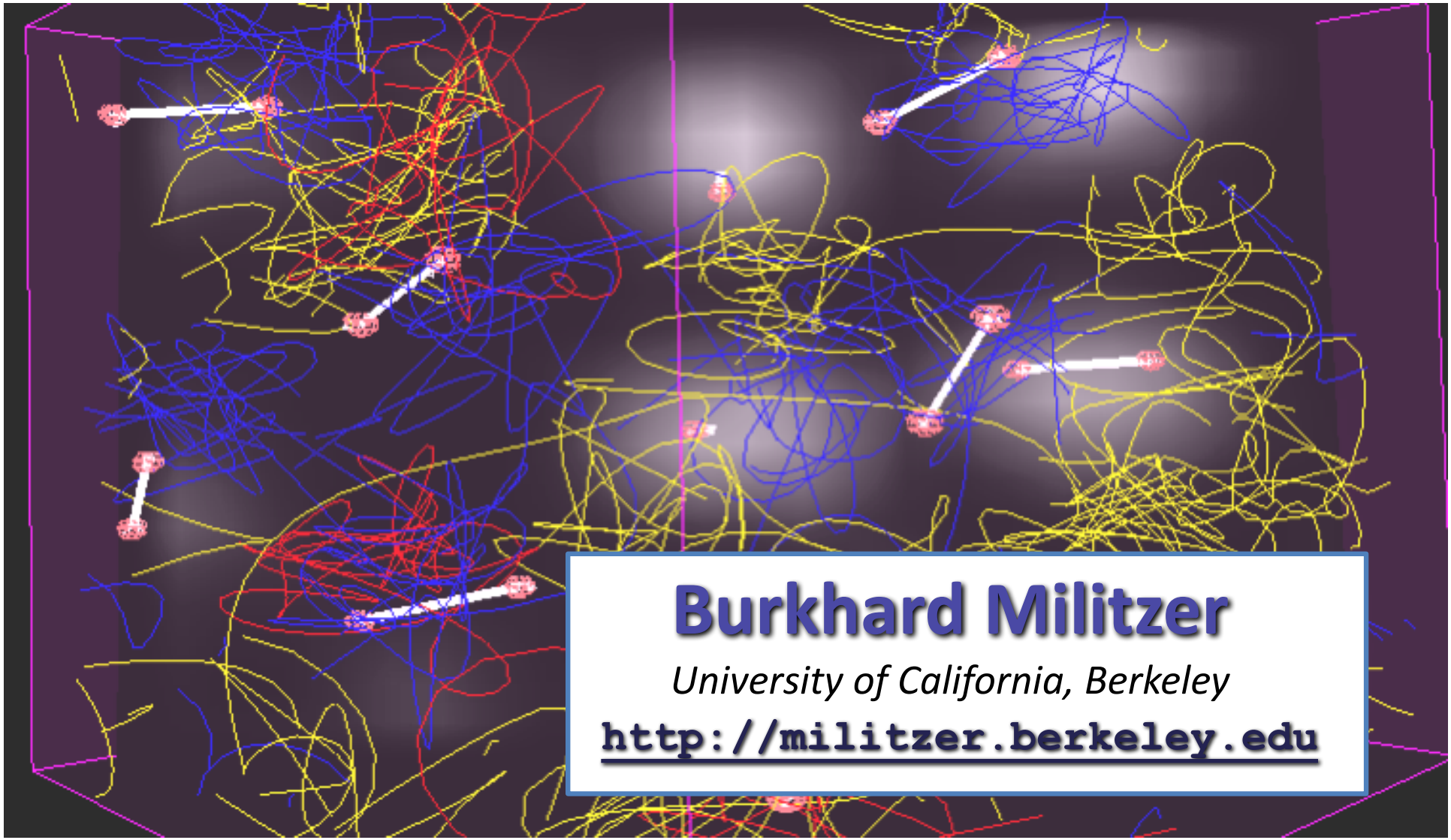
Path Integral Monte Carlo Simulations of Dense Plasmas



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Outline

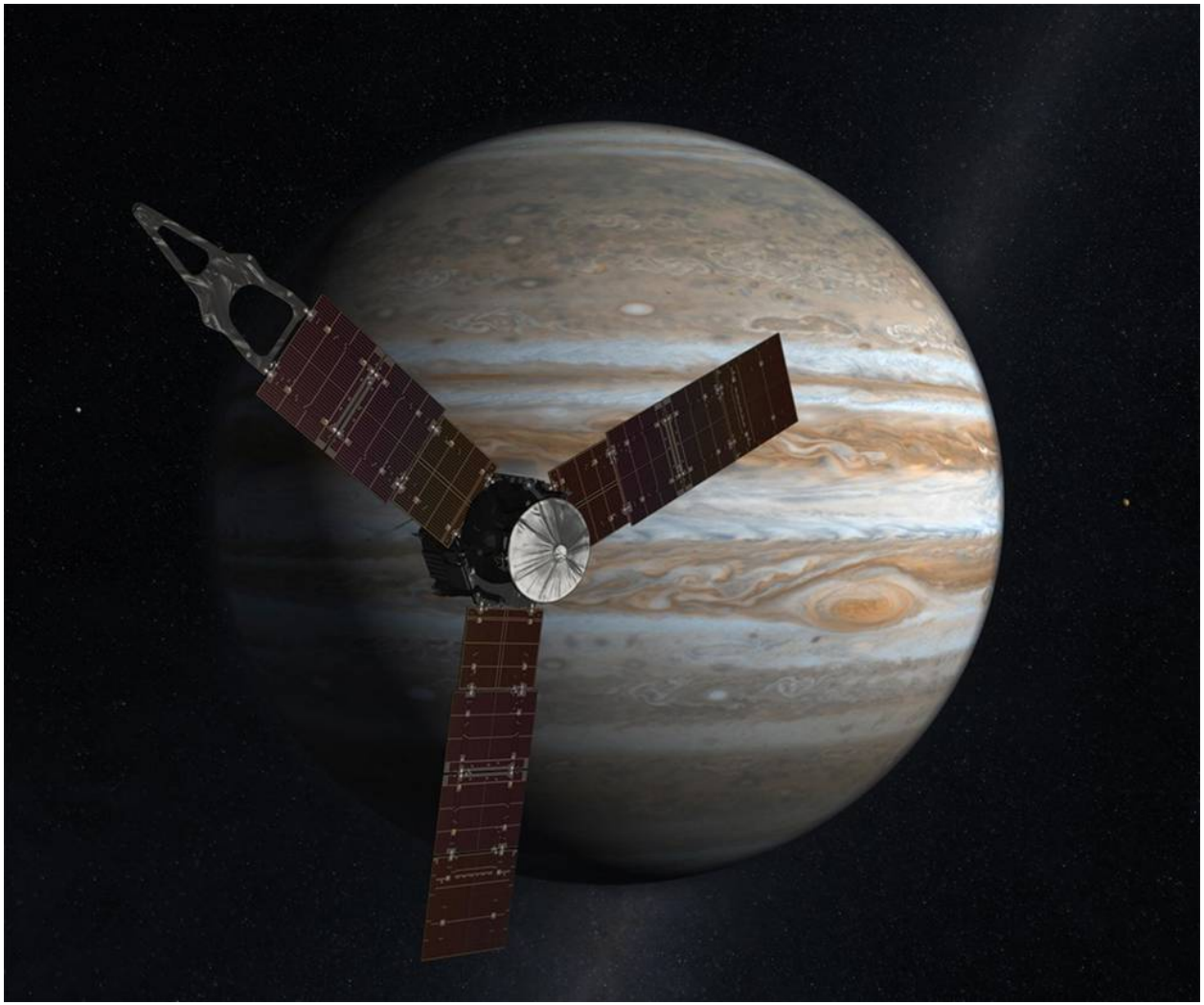
1. Juno mission in orbit around Jupiter
2. Transport properties of solid and liquid silicates
3. Path integral Monte Carlo simulation method
4. Application of CH plastic ablator materials
5. Application to warm, dense silicon

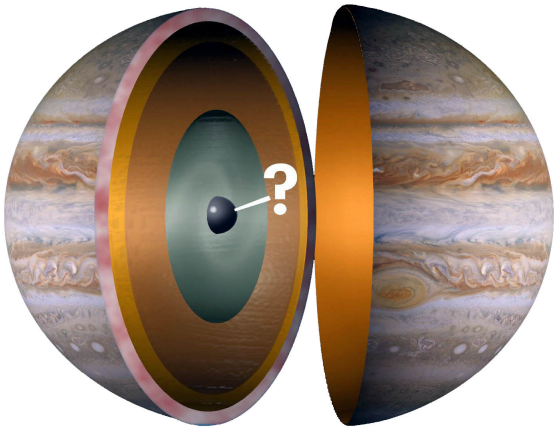
Kevin Driver, Shuai Zhang, Lorin Benedict (LLNL)
Francois Soubiran (UCB), and Suxing Hu (LLE)

Supported by DOE and NSF.

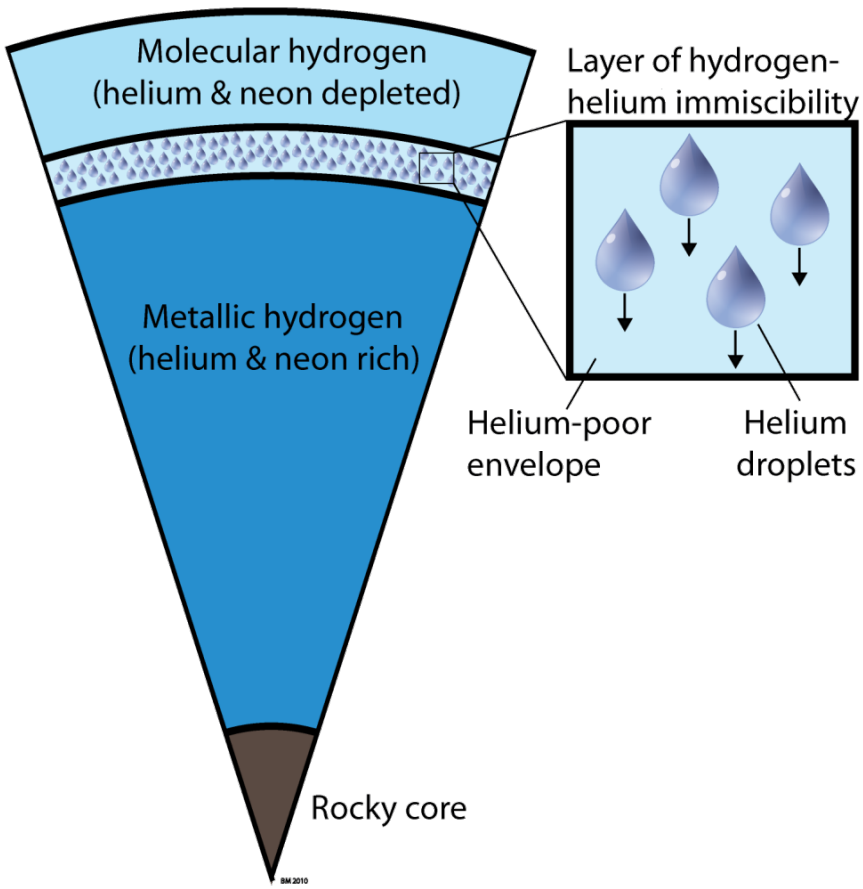
I. Matching Juno Gravity Data with Interior Models

