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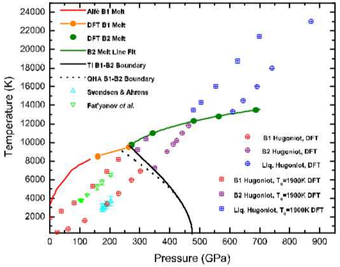

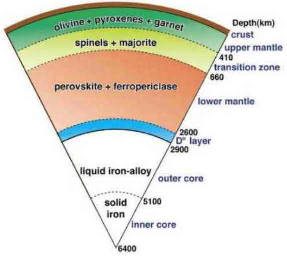

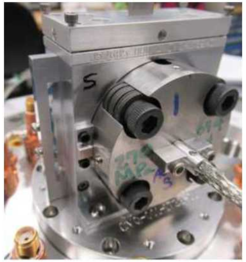

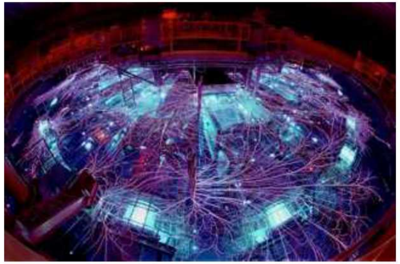

Integrating first-principles theory and multi-Mbar shock experiments for understanding phase transitions at extreme conditions

Thomas R. Mattsson, Sandia National Laboratories

Fall 2018 ACS Meeting, Boston, August 23, 2018

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Outline and acknowledgments

- Why study minerals under high pressure?
- What is Pulsed Power?
- MgO (periclase) – the end member of ferropericlase
- Mg_2SiO_4 (forsterite) – end member of olivine
- Outlook and other ongoing work

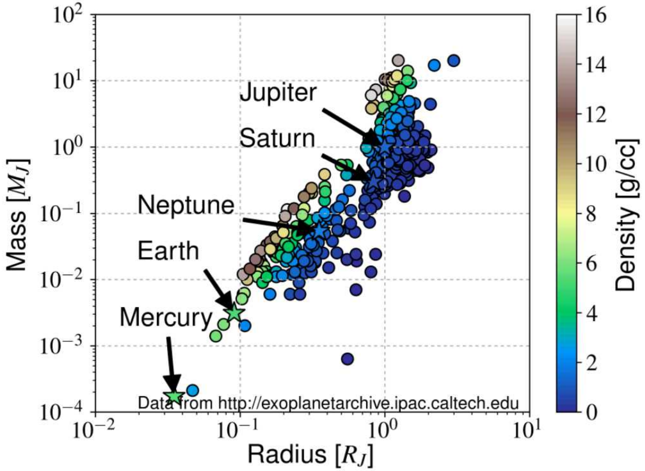
Sandia: Seth Root, Luke Shulenburger, Joshua Townsend, Raymond Clay, Kyle Cochrane, Dave Bliss, Dan Dolan, Ray Lemke, Mike Desjarlais, and Dawn Flicker

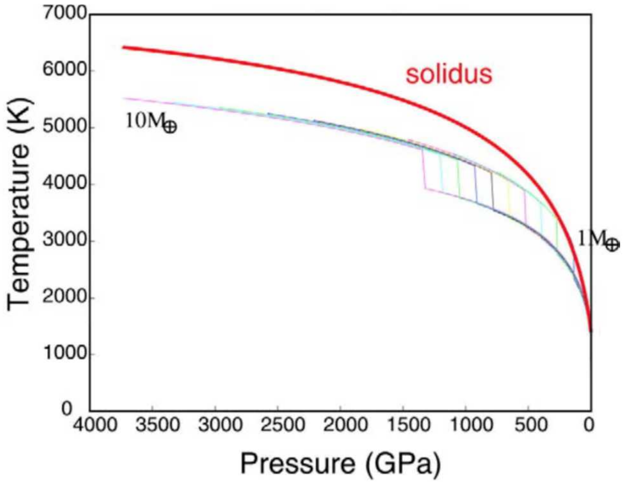
UC Davis: Sarah Stewart, Erik Davies, and Dylan Spaulding

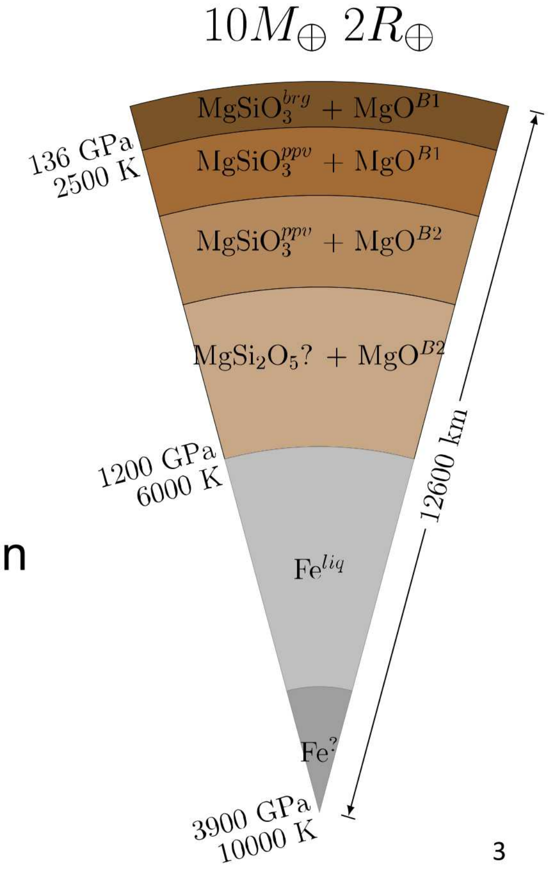
Harvard: Stein Jacobsen and Li Zeng

LLNL: Dayne Fratanduono, Rick Kraus, and Marius Millot

How to access extreme states for planetary science?

