Active Δ-learning with universal potentials for global structure optimization: Supporting Information

Joe Pitfield,* Mads-Peter Verner Christiansen, and Bjørk Hammer†
Center for Interstellar Catalysis, Department of Physics and Astronomy, Aarhus University, DK-8000 Aarhus C, Denmark
(Dated: November 21, 2025)

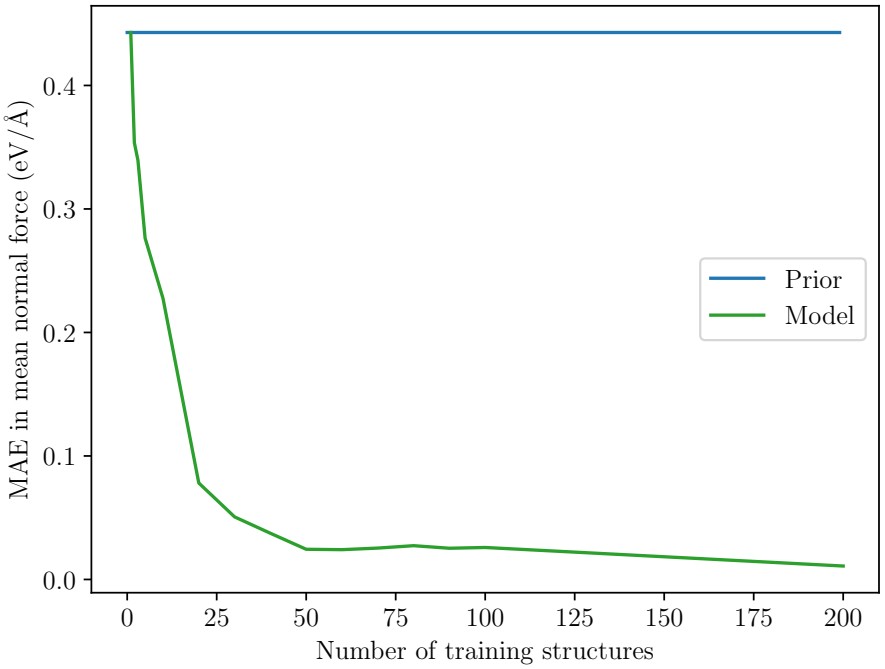

Fig. S1. Learning curve for Δ-model prediction on $Ag_{12}S_6$ clusters. MACE-MPA is the prior uMLIP for these models. Only DFT energies are used from the training data. The predicted forces are then compared with the DFT forces from the test data. The MAE in mean absolute force is calculated as a function of the quantity of training data, against this separate test set. Data is drawn from REX searches directly in MACE-MPA.

[1] F. Brix, M.-P. Verner Christiansen, and B. Hammer, The Journal of Chemical Physics **160**, 174107 (2024).

