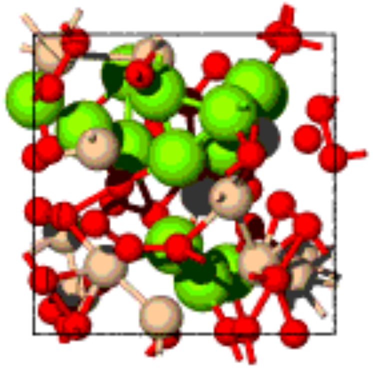
Quantum simulations at extreme conditions: warm dense matter and planetary interiors



Presentation to:
LLNL HEDS Seminar Series

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12/01/2022

ACKNOWLEDGEMENTS

DOE-NNSA
(DE-NA0003842)

CMEC
CENTER FOR MATTER UNDER EXTREME CONDITIONS

NERSC
National Energy Research Scientific Computing Center

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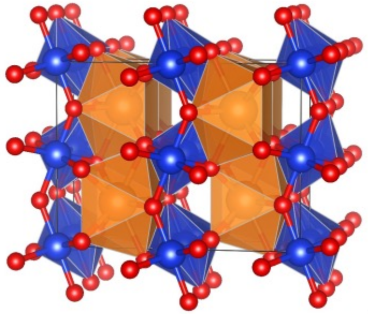
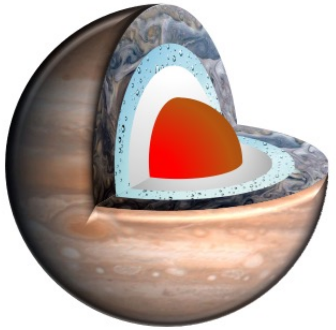
OVERVIEW AND MOTIVATION



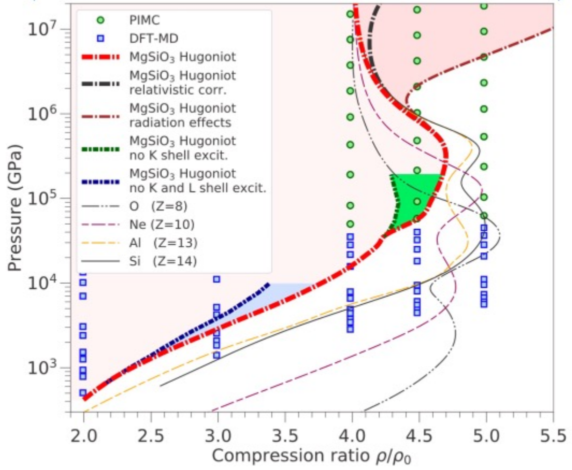
High pressure

- Solid-solid phase transitions
- Equations of state
- Melting



Warm dense matter

- Temperature ionization
- Pressure ionization
- Shock compression



Planetary Interiors

- Super-Earth interior models
- Core/mantle crystallization
- Element Partitioning
- Solubility / Miscibility

OUTLINE

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   - Rock/Ice mixtures in water planets
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   - Be & MgO: melting and anharmonicities
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   - Warm dense silicates: Mg, MgO & $MgSiO_3$
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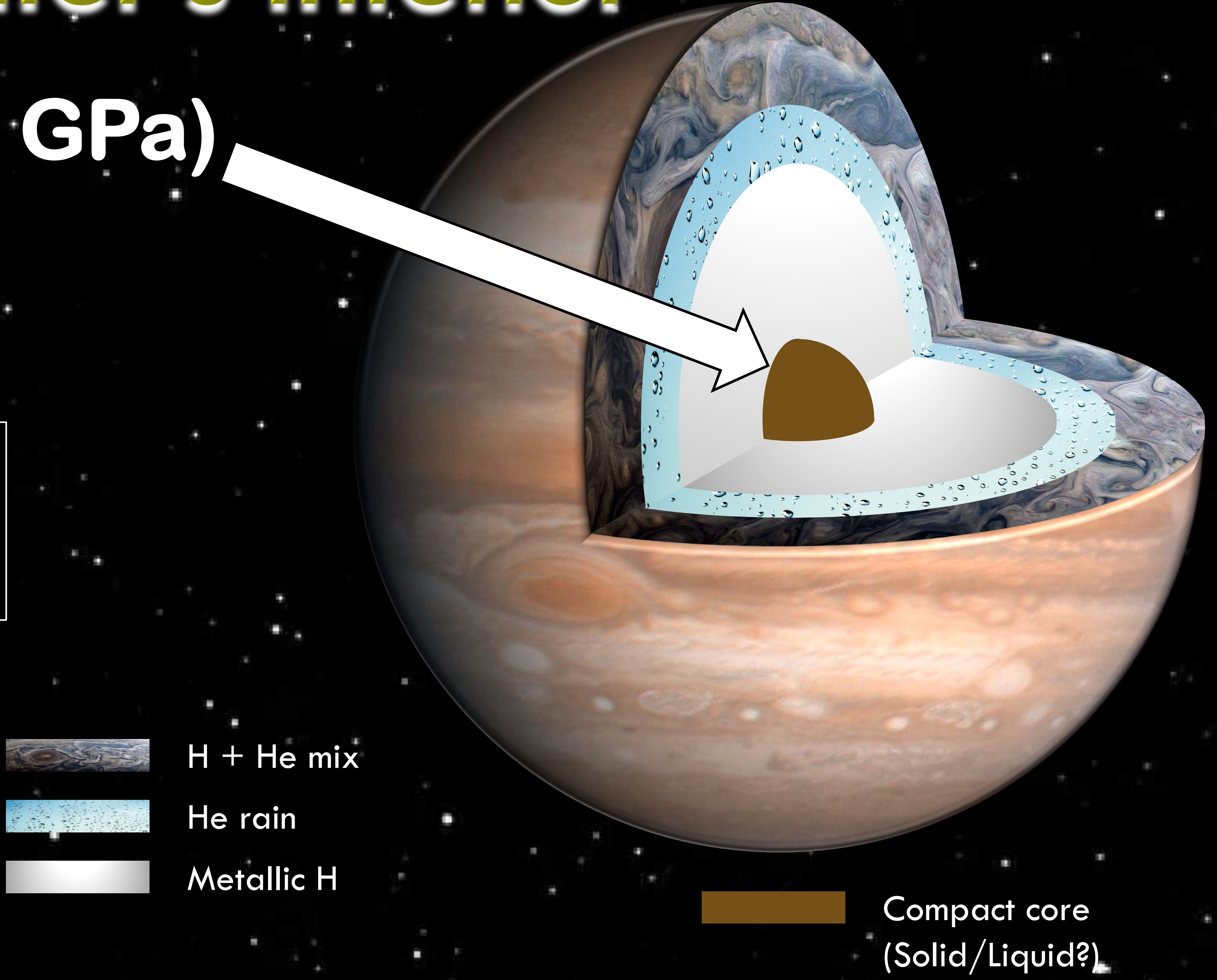
Jupiter's Interior

P~ 40 Mbar (4000 GPa)
T~ 16000 K

Can metallic H dissolve the rocky core?

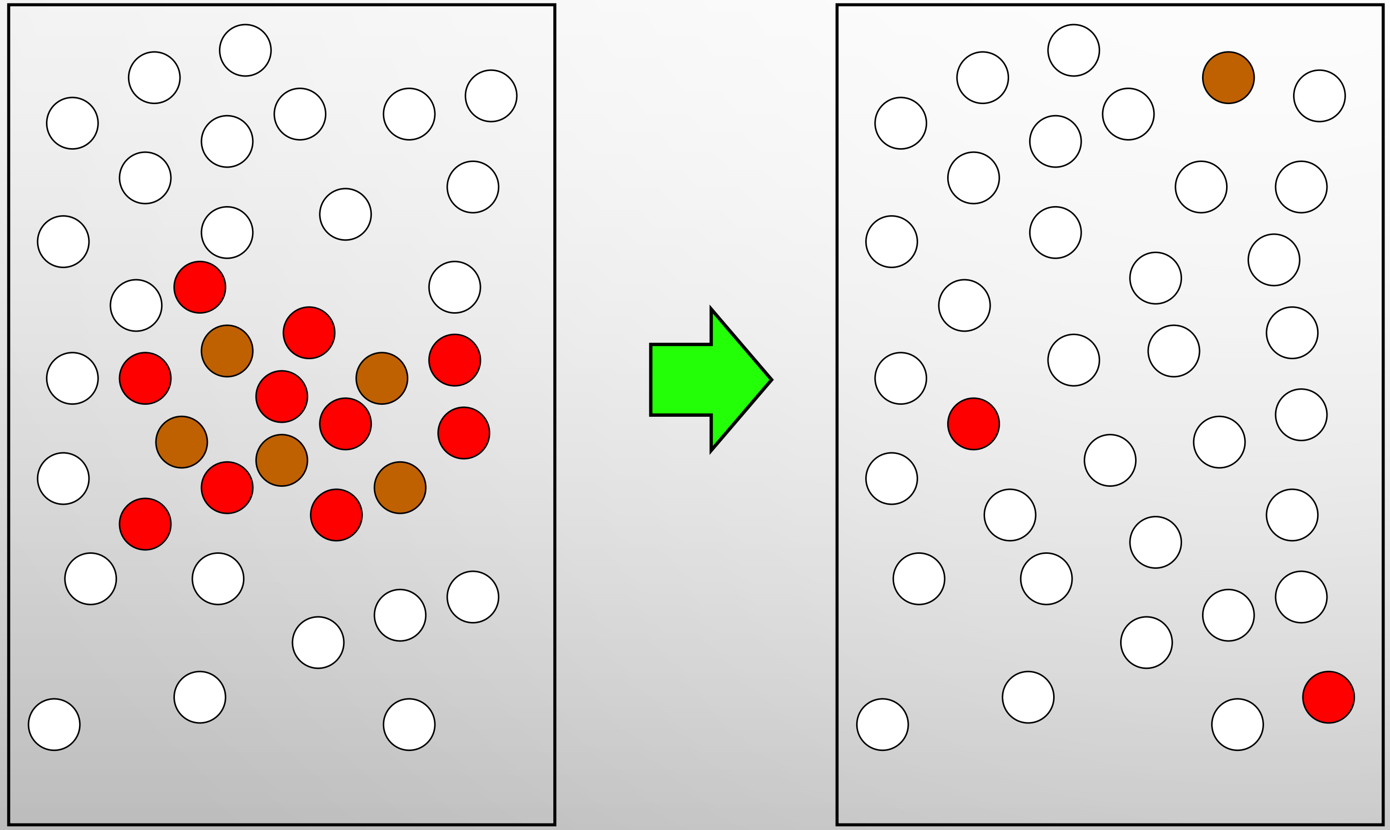


Solubility of SiO_2

$$H_n + SiO_2 \rightarrow H_nSiO_2$$

Free energy of solvation

$$\Delta G \equiv G(H_nSiO_2) - [G(H_n) + G(SiO_2)]$$



Free energy of the Dissolved System

$$\Delta G < 0 \quad \Rightarrow \quad \text{Dissolves}$$

How to calculate G(P,T)?

Gonzalez & Militzer, APJ (2014)

Thermodynamic Integration

$U_A \rightarrow U_B$

U_A: reference system

F_A: known

$$U(\lambda) = U_A + \lambda(U_B - U_A)$$

$$(U(0) = U_A;\ \ U(1) = U_B)$$

U_B: DFT system

F_B: unknown

$$\Delta F = \int_A^B dF = \int_0^1 d\lambda \frac{\partial F}{\partial \lambda} = -\int_0^1 d\lambda \frac{k_B\, T}{Z} \frac{\partial Z}{\partial \lambda}$$

$$= \int_0^1 d\lambda \frac{k_B\, T}{Z} \sum_s e^{-\frac{U(\lambda)}{k_B T}} \frac{1}{k_B\, T} \frac{\partial U(\lambda)}{\partial \lambda}$$

$$= \int_0^1 d\lambda \left\langle \frac{\partial U(\lambda)}{\partial \lambda} \right\rangle_\lambda = \int_0^1 d\lambda \, \langle U_B - U_A \rangle_\lambda$$

$$F_B = F_A + \Delta F$$

$$G_B = F_B + PV$$

$$F = -kT \ln Z$$

$$Z = \sum_s e^{-\frac{U(\lambda)}{k_B T}}$$

PLANETARY INTERIORS — A diluted core in Jupiter

TWO-STEPS TDI

LIQUIDS

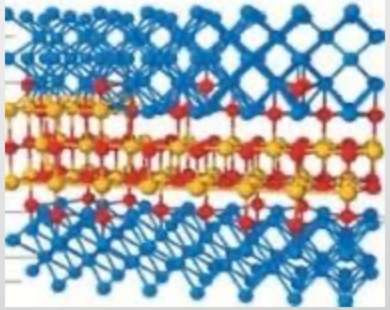
IDEAL GAS → PAIR POTENTIAL* → DFT

$U = 0$ $U = U_{PP}$ $U = U_{KS}$

- GGA-PBE
- Ecut = 900 eV
- 144 atoms
- Γ - point
- 14 val. elect.

SOLIDS

EINSTEIN CRYSTAL → ($\lambda = 0 \rightarrow \lambda = 1$) → PAIR POTENTIAL + EINSTEIN → ($\lambda = 0 \rightarrow \lambda = 1$) → DFT



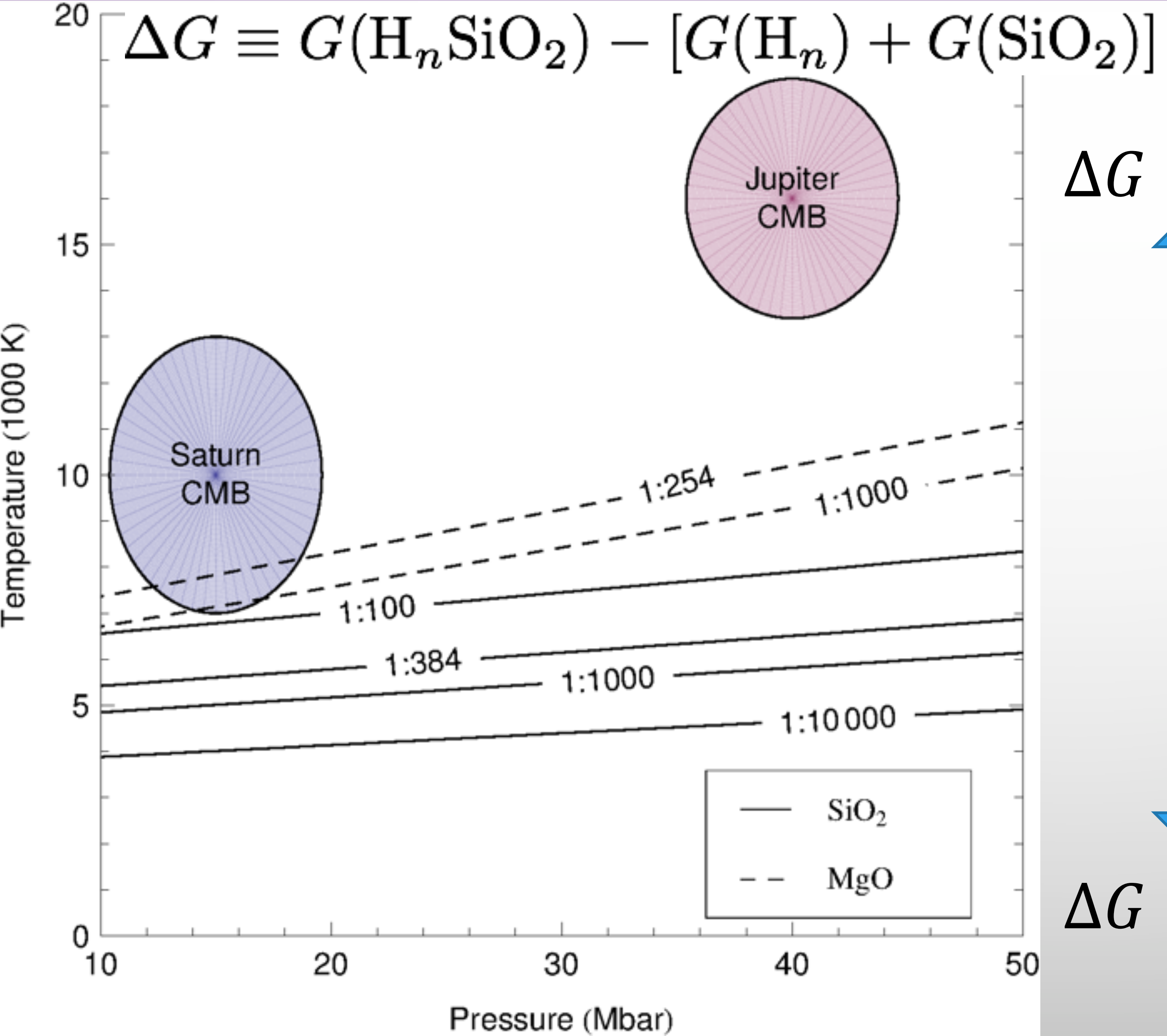
$\frac{1}{2} m\, \omega\, r^2$ (one body) (one body + two body)

$U = U_{Einstein}$ $U = U_{Einstein} + U_{PP}$ $U = U_{KS}$

Classical MD VASP

S. Izvekov & Parrinelo, JCP (2004)

A diluted core in Jupiter



$\Delta G < 0$

$\Delta G > 0$

$\Delta G < 0$ at CMB

- SiO_2 gets dissolved
- At SiO_2:H < 1:100

- SiO_2 more soluble than MgO
- Fe, H_2O also soluble in H

Wahl+, APJ (2013)

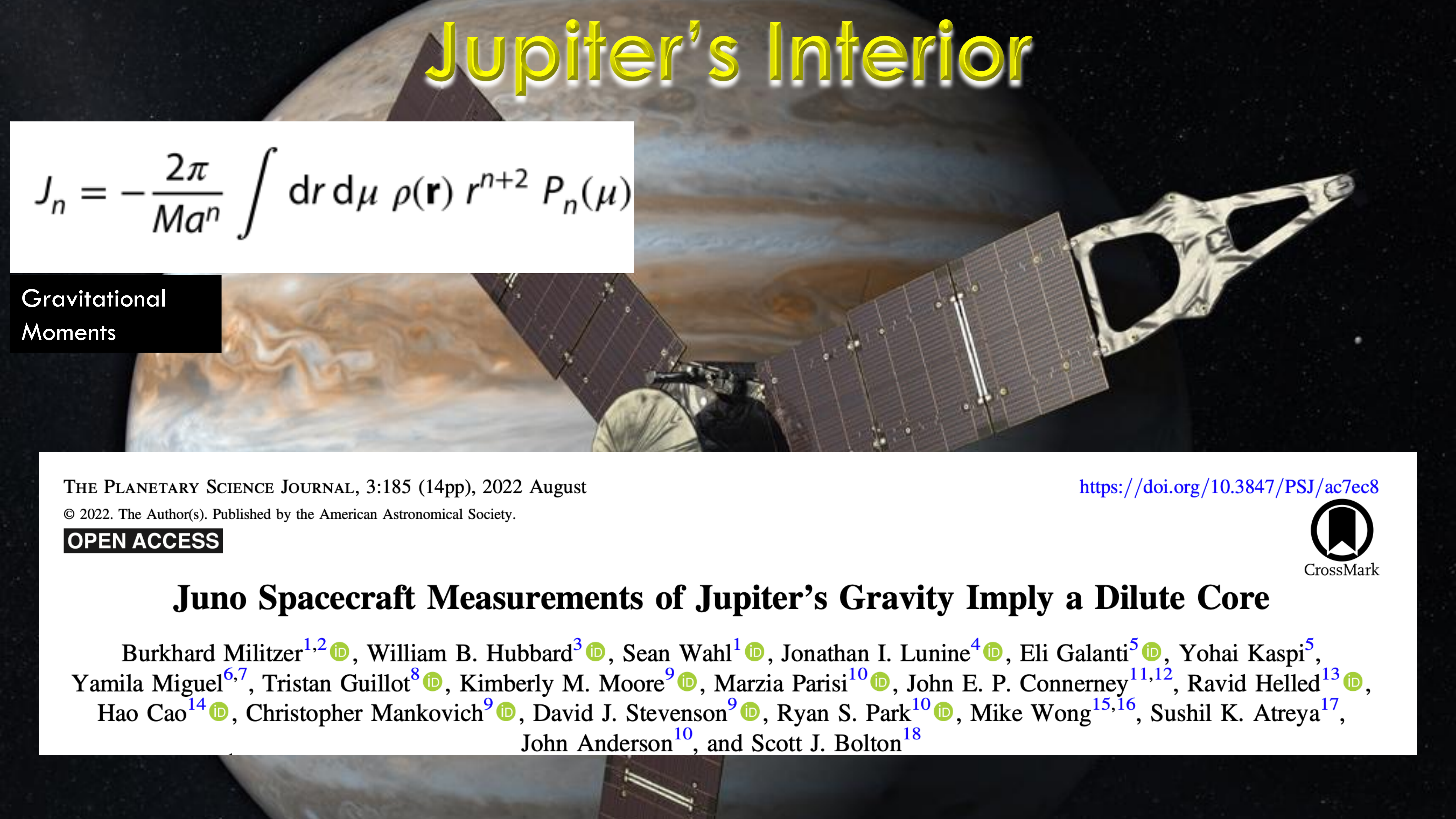
Wilson+ & Militzer APJ (2012)

Gonzalez & Militzer, APJ (2014)

Jupiter's Interior

$$J_n = -\frac{2\pi}{Ma^n} \int \mathrm{d}r\, \mathrm{d}\mu\; \rho(\mathbf{r})\; r^{n+2}\; P_n(\mu)$$

Gravitational Moments

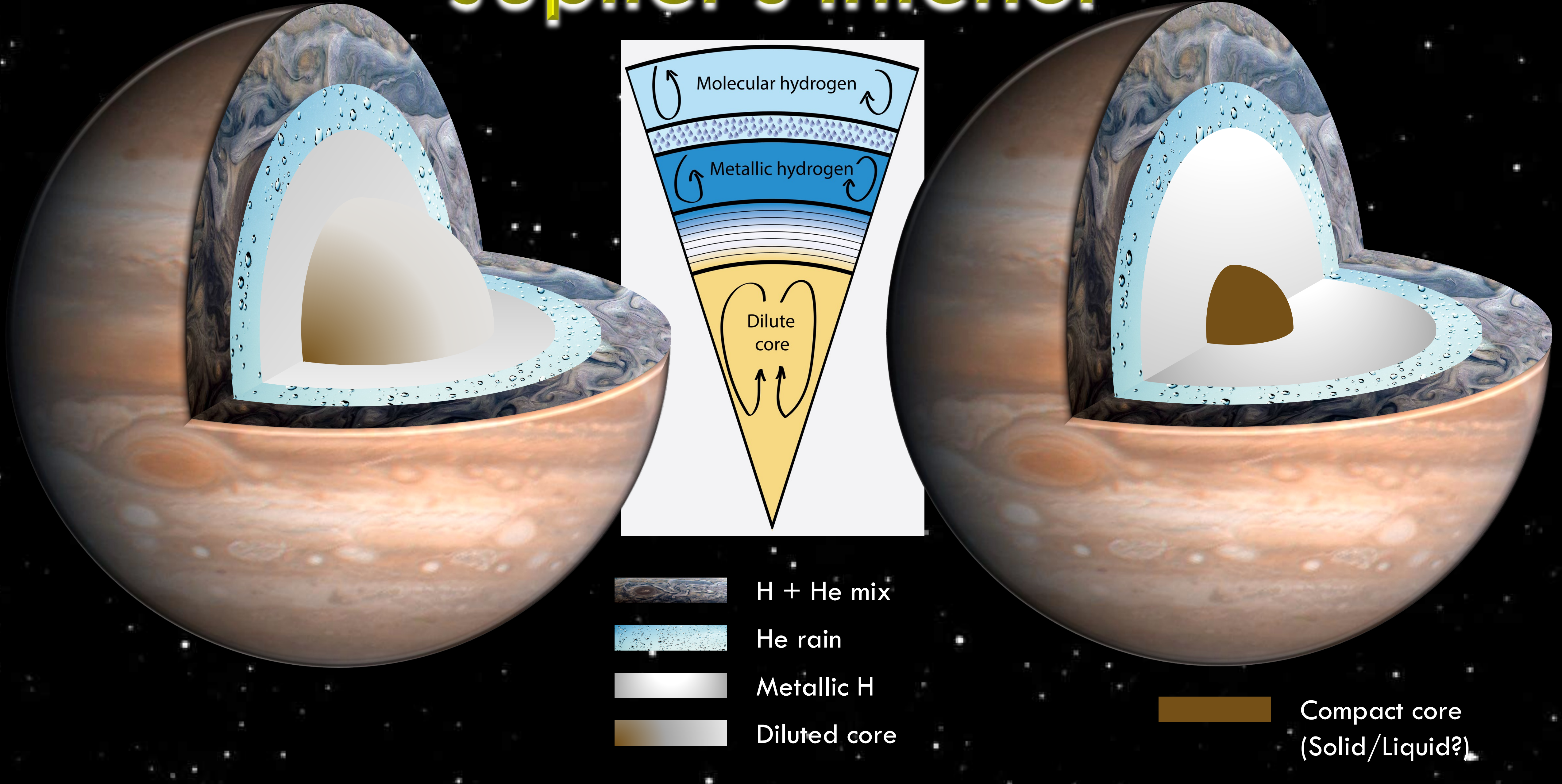


THE PLANETARY SCIENCE JOURNAL, 3:185 (14pp), 2022 August https://doi.org/10.3847/PSJ/ac7ec8

