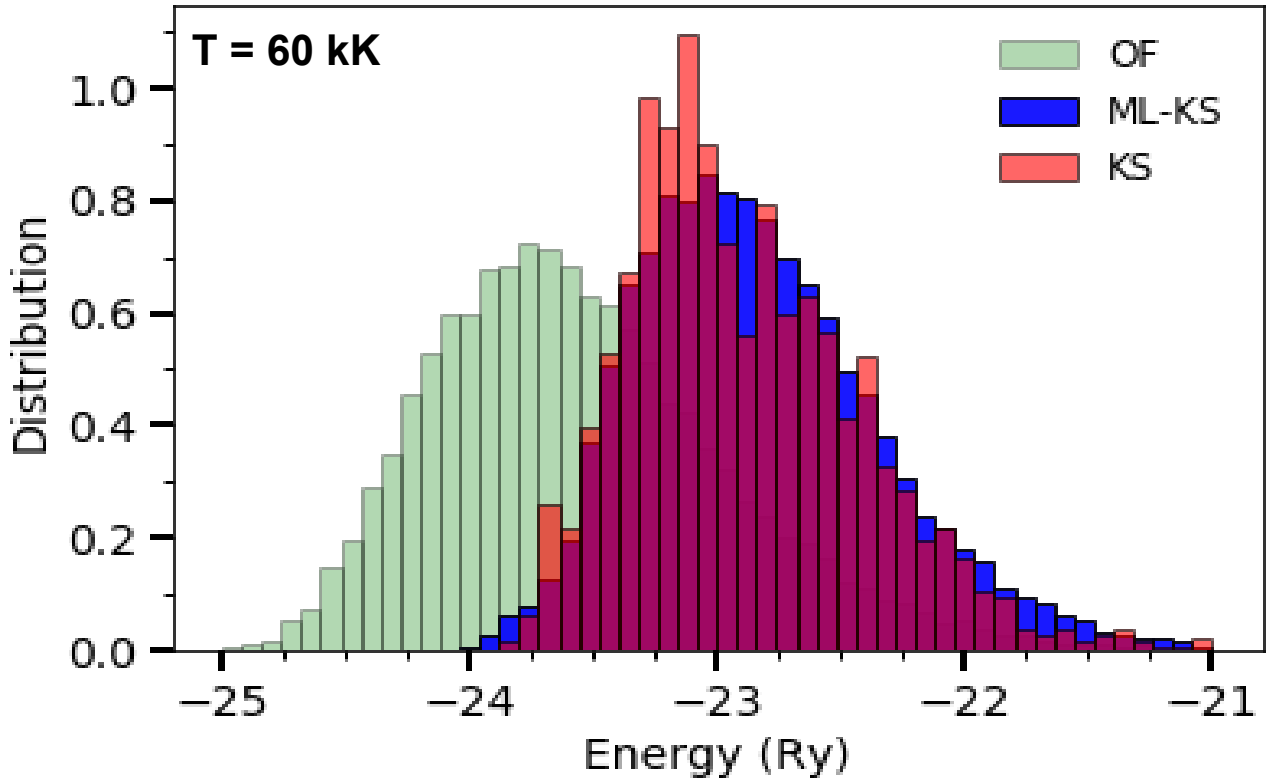
Development of a machine learning based ionic force correction model for quantum molecular dynamic simulations of warm dense matter.



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Collaborators and Funding

Valentin Karasiev, Suxing Hu, Deyan Mihaylov

Laboratory for Laser Energetics (LLE)

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Summary

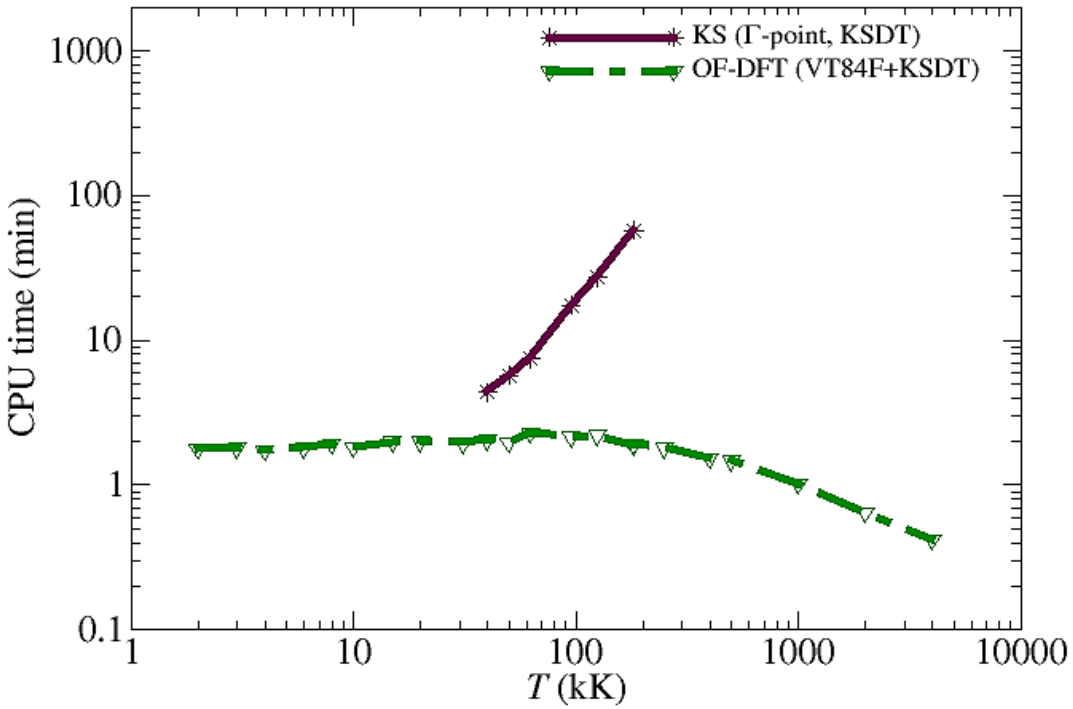
A neural network-based machine learning model has been constructed to predict ionic force differences between Kohn-Sham and orbital-free density functional theory.