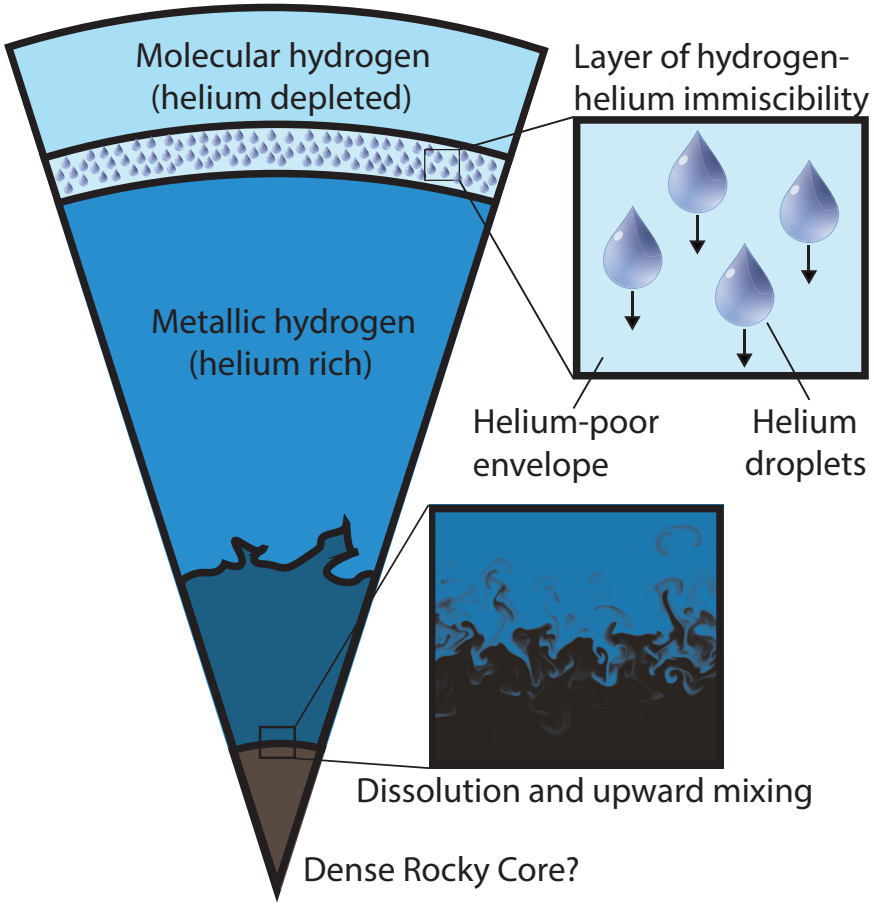
NASA Mission Juno visits Planet Jupiter



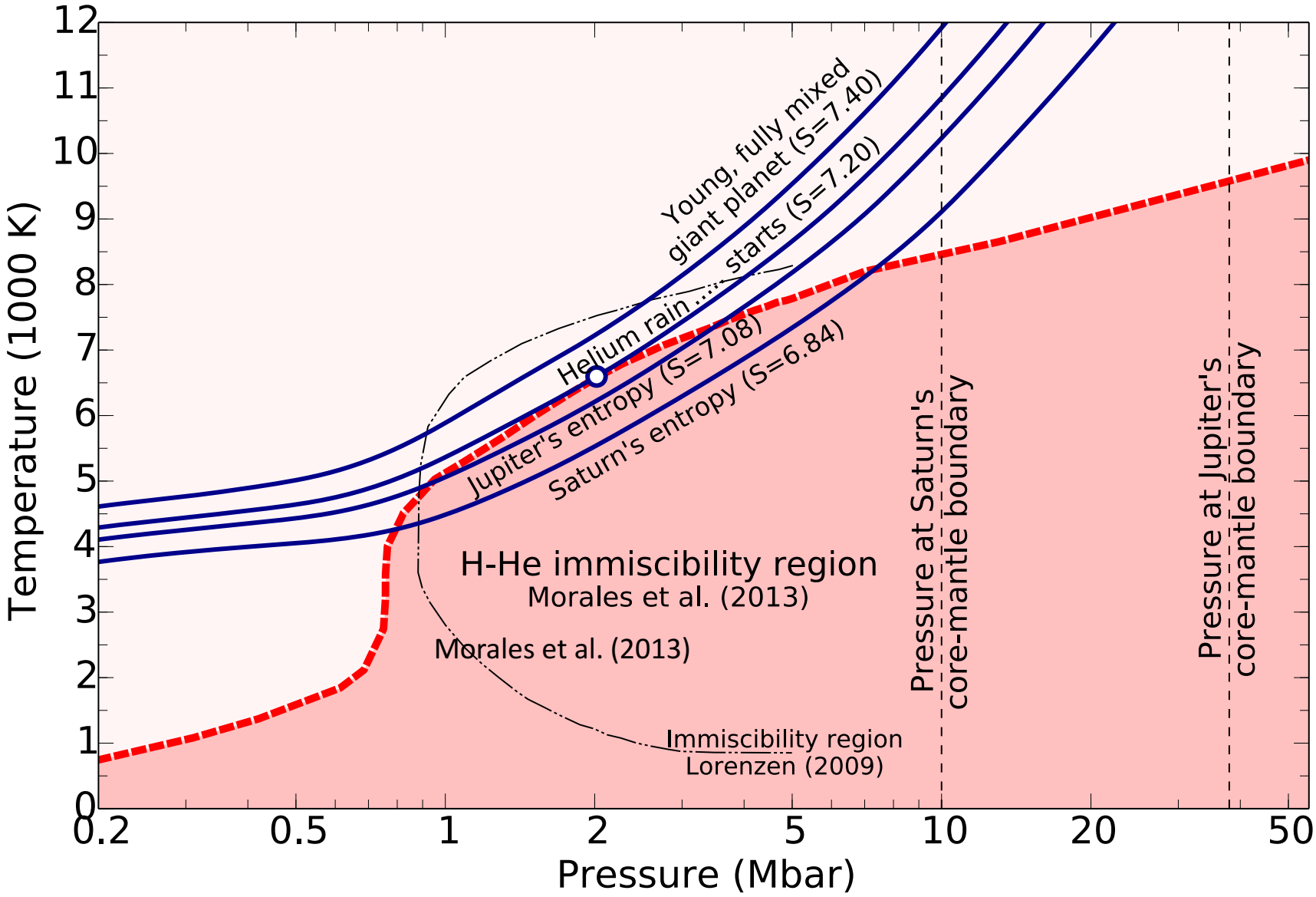


Two possible Jupiter Interior Models with a sharp and a dilute cores





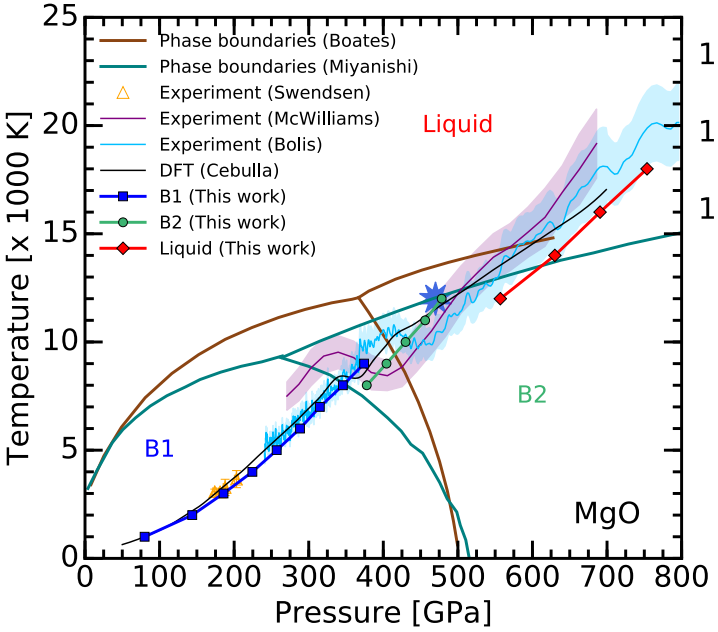
Giant Planet Adiabats and hydrogen-helium immiscibility