Juno Spacecraft Measurements of Jupiter's Gravity Imply a Dilute Core

Burkhard Militzer[1,2], William B. Hubbard[3], Sean Wahl[1], Jonathan I. Lunine[4], Eli Galanti[5], Yohai Kaspi[5], Yamila Miguel[6,7], Tristan Guillot[8], Kimberly M. Moore[9], Marzia Parisi[10], John E. P. Connerney[11,12], Ravid Helled[13], Hao Cao[14], Christopher Mankovich[9], David J. Stevenson[9], Ryan S. Park[10], Mike Wong[15,16], Sushil K. Atreya[17], John Anderson[10], and Scott J. Bolton[18]

Jupiter's Interior



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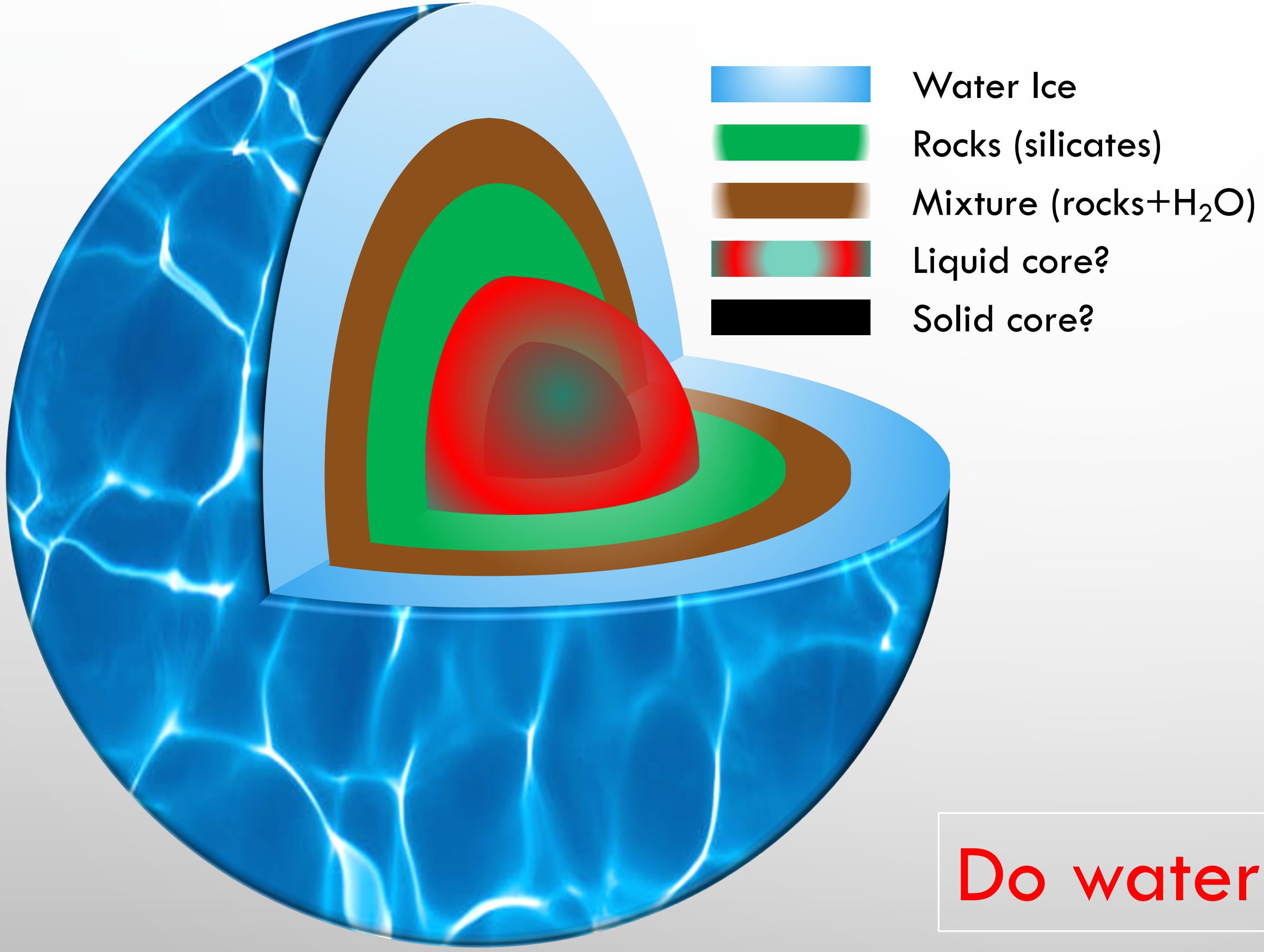
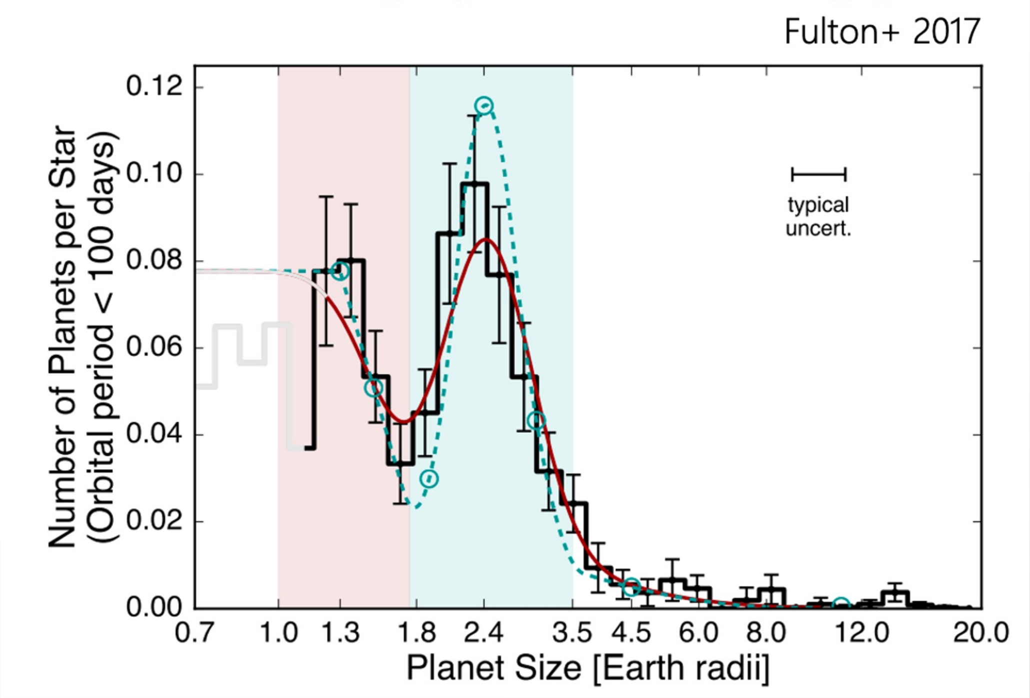
PLANETARY INTERIORS

WATER WORLDS



Radius Gap (Fulton Gap)



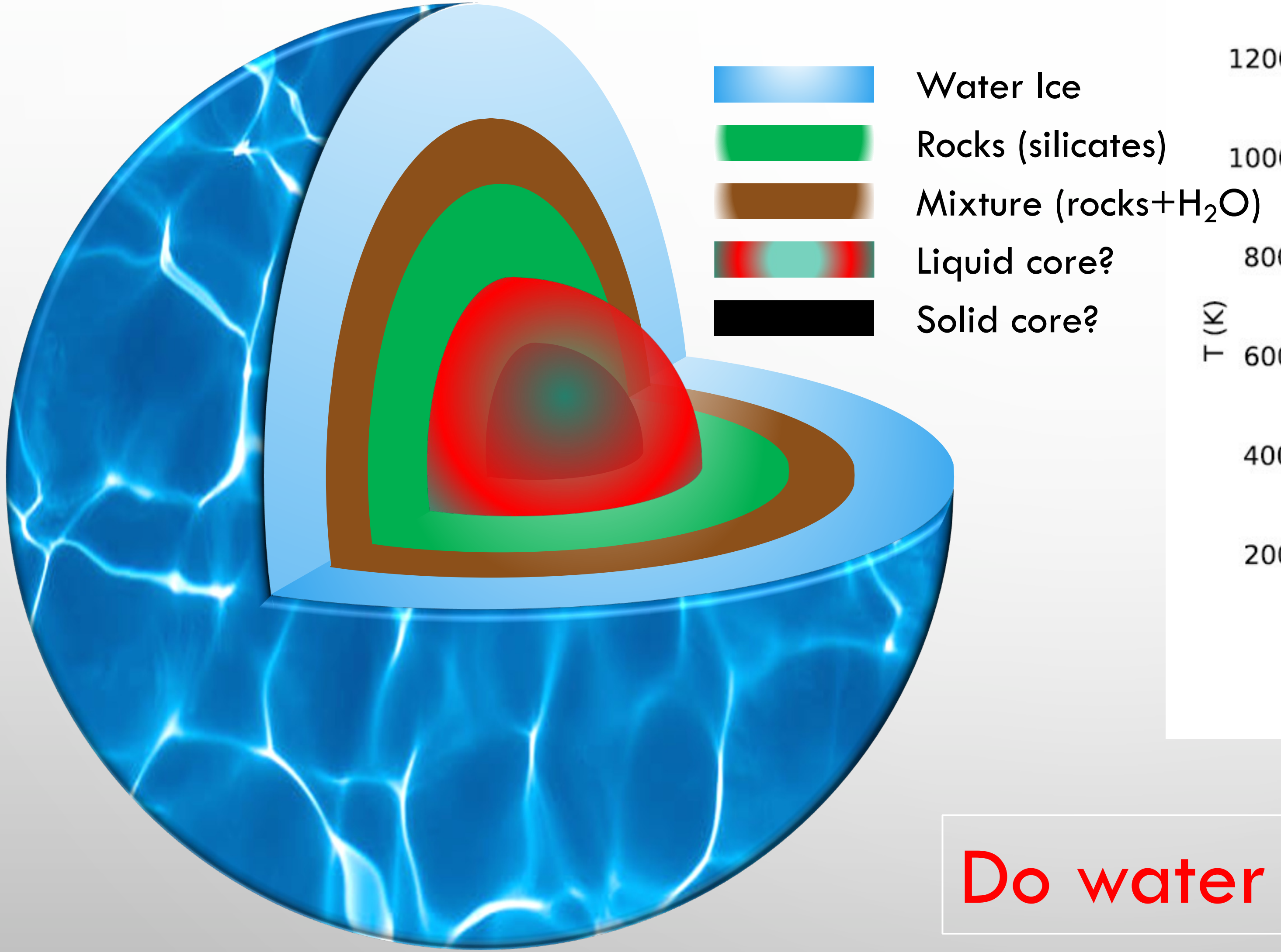
Super-Earths → Sub-Neptunes

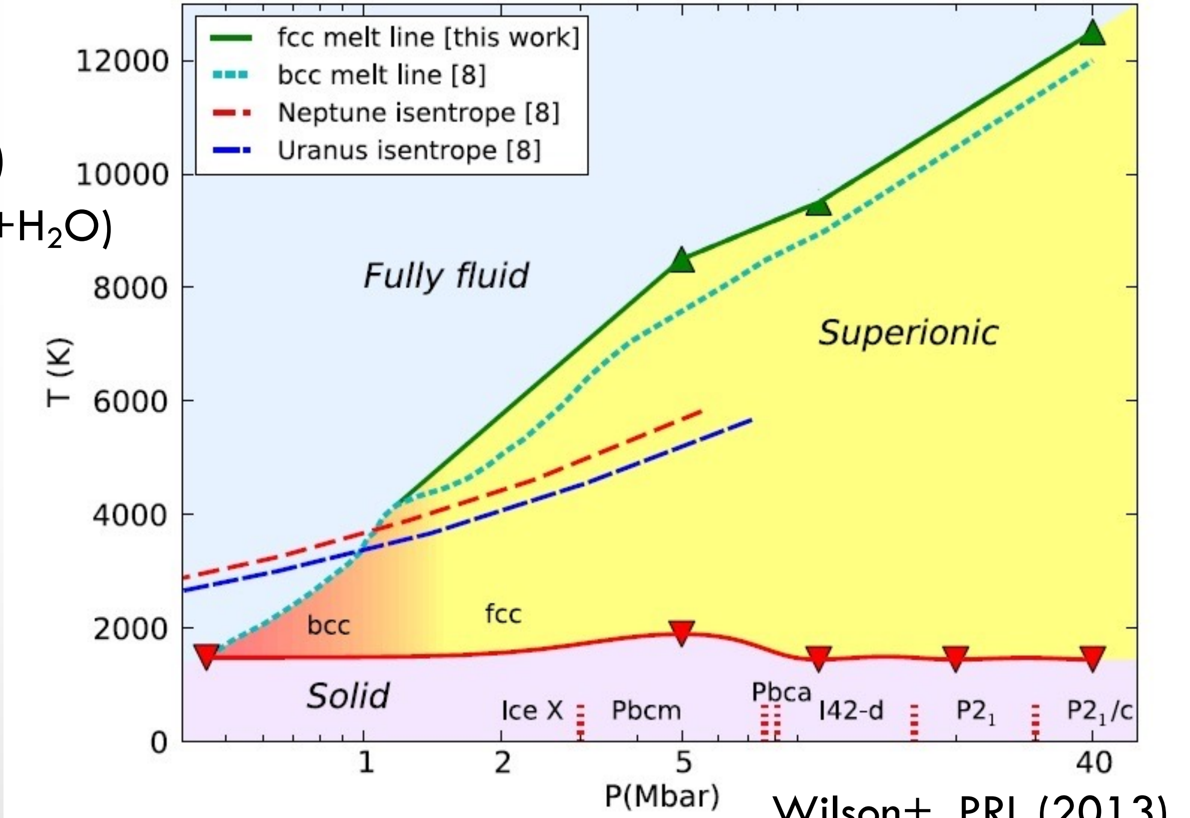
Do water and silicates mix?

Kovacevic+, Sci. Rep. (2022)

PLANETARY INTERIORS

WATER WORLDS





Wilson+, PRL (2013).

Do water and silicates mix?

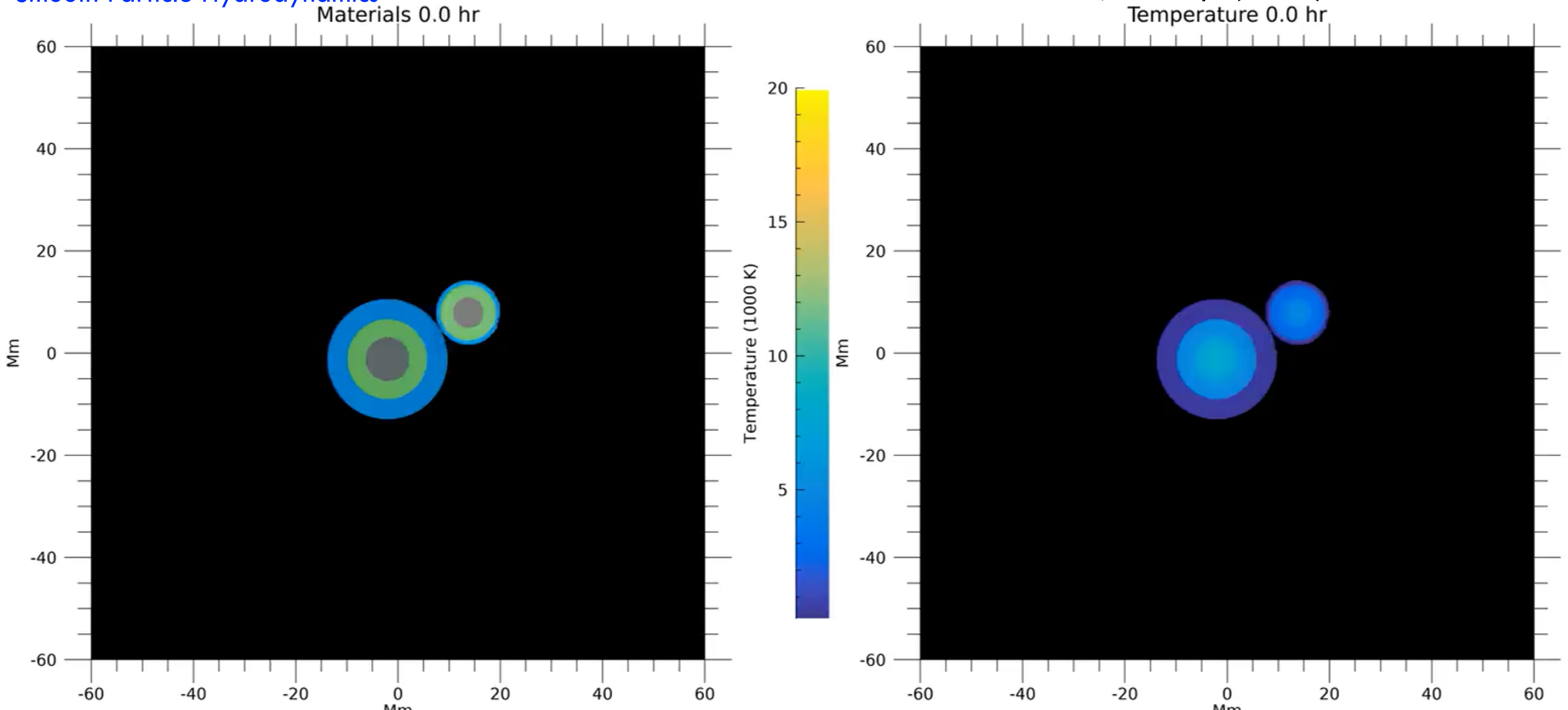
Kovacevic+, Sci. Rep. (2022)

PLANETARY INTERIORS

WATER WORLDS

Smooth Particle Hydrodynamics

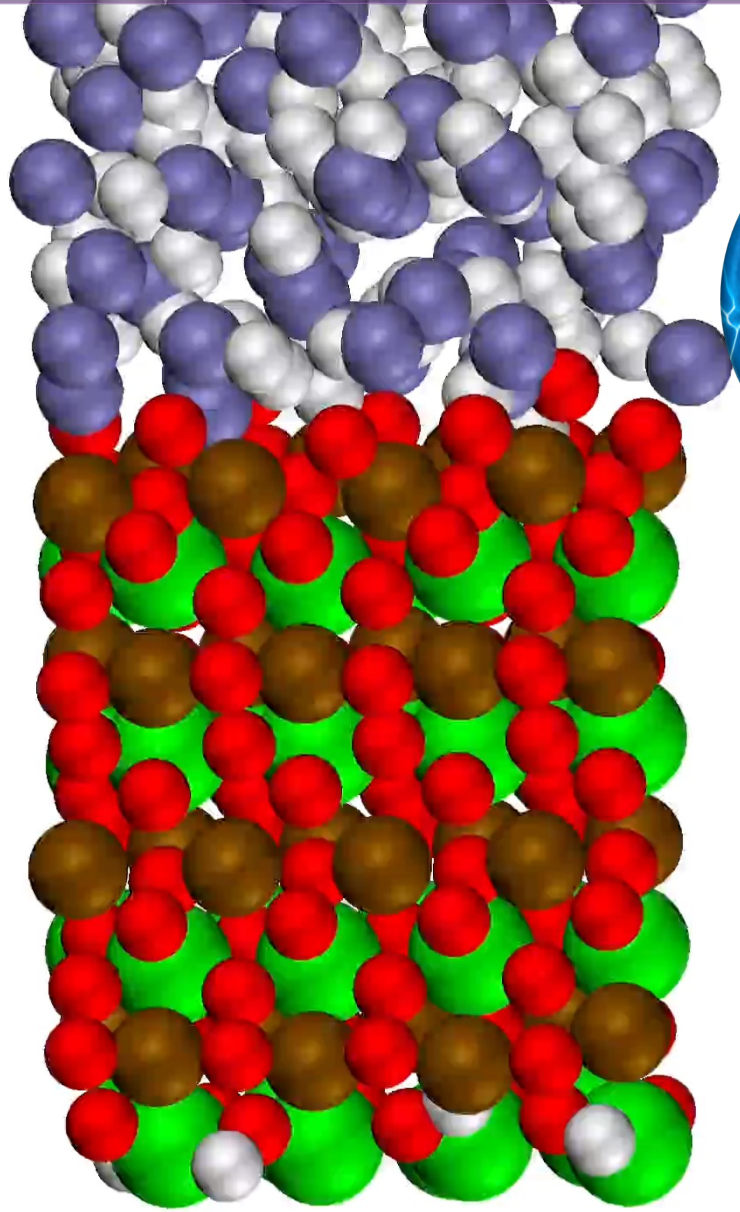
Kovacevic+, Sci. Rep. (2022)

