Supplementary Information for overcoming systematic softening in universal machine learning interatomic potentials by fine-tuning

CONTENTS

I. SURFACE BENCHMARK

The table below presents all the compounds and surface energy calculations with corresponding Miller indices, as discussed in the surface benchmark section. The surface energy values are in units of $eV/Å^2$.

mp-id	composition	miller index	M3GNet	CHGNet	MACE	DFT
mp-126	Pt	[1, 1, 1]	0.034337	0.067933	0.108786	0.094540
mp-126	Pt	[2, 2, 1]	0.041738	0.077731	0.120055	0.111288
mp-126	Pt	[2, 1, 0]	0.055588	0.087461	0.132217	0.124666
mp-126	Pt	[1, 1, 0]	0.049405	0.084198	0.129928	0.124951
mp-126	Pt	[2, 1, 1]	0.044232	0.079317	0.122664	0.110808
mp-126	Pt	[1, 0, 0]	0.045627	0.080184	0.129639	0.129515
mp-1639	BN	[1, 1, 0]	0.093675	0.081441	0.127251	0.173043
mp-733	GeO_2	[1, 0, 0]	0.073282	0.081616	0.079633	0.089101
mp-733	GeO_2	[0, 0, 1]	0.087457	0.018861	0.080309	0.029946
mp-733	GeO_2	[1, 0, 1]	0.066760	0.064233	0.062573	0.079778
mp-733	GeO_2	[2, 0, 1]	0.073349	0.076060	0.083530	0.094763
mp-1138	LiF	[2, 1, 0]	0.030142	0.025535	0.028741	0.033885
mp-1138	LiF	[2, 1, 1]	0.057645	0.047527	0.056996	0.067411
mp-1138	LiF	[2, 2, 1]	0.055239	0.043816	0.052588	0.058832
mp-1138	LiF	[1, 1, 0]	0.040302	0.034503	0.040202	0.047889
mp-1138	LiF	[1, 0, 0]	0.017387	0.013148	0.015115	0.019274
mp-19009	NiO	[1, 0, 0]	0.041740	0.034537	0.078835	0.075694
mp-19009	NiO	[1, 1, 0]	0.083462	0.069599	0.112546	0.146961
mp-19009	NiO	[2, 1, 0]	0.068462	0.054417	0.102001	0.116266
mp-19009	NiO	[2, 1, 1]	0.104637	0.087699	0.126782	0.179743
mp-19009	NiO	[2, 2, 1]	0.108441	0.088199	0.129272	0.176103
mp-19399	Cr_2O_3	[2, 1, 1]	0.088672	0.087783	0.103788	0.137950
mp-19399	Cr_2O_3	[0, 0, 1]	0.074112	0.070402	0.085364	0.103951
mp-19399	Cr_2O_3	[1, 0, 0]	0.091601	0.090690	0.099994	0.135110

mp-19399	Cr_2O_3	[2, 1, -2]	0.100278	0.108988	0.115123	0.176547
mp-19399	Cr_2O_3	[2, -1, 2]	0.100240	0.099631	0.113920	0.147635
mp-19399	Cr_2O_3	[2, 0, -1]	0.083573	0.083330	0.095162	0.126717
mp-124	Ag	[2, 1, 0]	0.023318	0.041564	0.040281	0.055204
mp-124	Ag	[2, 1, 1]	0.019012	0.036769	0.035553	0.054525
mp-124	Ag	[1, 1, 1]	0.014562	0.030217	0.028861	0.050079
mp-124	Ag	[1, 1, 0]	0.021264	0.039141	0.037516	0.055697
mp-124	Ag	[1, 0, 0]	0.019419	0.034914	0.034974	0.055207
mp-124	Ag	[2, 2, 1]	0.018113	0.036006	0.034438	0.054188
mp-841	Li_2O_2	[1, 0, 2]	0.046675	0.029191	0.039260	0.054088
mp-841	Li_2O_2	[2, 1, 2]	0.051622	0.032755	0.033052	0.060593
mp-841	Li_2O_2	[1, 0, 0]	0.086174	0.063395	0.065709	0.113353
mp-841	Li_2O_2	[2, 1, 0]	0.059645	0.038821	0.060778	0.070830
mp-841	Li_2O_2	[1, 1, 0]	0.047108	0.030317	0.044707	0.053582
mp-1143	Al_2O_3	[1, 0, -2]	0.086088	0.080818	0.103740	0.101860
mp-1143	Al_2O_3	[2, 1, -2]	0.164204	0.146616	0.166867	0.189583
mp-1143	Al_2O_3	[2, 0, -1]	0.106417	0.097795	0.116902	0.123334
mp-1143	Al_2O_3	[1, 0, 0]	0.111818	0.102915	0.119333	0.127045
mp-1143	Al_2O_3	[2, 1, 2]	0.116533	0.104785	0.130883	0.130165
mp-1143	Al_2O_3	[2, -1, 2]	0.128921	0.124779	0.133044	0.134912
mp-30	Cu	[2, 1, 1]	0.067982	0.065369	0.075433	0.095000
mp-30	Cu	[1, 1, 1]	0.054469	0.052346	0.064314	0.088602
mp-30	Cu	[2, 1, 0]	0.079509	0.075253	0.083459	0.100149
mp-30	Cu	[1, 0, 0]	0.068065	0.063664	0.075394	0.088411
mp-30	Cu	[2, 2, 1]	0.066242	0.063811	0.073944	0.090916
mp-30	Cu	[1, 1, 0]	0.074240	0.070257	0.079108	0.095428
mp-1894	WC	[1, 1, 0]	0.098792	0.143954	0.204065	0.232303
mp-1894	WC	[2, -1, 2]	0.114635	0.145034	0.201485	0.248596
mp-13	Fe	[2, 1, 1]	0.089164	0.093201	0.114475	0.163361
mp-13	Fe	[2, 1, 0]	0.089094	0.089230	0.122766	0.159772