* joepitfield@gmail.com
† hammer@phys.au.dk

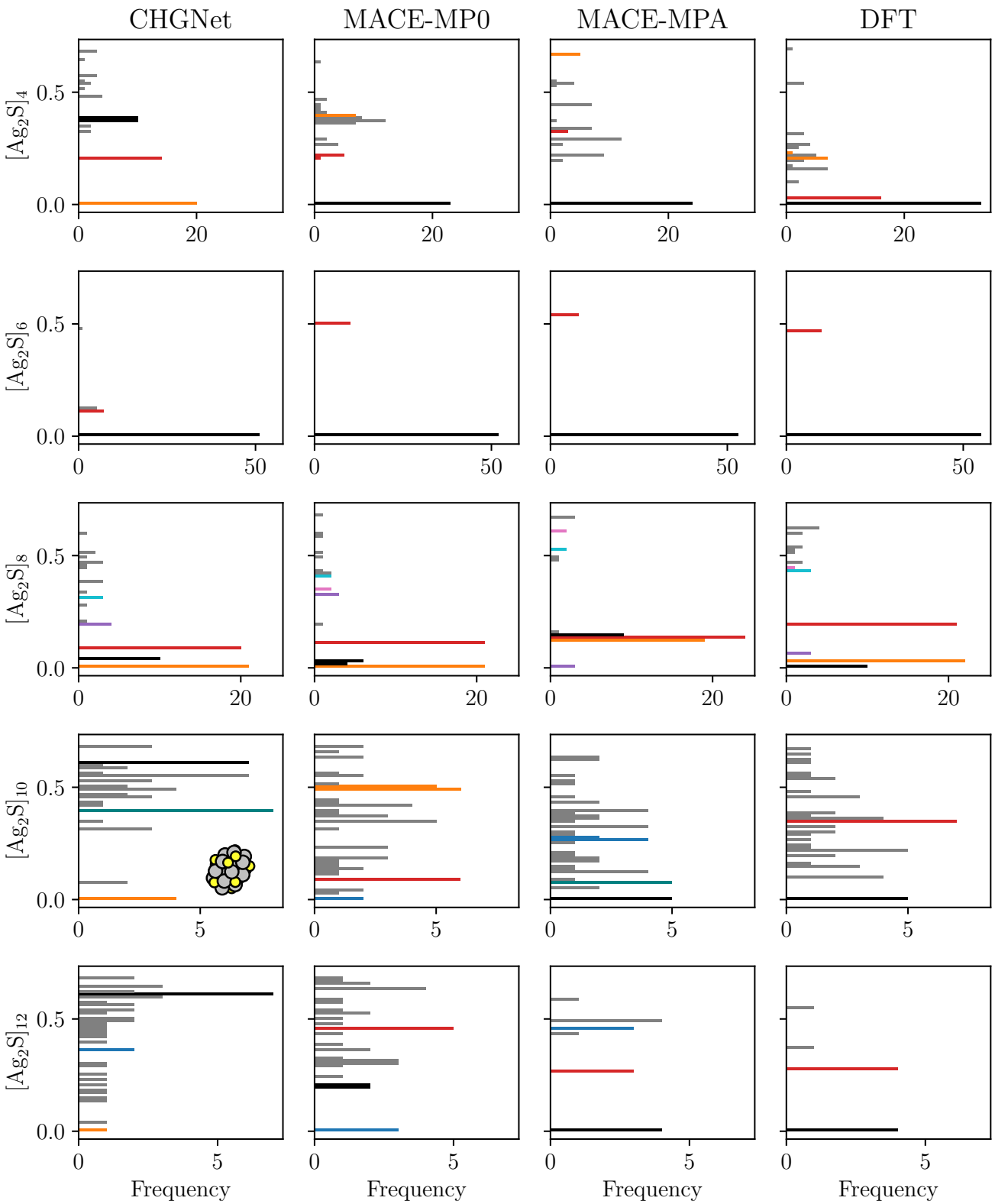

Fig. S2. For each stoichiometry of $[Ag_2S]_X$ clusters (having $X \in \{4, 6, 8, 10, 12\}$), 100 structures are selected according to the same methodology outlined in the main article. These structures are then relaxed in each of the uMLIPs/DFT, to a stringent 0.01 eV/Å force requirement, and a histogram of their relaxed energies is presented. Colors indicate corresponding structures are present within that bin, with stripes indicating degenerate energies. Colors used for $[Ag_2S]_6$ correspond to those in the main article, and the structural inset in CHGNet : $[Ag_2S]_{10}$ highlights the abnormal presence of sulfur coordinated sulfur in the minimum energy configuration.

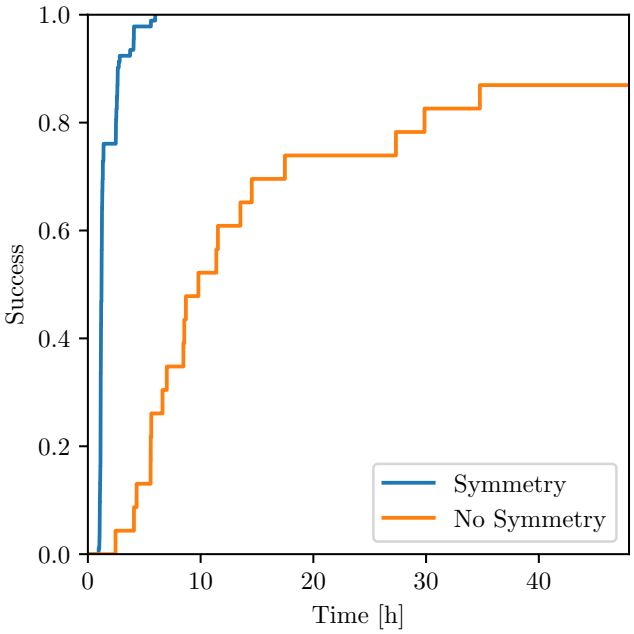

Fig. S3. Global optimization of $[Ag_2S]_{12}$ clusters using active Δ-learning with the REX algorithm and MACE-MP0 as the underlying potential. Here we compare the success profiles of 25 independent repeats either using a symmetry descent strategy[1], or using symmetry indifferent protocols.

