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UNIVERSITY OF SOUTH FLORIDA

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Phase Transitions in SiC in the Interiors of Carbon-Rich Exoplanets

ZFS Project: 2021-2023

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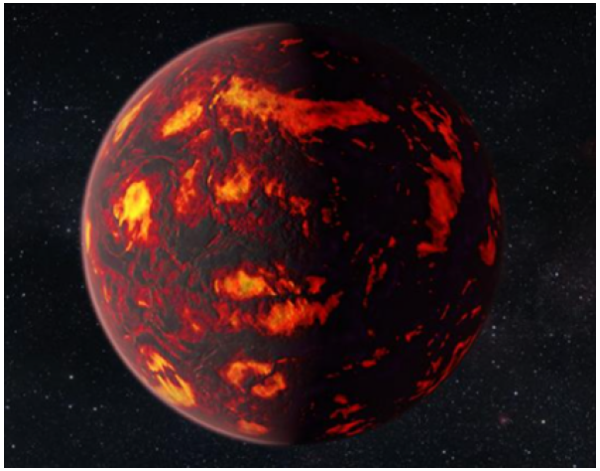
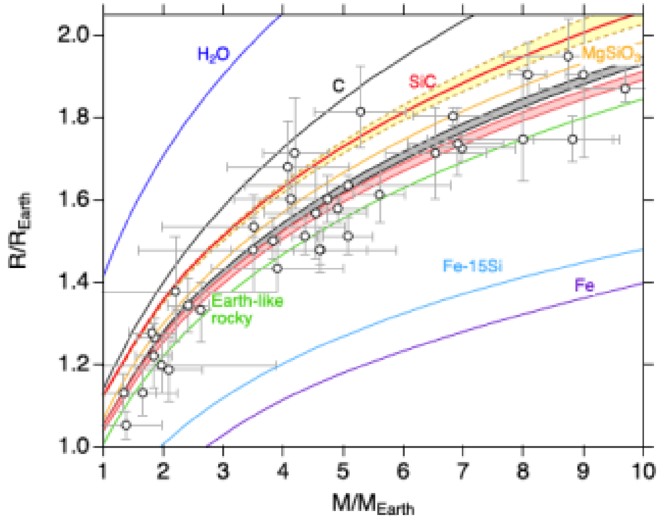
Silicon Carbide – major constituent of carbon-rich exoplanets

Mass-Radius relationship: Kim *et al Nat Comm (2022)*





- Host stars with high > 0.8 C/O ratio: potential existence of C-rich exoplanets

- SiC – major material of C-rich exoplanets: high-PT phase diagram and EOS - development of interior models of C-rich exoplanets

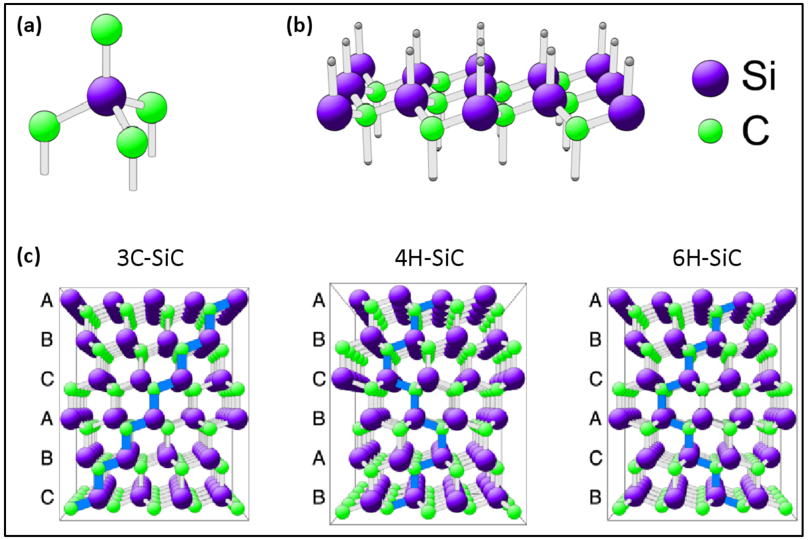
- EOS and phase diagram – poorly constrained: conflicting experimental results

Outline

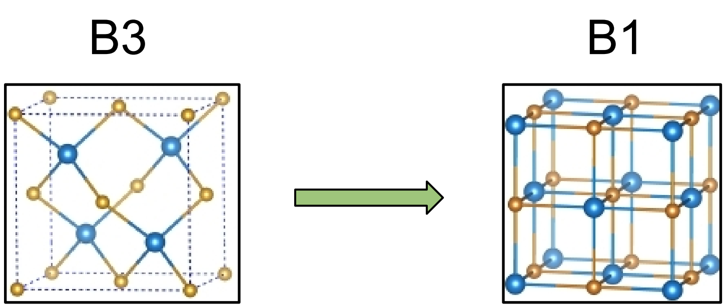
- Introduction: SiC major constituent of C-rich exoplanets, phase transitions and EOS
- Outstanding scientific questions: onset of B3(4H) to B1 solid-solid phase transition and SiC incongruent melting
- Predictions from quantum molecular dynamics - need experimental validation
- ZFS experiments: goals and results from first Z shot and STAR gas gun experiments
- Next steps: future Z shots and quantum-accurate MD simulations of B3 to B1 phase transition in SiC
- Conclusions

