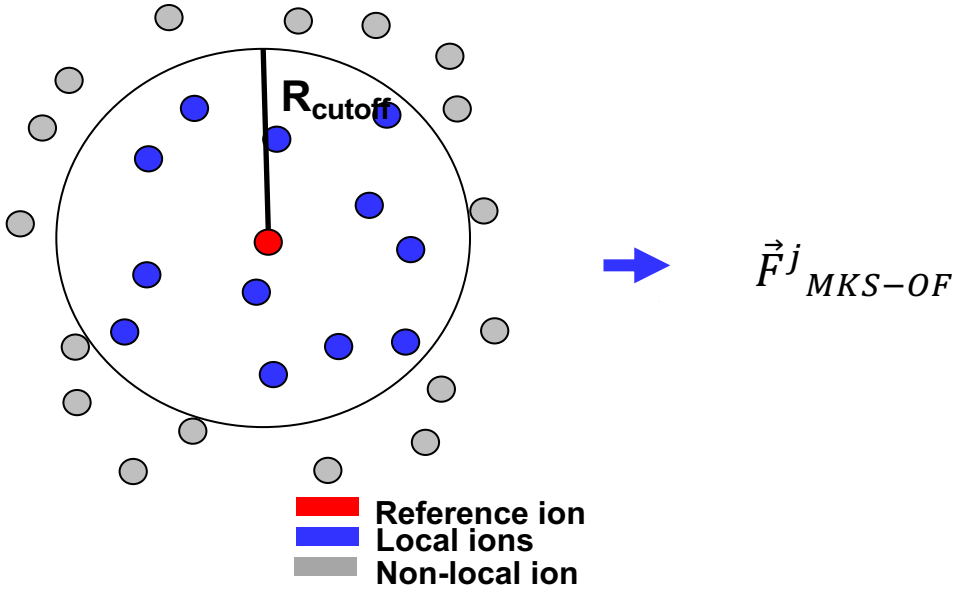
A Machine-Learned Orbital-Free Force-Correction Model: Extending the Thermodynamic Range of Affordable Kohn-Sham Level Accuracy



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Summary

A neural network-based machine learning model has been constructed to predict force differences between Kohn-Sham and orbital-free density functional theory.

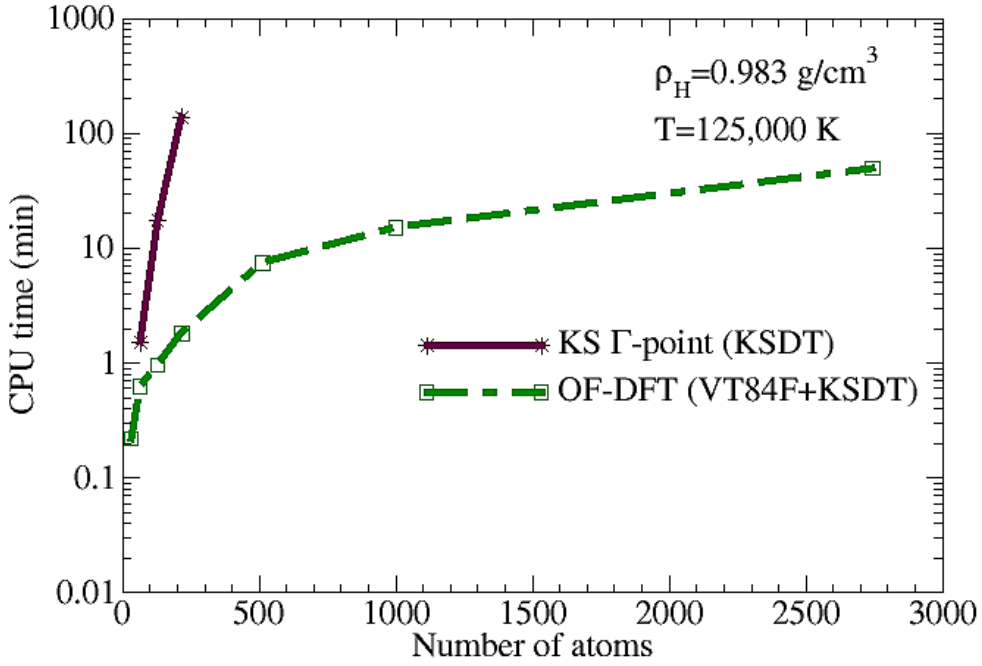
- The goal of the model is to address the trade off between accuracy and computational cost associated with the use of density functional theory (DFT) in *ab initio* molecular dynamic simulations.

