Theory for Transport Properties of Warm Dense Matter

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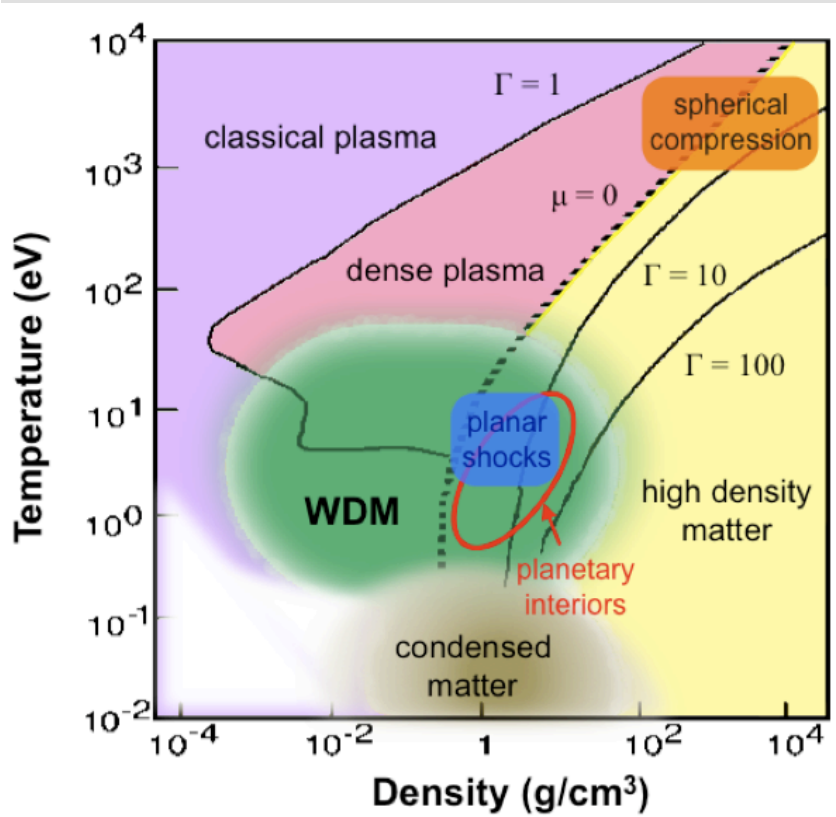
Collaborators

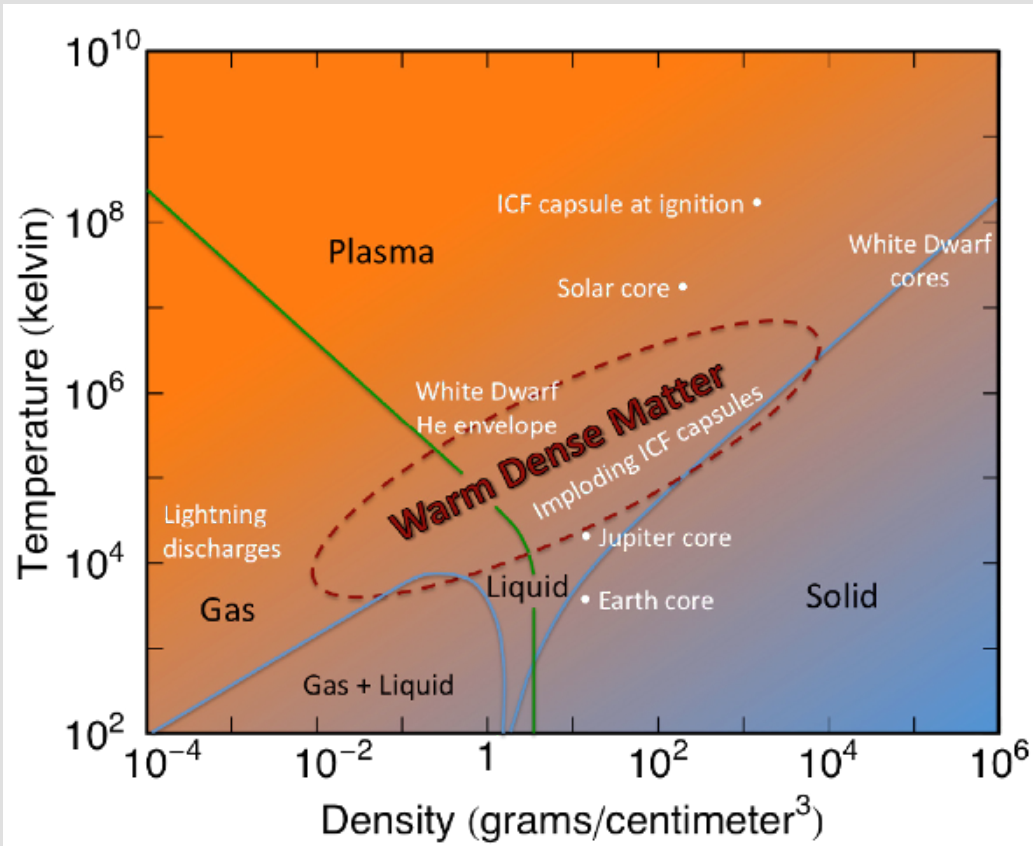
- Jerome Daligault (LANL)
  - All aspects of this work: theory, simulations
- Starrett, Saumon and Sjostrom (LANL)
  - Warm Dense Matter calculations
  - First-principles quantum simulations

Los Alamos
NATIONAL LABORATORY
EST. 1943

What is warm dense matter?



Lee, et al., J. Opt. Soc. Am. B 20, 770 (2003)



http://www.lanl.gov/projects/dense-plasma-theory/

Intersection of different disciplines
(plasma and condensed matter)

What is warm dense matter?

- Ions: Moderate-strongly coupled & classical

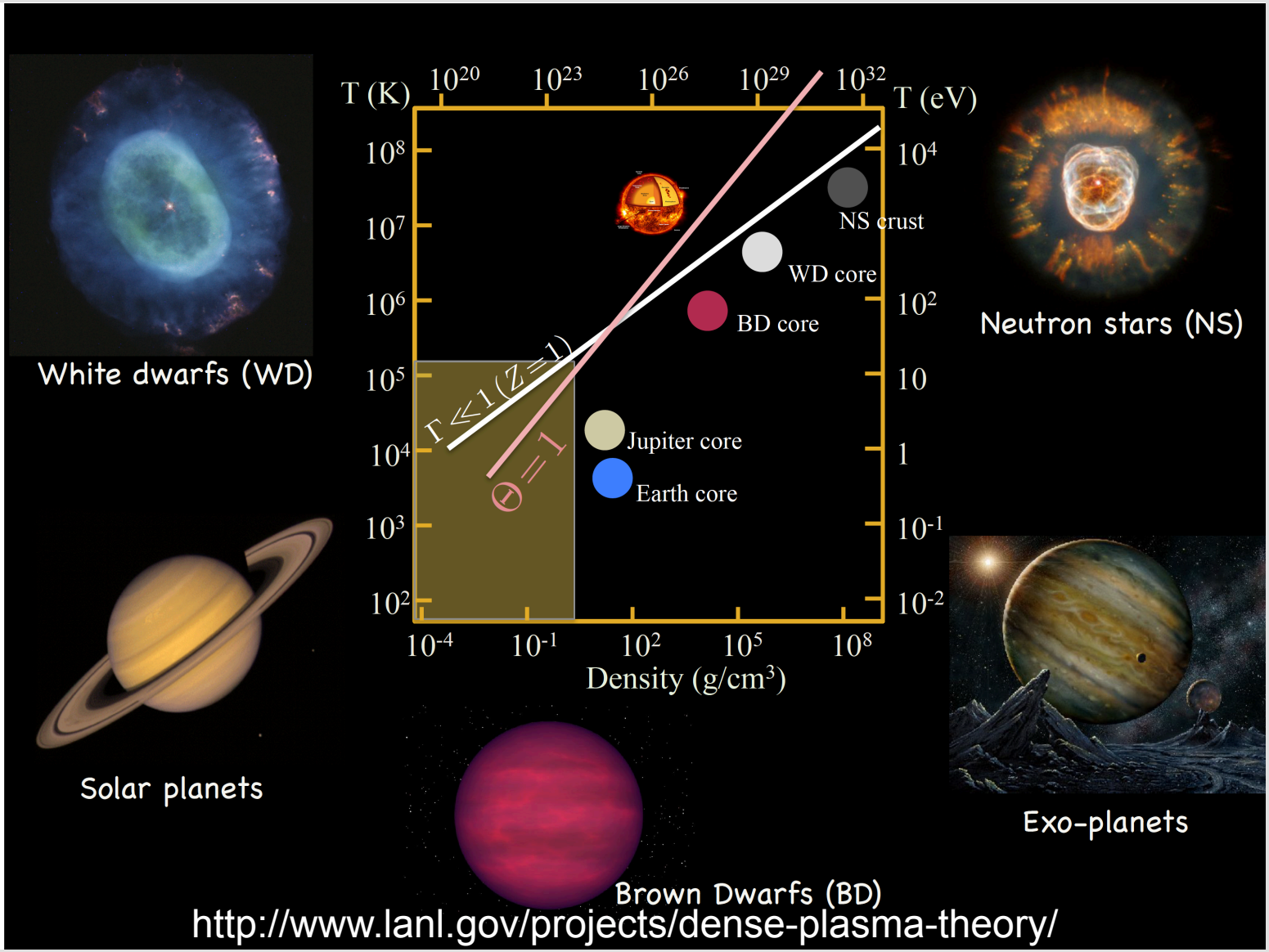
$$\Gamma \equiv \frac{e^2/a}{k_{\mathrm{B}}T} \sim \frac{\langle \text{potential energy} \rangle}{\langle \text{kinetic energy} \rangle}$$

- Electrons: Weak-moderately coupled & degenerate

$$\Theta \equiv \frac{k_B T}{E_F} \sim \left(\frac{a}{\lambda_{db}}\right)^2 \sim \frac{m a^2 k_B T}{\hbar^2}$$

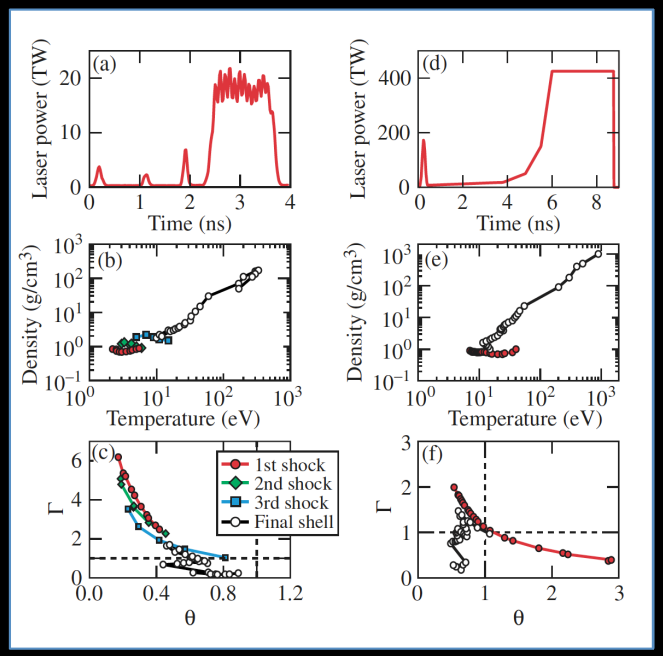
- Too dense for plasma theory, too hot for condensed matter theory