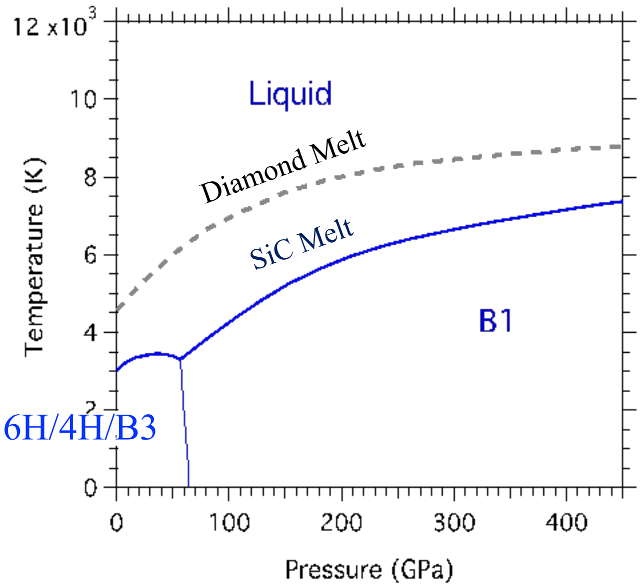
SiC ambient structures and phase diagram

- At 0 GPa: plethora of crystal structures with “diamond-like” sp^3 bonding network of C and Si atoms



- High pressure phase transition (70-110 GPa) from one of ambient to B1 rock-salt phase



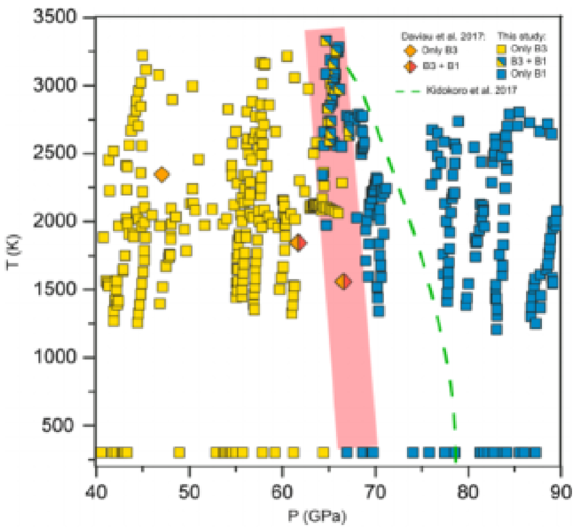


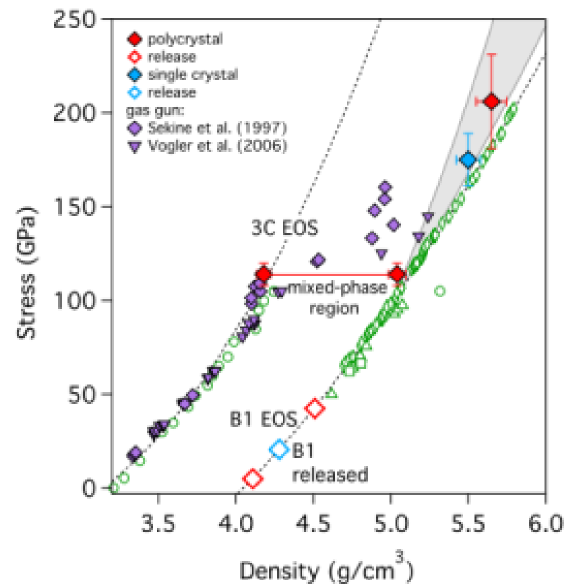
Solid-solid B3(4H) -> B1 phase transition: contradiction between static and dynamic compression experiments

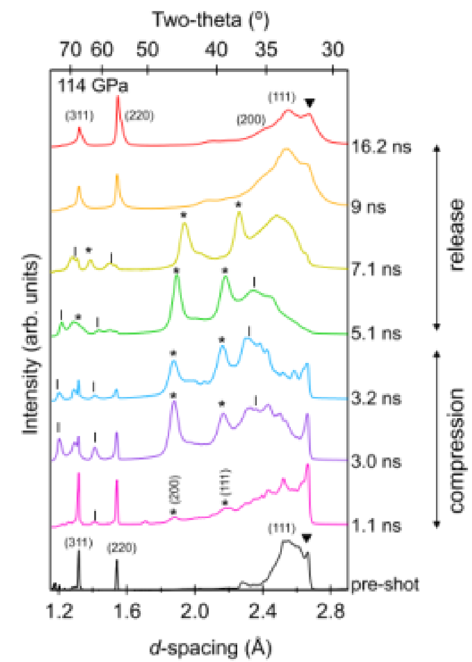
Laser heated DAC: B3 to B1 transition at **70 GPa (**Miozzi et al. (2018))

Gas-gun (Vogler et al (2006), Sekine et al (1996)) **& laser driven shock (**Tracy et al (2019)**):** B3(4H) to B1 transition at **~110 GPa**

Tracy et. al. PRB (2019) Shock experiments at MEC/LCLS: B3-B1 transition via XRD at **114 GPa**