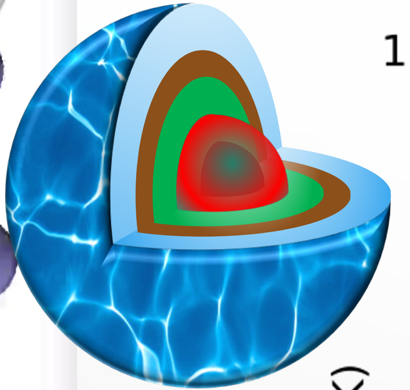


PLANETARY INTERIORS

WATER WORLDS

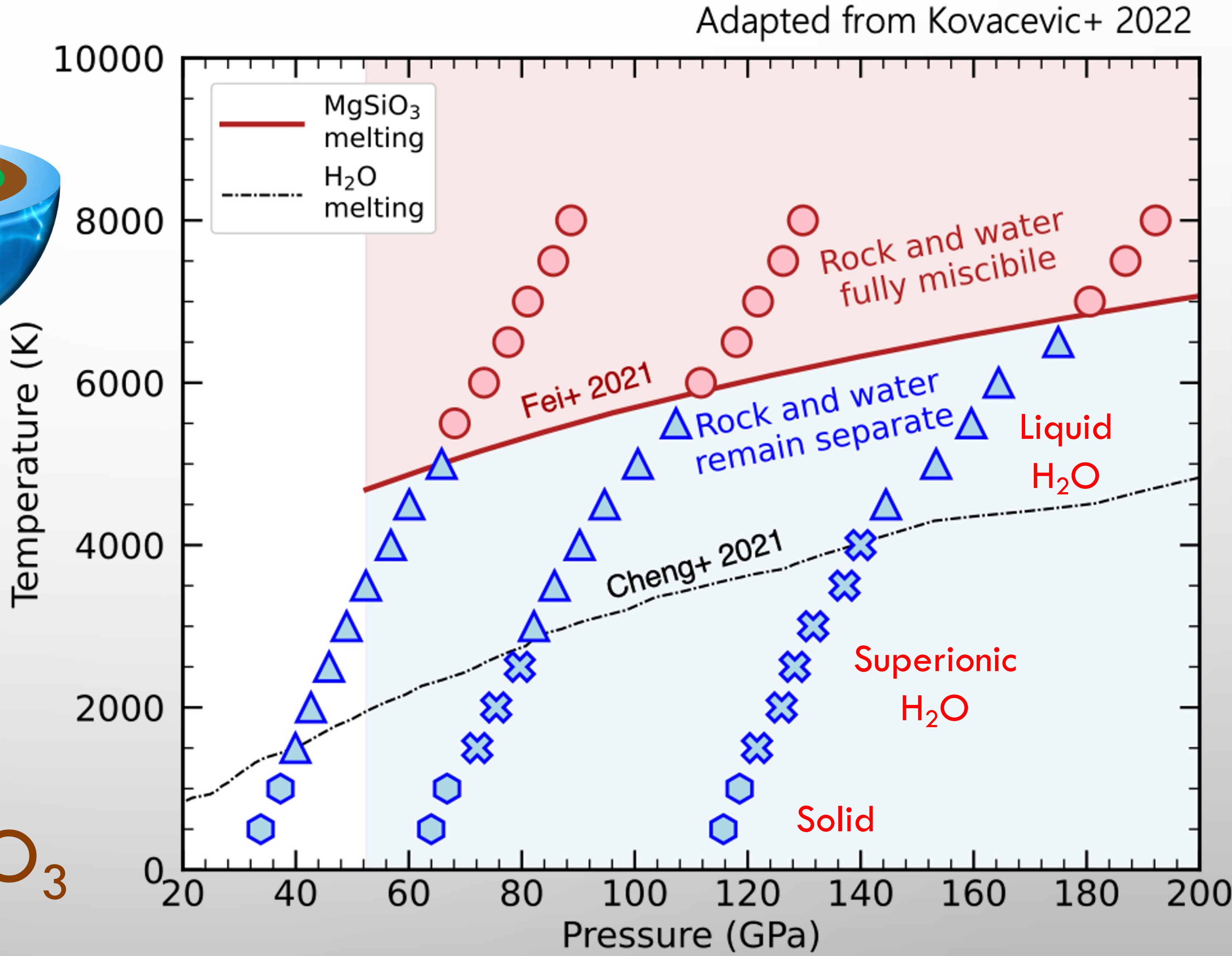
H_2O





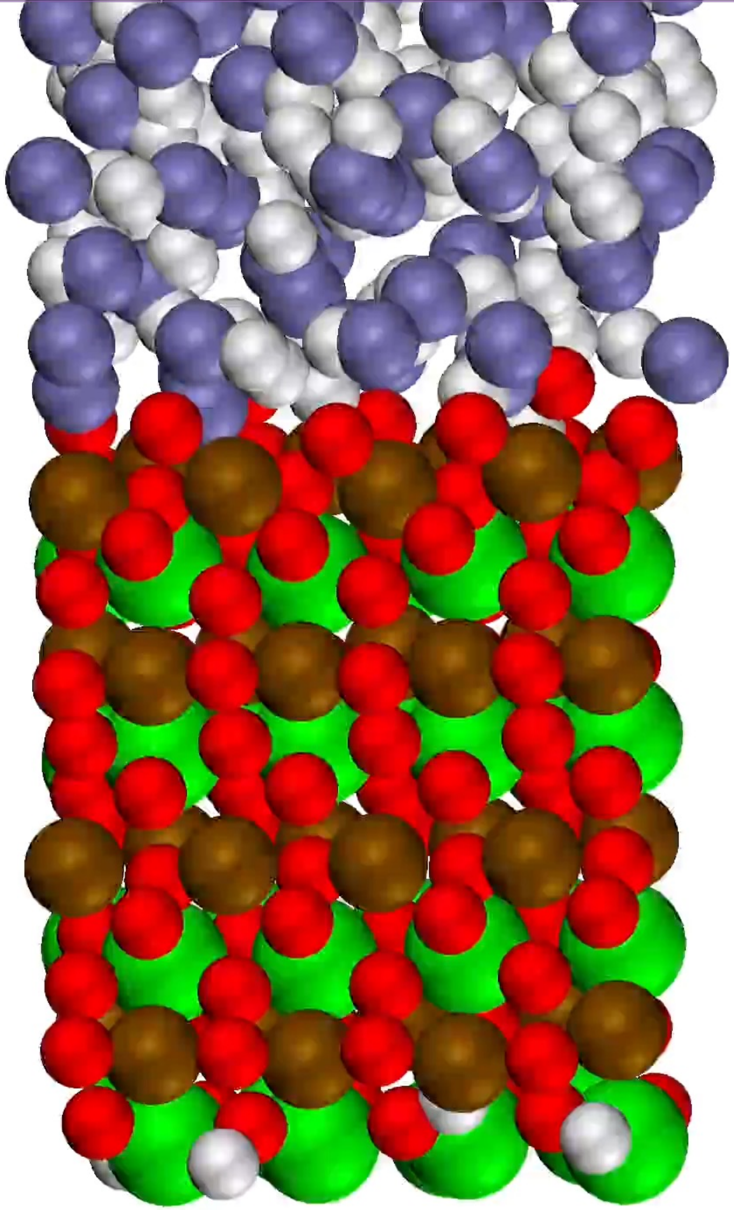
$MgSiO_3$

Kovacevic+, Sci. Rep. (2022)

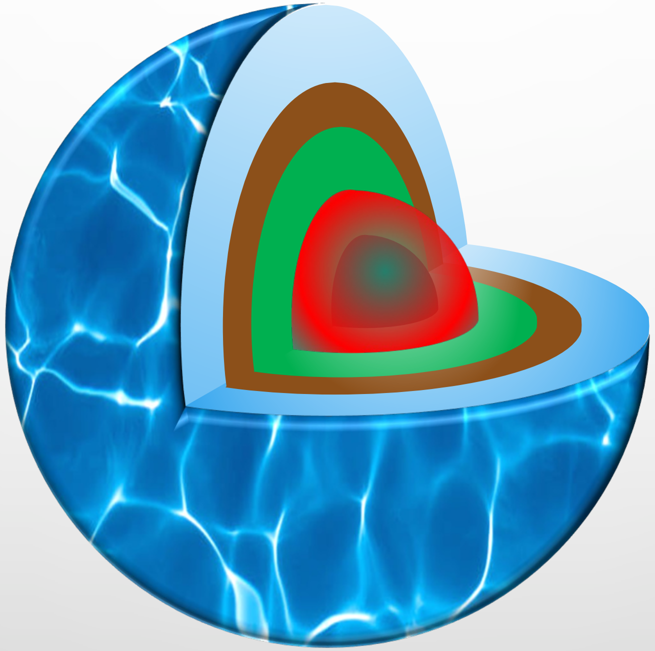


PLANETARY INTERIORS

WATER WORLDS

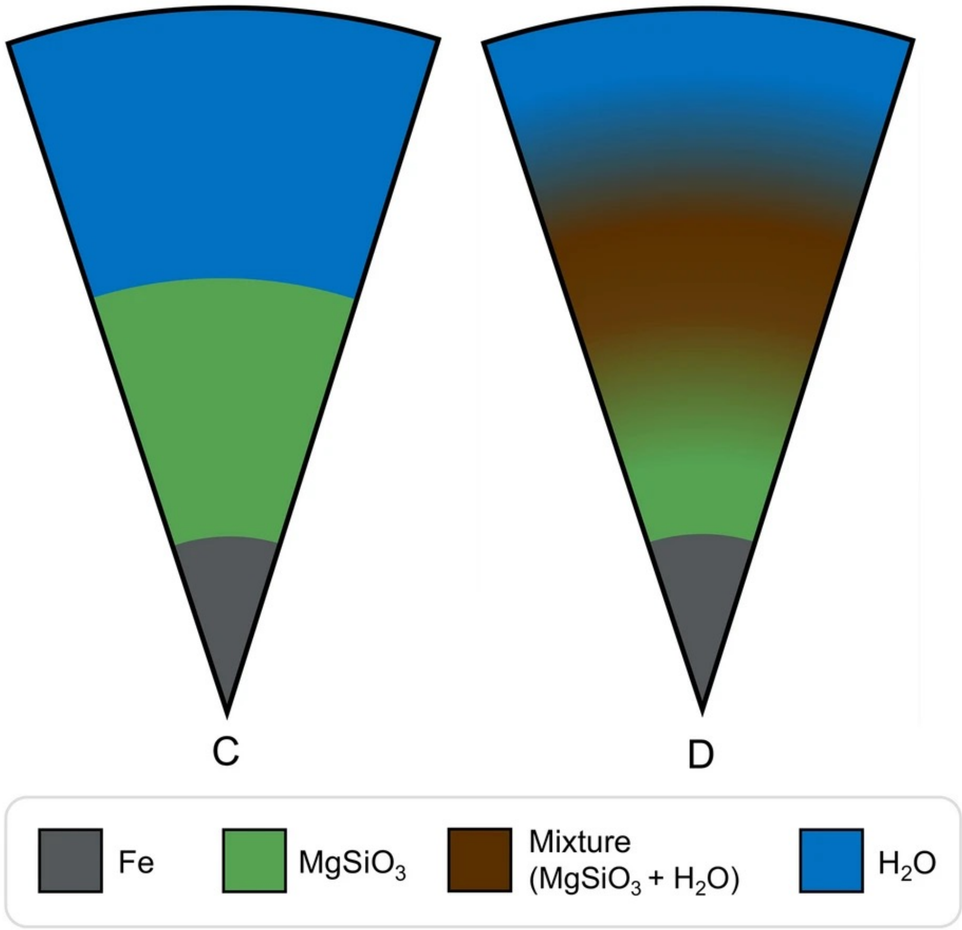


H_2O



$MgSiO_3$

Kovacevic+, Sci. Rep. (2022)



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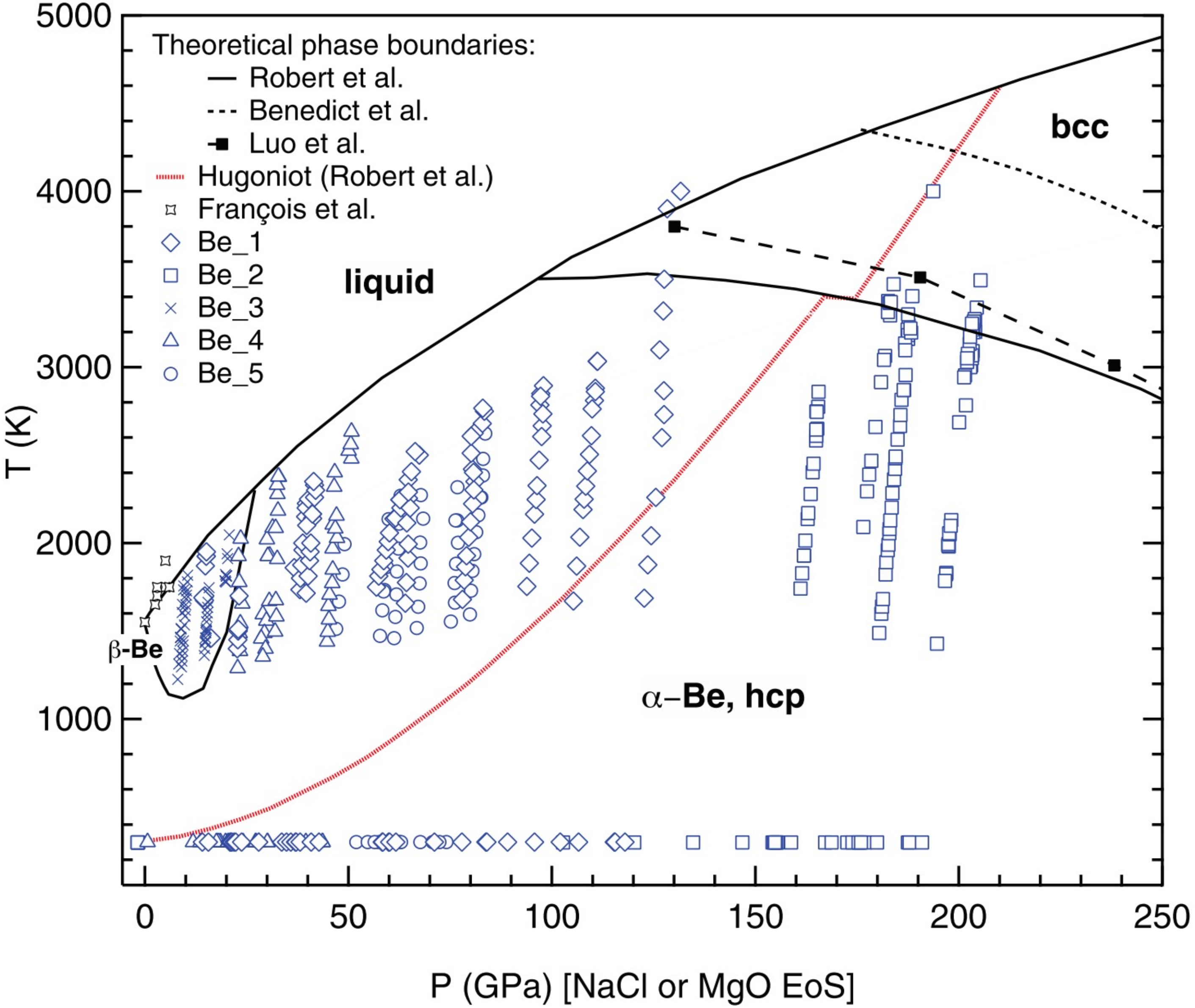
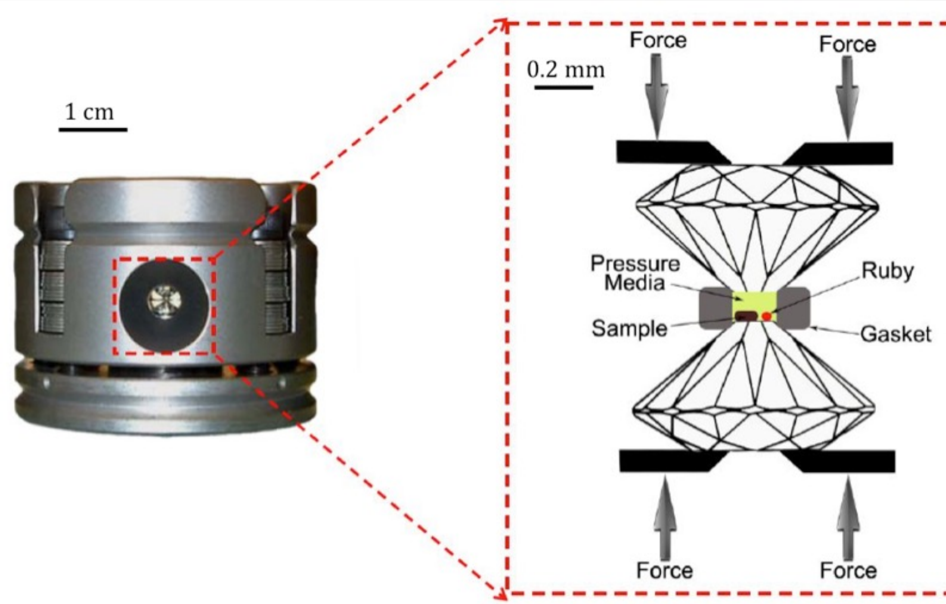
HIGH PRESSURE

Beryllium



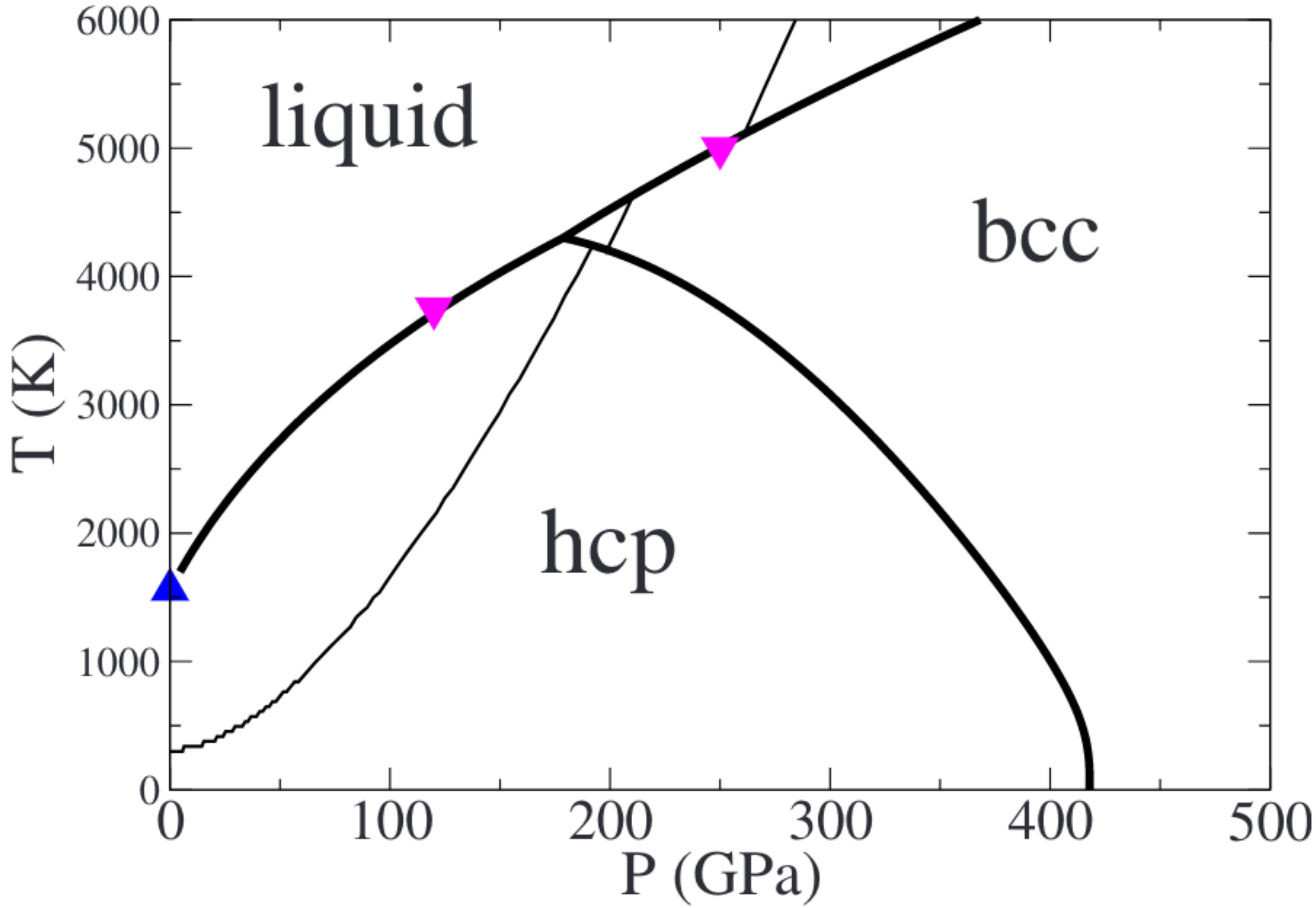
- Laser Heated DAC
- No signature of bcc



Lazicki+ PRB (2012)

HIGH PRESSURE

Beryllium



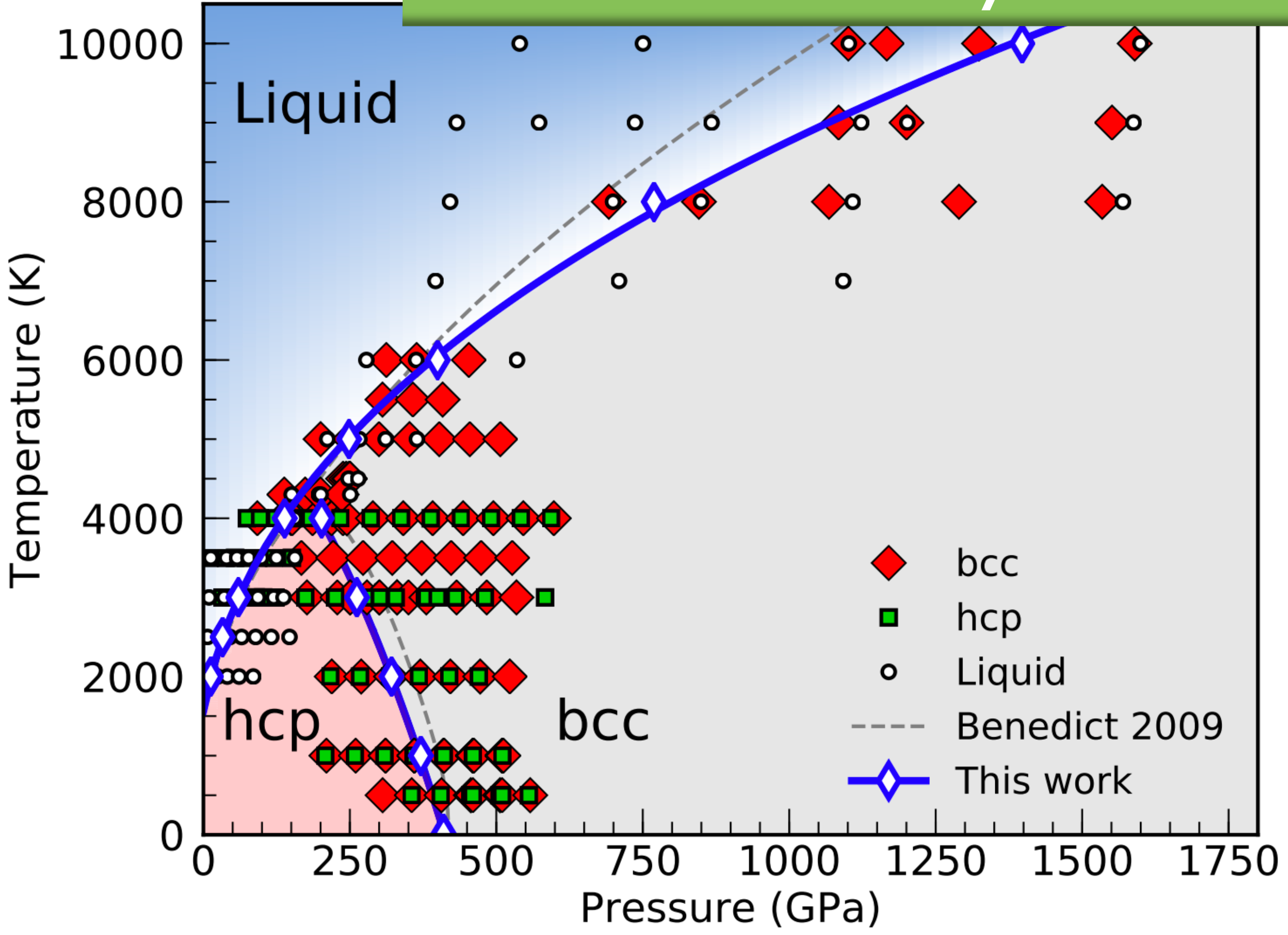
- QHA (quasi-harmonic approximation)
- No signature of bcc in experiments
- Two-phase simulations of melting

Benedict+ PRB (2009)

HIGH PRESSURE

Beryllium

Thermodynamic Integration



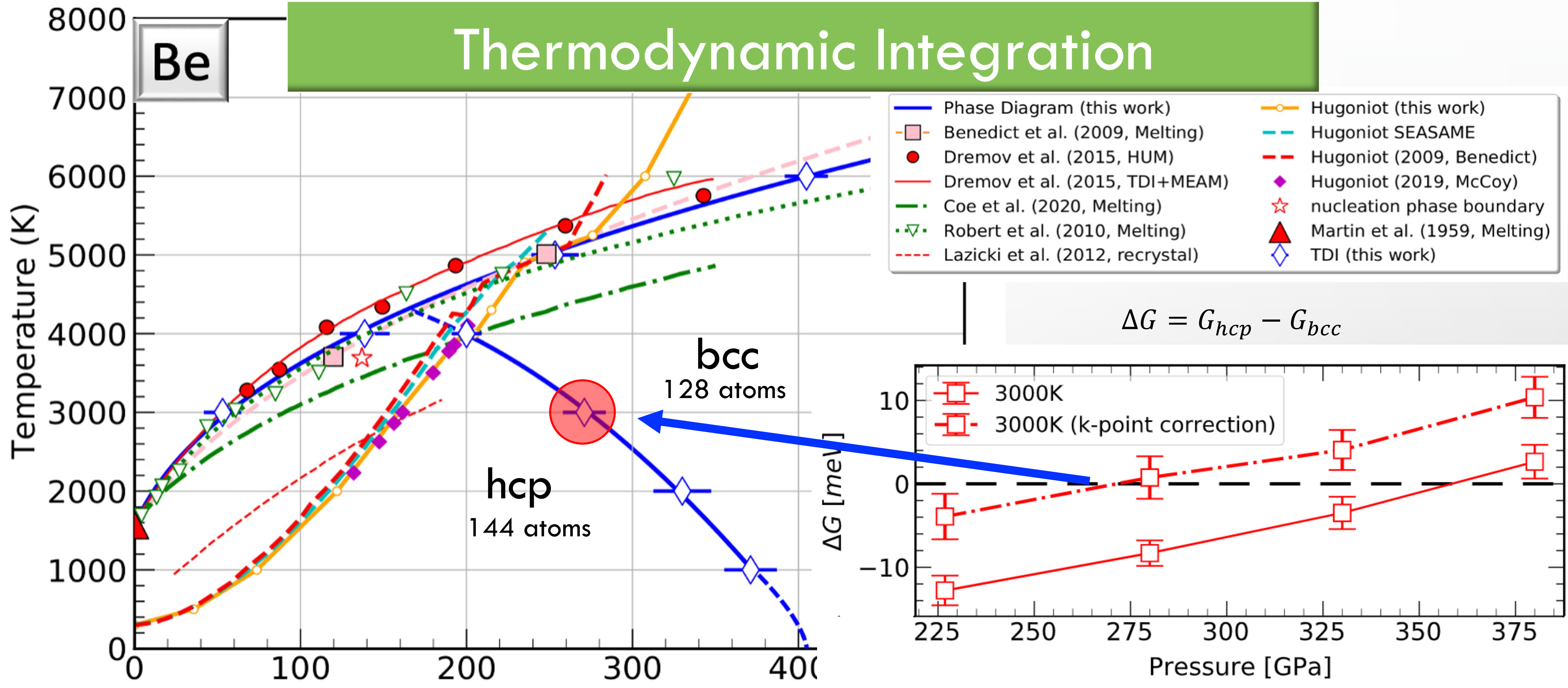
- DFT-MD
- hcp/bcc/liquid

Wu, Gonzalez, Militzer, PRB (2021)