WDM in Astrophysics

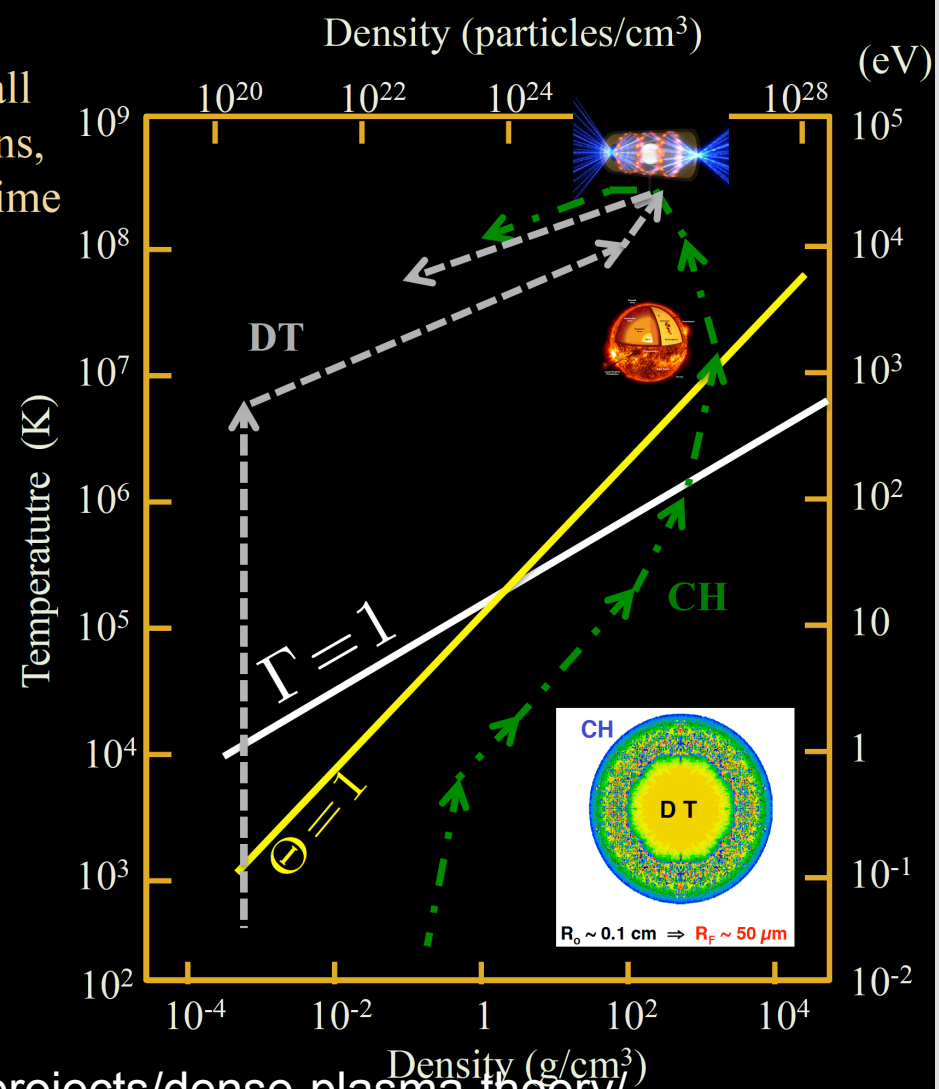


Inertial Confinement Fusion

ICF experiments start from a cryogenic sample of gas and solid, all the way to extreme ignition conditions, going across the hot dense matter regime



Hu et al. Phys Rev. Lett. (2004)



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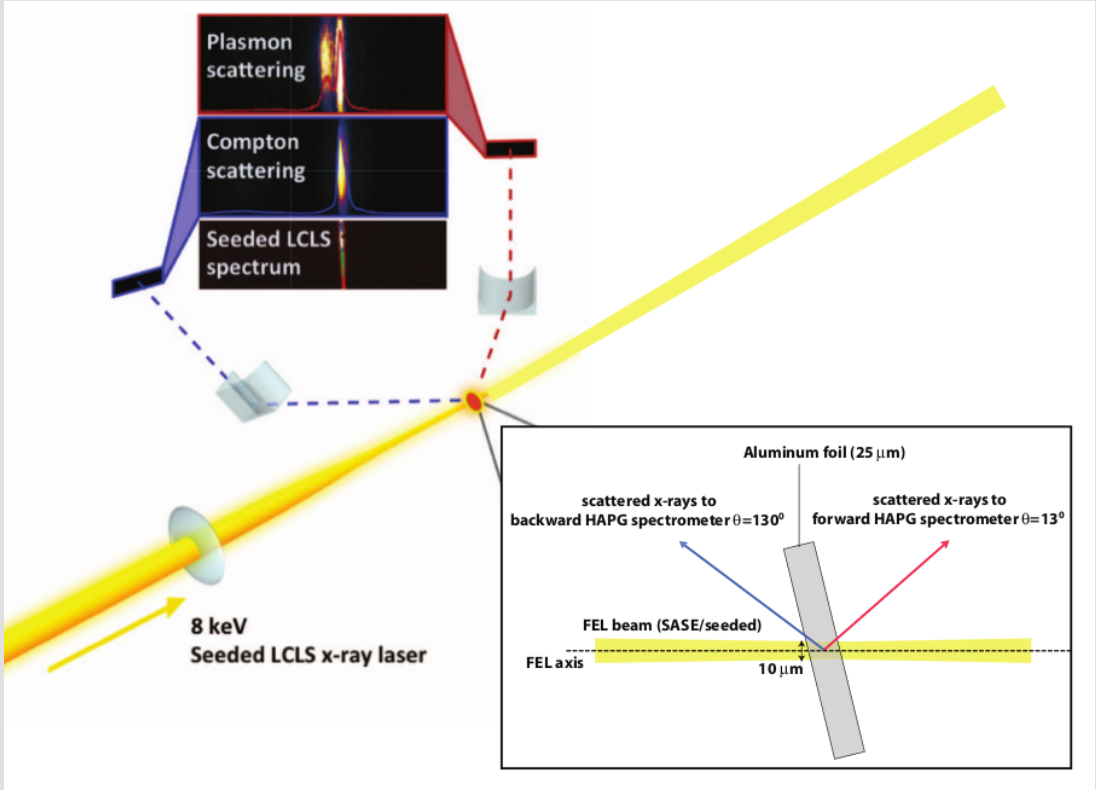
HEDP Experiments



- Significant advancements in X-ray free-electron lasers (XFELs) have enabled detailed WDM experiments
- World leading facility: Linac Coherent Light Source (LCLS) at SLAC

Research Needs

- Hydrodynamics (or rad-hydro) simulations
  - Transport coefficients
  - Equation of state
- Need to be cheap to evaluate
  - Table lookup
  - Formula
- Currently no accurate theory is available
  - Large-scale simulations (MD, DFT) dominate
  - Far too expensive to build tables with

Outline of this talk

- Effective Potential Theory
  - From static [g(r)] to dynamic (transport)
  - Very efficient to evaluate
- One-component plasmas
  - Hypernetted chain approximation (input)
  - Validation using molecular dynamics
- Warm dense matter
  - Quantum hypernetted chain approximation
  - Validation using orbital free density functional theory

Traditional Plasma Transport Theory

Coulomb Potential (with cutoff)

↓

Binary scattering cross section

↓

Boltzmann kinetic equation

↓

Chapman-Enskog solution
(or Grad, moment, etc)

↓

Diffusivity, viscosity, electrical and thermal conductivity

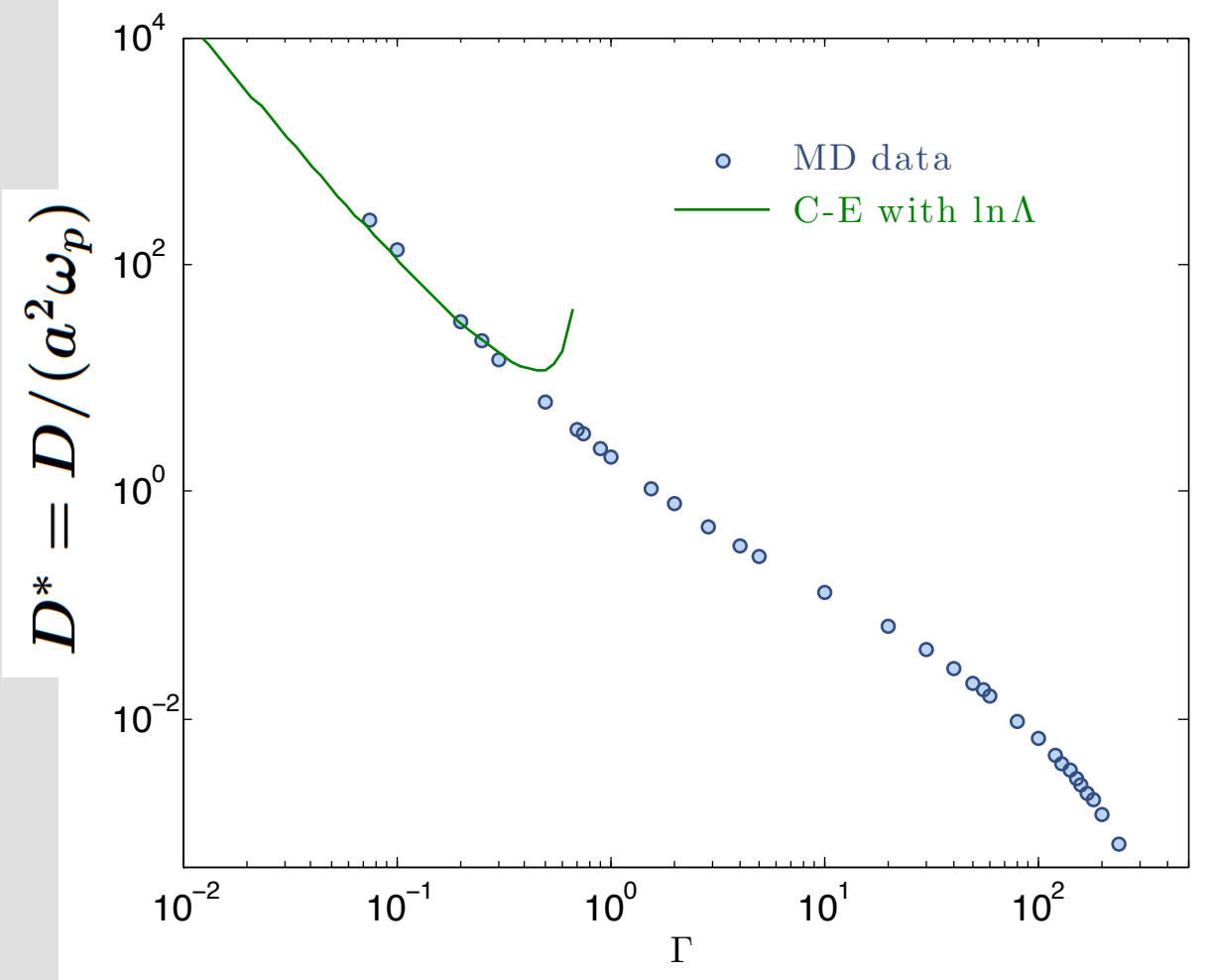
Classical Molecular Dynamics

- Solves **F**=m**a** for every particle
  - Large periodic box
  - We use P^3M method (separates close and distant interactions)
  - Transport coefficients from Green-Kubo relations