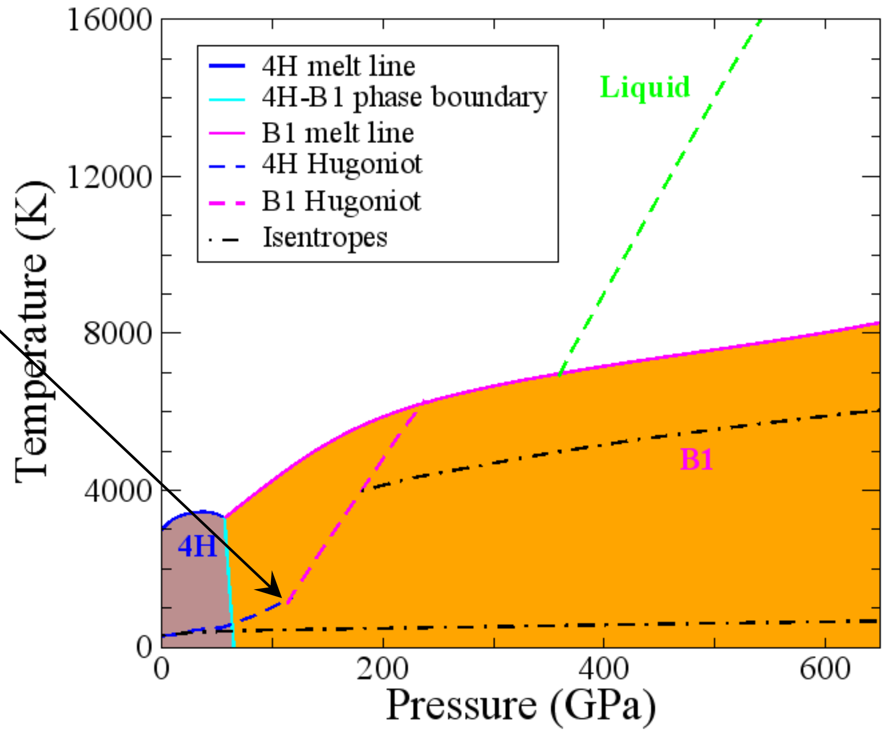


- QMD simulations – B3(4H)/B1 phase boundary at 65 GPa – similar to static DAC results
- Role of kinetics is counterintuitive: DAC and gas gun transition pressures should be similar, but surprisingly, both gas gun (μs) and laser-driven (ns) compressions – similar pressure

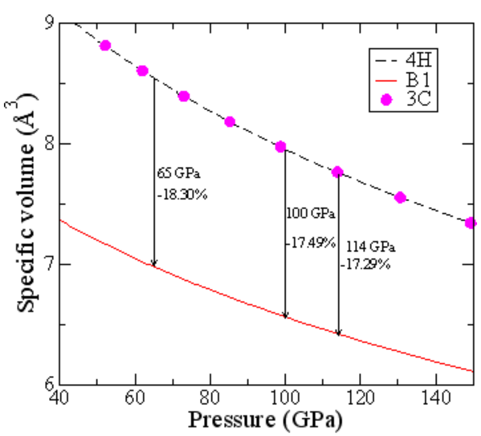
Delay of B3(4H) to B1 transition in shocked SiC: "Hugoniot frustration"

QMD simulations of PI's group:

- 4H to B1 phase transition at much higher pressure (110 GPa) - well above thermodynamic 4H/B1 phase boundary at 70 GPa

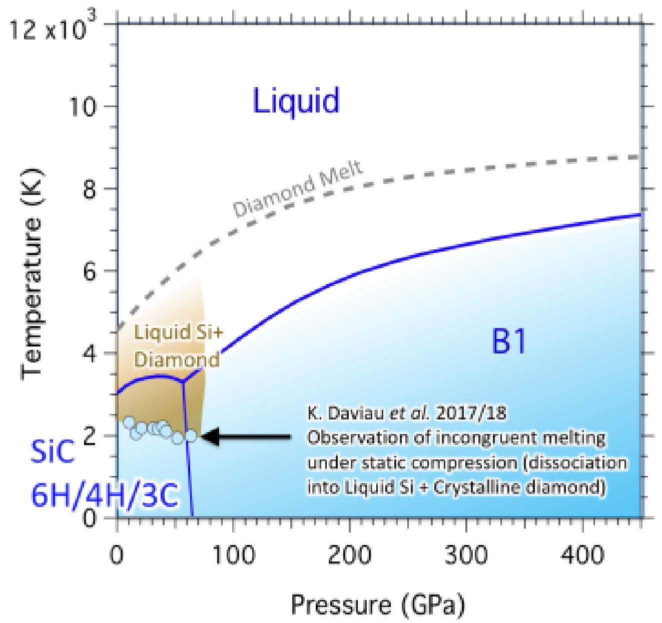


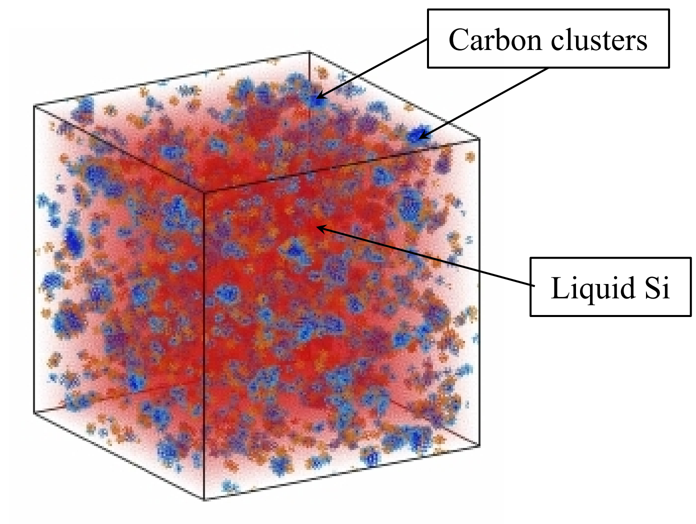
- Delay of phase transition up to 110 GPa: "Hugoniot frustration": impossibility to satisfy Rankine-Hugoniot relations:
  $\rho_0 U_s = \rho_1 (U_s - u_p)$
  $P_1 - P_0 = \rho_0 U_s u_p$
  $E - E_0 = \frac{1}{2}(P_1 + P_0)\left(\frac{1}{\rho_0} - \frac{1}{\rho_1}\right)$
  due to large (~20%) volume collapse:



Hypothesis: observation of B3 to B1 transition at thermodynamic phase boundary (70 GPa) in ramp compressions