mp-13	Fe	[1, 1, 1]	0.103324	0.107859	0.139042	0.170649
mp-13	Fe	[2, 2, 1]	0.093793	0.097154	0.126544	0.165998
mp-13	Fe	[1, 0, 0]	0.091056	0.095761	0.126991	0.156481
mp-13	Fe	[1, 1, 0]	0.073888	0.072561	0.104191	0.151700
mp-19770	Fe_2O_3	[0, 0, 1]	0.047551	0.048781	0.060852	0.135074
mp-19770	Fe_2O_3	[1, 0, 2]	0.069058	0.066659	0.077375	0.166870
mp-19770	Fe_2O_3	[2, 1, 0]	0.081511	0.081344	0.085917	0.181437
mp-19770	Fe_2O_3	[2, 1, 1]	0.067754	0.065974	0.076866	0.161943
mp-19770	Fe_2O_3	[1, 1, 0]	0.053869	0.052800	0.058509	0.130912
mp-19770	Fe_2O_3	[2, 0, -1]	0.057520	0.057341	0.065502	0.136786
mp-7000	SiO_2	[2, 0, 1]	0.141823	0.110030	0.137782	0.113992
mp-7000	SiO_2	[0, 0, 1]	0.145989	0.137888	0.129574	0.138955
mp-7000	SiO_2	[1, 0, 0]	0.153040	0.123052	0.127043	0.126585
mp-7000	SiO_2	[1, 0, 1]	0.062742	0.122331	0.100668	0.123040
mp-22862	NaCl	[2, 2, 1]	0.025123	0.027284	0.034891	0.030475
mp-22862	NaCl	[1, 1, 0]	0.015496	0.015306	0.026203	0.019928
mp-22862	NaCl	[1, 0, 0]	0.006124	0.007300	0.015636	0.009129
mp-22862	NaCl	[2, 1, 0]	0.010699	0.012592	0.023277	0.015305
mp-22862	NaCl	[2, 1, 1]	0.024815	0.025764	0.033700	0.029353
mp-135	Li	[1, 0, 0]	0.010798	0.020859	0.023871	0.031838
mp-135	Li	[1, 1, 0]	0.009445	0.020027	0.025692	0.032474
mp-135	Li	[1, 1, 1]	0.013633	0.024637	0.031637	0.035152
mp-135	Li	[2, 2, 1]	0.012928	0.024098	0.030276	0.034000
mp-135	Li	[2, 1, 0]	0.010720	0.021599	0.026752	0.032470
mp-135	Li	[2, 1, 1]	0.011725	0.023398	0.027753	0.036125
mp-149	Si	[2, 1, 1]	0.034148	0.042500	0.059773	0.102888
mp-149	Si	[1, 1, 0]	0.023614	0.027412	0.050974	0.110291
mp-149	Si	[2, 2, 1]	0.030926	0.037354	0.056349	0.101352
mp-149	Si	[1, 1, 1]	0.021687	0.023641	0.041046	0.094757
mp-149	Si	[2, 2, 1]	0.023567	0.027063	0.047707	0.098151

mp-149	Si	[2, 1, 0]	0.036716	0.046091	0.060881	0.092782
mp-804	GaN	[2, 1, 0]	0.080403	0.065448	0.091784	0.103071
mp-804	GaN	[1, 0, 0]	0.076561	0.062626	0.084137	0.097966
mp-804	GaN	[1, 0, 0]	0.218620	0.166211	0.178998	0.242491
mp-804	GaN	[2, 1, 0]	0.131125	0.107050	0.125705	0.158080
mp-804	GaN	[1, 1, 0]	0.079229	0.064616	0.091921	0.102659
mp-8062	SiC	[1, 1, 0]	0.075063	0.136799	0.159305	0.181694
mp-81	Au	[2, 2, 1]	0.014611	0.025625	0.034807	0.051627
mp-81	Au	[1, 0, 0]	0.016261	0.026563	0.039800	0.058522
mp-81	Au	[1, 1, 0]	0.016423	0.027964	0.039814	0.058177
mp-81	Au	[1, 1, 1]	0.013162	0.022272	0.031630	0.049855
mp-81	Au	[2, 1, 1]	0.015337	0.026356	0.036176	0.054002
mp-81	Au	[2, 1, 0]	0.017355	0.029708	0.040748	0.056562
mp-560588	ZnS	[1, 1, 0]	0.015619	0.024400	0.031528	0.022952
mp-560588	ZnS	[1, 0, 0]	0.055910	0.056939	0.066455	0.070868
mp-560588	ZnS	[1, 0, 0]	0.017069	0.024991	0.029862	0.023617
mp-560588	ZnS	[2, 1, 0]	0.016443	0.025202	0.031710	0.023441
mp-560588	ZnS	[2, 1, 0]	0.030725	0.037819	0.045833	0.040012
mp-32	Ge	[2, 2, 1]	0.006605	0.020387	0.034671	0.063867
mp-32	Ge	[2, 1, 1]	0.012166	0.027648	0.041963	0.066657
mp-32	Ge	[1, 1, 0]	0.007173	0.022050	0.036765	0.077589
mp-32	Ge	[2, 1, 0]	0.009510	0.023996	0.040576	0.059703
mp-32	Ge	[1, 1, 1]	0.005607	0.017935	0.030079	0.067034
mp-32	Ge	[2, 2, 1]	0.008362	0.021126	0.038605	0.062519
mp-1282	VC	[2, 1, 1]	0.093263	0.141647	0.129182	0.201552
mp-1282	VC	[1, 0, 0]	0.035456	0.053319	0.082388	0.074836
mp-1282	VC	[1, 1, 0]	0.082658	0.123809	0.110264	0.186008
mp-1282	VC	[2, 1, 0]	0.063290	0.089403	0.096957	0.133421
mp-1282	VC	[2, 2, 1]	0.096111	0.150875	0.131032	0.221928
mp-672	CdS	[1, 0, 0]	0.012007	0.013867	0.020398	0.020337

mp-672	CdS	[2, 1, 0]	0.011091	0.013713	0.021748	0.020396
mp-672	CdS	[2, 1, 0]	0.025093	0.023691	0.032142	0.033403
mp-672	CdS	[1, 1, 0]	0.010189	0.013384	0.021426	0.019865
mp-672	CdS	[1, 0, 0]	0.050317	0.038558	0.048120	0.056875
mp-127	Na	[1, 1, 1]	0.003365	0.010644	0.015300	0.016682
mp-127	Na	[2, 2, 1]	0.003111	0.009781	0.014608	0.014414
mp-127	Na	[1, 0, 0]	0.003661	0.008913	0.012753	0.012650
mp-127	Na	[1, 1, 0]	0.002443	0.007490	0.012011	0.013206
mp-127	Na	[2, 1, 1]	0.002638	0.009106	0.014097	0.014246
mp-127	Na	[2, 1, 0]	0.003077	0.008722	0.013129	0.012457
mp-510408	MnO_2	[1, 0, 2]	0.081781	0.094930	0.097742	0.145764
mp-510408	MnO_2	[0, 0, 1]	0.082990	0.082874	0.083948	0.109491
mp-510408	MnO_2	[2, 1, 1]	0.063035	0.060607	0.061940	0.077323
mp-510408	MnO_2	[1, 0, 0]	0.049726	0.047126	0.040640	0.059818
mp-510408	MnO_2	[1, 0, 1]	0.062869	0.058052	0.054840	0.081812
mp-510408	MnO_2	[2, 0, 1]	0.066334	0.063617	0.061789	0.085650
mp-390	TiO_2	[1, 0, 1]	0.059326	0.064625	0.079015	0.067956
mp-390	TiO_2	[2, 0, 1]	0.055256	0.069411	0.074662	0.067849
mp-390	TiO_2	[1, 0, 2]	0.072560	0.084707	0.096908	0.091490
mp-390	TiO_2	[1, 1, 2]	0.036109	0.041427	0.048947	0.044573
mp-390	TiO_2	[2, 2, 1]	0.076046	0.071177	0.082080	0.078796
mp-390	TiO_2	[1, 0, 1]	0.025502	0.029338	0.036004	0.028007
mp-66	C	[1, 1, 0]	0.163739	0.130145	0.223326	0.321780
mp-66	C	[2, 1, 0]	0.313806	0.280531	0.298584	0.355383
mp-66	C	[2, 1, 1]	0.256012	0.244349	0.241830	0.360210
mp-66	C	[2, 2, 1]	0.183087	0.137429	0.154165	0.301921
mp-66	C	[1, 1, 1]	0.585351	0.699314	0.595576	0.768543
mp-66	C	[1, 1, 1]	0.208489	0.140293	0.134630	0.317213
mp-19079	CoO	[1, 1, -1]	0.039383	0.045610	0.067481	0.180209
mp-19079	CoO	[1, -1, 1]	0.017043	0.024503	0.038119	0.302340

mp-19079	CoO	[2, 1, 1]	0.048293	0.055873	0.070322	0.209607
mp-19079	CoO	[1, 0, 2]	0.038874	0.045037	0.064643	0.156739
mp-19079	CoO	[1, 2, 0]	0.027128	0.035143	0.055198	0.191367

Supplementary Table I: Surface Energy Benchmark

II. DEFECT BENCHMARK

The table below presents all the compounds and defect types that have been benchmarked and included in the defect benchmark section. The defect energy values are in units of eV.