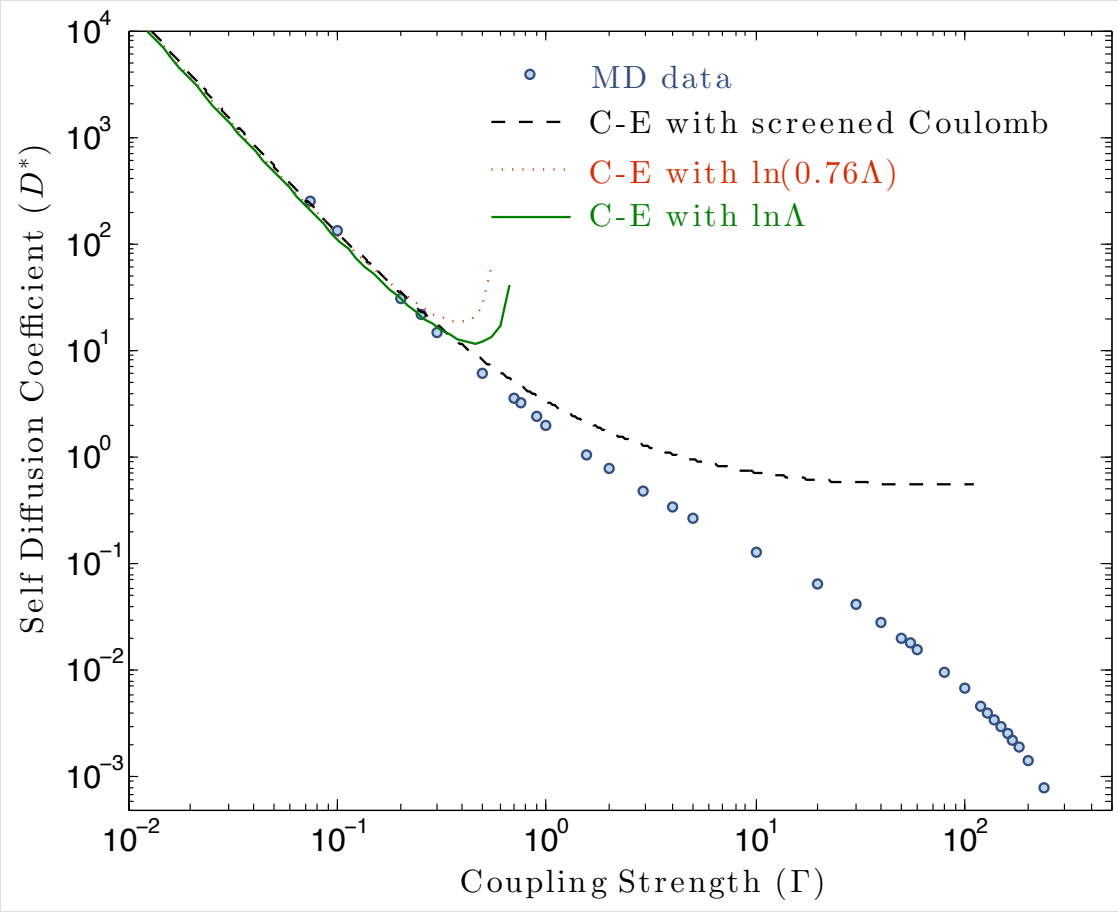
One Component Plasma (OCP)

- Model system that allows ab-initio simulations
  - Electrons form a uniform neutralizing background
  - Good model for ion transport in some systems
  - Good test bed for theory

Plasma Theory Breaks Down at Strong Coupling



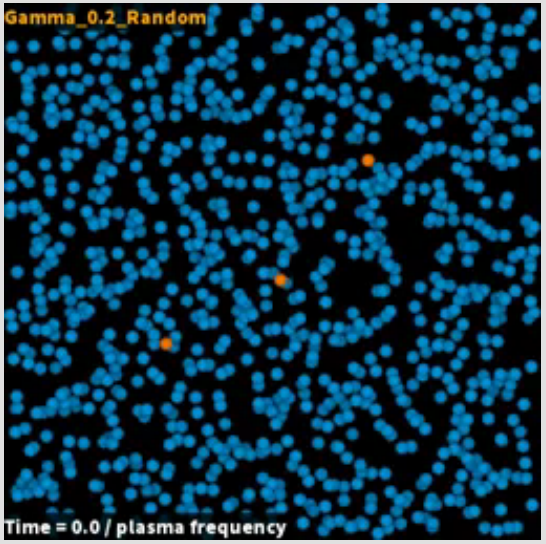
Using the Debye-Huckel Potential Fixes the Divergence



- But does not capture coupling effects

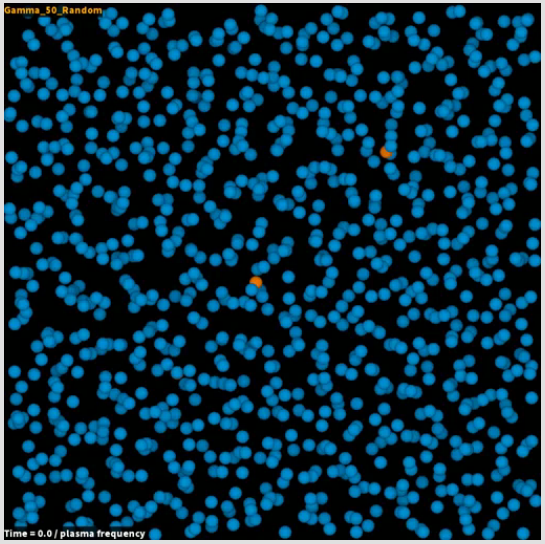
Particle trajectories in MD

Γ=0.2



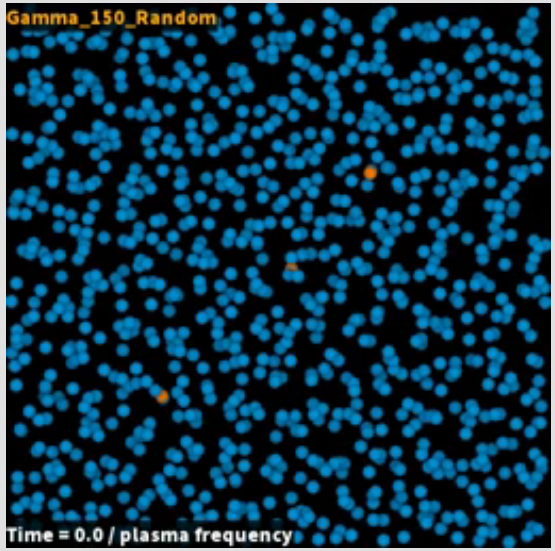
Small angle

Γ=50



Large angle, correlated

Γ=150



Trapped

http://www.lanl.gov/projects/dense-plasma-theory/

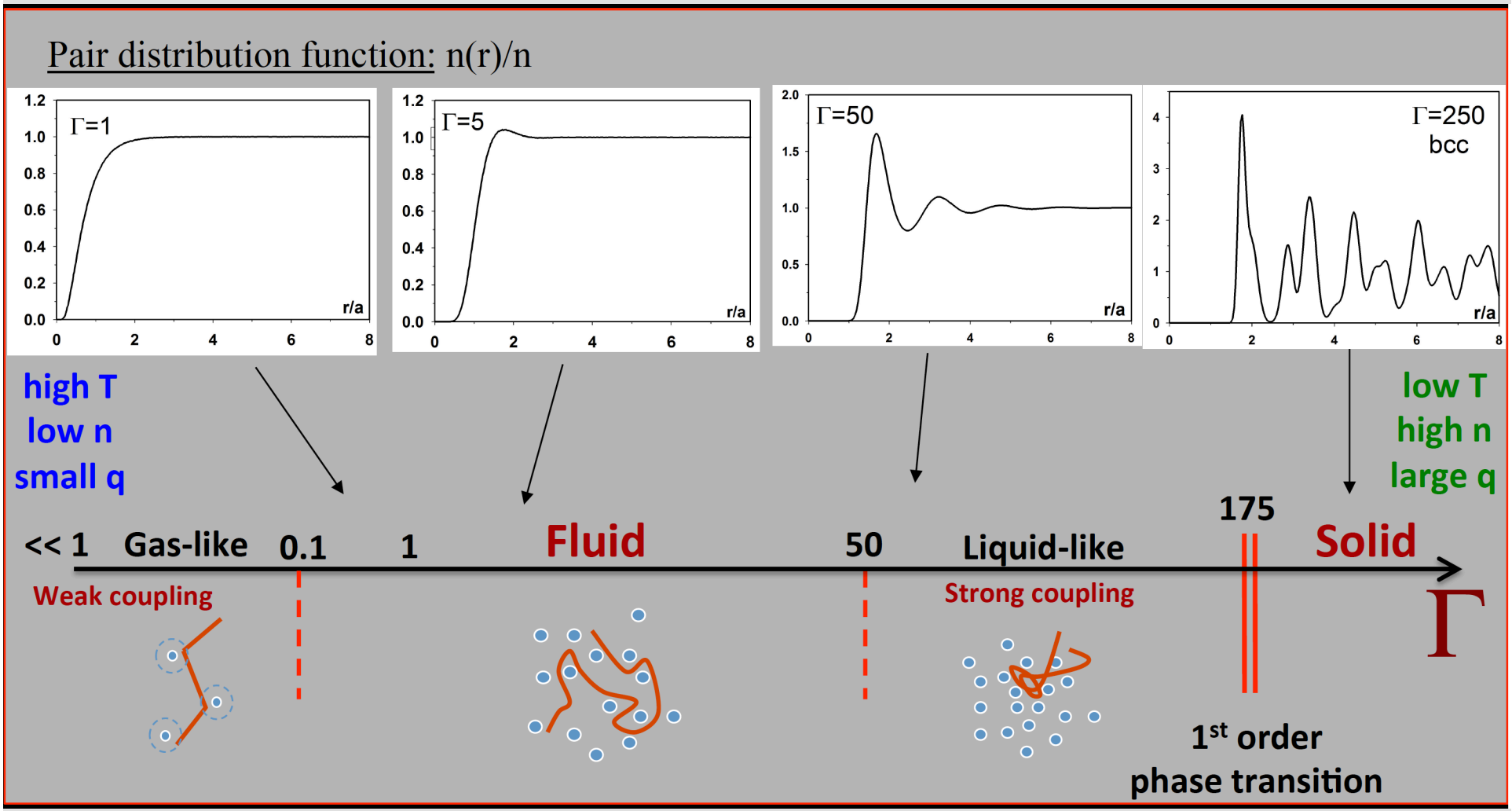
The Potential of Mean Force

- The potential obtained when taking two particles at fixed positions and averaging over the positions of all other particles

$$\begin{aligned}\vec{F}_{12} &= \int \left[-\nabla_{\vec{r}_1} U(\vec{r}_1, \vec{r}_2, \ldots, \vec{r}_N) \right] \frac{e^{-U/k_B T}}{\mathcal{Z}} d\vec{r}_3 \ldots d\vec{r}_N \\ &= -k_B T \nabla_{\vec{r}_1} \ln g(|\vec{r}_1 - \vec{r}_2|) \equiv -\nabla_{\vec{r}_1} \phi(\vec{r}_1 - \vec{r}_2)\end{aligned}$$

- = Debye-Huckel at weak coupling
- g(r) is the radial distribution function

Phase Diagram of the OCP



The Effective Potential Theory (1.0)

Potential of mean force

↓

Binary scattering cross section

↓

Boltzmann kinetic equation

↓

Chapman-Enskog solution
(or Grad, moment, etc)

↓

Diffusivity, viscosity, electrical and thermal conductivity

Hypernetted Chain Approximation

- A closure scheme to approximate g(r)

$$g(\vec{r}) = \exp\left[-v(\vec{r})/k_B T + n \int c(|\vec{r} - \vec{r}'|) h(\vec{r}') d\vec{r}'\right]$$

