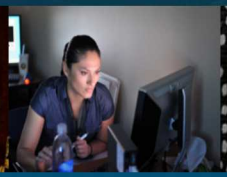
Sandia National Laboratories

SAND2019-9862PE

Effective Pair-Potentials for Modelling High Energy-Density Matter



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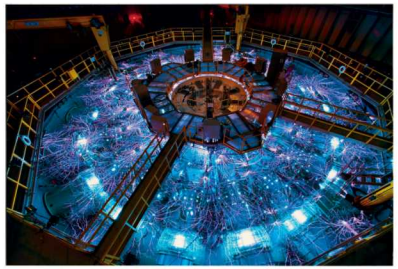
Plasma Interdiffusion

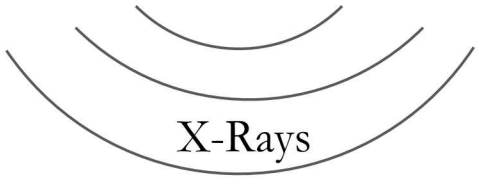
- Quantify interfacial mixing in ICF experiments
- Assess accuracy of transport models needed to design ICF experiments

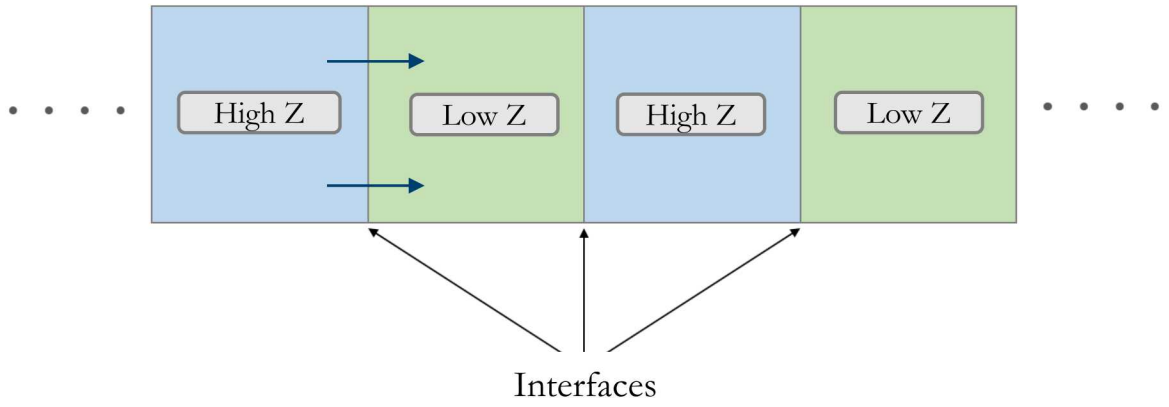
Experimental Set-up



Focus of this work

- Specifics:
  - High Z: Vanadium
  - Low Z: CH Foam





Inertial Confinement Fusion (ICF): Physical Regimes

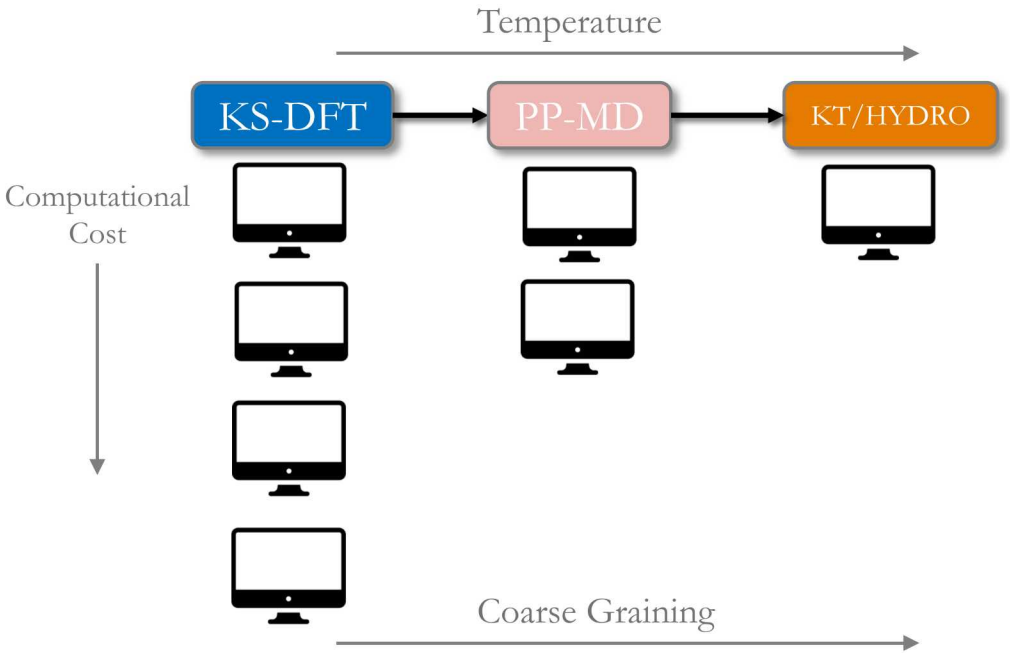
ICF experiments explore very disparate regimes