- An approximate force difference is constructed in terms of the neighboring ion positions allowing for a neural network to be used to predict each neighbor's contribution to the force difference.
- The input of the model is constructed from information about the configuration of neighboring ions.
- The model has been trained and tested on warm dense hydrogen at 1.0 g/cm^3 between 10 and 150 kK.
- The transferability of the model to temperatures outside of the training set indicates future work on descriptors for ML models of warm dense matter simulations is needed.

Results from molecular dynamic simulations indicate the model can reproduce the Kohn-Sham electronic energy and pressure within 1 and 2% respectively.

J. Hinz *et al*, Phys. Rev. Materials (under review).

Ab initio molecular dynamic simulations require a delicate balance of accuracy and computational cost.



- **Kohn-Sham density functional theory (KS-DFT) has become the primary work horse for the quantum treatment of electrons.**
- **However, KS-DFT has a computational cost that scales cubically with the system temperature.**
- **Alternatively, orbital-free (OF) DFT is order of magnitude faster but has a limited range of acceptable accuracy.**
- **Machine learning (ML) can be used to produce a model for the ionic force that captures the best of both DFT branches.**

$$\vec{F}^{KS} = \vec{F}^{OF} + \Delta\vec{F}^{ML}$$

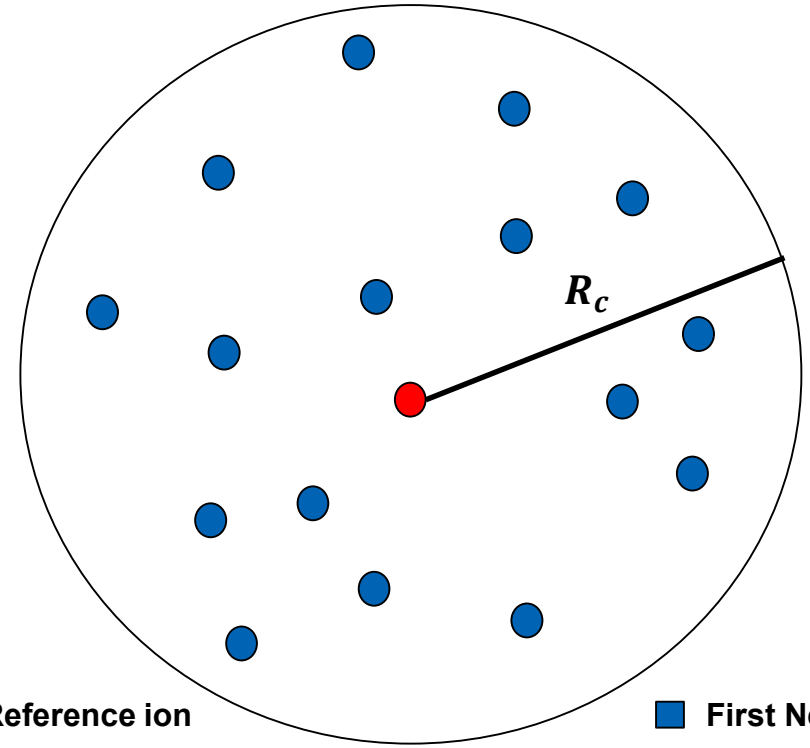
V. V. Karasiev, *et al*, Comput. Phys. Comm. 185, 3240, 2014.

An approximate force difference can be obtained in terms of the ionic positions.

Reference force difference:

$$\Delta\vec{F}_i^{ref} = kq^2 Z_i \int d^3r \frac{\Delta n_0(r)}{\left|\vec{r} - \vec{R}_i\right|^3}\left(\vec{r} - \vec{R}_i\right)$$

Approximations:



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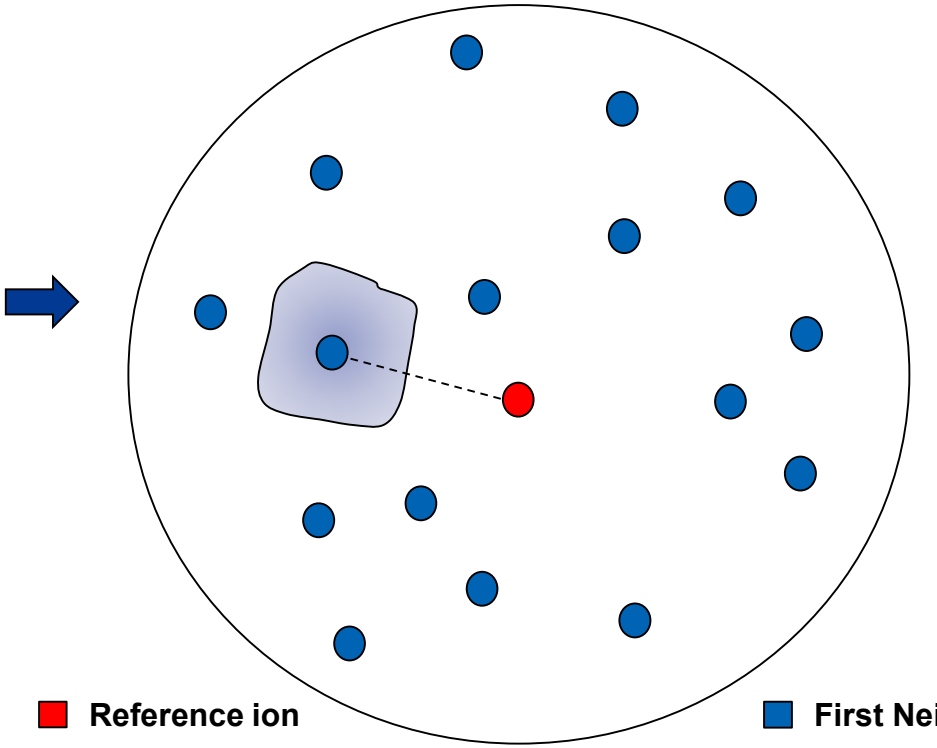

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Note: In practice we do not create these sub volumes but rather they will be learned implicitly through the learning of the weights.

