Supplementary Materials for

"Hydrogen Diffusion in Magnesium Using Machine Learning Potentials: a comparative study"

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In this section we show supplementary figures to the main work:

Fig. SF1 shows the mean square displacement of one the VASP-MLFF replicas at the highest investigated temperature, and the component decomposition along three directions. Overall across the various simulations we have not observed a consistent difference in the behavior between the different components.

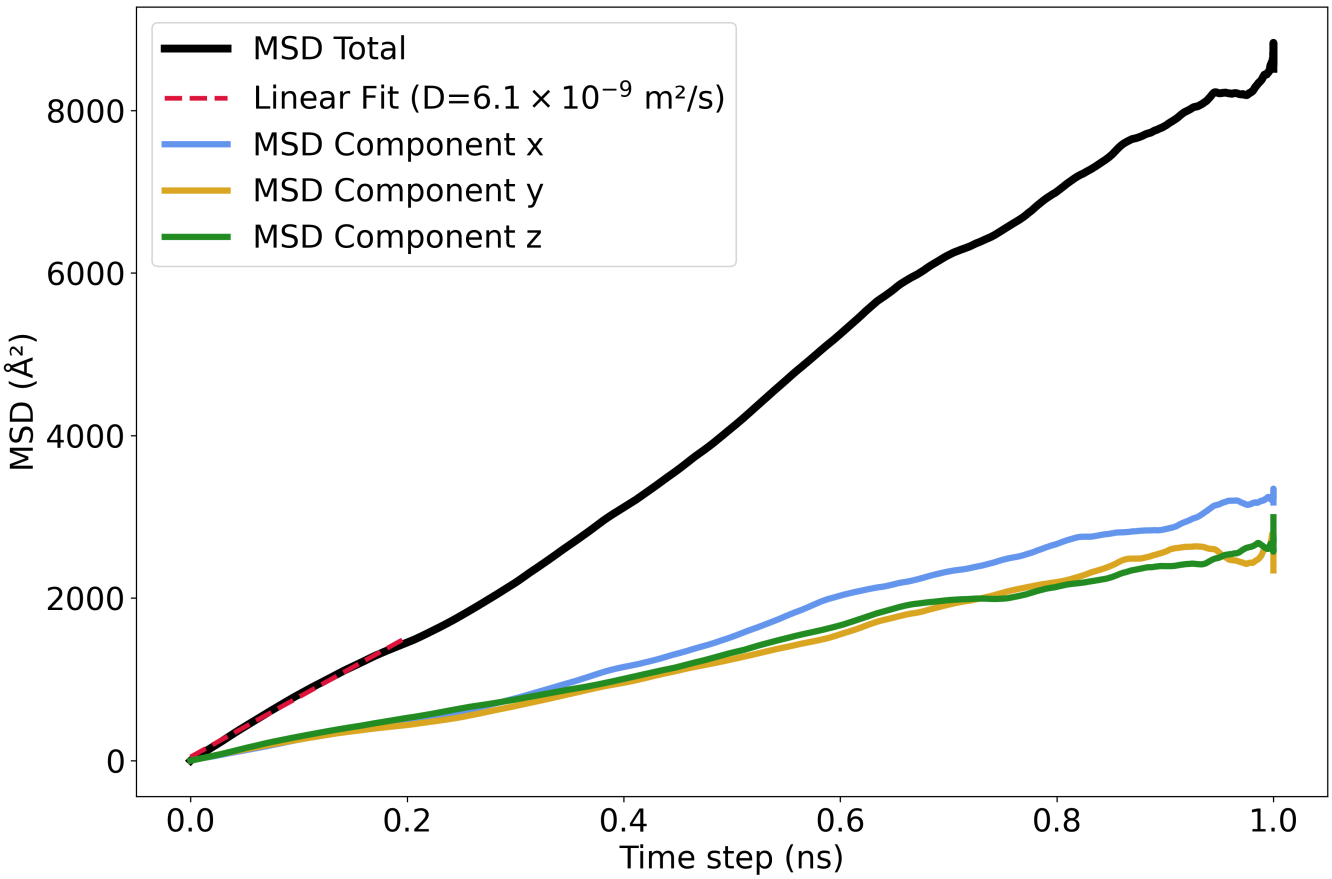
Fig. SF2 shows the un-shifted energy prediction errors of the models during their validation and the corresponding R^2 values.