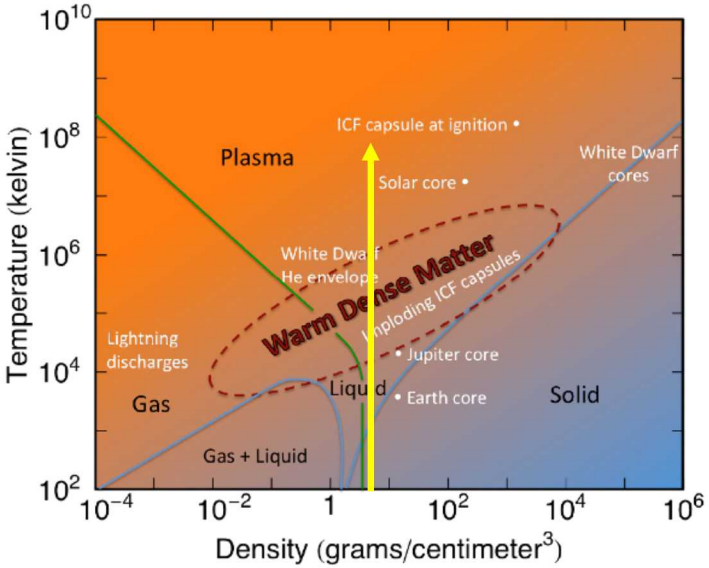
Solids

- Ions are very strongly coupled, electrons are extremely degenerate
- Density Functional Theory (DFT) – uses on-the-fly N-body potential

- **Warm Dense Matter (WDM)**
  - Ions are strongly coupled, electrons are partially degenerate
  - Pair-potential molecular dynamics (PP-MD) – pre-computed pair potentials

- **Hot Plasma**
  - Ions weakly coupled, electrons no longer degenerate
  - Solve a kinetic equation - McBGK





$1\ \text{eV} \approx 1.16\times10^4 K$

Modeling Techniques for WDM

- The **Yukawa (linear OF-DFT) model** is a general purpose widely used pair potential across plasmas physics. The following approximations are made:
  1. The bound electrons have no physical extent which shrinks bound cloud to a point. This gives us the effective ionization $< Z >$
  2. Considers a linearization of remaining free electrons density. This gives a spherical electron screening cloud with radius λ_{TF}

- **Kohn Sham DFT (KS-DFT)** includes richer QM effects and is a higher fidelity method
  - Solving the Kohn Sham equations gives the electron density

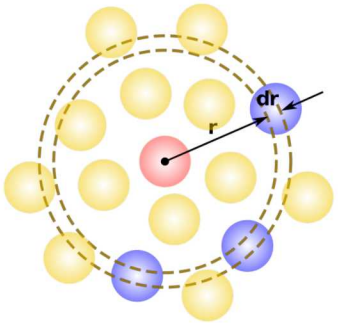
You can use either of the above methods in a molecular dynamics code to evolve the ion species

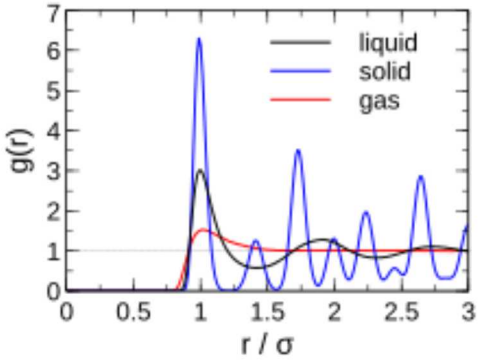
- Yukawa PP-MD uses **screened** Coulomb potential for ion-ion interactions
  - Sarkas – developed at MSU
- KS-DFT-MD uses **bare** Coulomb potential for ion-ion interactions
  - Vienna ab initio Simulation Package (VASP)

What are the macroscopic properties, like self-diffusion, that each model predicts?

Validating Results: Correlation Functions

- **Radial Distribution Function (RDF):** Spatial correlation function
  - Helps identify the physical state of the system

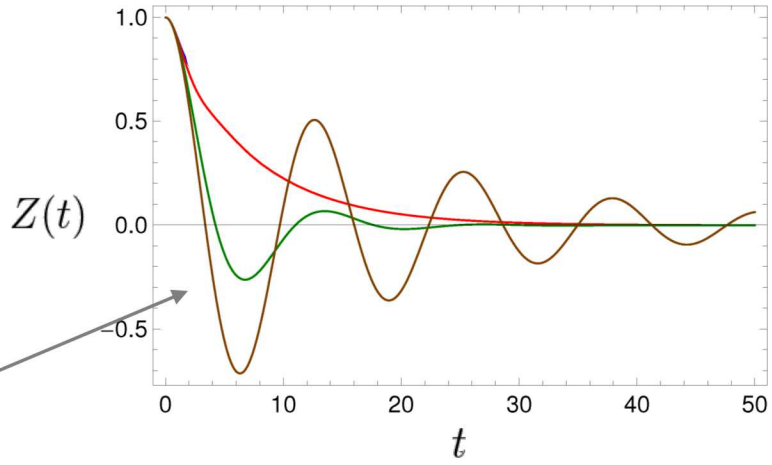




- **Velocity-Autocorrelation Function (VACF):** Time correlation function
  - Give us information on individual particle motion