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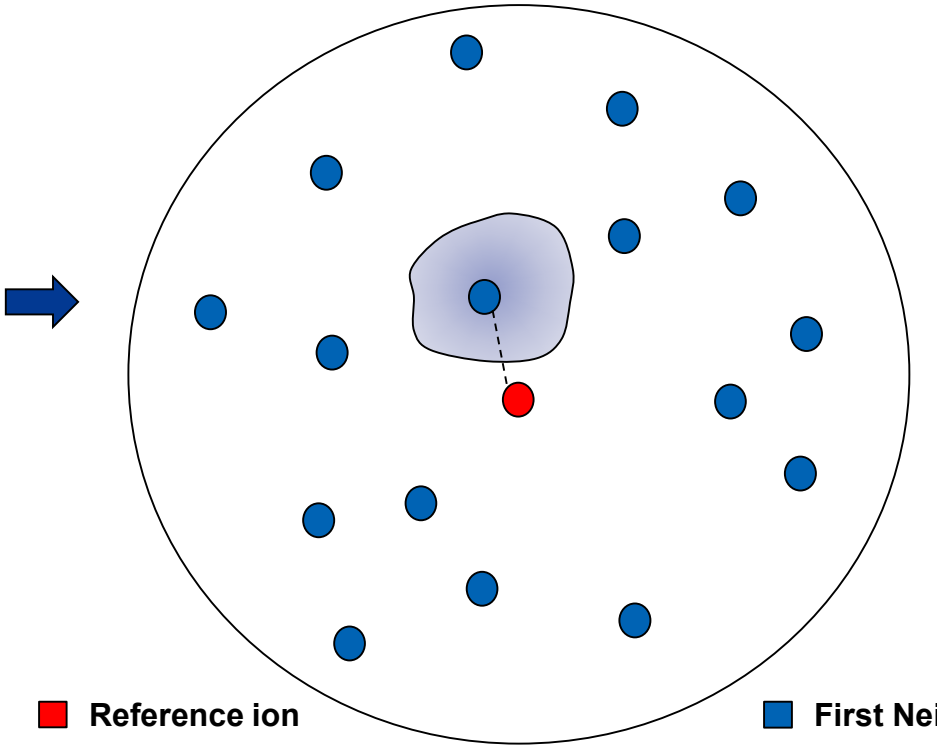

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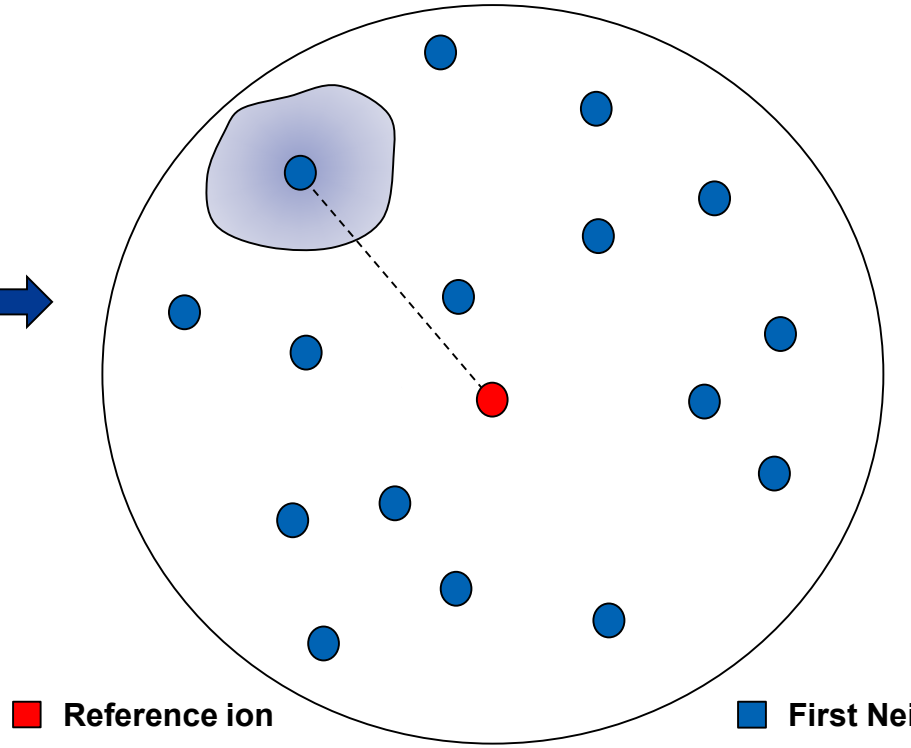

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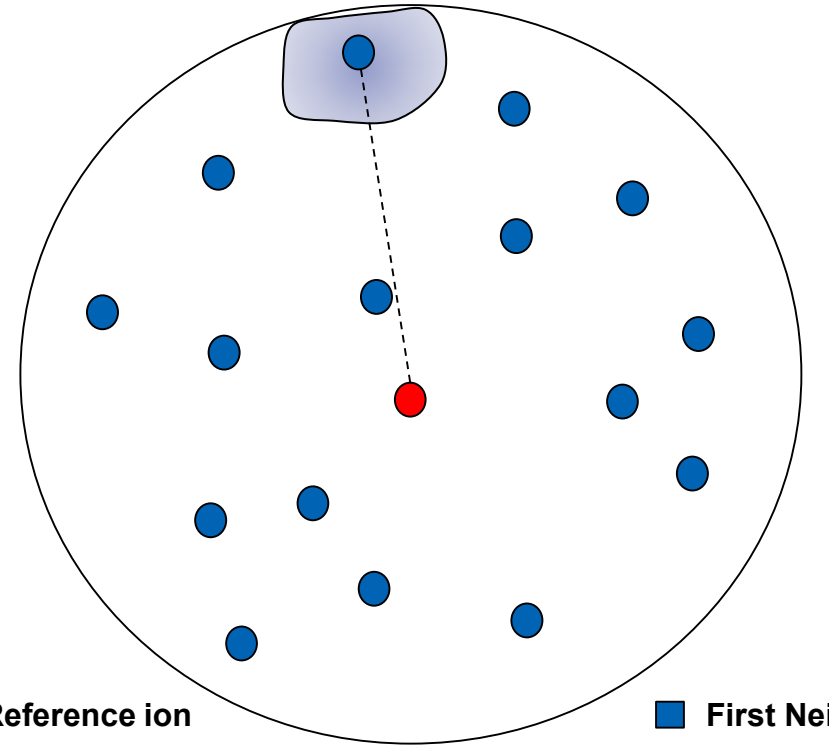