Fig. SF3 shows the octahedral O_i and tetragonal T_i interstitial hydrogen sites in the magnesium matrix, between which we computed the associated energy barrier via ciNEB, with the ML-models and VASP-DFT.

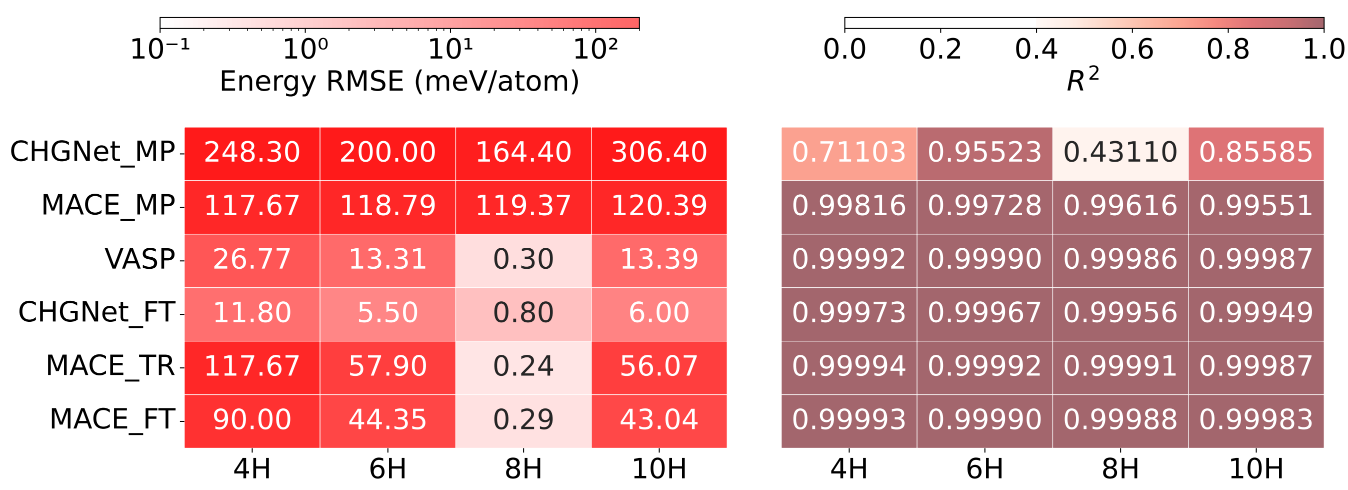
Fig. SF4 shows the drifting in energy for the CHGNet_MP model using a time step of 1 fs. By reducing it to 0.5 fs the drifting disappears as can be see in Fig. SF5. Moreover, we can also observe the temperature fluctuations during the molecular dynamics simulations of CHGNet_MP model, which turned to out to be most unstable model overall.

Fig. SF6 shows the Radial Distribution Function of all atom pairs for three ML-model at 480 K and 300 K.