Normalized VACF:

$$Z(t) = \frac{\langle \mathbf{v}(t) \cdot \mathbf{v}(0) \rangle}{\langle \mathbf{v}(0) \cdot \mathbf{v}(0) \rangle}$$



"Caging" or "backscattering" of particles

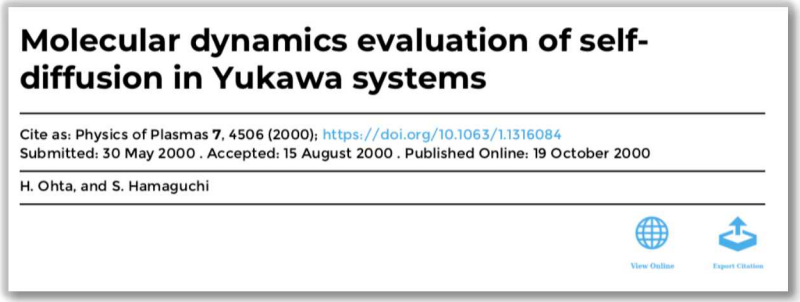
Macroscopic Comparisons: Transport

- **Macroscopic transport coefficients:** Self-diffusion
  - How far a particle moves over time as it moves from regions of high concentration to low concentration

Two examples of how to compute the self-diffusion coefficient

- From the VACF: $D = \frac{1}{3}\int_0^\infty dt\, \langle \mathbf{v}(t) \cdot \mathbf{v}(0) \rangle$
- From Einstein's Relation: $D = \lim_{t\to\infty} \frac{|\mathbf{r}_i(t) - \mathbf{r}_i(0)|^2}{6t}$

Can compare these expressions with those obtained from the **Stanton and Murillo** analytic theory



Molecular dynamics evaluation of self-diffusion in Yukawa systems

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H. Ohta, and S. Hamaguchi

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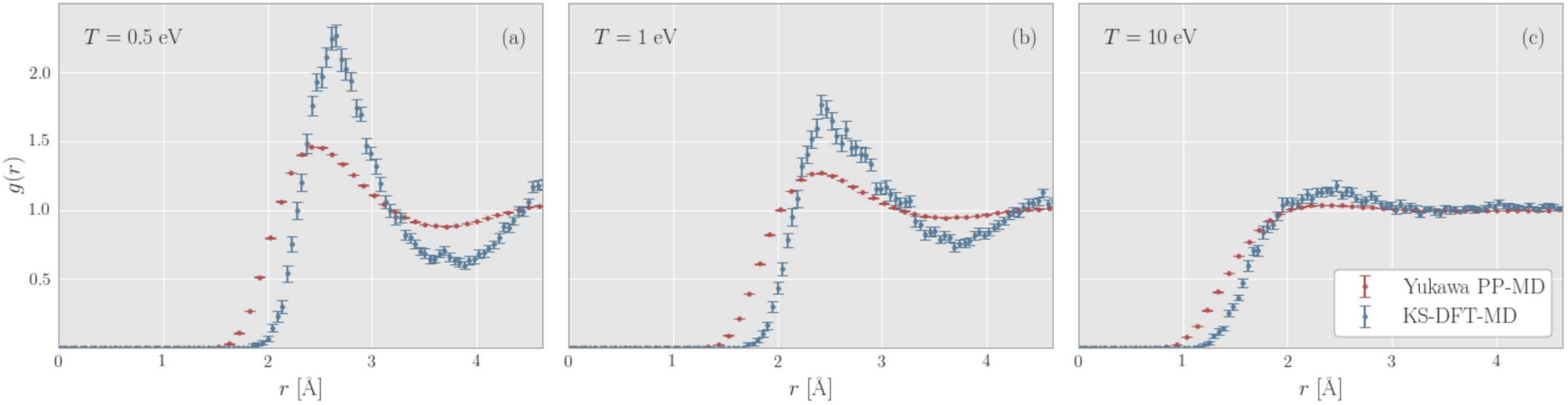
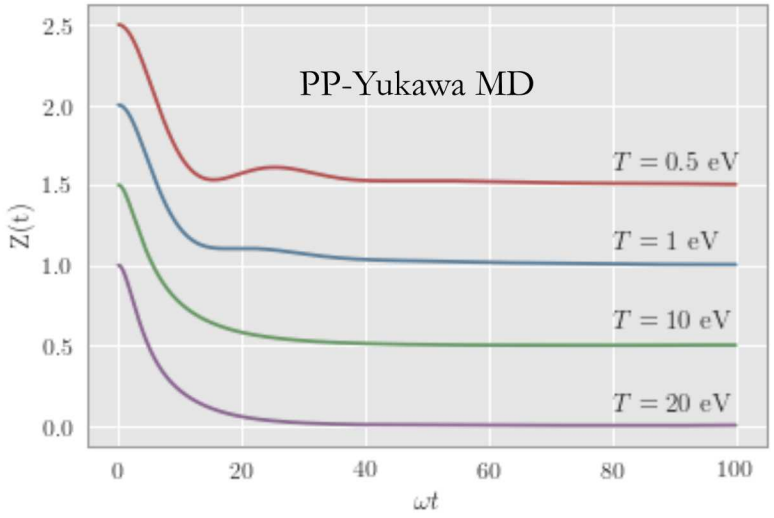
Value of D compared from two expressions above – only slight difference in values