$$\hat{h}(\vec{k}) = \hat{c}(\vec{k})[1 + n\hat{h}(\vec{k})]$$

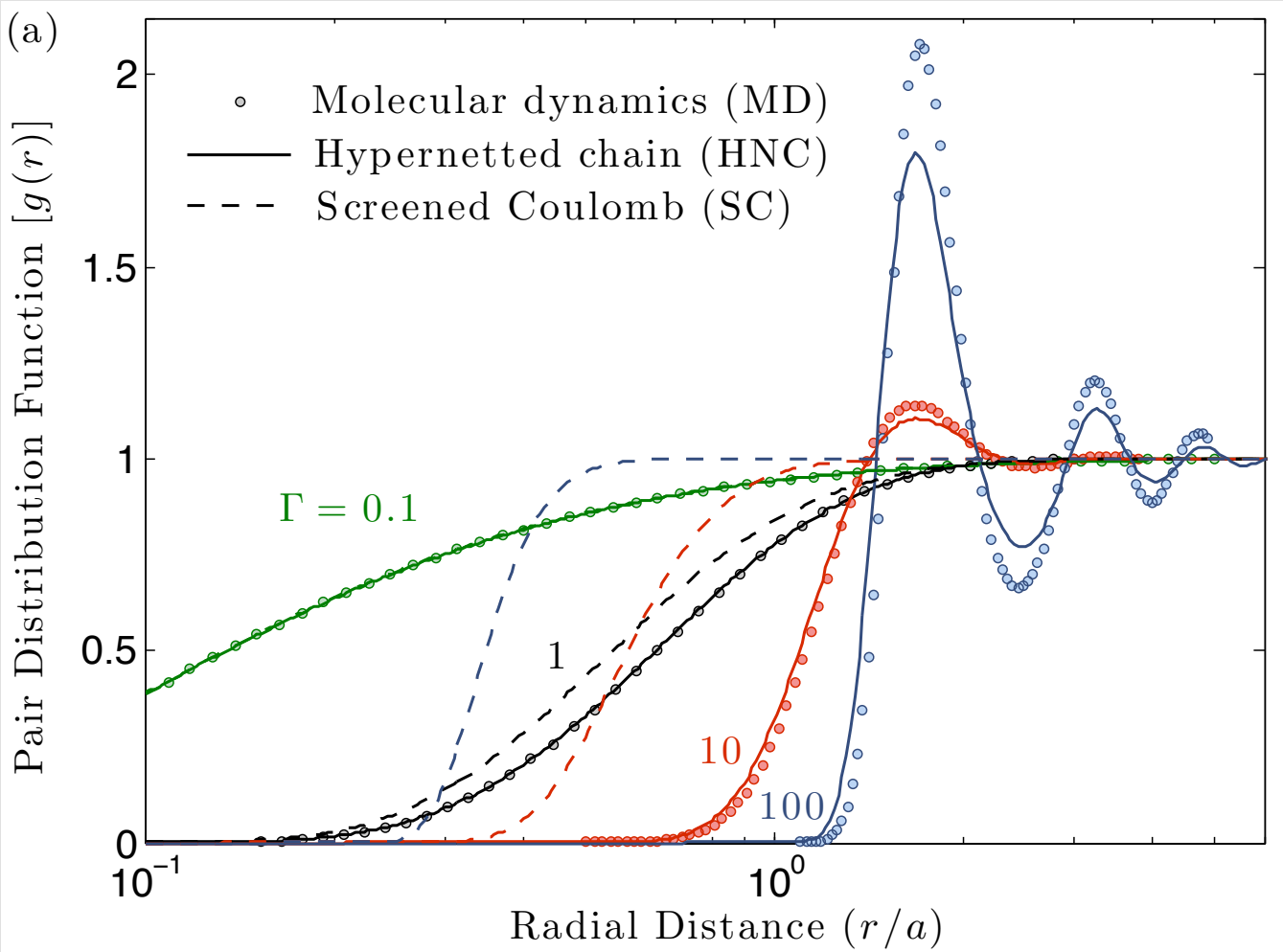
  – Coulomb potential: $v(\vec{r})/k_B T = \Gamma a/r$

  – Total correlation function: $h(r) = g(r) - 1$

- Theory input is then

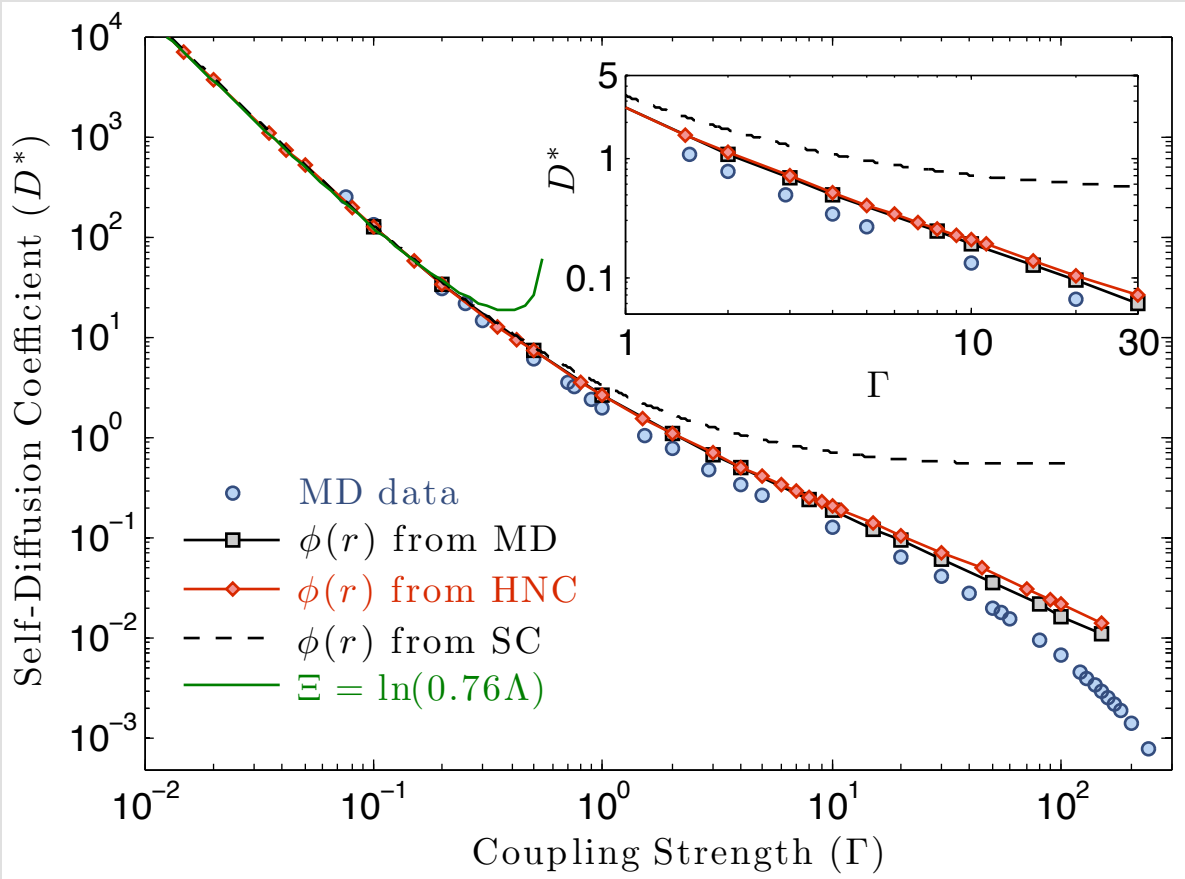
$$g(r) = e^{-e\phi/k_B T}$$

HNC works for Coulomb systems



EPT is accurate and efficient

- From equilibrium [g(r)] to dynamic [D]

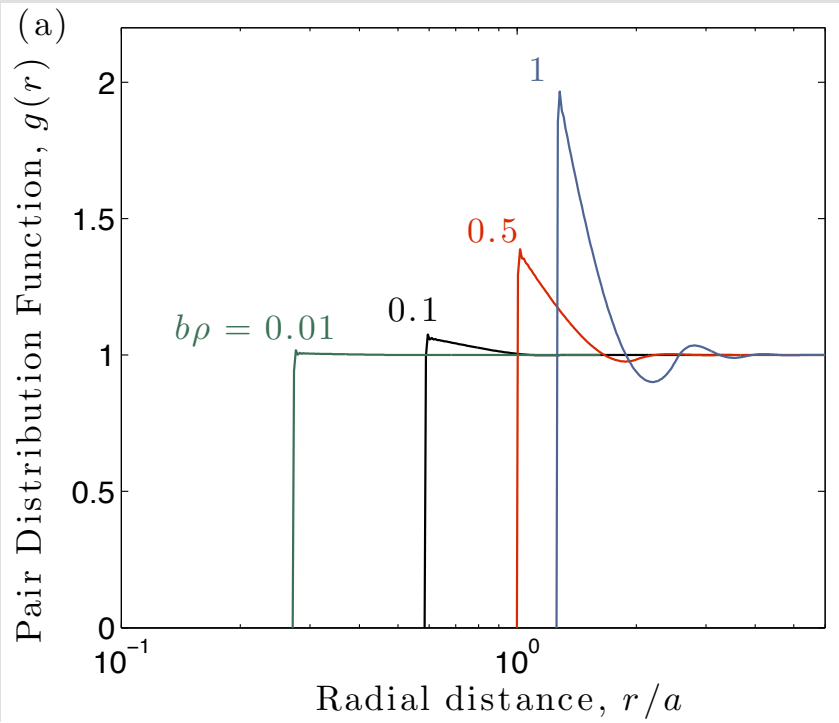


Baalrud and Daligault, PRL 110, 235001 (2013)

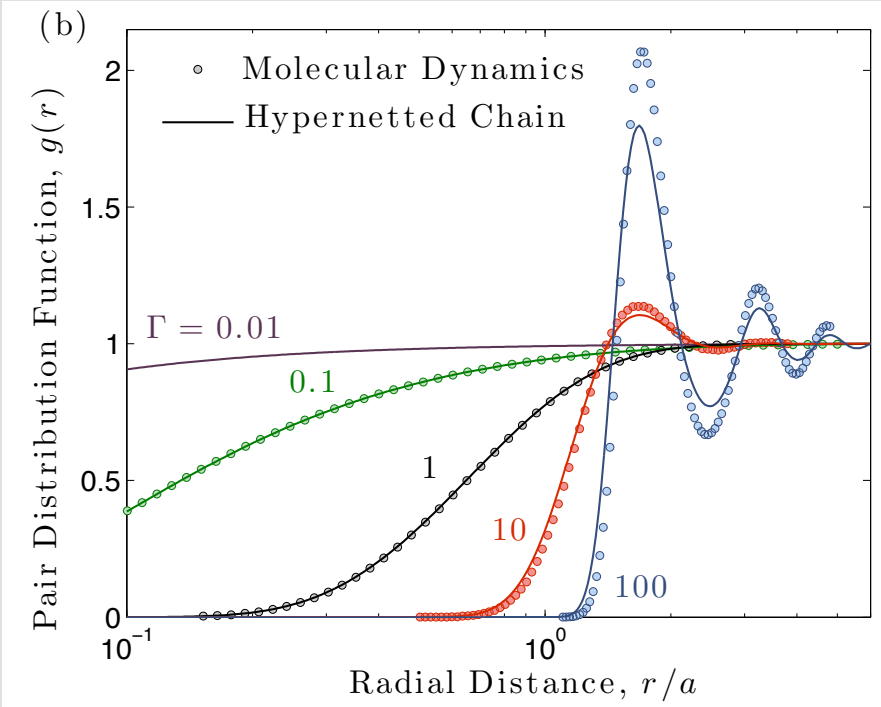
EPT can be improved

- Account for the “Coulomb hole”
  - Increased scattering due to excluded volume
  - Analogy with Enskog’s kinetic equation for hard spheres

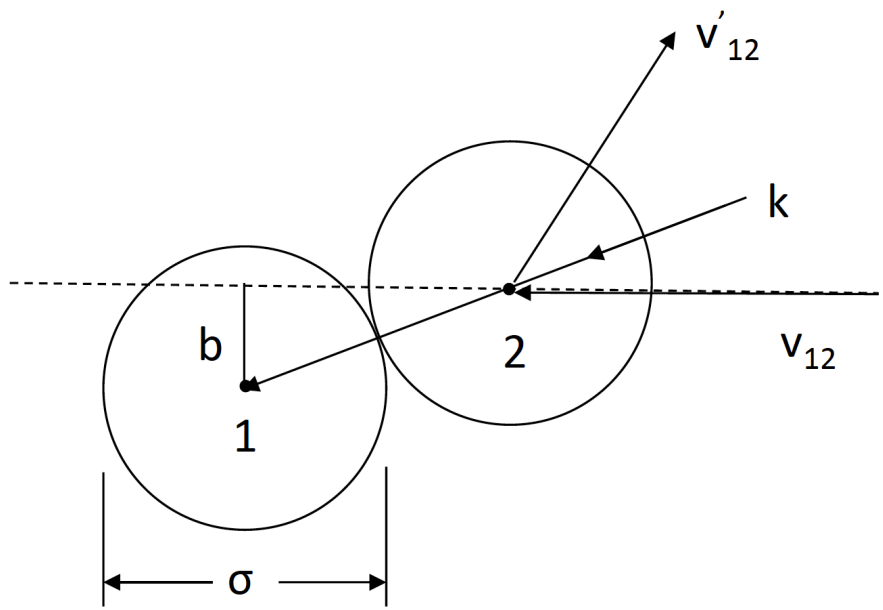
Hard Sphere



Coulomb



Enskog developed a kinetic theory for hard spheres



$$C = \int\int d^3v' d^2k \sigma^2 \vec{u}\cdot\vec{k}\left\{\chi(\vec{r}+\frac{1}{2}\sigma\vec{k})\hat{f}_s(\vec{r})\hat{f}'_{s'}(\vec{r}+\sigma\vec{k}) - \chi(\vec{r}-\frac{1}{2}\sigma\vec{k})f_s(\vec{r})f_{s'}(\vec{r}-\sigma\vec{k})\right\}$$