HIGH PRESSURE

Beryllium

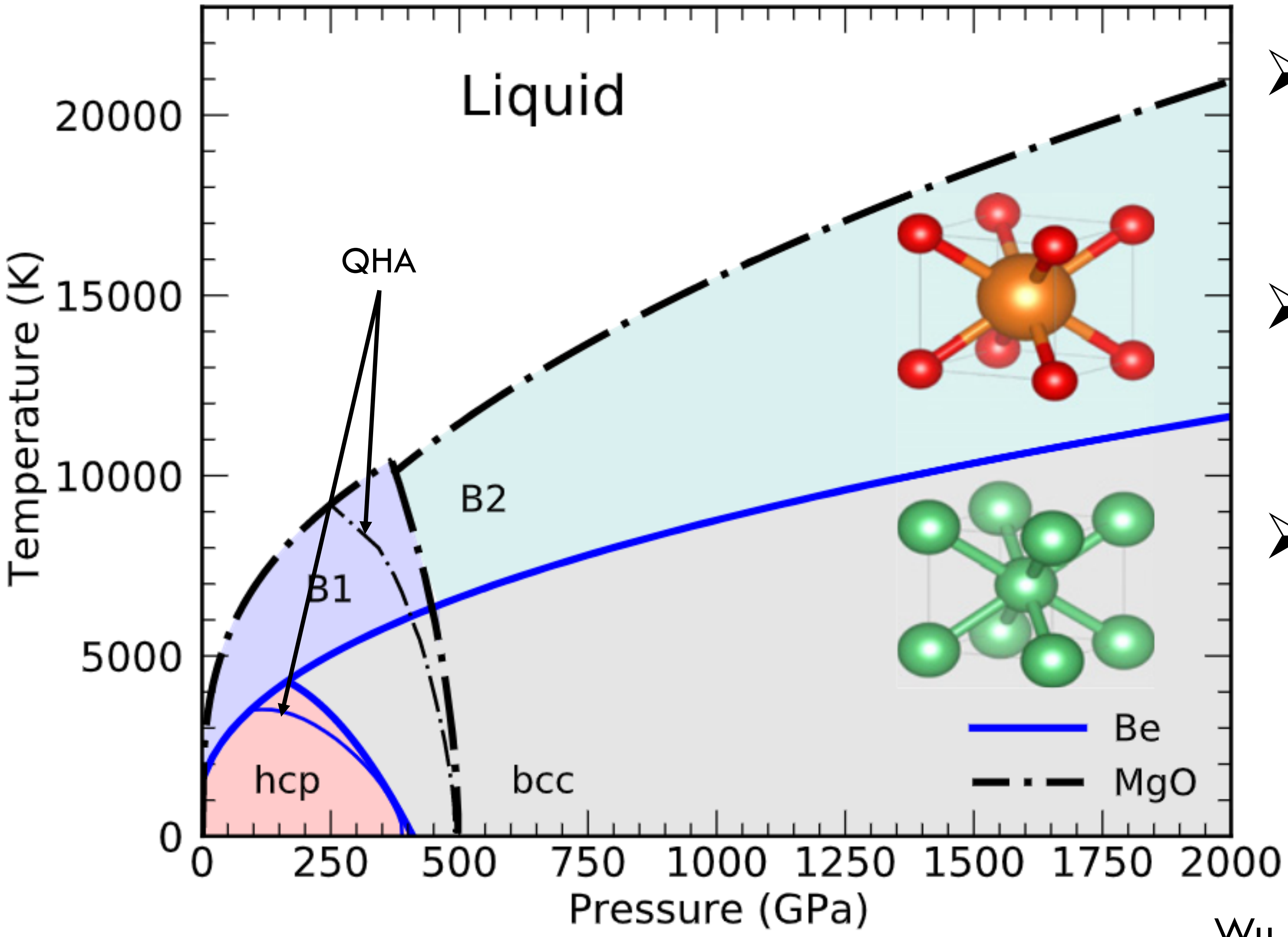
Thermodynamic Integration



Wu, Gonzalez, Militzer, PRB (2021)

HIGH PRESSURE

Beryllium & MgO



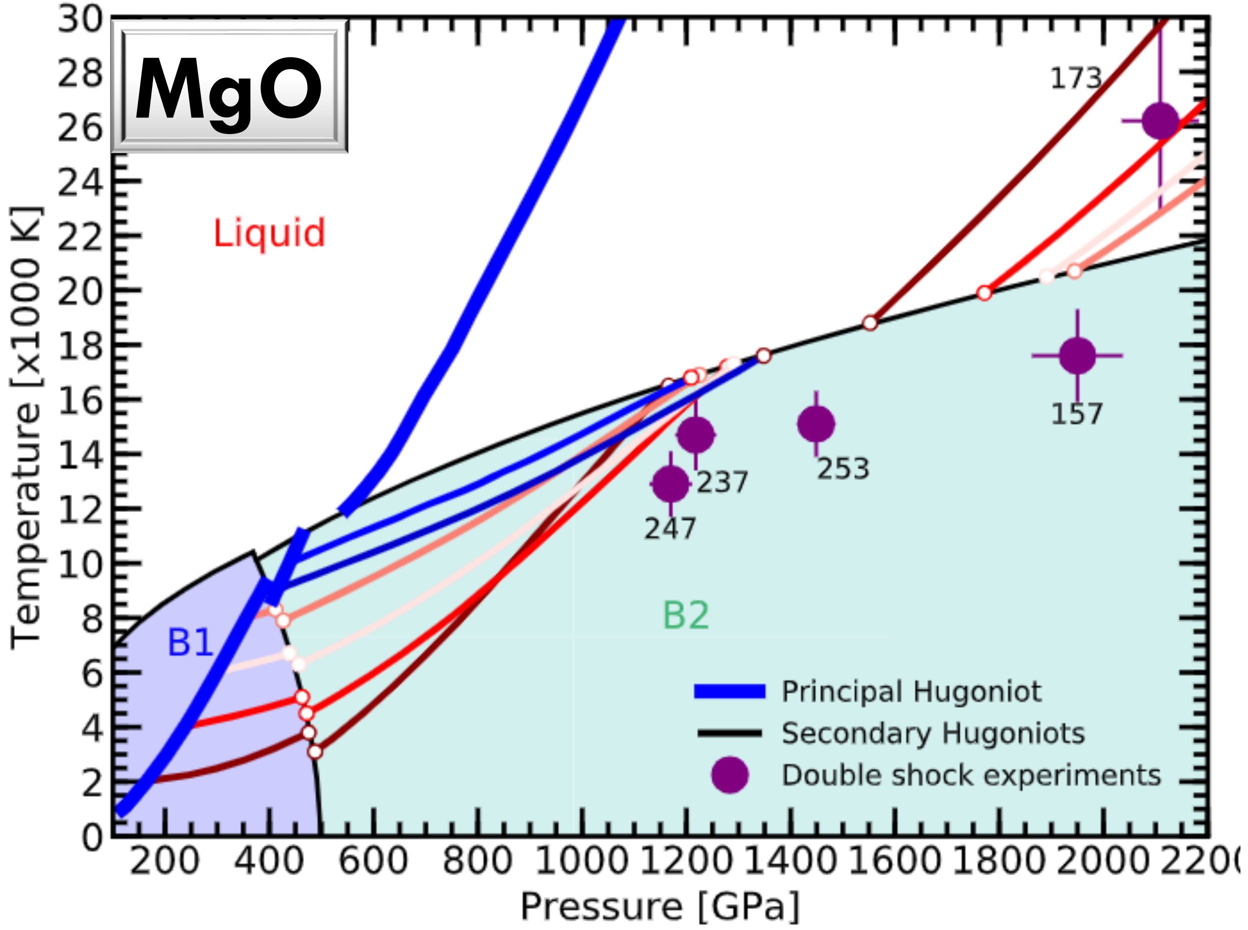
- $\Delta G = G_{liq} - G_{sol}$
  Melting lines Be & MgO
- 2x2x2 k-points in Be in ~100-atoms cells
- Strong anharmonicities:
  - QHA does not work well.
  - B1-B2 / hcp-bcc needs higher P.

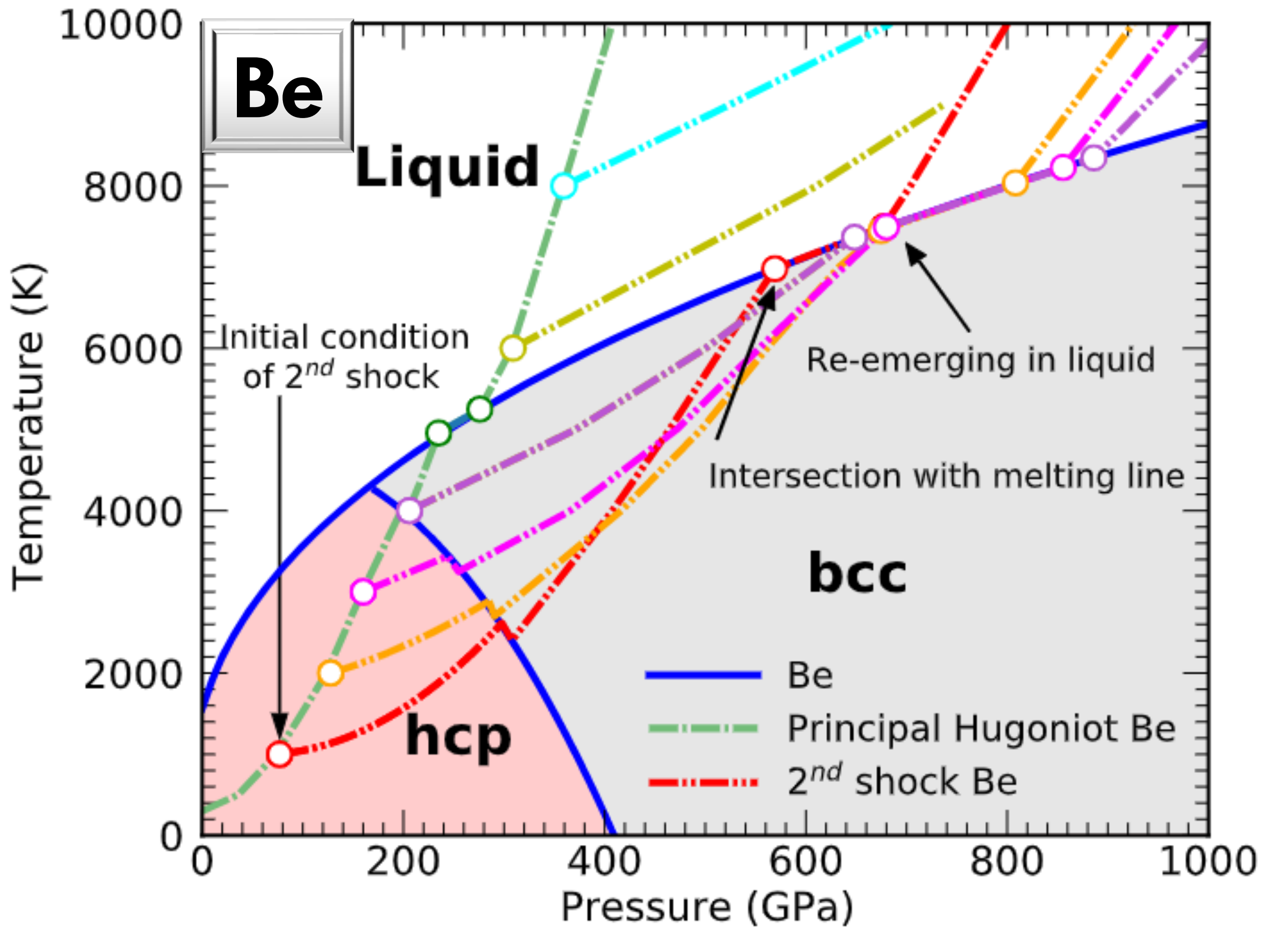
Wu, Gonzalez, Soubiran, Militzer, JPCM (2022)

HIGH PRESSURE

Beryllium & MgO

Double Shock compression





Wu, Gonzalez, Soubiran, Militzer, JPCM (2022)

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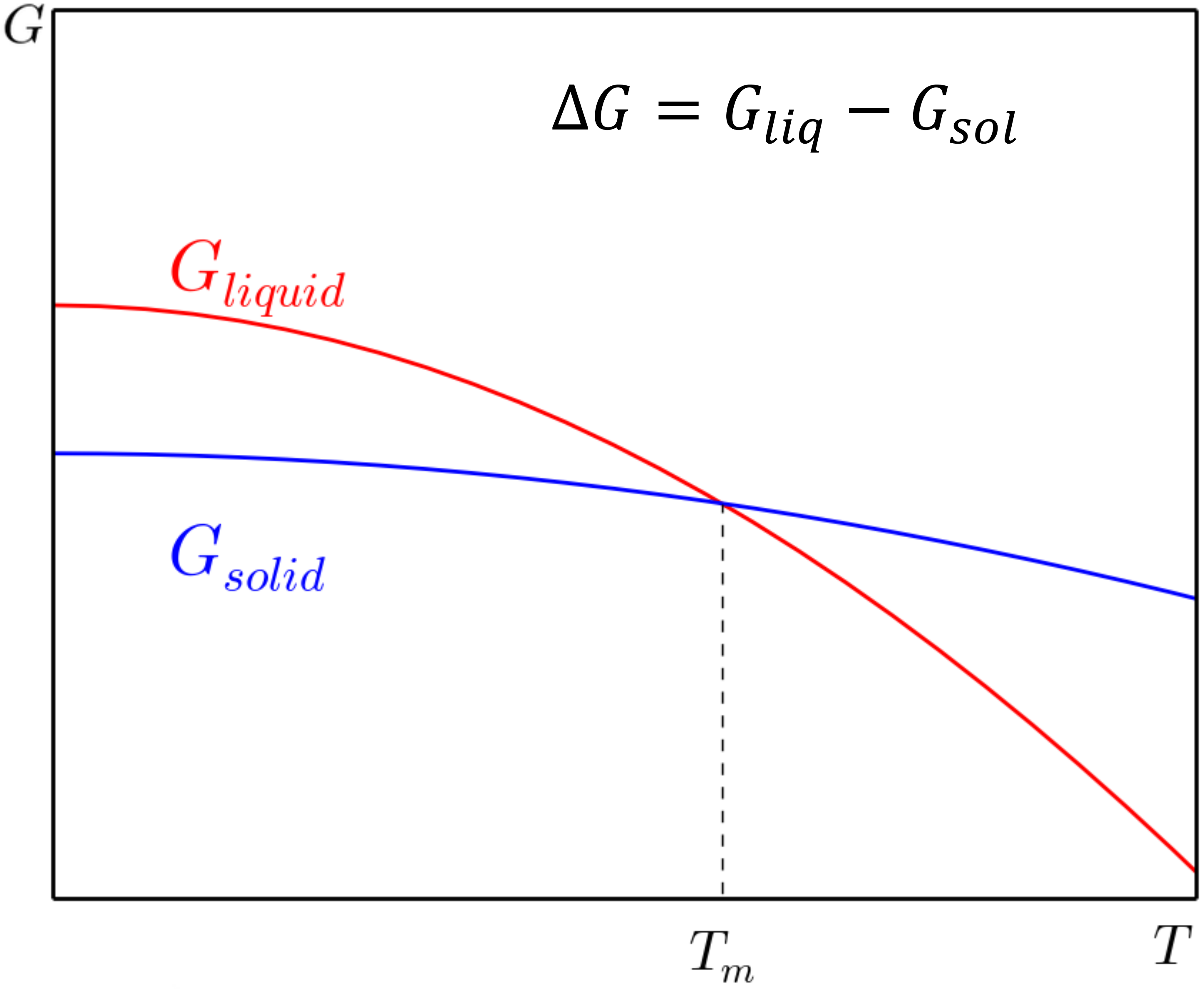
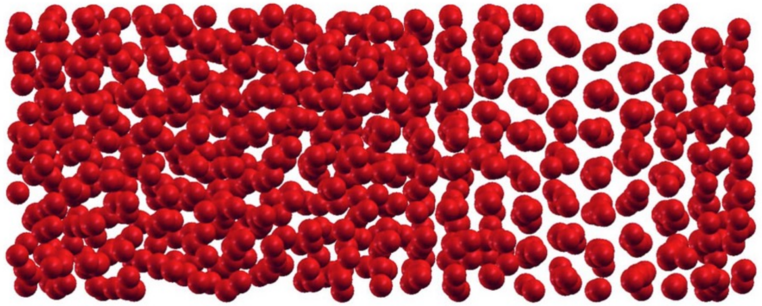
HIGH PRESSURE

Melting SiO_2

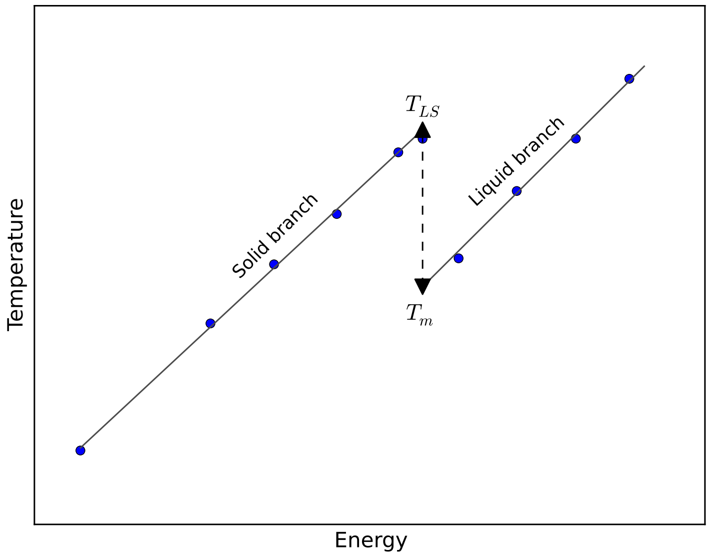


Coexistence (Two phase)



Alfe, PRB (2009)

Z method

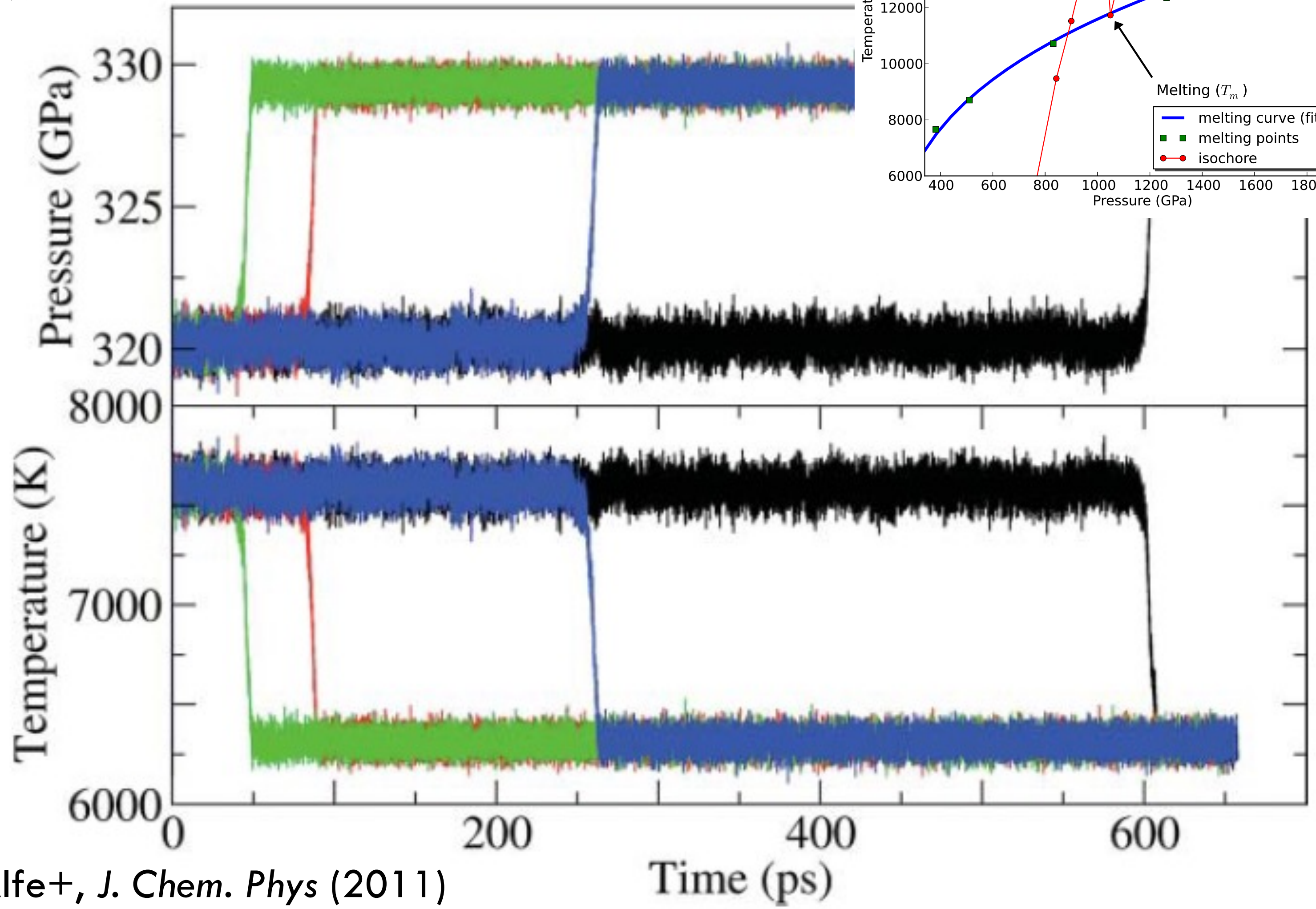


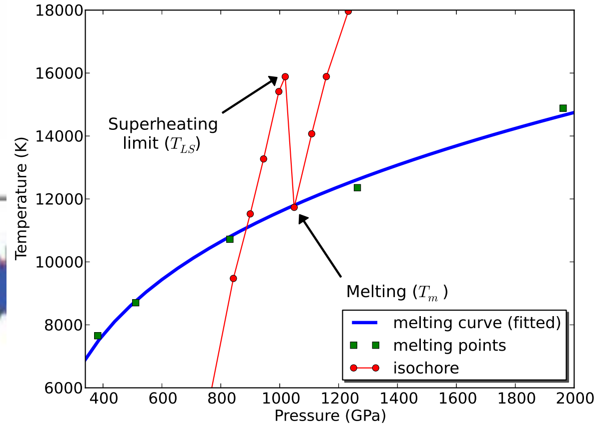
HIGH PRESSURE

Melting SiO_2

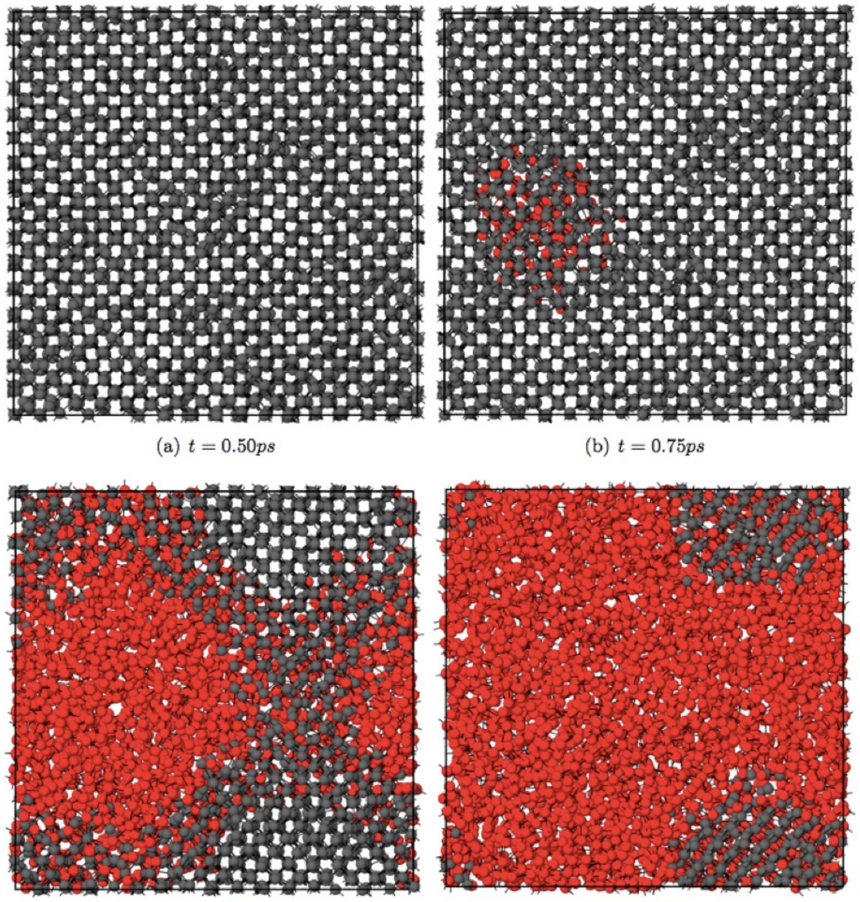
Z method

NVE heat until it melts





Alfe+, *J. Chem. Phys* (2011)