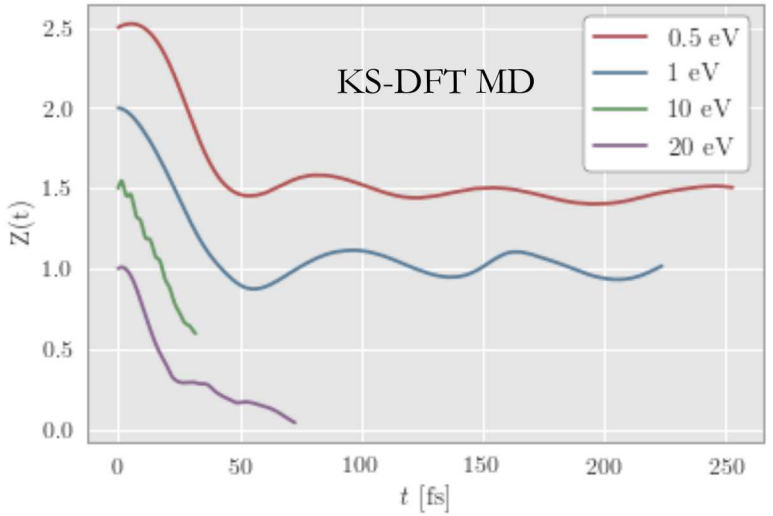
Initial Comparisons for Vanadium: KS-DFT-MD and PP-MD

Comparison of RDFs:



Comparison of VACFs:





Why Do We Need a Pair-Potential?

- **Goal:** Can we avoid using KS-DFT to determine the electronic structure by generating the "best" pair potential for a pair potential MD simulation?

> **Best** = pair-potential that incorporates as much information from KS-DFT as possible

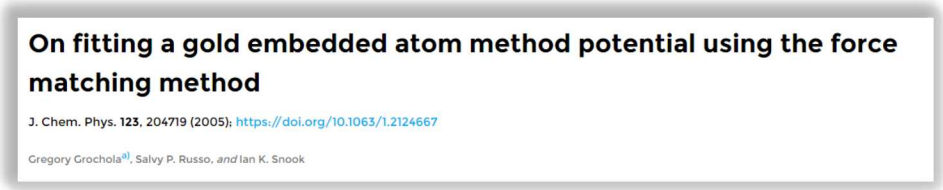
- Why do we care?

Temp [ev]	PP-MD CPU Hrs	KS-DFT-MD CPU Hrs
5	5.7e-1	1.20042e+4
10	4.6e-1	1.08590e+4
20	4.8e-1	5.80032e+3
	CPUs: 1 Particles: 500 Timesteps: 30K	CPUs: 36 Particles: 54 Timesteps: <=12K

- Simulations that can take multiple weeks across many CPUs can be reduced to minutes on a single CPU.
- This information can be used to inform transport coefficients in a hydro model.

Generating Effective Pair Potentials from Force Matching

- **Main idea of FM**: Take KS-DFT-MD data (e.g. positions, forces, energies, stresses, etc.) and minimize a cost function to fit a pair-potential.
- To perform the force matching. We make use of the open-source software **potfit**



- The target function to be minimized can be defined as

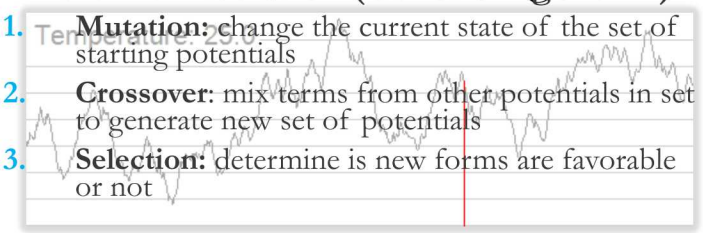
$$Z(\xi) = \sum_{k=1}^{3N} u_k (F_k(\xi) - F_k^0)^2 + \sum_{l=1}^{M} w_E (E_l(\xi) - E_l^0)^2 + \sum_{l=1}^{6M} w_S (S_l(\xi) - S_l^0)^2$$

Force Components Energy Components Stress Components

- The optimization routines in **potfit** include:
  - Simulated annealing
  - Differential evolution