- χ accounts for the occupied volume
- Relaxes the molecular chaos approximation

Analogy with Coulomb hole

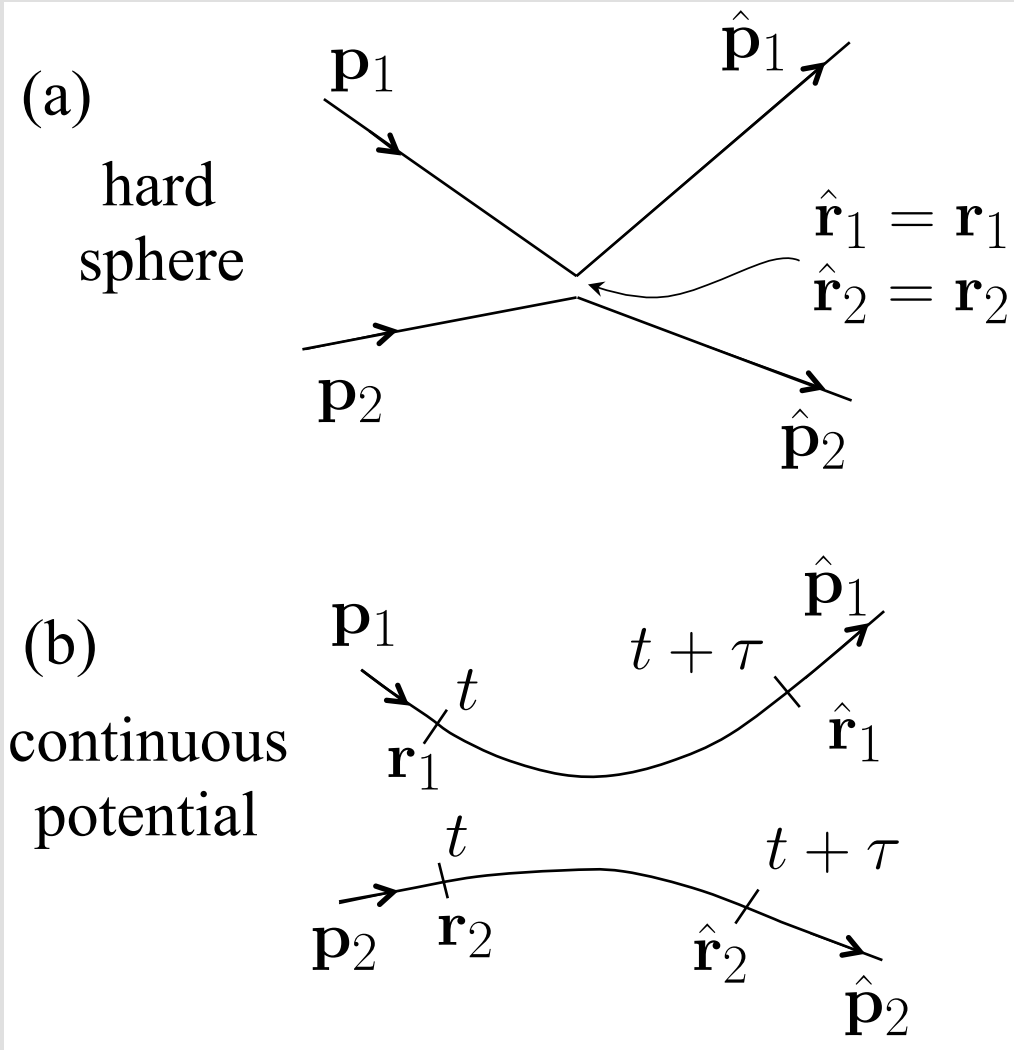
- Find radius from g(r)=0.87
  - This value was determined “empirically”
  - Expect the value to be universal
- Get χ by equating Enskog EOS and virial expansion

$$\chi = 1 + 0.6250 b\rho + 0.2869 (b\rho)^2 + 0.115 (b\rho)^3 + \ldots$$

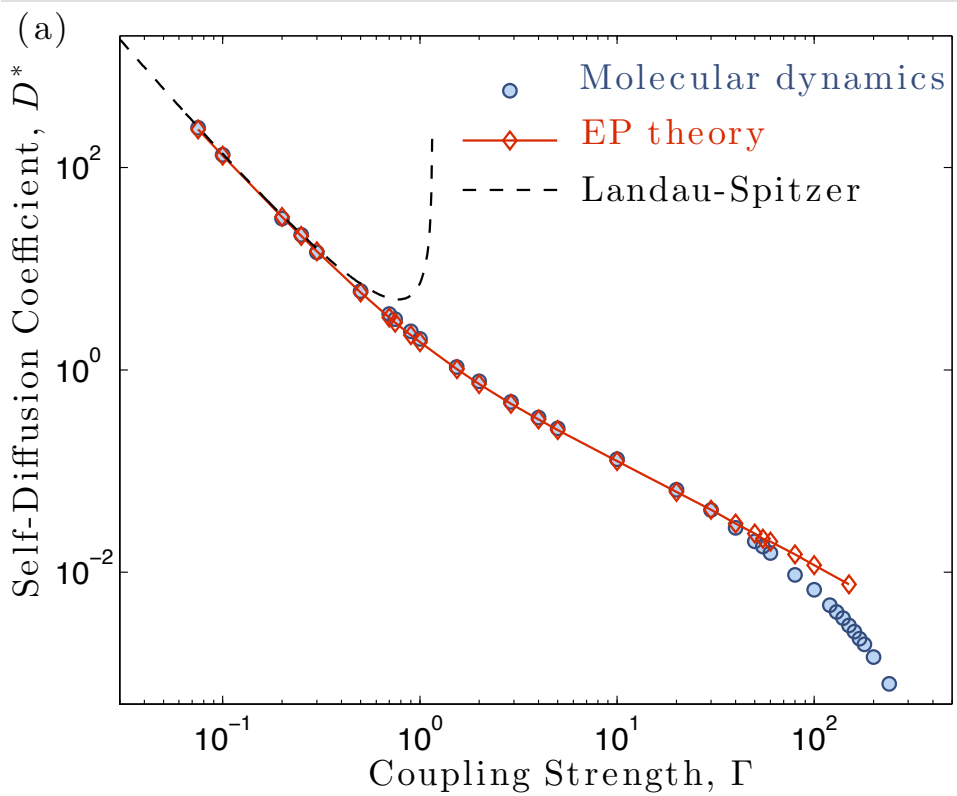
Co-Volume: $b\rho = \frac{2}{3}\pi n \sigma^3$

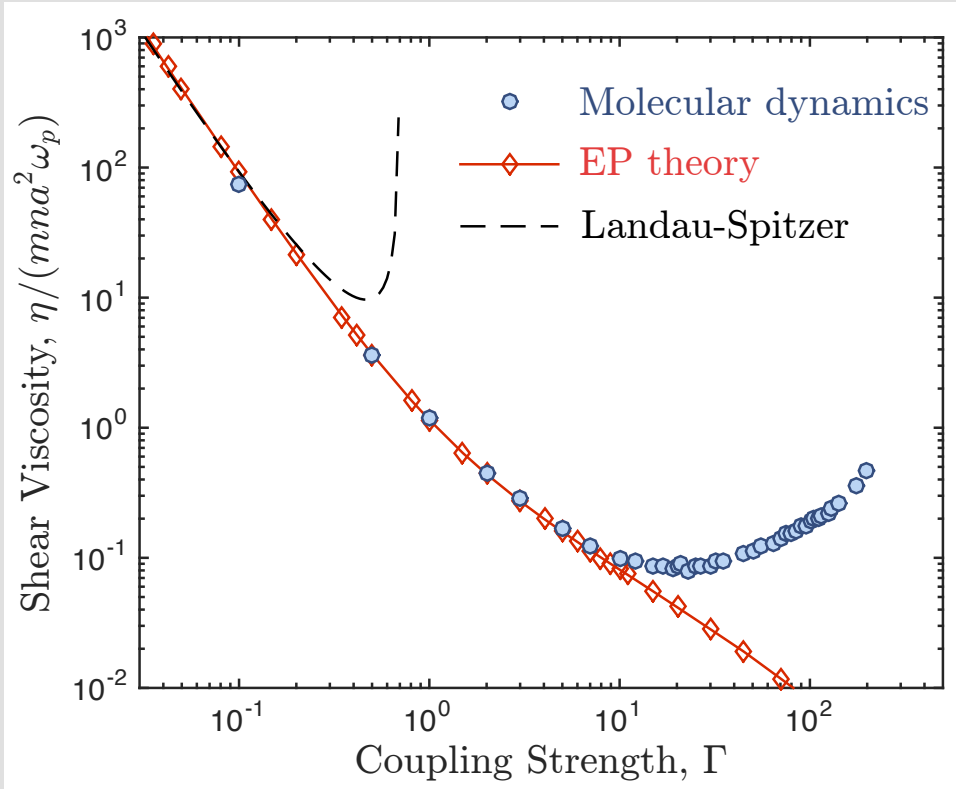
- Collision frequency multiplied by χ

The approximation is not perfect



Find excellent agreement with MD



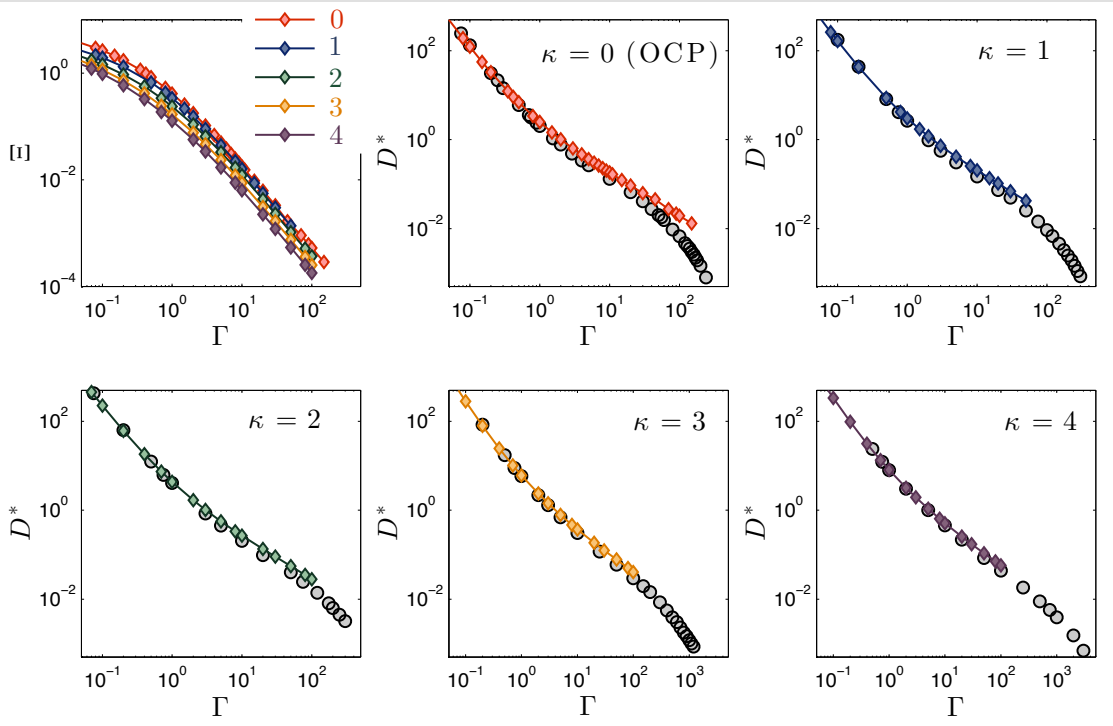


Baalrud and Daligault, PRE 91, 063107 (2015)

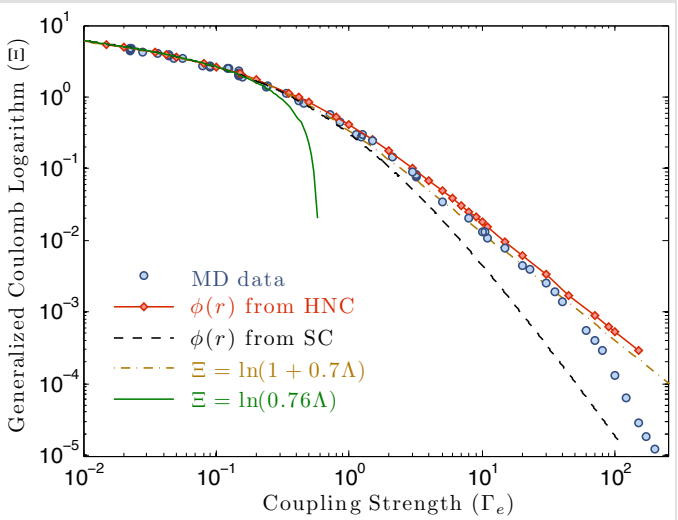
Daligault, Rasmussen and Baalrud, PRE 90, 033105 (2014)