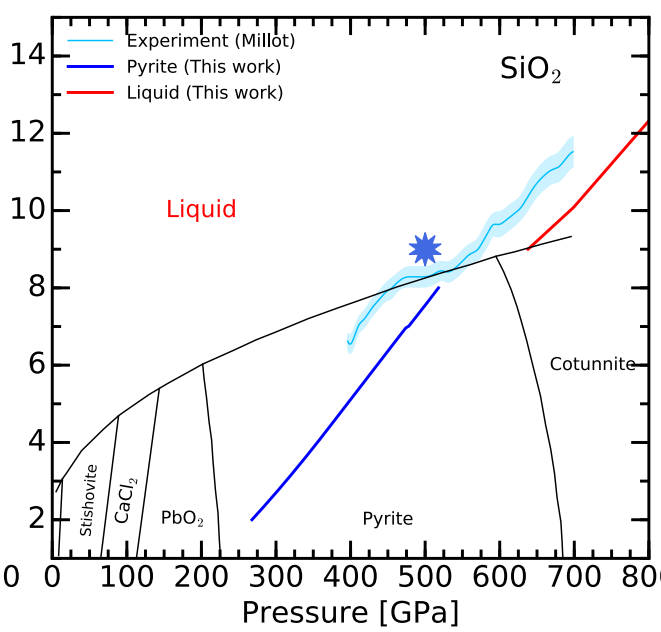
II. Solid and Liquid Silicates

Solid and Liquid Silicates at Super-Earth Interior Conditions

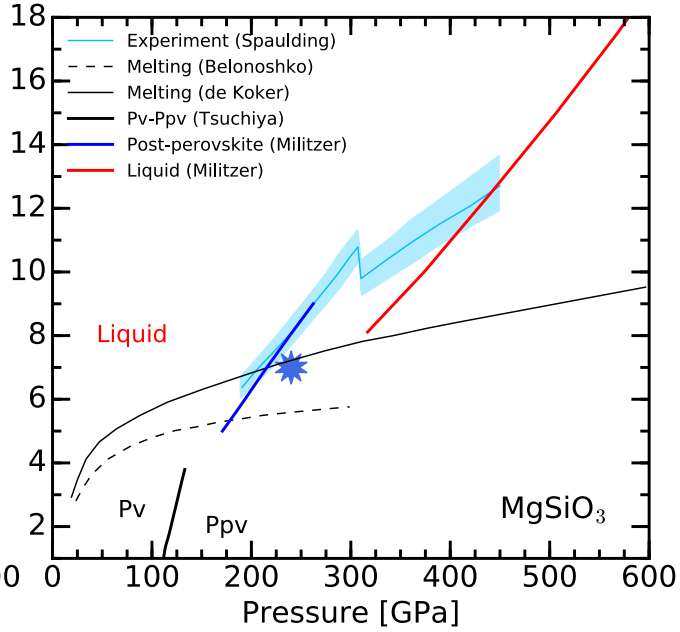
MgO



SiO_2

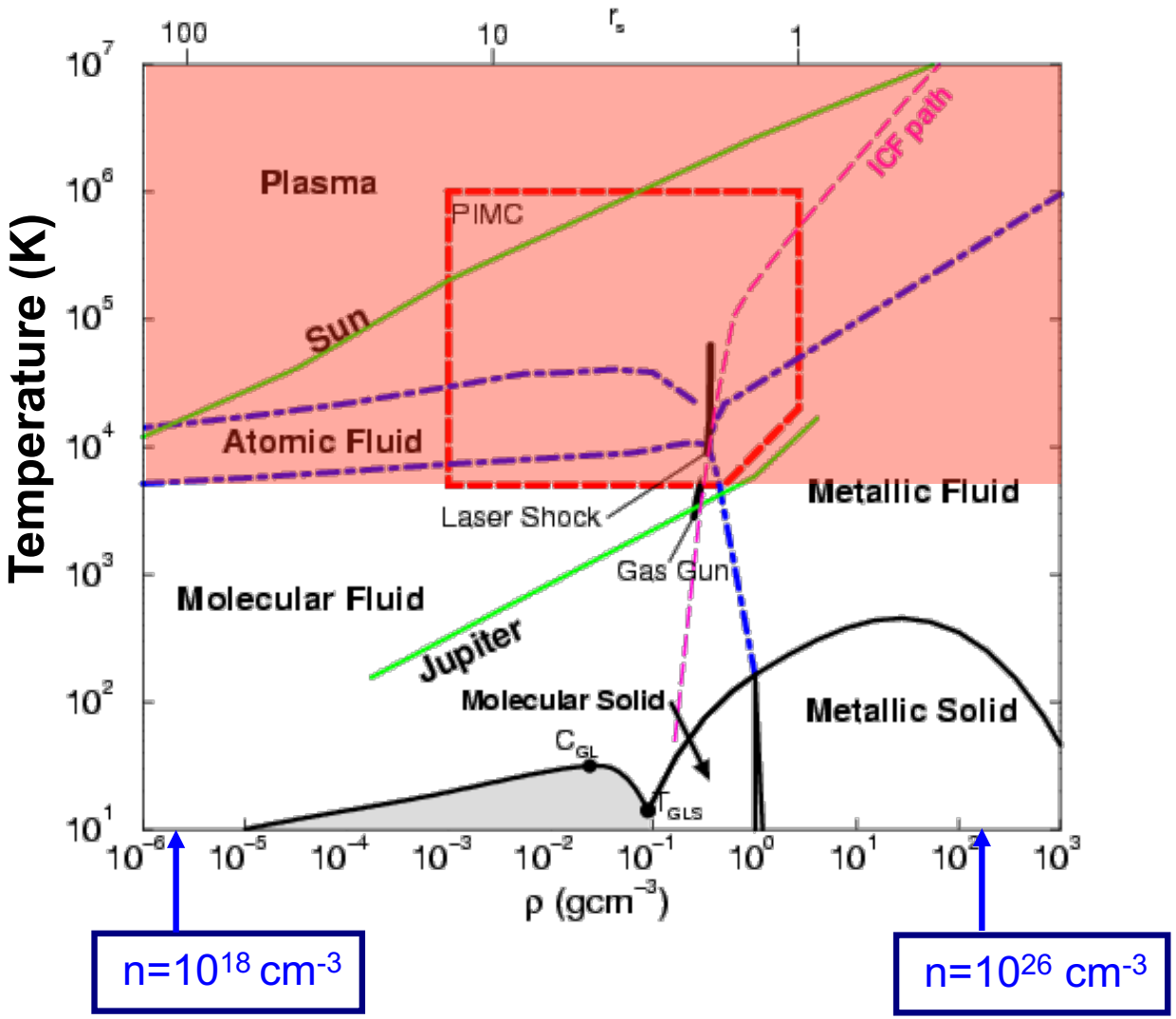


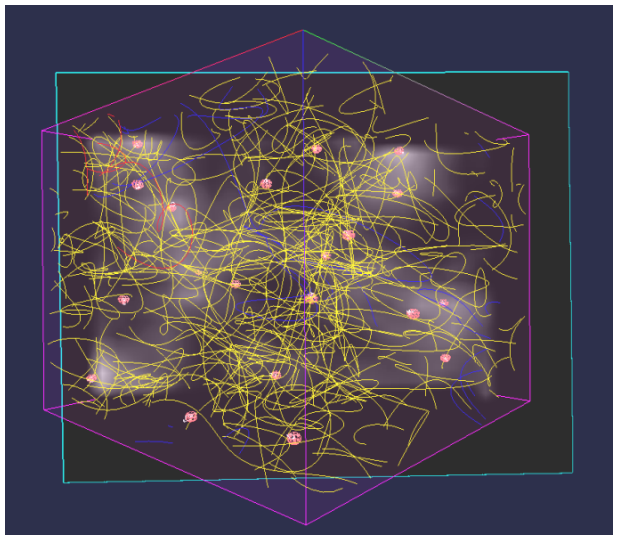
$MgSiO_3$

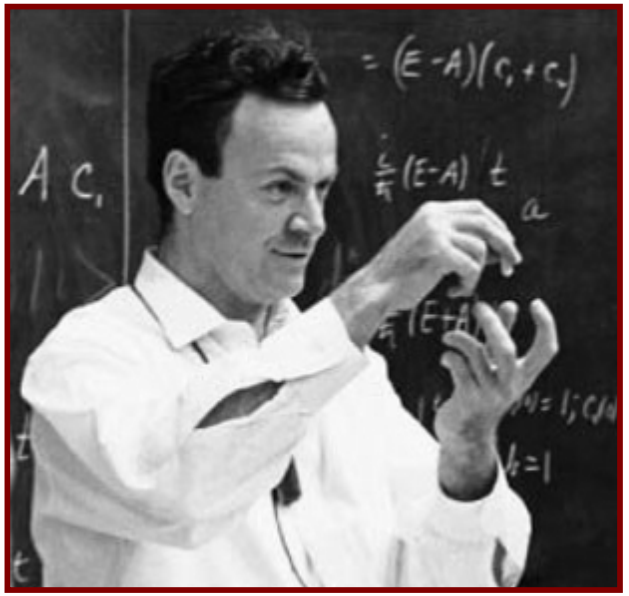


III. Path Integral Monte Carlo

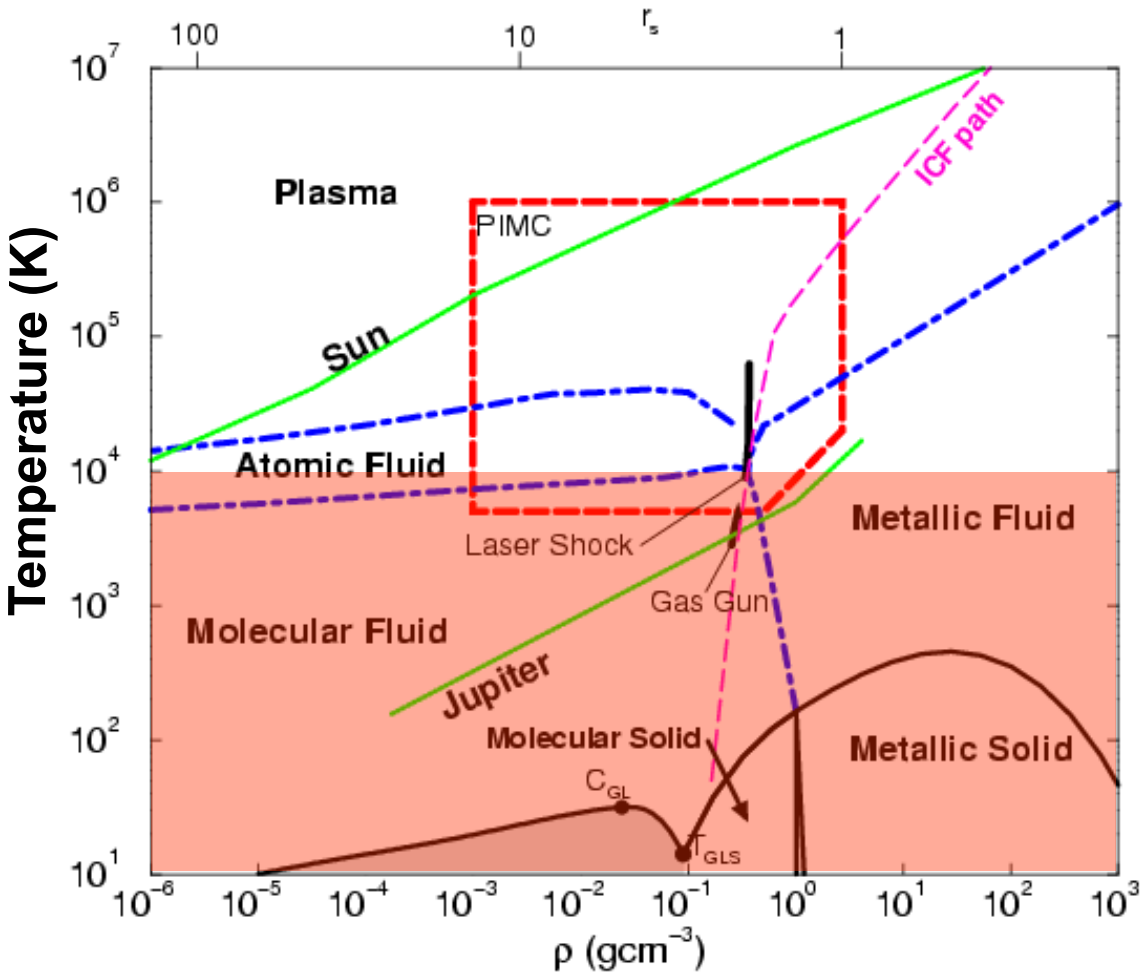
1) Path integral Monte Carlo for T>10000K

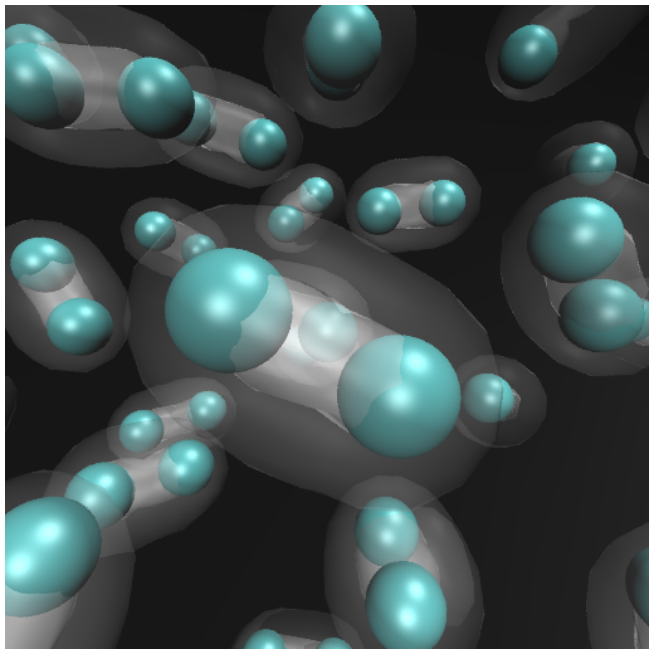






1) Path integral Monte Carlo for T>5000K
2) Density functional molecular dynamics below





Born-Oppenheimer approx.
MD with classical nuclei:

F = m a

Forces derived DFT with electrons in the instantaneous ground state.

Canonical Ensembles:

Classical

Boltzmann factor

$$e^{-E \,/\, k_B T}$$

Quantum

Density matrix

$$\hat{\rho} = e^{-\beta\, \hat{\mathrm{H}}}$$

$$\rho(R,R',\beta) = \left\langle R \middle| e^{-\beta\, \hat{\mathrm{H}}} \middle| R' \right\rangle$$

$$\rho(R,R',\beta) = \sum_S e^{-\beta\, E_S}\ \Psi_S^*(R)\ \Psi_S(R')$$

Thermodynamic averages:

$$Z_{Cl} = \sum_S e^{-\beta\, E_S}$$

$$Z_Q = Tr[\hat{\rho}] = \int dR\ \left\langle R \middle| e^{-\beta\, \hat{\mathrm{H}}} \middle| R \right\rangle$$

$$\left\langle \hat{O} \right\rangle = \frac{Tr[\hat{O}\ \hat{\rho}]}{Tr[\hat{\rho}]}$$

Step 1 towards the path integral

Matrix squaring property of the density matrix

Matrix squaring in operator notation:

$$\hat{\rho} = e^{-\beta\hat{\mathrm{H}}} = \left(e^{-(\beta/2)\hat{\mathrm{H}}}\right)\left(e^{-(\beta/2)\hat{\mathrm{H}}}\right), \quad \beta = \frac{1}{k_B T}$$

Matrix squaring in real-space notation:

$$\langle R \,|\, \hat{\rho} \,|\, R' \rangle = \int dR_1 \, \langle R \,|\, e^{-(\beta/2)\hat{\mathrm{H}}} \,|\, R_1 \rangle \, \langle R_1 \,|\, e^{-(\beta/2)\hat{\mathrm{H}}} \,|\, R' \rangle$$

Matrix squaring in matrix notation:

$$\begin{bmatrix} \cdots & R' & \cdots \\ R & \ddots & \vdots \\ \cdots & \cdots & \cdots \end{bmatrix} = \begin{bmatrix} \cdots & R_1 & \cdots \\ R & \ddots & \vdots \\ \cdots & \cdots & \cdots \end{bmatrix} * \begin{bmatrix} \cdots & R' & \cdots \\ R_1 & \ddots & \vdots \\ \cdots & \cdots & \cdots \end{bmatrix}$$

Repeat the matrix squaring step

Matrix squaring in operator notation:

$$\hat{\rho} = e^{-\beta \hat{\mathrm{H}}} = \left(e^{-(\beta/4)\hat{\mathrm{H}}} \right)^4, \quad \beta = \frac{1}{k_B T}$$

Matrix squaring in real-space notation:

$$\langle R | \hat{\rho} | R' \rangle = \int dR_1 \int dR_2 \int dR_3 \langle R | e^{-(\beta/4)\hat{\mathrm{H}}} | R_1 \rangle \langle R_1 | e^{-(\beta/4)\hat{\mathrm{H}}} | R_2 \rangle \langle R_2 | e^{-(\beta/4)\hat{\mathrm{H}}} | R_3 \rangle \langle R_3 | e^{-(\beta/4)\hat{\mathrm{H}}} | R' \rangle$$

Path Integrals in Imaginary Time

Simplest form for the paths' action: primitive approx.