(a) $t = 0.50ps$ (b) $t = 0.75ps$

(c) $t = 1.50ps$ (d) $t = 4.00ps$

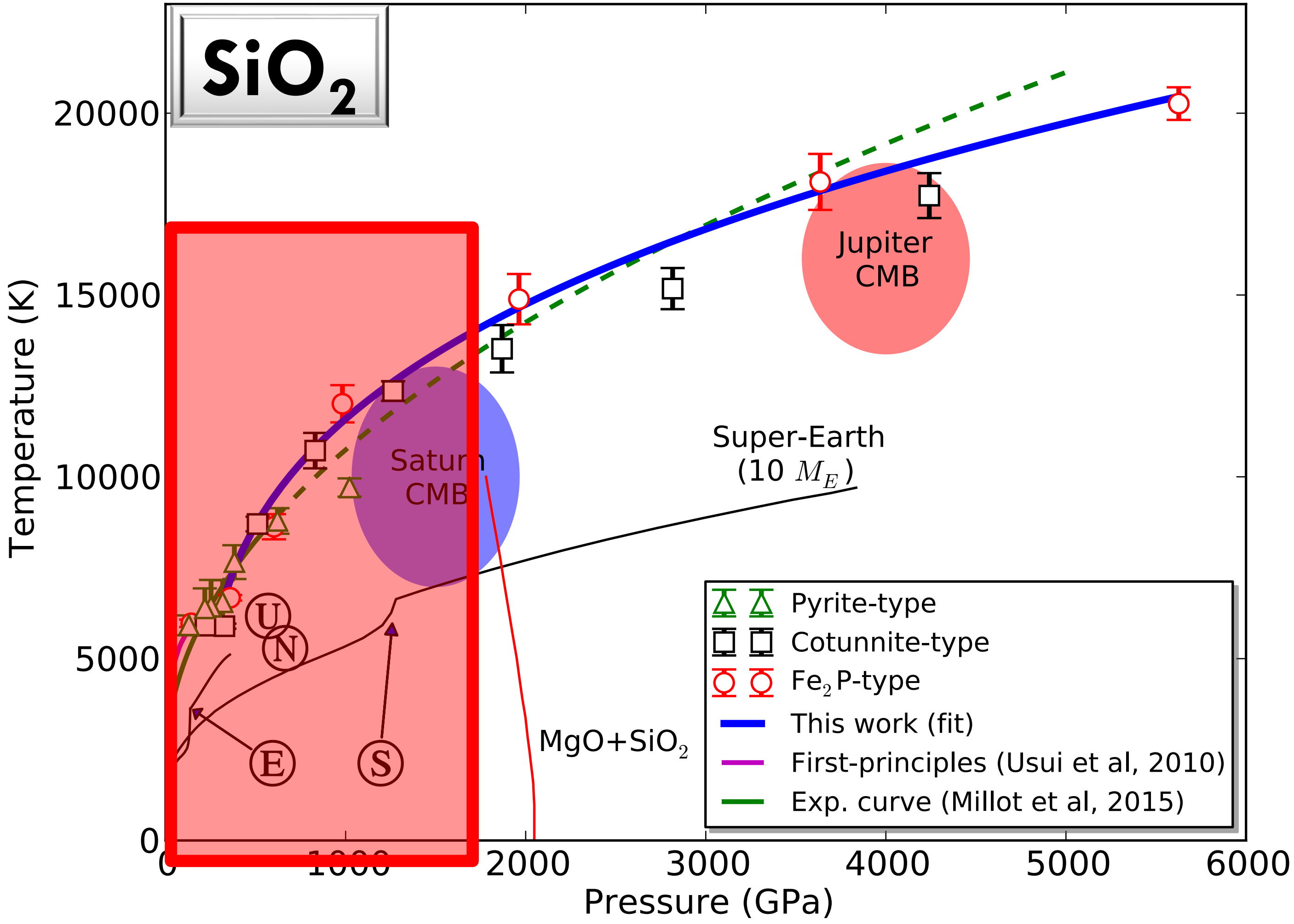
Gonzalez+*J. Phys. Conf. Series* (2018)

HIGH PRESSURE

Melting SiO_2

Z method

NVE heat until it melts



SiO_2 is solid at CMB of

- Super-Earths
- Gas Giants

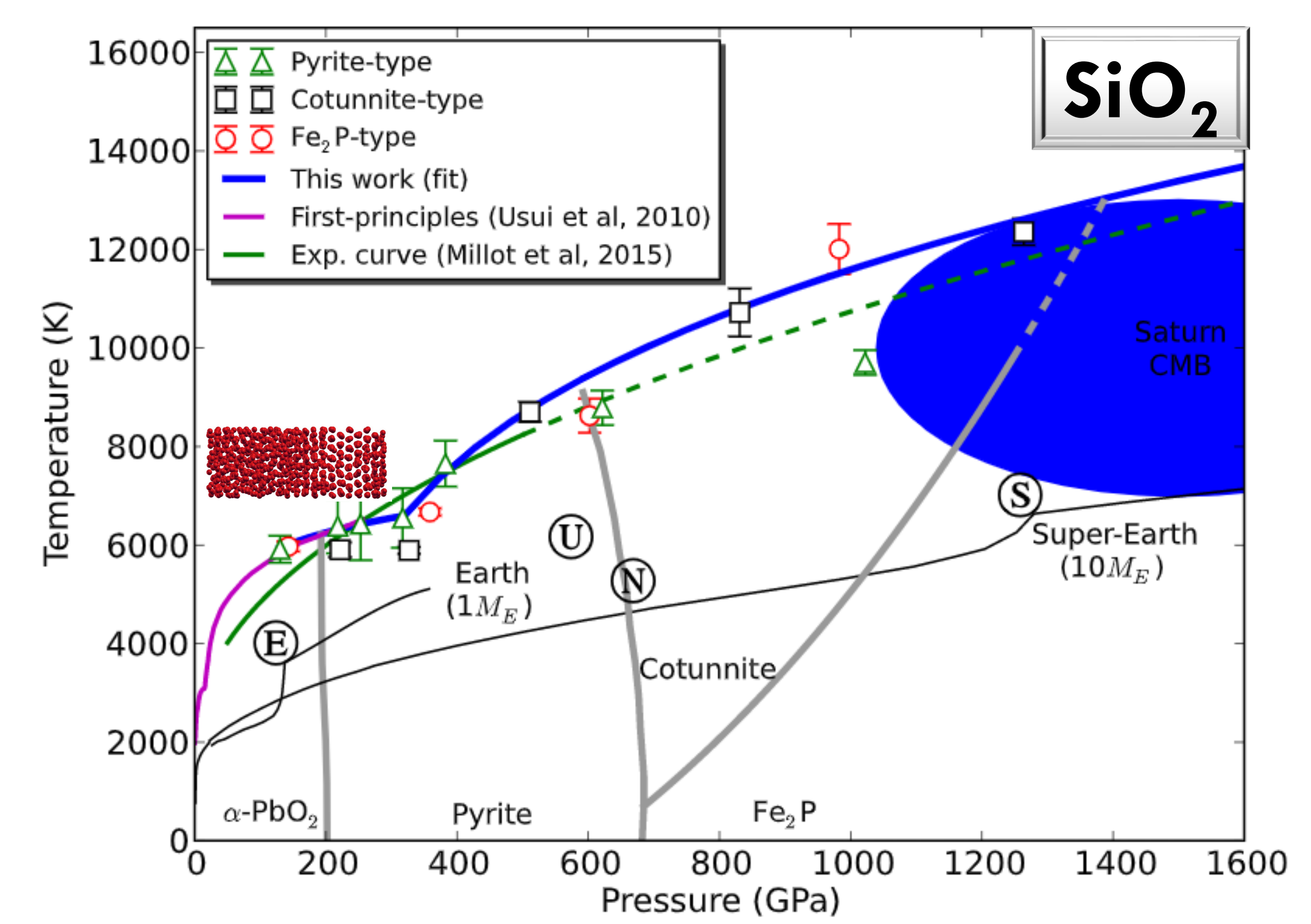
Gonzalez, Davis, Gutierrez, Sci. Rep. (2015)

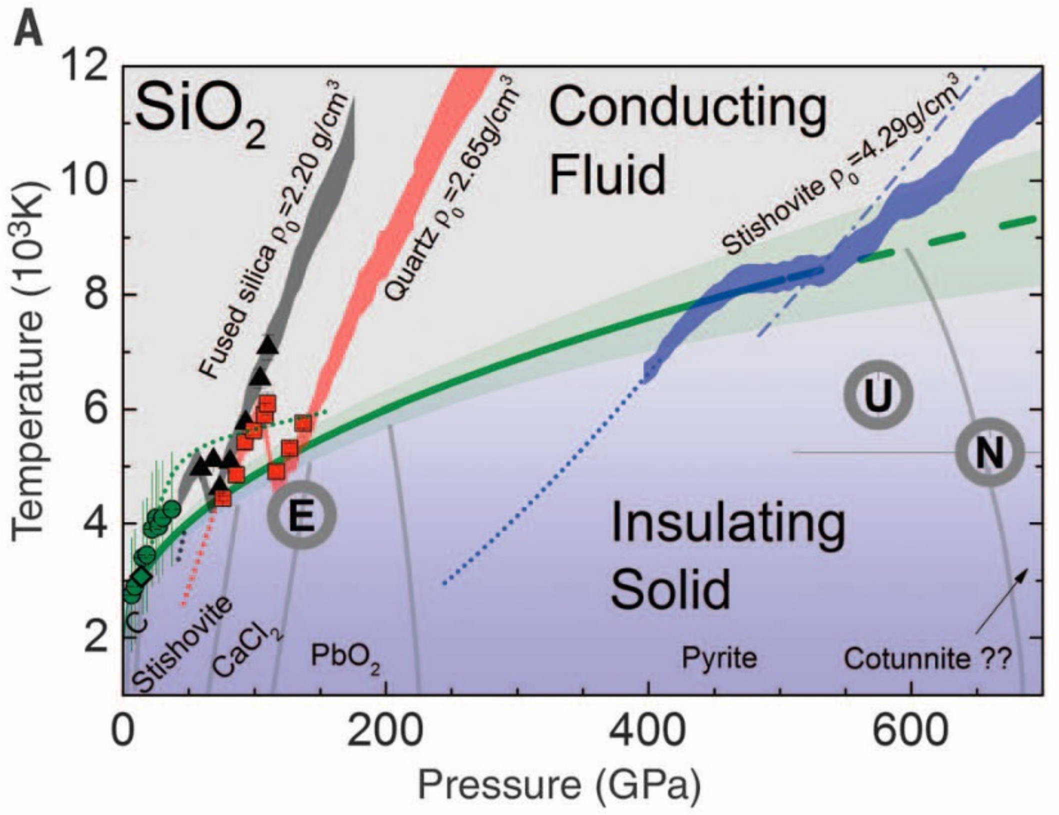
HIGH PRESSURE

Melting SiO_2

Z method

NVE heat until it melts





Millot+, Science (2015)

Z method agreement with

- Two-phase simulations
- Experiments

Gonzalez, Davis, Gutierrez, Sci. Rep. (2015)

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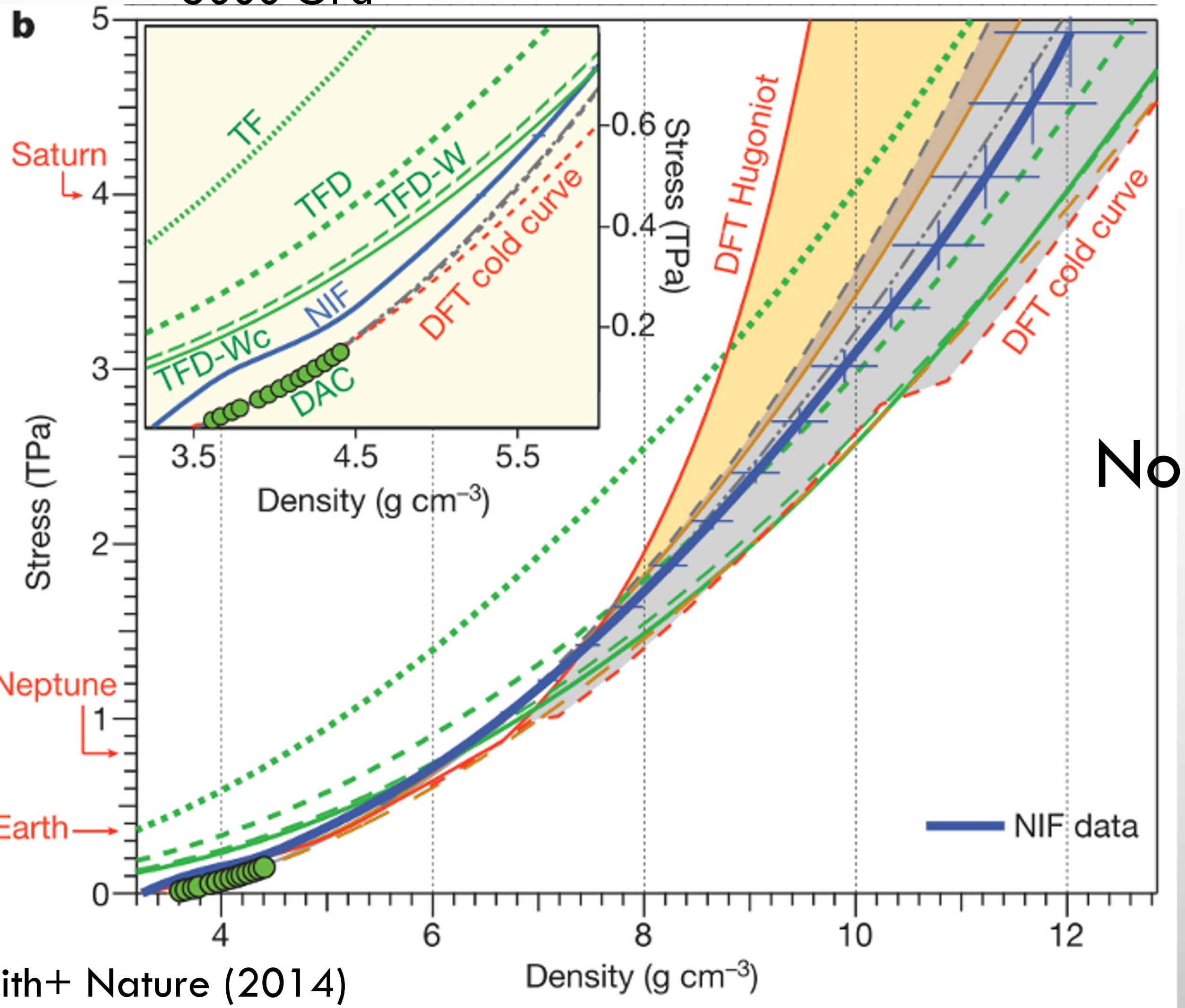
Ramp compression model

Diamond  from ab initio simulations

Gonzalez+, PRB (2021)

5000 GPa



Smith+ Nature (2014)

No signature of bc8

2000 GPa

Article

Metastability of diamond ramp-compressed to 2 terapascals

Lazicki+, Nature (2021)

https://doi.org/10.1038/s41586-020-03140-4
Received: 1 June 2020
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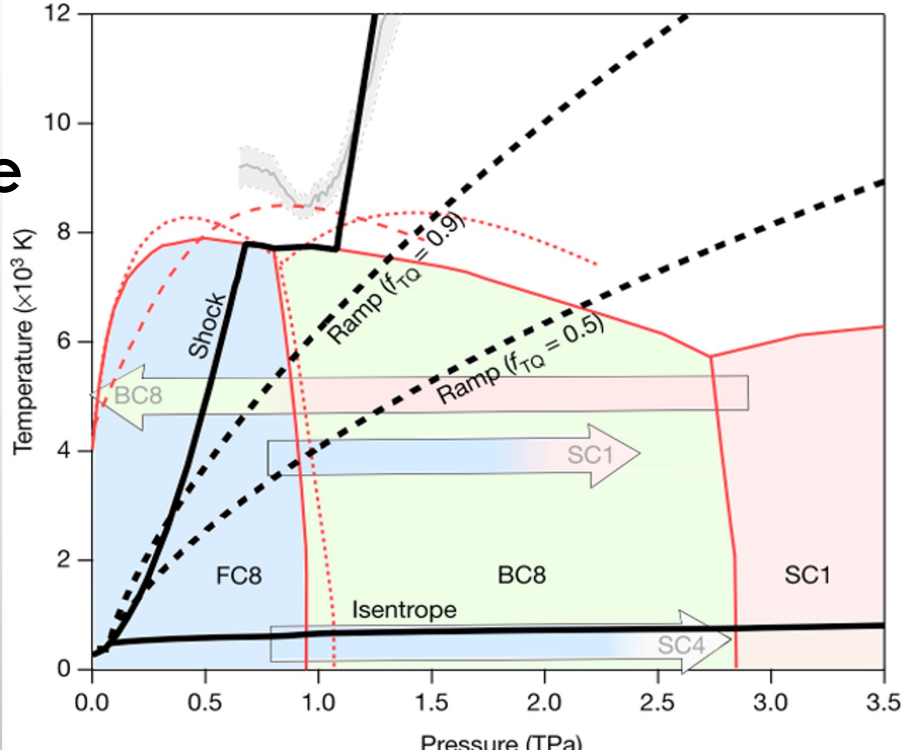
A. Lazicki[1,22], D. McGonegle[2], J. R. Rygg[3,4,5], D. G. Braun[1], D. C. Swift[1], M. G. Gorman[1], R. F. Smith[1], P. G. Heighway[2], A. Higginbotham[6], M. J. Suggit[2], D. E. Fratanduono[1], F. Coppari[1], C. E. Wehrenberg[1], R. G. Kraus[1], D. Erskine[1], J. V. Bernier[1], J. M. McNaney[1], R. E. Rudd[1], G. W. Collins[3,4,5], J. H. Eggert[1] & J. S. Wark[2]

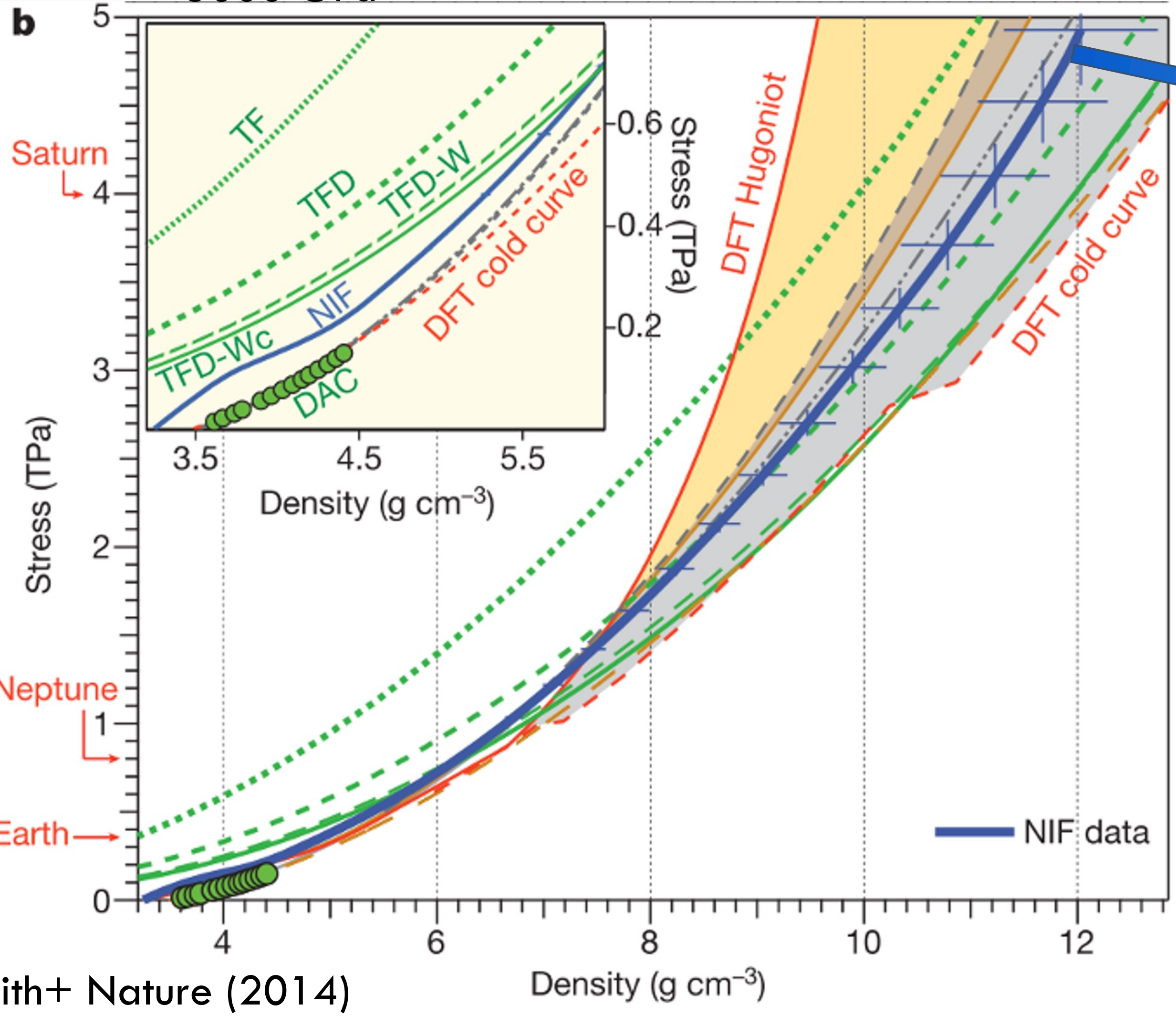


Ramp compression model

Gonzalez+, PRB (2021)

from ab initio simulations

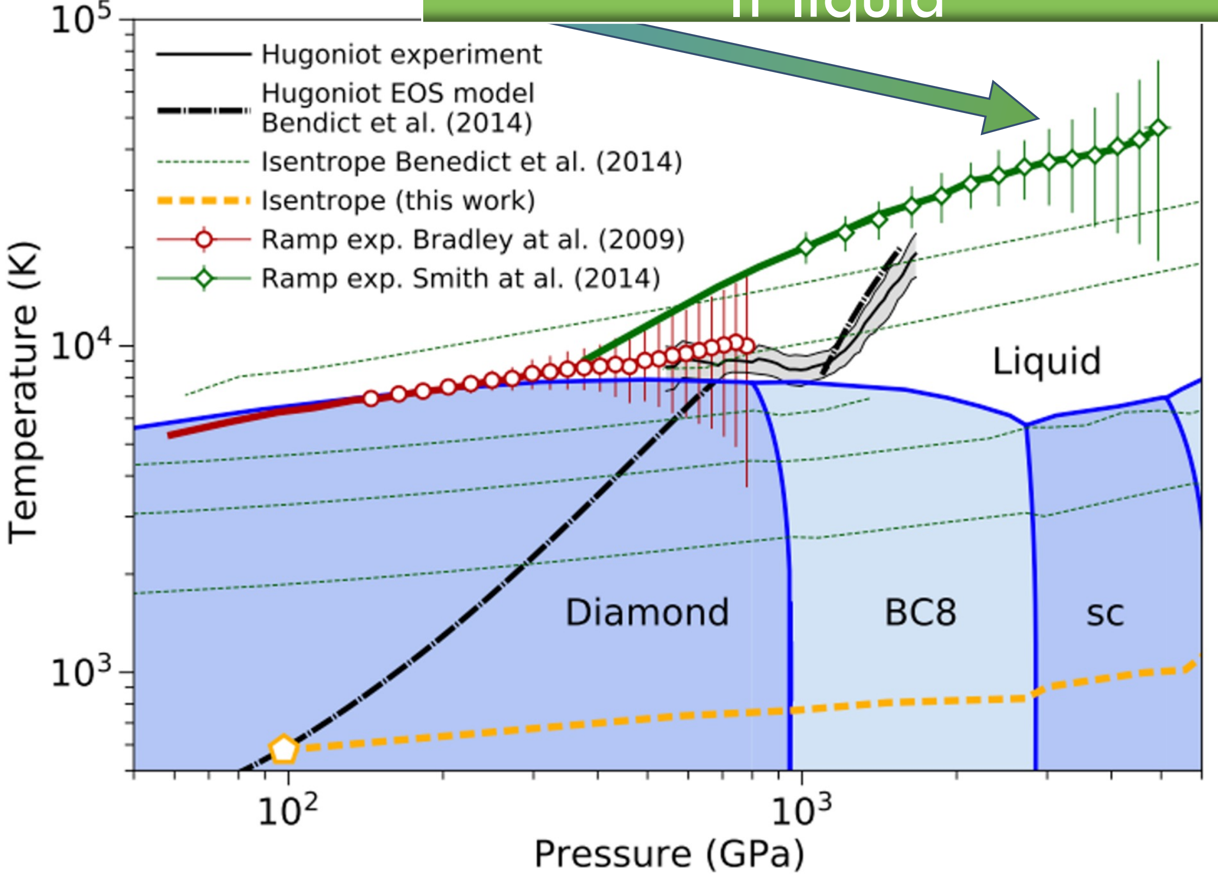
5000 GPa



Smith+ Nature (2014)