Similar results for other systems

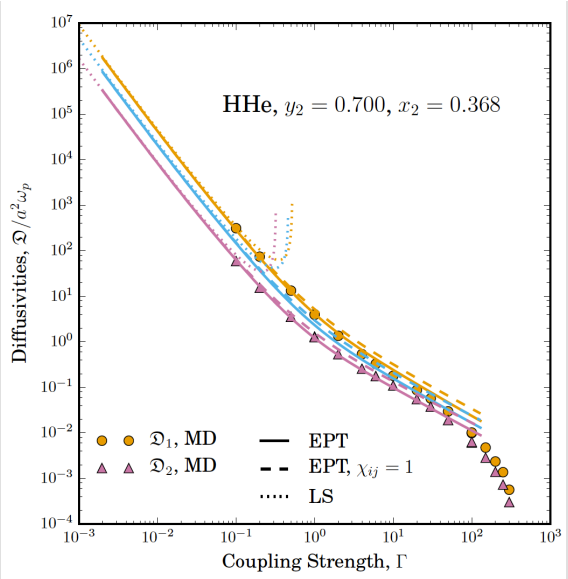
Yukawa OCP



Temperature relaxation



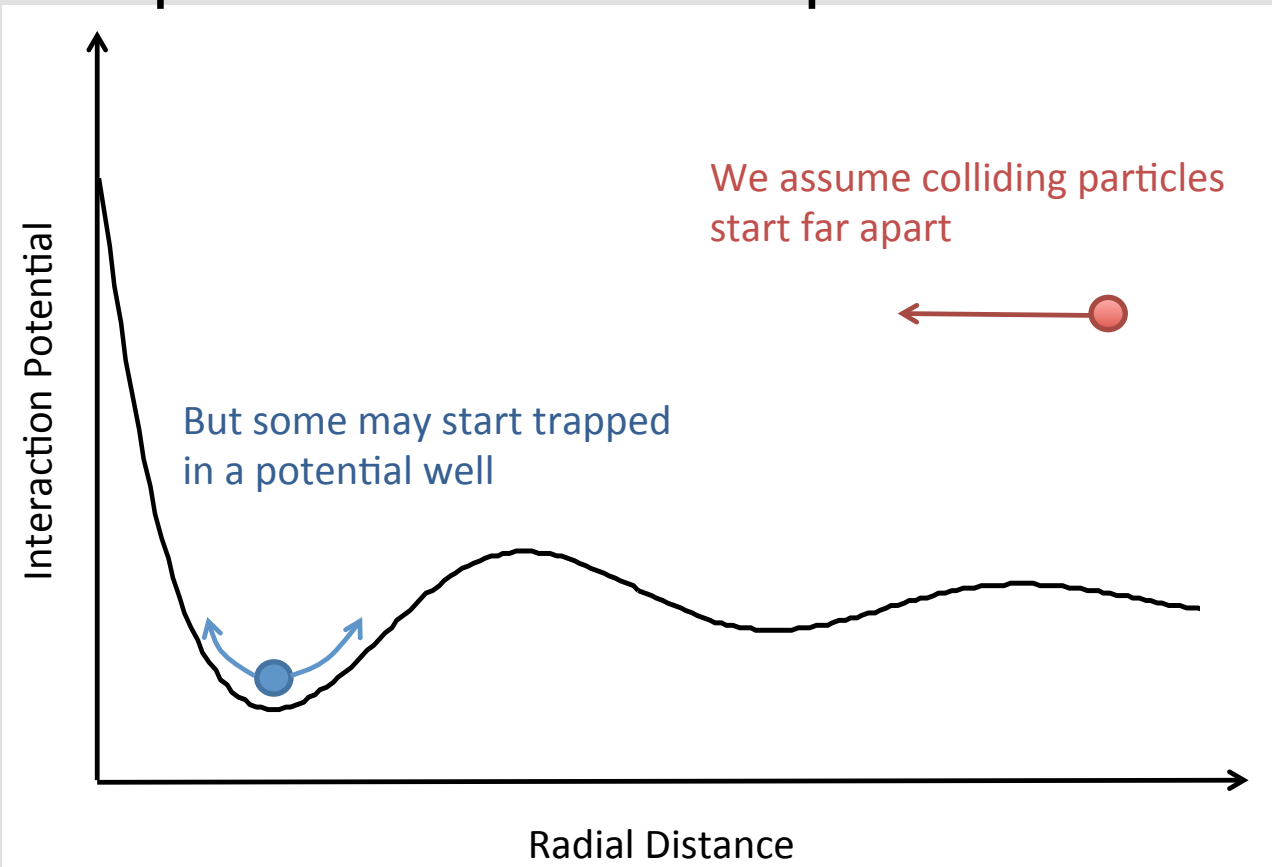
Mixtures:



...and several others

Limitations

- Caging
  - Particles get trapped in potential wells
  - Corresponds to onset of liquid-like behavior

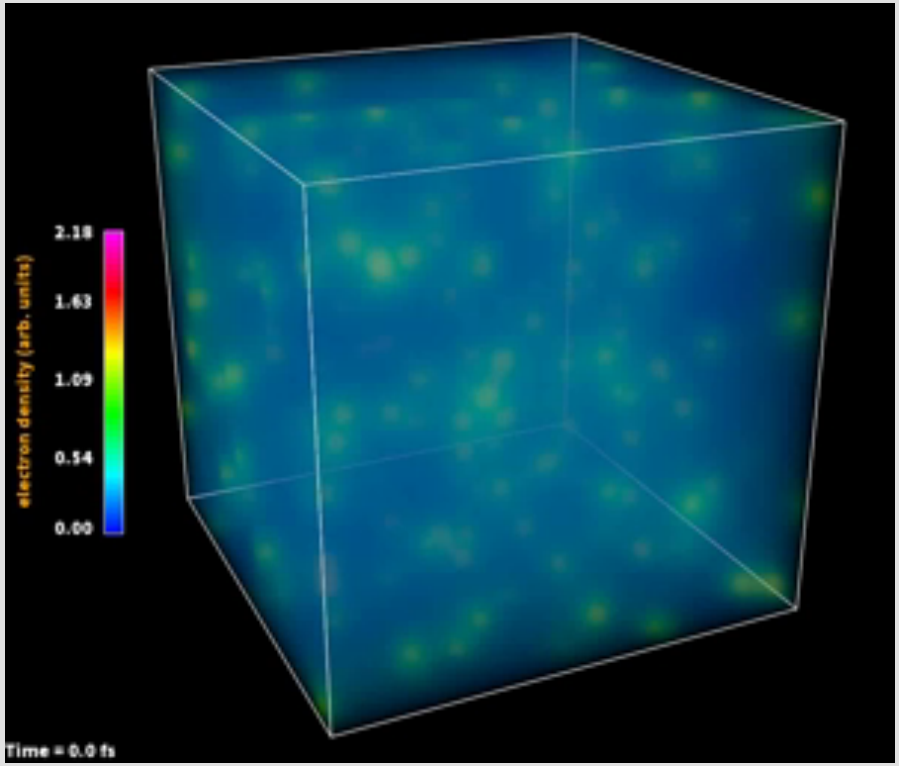


EPT for Warm Dense Matter

- So far, we have discussed simple electron models (OCP or YOCP)
- Ion transport in WDM
  - Electron behavior is not simple
  - Must account for degeneracy
  - Need $g_{ii}(r)$ as input to EPT
- First principles simulations
  - Quantum orbital free MD (QOFMD) (state of the art, but expensive)
  - Puedoatom MD (approximate, but accurate)

