Supporting Information for "Charting electronic-state manifolds across molecules with multi-state learning and gap-driven dynamics via efficient and robust active learning"

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Contents

S1 Performance of MS-ANI

Table S1: Performance of the MS-ANI and single-state ANI models for predicting the energies of the first eight electronic states of pyrene, kcal/mol.

State	Single-state RMSE	Multi-state RMSE	Single-state MAE	Multi-state MAE
S_0	1.32538	0.96832	1.01809	0.74647
S_1	1.33954	1.00046	1.04498	0.77016
S_2	1.29239	1.02189	1.01633	0.80134
S_3	2.1493	1.47939	1.6647	1.15242
S_4	1.92178	1.49865	1.48902	1.18519
S_5	1.72372	1.23457	1.35766	0.98109
S_6	1.96994	1.28371	1.52209	1.0147
S_7	1.64697	1.33066	1.28604	1.05033

Table S2: Performance of the MS-ANI and single-state ANI models for predicting the energy gaps of the first eight electronic states of pyrene, kcal/mol.

States	Single-state RMSE	Multi-state RMSE	Single-state MAE	Multi-state MAE
S_0 - S_1	1.72368	1.32452	1.30568	1.02068
S_1 - S_2	1.54319	1.23684	1.1957	0.94517
S_2 - S_3	2.38496	1.89723	1.83622	1.47973
S_3 - S_4	2.73568	2.09397	2.13051	1.68351
S_4 - S_5	2.47677	1.93942	1.91491	1.54836
S_5 - S_6	2.19074	1.42764	1.69054	1.15271
S_6 - S_7	2.1447	1.22756	1.63729	0.97736

Table S3: Difference between the MAE of single-state ANI models and MS-ANI in terms of energies and energy gaps.

Quantity	S_0	S_1	S_2	S_3	S_4	S_5	S_6	S_7
Energy MAE	0.27161	0.27482	0.21499	0.51228	0.30383	0.37657	0.50739	0.2357
Gap MAE	-	0.28500	0.25054	0.35649	0.44701	0.36655	0.53783	0.65992

S2 Uncertainty quantification thresholds

Table S4: Uncertainty quantification thresholds for AL runs described in the main text, in kcal/mol.

System	S_0	S_1	S_2	S_3
Fulvene, MS-ANI	5.58	7.72	-	-
Fulvene, MACE	4.06	9.75	-	-
Ferro-wire	27.02	24.46	21.23	34.79
Azobenzene	11.06	8.75	-	-

S3 Computational performance

Table S5: Computational performance of MS-ANI for the three studied systems, in ps/day.

System	Fulvene	Ferro-wire	Azobenzene
Computational performance [ps/day]	2725	1660	2340