First-Principles Simulations and Shock-Hugoniot Calculations of Warm Dense Neon

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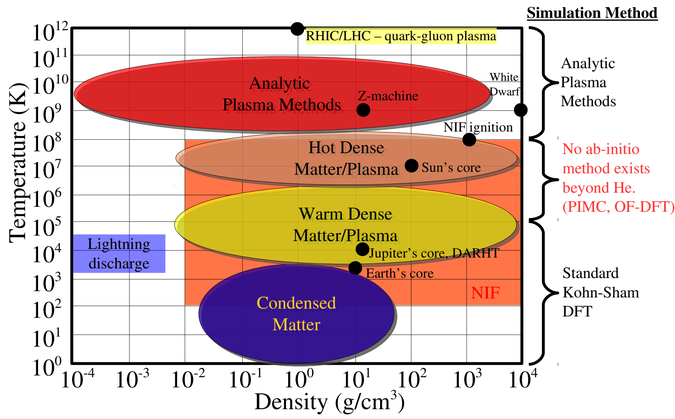
Introduction:first-principles mehods for warm dense matter (WDM)



- In WDM, effects of bonding, ionization, XC, and quantum degeneracy are all important.
- Standard KS-DFT intractable by T=10 eV due to large number of occupied orbitals.
- No reason to assume ground-state XC-functionals are valid for T > 0 K.
- PIMC offers a highly accurate route to simulate WDM. [1,2,3,4]
- WDM methods are important for inertial confinement fusion (ICF) and planetary cores.

Path Integral Monte Carlo Method

- Based on Feynman's path integral formulation of quantum statistics.
- A quantum, many-body generalization of classical Lagrangian action principle.
- Thermal density matrix determines thermodynamics of a many-body system.

$$\hat{\rho}=e^{-\beta\hat{H}}=e^{-E/k_bT}=\left[e^{-\frac{\beta}{M}\hat{H}}\right]^M=\left[e^{-\tau\hat{H}}\right]^M \quad \text{(Time slicing)}$$

- ρ is expressed as an imaginary time path integral at temperature T with time step τ.
- Sample paths from R to R' using action to accept/reject moves.

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

- Density matrix approximated as pair density; valid for converged time step or T large.

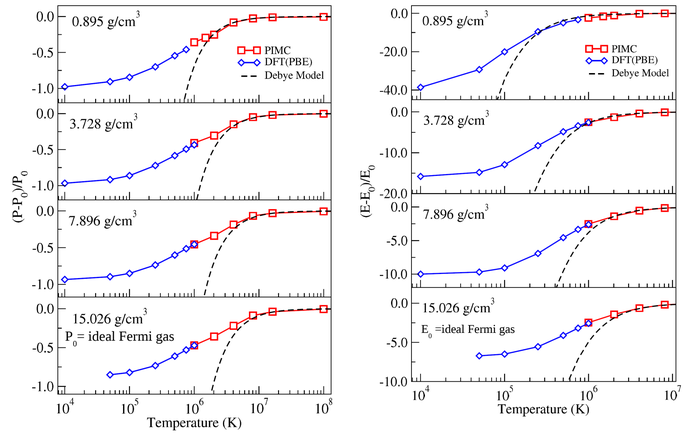
$$\hat{\rho}(\boldsymbol{R},\boldsymbol{R}';\beta)\rightarrow\hat{\rho}(\boldsymbol{r}_{ij},\boldsymbol{r}'_{ij};\beta) \quad \text{(Coulomb interaction: pair density matrices)}$$

- Fermion sign problem: positive and negative contributions to observable cancel.

$$\hat{\rho_{Trial}}(\boldsymbol{R},\boldsymbol{R}';\beta)>0 \quad \text{(Use trial nodes of } \textbf{free-particle} \text{ density matrix.)}$$