Quantum Orbital Free MD

electron density

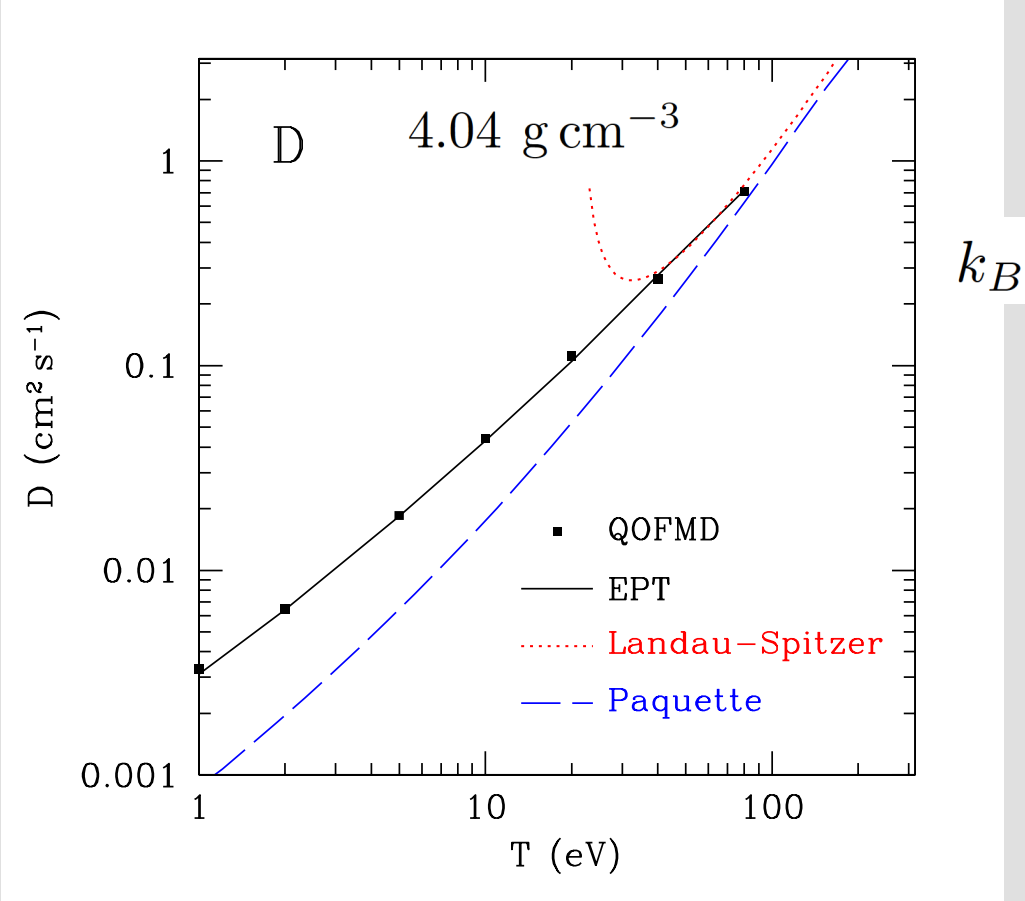


Deuterium at 10 g/cc and 10 eV

http://www.lanl.gov/projects/dense-plasma-theory/

Benchmark using QOFMD

- g(r) came from the QOFMD simulation



$$\Gamma_{\rm eff} = 9.5 \text{ to } 0.25$$

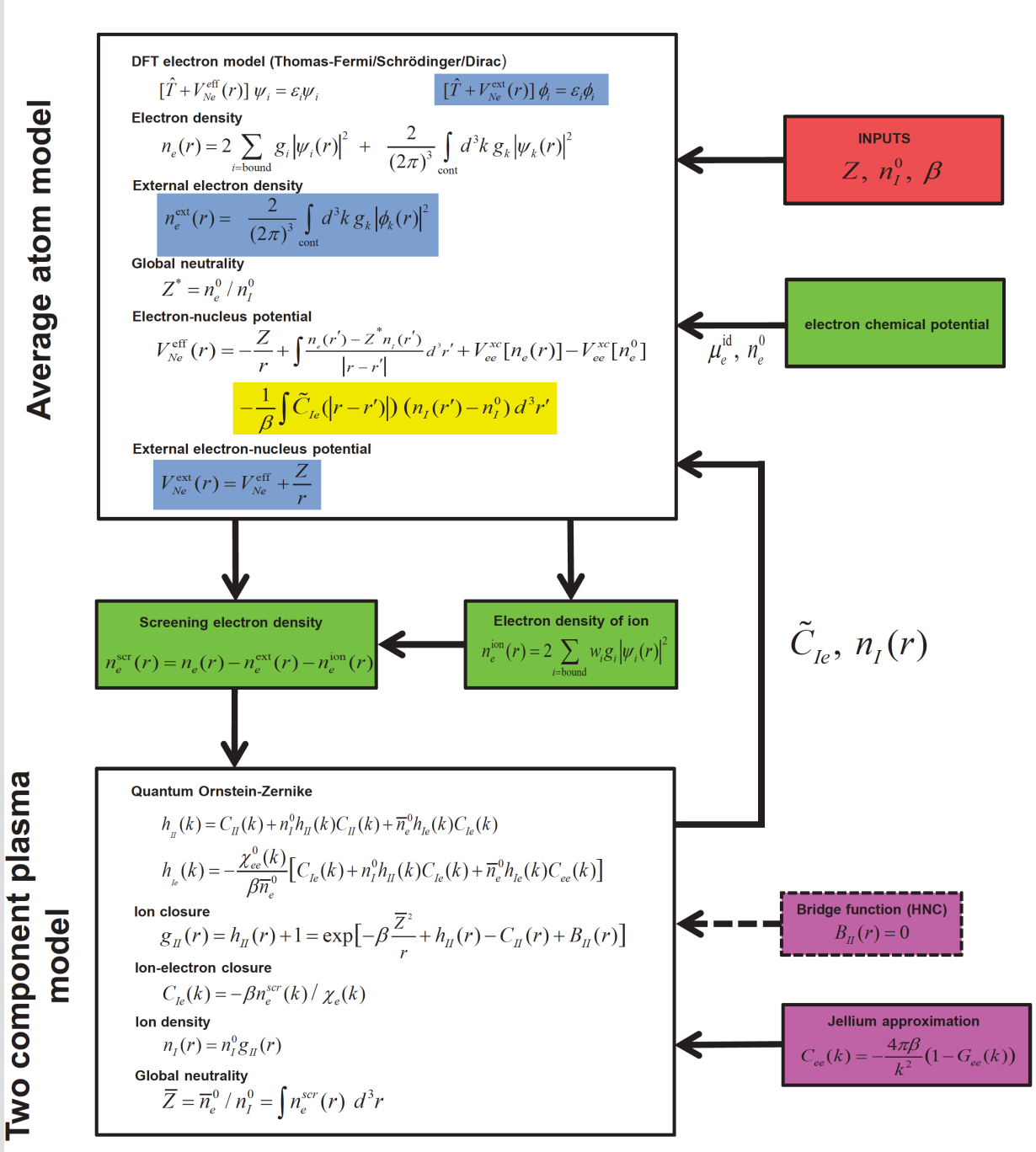
$$k_B T/E_F = 0.0024 \text{ to } 1.9,$$

Daligault, Baalrud, Starrett, Saumon, Sjostrom PRL 116, 075002 (2016)

Average Atom + HNC

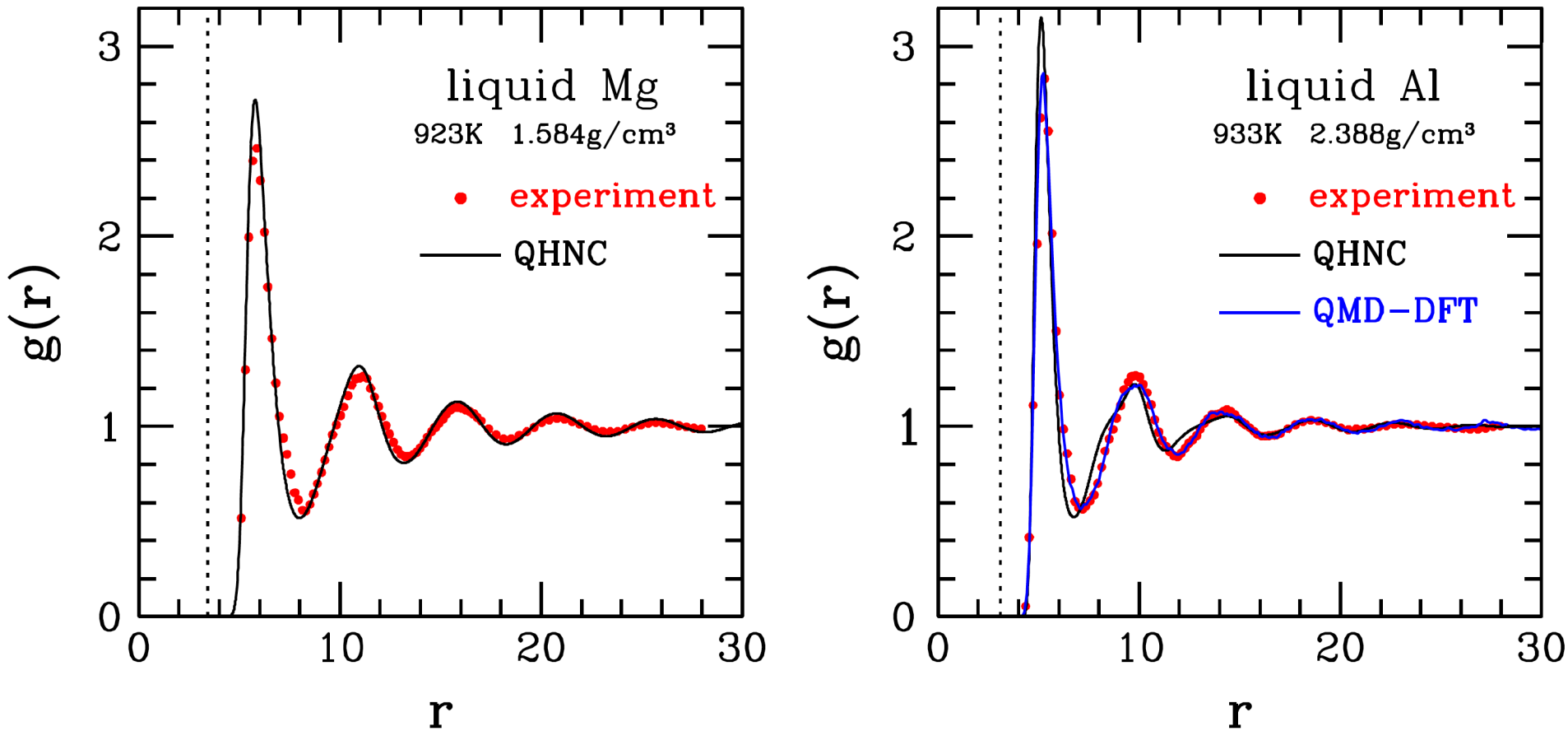
- Average atom model
  - Describes electron cloud around an ion of charge Z
  - Finite temperature Density Functional Theory
- Two-component plasma model
  - AA is coupled with the integral equation theory of fluids
  - Called the Orstein-Zernike Equation
- Result is a quantum HNC approximation

Starrett, Saumon PRE 85, 026403 (2012)



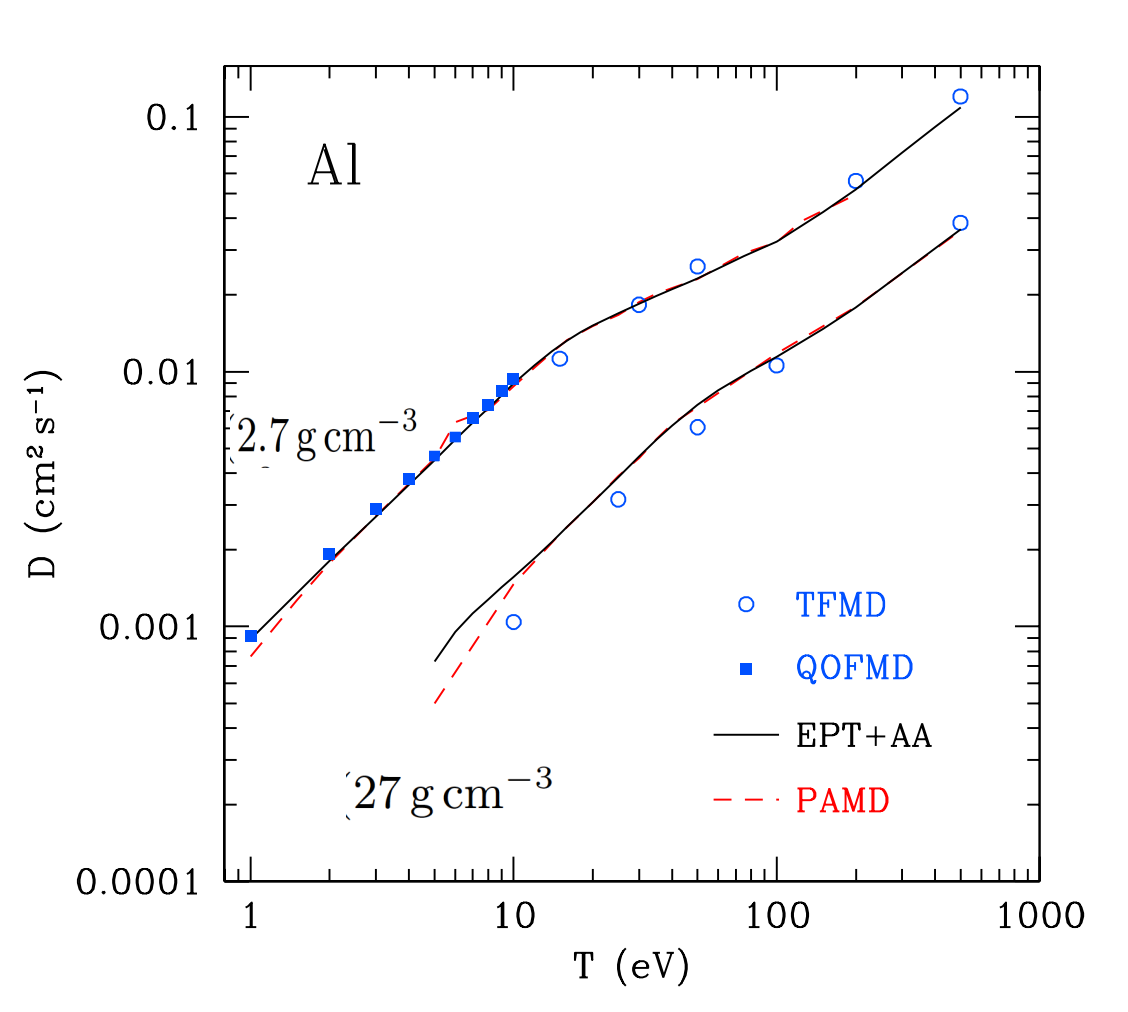
Saumon, Starrett, et al. in Frontiers and Challenges in WDM (2014)

Average Atom Two-Component Plasma (AA-TCP)



Saumon, Starrett, Kress, Clerouin, HEDP 8, 150 (2012)