  High-level optimization
  - Least-squares ← Low-level optimization

- **Differential Evolution (Genetic Algorithm)**



Analytic Potential Forms for Force Matching

- We use the VASP data to fit the analytic forms of the following potentials
  1. Vanilla **Yukawa**

$$u^{Y}(r) = \frac{Z^2 e^2}{r} e^{-r/\lambda_{TF}}$$

parameters to fit: **2**

  2. Yukawa with a **short-range repulsion** term (SRR Yukawa)

$$u^{Y+SRR}(r) = \frac{a}{r^4} + \frac{Z^2 e^2}{r} e^{-r/\lambda_{TF}}$$

parameters to fit: **3**

  3. The **exact gradient-corrected** potential (EGS)

$$u^{EGS}_{\text{mon}}(r) = \frac{Z^2 e^2}{2r}[(1+\alpha)e^{-r/\lambda_-} + (1-\alpha)e^{-r/\lambda_+}]$$

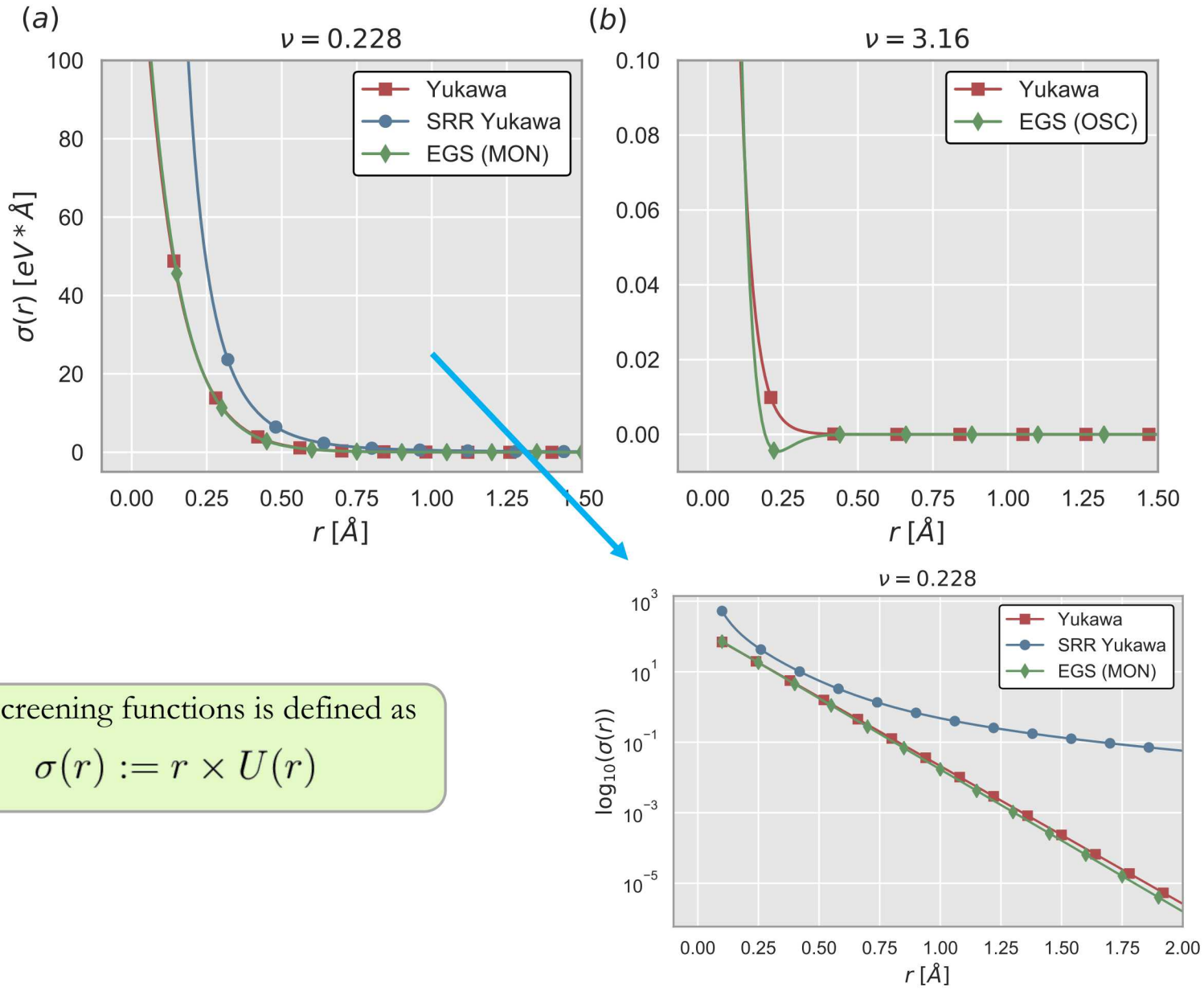
$$u^{EGS}_{\text{osc}}(r) = \frac{Z^2 e^2}{2r}[\cos(r/\gamma_-) + \alpha'\cos(r/\gamma_-)]e^{-r/\gamma_+}$$

parameters to fit: **4**

$$\nu = \frac{3\sqrt{8\beta}}{\pi}\lambda\mathcal{I}'_{-1/2}(\bar{\eta})$$

gradient correction strength parameter:
When $\nu > 1$, the oscillatory form of EGS is used