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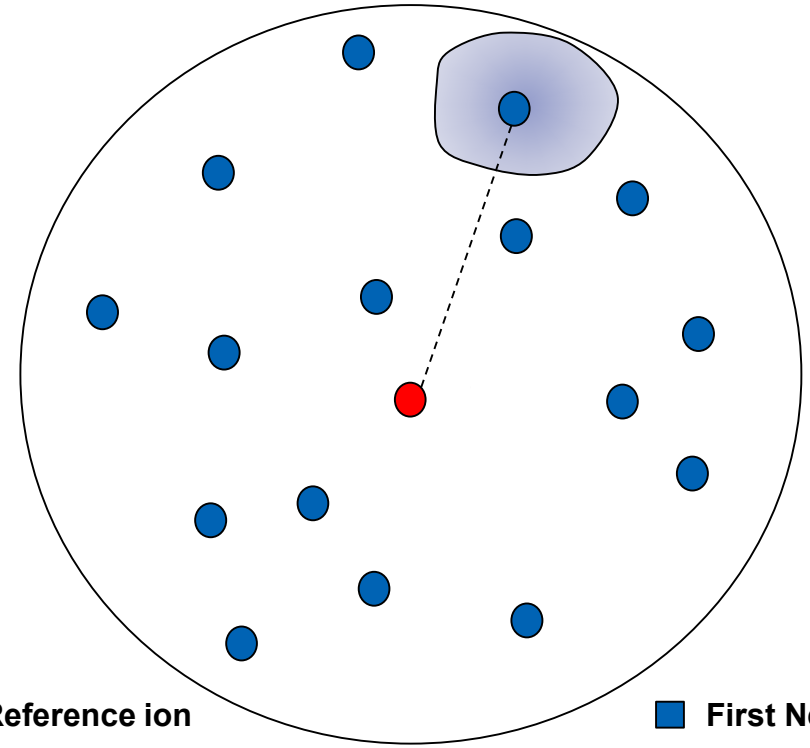

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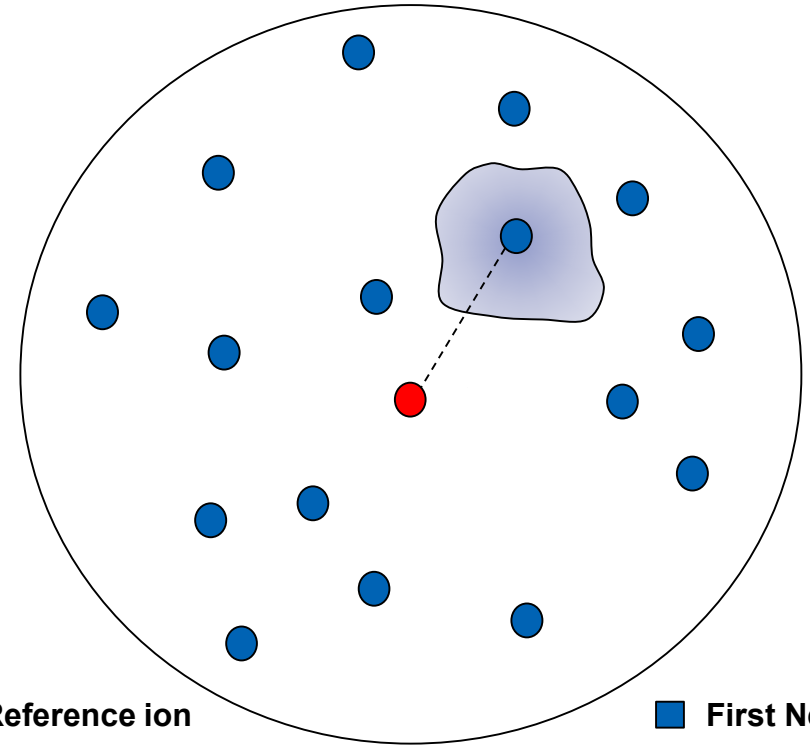