Solid-liquid phase transformations in SiC: potential incongruent melting

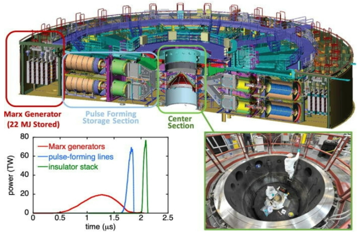




- SiC melting line is poorly constrained
- SiC- binary system – might melt into solid C and liquid Si or any other Si_xC_y compounds
- Earlier DAC –studies – contradictory – incongruent melting of B3 phase from 0 to 10 GPa (Togaya et al, 1998), congruent melting (Sokolov et al, 2012)
- Thermal decomposition of B3 phase onto elemental Si and C up to 70 GPa and 4,000 K (Daviau et al, 2017) interpreted as incongruent melting
- No studies above 70 GPa: does high-P phase B1 – melt incongruently?

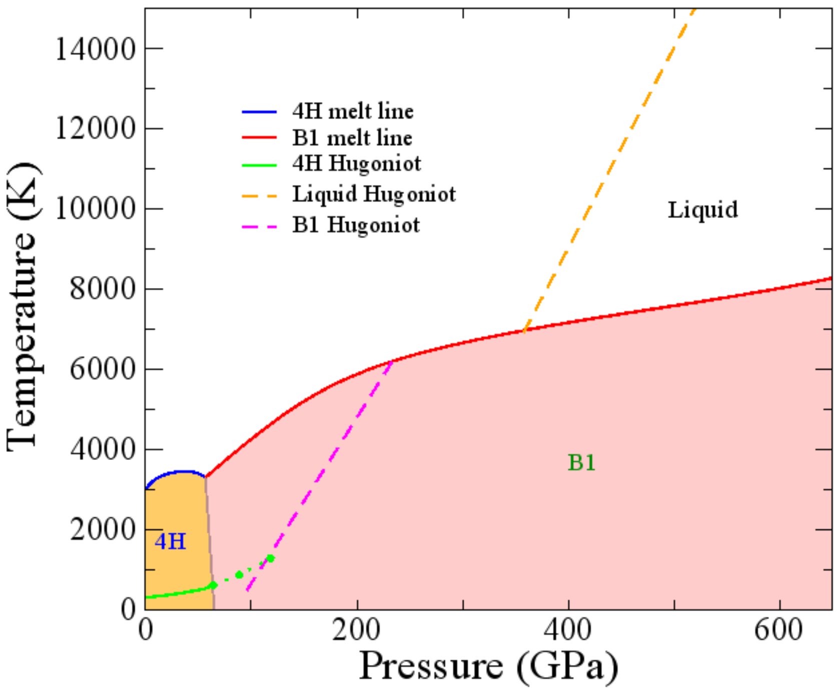
ZFS project: Scientific Objectives

- Unique coupling of experiments on Z, Thor and STAR and predictive simulations to uncover fundamental mechanisms of solid-solid and solid-liquid phase transitions in SiC under pressure and temperature conditions representative of the interiors of carbon-rich exoplanets
  - Validate prediction of "Hugoniot frustration" by ramp compressing SiC to observe B3 to B1 transition at phase boundary at ~70 Gpa using X-ray diffraction on Z
  - Prove/disprove occurrence of incongruent melting of SiC: mechanisms and kinetics of solid-solid and solid-liquid phase transitions
  - To obtain experimentally validated multi-phase EOS of SiC
  - Study kinetics of solid-solid and solid-liquid phase transitions
- Employ combination of dynamic x-ray diffraction (DXRD), spectroscopy and velocimetry
- Support and guide experiments by QMD and quantum accurate SNAP MD simulations