Density matrix:

$$\hat{\rho} = e^{-\beta \hat{\mathrm{H}}} = \left(e^{-\tau \hat{\mathrm{H}}}\right)^M, \quad \beta = \frac{1}{k_B T}, \quad \tau = \frac{\beta}{M}$$

$$\langle \hat{O} \rangle = \frac{\mathrm{Tr}[\hat{O}\hat{\rho}]}{\mathrm{Tr}[\hat{\rho}]}$$

Trotter break-up:

$$\langle R | \hat{\rho} | R' \rangle = \langle R | (e^{-\tau \hat{\mathrm{H}}})^M | R' \rangle = \int dR_1 \ldots \int dR_{M-1} \langle R | e^{-\tau \hat{\mathrm{H}}} | R_1 \rangle \langle R_1 | e^{-\tau \hat{\mathrm{H}}} | R_2 \rangle \ldots \langle R_{M-1} | e^{-\tau \hat{\mathrm{H}}} | R' \rangle$$

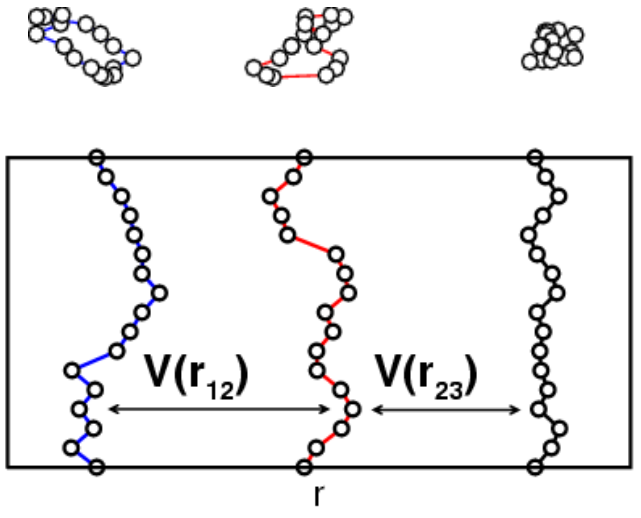
Trotter formula:

$$e^{-\beta(\hat{T}+\hat{V})} = \lim_{M \to \infty} \left[e^{-\tau \hat{T}} e^{-\tau \hat{V}} \right]^M$$

Path integral and primitive action S :

$$\langle R | \hat{\rho} | R' \rangle = \oint_{R \to R'} dR_t e^{-S[R_t]}$$

$$S[R_t] = \sum_{i=1}^{M} \frac{(R_{i+1} - R_i)^2}{4\lambda\tau} + \frac{\tau}{2}\left[V(R_i) + V(R_{i+1})\right]$$



Pair action: Militzer, Comp. Phys. Comm. (2016)

Bosonic and Fermionic Path Integrals

Bosonic density matrix:
Sum over all symmetric eigenstates.

$$\rho_B(R,R',\beta) = \sum_i e^{-\beta E_i} \Psi_S^{[i]*}(R) \Psi_S^{[i]}(R')$$

Fermionic density matrix:
Sum over all antisymmetric eigenstates.

$$\rho_F(R,R',\beta) = \sum_i e^{-\beta E_i} \Psi_{AS}^{[i]*}(R) \Psi_{AS}^{[i]}(R')$$

Project out the symmetric states:

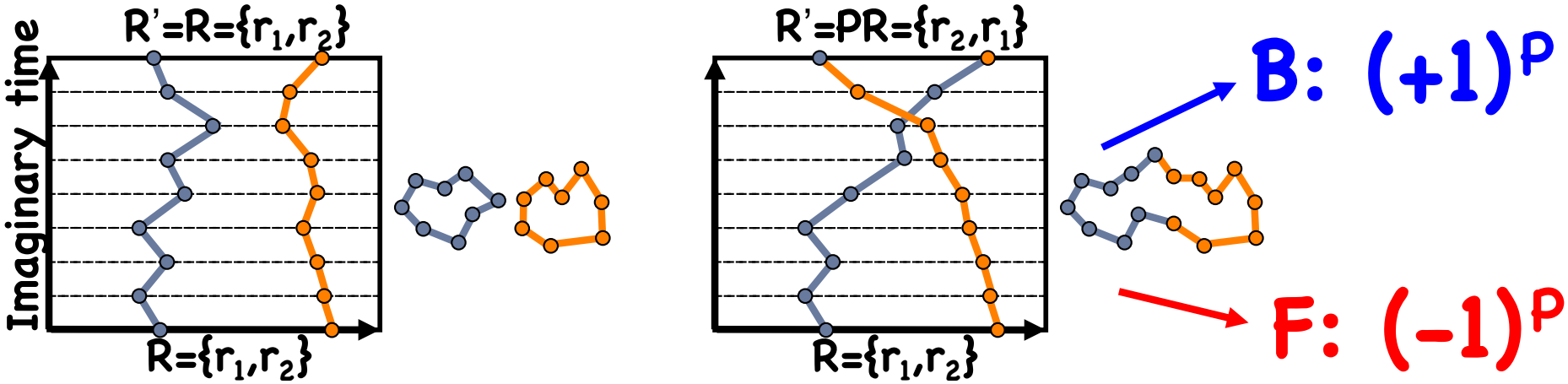
$$\rho_B(R,R',\beta) = \sum_P (+1)^P \rho_D(R,PR',\beta)$$

Project out the antisymmetric states:

$$\rho_F(R,R',\beta) = \sum_P (-1)^P \rho_D(R,PR',\beta)$$

$$\langle R | \hat{\rho}_{F/B} | R' \rangle = \sum_P (\pm 1)^P \int dR_1 \ldots \int dR_{M-1} \langle R | e^{-\tau \hat{H}} | R_1 \rangle \ldots \langle R_{M-1} | e^{-\tau \hat{H}} | PR' \rangle$$

D:



Based on Groundbreaking PIMC work started at LLNL

PHYSICAL REVIEW B VOLUME 30, NUMBER 5 1 SEPTEMBER 1984

Simulation of quantum many-body systems by path-integral methods

E. L. Pollock and D. M. Ceperley
Lawrence Livermore National Laboratory, Livermore, California 94550
(Received 2 April 1984)

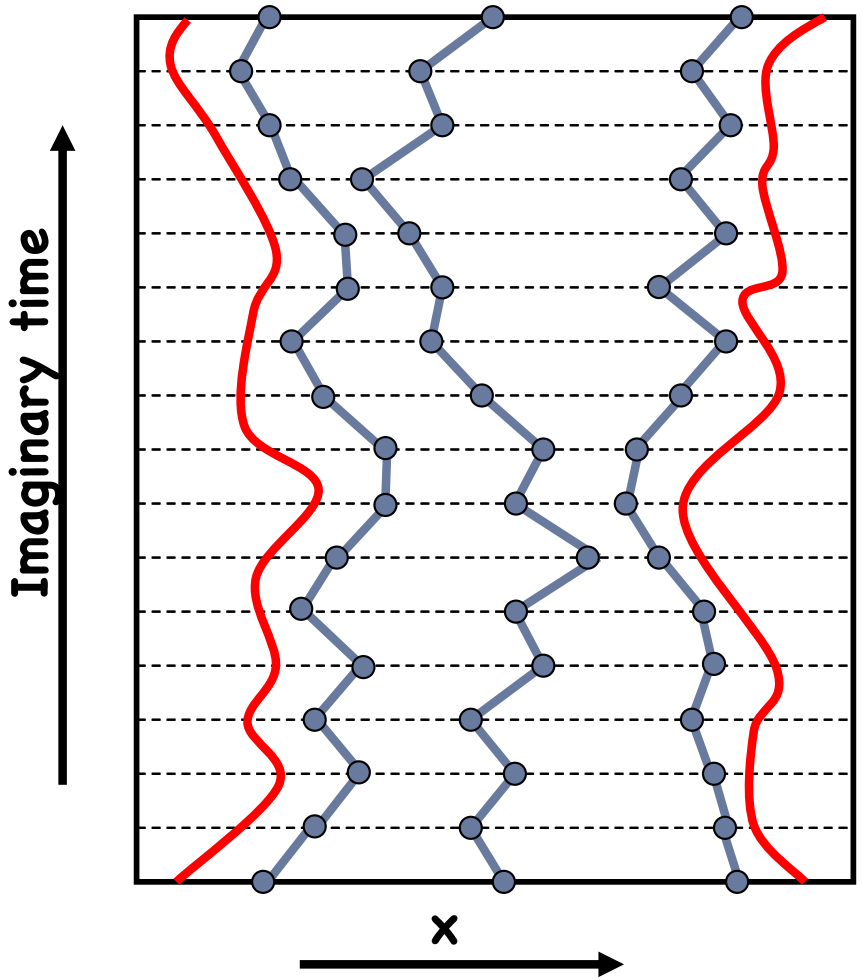
PHYSICAL REVIEW
LETTERS

VOLUME 73 17 OCTOBER 1994 NUMBER 16

Equation of State of the Hydrogen Plasma by Path Integral Monte Carlo Simulation

C. Pierleoni,[1,2,*] D. M. Ceperley,[3] B. Bernu,[1] and W. R. Magro[3]

Restricted PIMC for fermions: How is the restriction applied?



Construct a fermionic trial density matrix in form of a Slater determinant of single-particle density matrices:

$$\rho_T(R,R',\beta) = \begin{vmatrix} \rho(r_1,r_1',\beta) & \cdots & \rho(r_1,r_N',\beta) \\ \vdots & \ddots & \vdots \\ \rho(r_N,r_1',\beta) & \cdots & \rho(r_N,r_N',\beta) \end{vmatrix}$$

Enforce the following nodal condition for all time slices along the paths:

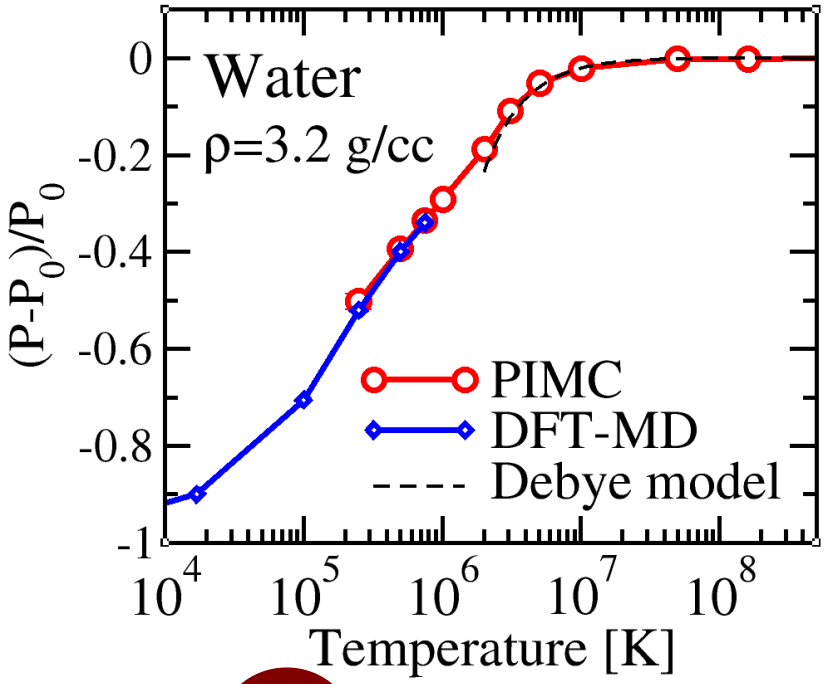
$$\rho_T\left[R(t),R(0),t\right] > 0$$

This 3N-dimensional conditions eliminates all negative and some positive contribution to the path → Solves the fermion sign problem approx.