Composition	Defect Type	M3GNet	CHGNet	MACE	DFT
Na	Vacancy	0.098512	0.183401	0.430823	0.263000
Na	Vacancy	0.120645	0.477933	0.517838	0.455000
Cd	Vacancy	-0.012552	0.153877	0.140947	0.320000
K	Vacancy	Failed	0.223832	0.210363	0.513000
Bi	Vacancy	-0.139624	-0.579367	-0.392083	0.313000
Bi	Vacancy	-0.044672	0.206740	0.372396	0.922000
In	Vacancy	-0.002449	0.100538	0.274098	0.345000
In	Vacancy	0.012018	0.099818	0.283277	0.503000
Sn	Vacancy	0.325116	-1.151863	0.368152	1.384000
Ti	Vacancy	-16.462205	0.421127	1.776438	2.951000
V	Vacancy	2.332988	1.732452	2.584888	2.328000
Al	Vacancy	0.154319	0.141712	0.475685	0.262000
Cu	Vacancy	0.888738	0.718282	1.042848	1.038000
Zn	Vacancy	0.083270	0.145502	0.247718	0.521000
Pb	Vacancy	-0.018524	0.147121	0.162681	0.155000
Ti_4Sn_2	Vacancy_Sn	1.076550	1.107670	0.711711	0.554576
Ti_4Sn_2	Anti_Site_Ti_Sn	1.064445	1.139473	1.139240	0.423105
Na_4Au_2	Vacancy_Au	0.844603	0.871855	1.098101	1.216742
Na_4Au_2	Interstitial_Na_2	0.616568	0.407622	0.923492	1.014546
Na_4Au_2	Anti_Site_Na_Au	0.899678	1.060992	1.060488	1.137531
Na_4Au_2	Interstitial_Au_2	-0.153044	-0.420460	0.133420	0.487577
Na_4Au_2	Vacancy_Na	0.315789	0.436413	0.509782	0.633643
Na_4Au_2	Anti_Site_Au_Na	-0.335439	-0.208590	0.005736	0.015104
Cu_1Au_3	Vacancy_Au	0.263364	0.206642	0.609131	0.575274
Cu_1Au_3	Interstitial_Au_1	3.010926	3.071402	4.541988	5.189230

Cu_1Au_3	Vacancy_Cu	0.283198	0.296684	0.395783	0.236827
Cu_1Au_3	Anti_Site_Cu_Au	-0.078672	-0.076843	-0.074373	-0.056926
Cu_1Au_3	Interstitial_Cu_1	2.152988	2.014165	2.967708	3.505615
Cu_1Au_3	Anti_Site_Au_Cu	0.145199	0.138924	0.162504	0.130139
Al_1Ni_3	Anti_Site_Ni_Al	1.811493	1.829491	1.940757	1.927665
Al_1Ni_3	Interstitial_Ni_3	3.963899	2.815650	3.165469	4.009906
Al_1Ni_3	Vacancy_Al	3.019099	2.937929	3.083741	3.560679
Al_1Ni_3	Interstitial_Ni_1	3.781316	3.281920	3.391787	3.916801
Al_1Ni_3	Interstitial_Al_3	2.294657	1.329771	1.290916	2.547053
Sc_1Ag_4	Anti_Site_Sc_Ag	-0.676006	-0.379025	-0.456614	0.279505
Sc_1Ag_4	Anti_Site_Ag_Sc	0.937587	1.222941	0.879038	1.320786
Sc_1Ag_4	Vacancy_Sc	1.138478	1.738402	1.627161	2.100641
Sc_1Ag_4	Interstitial_Sc_1	0.668270	1.482140	1.516516	2.692651
Sc_1Ag_4	Interstitial_Ag_2	1.787543	2.515844	2.690293	3.230396
Sc_1Ag_4	Interstitial_Sc_2	0.808300	1.746278	1.902472	2.819566
Sc_1Ag_4	Vacancy_Ag	0.359384	0.858395	0.866256	1.153118
Sc_1Ag_4	Interstitial_Ag_1	1.342566	1.928889	2.003800	2.719110
Na_2Ag_4	Interstitial_Na_3	3.426612	2.914153	4.045333	7.173276
Na_2Ag_4	Interstitial_Ag_3	3.589606	2.753056	3.987016	6.627579
Na_2Ag_4	Anti_Site_Ag_Na	0.762848	0.529697	0.716249	3.272565
Na_2Ag_4	Interstitial_Na_2	0.868781	0.654781	1.221725	3.835408
Na_2Ag_4	Interstitial_Na_1	0.897589	0.678202	1.200698	3.927602
Na_2Ag_4	Anti_Site_Na_Ag	0.286011	0.068325	0.332060	2.970150
Na_2Ag_4	Interstitial_Ag_2	0.981940	0.665227	1.398667	4.052211
Na_2Ag_4	Vacancy_Ag	0.420526	0.353202	0.650741	3.394687
Na_2Ag_4	Interstitial_Ag_1	1.028540	0.713818	1.246110	3.936566
Na_2Ag_4	Vacancy_Na	0.815698	0.817853	0.965256	3.767730
Na_1Pb_3	Interstitial_Pb_1	0.597958	0.686051	1.253197	4.105730
Na_1Pb_3	Interstitial_Na_3	0.072221	-0.138940	0.284905	3.408402
Na_1Pb_3	Vacancy_Pb	0.171348	0.252982	0.217706	2.875408