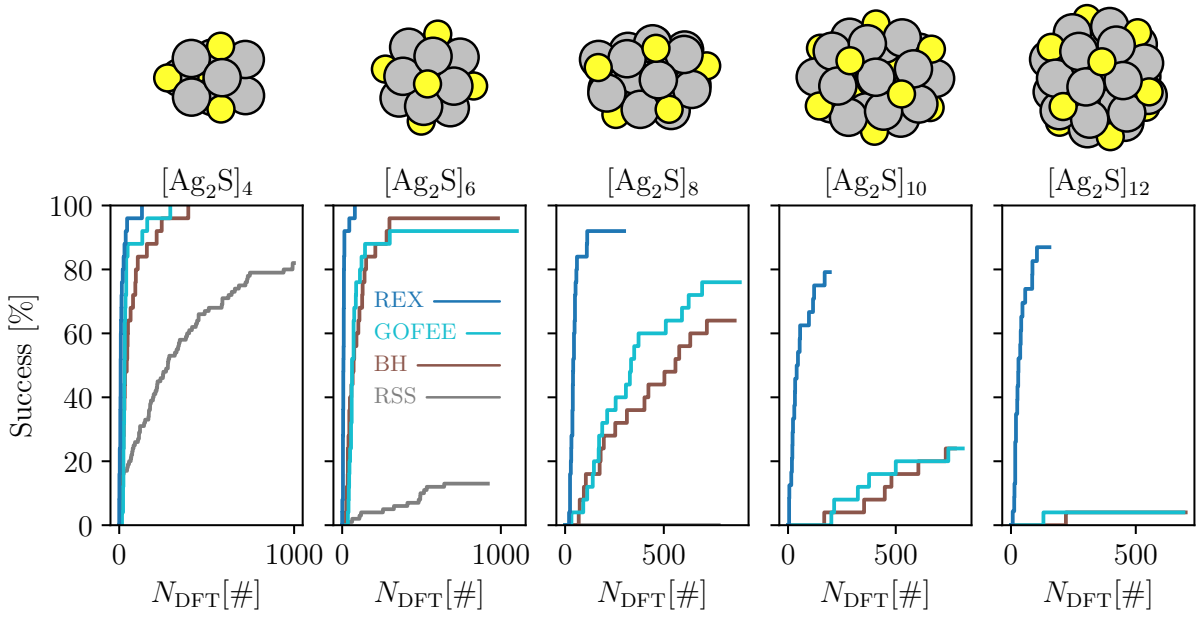

Fig. S4. The data presented in this figure is identical to that presented in Fig. 4 of the main article, with number of DFT calculations performed as the metric of comparison, rather than elapsed time. Global optimization is performed on $[Ag_2S]_X$ clusters using active Δ-learning with the MACE-MP0 universal potential, employing the four search methods outlined in the Global Optimization section of the main article. For each search 25 (100 for RSS) independent repeats were conducted and the success curves report the accumulated share of repeats that have found the GM as a function of the number of performed DFT calculations. The finding of the GM is determined according to a strict spectral graph decomposition, as described in the main article