Aluminum: HEDP experiments



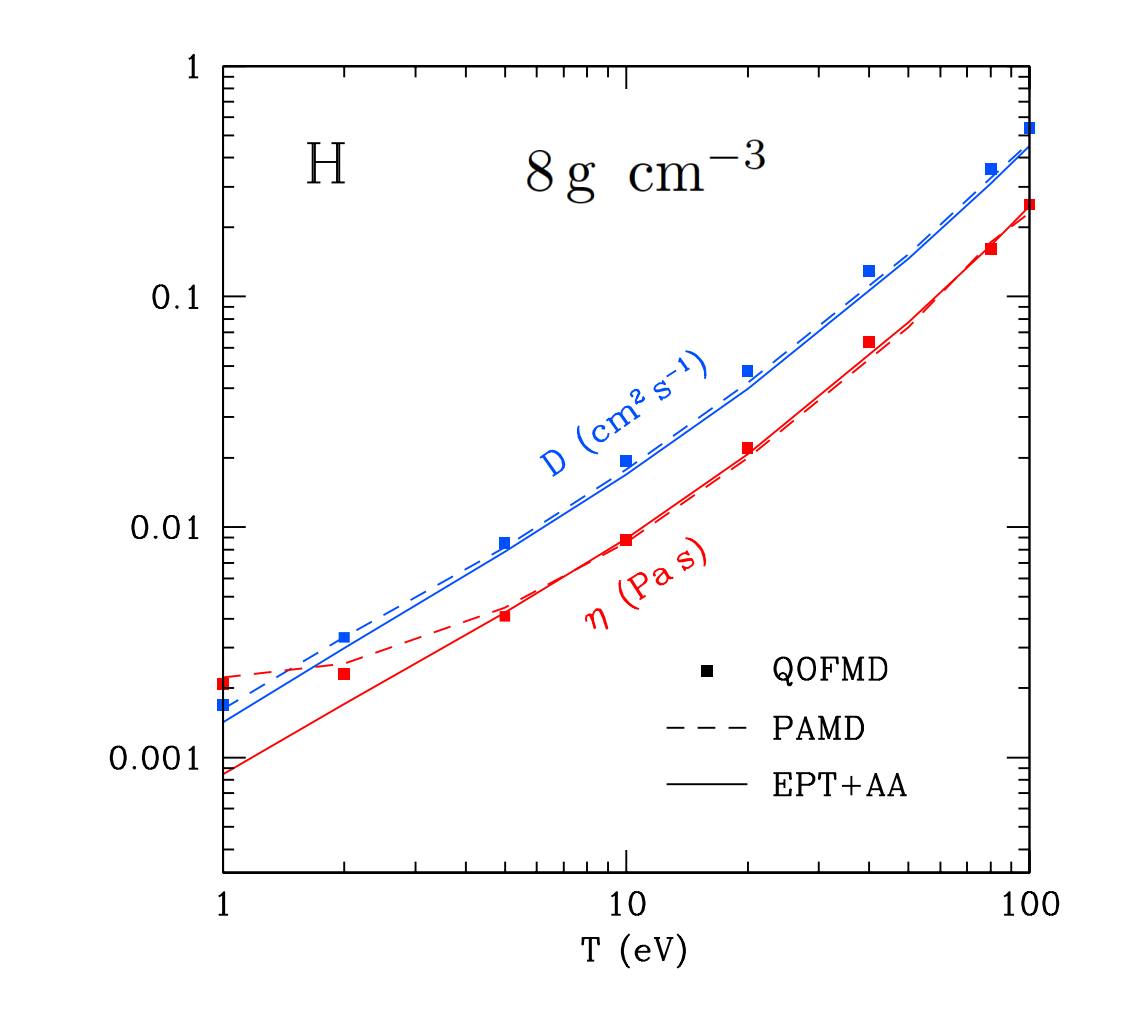
$$\bar{Z} = 3 \text{ to} \sim 12,$$

$$\Gamma_{\text{eff}} \sim 72 \text{ to} \sim 2.$$

Pressure ionization is important here

Daligault, Baalrud, Starrett, Saumon, Sjostrom PRL 116, 075002 (2016)

Hydrogen: ICF

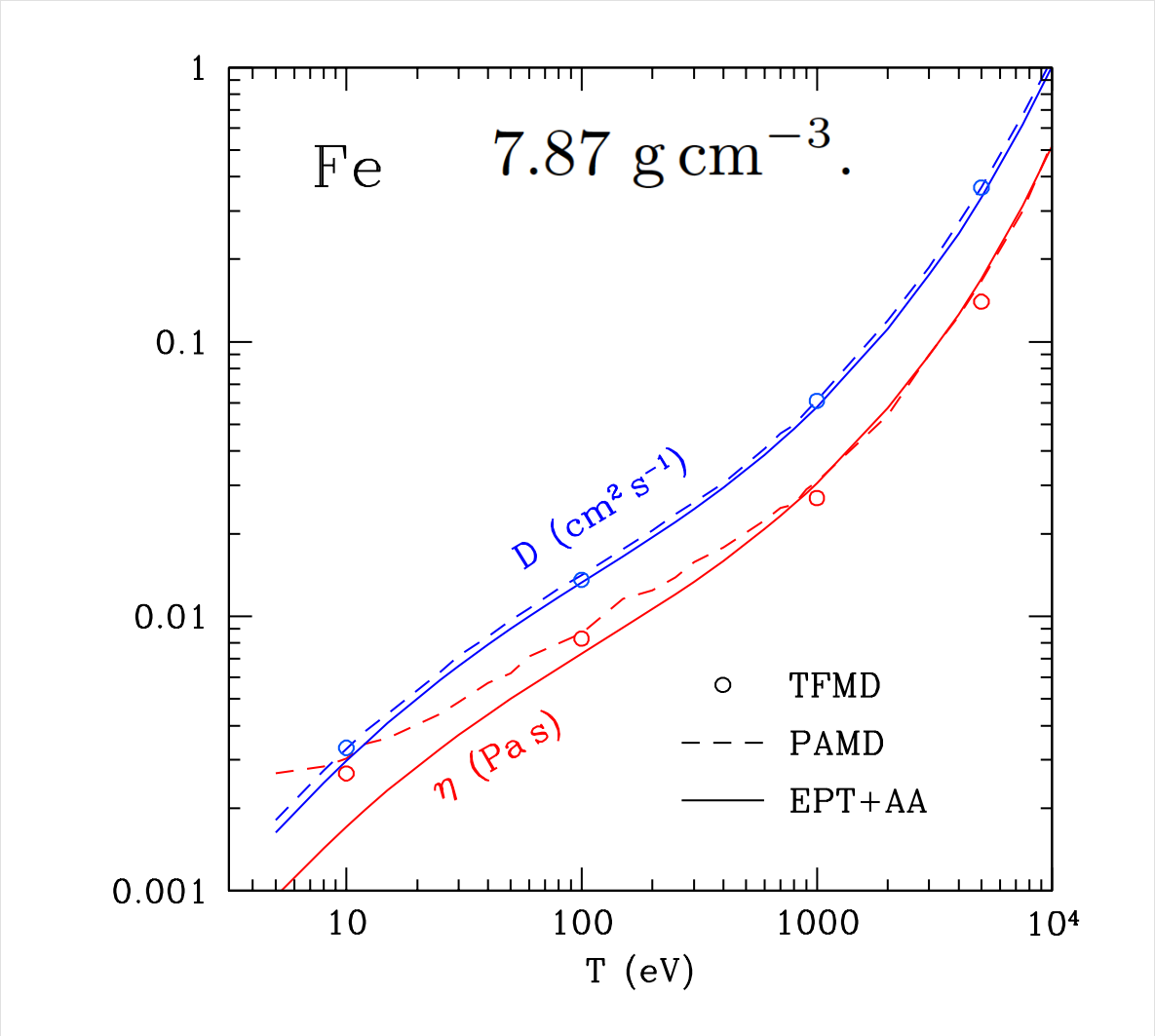


$$\Gamma_{\rm eff} = 21 \text{ to } 0.4,$$

$$k_B T / E_F = 0.01 \text{ to } 1.$$

Daligault, Baalrud, Starrett, Saumon, Sjostrom PRL 116, 075002 (2016)

Iron: Giant Planet Interiors



$$\bar{Z} = 8 \text{ to } \sim 18.4,$$

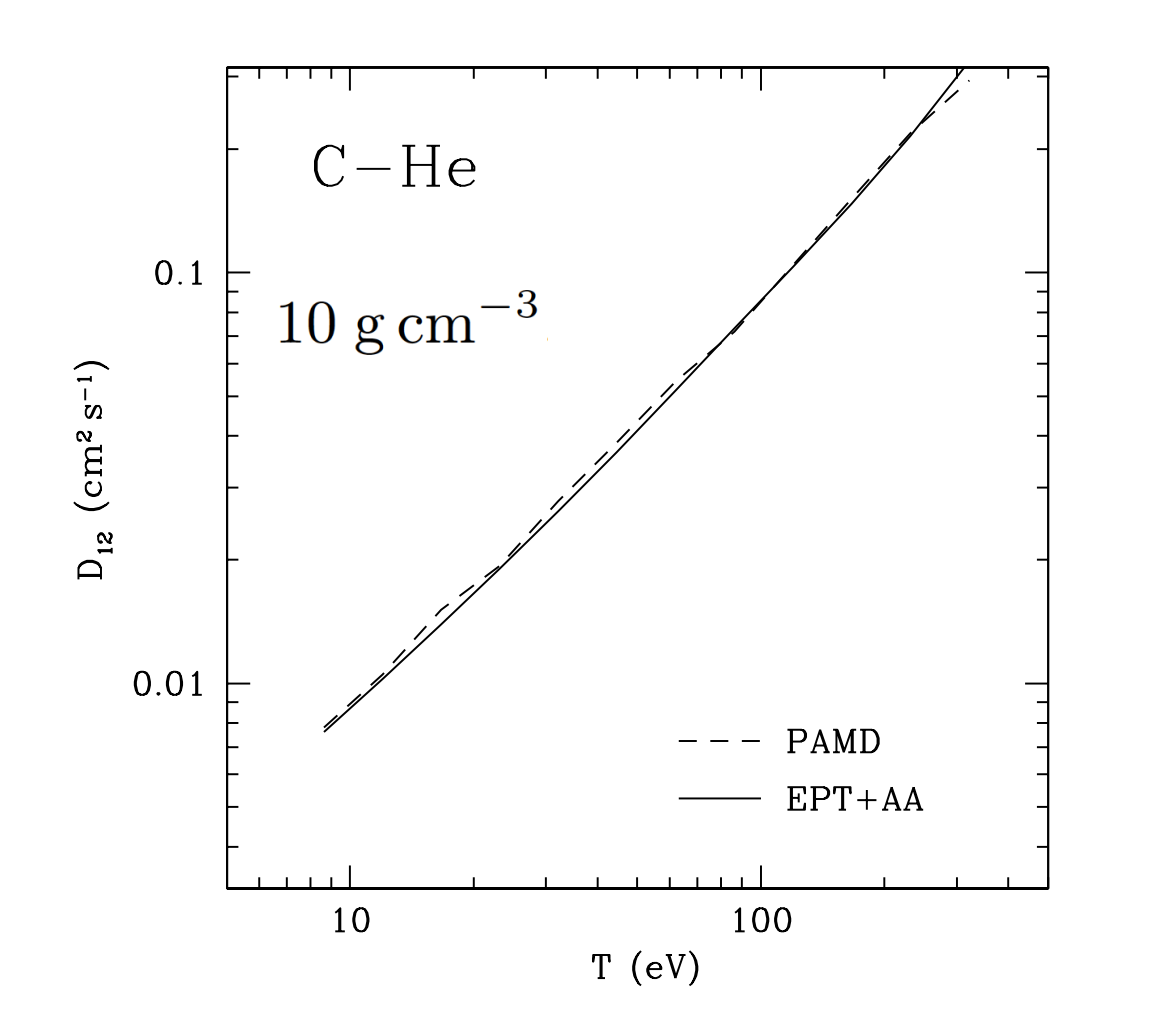
$$\Gamma_{\text{eff}} = 39 \text{ to } 0.6,$$

$$k_B T / E_F = 0.4 \text{ to } 200.$$

Daligault, Baalrud, Starrett, Saumon, Sjostrom PRL 116, 075002 (2016)

Carbon-Helium: White Dwarfs

Mutual diffusion coefficient



60% C, 40% He

Relevant to carbon "pollution" at the surface of white dwarf stars
(otherwise, pure He)

Daligault, Baalrud, Starrett, Saumon, Sjostrom PRL 116, 075002 (2016)

Computational Cost

- A few minutes on your PC (per point)
  - Would take hundreds of processor hours with MD
  - Many regimes are simply unattainable with DFT (or any QMD)
- Building tables becomes feasible

Future directions

- Electron transport
  - Use the quantum Boltzmann equation
  - Diffraction in addition to degeneracy
- Liquid-like regime
  - Promising avenues for caging models
- Magnetized plasmas
  - Gyroradius smaller than interparticle spacing
- g(r) from experimental data

Summary

- EPT is efficient and accurate
  - Very low computational cost
  - Accurate for $\Gamma<30$ (up to liquid-like state)
- First practical theory for computing transport coefficients in WDM
  - Ionic transport
  - Can build tables for real plasmas