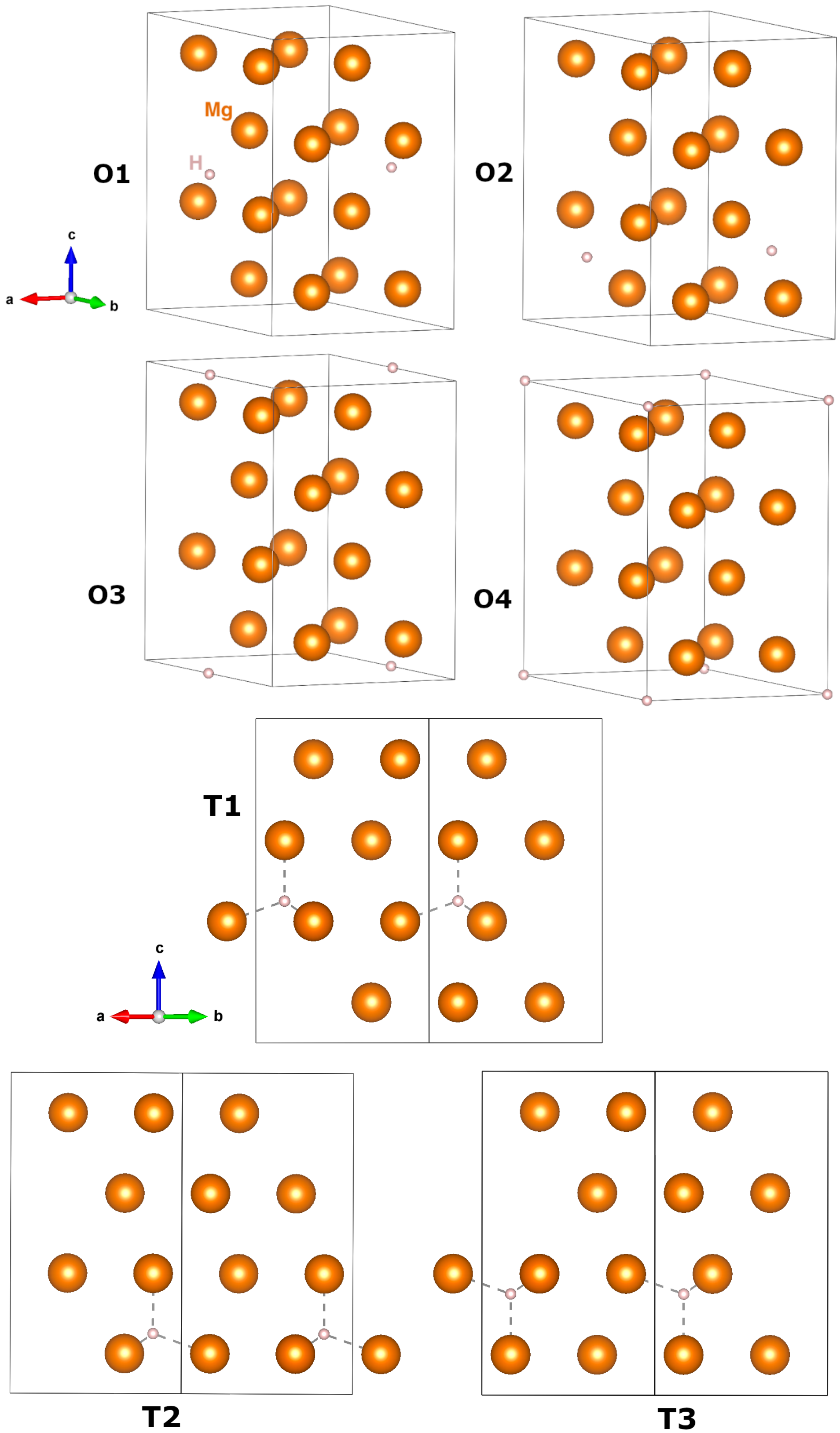
Fig. SF7 shows estimates of the MSD at different temperature for the MACE_FT model using different time lags (parameter δ of MSD formula in Ref. [1]). In particular, the results are calculated for the standard $\sigma = 1$ and the fully uncorrelated $\sigma = \tau$ cases, estimating also the diffusion coefficient to show a possible dependence on the parameter. The two estimates showed nearly the same linear trend with differences mainly related to the lower statistic available to the $\delta = \tau$ case that makes it more noisy. This is well reflected by the estimates of D fitted in the range described previously for the $\delta = 1$ case and in a smaller range of 0 and 0.1 ns for the uncorrelated case, where the smaller range is needed due to the much higher noise in longer times. The estimates deviates of at most 2.5 % between the two cases still remaining perfectly compatible within the estimated error, leading to variations in the estimates of the energy barrier of less than 1 %. This shows how no major correlation is present inside the MSD of the case under study, making the values obtained with $\delta = 1$, corresponding to the formula reported in the main text, more reliable due to their much larger statistic that allows for a converged estimates in the simulation time under investigation.