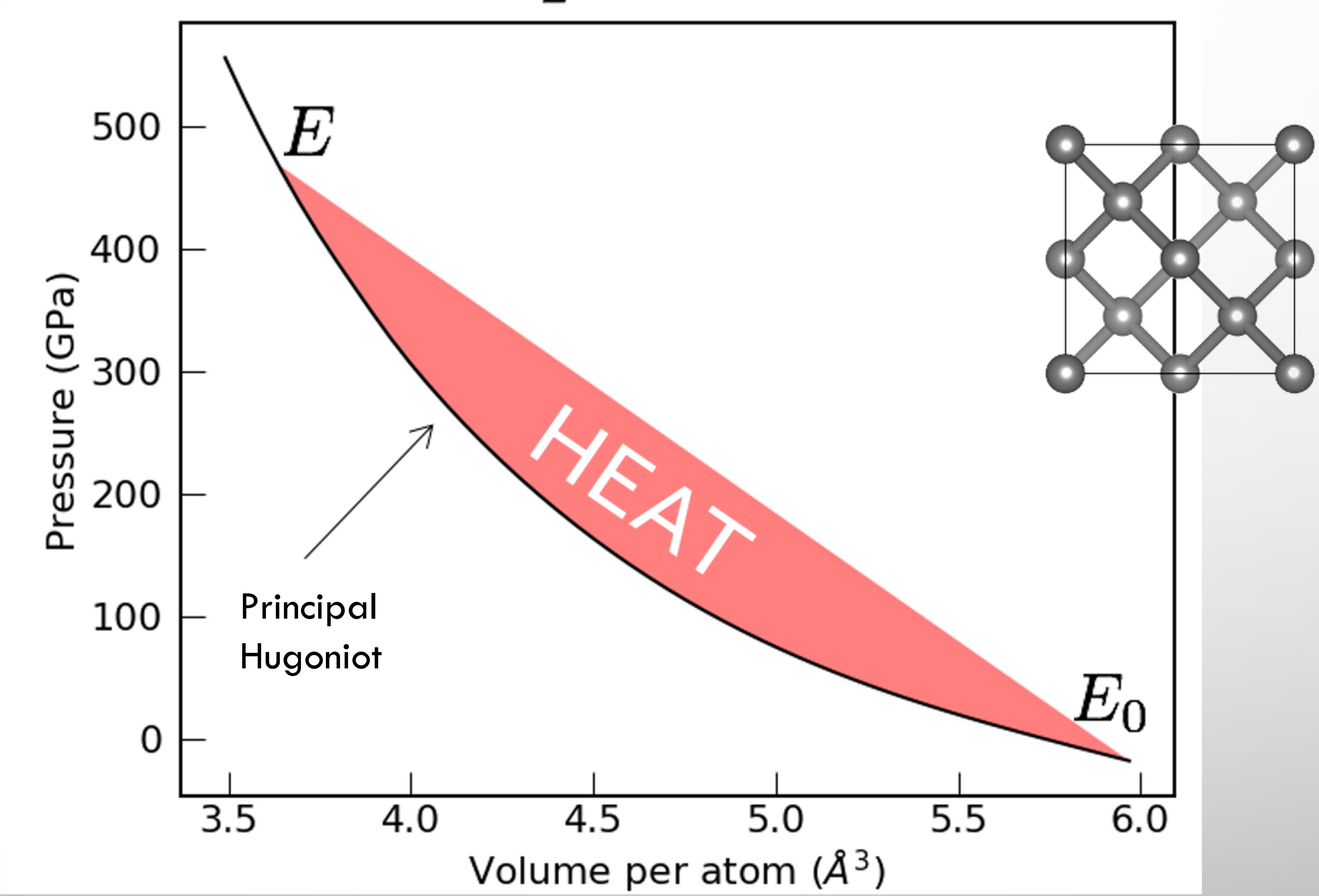
$T > 20\ 000$ K

If liquid



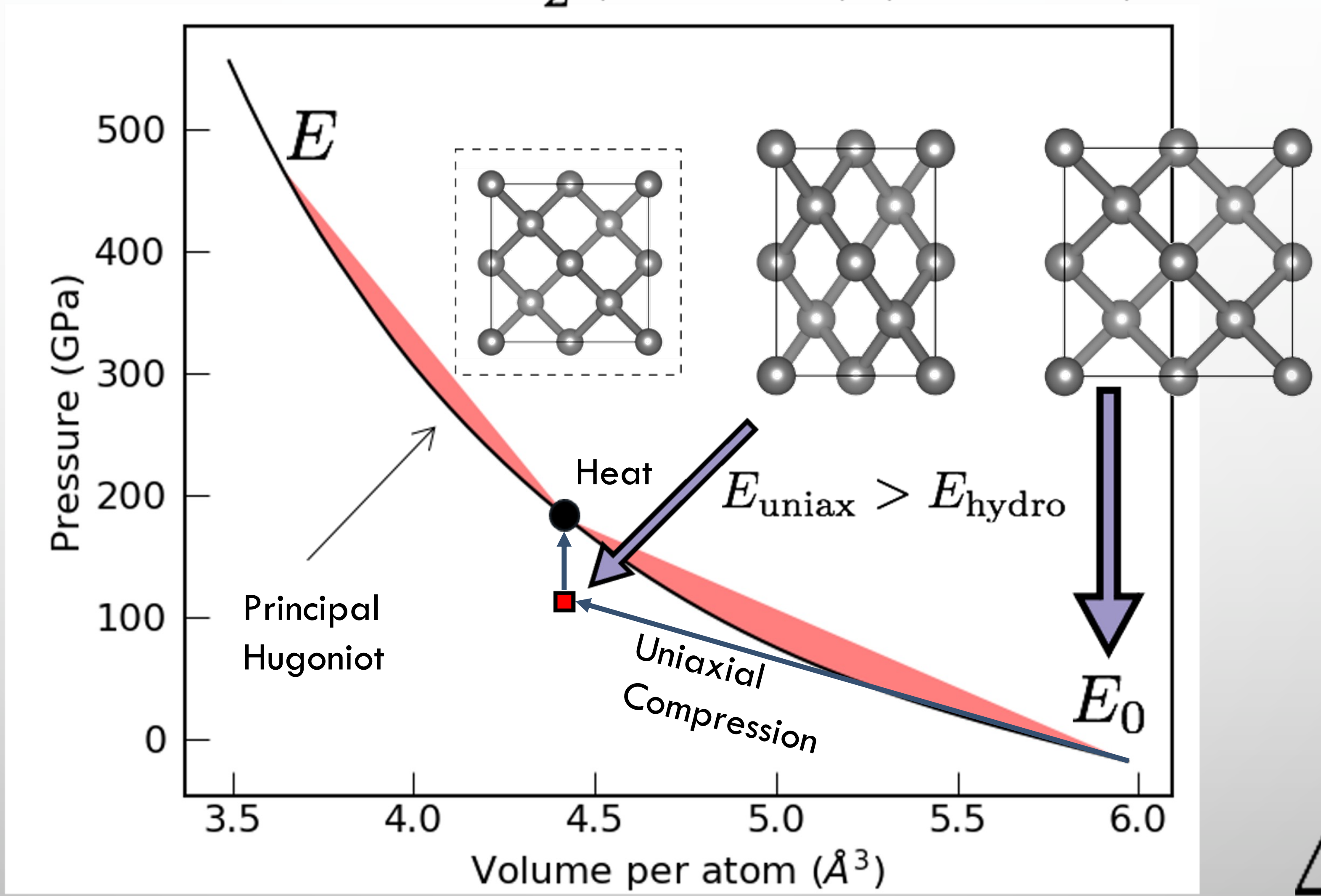
Ramp ~ multishocks

$$E - E_0 = \frac{1}{2}(P + P_0)(V_0 - V)$$



Ramp ~ multishocks

$$E - E_0 = \frac{1}{2}(P + P_0)(V_0 - V)$$

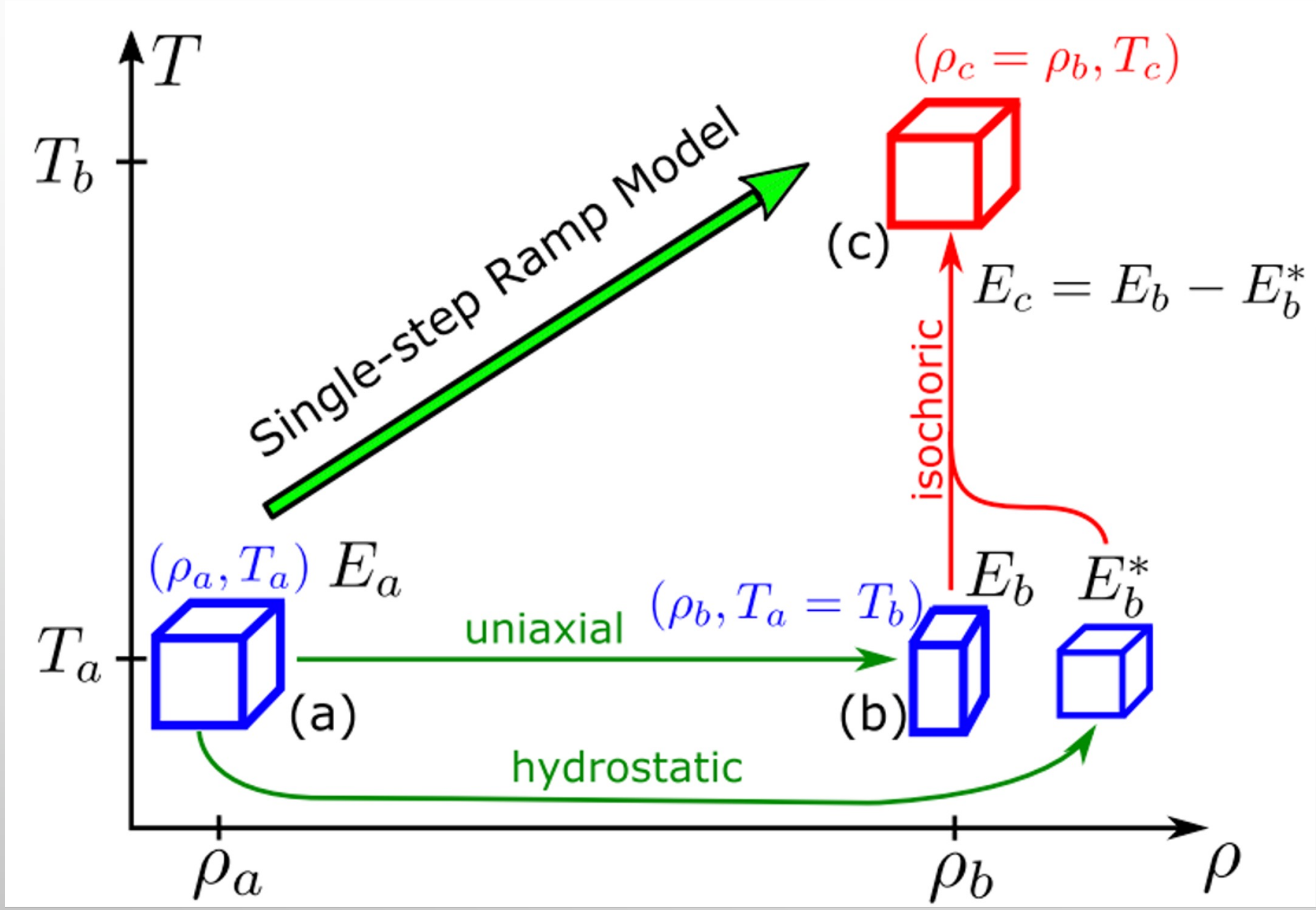


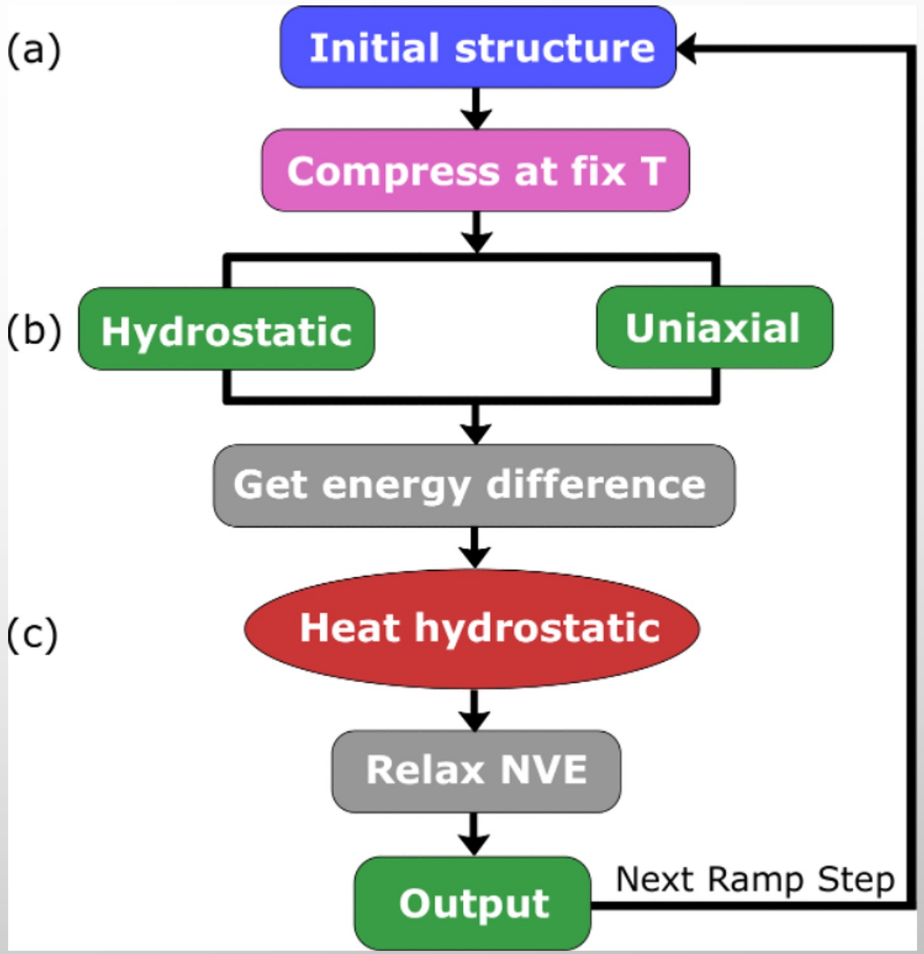
1. Initial state
2. Uniaxial compression
3. Ediff → Kinetic energy

$$\Delta E \to K$$

OUR MODEL OF RAMP COMPRESSION

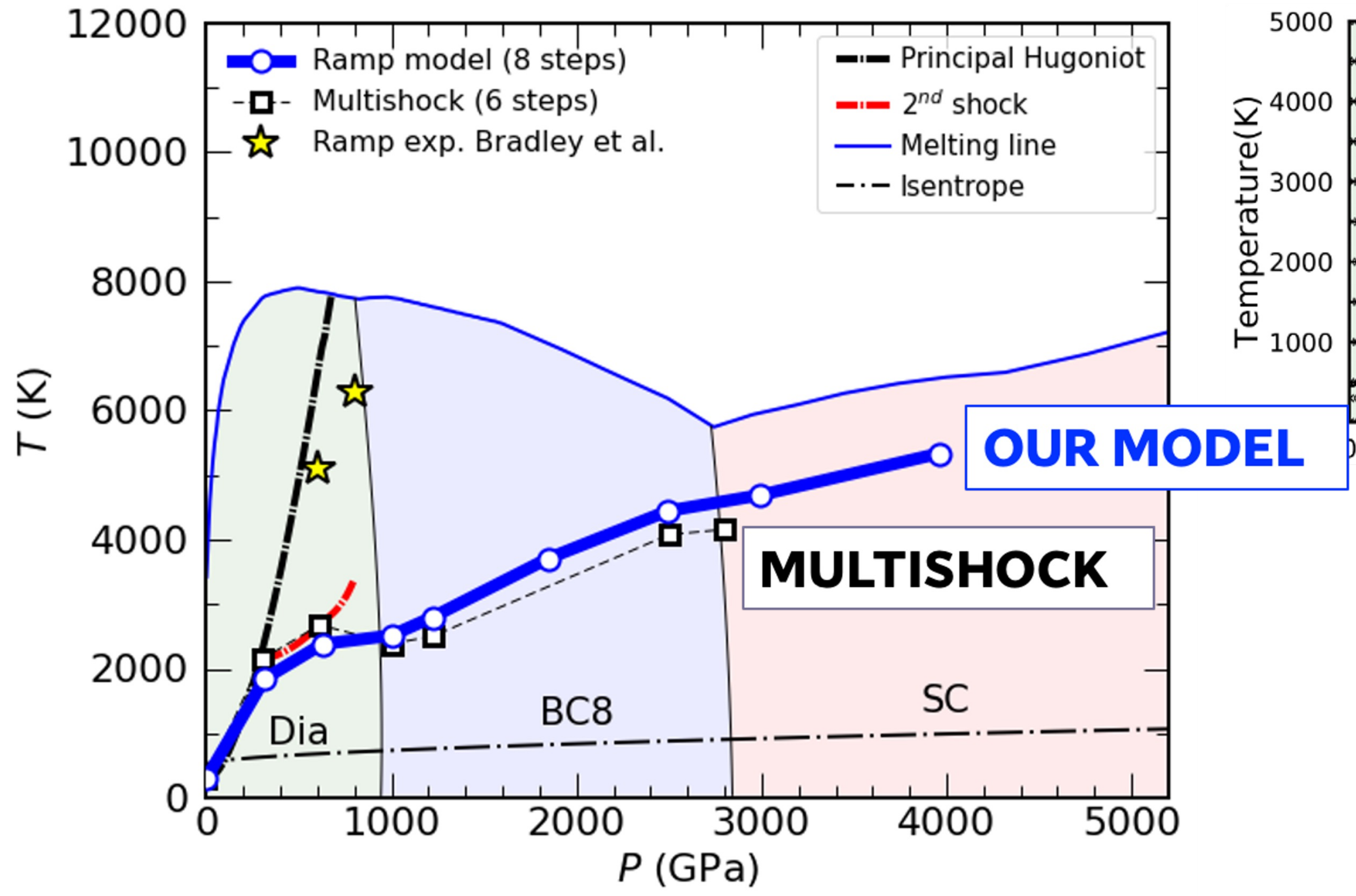
for ramp compression from ab initio simulations

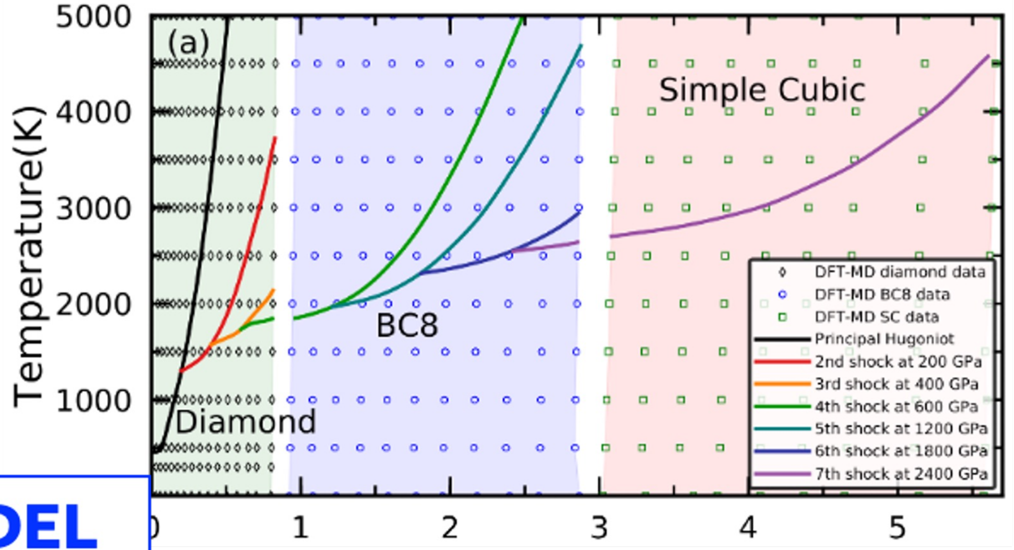


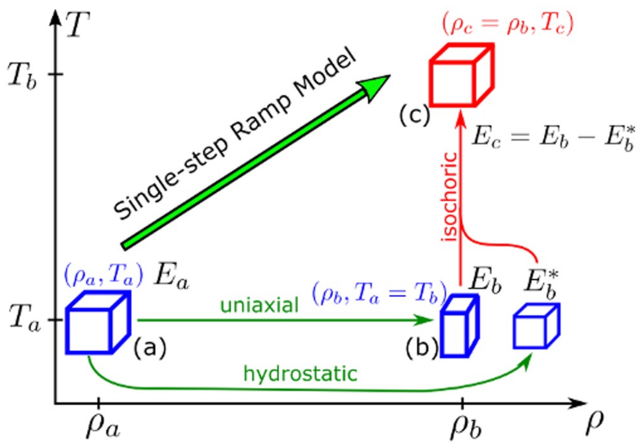


Gonzalez+, PRB (2021)