Na_1Pb_3	Interstitial_Na_2	0.348191	0.284684	0.866631	3.561036
Na_1Pb_3	Interstitial_Pb_3	0.438412	0.138439	0.755181	3.760386
Na_1Pb_3	Interstitial_Na_1	0.353013	0.413453	0.972740	3.464343
Na_1Pb_3	Anti_Site_Na_Pb	-0.238924	-0.289047	-0.247616	2.198242
Na_1Pb_3	Anti_Site_Pb_Na	0.326345	0.423483	0.576669	3.048626
Na_1Pb_3	Vacancy_Na	0.483890	0.614678	0.707204	3.331136
Cu_3Au_1	Vacancy_Au	1.175890	0.908096	1.421057	1.475802
Cu_3Au_1	Interstitial_Cu_2	1.205956	1.026899	1.247107	1.788362
Cu_3Au_1	Interstitial_Au_1	3.505941	3.032763	3.669466	4.727760
Cu_3Au_1	Interstitial_Cu_3	1.232872	1.077133	1.293764	1.750586
Cu_3Au_1	Vacancy_Cu	0.514480	0.411493	0.754741	0.737859
Cu_3Au_1	Anti_Site_Cu_Au	0.270316	0.211728	0.212101	0.288406
Cu_3Au_1	Interstitial_Cu_1	2.728051	2.363988	2.803438	3.432098
Cu_3Au_1	Interstitial_Au_3	1.268331	1.109756	1.421449	2.056506
Cu_3Au_1	Anti_Site_Au_Cu	0.044663	0.040125	-0.010673	0.106534
Ca_2Al_4	Vacancy_Ca	-0.768699	-0.341304	-0.059612	1.685380
Ca_2Al_4	Interstitial_Ca_3	4.724327	5.036987	5.465129	9.071019
Ca_2Al_4	Interstitial_Ca_1	3.991722	4.259613	4.920391	7.516047
Ca_2Al_4	Interstitial_Ca_2	4.111412	4.415603	5.098552	7.625161
Ca_2Al_4	Interstitial_Al_1	-0.585865	-0.480255	0.083618	2.318874
Ca_2Al_4	Vacancy_Al	2.580433	2.735560	2.615052	4.740785
Ca_2Al_4	Anti_Site_Al_Ca	-2.793529	-2.712170	-2.269451	-0.492433
Ca_2Al_4	Anti_Site_Ca_Al	4.335521	4.429608	4.298504	6.443542
Ca_2Al_4	Interstitial_Al_3	-0.600391	-0.423151	0.098938	2.667655
Ca_2Al_4	Interstitial_Al_2	-0.657154	-0.487158	0.077697	2.318743
Ca_2Sn_2	Vacancy_Sn	0.584064	0.974910	1.261180	1.553902
Al_2Au_1	Vacancy_Au	1.318419	1.511772	1.853213	2.587571
Al_2Au_1	Anti_Site_Au_Al	0.029184	-0.002333	0.340075	0.643577
Al_2Au_1	Interstitial_Al_1	0.492729	0.585401	0.963298	0.978751
Al_2Au_1	Vacancy_Al	0.184090	0.235345	0.782002	1.531555

Al_2Au_1	Interstitial_Au_1	0.012178	0.104948	0.643674	0.376088
Al_2Au_1	Anti_Site_Al_Au	1.247092	1.470930	1.352705	1.582227
Ca_1Sn_3	Vacancy_Ca	1.625287	1.740235	1.795373	1.576081
Ca_1Sn_3	Vacancy_Sn	0.175633	0.474164	0.468784	0.570375
Ca_1Sn_3	Interstitial_Ca_3	-0.345485	-1.693932	-1.071052	0.274956
Ca_1Sn_3	Interstitial_Ca_1	-0.325099	-0.226075	1.537926	3.046946
Ca_1Sn_3	Interstitial_Sn_1	0.476850	0.309163	1.380965	2.848980
Ca_1Sn_3	Interstitial_Sn_3	0.744946	-0.741909	0.349440	1.840613
Ca_1Sn_3	Anti_Site_Ca_Sn	-1.190536	-1.044185	-0.754178	-0.433526
Ca_1Sn_3	Anti_Site_Sn_Ca	1.608705	1.403155	1.904647	2.042548
Ca_1Sn_3	Interstitial_Sn_2	0.691297	-0.804876	0.376417	1.846830
V_6Sn_2	Anti_Site_V_Sn	0.012456	0.672186	0.361463	-0.364769
Cu_3Sn_1	Vacancy_Sn	-0.352039	-0.353140	-0.141500	-0.298237
Cu_3Sn_1	Anti_Site_Cu_Sn	-0.155258	0.013906	0.054421	-0.186690
Cu_3Sn_1	Vacancy_Cu	-0.674016	-0.757391	-0.465360	-0.228936
Cu_3Sn_1	Anti_Site_Sn_Cu	0.188827	0.321726	0.836204	1.860569
V_6Pb_2	Anti_Site_V_Pb	-1.016977	-1.387503	-1.752316	-2.004045
Al_1Ni_1	Anti_Site_Ni_Al	0.882606	0.529421	0.577434	1.052144
Al_1Ni_1	Anti_Site_Al_Ni	0.825402	1.133420	1.605916	1.351988
Al_1Ni_1	Interstitial_Al_1	2.831688	2.820018	3.927898	4.645804
Al_1Ni_1	Vacancy_Al	1.449867	1.371163	1.698506	2.517611
Al_1Ni_1	Interstitial_Ni_1	2.559204	2.205117	2.777731	3.398817
Al_1Ni_1	Vacancy_Ni	0.877869	1.008799	1.083292	1.073481
Cu_2Sn_2	Vacancy_Sn	0.237682	0.878650	1.188014	1.615946
Cu_2Sn_2	Interstitial_Sn_1	1.107487	0.916038	1.392385	2.243797
Cu_2Sn_2	Anti_Site_Cu_Sn	0.481003	0.725072	1.045189	1.061208
Cu_2Sn_2	Interstitial_Cu_1	0.117741	-0.020504	0.021802	0.284972
Pb_4Au_2	Vacancy_Au	-0.234607	0.051087	-0.162155	0.153663
Pb_4Au_2	Interstitial_Pb_1	-0.088194	0.296182	1.066526	1.733611
Pb_4Au_2	Vacancy_Pb	0.488616	0.824970	0.794894	1.061960

Pb_4Au_2	Anti_Site_Pb_Au	-0.656511	-0.367160	-0.451769	-0.056997
Pb_4Au_2	Anti_Site_Au_Pb	0.834245	1.016932	1.043932	1.135593
Pb_4Au_2	Interstitial_Pb_2	-0.067717	0.342806	1.096555	1.825586
Sc_1Au_1	Vacancy_Au	1.668059	1.584674	1.803652	3.120869
Sc_1Au_1	Vacancy_Sc	0.831120	0.875999	1.744867	2.944137
Sc_1Au_1	Interstitial_Au_1	0.452058	0.293085	0.952268	2.801970
Sc_1Au_1	Anti_Site_Au_Sc	-0.044703	-0.071914	0.143485	1.473809
Sc_1Au_1	Interstitial_Sc_1	1.381893	1.335726	1.515021	3.516906
Sc_1Au_1	Anti_Site_Sc_Au	1.594263	1.091141	1.407540	2.823275
Cu_1Au_1	Vacancy_Au	0.668143	0.620434	1.091845	0.925546
Cu_1Au_1	Vacancy_Cu	0.289417	0.299326	0.610156	0.446427
Cu_1Au_1	Anti_Site_Cu_Au	0.175731	0.248570	0.167038	0.070113
Cu_1Au_1	Interstitial_Cu_1	1.237851	1.143912	1.604200	2.153533
Cu_1Au_1	Anti_Site_Au_Cu	0.122731	0.166622	0.152603	0.162015

Supplementary Table II: Defect Energy Benchmark

III. PHONON VIBRATIONAL FREQUENCIES

The table below presents the maximum phonon frequencies for 229 materials we benchmarked in the main text, in units of terahertz. Note that the high K-path for each material is fixed for all the calculation methods tested.