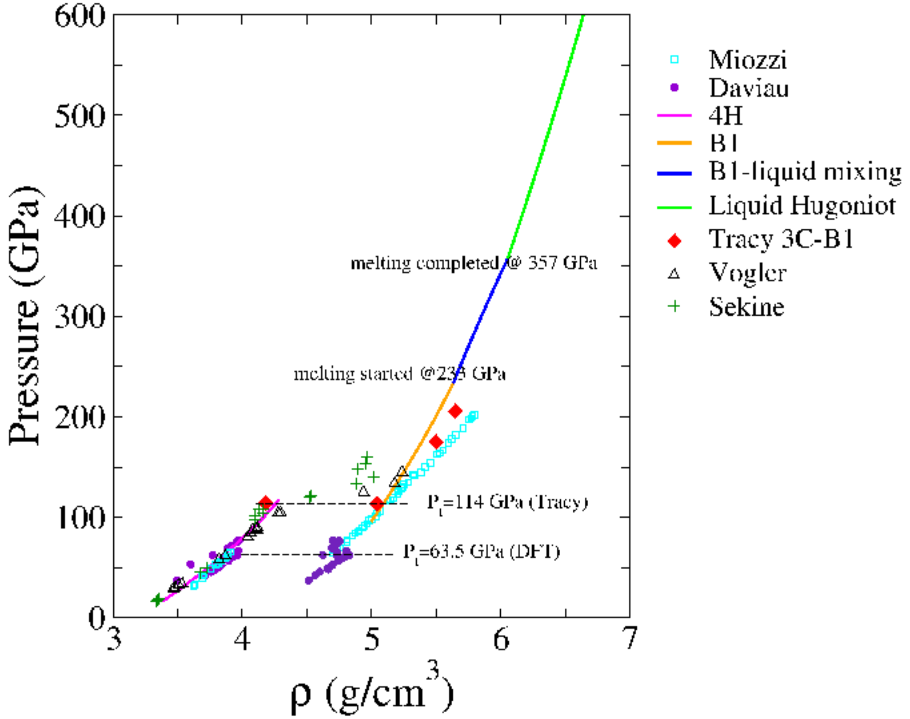


QMD simulations of SiC EOS and phase diagram to aid in design of experiments

SiC Phase diagram



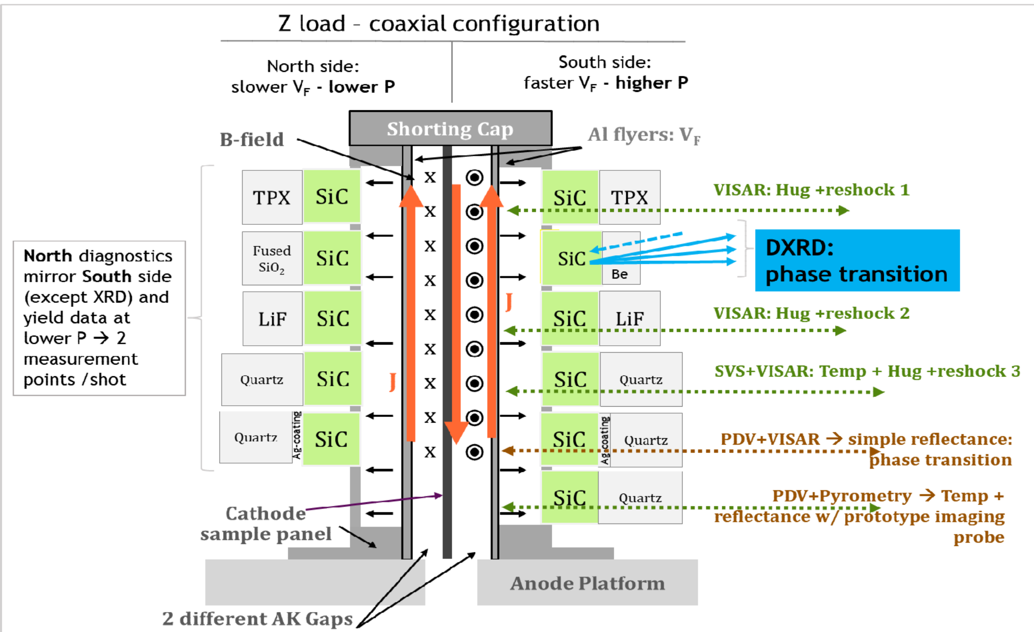
P-ρ Hugoniot



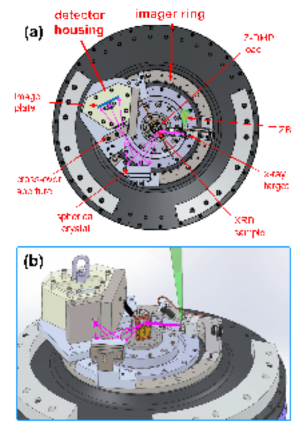
- 4H/B1 phase boundary at 0 K at 65 GPa
- Melting curve QMD calculations using 2-phase method: $T_m = 2{,}900$ K; dT_m/dP – maximum at (45 GPa, 3,450 K) , triple 4H/B1/liquid point at (57 GPa, 3,450 K), large positive dT_m/dP for B1 phase, reaching 9,100 K at 1 TPa
- EOS: good agreement with static experiments, disagreement with laser experiments of Tracy et al (2019), resolved in recent experiments by Smith et al (2022)
- Shock melting starts at 233 GPa and completes at 357 GPa

ZFS SiC project: Experiments on Z

- Combine DXRD, velocimetry and spectroscopy on Z to study B3(4H) -> B1 phase transition and incongruent melting

