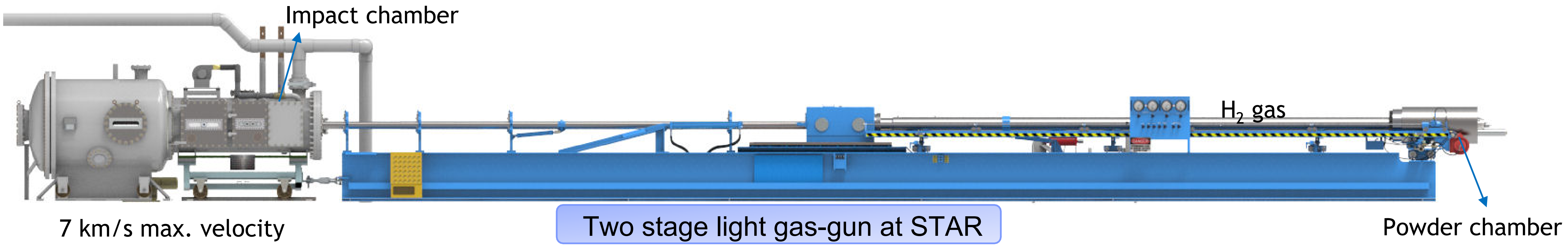


- Temperature measurements from four different shots: 5, 000K to 52, 000K for U_s of 10.9 to 24 km/s
- Shock temperature increases monotonically with increasing U_s: no kink or plateau observed
- Increase in reflectivity suggests liquid phase
- Measured data consistent with QMD calculations
- Combined the temperature data from Omega and the pressure data from Z-experiments