MULTISHOCKS / OUR MODEL

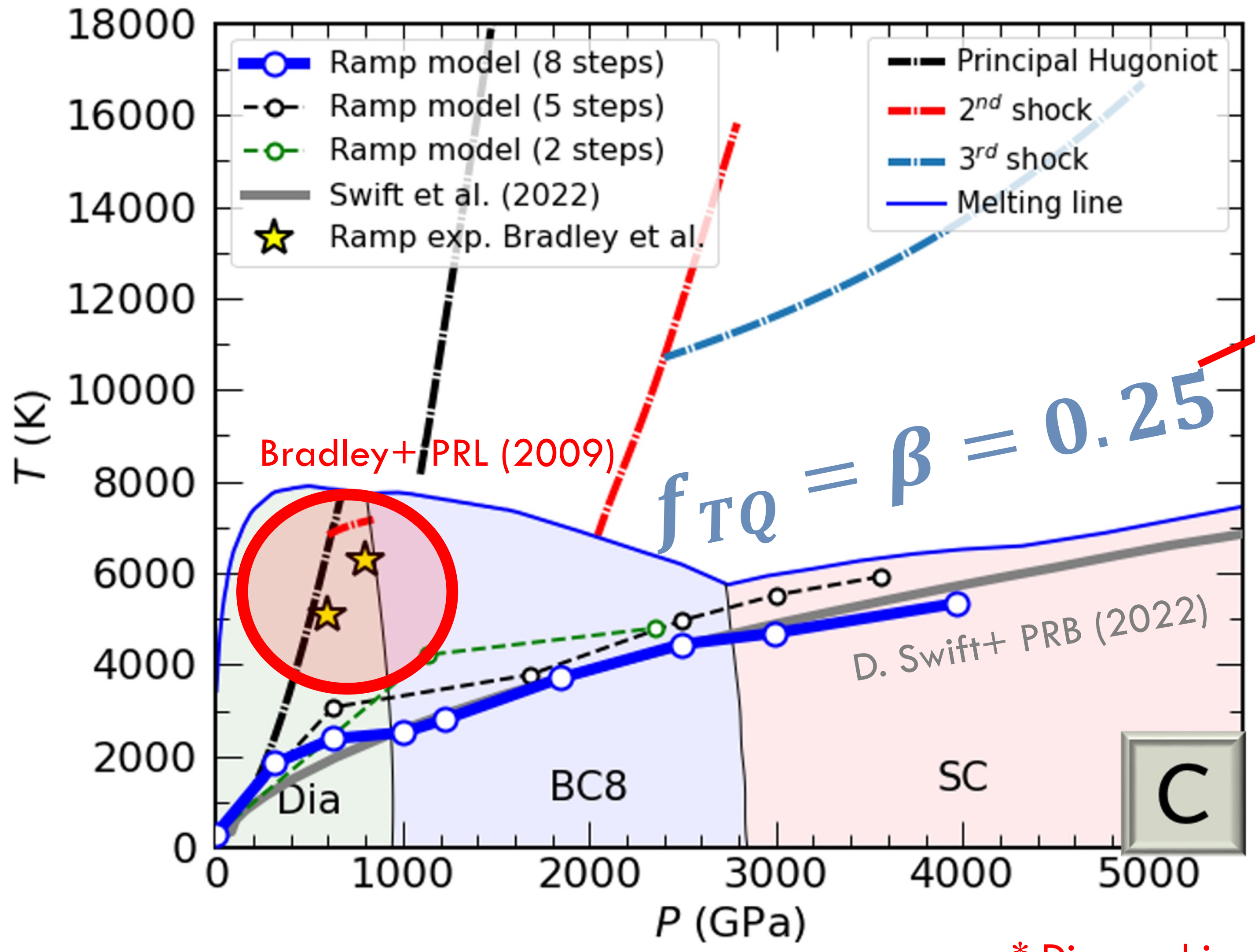




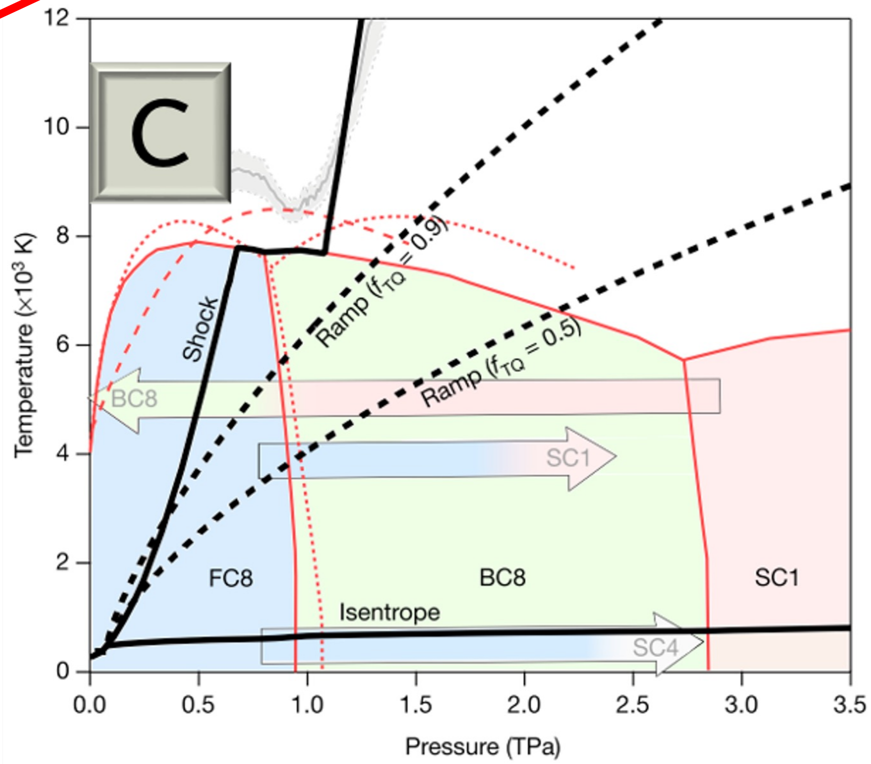


Gonzalez+, PRB (2021)

MULTISHOCKS / OUR MODEL



$$P_{isen} = \sigma_x - \frac{2}{3}Y - \int \beta dW_p - \gamma\rho_H(E_H - E_{isen})$$



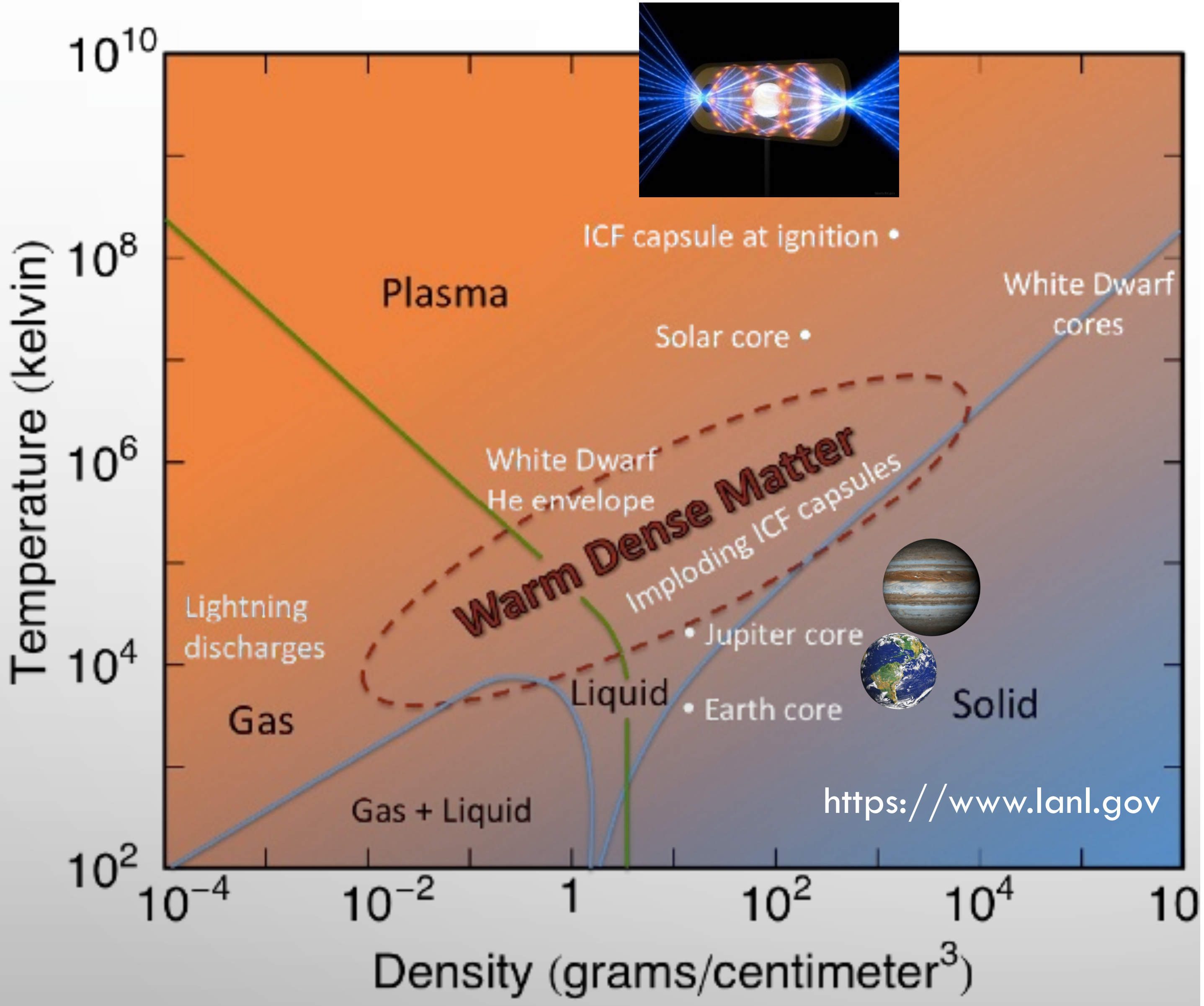
Lazicki+, Nature (2021)

* Diamond is weaker than expected
* 3/4 plastic work absorbed by defects!

OUTLINE

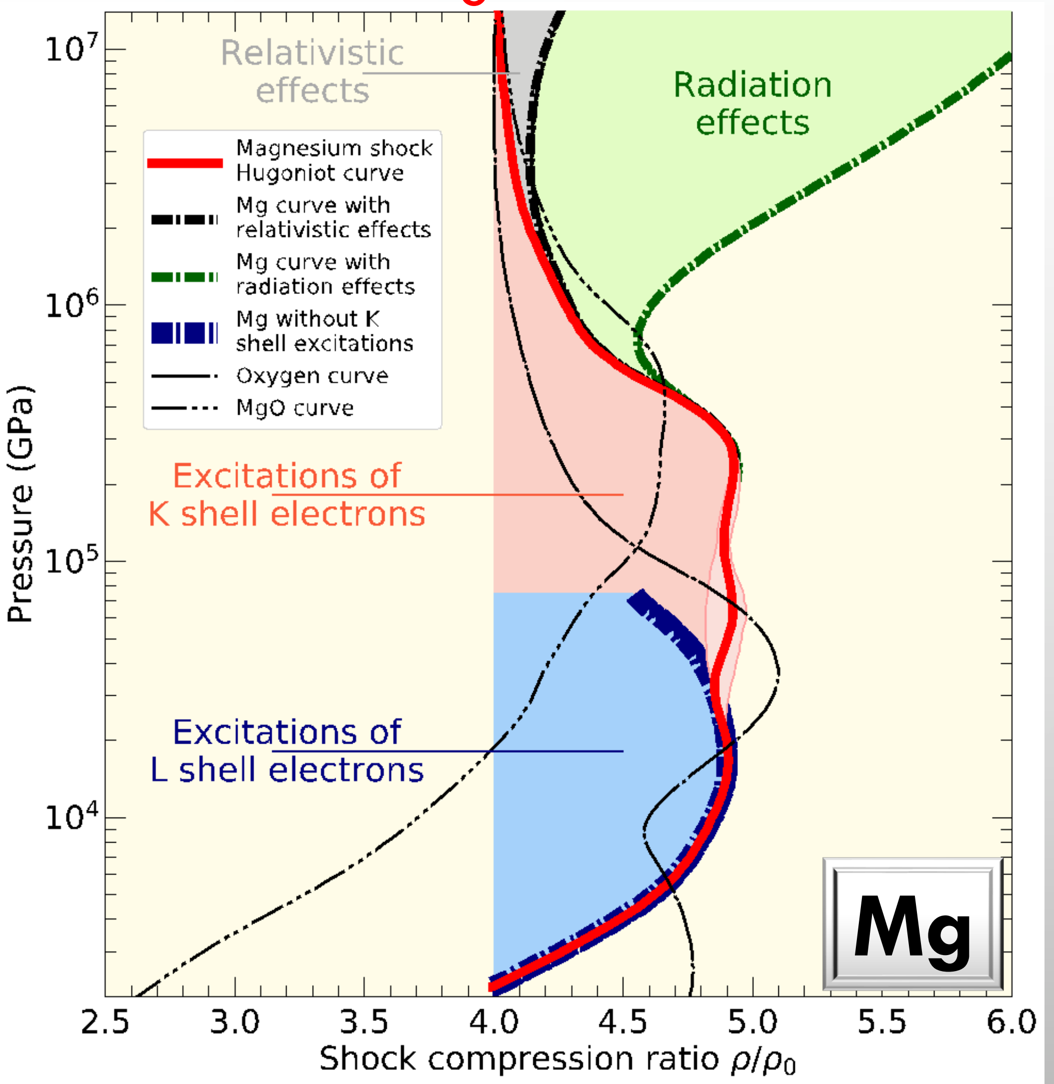
1. **Planetary Interiors**
    - A diluted core in Jupiter
    - Rock/Ice mixtures in water planets
2. **High Pressure Phase Transitions**
    - Be & MgO: melting and anharmonicities
    - Melting of SiO_2
    - Ramp compression from DFT
3. **Warm Dense Matter**
    - Warm dense silicates: Mg, MgO & $MgSiO_3$
    - FPEOS

WARM DENSE MATTER

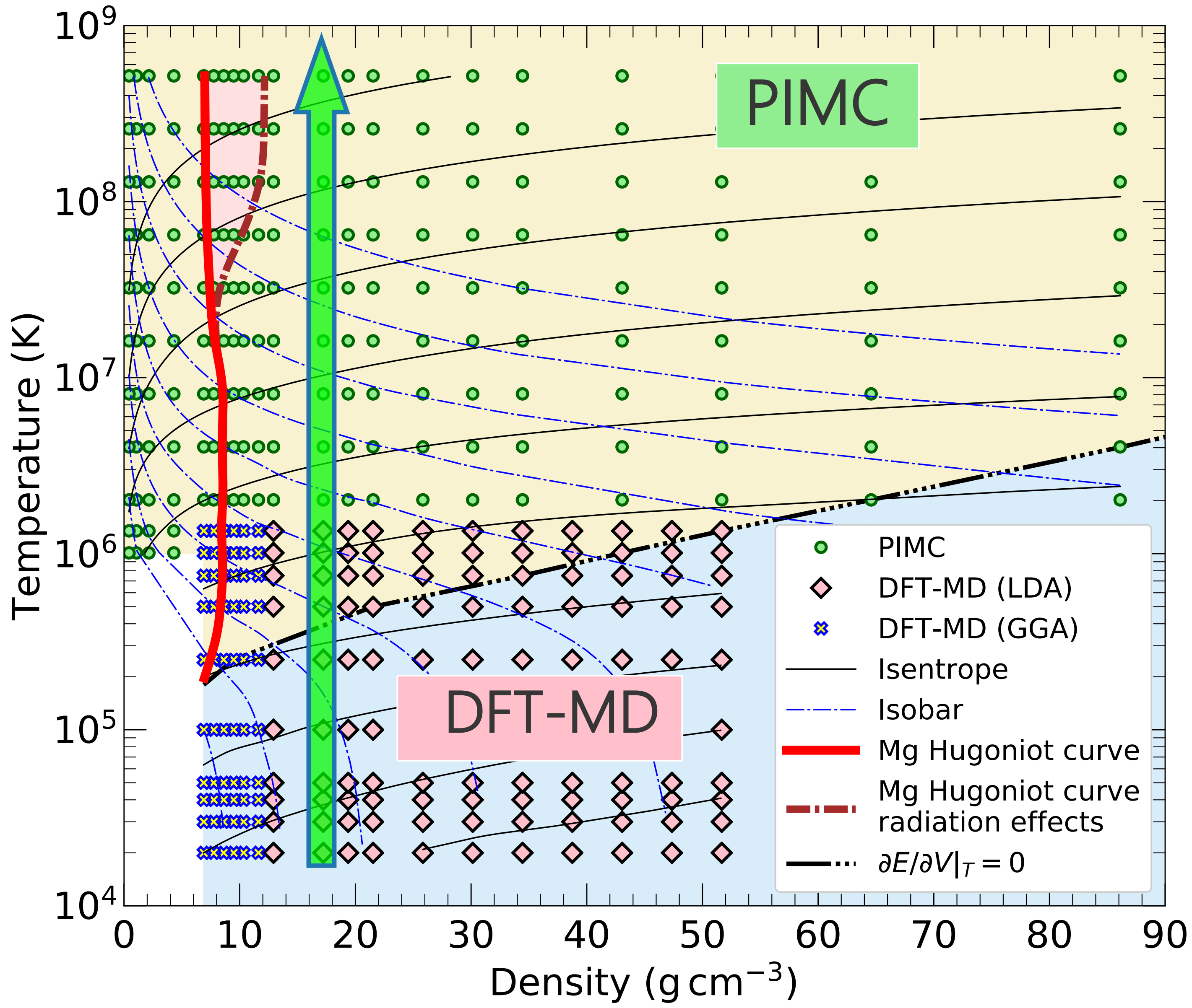


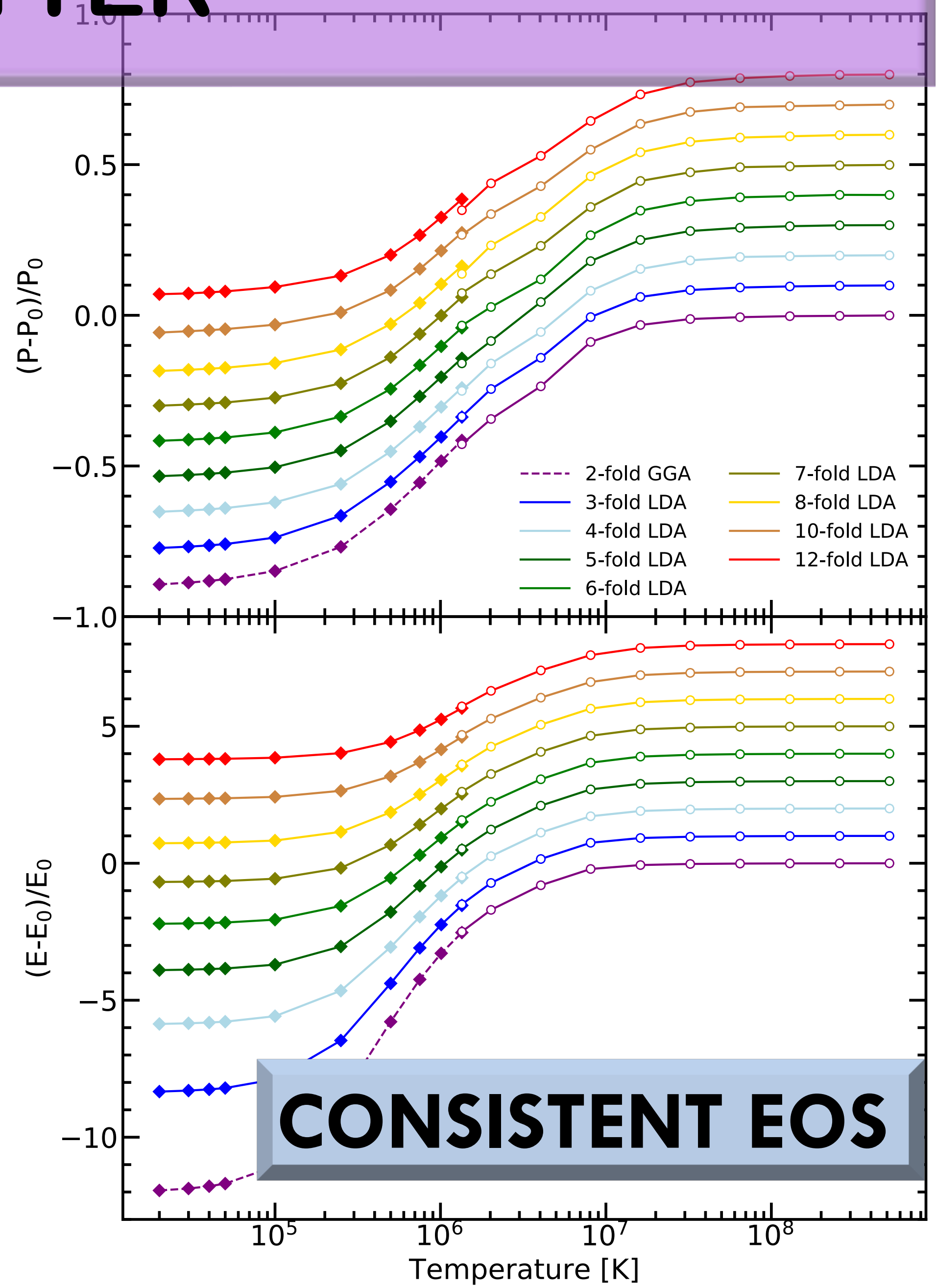
Shock compression:

The Hugoniot curve



WARM DENSE MATTER

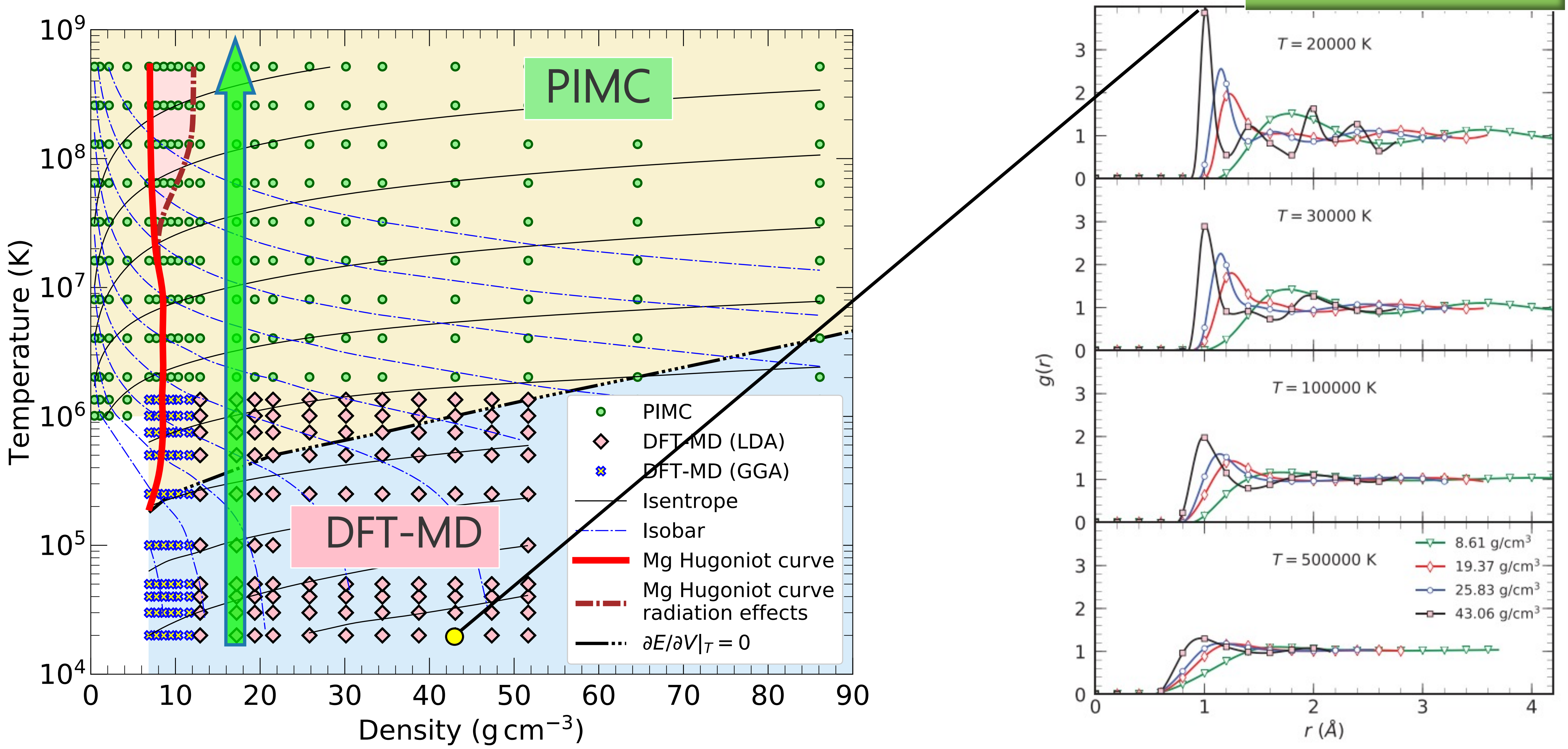




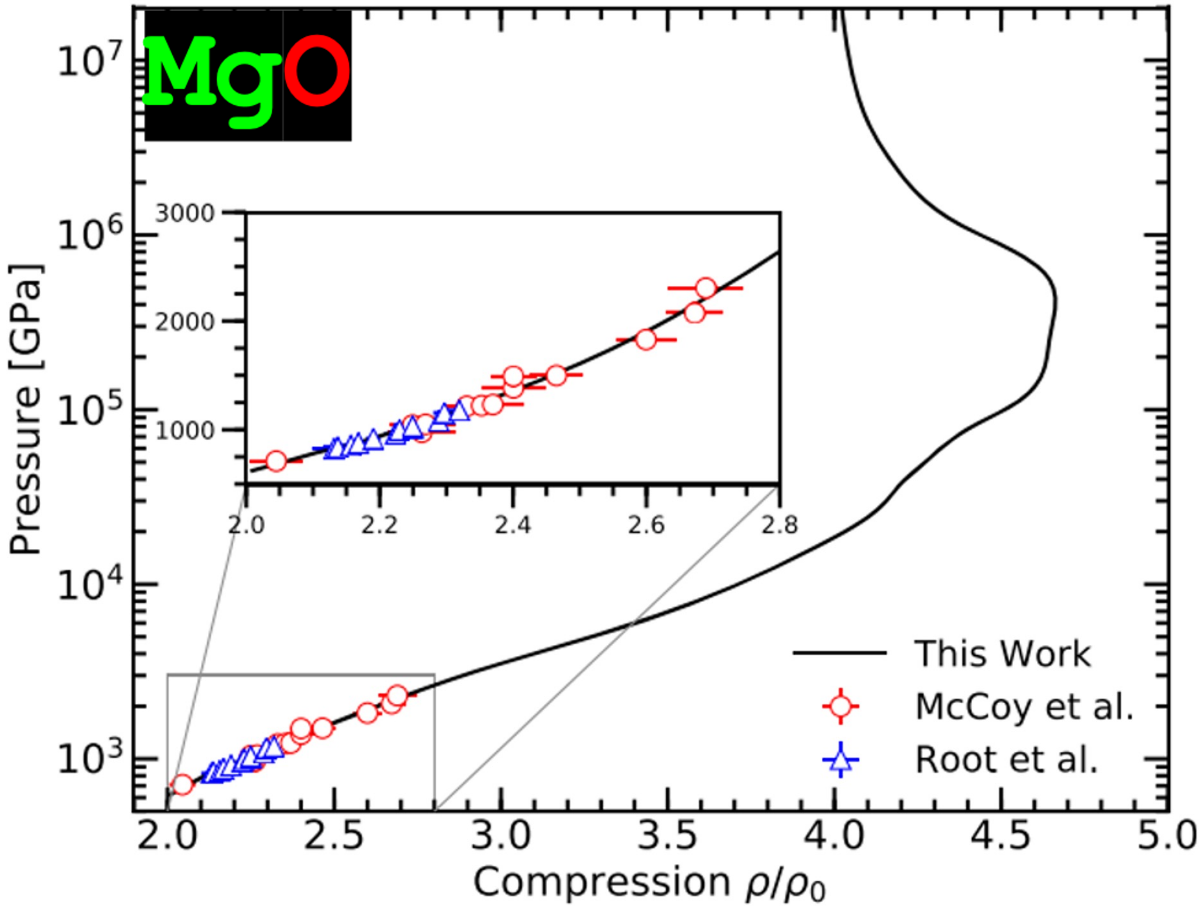
CONSISTENT EOS

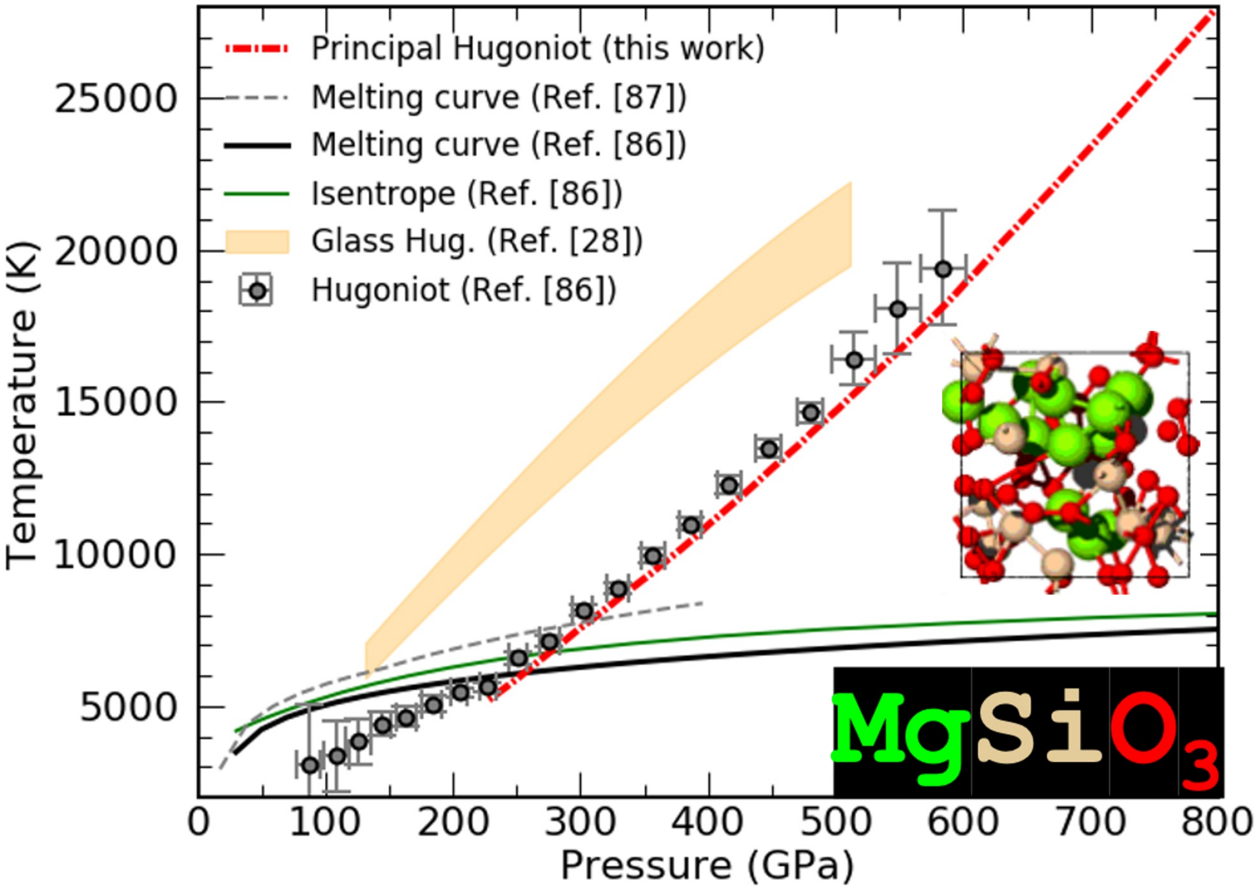
WARM DENSE MATTER

$g(r)$ Structural properties



WARM DENSE MATTER

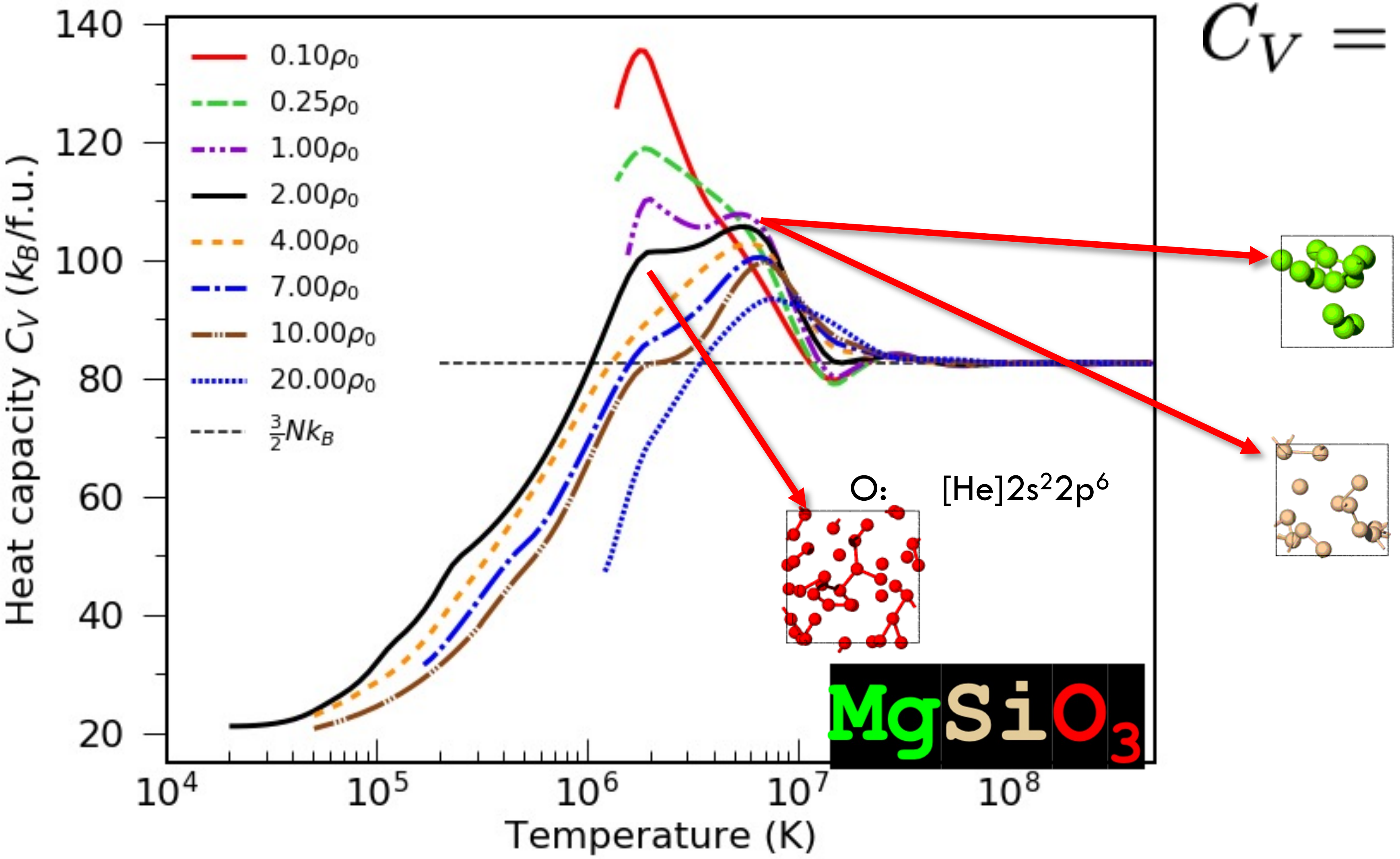




Soubiran+ J. Phys. Chem. (2019)

Gonzalez-Cataldo+ PRB (2020)

WARM DENSE MATTER



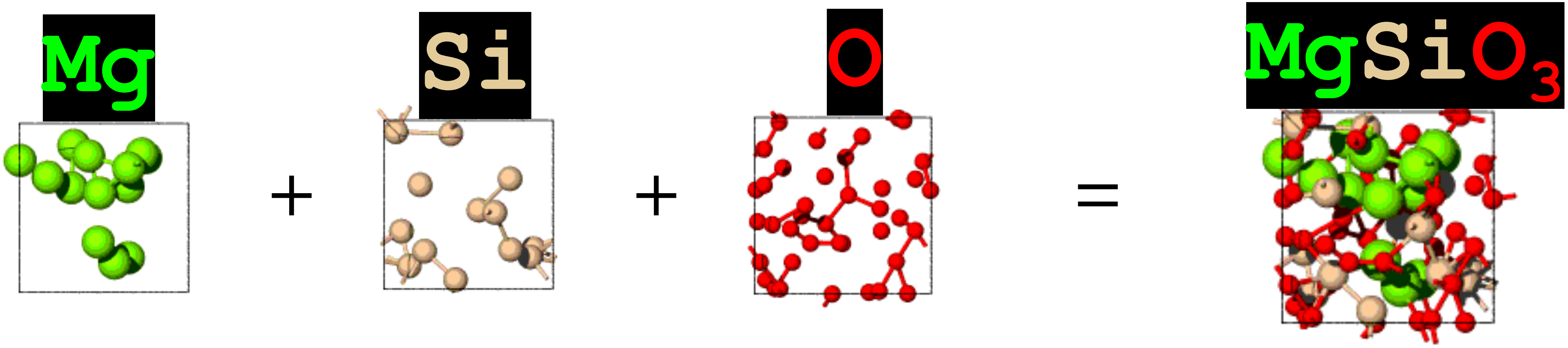
$$C_V = \left(\frac{\partial E}{\partial T}\right)_V$$

Mg: [He]$2s^2 2p^6 3s^2 3p^2$

Si: [He]$2s^2 2p^6 3s^2 3p^2$

WARM DENSE MATTER

Non ideal mixing



Linear Mixing Approximation: (at constant P and T)

$$V_{mix} = N_1 V_1 + N_2 V_2 + N_3 V_3$$

(additive volume rule)

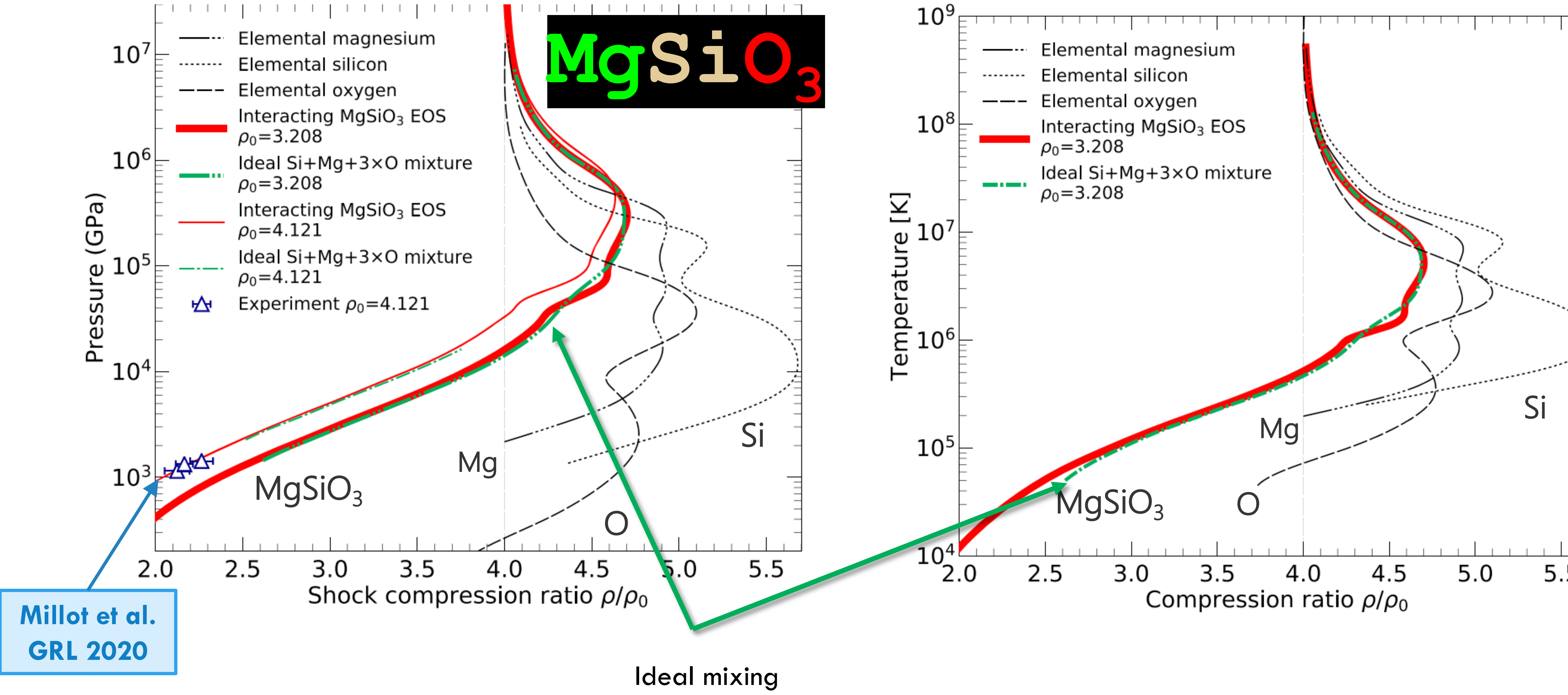
$$m_{mix} = N_1 m_1 + N_2 m_2 + N_3 m_3$$

$$\rho_{mix} = m_{mix} / V_{mix}$$

$$E_{mix} = N_1 E_1 + N_2 E_2 + N_3 E_3$$

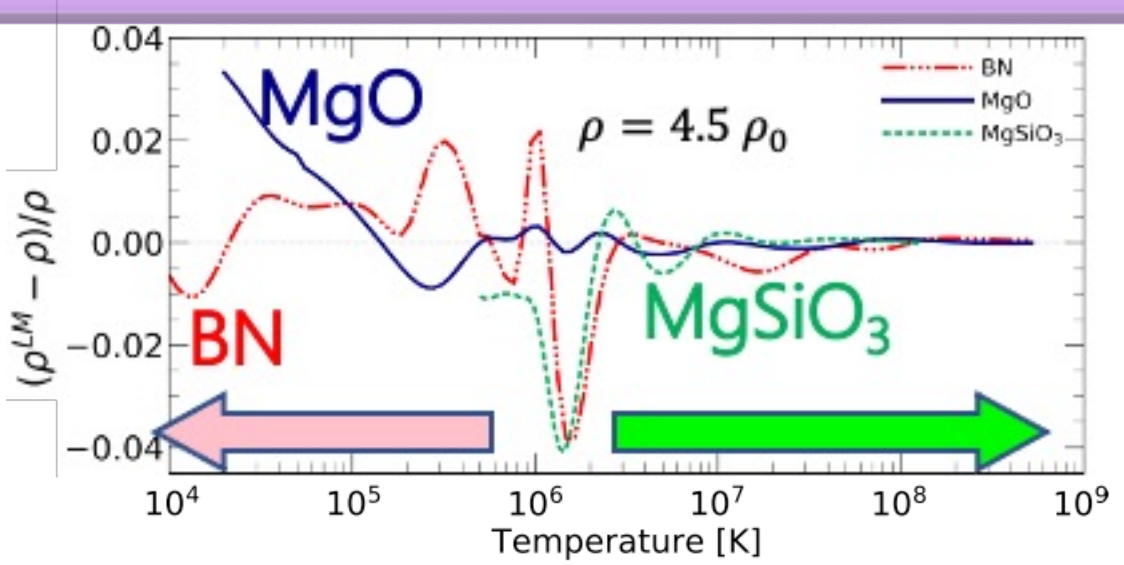
Militzer, Gonzalez, Zhang, Whitley, Swift, Millot, JCP (2020)

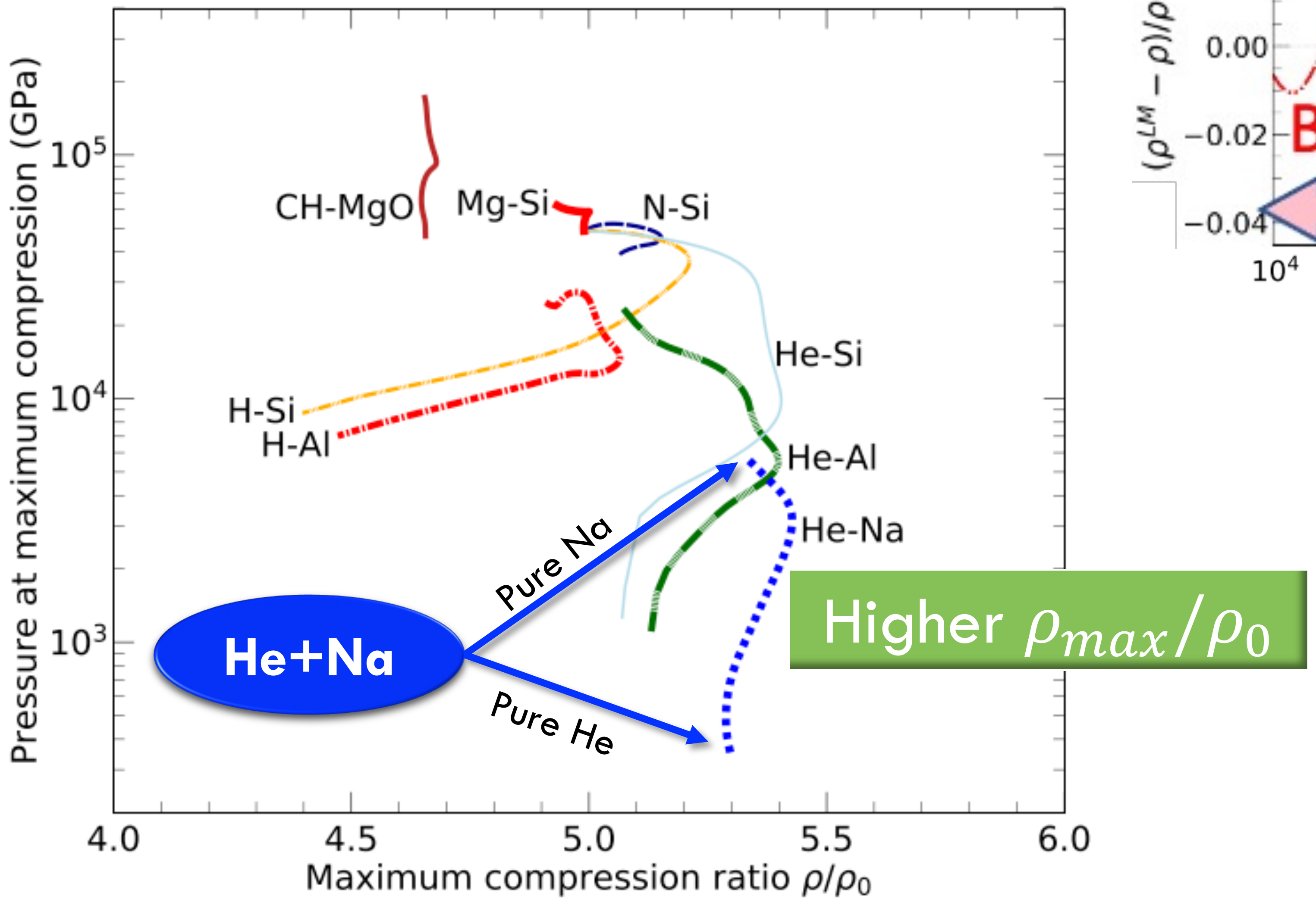
WARM DENSE MATTER

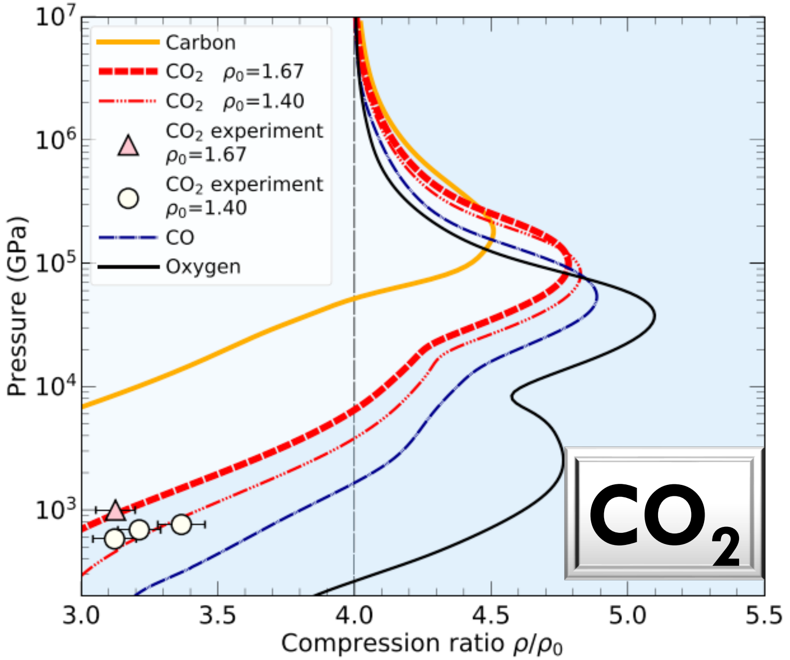


FIRST PRINCIPLES EQUATION OF STATE (FPEOS)

http://militzer.berkeley.edu/FPEOS







CONCLUSIONS

1. Planetary interiors: mixing, erosion, crystallization.
2. Thermodynamic Integration: melting, anharmonicity
3. Z method agrees with two-phase and ΔG.
4. Ramp compression: better models needs
5. Validated Linear Mixing for MgO, $MgSiO_3$, and BN plasmas.
6. http://militzer.berkeley.edu/FPEOS/

Thanks!

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