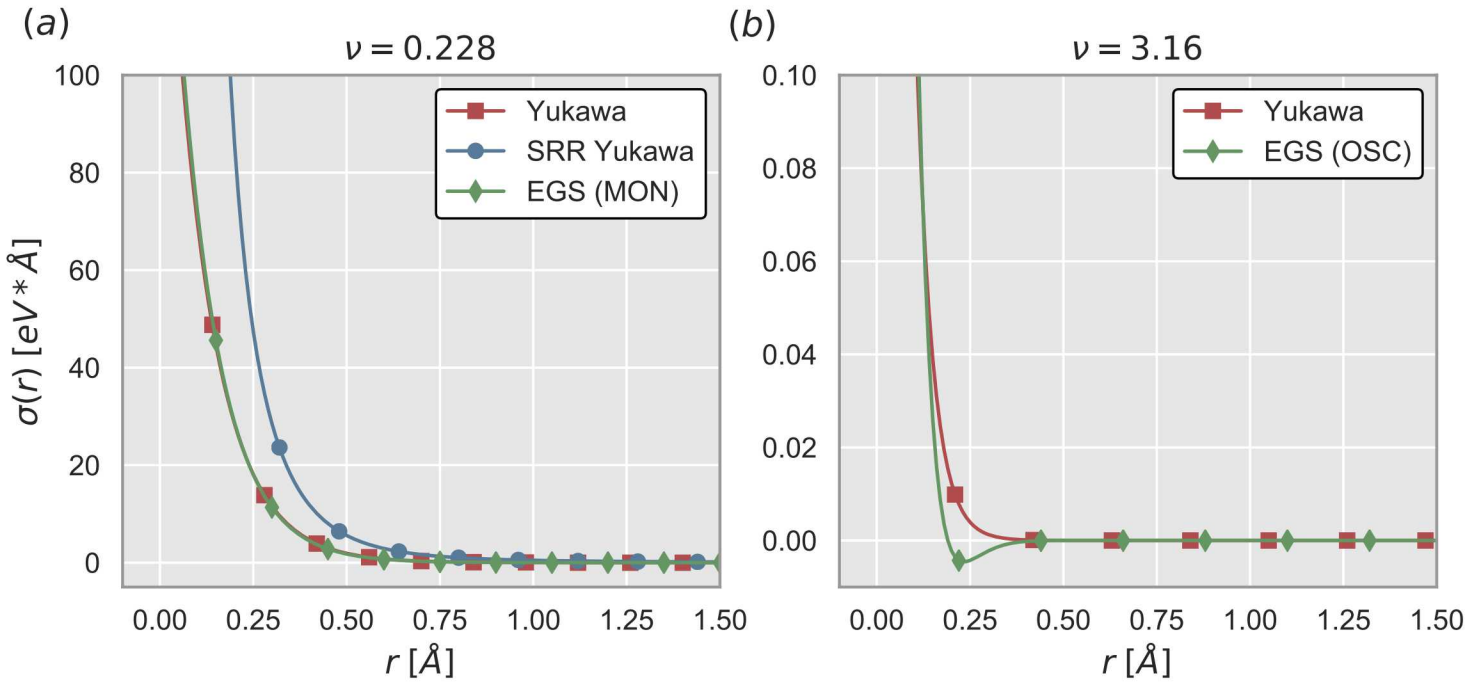
Comparison of Screening Functions



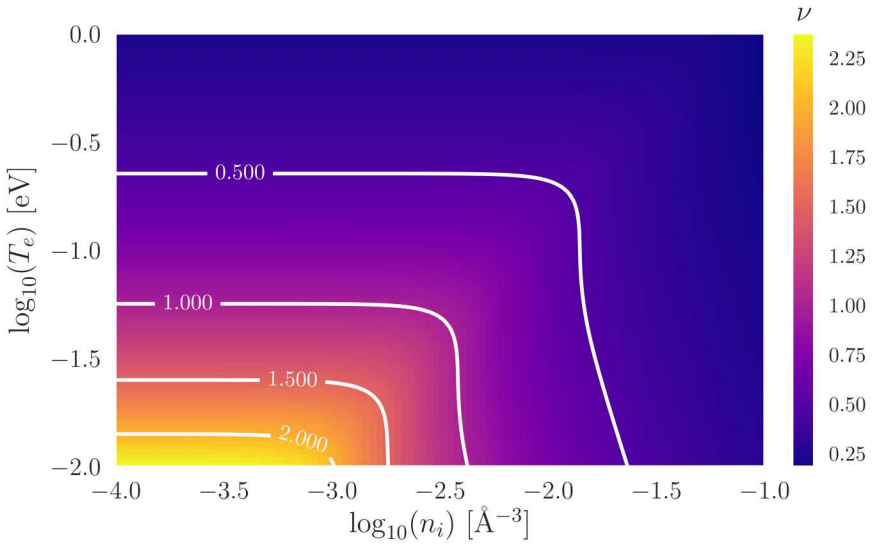
The screening functions is defined as

$$\sigma(r) := r \times U(r)$$

Comparison of Screening Functions



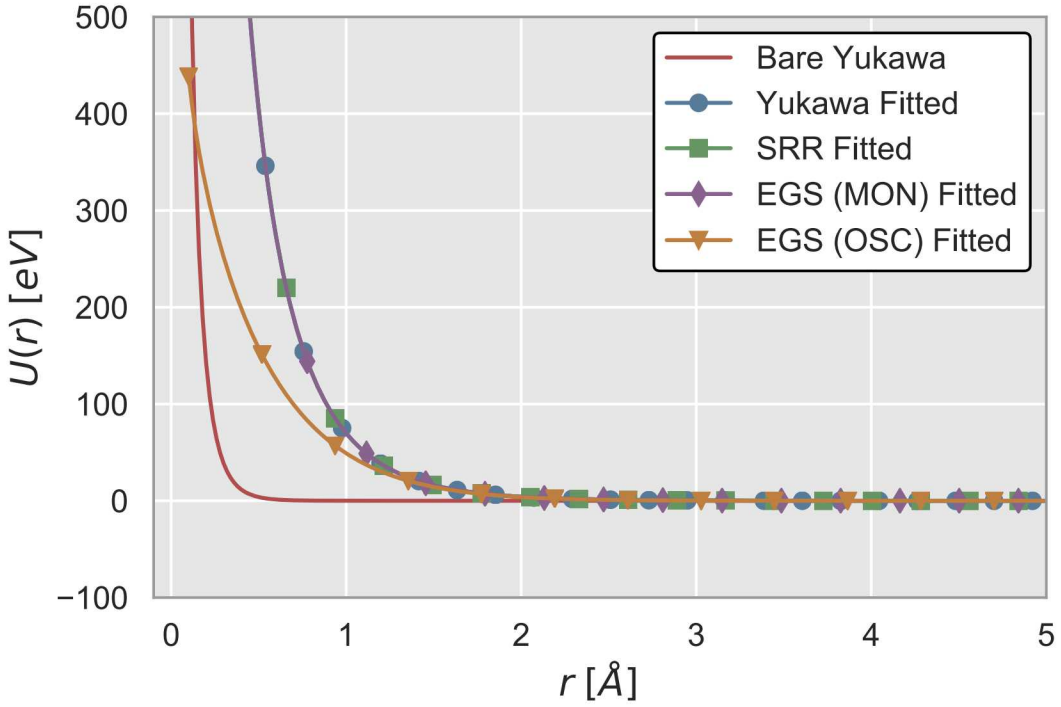
The heatmap to the right shows the regions when the oscillatory behavior from the EGS potential is expected ($\nu > 1$)