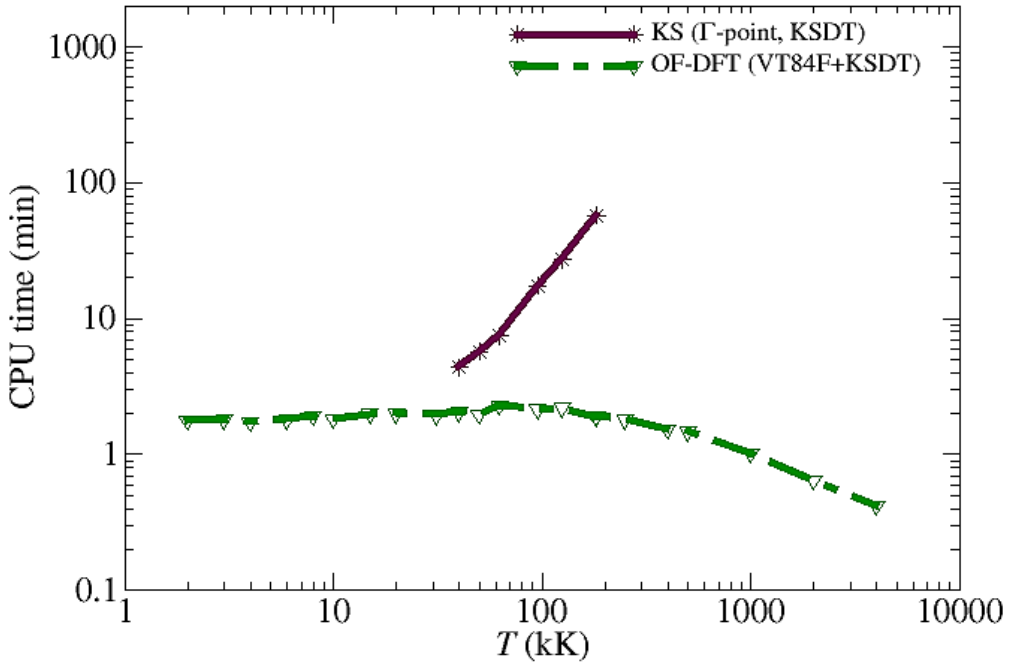
- With the use of descriptor vectors, the local ion configuration is used to predict the difference in ionic forces from Mermin-Kohn-Sham (MKS) and orbital free (OF) DFT.

- Initial tests of the model on warm dense fluid hydrogen have been performed demonstrating a significant improvement on the computational cost associated with each molecular dynamic step.

The force correction model shows encouraging results, but further work is required to improve the accuracy and validate the reliability of the model.

Ab initio molecular dynamic simulations require balancing the desired accuracy with the computational cost.

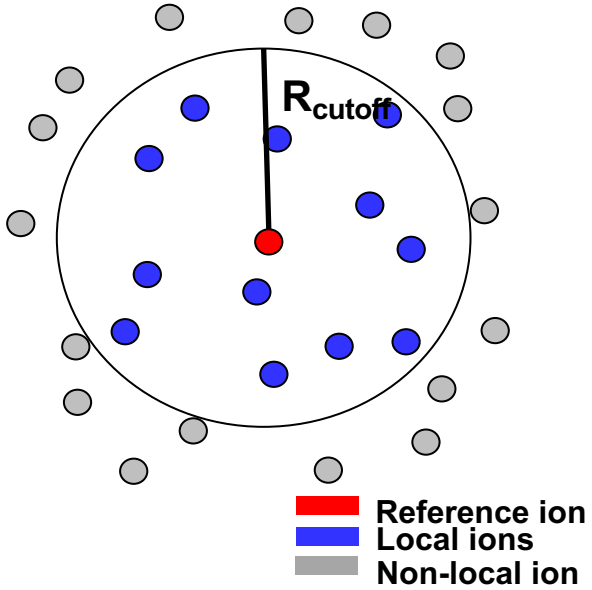




MKS DFT is limited to systems of a few hundred atoms and temperatures below 100 kK.