mp-id	composition	M3GNet	CHGNet	MACE	DFT
mp-30300	$Cs_3As_5O_9$	19.498332	24.005564	23.229576	24.927849
mp-8725	$HfSnS_3$	7.657295	9.178484	9.465515	10.164333
mp-27400	$RbCdBr_3$	3.642681	5.328183	4.996765	5.268003
mp-677179	$BaNaIn_2(PO_4)_3$	28.857168	33.666028	33.054118	34.365236
mp-28891	Li_7TaO_6	20.093920	22.002941	21.573040	24.231359
mp-5487	Sr_3SiO_5	26.222964	26.882499	26.888053	29.115291
mp-1265	MgO	15.662517	16.278631	15.695725	18.291675
mp-4662	ScOF	15.671689	18.296357	21.997349	22.139717
mp-558267	$NaLiSiB_3HO_8$	92.727786	101.787191	104.762855	100.384036
mp-1666	Th_2S_5	9.221056	13.162428	8.516954	11.320001
mp-600033	SiO_2	36.172144	36.462391	36.458076	37.155855
mp-4511	La_2SO_2	9.693148	11.547466	12.845953	15.446820
mp-9518	NdZnPO	11.433350	9.894731	12.743912	16.731082
mp-504922	$UPbO_4$	17.486796	20.738354	23.312188	23.420553
mp-556784	ZnS	8.006407	7.490294	8.658517	10.572585
mp-6065	Sr_2GaSbO_6	21.618999	21.540622	22.654707	24.148578
mp-754710	Sr_2CaI_6	3.050949	4.510335	4.416511	5.465568
mp-778364	$Na_6Ti_2O_7$	21.213630	24.014932	23.937547	24.458999
mp-684015	Bi_9AuBr_9	3.290173	3.182236	4.440894	4.538602
mp-559719	$LiPr(SO_4)_2$	31.933009	34.832244	35.630050	37.624216
mp-2782	ZnP_2	11.789810	9.618362	9.067072	14.253712
mp-626412	$In(HO)_3$	87.250615	94.680967	94.588404	90.223996
mp-8191	Cs_2PtF_6	15.318945	15.381937	16.090010	17.761747
mp-561394	$SrTeO_3$	16.810258	19.622328	22.457252	23.468848

mp-760184	$Na_4Ti_5O_{12}$	17.825455	21.701842	21.465890	22.635884
mp-2798	SiP	10.653343	10.959714	11.114200	16.275461
mp-571240	$KLa_5(CCl_5)_2$	32.153622	32.831233	32.150644	30.967270
mp-18610	Sm_2SiSeO_4	23.901672	26.313458	26.820909	29.187366
mp-570301	AgBr	3.179476	2.914059	3.534861	4.193857
mp-754646	$Dy_2Ge_2O_7$	16.479185	22.374415	22.242575	22.490935
mp-8179	$LiTlO_2$	15.326730	14.617085	16.473533	15.976848
mp-23980	$Cs_3H(SeO_4)_2$	36.640962	43.133931	55.710913	44.702111
mp-3164	$BaSO_4$	28.914065	33.371809	34.502927	35.237333
mp-24566	$MgH_8(IO_5)_2$	94.332392	98.942998	99.952573	98.799605
mp-541732	$Al_3Pb_5F_{19}$	16.463954	17.777449	18.303016	19.899125
mp-4056	$CaSc_2O_4$	15.069466	20.001733	20.702613	20.823375
mp-2697	SrO_2	23.807163	21.223020	27.831482	28.063326
mp-5368	Li_2GeF_6	15.420387	17.317993	17.026133	19.082456
mp-642996	Li_4H_6Os	41.664978	40.603076	50.062121	58.342346
mp-755759	ZrO_2	15.006227	21.482262	20.626738	22.190519
mp-531840	$MgAl_2O_4$	22.187780	24.143385	23.303942	26.222239
mp-17035	$K_3Sc(PO_4)_2$	30.067616	34.385703	32.649151	34.201410
mp-654956	$CaAlH_5$	42.712351	45.674957	49.621533	52.024846
mp-571390	Cs_2LiCl_3	8.802312	9.558510	10.040789	11.162049
mp-8985	$Ba_2YC_2(O_2F)_3$	40.817986	44.656237	45.939592	47.272489
mp-5967	TiCoSb	6.249633	5.088310	6.568478	9.713533
mp-557301	$K_2Ti(Si_2O_5)_3$	29.910673	33.030238	32.802950	34.951543
mp-567710	KPr_3Te_8	3.232482	2.975411	3.710943	4.696342
mp-558128	KNd_2NbO_6	20.622653	21.484120	22.015203	23.610312
mp-5285	$SrSO_4$	28.421384	35.767180	34.707110	35.820274
mp-1224	HgO	13.104979	14.316341	16.864588	18.019610
mp-13000	Ba_2HoTaO_6	22.003758	22.393686	23.435321	24.275170
mp-23660	$KNa_{22}C_2S_9ClO_{42}$	40.137678	42.816564	41.346072	44.235735
mp-561121	$CsTi_3P_5O_{19}$	33.018993	33.890238	34.679923	36.648114

mp-554272	Cu_3SbS_3	7.752338	8.585867	9.951112	10.806783
mp-755362	Sm_2Se_2O	9.101126	10.807402	11.962620	14.161454
mp-23803	$GaHO_2$	83.721835	77.070048	80.893042	67.860536
mp-582080	Cs_3LaCl_6	6.405247	7.266318	7.162932	7.830748
mp-561499	Dy_3CuSnS_7	8.571407	9.107696	10.333545	11.653242
mp-29143	Na_3VS_4	10.802963	13.899317	14.233054	15.375085
mp-2789	NO_2	43.612687	49.851184	53.718656	55.614781
mp-27275	Li_3BO_3	35.204417	38.486714	38.305306	40.973100
mp-2672	K_2O_2	21.370411	24.922505	23.134637	25.516471
mp-570536	$Rb_2AgAu_3I_8$	5.414692	5.042288	5.687573	5.852634
mp-4608	KPF_6	23.647910	24.428451	24.211795	26.312823
mp-754266	Li_8TiS_6	8.377273	10.912754	12.038215	13.665810
mp-14052	$Ca_3In_2(SiO_4)_3$	24.095039	27.362317	26.674481	28.783854
mp-2741	CaF_2	10.703395	10.877435	11.998484	13.675218
mp-31276	Cd_2PCl_2	8.927222	11.132247	9.742696	14.175288
mp-571164	$Rb_3AgSn_3Se_8$	5.631309	6.739810	6.861746	7.825334
mp-22766	$CaIn_2O_4$	13.792242	18.701847	16.494154	17.374150
mp-16557	$SrAlF_5$	18.157312	19.666204	19.827642	21.274673
mp-6037	Ba_2SmSbO_6	21.569757	21.795696	21.672128	22.614055
mp-556806	$BaNa_2(SiO_3)_2$	29.293107	30.380491	29.798118	31.306275
mp-650026	$PrIn_2Cl_5$	5.822375	6.218022	7.139283	7.447356
mp-11114	$KCuPdSe_5$	5.706417	6.196719	6.725145	8.265870
mp-7652	Na_5TlO_4	12.011351	12.249753	14.114787	14.991474
mp-753360	BiOF	11.453941	14.890706	14.830247	15.946454
mp-7249	$Ba_3Nb_2ZnO_9$	20.348832	23.088228	23.389268	23.751718
mp-17196	$TiAs_2O_7$	19.686094	27.750446	28.863078	29.101985
mp-775918	$LaTiNO_2$	16.714191	22.132294	24.851736	26.829656
mp-29892	$LiSb_3O_8$	15.951147	21.683347	22.397243	24.686726
mp-756624	Ba_2SrI_6	2.076098	2.823466	2.922523	3.730838
mp-11695	$Rb_2Pd_3S_4$	7.986149	9.355513	9.203478	10.823883