Force Matched Potentials: Fitted Analytic Forms

- Optimization parameters:
  - Algorithm: Genetic Algorithm
  - Number of configurations from VASP: ~400

Results for 0.5 eV VASP data



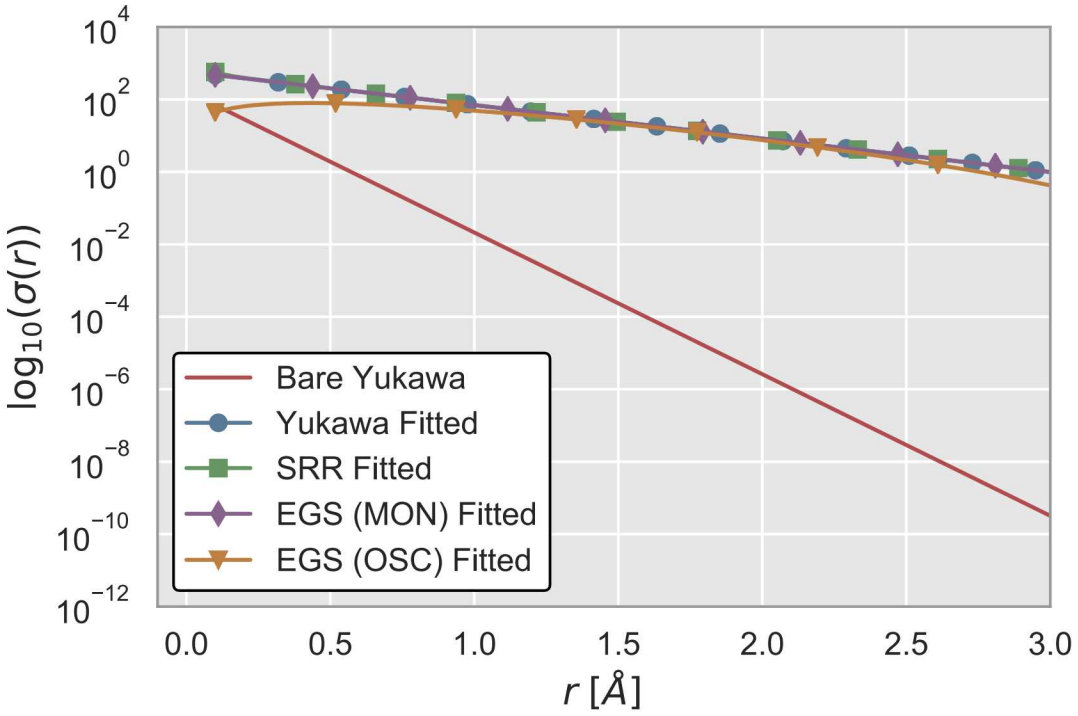
The **VASP** configurations are used as input to **potfit** and the objective function is minimized for the analytic forms on the previous slides