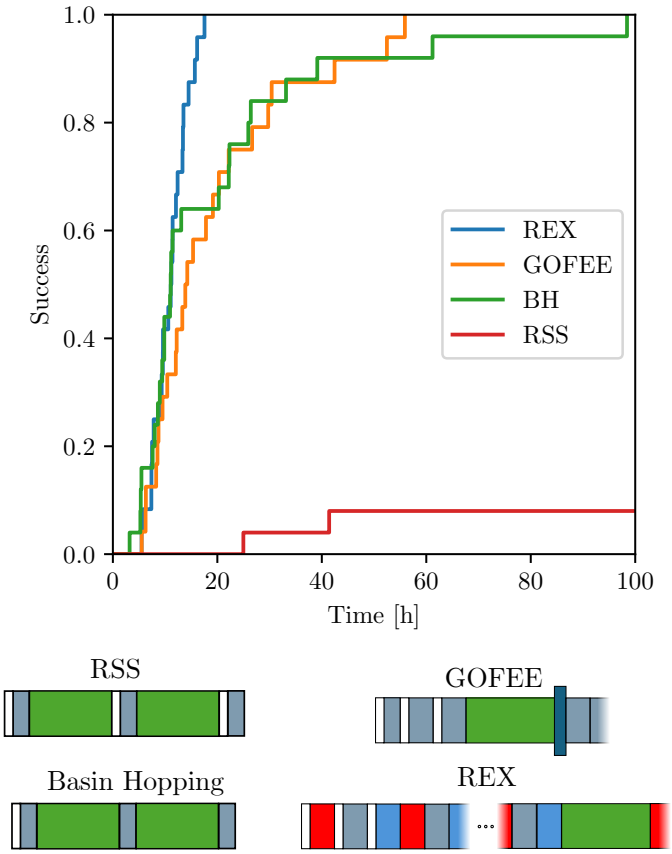

Fig. S5. Above: Global optimization of $[Ag_2S]_6$ clusters with serialized (1 core only) versions of the RSS, BH, GOFEE and REX algorithms. The success curves of 25 independent experiments for each method are shown, with the same success criteria as in the main article. Beneath: Schematic representation of the serialized version of the algorithms, highlighting that RSS and BH now no longer have idle CPUs while performing surrogate model relaxation.

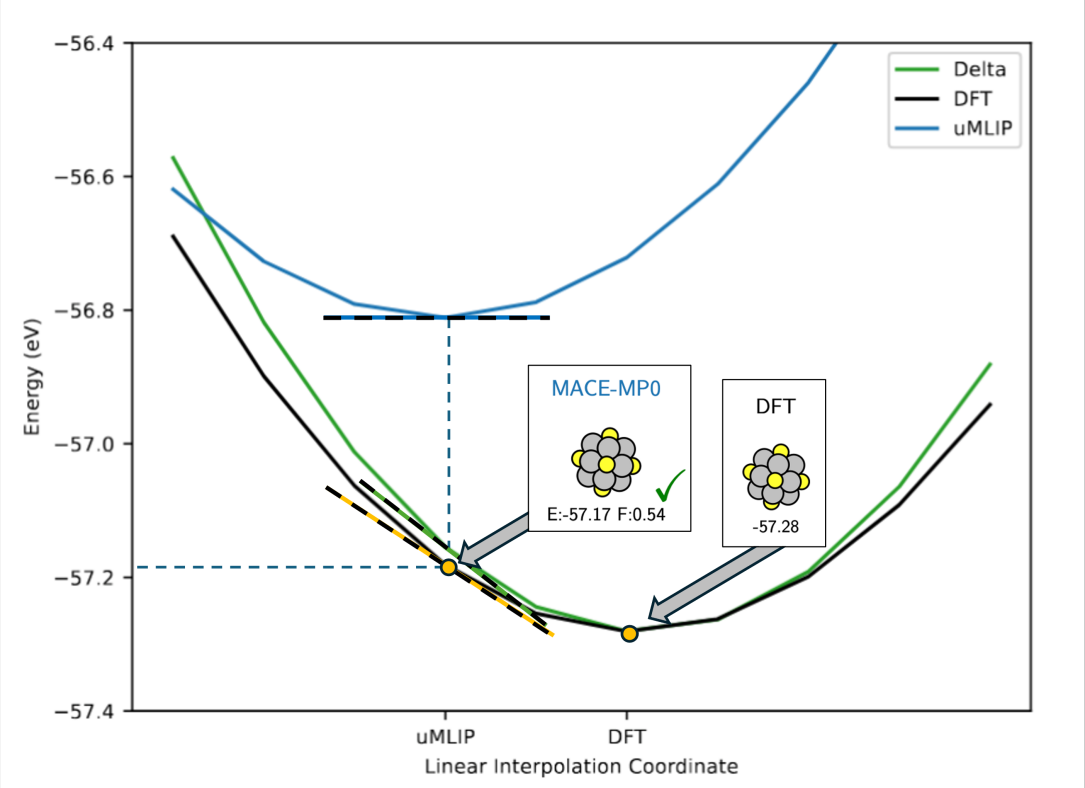

Fig. S6. In this figure, we show the energy predictions for a MACE-MP0 Δ-model, obtained from a single $Ag_{12}S_6$ REX search, on the linear interpolation of structures between and surrounding the DFT/MACE-MP0 GM structures. The curves are annotated with schematic representations of the forces present, corresponding to the information provided in Fig. 2.