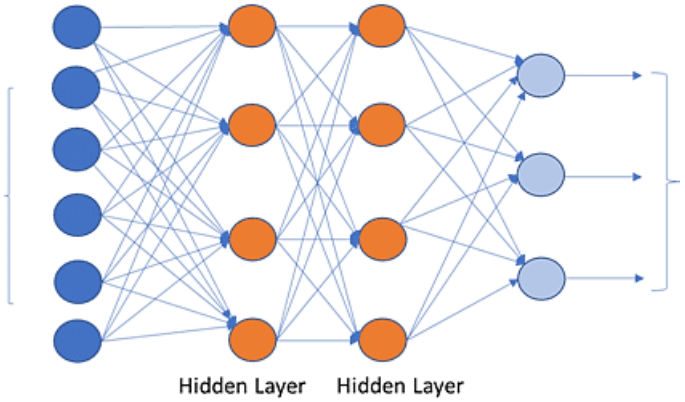
V. V. Karasiev, *et al*, Comput. Phys. Comm. 185, 3240, 2014.

For each ion, a coordinate system based on the local surrounding ion configuration is constructed.



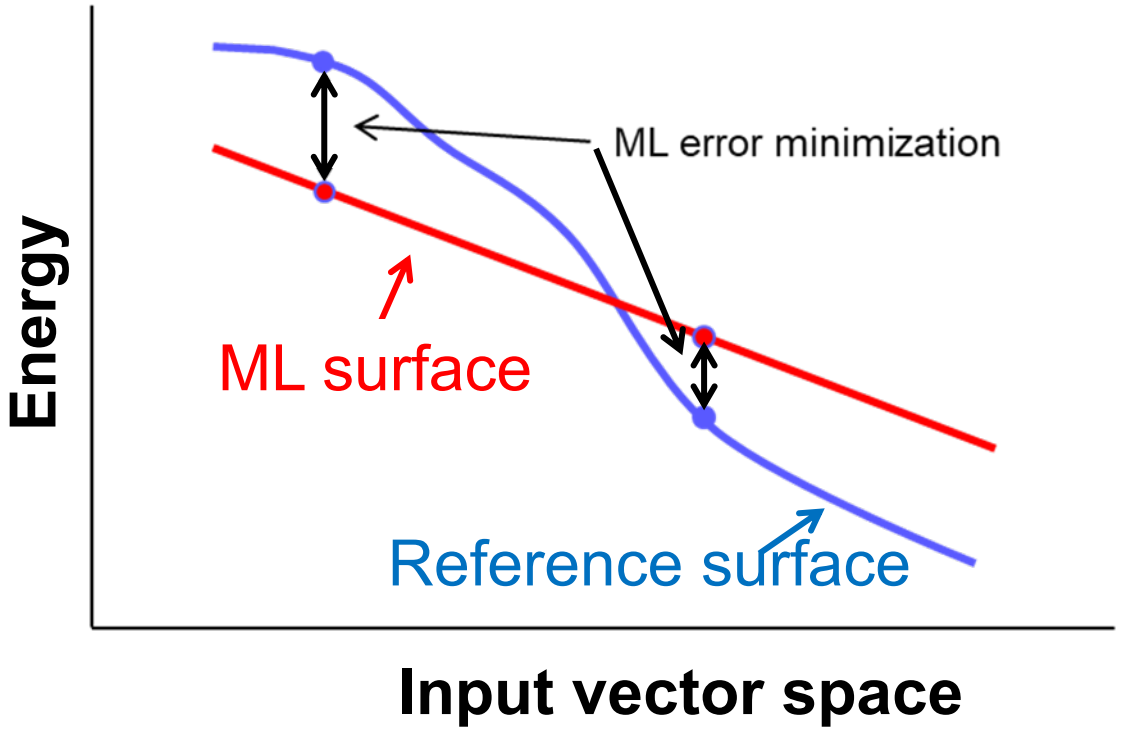
- It is assumed only the local ion configuration contributes to the force on any given ion.
- A set of descriptor vectors are constructed to describe the local ion configuration.