Can obtain effective ionization and screening lengths from these fitted potentials

Screening Functions for Force Matched Analytic Potentials

- Optimization parameters:
  - Algorithm: Genetic Algorithm
  - Number of configurations from VASP: ~400

Results for 0.5 eV VASP data

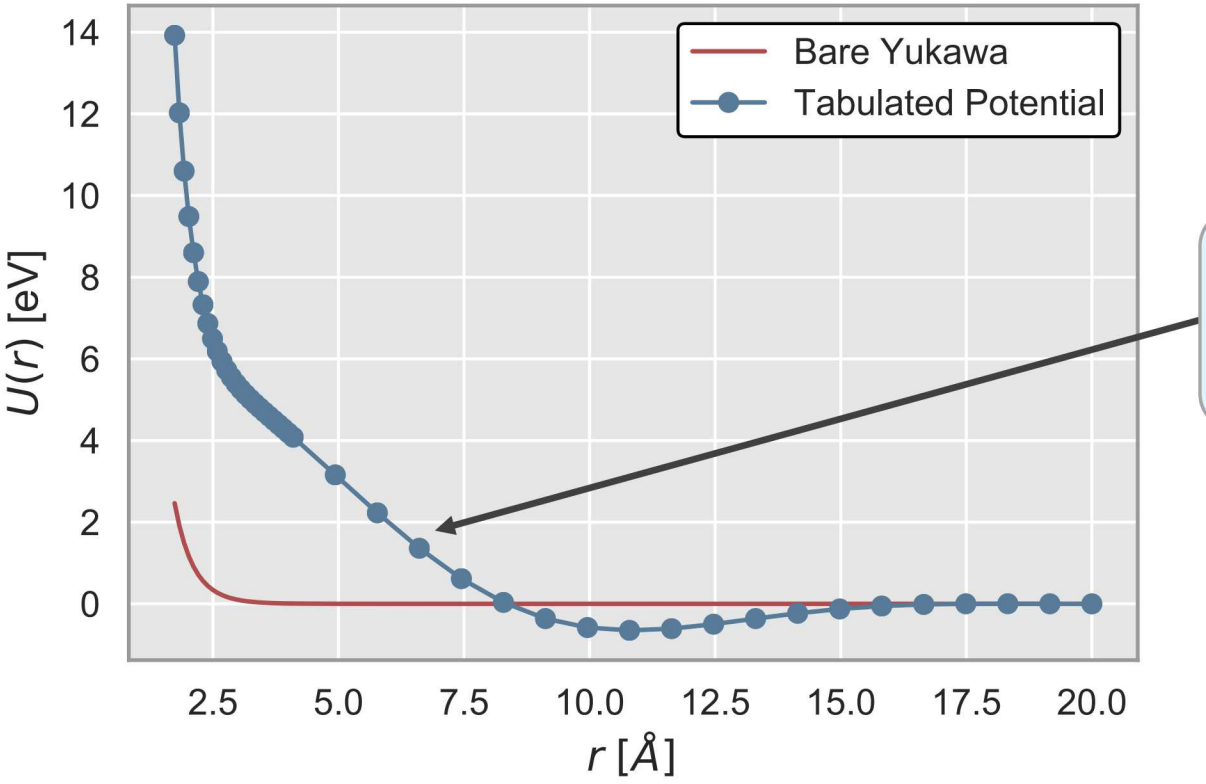


Even at 3 Angstroms, forces non-negligible relative to **bare Yukawa** potential

Force Matched Potentials: Tabulated Form*

- Tabulated potentials: A series of splines fit to VASP data

Pros: No restriction on the functional form of the potential
Cons: Number of nodes/knots results in a very high number of parameters to optimize



Each point on the blue curve is a parameter to optimize (45 parameter optimization problem!)

*Preliminary results

Next Steps and Future Work

- Compare RDFs, VACFs, and self-diffusion coefficients for fitted potentials
- Finalize tabulated potential work
  - Lots of options to play around with here, potfit metric for accuracy is ambiguous
- Obtain data for more temperature and densities to determine transferability to other materials (e.g. Al, Be, Fe)