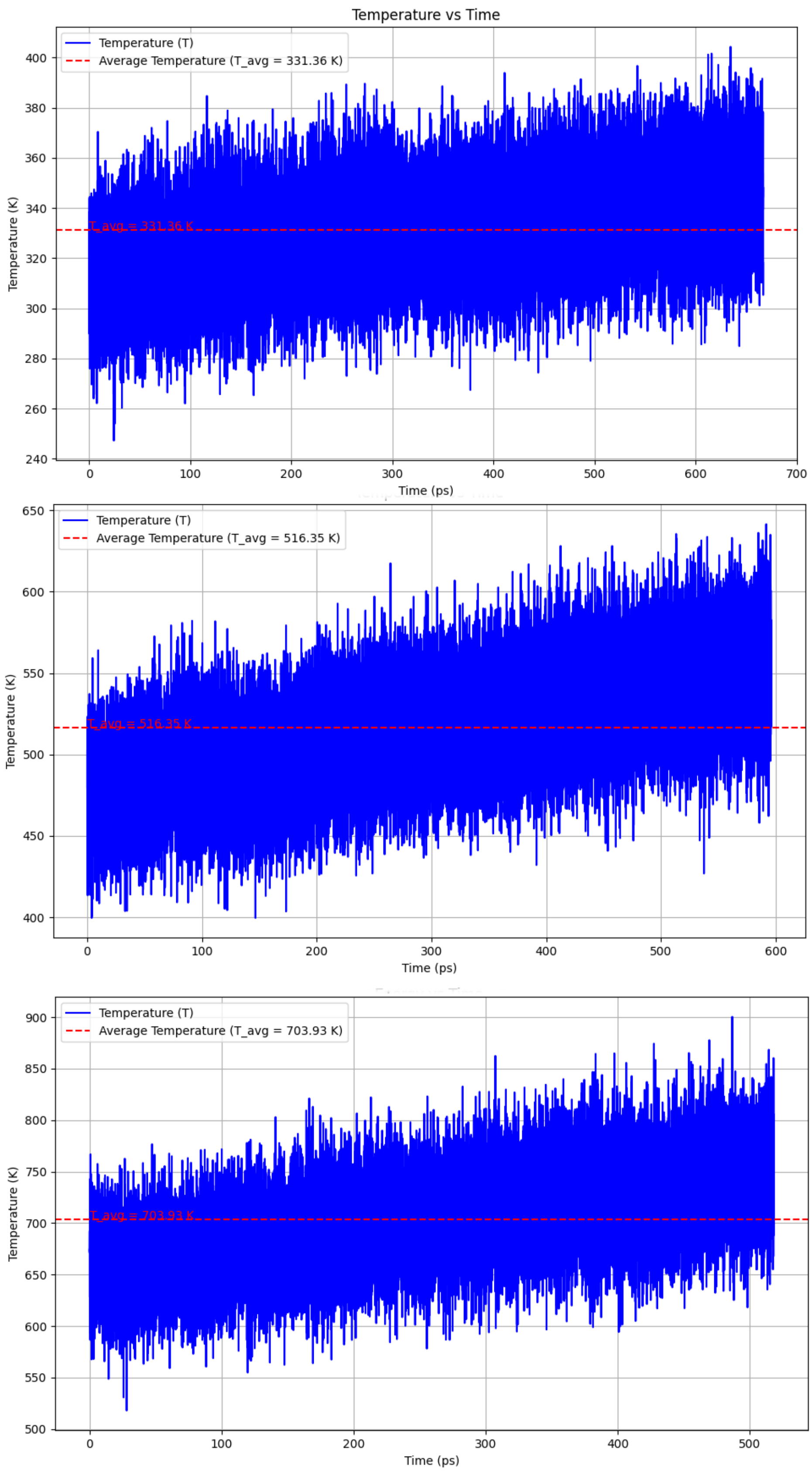
Supplementary Figure 1: Example of MSD fitting procedure for the case of VASP MLFF model at 673K and the values along different directions.