$$\vec{V}_i^{\,j} = \sum_{q}^{Neighbors} \frac{\vec{r}_q^{\,j}}{r_q^{\,j}} \exp\left(-\left(\frac{r_q^{\,j}}{\tau_i}\right)^{p_i}\right) \rightarrow \boldsymbol{X}^j{}_{i,k} = \frac{\vec{V}_i^{\,j} \cdot \vec{V}_k^{\,j}}{\left\|\vec{V}_k^{\,j}\right\|} \rightarrow \vec{F}^j{}_{MKS-OF}$$



Z. Li, et al, Phys. Rev. Lett. 114, 096405, 2015.

The model is built on ionic forces directly to provide a more reliable prediction.

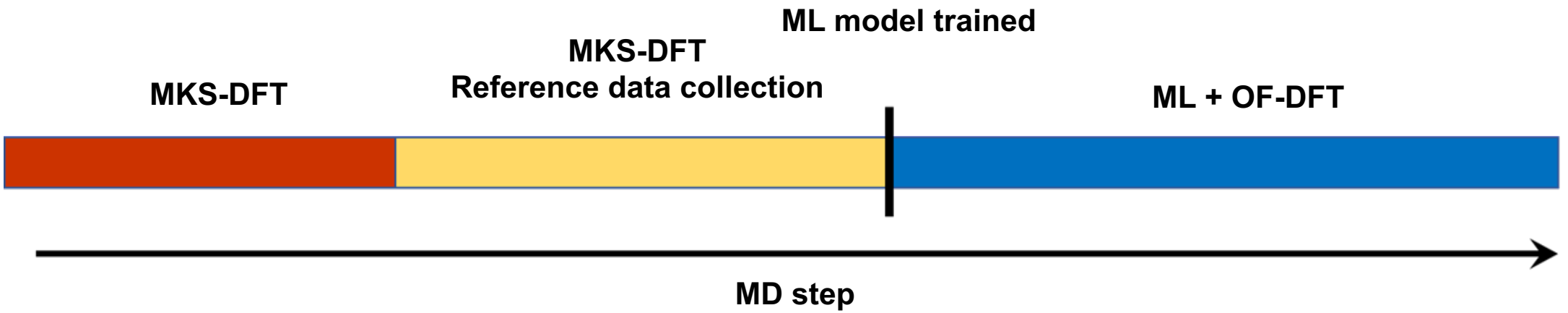


- **ML in its simplest form represents an interpolation of a surface of interest.**
- **The average distance between the ML and reference surface are minimized at a select number of points.**
- **The accuracy of The ML model may not translate well to its derivatives.**

The reference data is collected, and the model is trained at a fixed set of thermodynamic conditions.

- With every ML problem there is also an important and typically unknown distribution on the sample space.
- $P(\{\vec{R}_i\}, \{\vec{F}_i\}) = P(\{\vec{R}_i\})P(\{\vec{F}_i\}|\{\vec{R}_i\})$

- In MKS DFT the electron ground state depends on the system temperature.
- A fixed ion configuration from H at 1.0 g/cm^3 has a median change of 24% in the components of force for T =1000 K $\rightarrow$ T = 1200 K



The force correction model is benchmarked on warm dense fluid hydrogen.