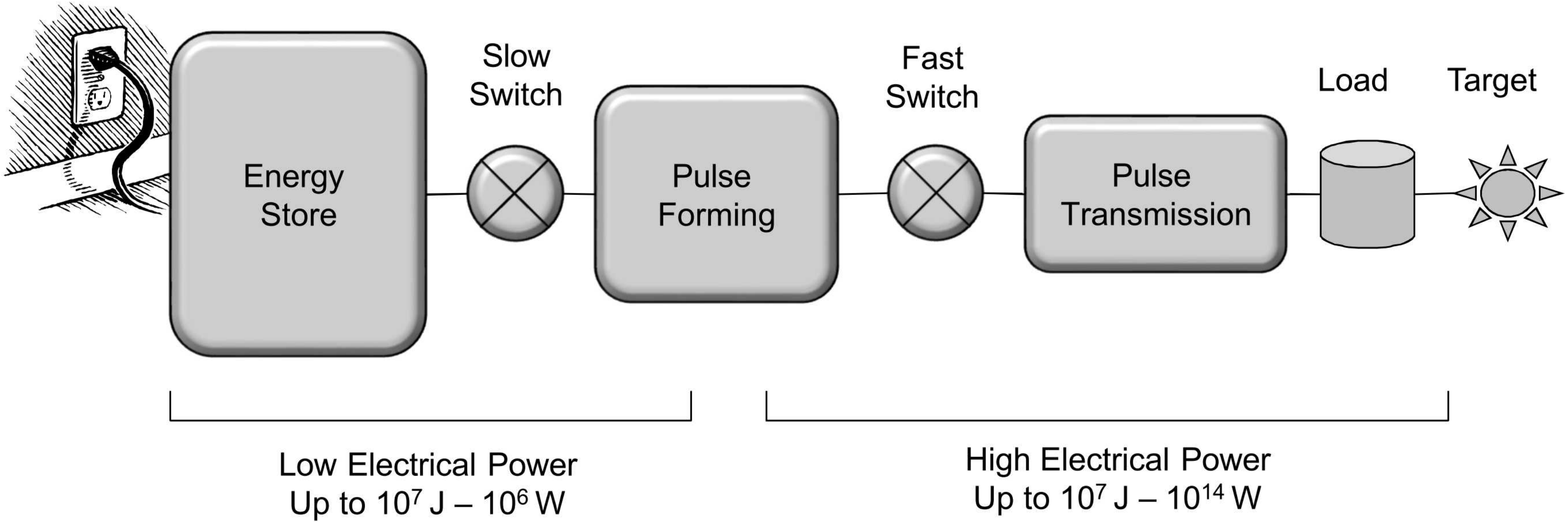






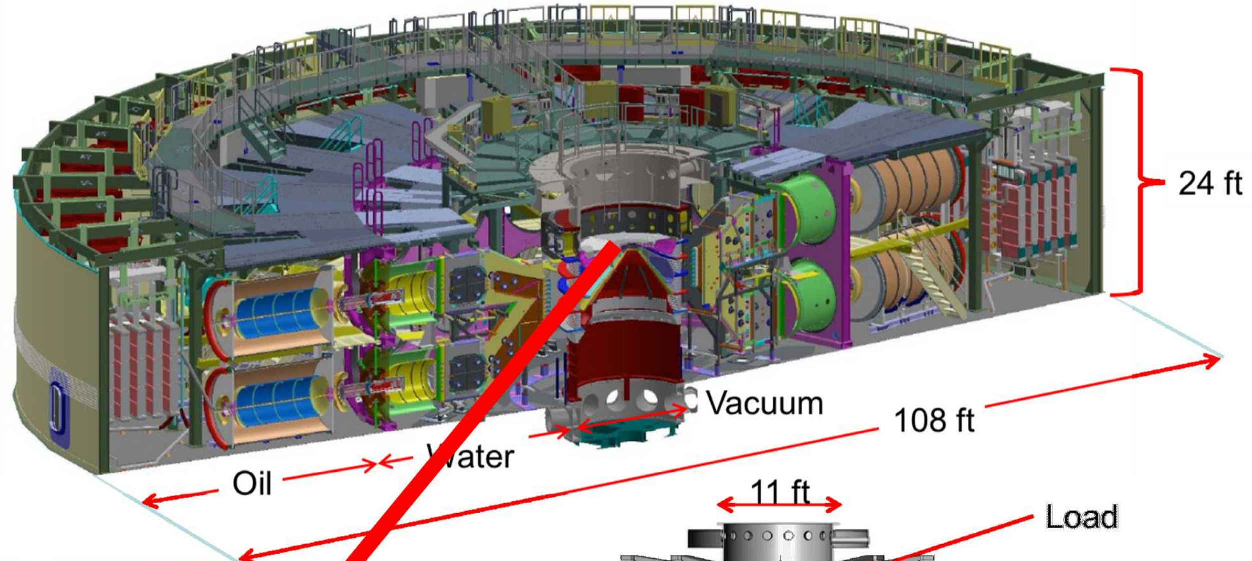
- Rapid planetary discovery driven by NASA's Kepler mission
- Challenges our understanding of structure, dynamics, etc.
- Static compression techniques span Earth conditions
- Dynamic compression techniques allow direct measurements of planetary materials at relevant *V,P,T*

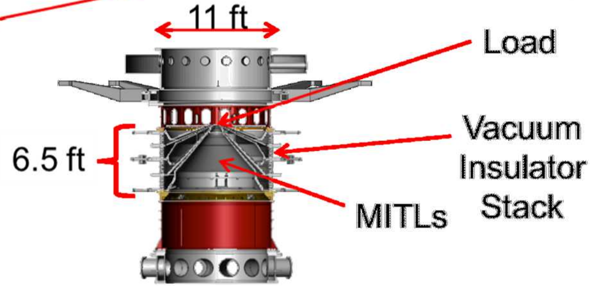
Pulsed power: The temporal compression of electrical energy to produce short bursts of high power

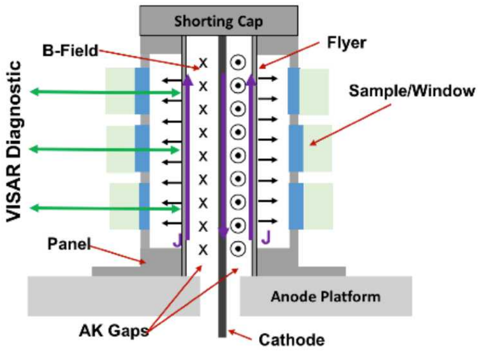


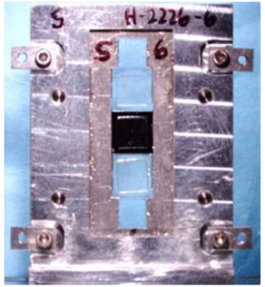
Dynamic compression experiments on the Z Machine

- Z is 33 x 8 m to store the energy and shape the pulse
- The load is 4x1x1 cm





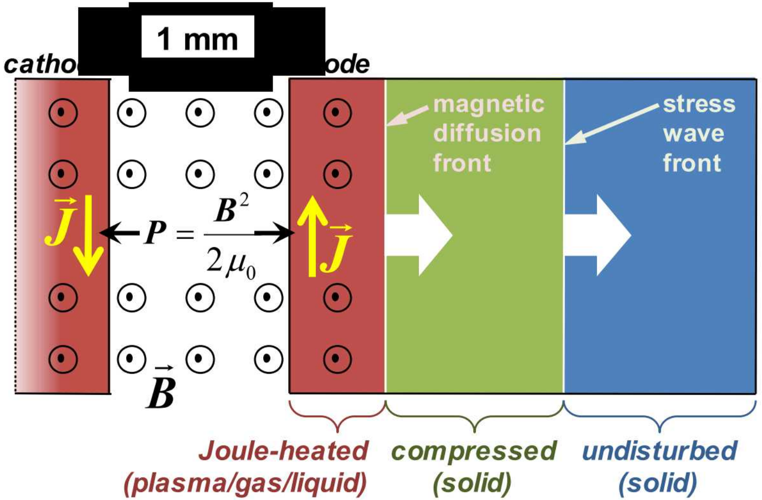




Credit: S. Root (SNL)

Using magnetic pressure as a source has unique advantages

- In Magneto Hydrodynamics (MHD) – a magnetic field is equivalent to a scalar pressure
- *Can create high pressures without heating*
- Generated over long time scales (100-1200 ns) with control over the pressure pulse
- Large samples (mm to cm)
  - Allows HED conditions in sample sizes >> sample grain boundary dimensions
  - One Z experiment can field 6-20 samples all experiencing identical drive



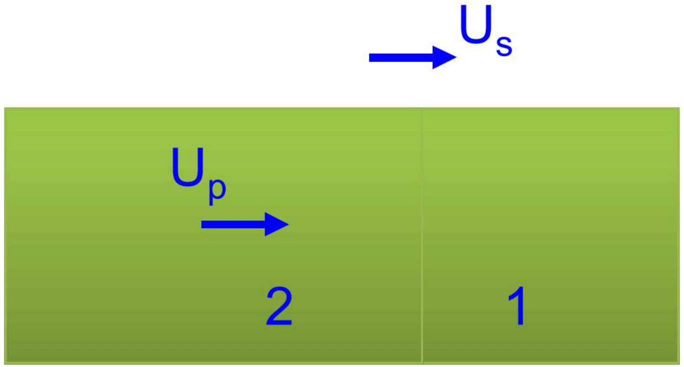
The stress/pressure front runs faster than the magnetic field/melt – a fluke of nature…

Opportunity for theory and experiments to complement each other in analyzing shock compression

- Dynamic experiments on Z have limitations
  - Main diagnostics is velocimetry – yields ρ,P
  - Measuring temperature is only a recent capability
  - No XRD for microstructure
- Ab initio methods have limitations
  - Free energy is difficult and has uncertainties
  - “Band gap problem” in Density Functional Theory (DFT)
  - Modest system sizes / finite size effects

We use high precision dynamic experiments to tightly constrain knowledge of the material behavior, calculations are then validated and used to fill in missing information

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

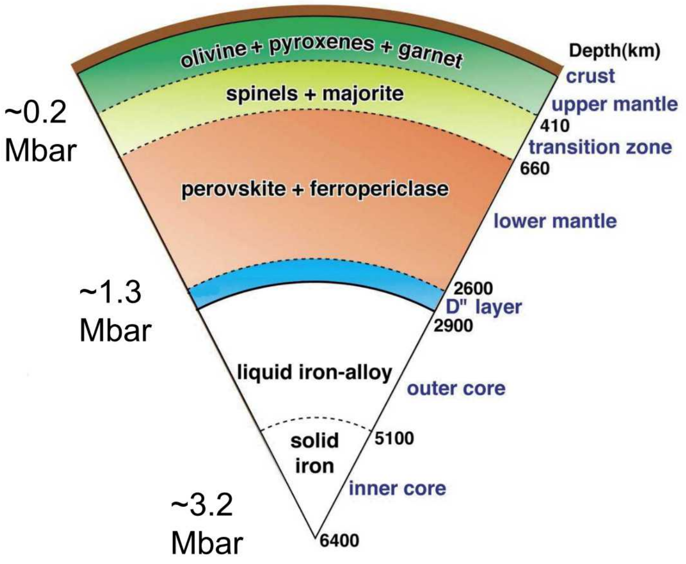


$$2\left(E_2 - E_1\right) = \left(P_2 + P_1\right)\left(\upsilon_1 - \upsilon_2\right)$$

We utilized the Z-machine to investigate the behavior of MgO at multi-Mbar pressure