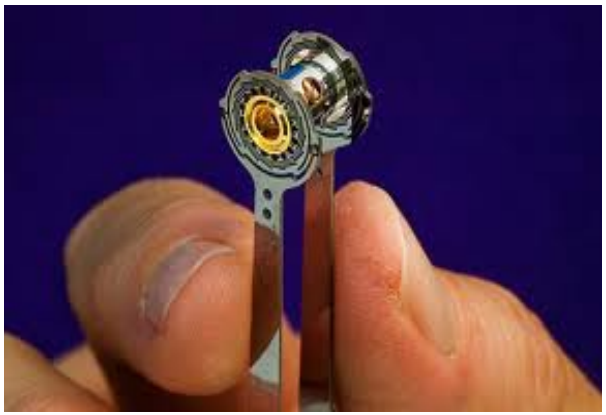
Free-particle nodes:

$$\rho_0^{[1]}(r,r';\beta) = \sum_k e^{-\beta E_k}\,\Psi_k(r)\,\Psi_k^*(r')$$

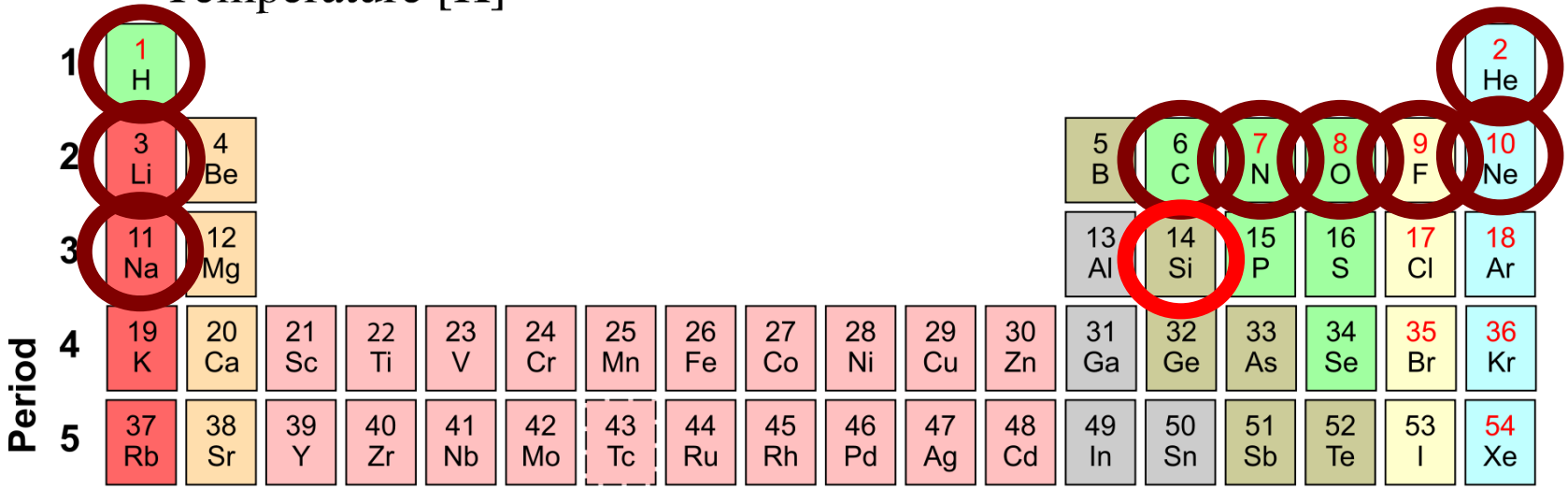
New Path Integral Monte Carlo Simulations of Heavier Elements Aid Fusion Capsule Design



ICF Hohlraum

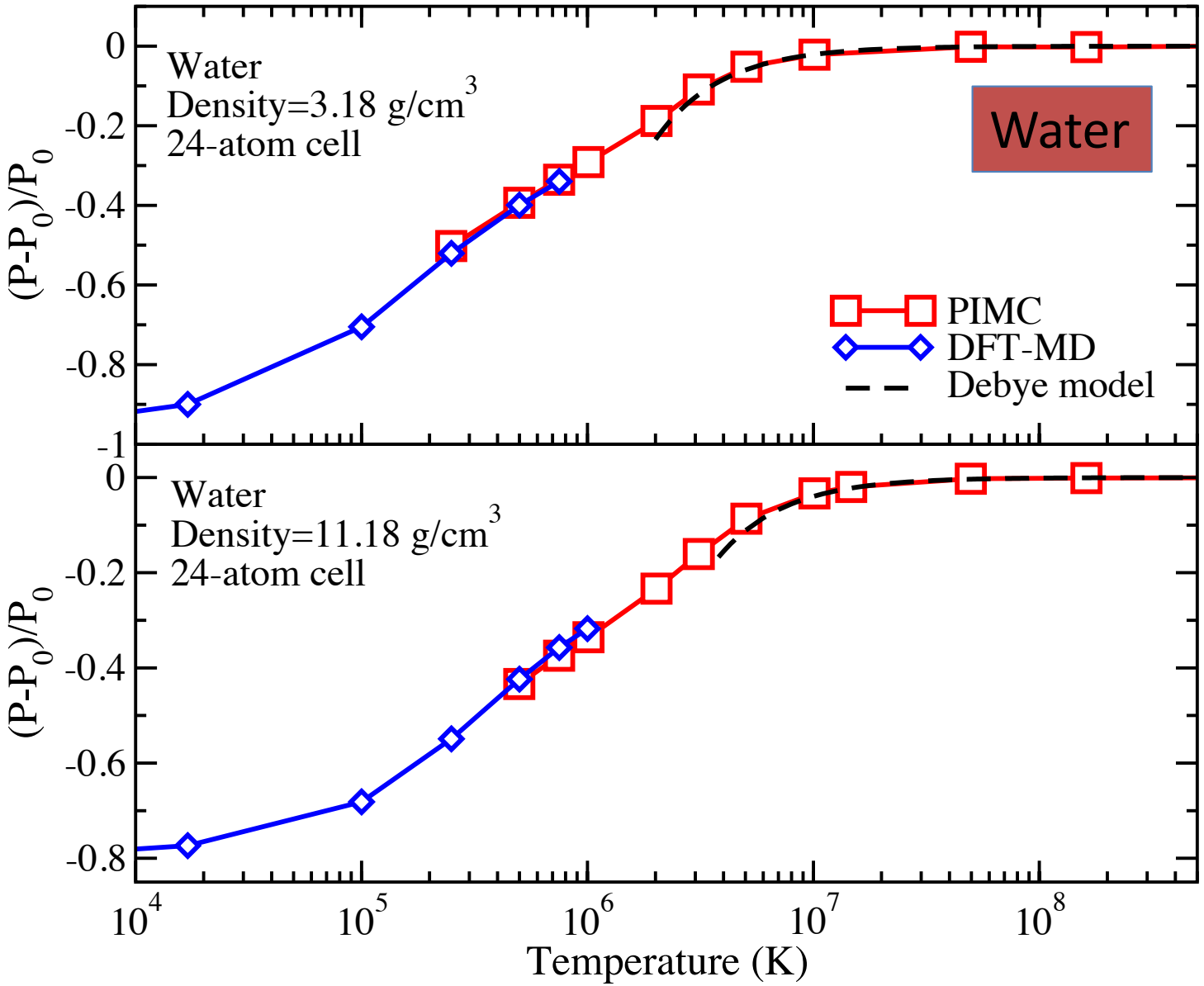


ICF Capsule

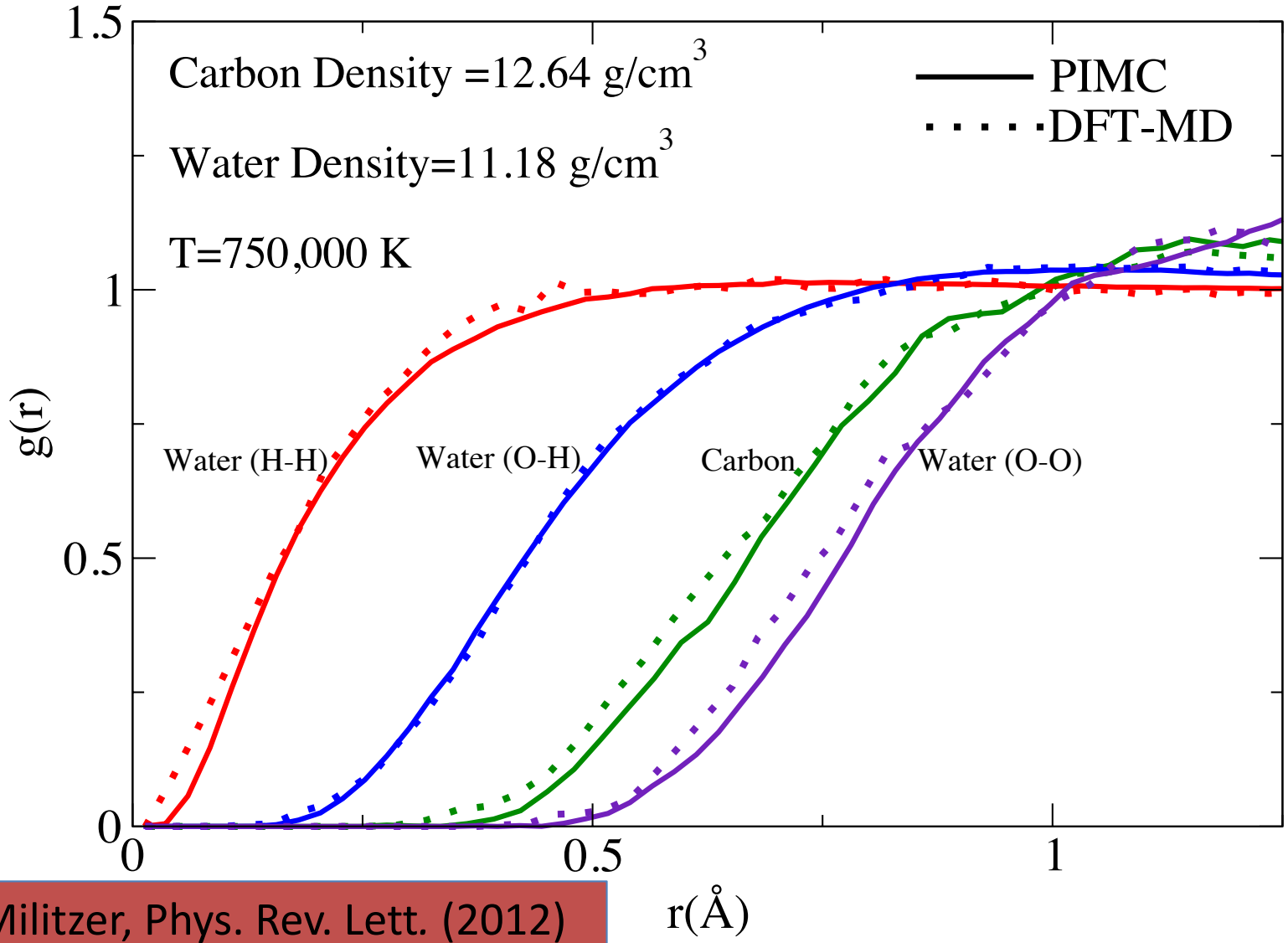


Water

First Path Integral Monte Carlo Simulations for Heavier Elements Fill this Gap in Temperature



Path Integral Monte Carlo and DFT-MD are in very good agreement



Driver, Militzer, Phys. Rev. Lett. (2012)

Silicon

Path Integral Monte Carlo with localized nodal surfaces and application to silicon plasmas

How the nodes are enforced:

$$\rho_F(\mathbf{R},\mathbf{R}';\beta) = \frac{1}{N!}\sum_{\mathcal{P}}(-1)^{\mathcal{P}}\int_{\mathbf{R}\to\mathcal{P}\mathbf{R}',\,\rho_T>0} d\mathbf{R}_t\; e^{-S[\mathbf{R}_t]}$$

Nodes are a Slater determinant:

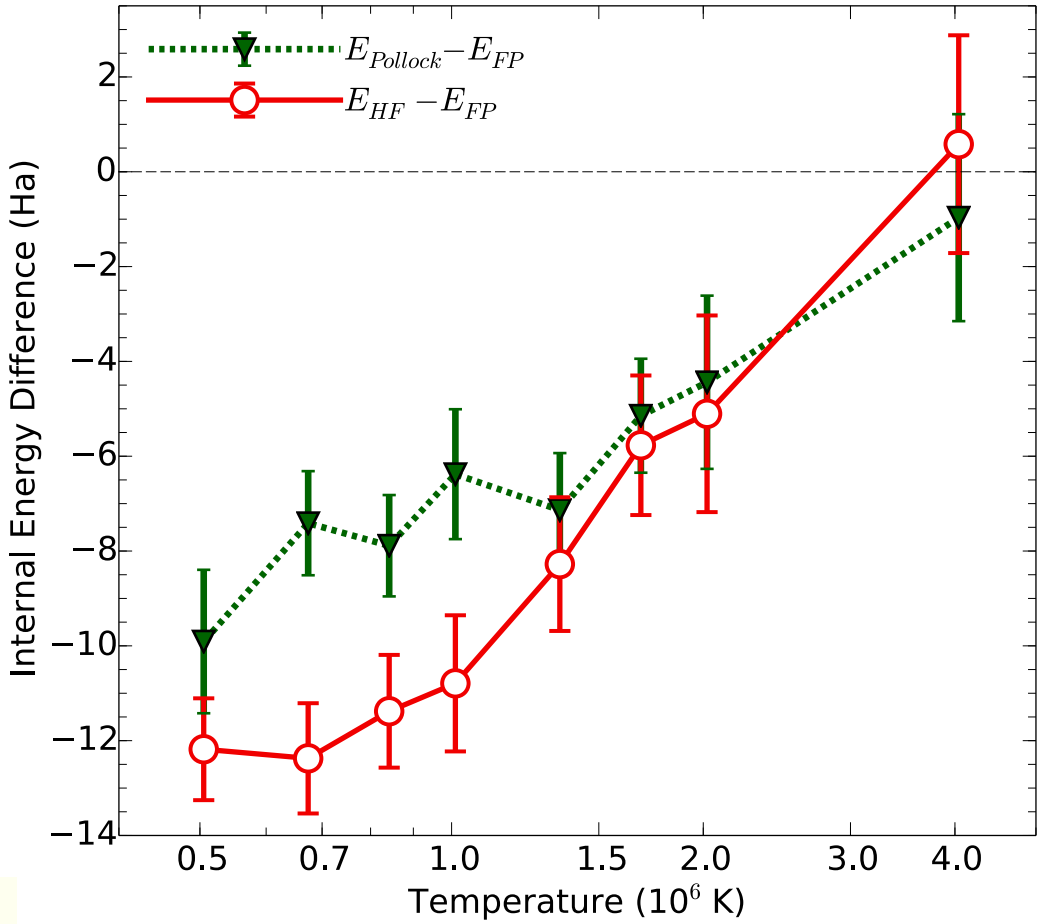
$$\rho_T(\mathbf{R},\mathbf{R}';\beta) = \left|\left|\rho^{[1]}(r_i,r'_j;\beta)\right|\right|_{ij}$$

Before we used only free-particle orbitals (plane waves):

$$\rho_0^{[1]}(r,r';\beta) = \sum_k e^{-\beta E_k}\,\Psi_k(r)\,\Psi_k^*(r')$$

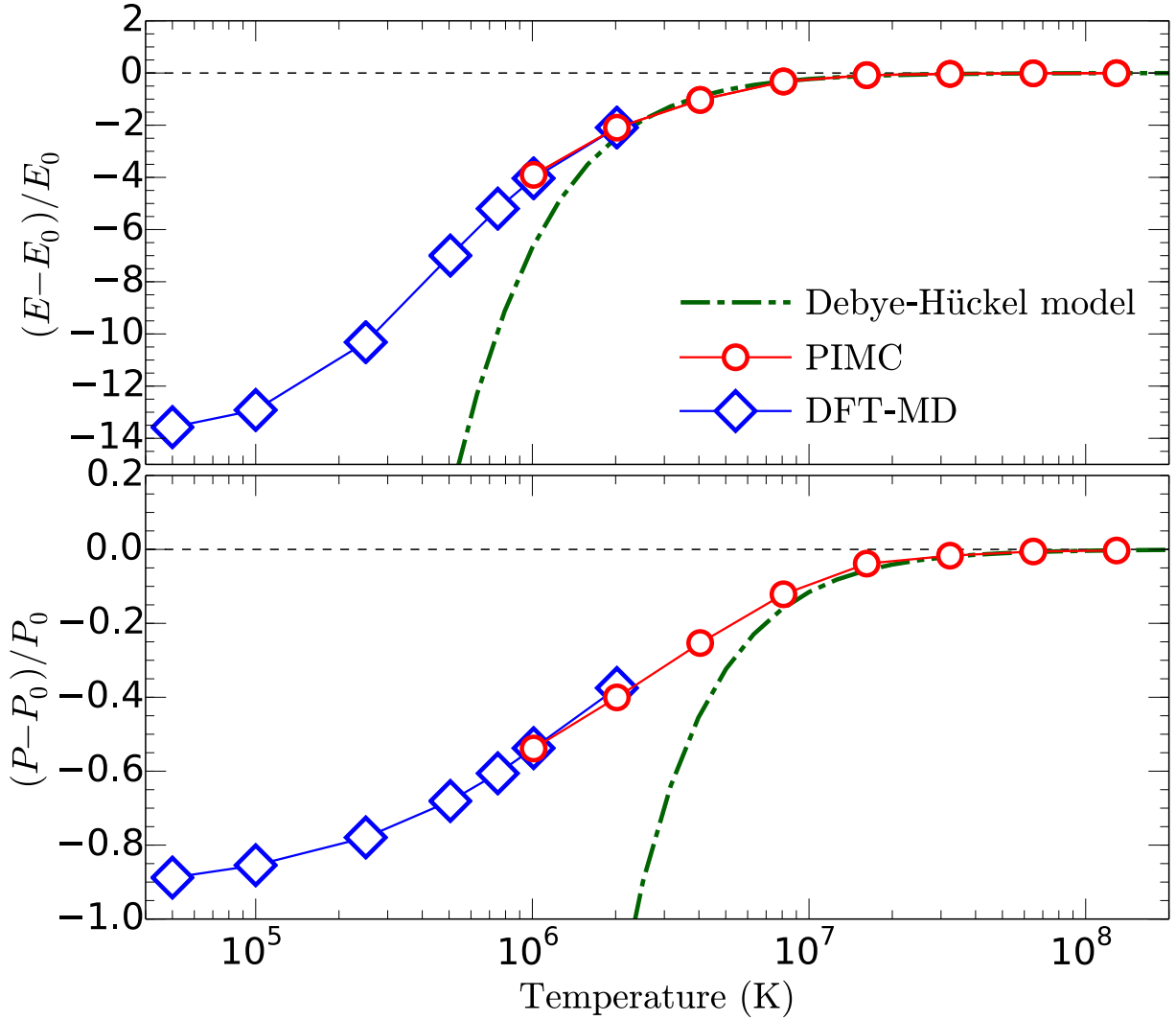
New idea: Add Hartree-Fock orbitals

$$\rho^{[1]}(r,r',\beta) = \sum_{I=1}^{N}\sum_{s=0}^{n} e^{-\beta E_s}\Psi_s(r-R_I)\Psi_s^*(r'-R_I)$$



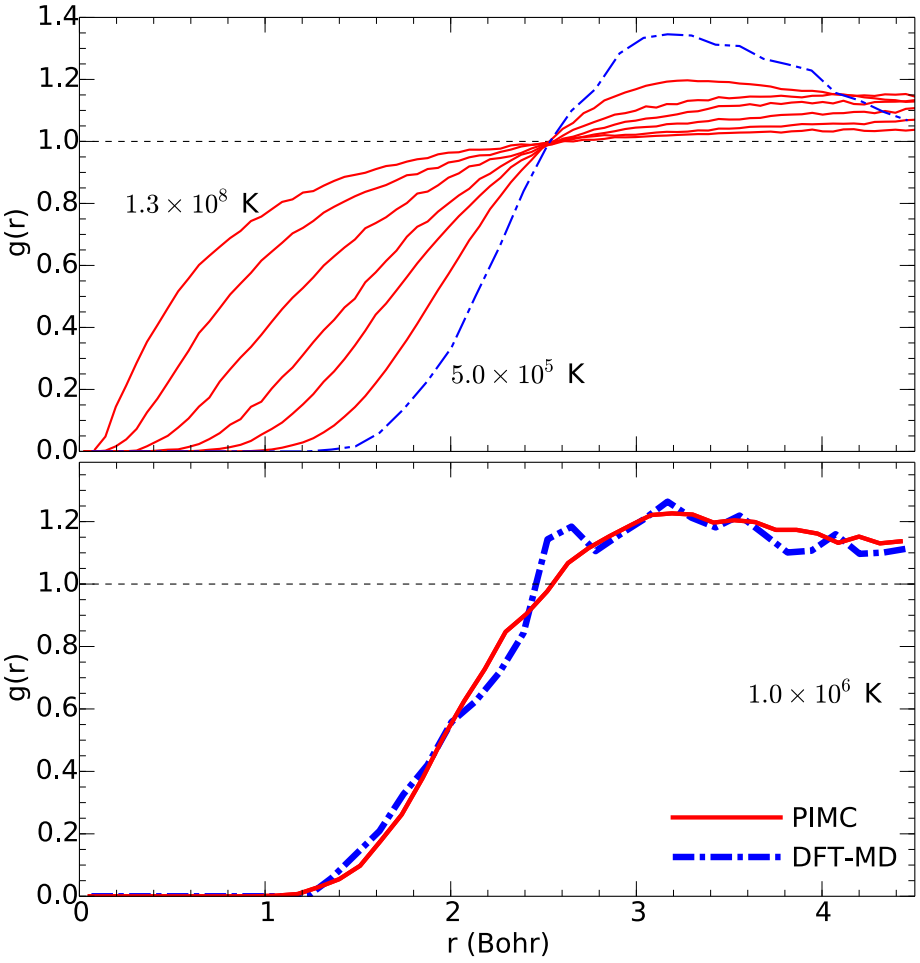
Militzer, Driver, Phys. Rev. Lett. (2015)

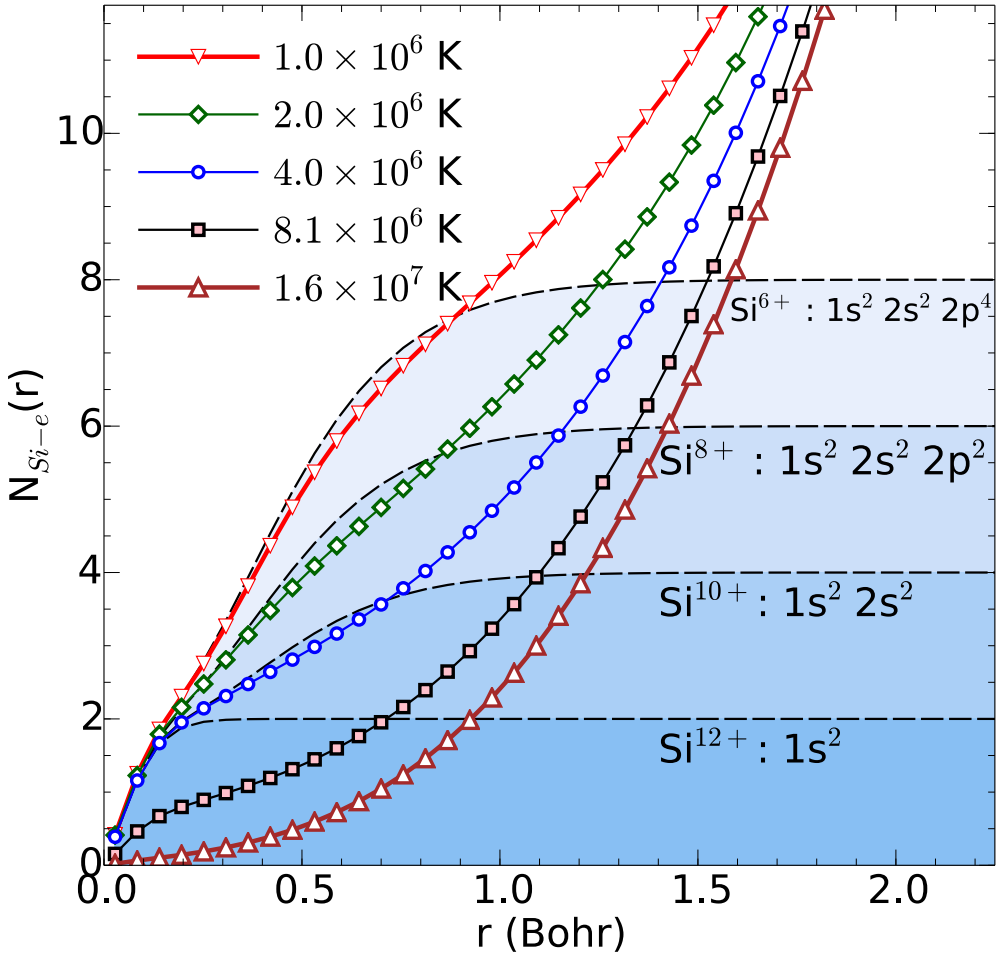
Silicon: Energy and Pressure Comparison of Path Integral Monte Carlo and Kohn-Sham DFT