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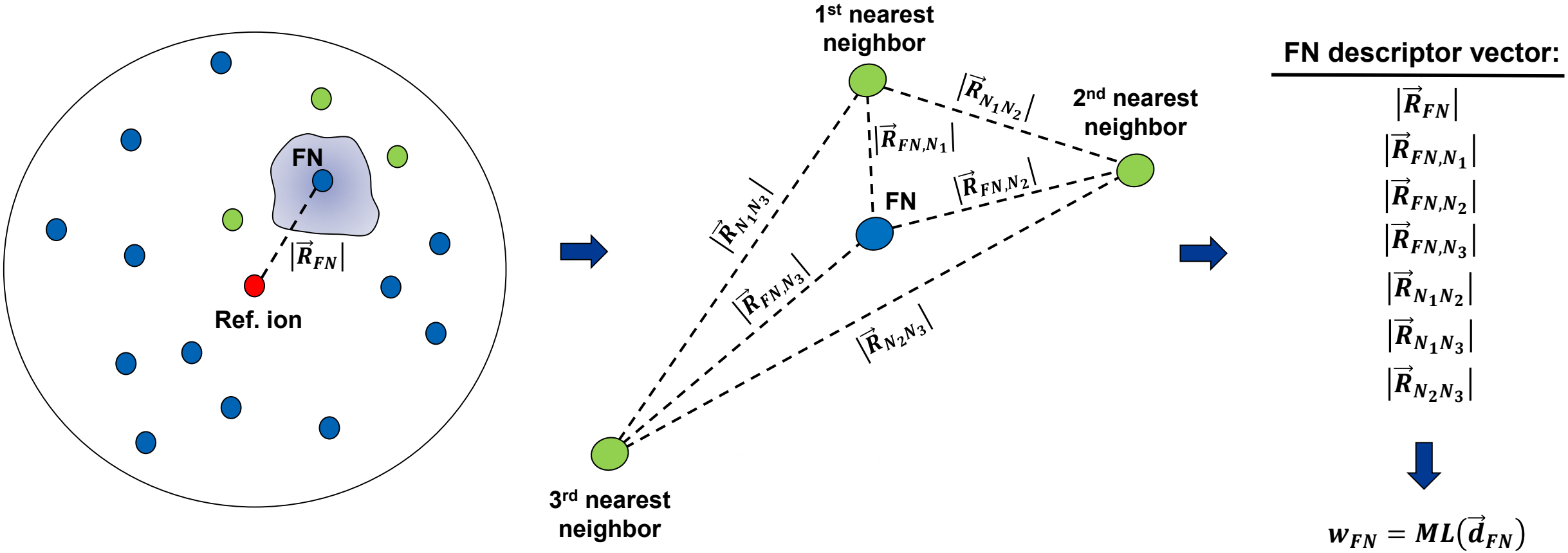
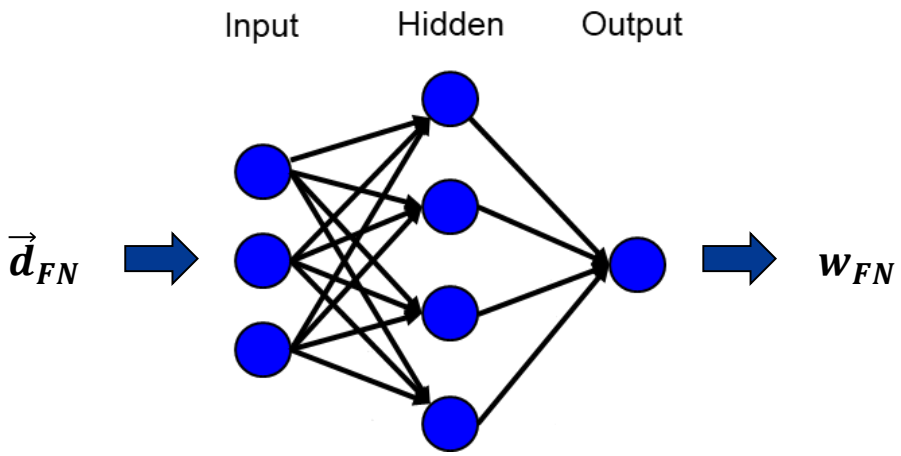
For each FN, information about its neighbors is used to construct the descriptor vector needed to predict that FN's contribution to the force difference.



A neural network is used to implicitly learn the FN weights for warm dense hydrogen at 1.0 g/cm^3 and at temperatures between 10 and 150 kK.



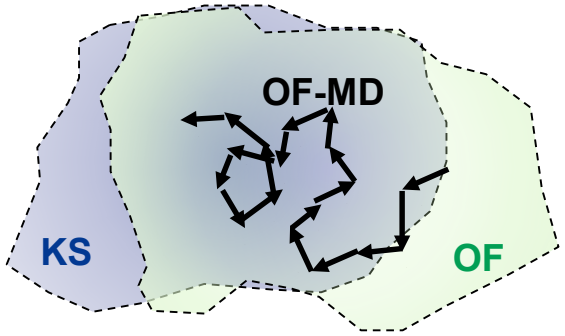
Technical parameters:

 # hidden layers (HL)= 1
 # nodes in HL = 40
Activation function= ReLu

Cost:

$$C = \frac{1}{2N_s}\sum_{i}^{N_s}\left\{\left|\Delta\vec{F}_i^{ref} - \Delta\vec{F}_i^{ML}\right|^2 + A exp\left(-\alpha\left|\Delta\vec{F}_i^{ML}\right|\right)\right\}$$

Domain of ion configurations:



F_S: LKTFγTF E_{XC}: SCAN-L

- **At each temperature considered a test and master training set are constructed.**

Number of local configurations:

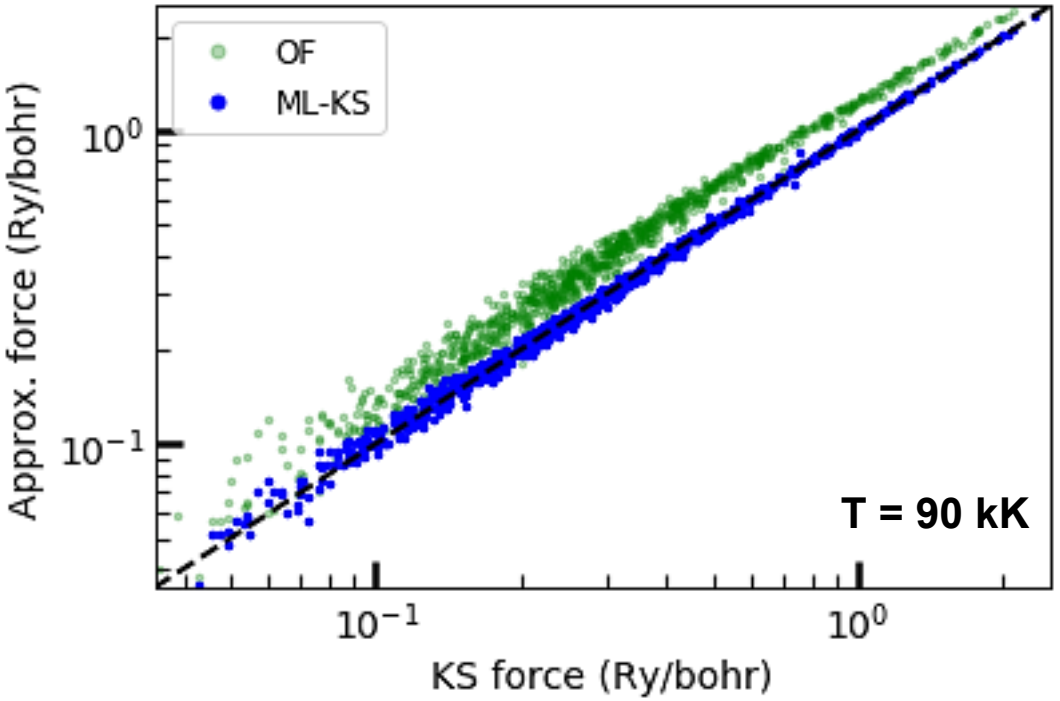
Test: 2000
Master Training:10000 → **Ensemble (15 models)** → **Training: 5800**, **Validation: 2500**, **Pseudo-test: 1700**

Randomly sampled

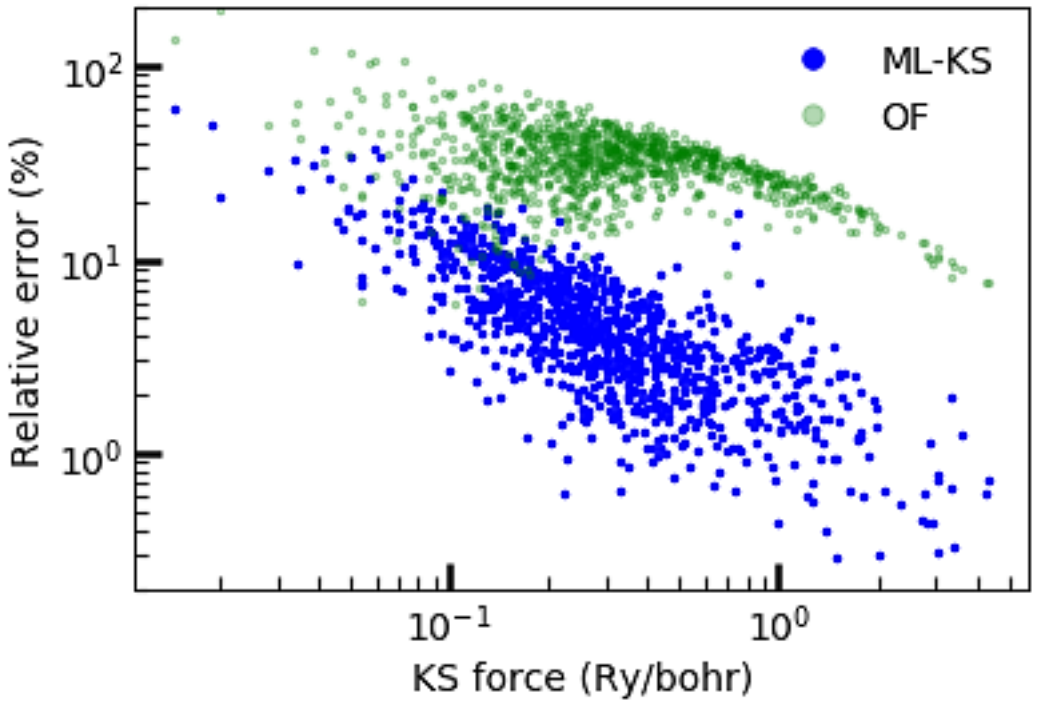
V. V. Karasiev, *et al*, Comput. Phys. Comm. 185, 3240, 2014. D. Mejia-Rodriguez and S. B. Trickey, Phys. Rev. A 96, 052512 (2017). K. Luo, *et al*, Phys. Rev. B 101, 075116 (2020). R. P. Feynman, *et al*, Phys. Rev. 75, 1561 (1949).

Results of the force correction model on the test set at 90 kK show significant improvement over the underlying orbital-free ionic forces.

$$\vec{F}^{ML-KS} = \vec{F}^{OF} + \Delta\vec{F}^{ML}$$



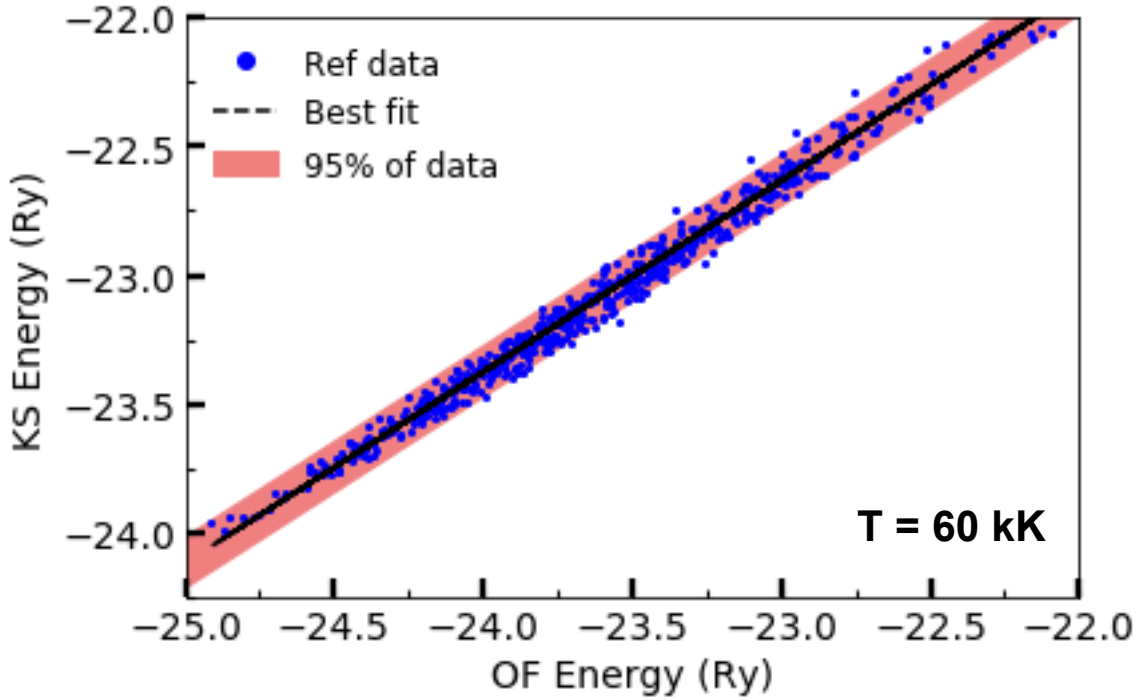
$$RelErr = abs\left(\frac{|\vec{F}^{approx}| - |\vec{F}^{KS}|}{|\vec{F}^{KS}|}\right) x100$$