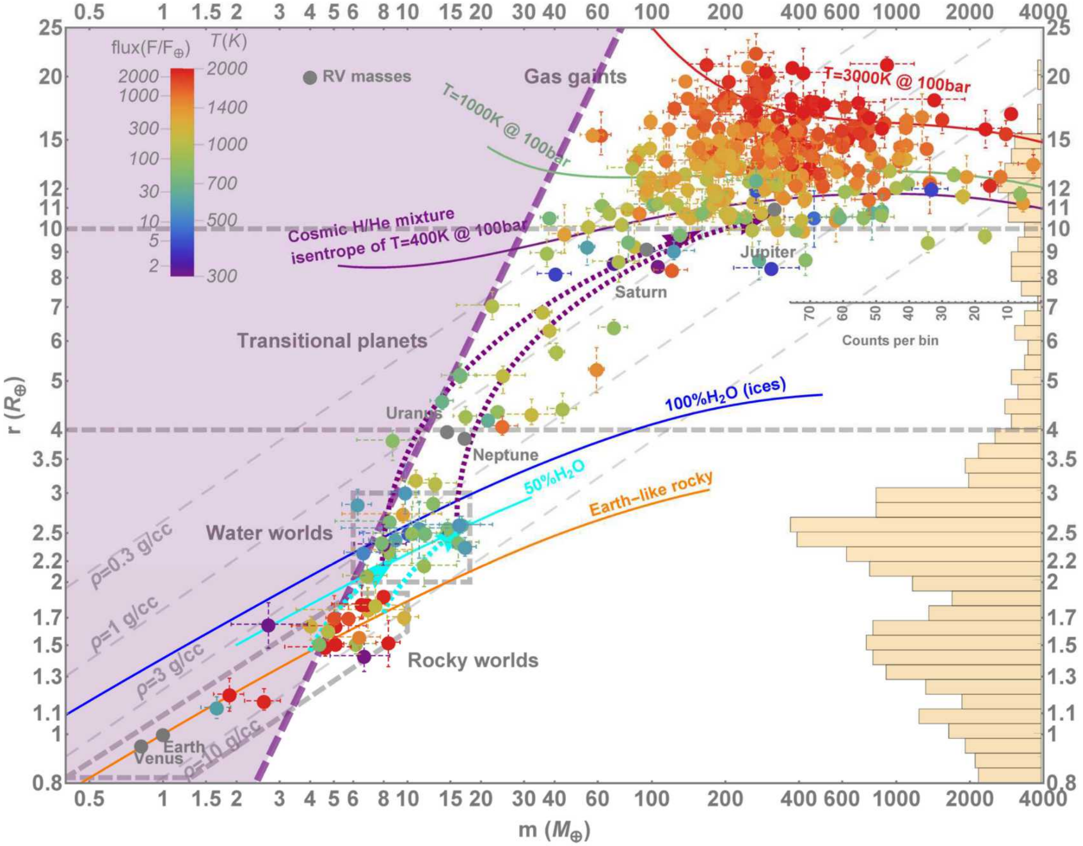
- MgO (periclase) is an end member of the $Mg_{1-x}Fe_xO$ mineral ferropericlase that comprises a large fraction of the earth's mantle
- Useful check of experimental methodology: simple structure and transport properties
- The range of conditions inside earth is relatively small – from DAC we know that MgO is boring under terrestrial conditions

Earth Composition vs Depth



Exo-planet distribution: rocky worlds, water-worlds, and gas giants

- Exo-planets are clustered
- Gas giants (Jupiter and super-Jupiters)
- Rocky worlds (Earth, Mars, Venus, super-Earths)
- Water worlds (*missing in the solar system*)



Li Zeng, Stein B. Jacobsen,…,TRM, et. al. Submitted to PNAS (2018).

Exo-planet distribution: rocky worlds, water-worlds, and gas giants

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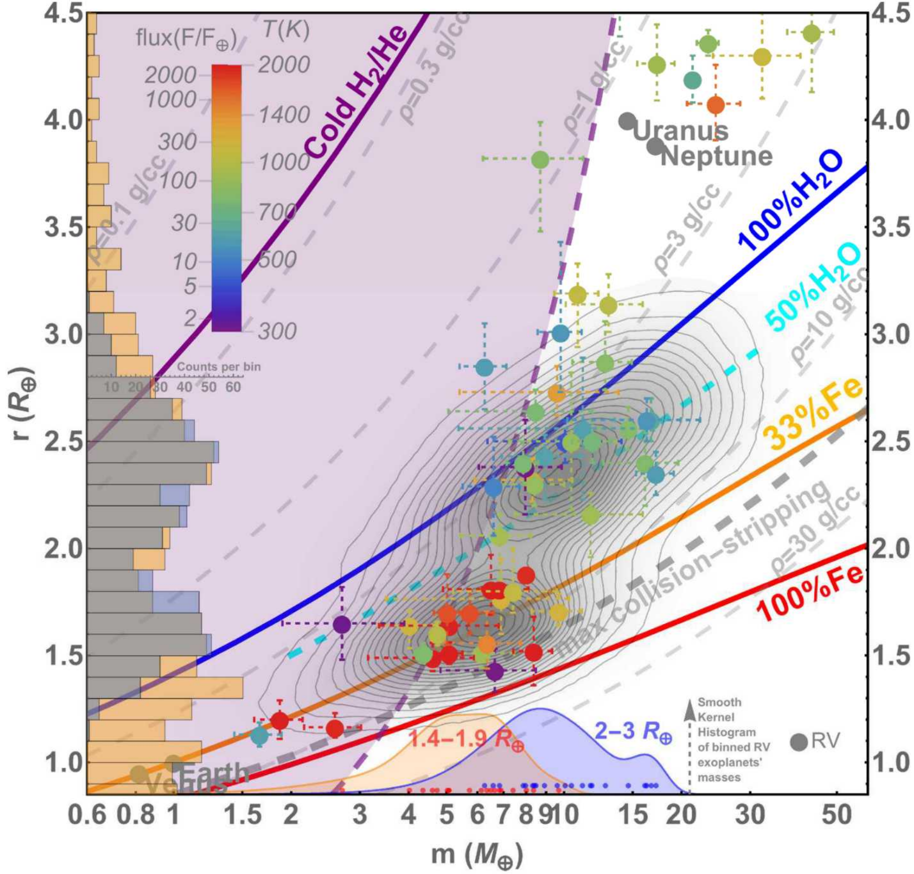
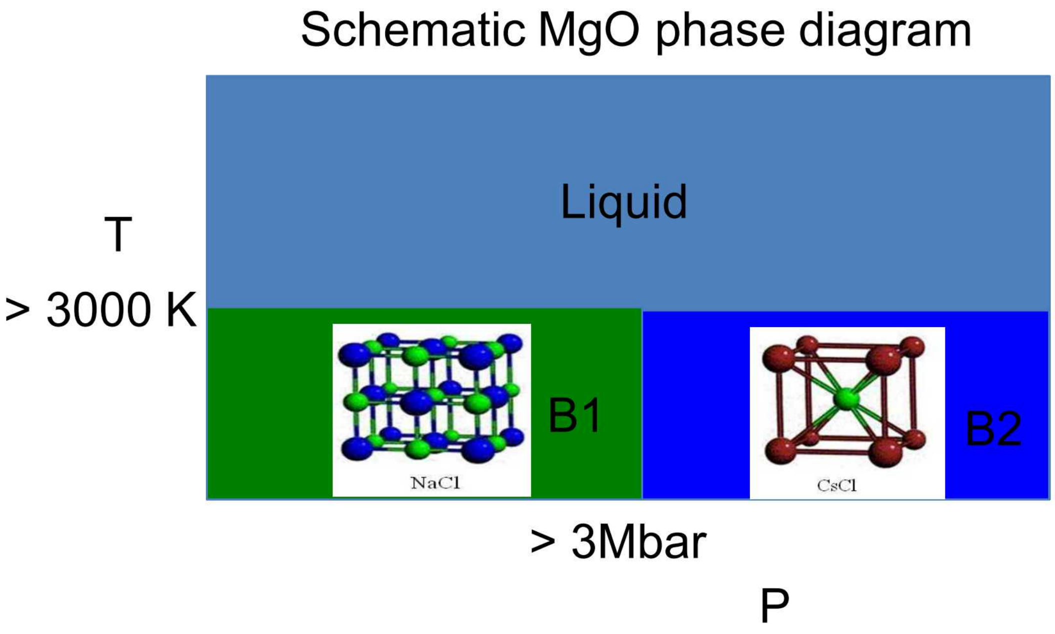
- The center of rocky planet distribution is
  - 5-6 Earth masses
  - 1.5 Earth radii
- Significantly higher pressure in the mantle and core
- Where is melt, for plate tectonics, magnetic dynamo, and thus life?



Li Zeng, Stein B. Jacobsen,…,TRM, et. al. Submitted to PNAS (2018).