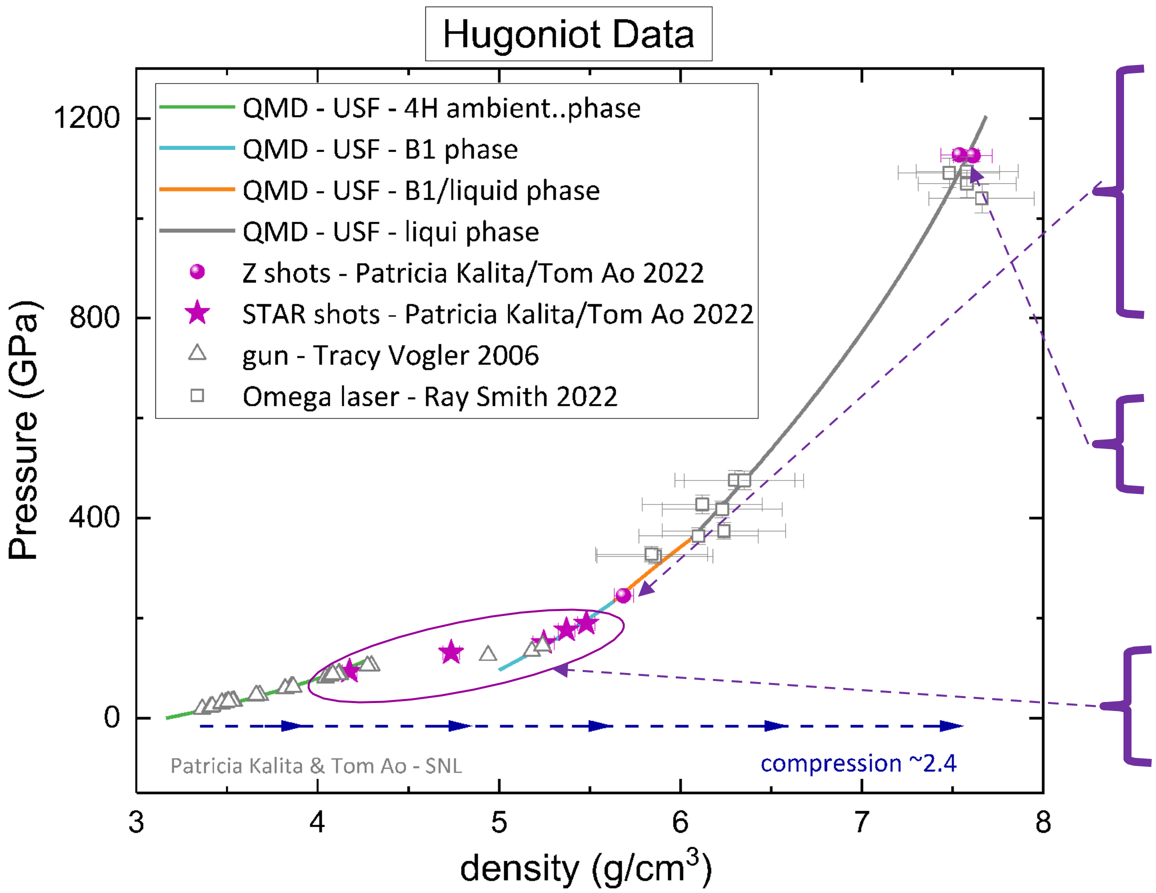


DXRD on Z



- Dynamic materials properties configuration: Al impactor, flyer plate velocities 10-15 km/s, shock states for few tens of ns
- Multiple diagnostics (VISAR, PDV, SVS, XRD), one sample for RXRD measurements in each Z shot
- DXRD using Spherical Crystal Diffraction Imager (SCDI) diagnostic
- Shock, ramp, shock-ramp using Z pulse shaping to reach several off-Hugoniot states
- Thor and STAR 2-stage gun to probe strength effects at low pressure and get EOS between 100 and 250 GPa

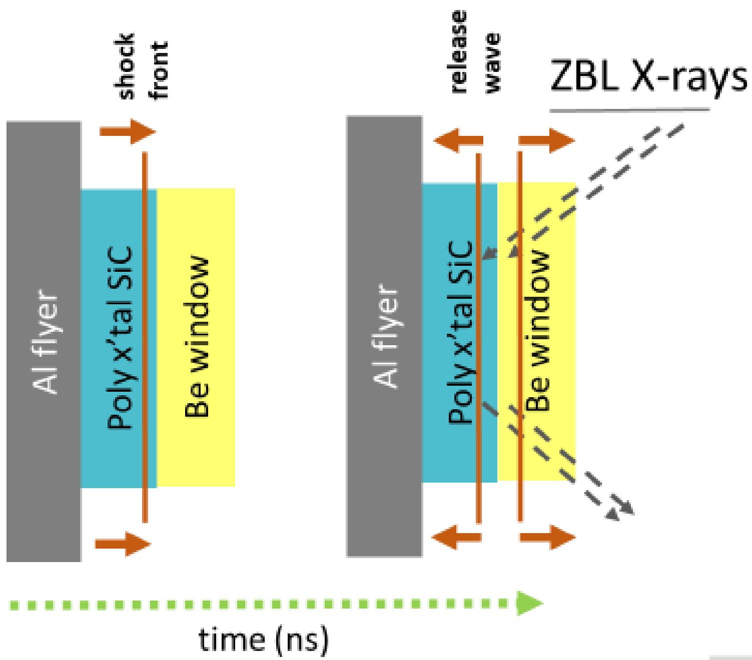
First Z shot for Z project Z3708 in May 2022
STAR shots in Feb 2022



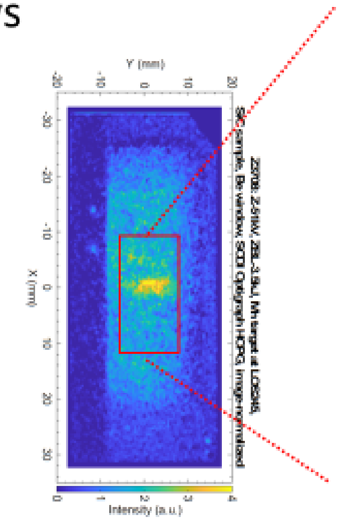
Accomplishments

- **Z shot** of the SiC series: Z3708, May'22
- **Hugoniot** measurement at 245 GPa at the solid liquid boundary
- **dynamic XRD & phase transition**
- **Hugoniot** data obtained on other Z shots at ~1.1 TPa
- **SVS temperature** measurements : data being analyzed
- Series of **5 STAR** Hugoniot shots in Feb. 2022

Shock-driven phase transition in SiC measured in situ with Dynamic XRD on the SNL Z machine – Z3708