mp-752753	SbO_2F	19.971860	24.697845	23.023076	25.664722
mp-21943	$Ba(InS_2)_2$	7.077408	7.359352	9.309312	10.608161
mp-558781	$ScTaO_4$	19.412596	24.411689	23.620404	25.787249
mp-6586	K_2NaAlF_6	15.217256	16.045300	15.875068	17.984555
mp-558428	$Ba_9Sc_2(SiO_4)_6$	27.900467	30.505614	29.123236	31.692278
mp-22694	$LiPPbO_4$	27.275948	30.382271	30.345163	31.963742
mp-661	AlN	19.674519	22.882298	20.244257	25.587593
mp-674508	$RbPbF_3$	11.190651	11.274886	11.143496	12.164374
mp-6233	$NaSr_3RuO_6$	16.600006	17.174052	17.224984	18.868252
mp-2784	Na_2Te	3.211512	4.735223	4.699274	6.061981
mp-979932	Sr_3SiO_5	26.256988	27.340525	27.049567	29.563165
mp-14636	Rb_2NbAgS_4	9.998213	11.491546	11.864735	13.160797
mp-541837	Bi_2Se_3	3.786642	4.242295	4.899462	5.522900
mp-569346	CuI	4.516904	4.125959	3.959441	4.953035
mp-40484	$NaNd_3Ti_2(SbO_7)_2$	18.146363	23.676385	22.384537	24.069777
mp-2739	TeO_2	14.360576	20.833018	18.504872	22.853212
mp-2659	LiN_3	62.882655	69.710957	65.875811	70.973477
mp-2998	$BaTiO_3$	20.745865	21.651299	22.237633	22.835234
mp-17651	$HoPtF_7$	17.199820	18.153760	18.556052	20.108489
mp-2892	$BaNd_2O_4$	14.912531	15.740207	16.178788	17.059619
mp-769262	K_5YO_4	13.474036	16.380128	15.986780	16.906324
mp-4809	Ga_2HgS_4	8.163614	8.646606	9.305835	11.330612
mp-780246	$Na_2Bi_4O_7$	13.311275	15.299807	17.506300	18.683400
mp-541081	$NaMg_2H_3(SO_4)_2$	105.429526	101.646147	106.011376	104.333704
mp-30004	Rb_4Br_2O	11.246984	8.485107	8.242653	9.581160
mp-644285	$KAlH_2CO_5$	86.162067	103.645932	103.250247	100.988552
mp-673682	Tb_2YbS_4	7.696280	7.786482	8.672647	9.483012
mp-7084	SrCaSi	4.979672	5.986852	5.723764	7.962960
mp-556282	Ba_2AlInO_5	29.415501	27.138005	29.053588	30.715893
mp-17446	$PrPtF_7$	16.596853	19.237362	18.100520	19.708645

mp-23465	Cs_2TeCl_6	7.391135	9.398186	7.676194	8.374533
mp-570448	CdI_2	2.871324	3.539854	3.754721	4.475238
mp-6175	$BaLi_2P_2O_7$	31.092281	33.421573	33.555968	34.675902
mp-1105	BaO_2	26.485322	19.633321	26.841741	27.699630
mp-27716	KTlO	8.826034	11.429552	9.826243	12.808043
mp-23029	$Cs_3Sb_2I_9$	3.372674	3.569566	4.288871	5.220400
mp-16847	$K_3Na_2SnP_3$	6.610451	6.905020	7.517071	9.115734
mp-618177	K_2ZnCl_4	7.687193	8.730602	8.814666	9.813377
mp-754196	Ba_2SrI_6	4.402462	3.124910	3.393254	3.969172
mp-29008	Li_6MgBr_8	7.579386	9.899036	9.970130	10.836195
mp-541040	$Y_2Be_2GeO_7$	24.257677	28.267514	28.476573	30.158182
mp-756122	Cs_3TlO_3	12.305235	13.062627	13.748112	15.086195
mp-558054	$SrLi_2Ta_2O_7$	22.841178	25.973971	25.957104	27.701731
mp-8590	K_2HfF_6	15.626526	16.677679	17.091821	17.726222
mp-3193	$Na_2Si_2O_5$	31.221700	31.953395	31.976841	33.947892
mp-2763	Nd_2O_3	15.144629	16.528169	16.732876	17.257567
mp-4192	Ca_3AsN	7.802000	11.599185	11.793036	14.618685
mp-7765	Ba_3SiO_5	24.157841	24.908949	25.677704	27.992085
mp-696195	YSeF	10.870769	12.625762	13.092855	14.330367
mp-756148	$Sm_2Pt_2O_7$	15.760187	16.246733	18.942525	19.672175
mp-755659	$DyHO_2$	90.854067	91.099401	97.759026	87.812769
mp-556179	$Pr_3Si_2S_8Cl$	10.391575	11.122965	13.997019	16.738493
mp-582753	Tl_2Te	2.127984	2.518933	2.711719	3.356966
mp-9607	Li_2UO_4	19.187211	21.756339	22.709055	23.424290
mp-568612	$SnHg_6(P_2Cl_3)_2$	7.579738	8.537377	8.308079	14.275036
mp-35659	$HgPbF_6$	12.602519	13.957240	14.058152	16.191283
mp-8614	$K_3NdSi_2O_7$	28.401351	29.736088	28.949918	30.746816
mp-5309	$KAuF_4$	16.361213	19.264957	17.575548	18.842919
mp-733604	$Ba_2Mg_3H_{10}$	33.112571	40.976294	39.396168	42.492648
mp-5387	$YNbO_4$	19.100370	24.541295	24.364710	25.249000