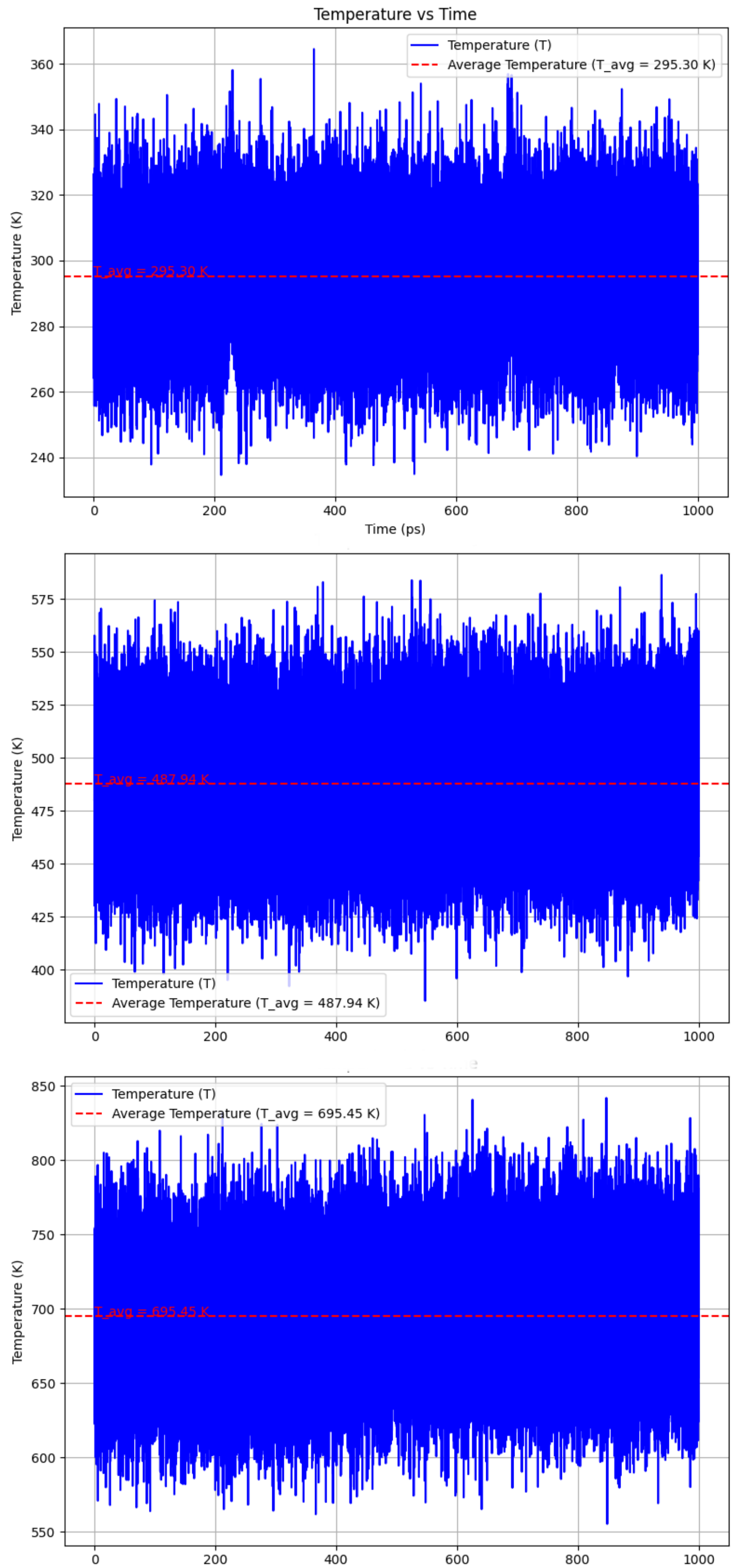
Supplementary Figure 2: The left table shows the RSME for energies and forces for all models sand concentrations, without energy shifting. The corresponding R^2 values for the predictions are on the right table.