MD simulation parameters

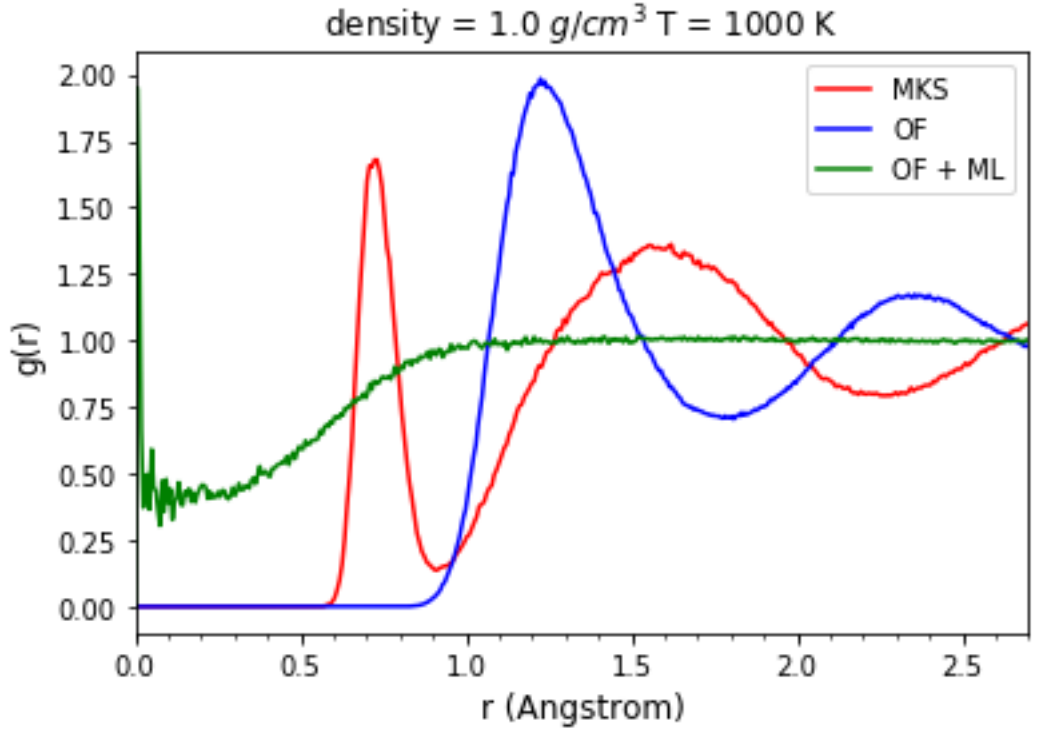
- 256 atoms in a cubic supercell
- 5000 MD steps
- NVT ensemble
- Γ point sampling
- F_{XC} = SCAN-L
- F_S, convex combination LKT and TF