Key features of the MgO phase diagram

- MgO occurs in two solid phases
- The nature of solid phases can have large impact on structure and transport in planets
- The melt boundary at high pressures is not well known
  - Important for giant impacts and super-earth planets



Preliminary analysis showed MgO data was inconsistent with extrapolation from low pressure B1 state

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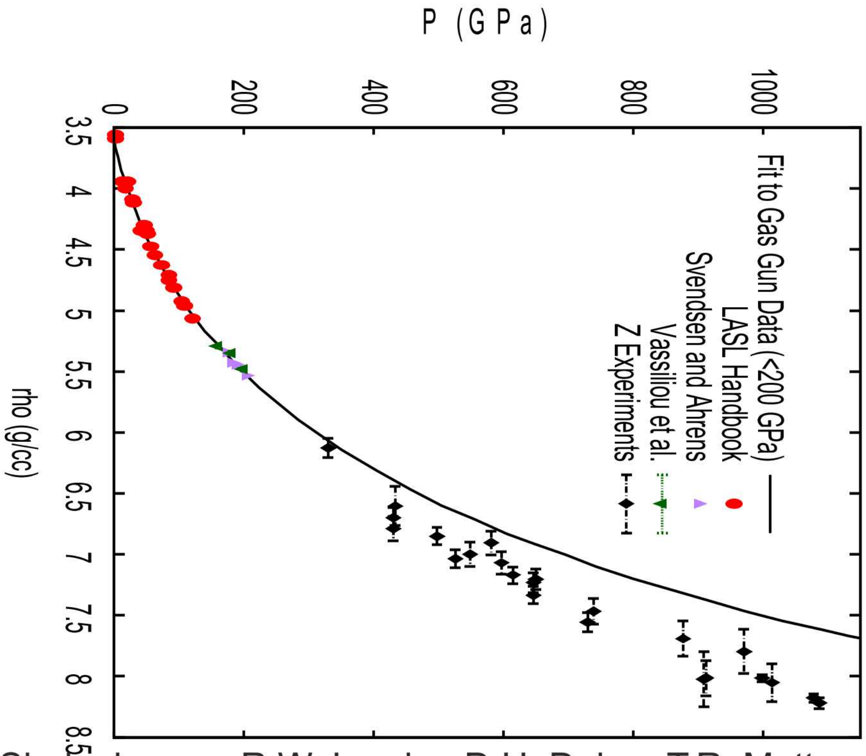
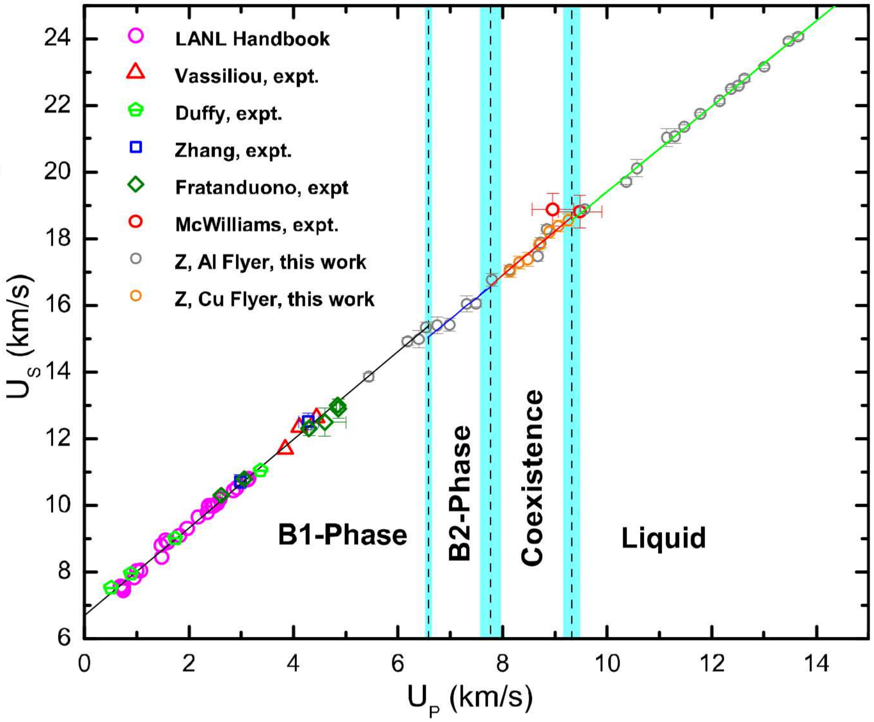
- At 330 GPa, the Hugoniot point lies on the extrapolation of the gun data fit
- Slope change starting at 440 GPa – suggests a phase transition
- *No obvious slope changes at higher pressures that would suggest melt*



S. Root, L. Shulenburger, R.W. Lemke, D.H. Dolan, T.R. Mattsson, and M.P. Desjarlais, Phys. Rev. Lett **115**, 198501 (2015).

Careful analysis of refined shock data suggests a rich behavior

- Use *many* shots to map $u_s(u_p)$ along the Hugoniot – *with high accuracy*
- Phase changes are inferred by assuming $u_s(u_p)$ is linear in any given phase
- Similar method was applied to diamond (Knudson et al, Science **322**, 1822 (2008))
- Coincident with last break, the shock becomes reflective
- Assuming some knowledge of the phase diagram, suggests large region of coexistence between B2 and liquid

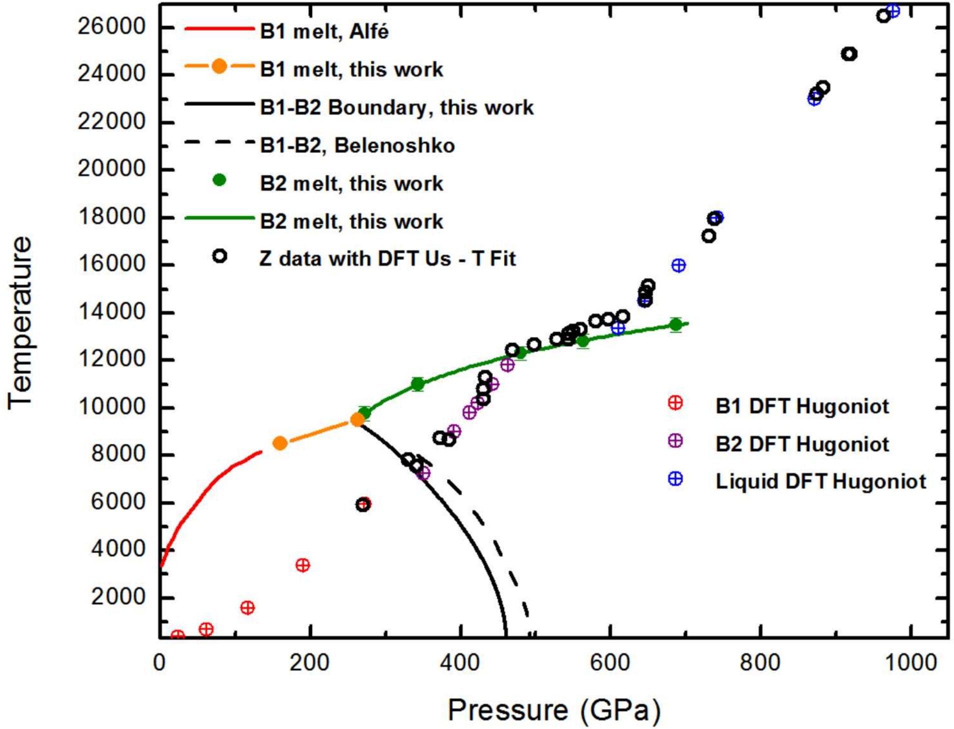


S. Root, L. Shulenburger, R.W. Lemke, D.H. Dolan, T.R. Mattsson, and M.P. Desjarlais, Phys. Rev. Lett **115**, 198501 (2015).

Theory and experiment together give a comprehensive picture of MgO at higher pressures

- Use U_s vs T fit from DFT calculations and apply to experimental data
- DFT confirms a large coexistence region between B2 and liquid on the Hugoniot

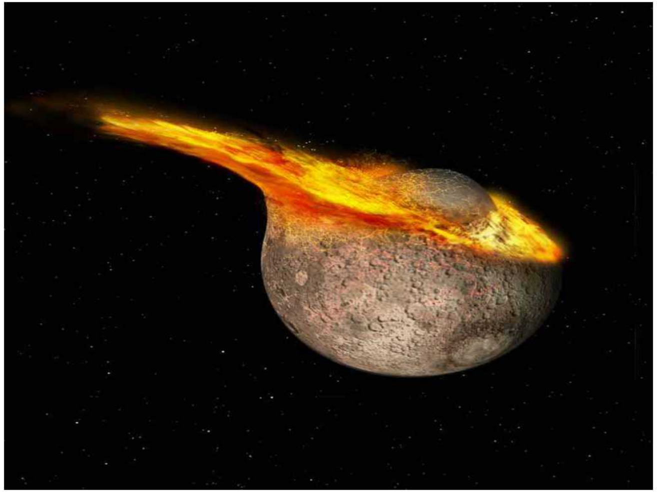
- Density driven phase change between B1 and B2
- Large difference in entropy between B2 and liquid along Hugoniot



S. Root, L. Shulenburger, R.W. Lemke, D.H. Dolan, T.R. Mattsson, and M.P. Desjarlais, Phys. Rev. Lett **115**, 198501 (2015).