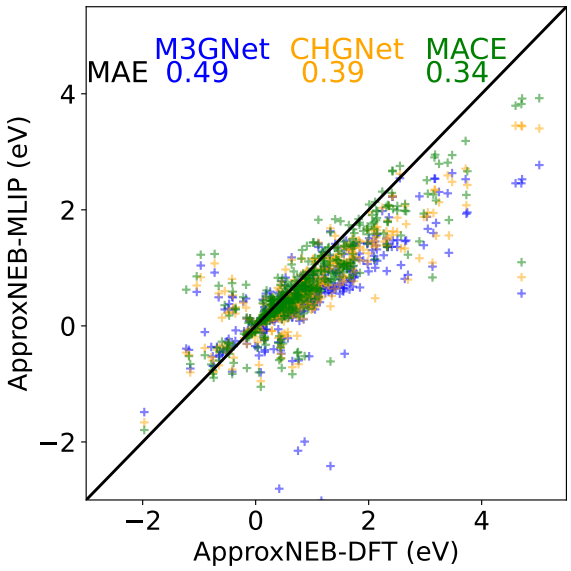
mp-17747	Y_3CuSnS_7	8.677936	9.529684	10.323484	11.606889
mp-6169	$Ba_3Ta_6(Si_2O_{13})_2$	32.099686	35.386415	34.591289	36.287347
mp-30905	$Ba_3(BN_2)_2$	48.429440	45.992541	48.238694	51.318387
mp-23297	BrF_3	14.707614	18.006457	16.206366	19.516196
mp-752501	$LiSiBiO_4$	17.520473	25.876954	27.081228	30.021508
mp-28995	Cu_4SiP_8	7.514909	9.181596	10.921656	15.409987
mp-556423	$Na_2Si_2O_5$	30.977654	32.024351	31.896352	34.325569
mp-6875	$BaNa_2Al_4(SiO_4)_4$	28.977915	31.726806	31.340085	33.182984
mp-4473	$Ba(CuSe)_2$	4.198372	4.363719	5.342831	6.202939
mp-12637	Ba_2NdNbO_6	21.629287	21.652415	23.095729	24.194169
mp-861618	$BaBiBS_4$	19.269514	19.139399	23.573749	27.129328
mp-7293	$HgPSe_3$	9.109851	11.659809	10.549999	13.481428
mp-570044	NbI_4	5.084630	4.815090	5.397571	7.258930
mp-554082	$Ho_2Hf_2O_7$	19.661509	25.868937	23.697774	24.387995
mp-4718	$NdTaO_4$	19.795819	22.715476	23.282827	24.703618
mp-760311	Na_7SbO_6	15.936250	17.658392	17.693253	18.216444
mp-505607	$In_2Ag_2GeSe_6$	5.434015	6.254502	7.117750	9.189476
mp-20790	$InPS_4$	10.085534	11.291916	12.743677	17.157412
mp-753520	Li_3BiS_3	6.507469	9.512007	10.809531	13.817166
mp-5970	$Ba(CuS)_2$	8.375245	7.501258	9.007692	9.310916
mp-9077	$Nd_2Be_2SiO_7$	24.118093	27.559569	29.952582	31.567342
mp-540631	$KTa(GeO_3)_3$	23.052293	26.754670	27.371169	28.803721
mp-756117	Li_2TiTeO_6	16.259084	20.082248	19.394177	24.092821
mp-758868	$MgBP(H_3O_4)_2$	91.686383	101.845958	101.005491	101.897993
mp-1392	ZnP_2	11.541882	10.403141	9.711955	13.944481
mp-3700	$LiYF_4$	14.017649	14.752331	15.357062	17.042099
mp-17885	$Ba_2Ge_2Te_5$	4.605541	4.591247	6.458764	7.953390
mp-3707	$CaGe_2O_5$	20.569384	24.065241	24.682670	25.426306
mp-864617	NdI_3	3.574539	3.936739	4.160366	5.358786
mp-558325	$K_2YNb_5O_{15}$	23.622335	29.406875	29.836962	29.012973

mp-22956	K_2PdCl_4	8.455659	8.383192	9.221956	10.345035
mp-571667	CdI_2	2.871245	3.542372	3.759112	4.475804
mp-540881	Cs_2AgI_3	2.443763	2.469741	2.869041	3.653978
mp-8774	Rb_3CO_3F	39.895937	42.915725	40.775895	41.463011
mp-28490	Tl_3BSe_3	11.821513	19.797006	19.129984	21.712173
mp-553342	$CsBe_2BO_3F_2$	32.996902	39.164675	39.883361	41.570730
mp-5338	$ZnSeO_3$	17.112570	21.430390	22.738495	24.727762
mp-27033	$Li_2SnP_2O_7$	30.655747	33.026538	34.264959	35.631151
mp-545346	$LiGaAs_2O_7$	22.205481	26.748083	27.278868	28.740909
mp-554825	$K_2SnP_2O_7$	28.824142	32.296493	34.130862	35.088915
mp-9263	$KErTe_2$	3.199691	3.597238	3.615759	5.151028
mp-667369	$Ca_2Zr_5Ti_2O_{16}$	18.826108	24.135513	23.185026	24.673120
mp-6574	$Er_2C(NO)_2$	57.470893	66.739418	65.673907	66.978486
mp-643727	HPbBrO	101.843460	108.568779	107.276480	107.362559
mp-555211	SiO_2	35.286678	36.462953	35.768687	36.992401
mp-2691	CdSe	4.219424	5.234043	5.124339	5.876200
mp-541097	$Zr_3Pb(O_2F_3)_2$	15.890349	19.030853	19.178444	20.032133
mp-571162	$Ca_3(GaN_2)_2$	13.774918	15.144857	17.274702	20.675739
mp-765970	$Ti_3Sn_7O_{20}$	16.064156	20.932704	21.672893	22.941323
mp-2657	TiO_2	17.788619	24.198959	22.215556	25.051936
mp-23451	$Cs_3Cr_2Br_9$	7.296592	6.519230	8.265594	9.384516
mp-10429	$LaYO_3$	14.265984	17.101780	18.068090	18.884700
mp-696329	$RbCa(H_2N)_3$	95.501864	105.309778	102.647165	101.816651
mp-557758	$LaMg(BO_2)_5$	32.872326	39.687567	41.653019	43.105451
mp-7297	Cs_2SnF_6	14.514149	14.969242	15.852455	16.612606
mp-583454	$Nb_2Bi_2PbO_9$	20.949899	24.894232	24.951274	26.148640
mp-680674	$Cs_3Bi_2Br_9$	4.730753	5.014445	5.135577	5.783075
mp-676154	$Yb(DyS_2)_2$	8.214336	7.797222	8.800157	9.544256
mp-23969	ZnH_2SeO_5	78.705468	93.685044	91.429204	90.989839
mp-662535	$PbSe_2O_5$	17.757292	22.836639	23.712959	25.065754

mp-19941	As_2PbS_4	6.240551	8.326194	8.597937	10.106015
mp-20337	$ZrPbO_3$	18.005835	22.941491	23.316683	24.054353
mp-6332	Li_2TiSiO_5	25.429806	28.124967	29.953335	30.962226
mp-723064	$Ti_3(BiO_3)_4$	19.457918	22.671688	23.662213	25.590143
mp-28295	$CsIO_3$	19.226609	21.377101	22.535252	23.326606
mp-6265	Ba_2TbSbO_6	21.243219	22.157103	21.865874	22.627028
mp-568543	Na_2SnSe_3	4.751692	6.582593	6.892233	7.921720
mp-2667	CsAu	1.139430	1.398299	1.727349	2.348112
mp-768695	$Ho_2Ti_2O_7$	18.055287	23.625852	24.461490	25.190504
mp-505121	$Na_2Hg_3S_4$	7.028159	8.750792	9.903008	10.473579
mp-29213	$Cd_2Ge_7O_{16}$	18.736704	24.066233	24.078755	25.581670
mp-752484	YbB_4O_7	30.813165	34.494088	33.305846	39.685472
mp-558543	$Ca(CO_2)_2$	45.483431	49.360940	48.133331	51.710054
mp-626724	$LaHO_2$	100.587769	108.547023	109.167605	110.052210
mp-27281	LiH_3O_2	100.353605	110.793879	108.617340	108.772105
mp-540634	$TaTl(GeO_3)_3$	23.386024	27.441990	27.383030	28.764906
mp-684679	Sb_6O_{13}	15.866763	22.749707	23.396280	26.529795
mp-18101	$K_2Cd_3S_4$	6.821617	7.844766	8.103051	9.343665
mp-3491	$NbSbO_4$	20.594411	23.797489	25.903608	27.072541
mp-861914	$Ba_3Co(CN)_3$	45.097845	41.752742	52.882651	52.354671
mp-551873	Ca_4Bi_2O	6.471748	6.589033	10.028025	12.138939
mp-984055	$Ca_7(H_6Cl)_2$	25.810438	29.535589	34.375237	34.512397
mp-3107	Ga_3PO_7	25.725351	31.823884	32.725755	33.602760
mp-8866	$La_{10}Se_{19}$	4.500700	6.313055	4.549349	5.808983
mp-13282	$Sm(ErSe_2)_3$	4.714418	5.320010	5.702891	6.585084

Supplementary Table III: Phonon Max Frequencies

IV. ION MIGRATION BARRIERS



Supplementary Figure 1. Parity plot of the kinetic resolved activation barriers calculated in uMLIPs *vs.* DFT.