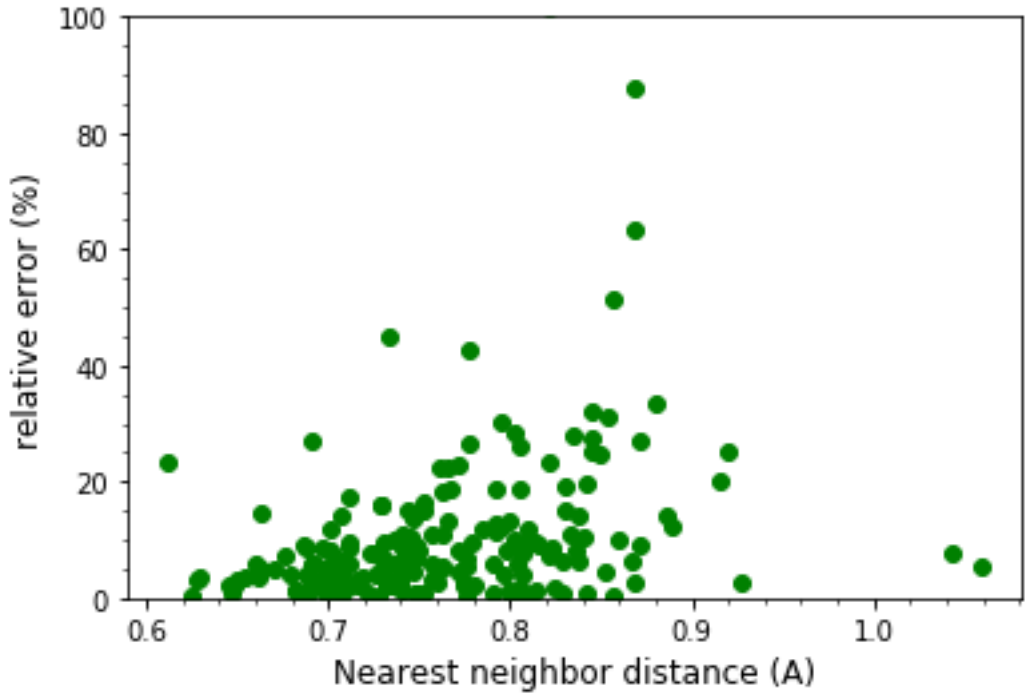
ML parameters

- Activation function: Hyperbolic tangent
- 20 x 20 Neural Network
- Learning rate = 0.5
- 1000 epochs
- Training set: 3000 vectors
- Validation set: 400 Vectors
- Test Set: 400 Vectors

K. Luo, *et al*, Phys. Rev. B, 98, 041111®, 2018. J. Sun, *et* al, Phys. Rev. Lett. 115, 036402, 2015. D. Mejía-Rodríguez, S. B. Trickey,. Phys. Rev. A 96, 052512, 2017.

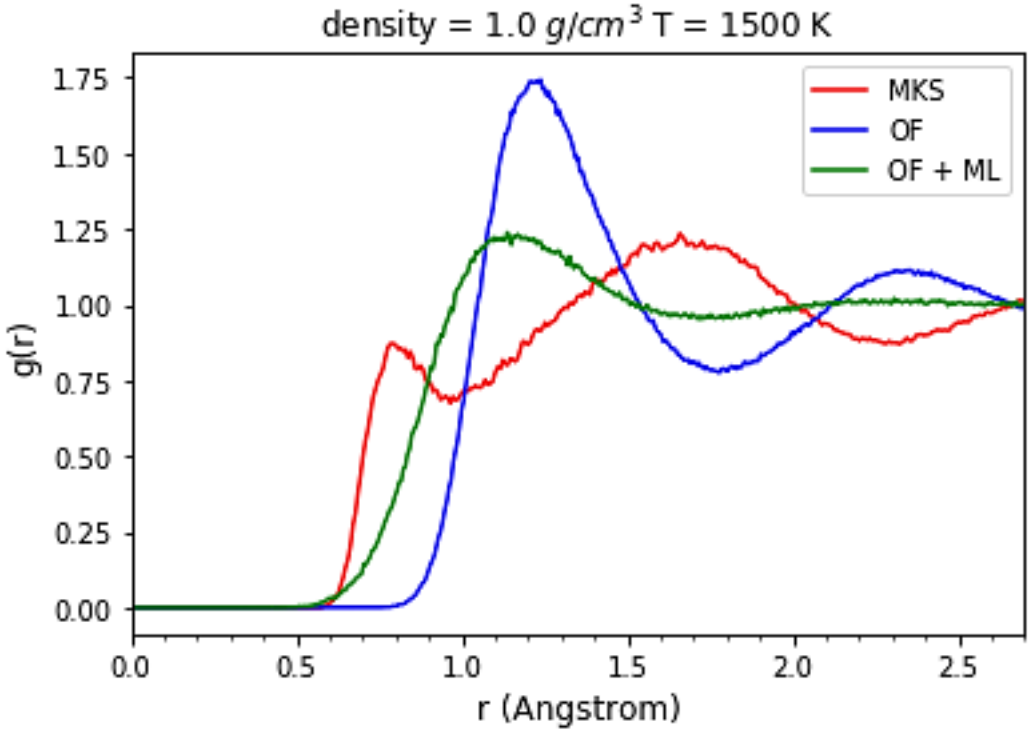
Further care must be taken when constructing the reference data set to ensure there is enough resolution of the entire input vector space.





- With two standard and one nearest neighbor descriptor vector the average and median relative error on the force components are 28% and 5.5% respectively.