Most recently, we have investigated Forsterite – Mg_2SiO_4

- Forsterite (Mg_2SiO_4) end member of olivine
- Major constituent of the Earth and other terrestrial planetary interiors
- Analytic EOS's fit to low pressure gun data P<200 GPa, higher pressure behavior poorly understood
- High-precision shock experiments on Z and extensive Density Functional Theory simulations

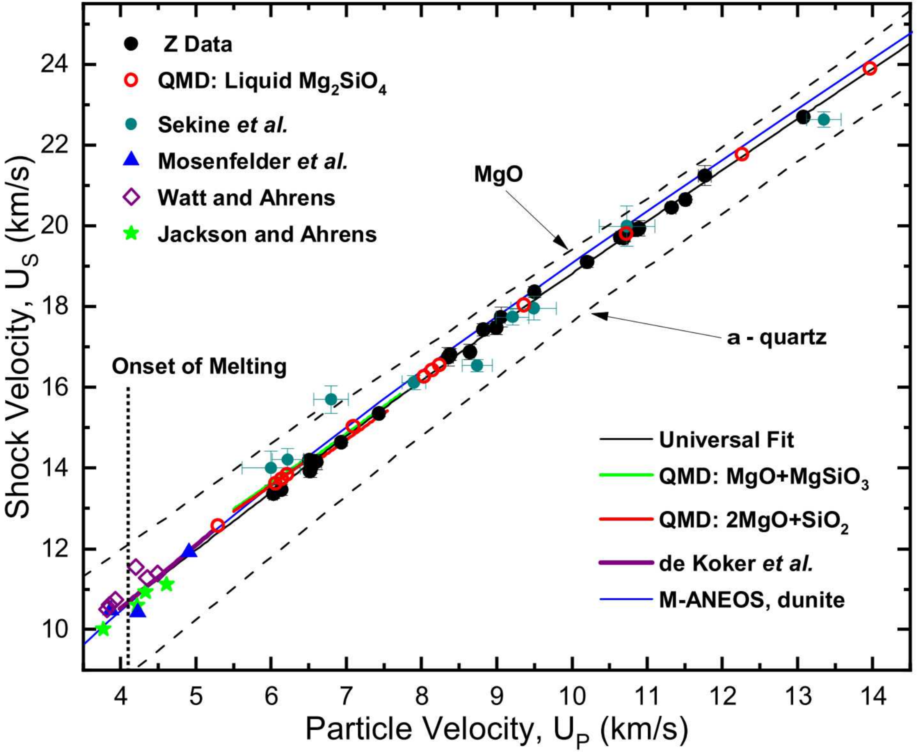


What happens in a giant impact scenario depends sensitively on principal Hugoniot states of planet

S. Root, J. Townsend, E. Davies, et. al. Geophysical Research Letters (2018).

The principal Hugoniot of forsterite to 950 GPa

- Sandia, Harvard, UC Davis, and LLNL collaboration
- 44 data points form Z spanning u_P 6-13 km/s
- Excellent agreement with previous low *P* data
- Curvature in Hugoniot due to compressibility increase in liquid



S. Root, J. Townsend, E. Davies, et. al. Geophysical Research Letters (2018).

Temperature measurements along the principal Hugoniot of forsterite to 950 GPa / 35,000 K validate DFT simulations

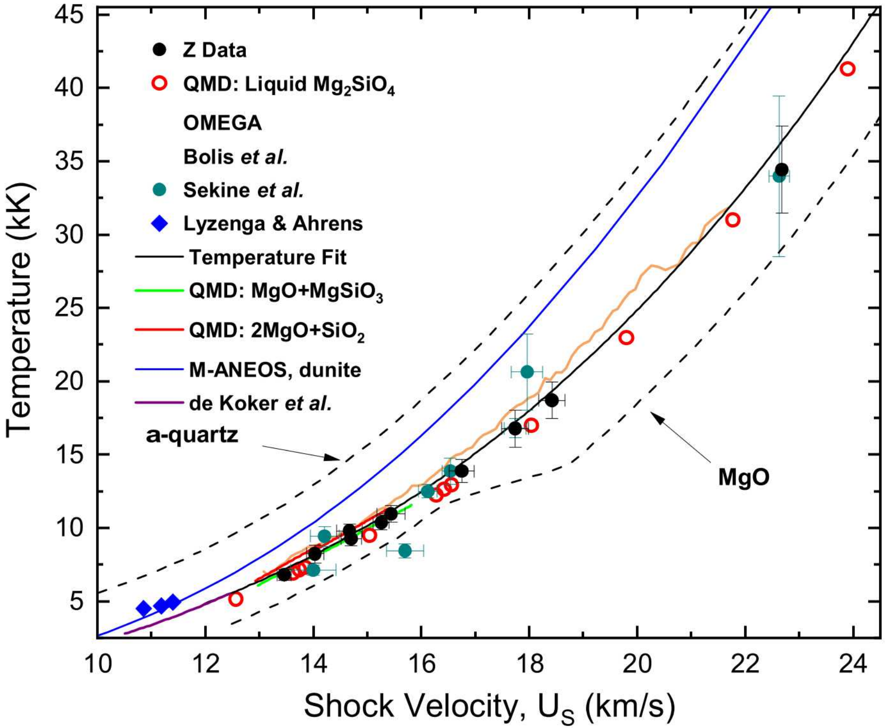
- *First Z optical pyrometry experiments*
- Important cross validation between OMEGA and Z experiments
- *Both agree with DFT-MD predictions*
  - *Validates the earlier MgO temperature inference*

Z + DFT is a powerful method to elucidate the properties of materials in extreme conditions

But, this is a multi-component system.

Does phase separation/recrystallization occur on principal Hugoniot?

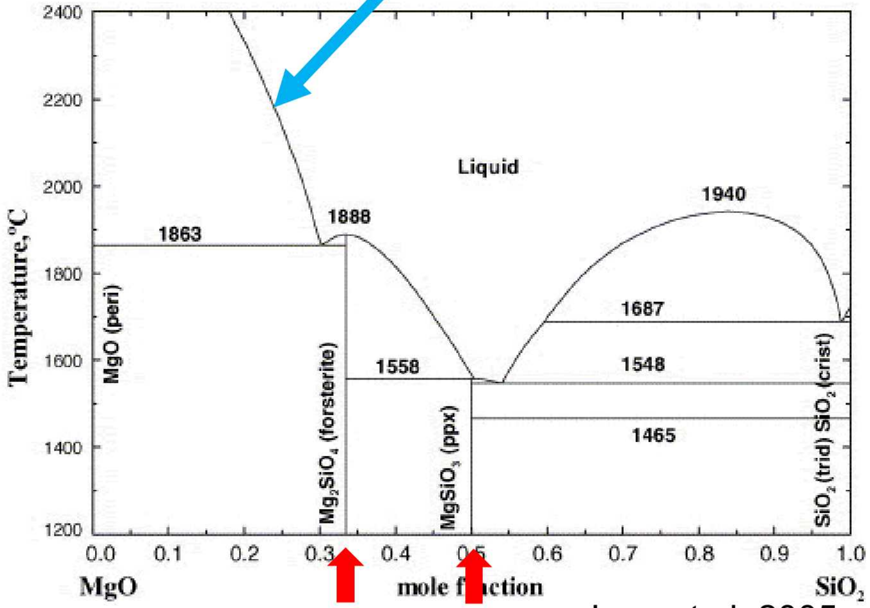
Sekine et. al. Sci. Adv., 2, e1600157 (2016).



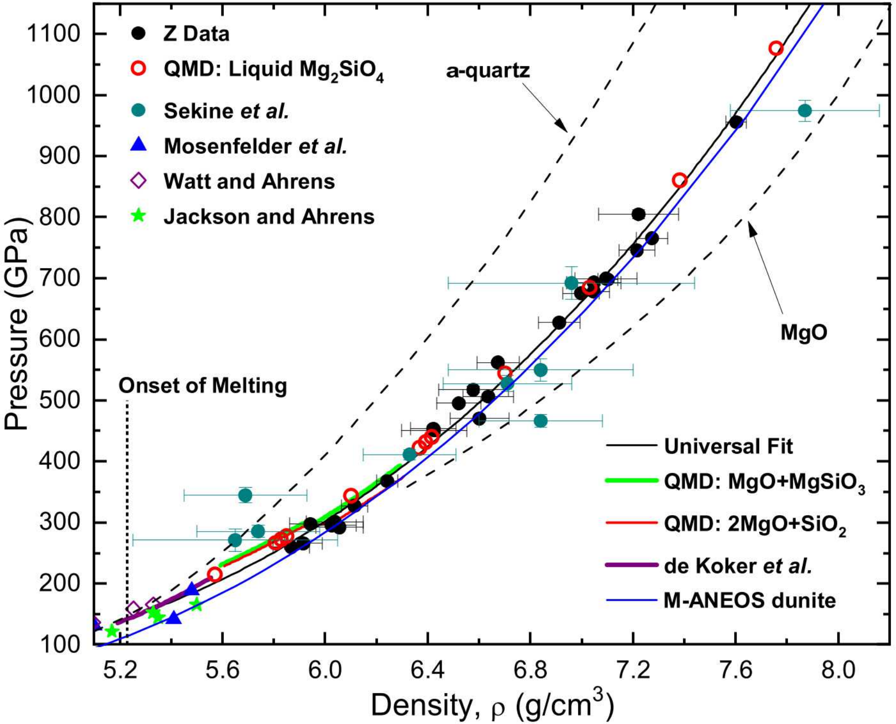
S. Root, J. Townsend, E. Davies, et. al. Geophysical Research Letters (2018).