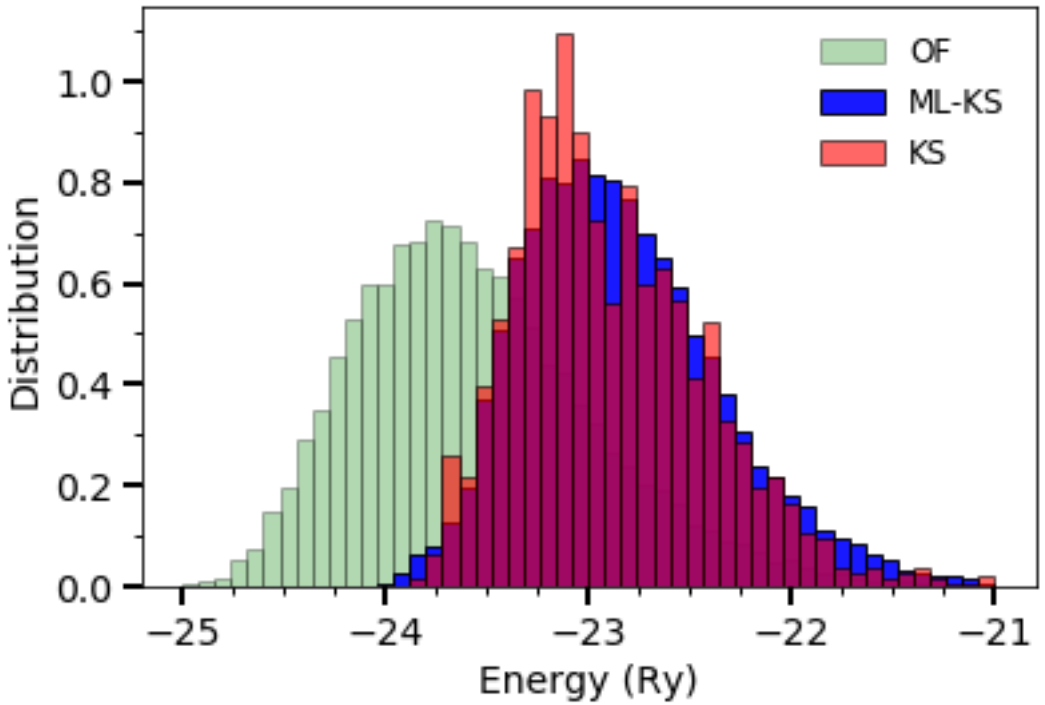
1 Ry/bohr = 25.711 eV/Å

Correlations between the orbital-free and Kohn-Sham energy are used to obtain the energy from the force correction model during molecular dynamic simulations.

Reference energies



Distributions from MD

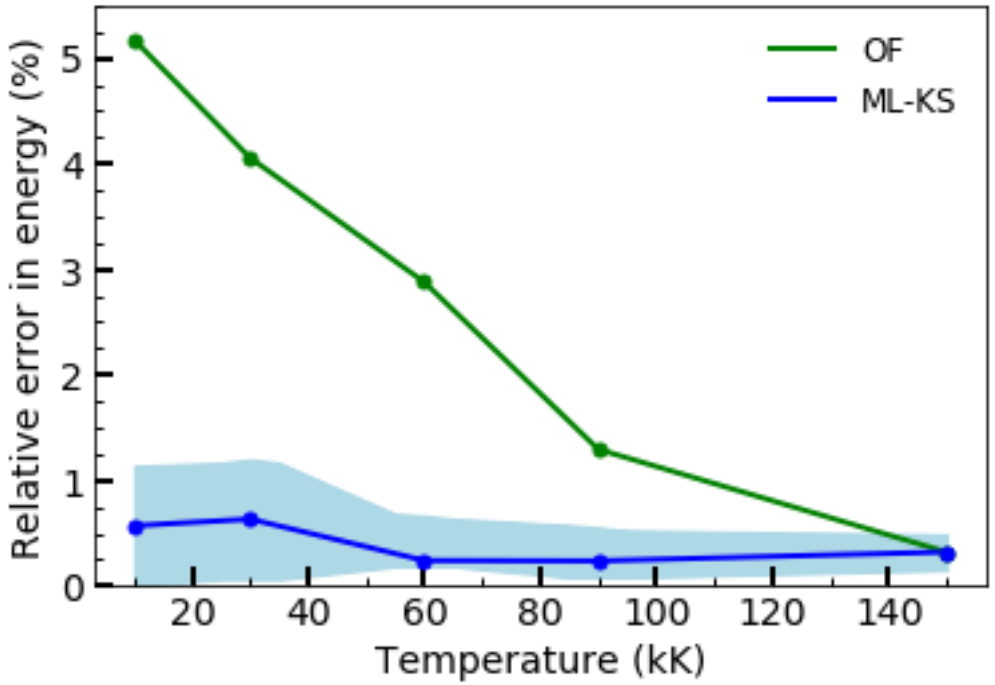


Similarly, a linear correlation between the orbital-free and Kohn-Sham electronic pressure exists for a given snapshot.