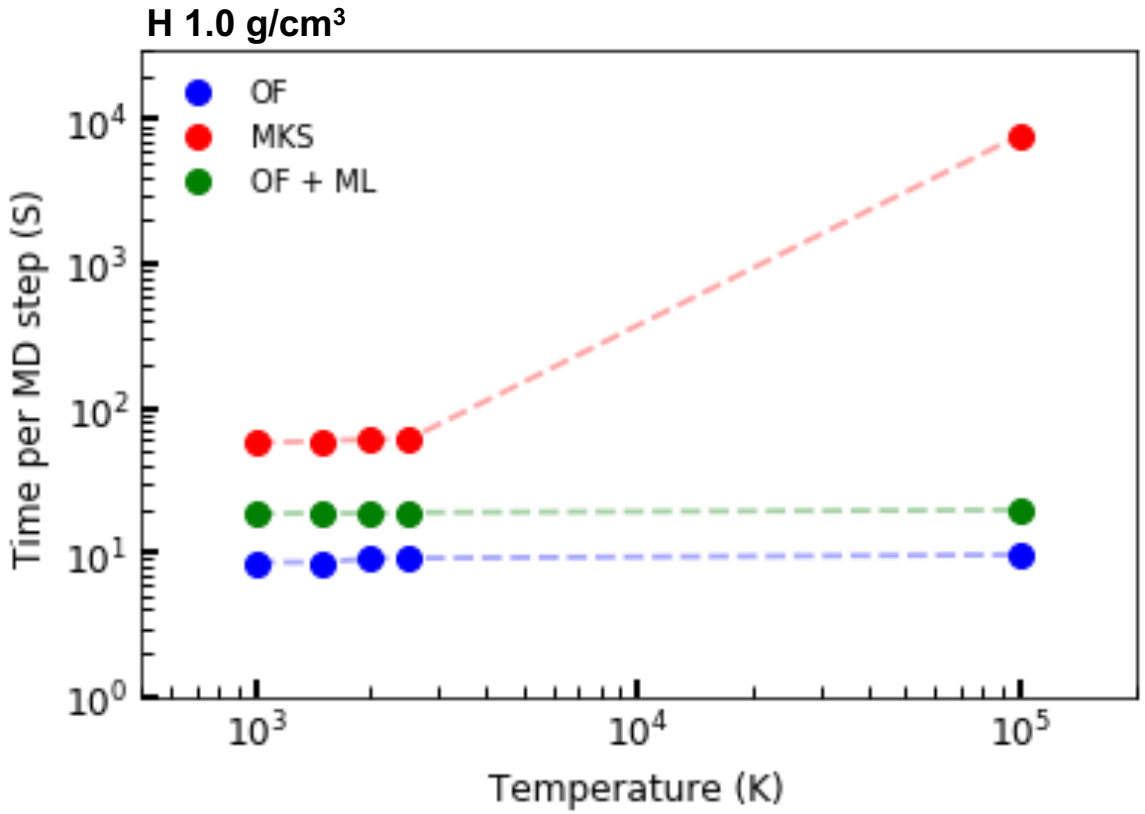
The ionic pair correlation function indicates further work must be done to improve the force correction model.



- **At its current state, the model is able to produce a fluid hydrogen system with a molecular character.**
- **The equilibrium bond length and the total fraction of molecules lies between the MKS and OF predictions.**

Time analysis of the force correction model shows significant improvement from MKS based MD simulations.

- The computational time per MD step of MKS DFT is a factor of three larger than the OF+ML model.
- The optimization of the hyperparameters costs what is equivalent to 2s on each MD step.
- From an implementation standpoint, improvements can be made to reduce the computational time.



The force correction model shows promise but requires further work to improve both the accuracy and validate the results.

- The force correction model shows promise in terms of the reduction in computational time as well being able to predict most force components within 5.5% error.
- Adjustments to the acquisition of reference data is required.
- Introducing descriptor vectors to provide information on the angular distribution of the local ion configuration is required.
- Further validation must be carried out to confirm result are representative of MKS calculation for longer time scales and for larger systems.

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