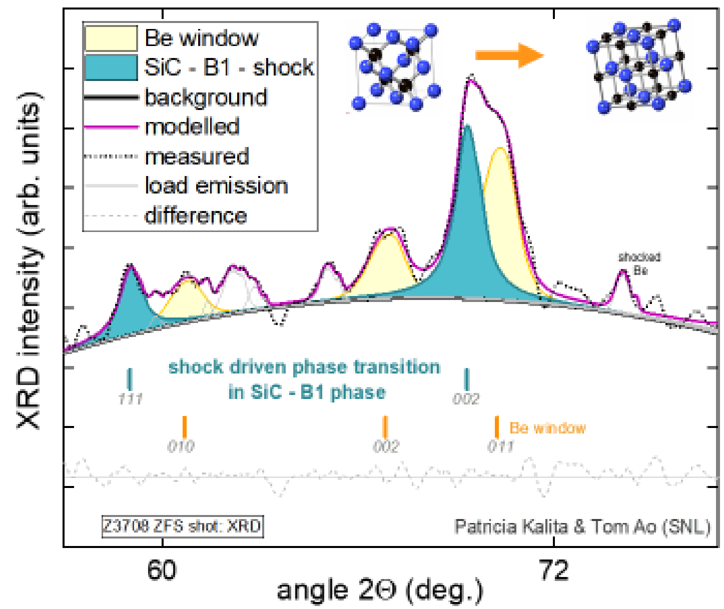


XRD geometry



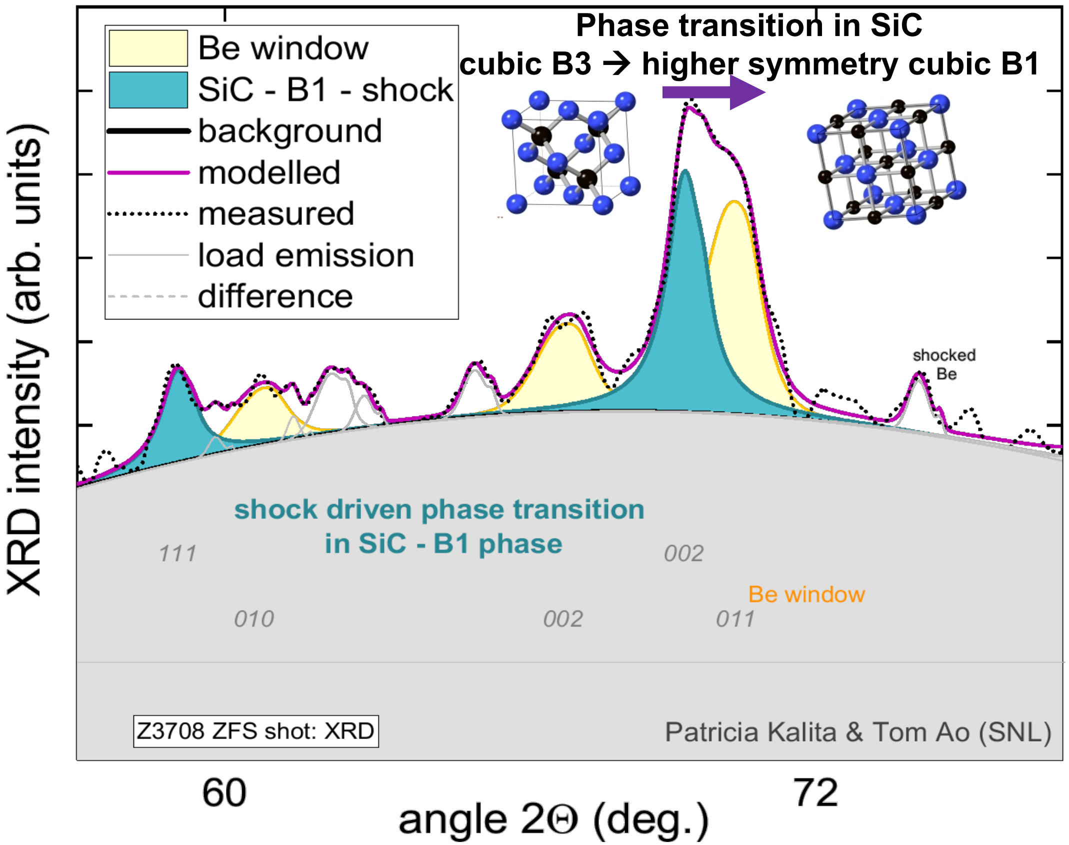
XRD image with SCDI

Phase transition in SiC
cubic B3 or zincblende-type → higher symmetry cubic B1 or rocksalt-type phase



XRD analysis

Shock-driven phase transition in SiC measured in situ with Dynamic XRD on the SNL Z machine

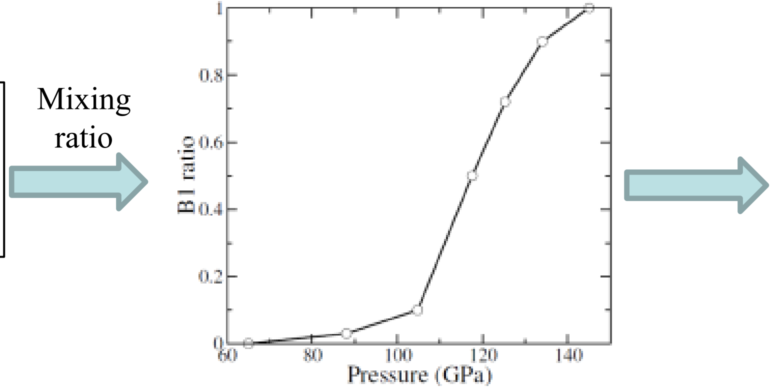


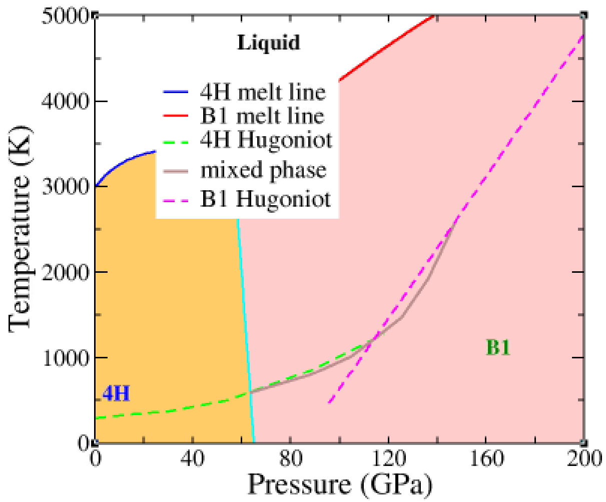
XRD analysis

- We successfully measured a shock-driven phase transition in SiC with the SCDI XRD diagnostic on SNL's Z machine
- Rietveld analysis of in-situ XRD consistent with a B3 → B1 solid-solid phase transition
- XRD pattern shows an important contribution from the beryllium backing window, as well as unwanted emission from the experimental load
- Measurement made off-Hugoniot on shockwave release from the Be window for better control of timing
- Simple compression of B3 phase is ruled-out, based on the relative intensity of diffractions lines in the B1 vs. the B3 phase;
- More quantitative analysis is ongoing
- Future experiments may use the newly implemented DISCO XRD diagnostic on Z