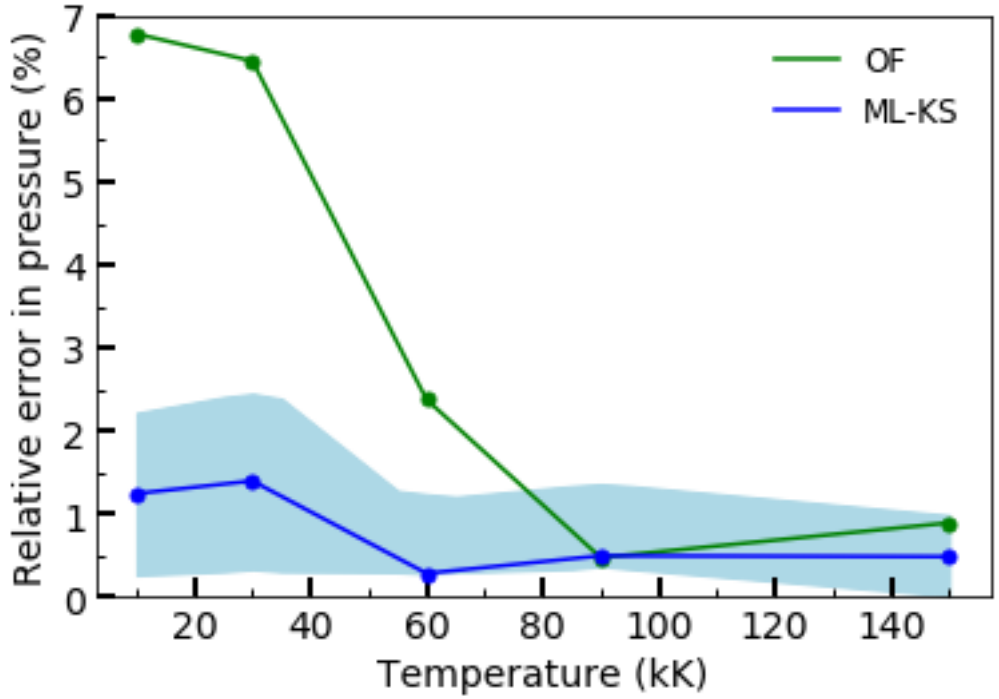
1 Ry = 13.606 eV

The average electronic energy and pressure from molecular dynamic with the force correction model are an improvement over those from orbital-free DFT.

Error in predicted energy



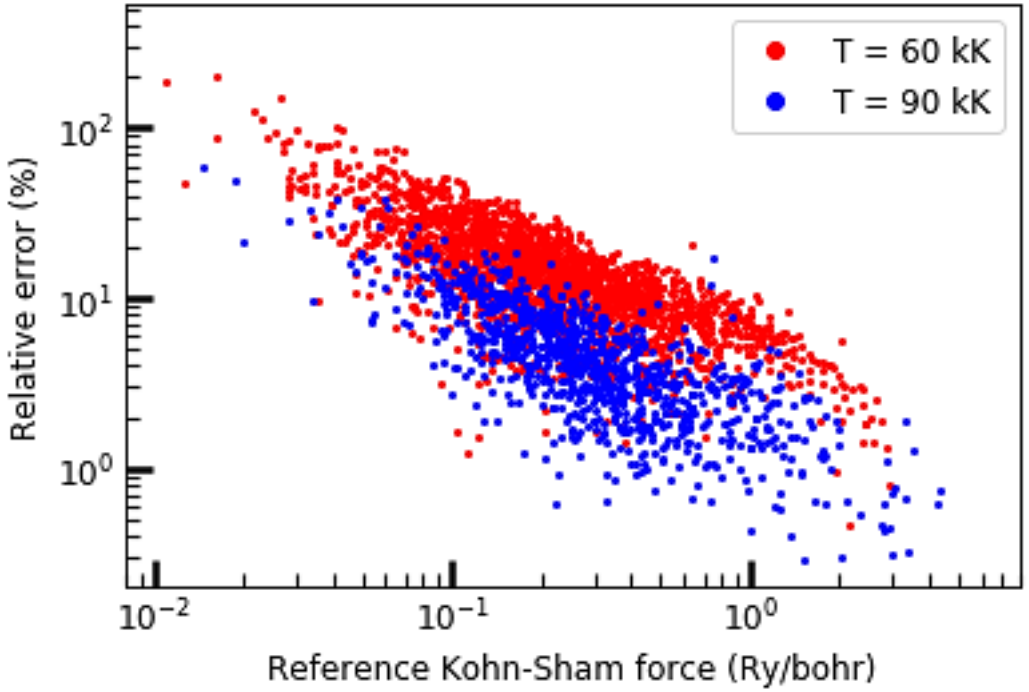
Error in predicted pressure



Errors in the electronic energy and pressure are consistently below 1 and 2 % respectively down to 10 kK.

Transferability tests indicate a trained model will have a limited temperature range of applicability.

- All model up to this point have been constructed with reference data obtained at a single temperature.
- The force correction model trained at 90 kK is applied to a test set generated at 60 kK
- Loss in model accuracy can be attributed to the ionic forces being temperature dependent.



At present, current schemes of descriptors do not contain all relevant information needed to construct a single model from wide ranging reference data in the warm dens matter regime.

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Summary

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- An approximate force difference is constructed in terms of the neighboring ion positions allowing for a neural network to be used to predict each neighbor's contribution to the force difference.
- The input of the model is constructed from information about the configuration of neighboring ions.
- The model has been trained and tested on warm dense hydrogen at 1.0 g/cm^3 between 10 and 150 kK.
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