No discontinuous change on the principal Hugoniot of forsterite: likely no phase separation at low P

$$\mathrm{Mg_2SiO_4^{\mathit{liq}}} \leftrightarrow \mathrm{MgO^{\mathit{sol}}} + \mathrm{MgSiO_3^{\mathit{liq}}}$$



Forsterite Mg2SiO4 **Enstatite MgSiO3** Jung et al. 2005

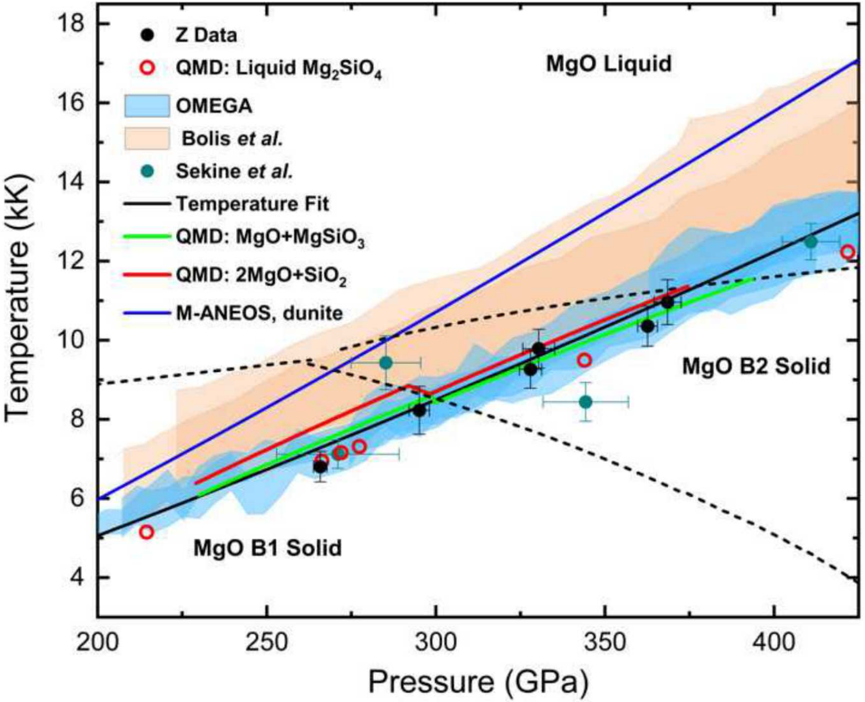


S. Root, J. Townsend, E. Davies, et. al. Geophysical Research Letters (2018).

The possibility of phase separation investigated with simulations – the effect is within experimental uncertainties

- Sekine et al. proposed recrystallization of MgO from the Mg_2SiO_4 liquid caused the observed changes in their data
- *Using DFT-MD, we can calculate the Hugoniot for two mixed-phase assemblages*:
  - **MgO (solid) + $MgSiO_3$ (liquid)**
  - **2MgO (solid) + SiO_2 (liquid)**
- *Mixed-phase assemblages are consistent with the uncertainties in our data*
- Lower temperature in the MgO+$MgSiO_3$ assemblage suggests IF phase separation occurs, it may be more likely

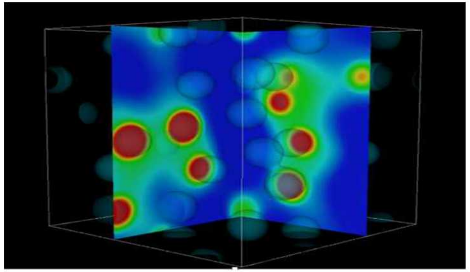
Sekine et. al. Sci. Adv., 2, e1600157 (2016).



S. Root, J. Townsend, E. Davies, et. al. Geophysical Research Letters (2018).

Atomistic simulation advances and Z experiments are being successfully used together to address transport, phase dynamics and multiphase EOS

- DFT/QMD predicted/explained Z&NIF results
  - EOS & Phase Boundaries
    - Deuterium, H_2O, LiD, CH_2, Xe, Kr, SiO_2, MgO
  - Electrical and Thermal Transport
    - Cu, Al, H_2O, Be, Li, Deuterium



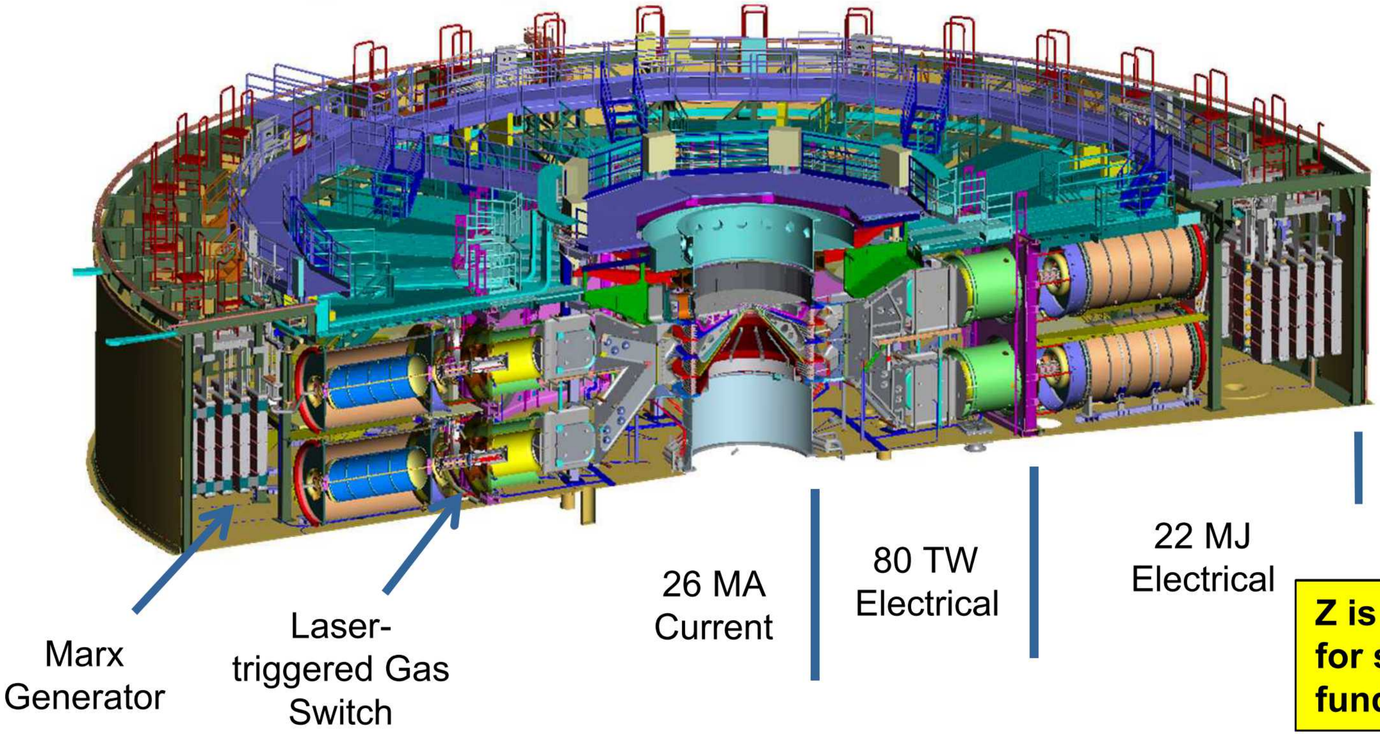
TDDFT simulation of moving ion exciting the electronic structure

Exciting future and ongoing work

- Improved capabilities for phase transitions, transport, and response of dense plasmas/ warm dense matter using time-dependent DFT (TDDFT)
- Quantum Monte Carlo: high-precision results for challenging materials – *new results on D2 Hugoniot and phase transitions in strongly correlated systems (Clay et al., submitted PRL, Townsend et al submitted PRB)*
  - *Computational Materials Science Center*
  - *ECP - Exascale Computing Project*
- Develop high fidelity potentials suitable for exploring transport or kinetics of phase transitions - *collaboration with LLNL/Jon Belof*

BACKUPS

Z is the world's largest pulsed power machine, and compresses energy in space ($>10^9$ x) and time ($>10^9$ x) to generate high energy density conditions



Z is an "engine of discovery" for stewardship and fundamental HED science

There are two new opportunities for Fellowships to perform Thesis work on Thor and other Sandia DMP facilities

Sandia National Laboratories

- **SUPER Center Fellowships [S**andia/**U**NM **P**ulsed power **E**xtreme condition **R**esearch (SUPER) Center]
  - Two joint UNM/SNL Ph.D. Fellowships
  - The successful applicant will have a primary advisor at UNM but will also be expected to collaborate with scientists at SNL.