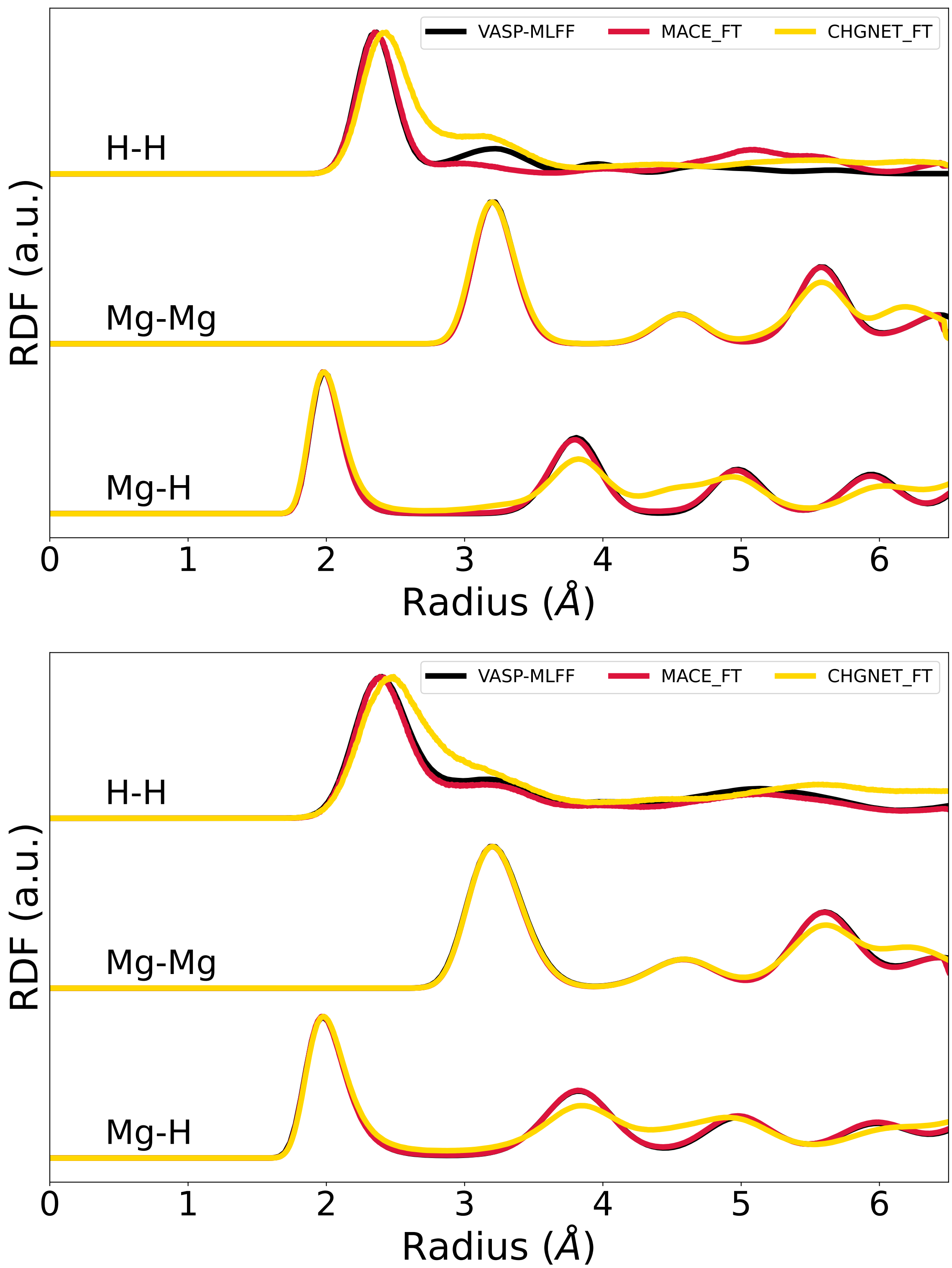
Supplementary Figure 3: Interstitial states considered for NEB calculations with all models.