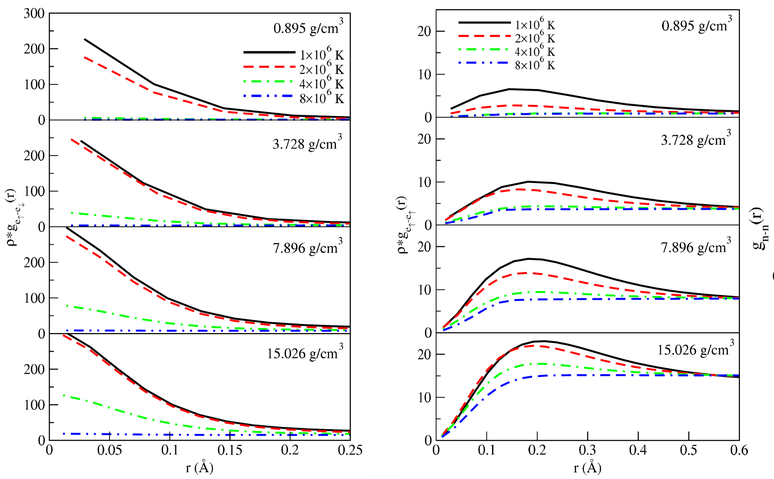
- Temperature explicitly included via density matrix
- Particles treated as paths which become shorter for higher temperatures
- Efficiency improves with temperature (unlike DFT efficiency, which decreases with T)
- Previously applied to only H[1] and He[2]. Extended to C [3,4] and H_2O [3]
- Free-particle nodes work at high temperature as atoms are sufficiently ionized

Neon Equations of State: Excess Pressure and Internal Energy vs. Temperature



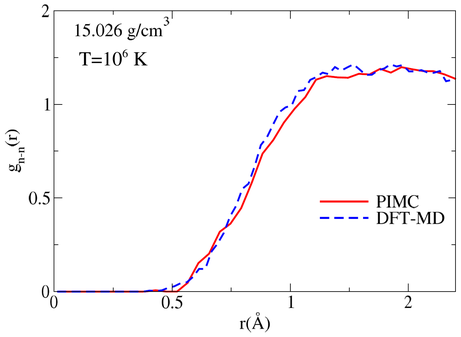
- DFT and PIMC form a coherent equation of state; PIMC converges to classical Debye-Hückel model
- PIMC and DFT overlap at 10^6 K; confirms DFT functionals work at high T

Neon Pair-Correlation Functions, g(r)



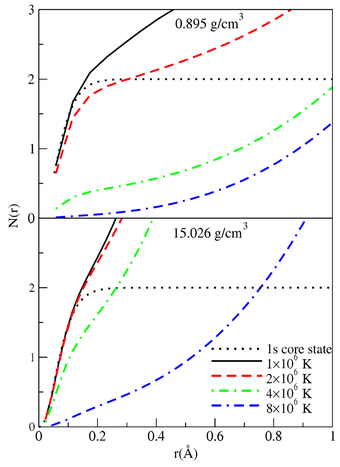
- Opposite-spin electrons remain near the nuclei until excited away by temperature.

- Same-spin electrons have zero correlation near the nuclei due to Pauli exclusion.



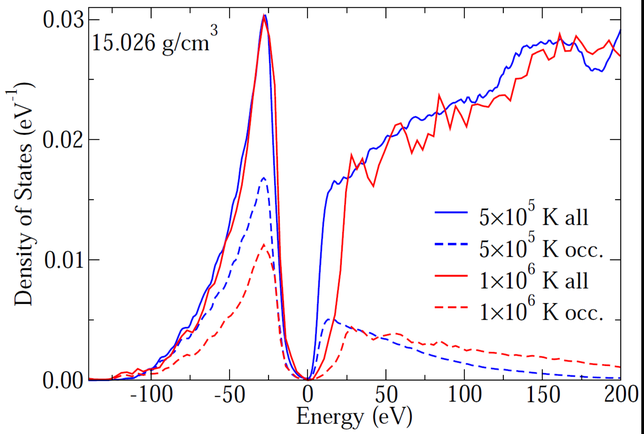
- The nuclear-nuclear g(r) function from PIMC and DFT-MD agree at 10^6 K