Militzer, Driver, Phys. Rev. Lett. (2015)

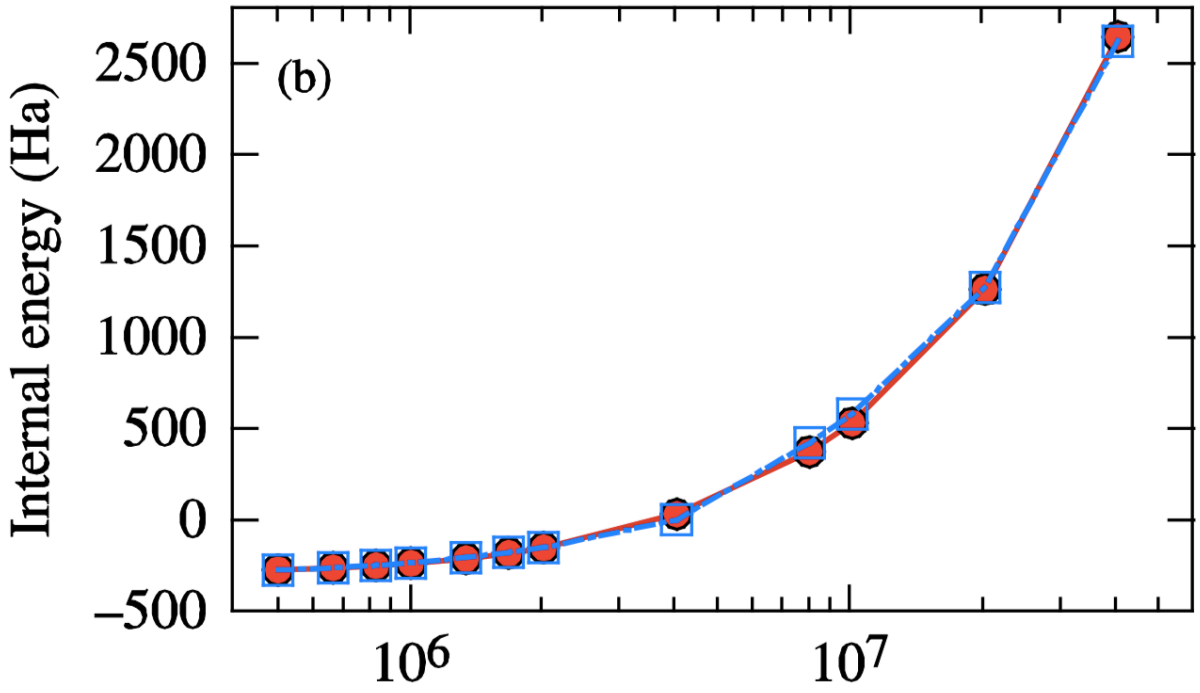
g(r) Comparison of Path Integral Monte Carlo and Kohn-Sham DFT

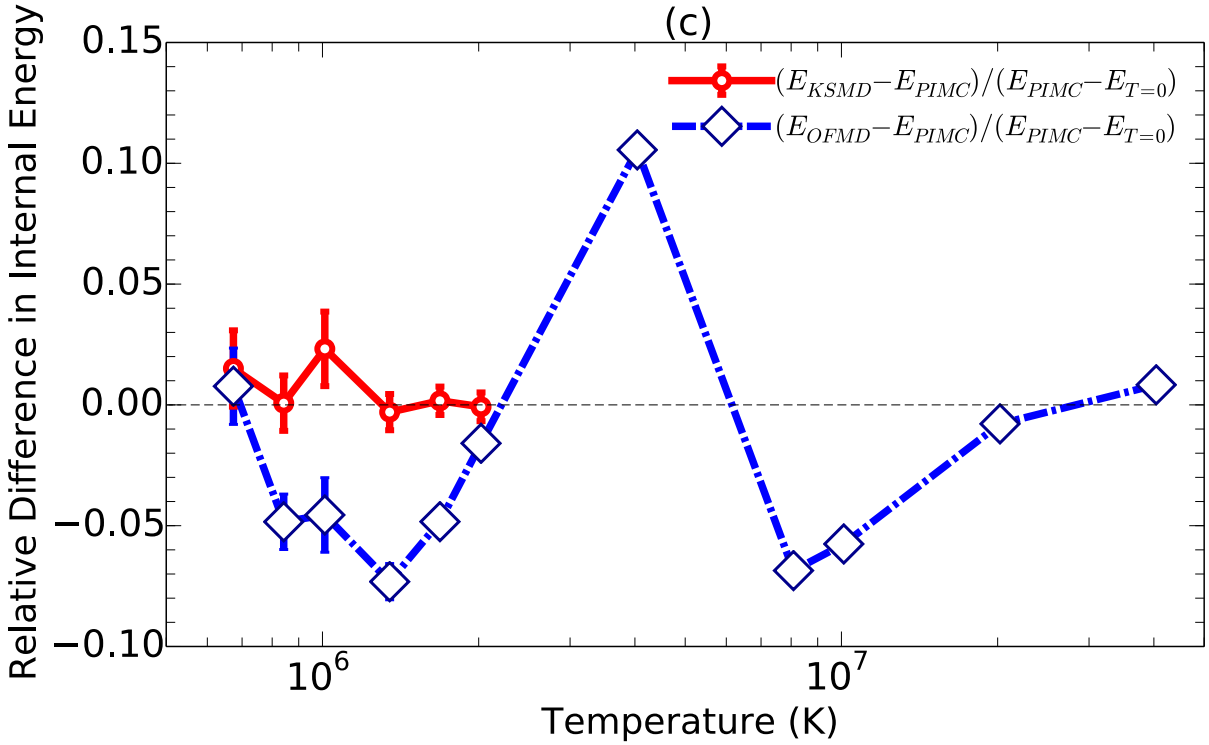




Militzer, Driver, Phys. Rev. Lett. (2015)

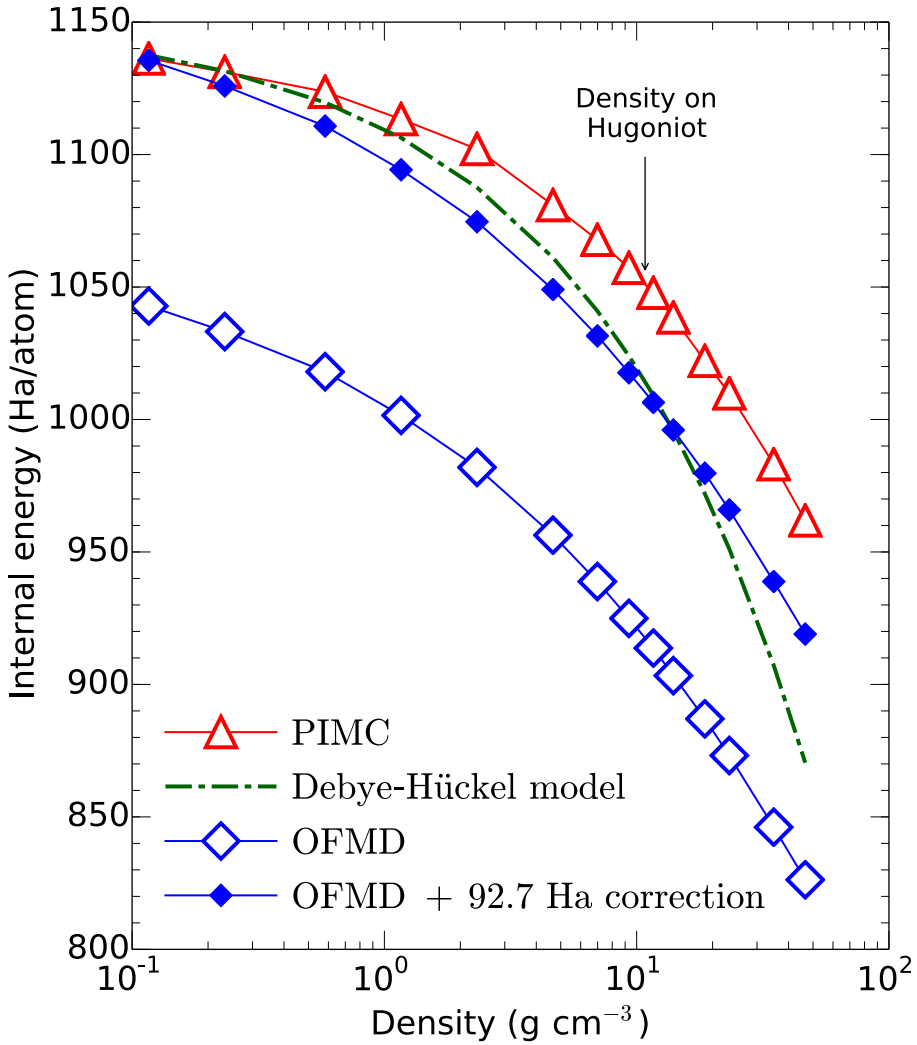
Energy of isolated silicon atom: Path Integral Monte Carlo and Orbital-Free DFT