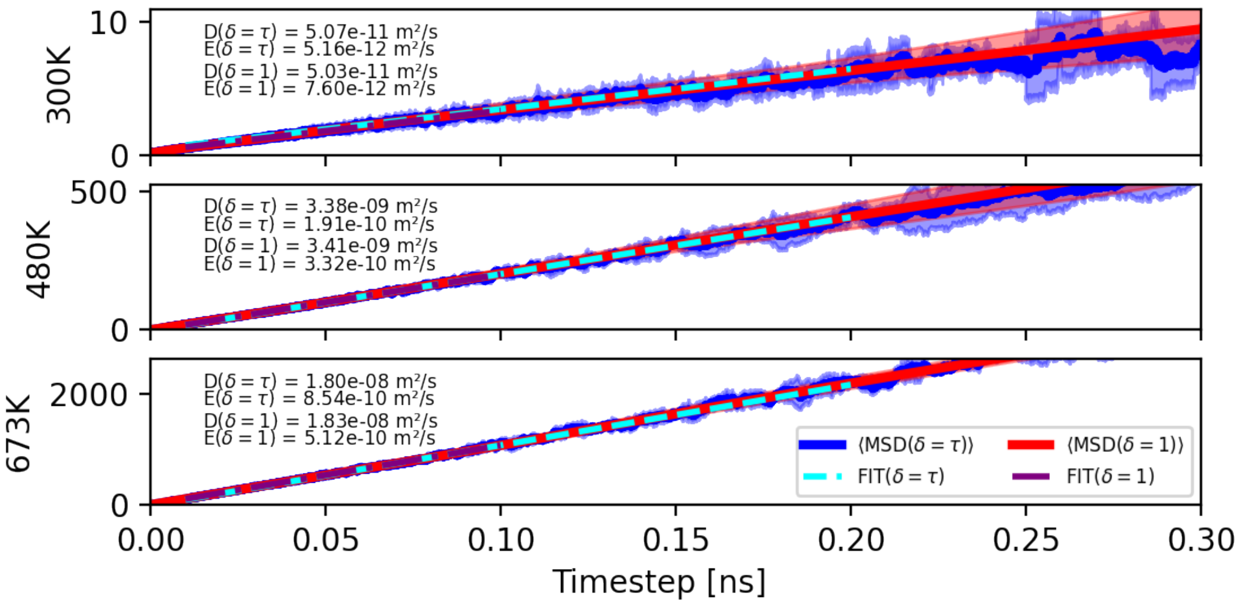
Supplementary Figure 4: Example of drifting in the NVE temperatures for the case of 1 fs timestep simulations with the pretrained CHGNet_MP model.



Supplementary Figure 5: Example of stable temperature oscillations in the NVE simulations for the case of 0.5 fs timestep with the CHGNet_MP model.



Supplementary Figure 6: Radial distribution function at different temperatures for the best performing models, at 300 K on top and 480K at the bottom.



Supplementary Figure 7: Values of the MSD at different temperatures, with following linear fit, for the MACE_FT model estimated for different time lags, δ, following the formula in Ref. [1].

[1] G. Pranami and M. H. Lamm, Journal of Chemical Theory and Computation **11**, 4586 (2015), https://doi.org/10.1021/acs.jctc.5b00574.