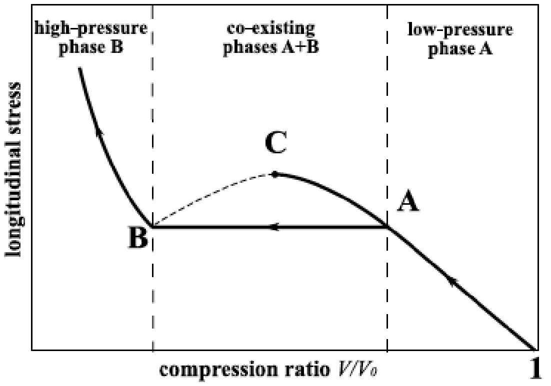
QMD simulations: mixed phase B3/B1 region: "Hugoniot frustration"

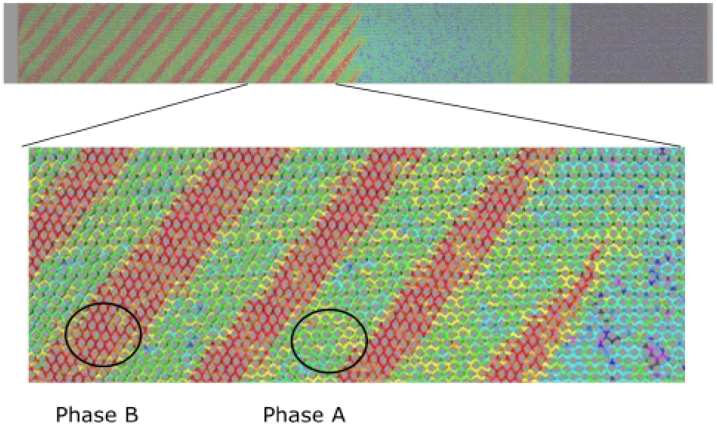
Pressure-density from STAR experiments





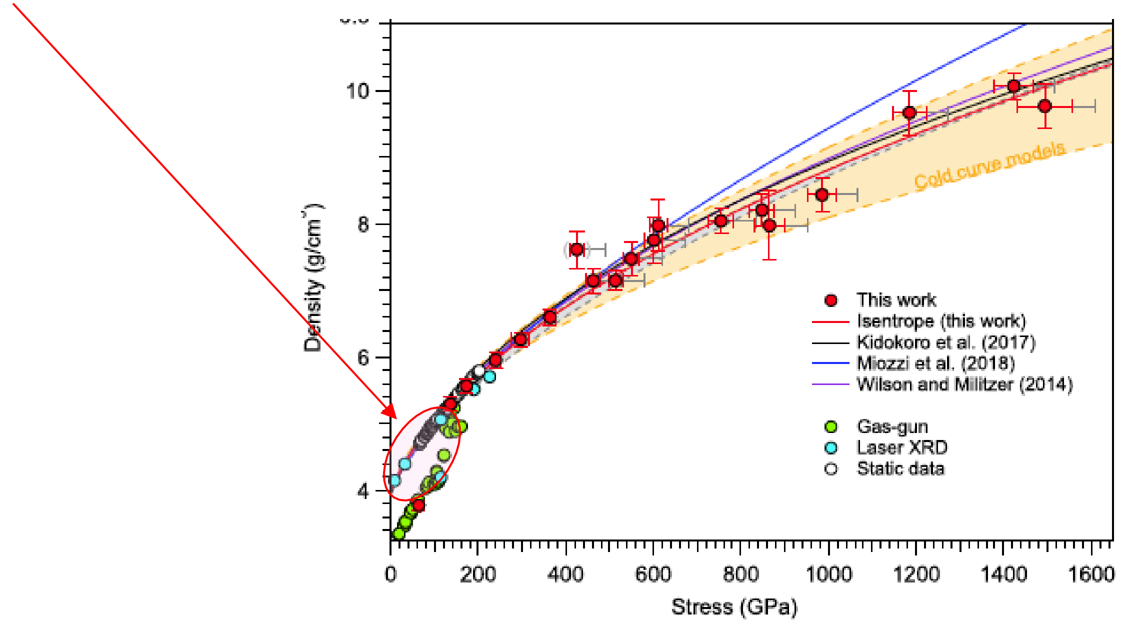
- Kinematic Hugoniot frustration: very little B1 formed based simple mixing model based on experimental data from STAR experiments
- Explicit MD shock simulations of phase transitions will provide insight into fundamental mechanisms of phase transitions





Next step: Experiments and Simulations

- Focus on B3 to B1 phase transition: ramp compression on Z with dynamic XRD
  - Recent ramp compression experiments (Kim et al 2022) missed interesting mix-phase region



- Develop quantum-accurate SNAP machine learning interatomic potential and perform explicit MD shock simulations of B3/B1 phase transition

- Develop experimentally validated model of B3/B1 phase transition from combined experiment/simulation studies

Acknowledgements

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