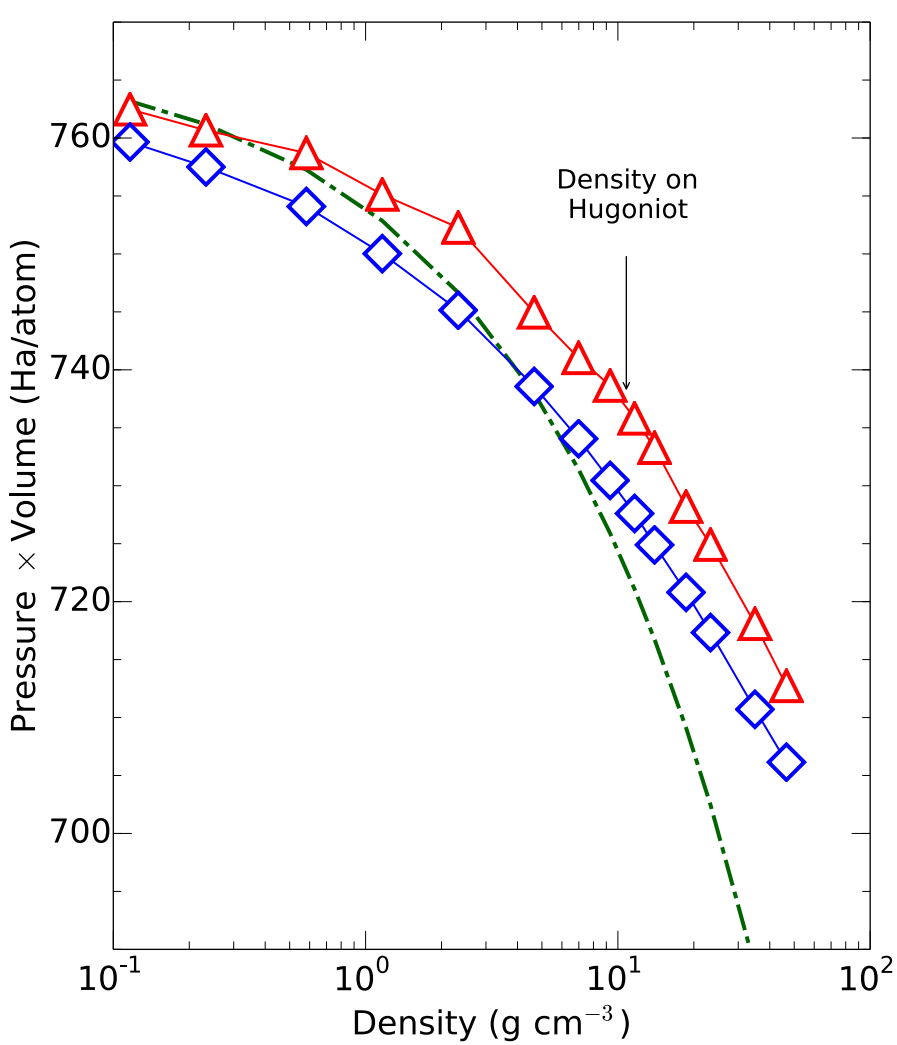




Hu, Militzer, Collins, Driver, Kress, Phys. Rev. B 94 (2016) 094109

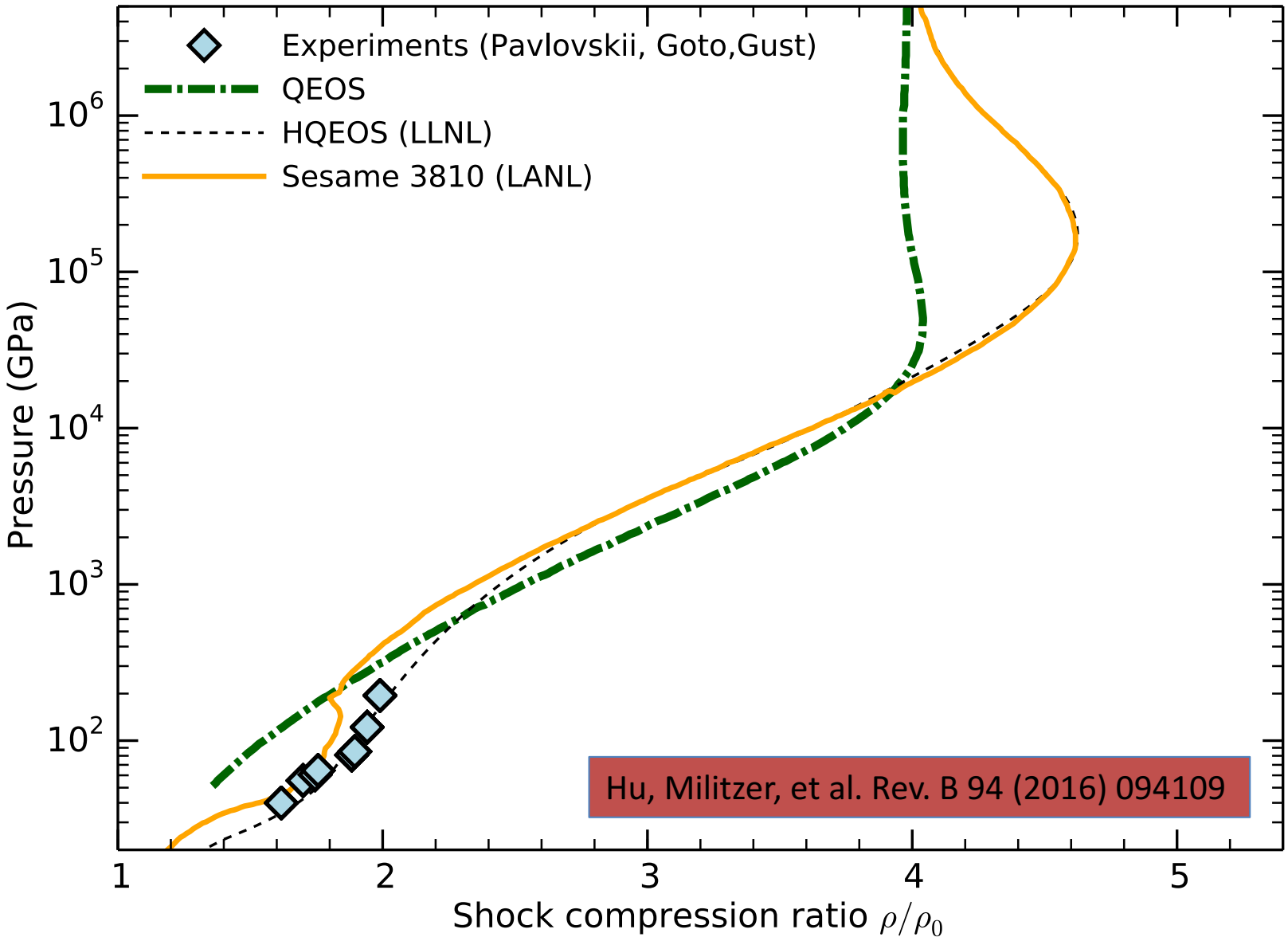
Energy and Pressure Comparison of Path Integral Monte Carlo & Orbital-Free DFT



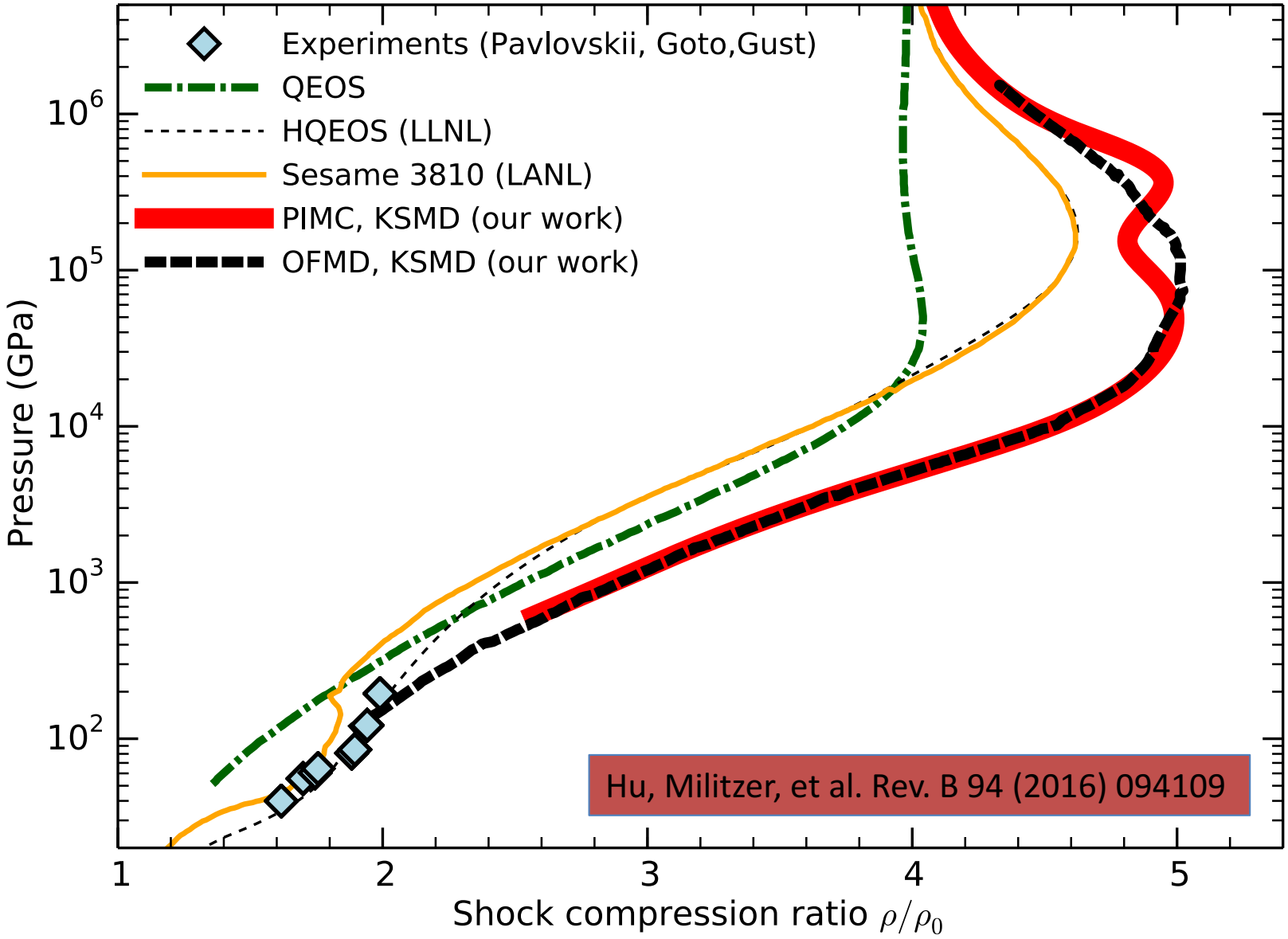


Hu, Militzer, Collins, Driver, Kress, Phys. Rev. B 94 (2016) 094109

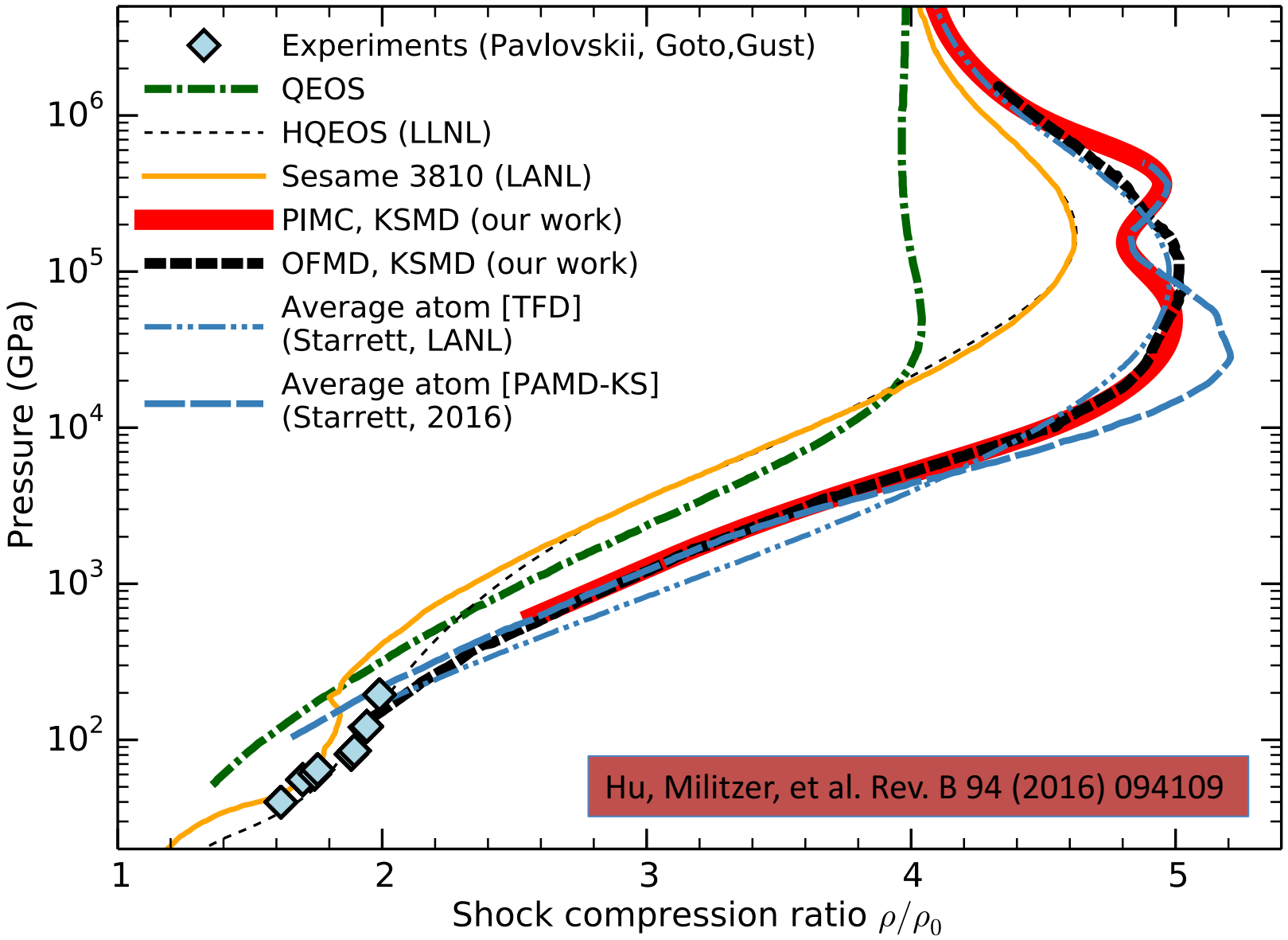
Silicon Hugoniot Curve: Experiments and Semi-analytical EOS models



Silicon Hugoniot Curve:
Path Integral Monte Carlo, Orbital-Free DFT



Silicon Hugoniot Curve:
Average-Atom models



Conclusions

- For elements from hydrogen to through silicon, the energy, pressure, and g(r) functions computed with PIMC and KS-DFT agree well at 2×10^6 K.
- Internal energy agrees to better than 5 Ha/atom; pressure to 2%.
- So far, we could not detect any problem with the zero-temperature exchange-correlation functionals.
- We will provide more EOS data to improve orbit-free methods.
- Second row elements: More comparison between simulations and experiments
- Extending PIMC to third row elements.

The End