- **Ph.D. Fellowship in High Pressure Earth and Planetary Sciences**
  - The Institute of Meteoritics (IOM) – Prof. Carl Agee
  - HEDP load dynamics research as applied to the extreme conditions in the Earth and planetary interiors.

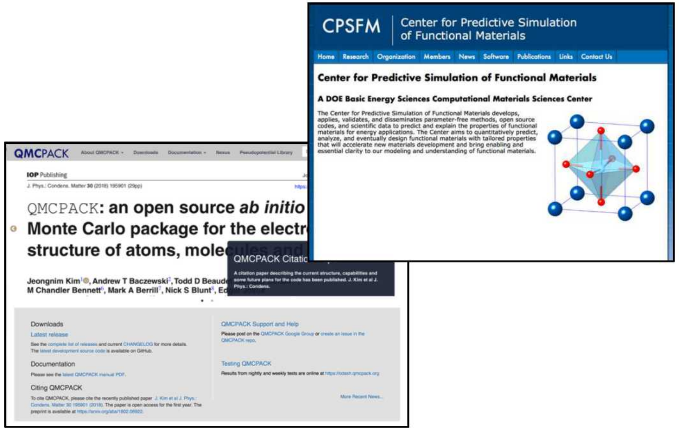
- **Ph.D. Fellowship in Pulsed Power for High Energy Density Science**
  - Applied Electromagnetics Group - Prof. Edl Schamiloglu
  - HEDP load dynamics (both experimental and modeling), pulsed power machine design and optimization (both experimental and simulation), diagnostics, and modeling machine/load coupling

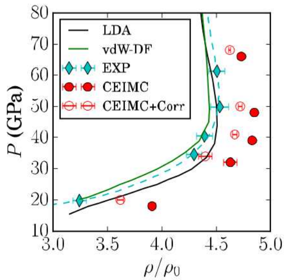
For more information, please contact Dawn Flicker (dgflick@sandia.gov)

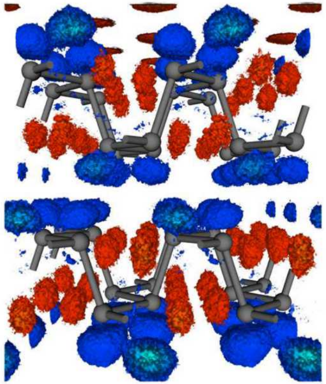
Sandia plays a key theoretical and modeling role in the Center for the Predictive Simulation of Functional Materials

Sandia National Laboratories

- Computational Materials Science Center
  - Funded by the DOE Office of Basic Energy Sciences
- Joint Collaboration between four National Laboratories (ORNL, SNL, ANL and LLNL) and two universities (UC Berkeley and NCSU)
- Produces open source software (qmcpack.org)
- Accuracy of standard electronic structure techniques like DFT is not known a priori
  - Calculations may even fail qualitatively for strongly correlated materials where charge and spin degrees of freedom are both important (eg magnets, oxides, actinides)
- The center develops Quantum Monte Carlo techniques that aim for systematically improvable approximations
  - Understanding of errors leads to confidence in predictions



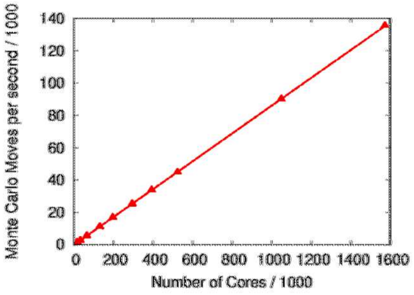


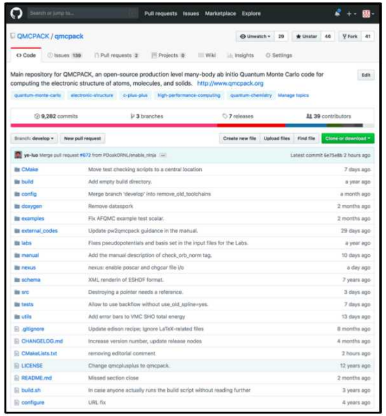


Sandia is a key partner for QMCPACK: Predictive and Improvable Quantum Mechanics – Based Simulations

- QMC is ideally suited to today's largest supercomputers
  - Nearly perfect parallel scaling on 1.5 million processing elements on LLNL's biggest machine
- Next generation of supercomputers will involve more complex architectures: GPUs, FPGAs etc.
- Working as part of the Exascale Computing Project (exascaleproject.org) to develop performance portable code for these more complex machines
- Goal is to improve parallelism while writing code that can run across a wide variety of machines
- Multi – Year effort leveraging Sandia's strengths in computing (e.g., kokkos) in collaboration with ORNL, LLNL and ANL

Scaling of throughput on Sequoia





ECP
EXASCALE COMPUTING PROJECT

If you are interested in pulsed power related research as a career, one option is to apply for the LRGF

Sandia National Laboratories

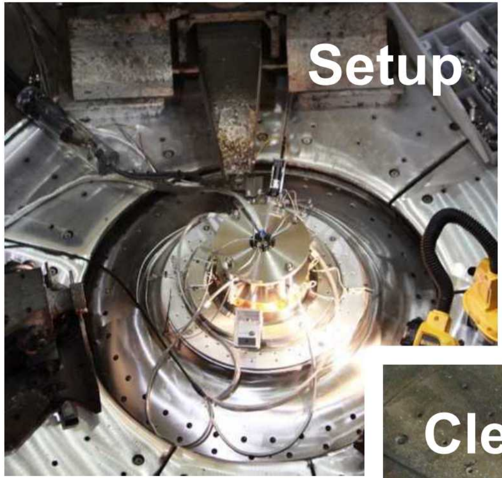
LRGF **DOE NNSA Laboratory Residency Graduate Fellowship**

- Managed by the Krell Institute for NSNA (https://www.krellinst.org/lrgf/)
- Applications for academic year 2019-2020 will open in late November
- Must be at least a second year graduate student
- Proposals to perform thesis work on national laboratory facilities
- Two 12 week residencies at the laboratory required; more is desired
- Fellowships for study in:
  - Pulsed Power Science and Engineering
  - Radiation MHD
  - Atomic physics and Spectroscopy
  - Dynamic Materials Properties/Shock Physics
  - Accelerator Design

Sandia National Laboratories is a world leader in dynamic materials science and applications

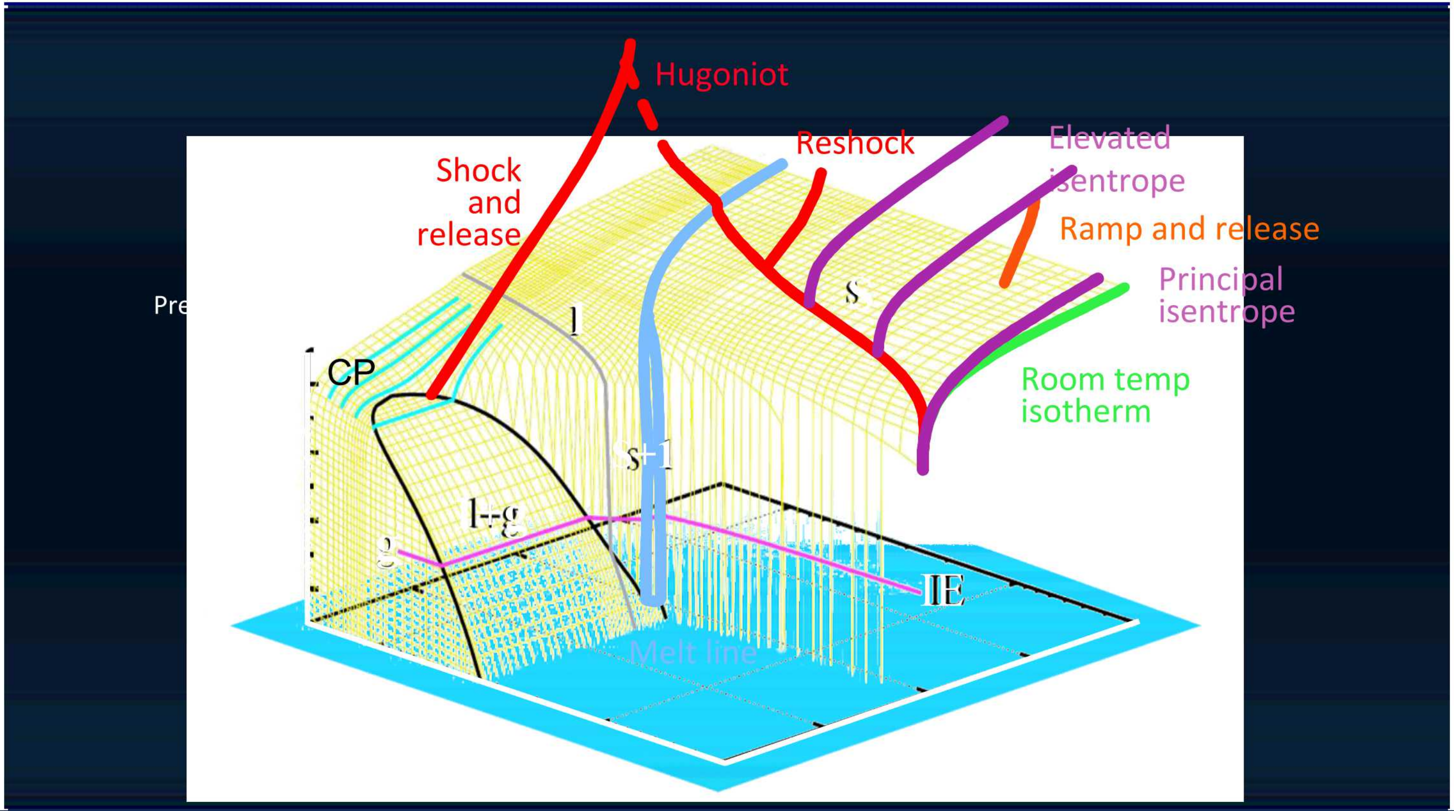
- We apply a unique suite of driver technologies covering a wide range of strain rates, time scales, and pressures in large sample sizes (mm to cm)
  - Pulsed power
  - Gas guns
- We are working at the forefront of dynamic material properties science
  - Novel, high-precision platforms for materials research (e.g., shock+ramp)
  - First-principles density functional theory
  - New Quantum Monte Carlo computational tools for exascale computers
- We are developing new experimental capabilities that will greatly advance our program in the near future
  - New diagnostics for pulsed power platforms
  - New pulsed power drivers with unprecedented pulse shaping control