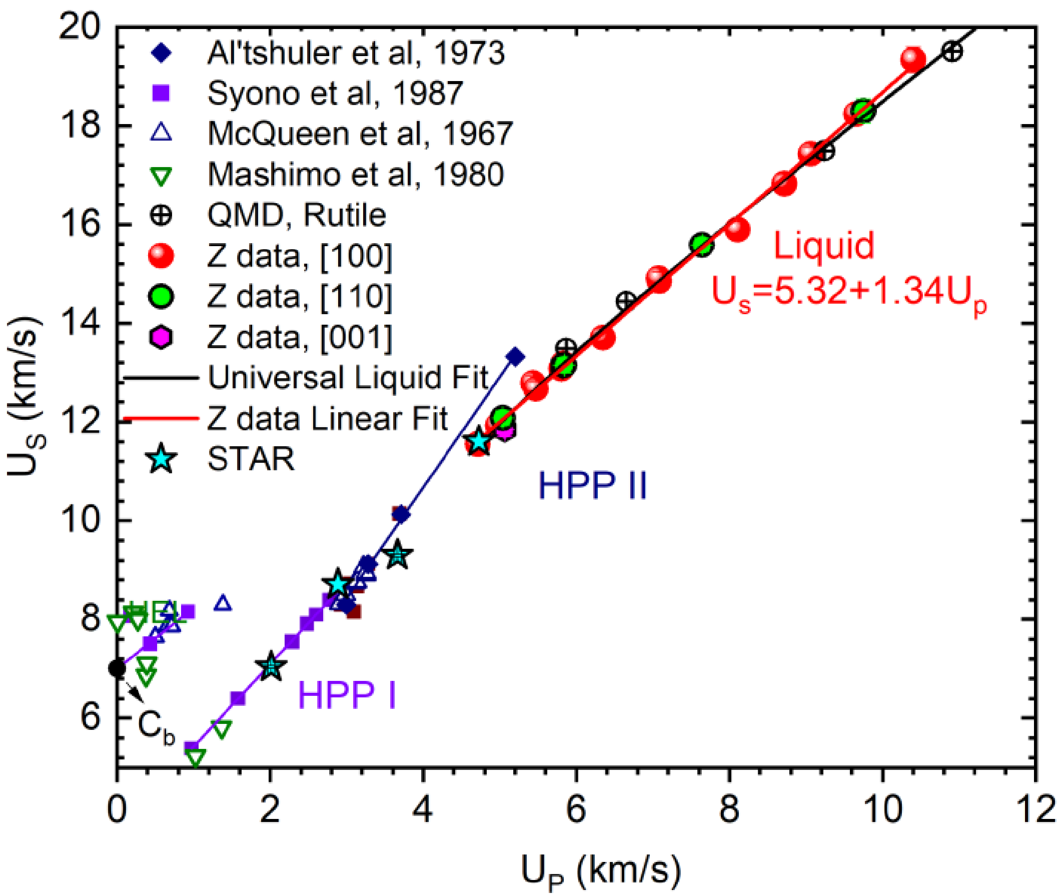
Transition from HPPII to liquid

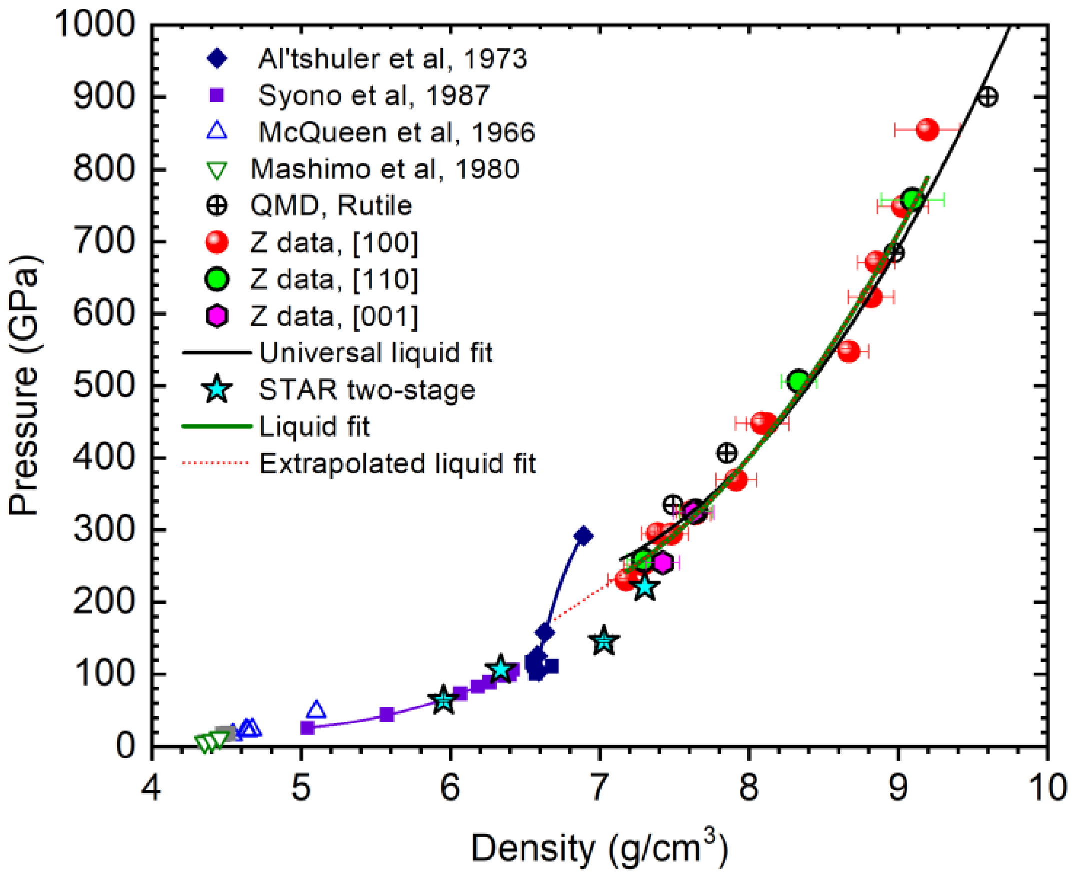






Transition from HPPII to liquid





- Gas-gun data agrees with the literature data
- HPPII data deviates from Al'tshuler's data
- More experiments planned to resolve the discrepancy

Acknowledgements

- Org. 1646: Chris Seagle, Marcus Knudson, Seth Root, Chad McCoy, Dan Dolan, Tommy Ao, Jus
  Jean-Paul Davis, Paul Specht, Sean Grant
- DICE: Josh Usher, Keith Hodge, Steven Dean, Lena Pacheco, Sheri Payne, Richard Hack
- STAR: Bill Reinhart, Scott Alexander, John Martinez, Bernardo Farfan, Robert Palomino, Rafael
- Z: Leo Molina, Roger Harmon and all of the Z technologists
- Heath Hanshaw, Jim Williams, John Fisher
- Diagnostic Support Staff

Thank you!