Neon Number Density, N(r): 1s excitation



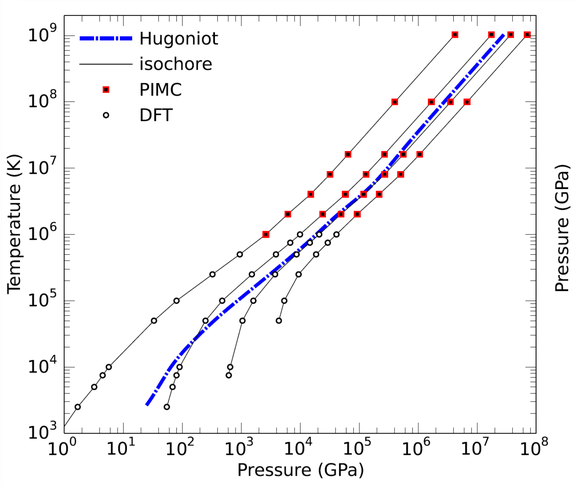
- N(r) is average number of electrons near the nucleus
- Reveals occupation dependence of the 1s state on temperature

Neon Electronic Density of States

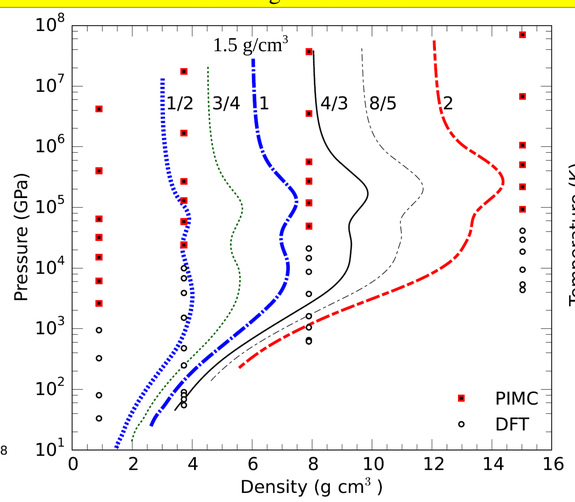


- An electronic gap persists even at 15 g/cm^3 and 10^6 K
- Neon does not become metallic at these conditions

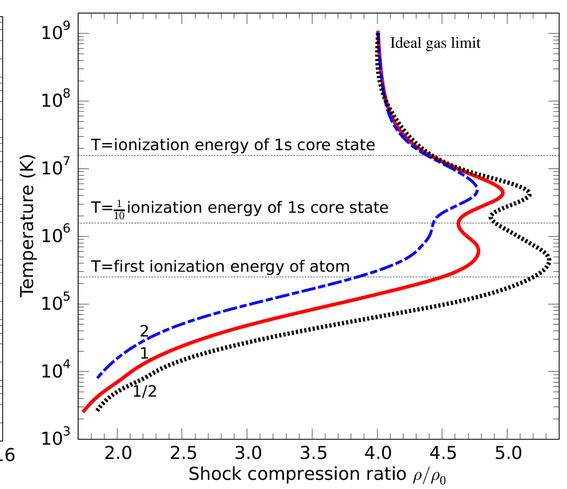
Neon Hugoniot Curves



- PIMC and DFT isochores in the T-P space explored
- Hugoniot curve for an initial, ambient density of 1.5 g/cm^3



- Shock Hugoniot pressures reached for different initial densities
- 2-fold precompression doubles the maximum shock density



- Temperatures vs. shock density divided by initial density
- Precompression leads to slight reduction in compression ratio

Conclusions

- We have a PIMC method that produces accurate results for WDM for first and second row elements.
- PIMC and DFT-MD together form a coherent equation of state from condensed matter to the plasma limit. PIMC pressures, internal energies, and pair correlation functions agree with DFT-MD at 10^6 K.
- Neon is not metallic for the most extreme conditions studied here
- Hugoniot curves reveal how particles interact at high density by showing how shock density depends on initial density.
- Neon results submitted to Phys. Rev. B.

Acknowledgements

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[1]B. Militzer and D.M. Ceperley, Phys. Rev. E **63**, 066404 (2001).
[2]B. Militzer, Phys. Rev. B **79**, 155105 (2009).
[3]K.P. Driver and B. Militzer Phys. Rev. Lett. **108**, 115502 (2012)
[4]L.X. Benedict, K.P. Driver, *et al.*, Phys. Rev. B, **89**, 224109 (2014)