A challenge for Z experiments is that they release the energy of a few sticks of dynamite



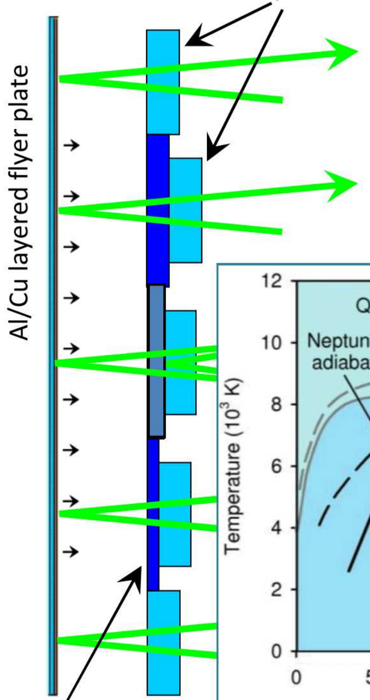
- Harsh debris, shock, and radiation environment make fielding experiments unique and challenging

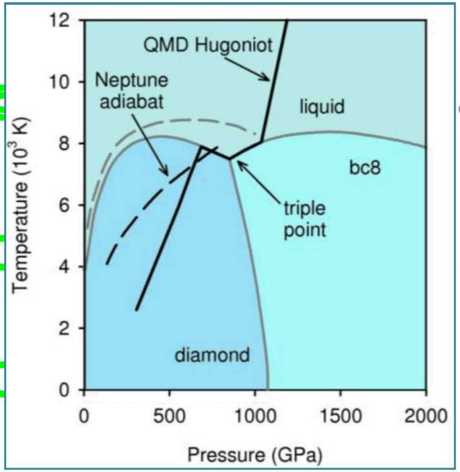
Collectively, we have a wide range of tools on Z allowing dynamic compression experiments to probe large regions of a material's equation-of-state surface



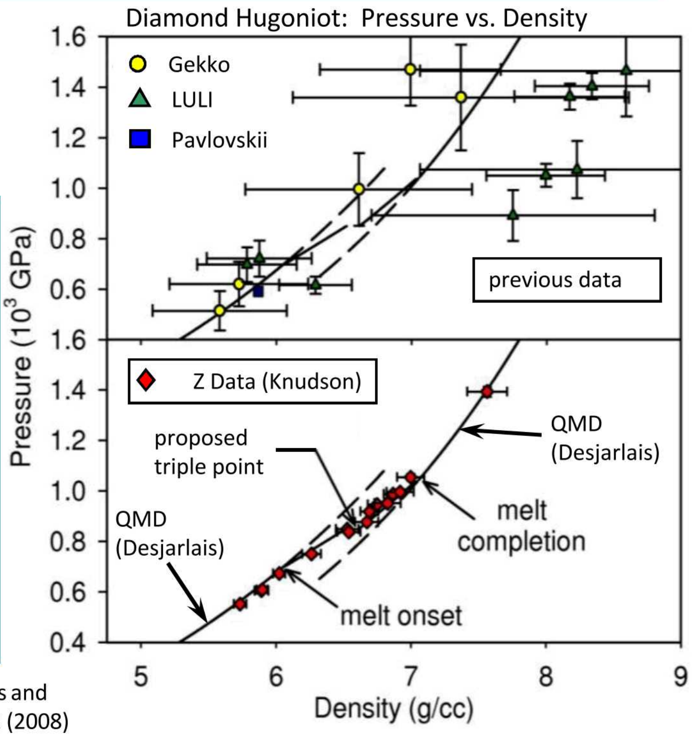
Z flyers provided first experimental evidence of diamond-liquid-BC8 triple point in carbon, important for determining at what shock pressure diamond ICF capsule ablators on NIF would melt

Order-of-magnitude improvement in precision over laser-driven shock techniques (larger spatial/temporal scales)





M.D. Knudson, M. P. Desjarlais and D. H. Dolan, *Science* **322**, 1822 (2008)



Z flyer experiments and theory provided new understanding of high pressure Xenon

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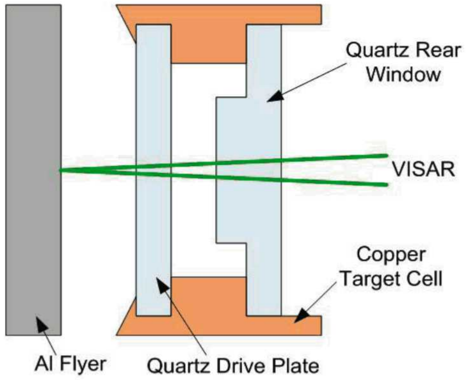
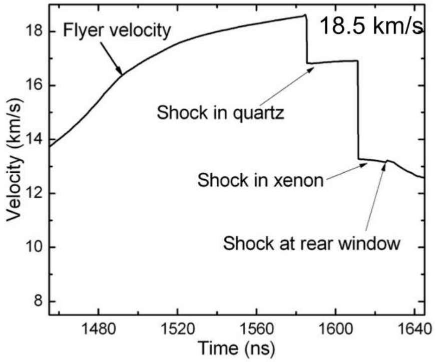
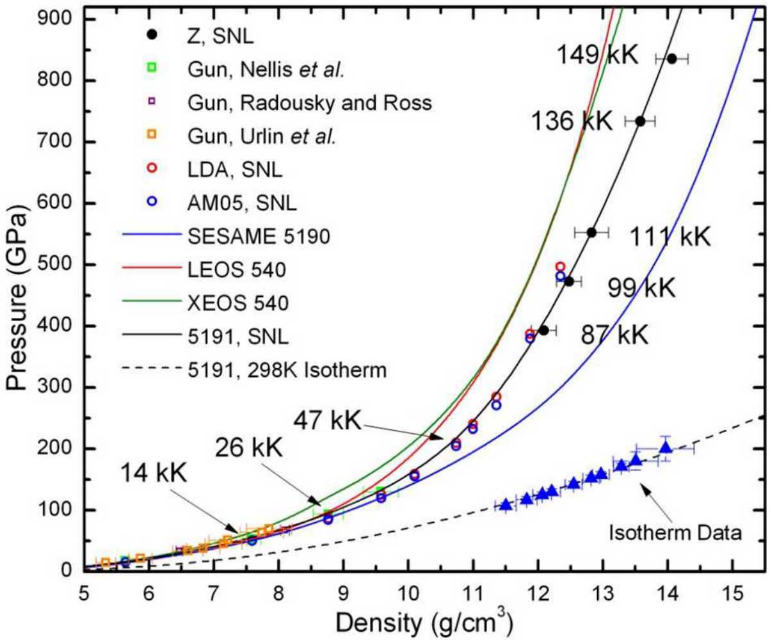




Shock velocities in transparent materials measured with sub-percent accuracy



Theory & data almost always diverge in previously unreachable regimes

S. Root *et al.*, Phys. Rev. Lett. 105, 085501 (2010).

The possibility of phase separation investigated with simulations – the effect is within experimental uncertainties

Recent observations of crystallization of pure SiO_2 in pure SiO_2 liquid at LCLS by Gleason et al. give an upper bound on crystallization rate for MgO in silicate liquid

Rudimentary crystallization model suggests that phase separation may not be observable over timescale of experiment.