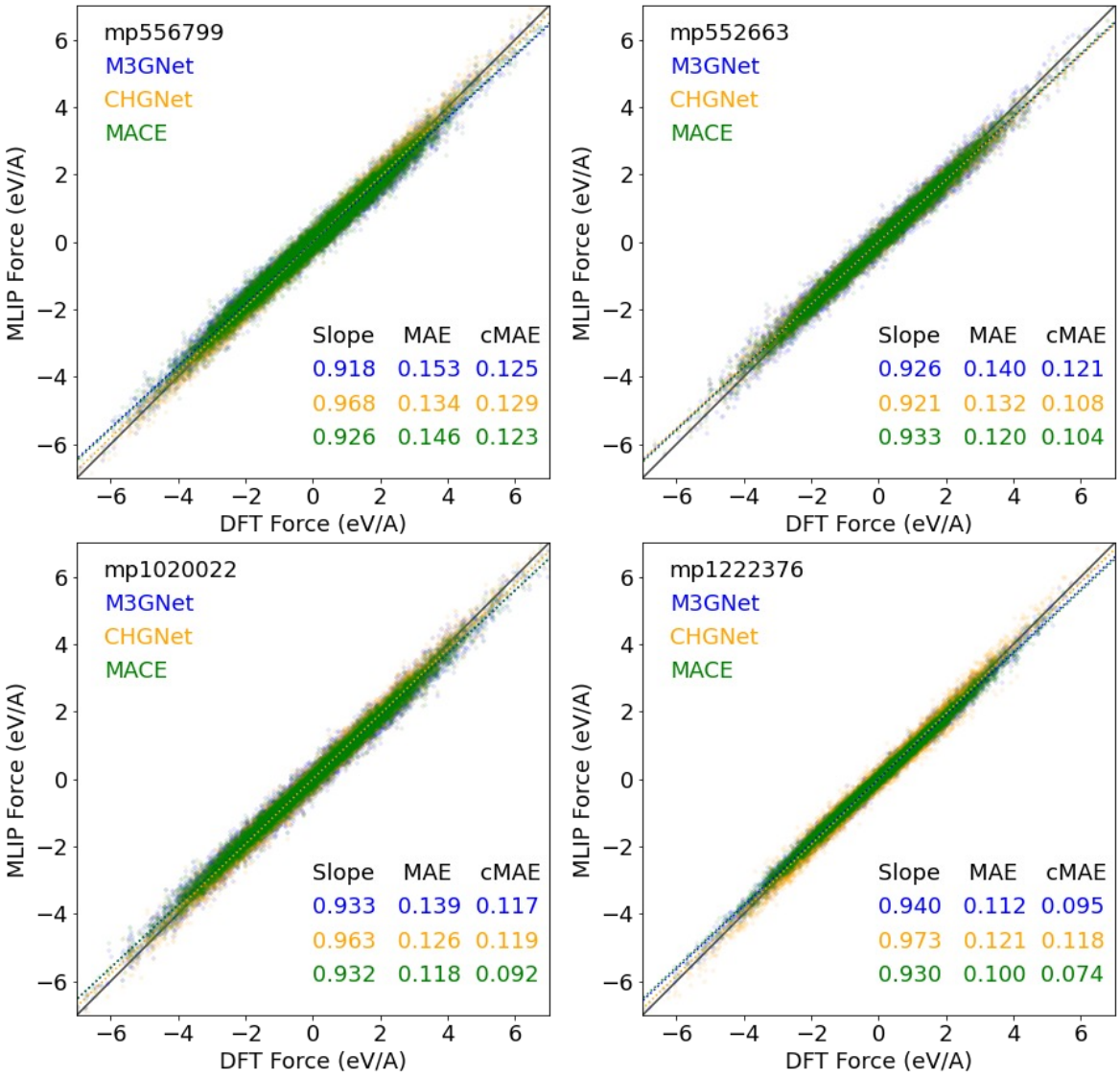
Supplementary Figure 1 shows the comparison between the KRA barriers calculated using uMLIPs and DFT. The majority of uMLIP ion migration barriers are underestimated compared to the DFT results. Some KRA barriers are predicted to be negative due to the approximated NEB method being used.

V. SOFTENING SCALES

Supplementary Fig. 2 shows four more examples of softening scales from high-energy states sampled in high-temperature MDs like Fig 5(a) in main text. These structures and AIMD trajectory are taken from the work of Jun *et al.* [1].

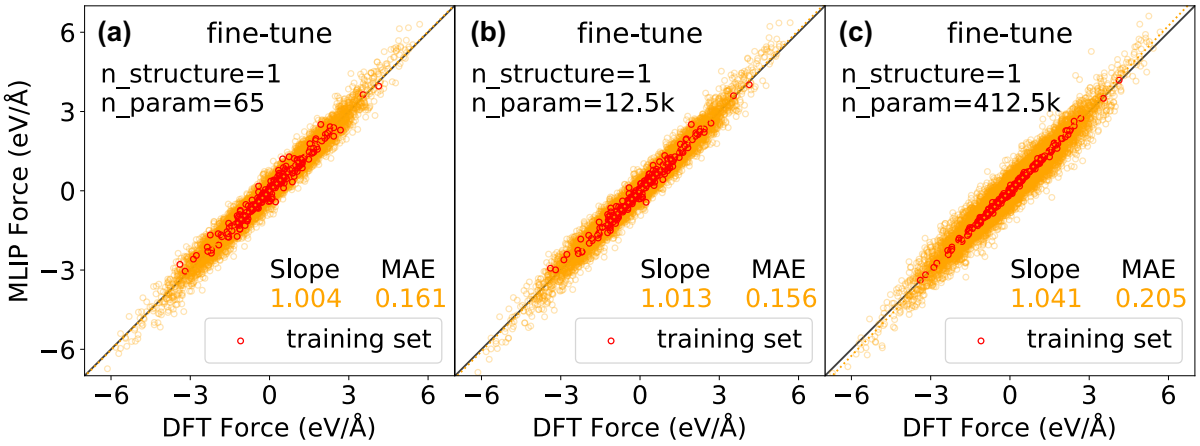
VI. FINE-TUNING VS TRAINING FROM-SCRATCH

To better show advantage of pre-trained uMLIPs in achieving data-efficient error reduction, we present a series of experiments comparing the behavior of force errors in fine-tuned CHGNet models to those trained from scratch. In Supplementary Fig. 3(a-c), We use one



Supplementary Figure 2. More examples of softening scales from high temperature MDs.

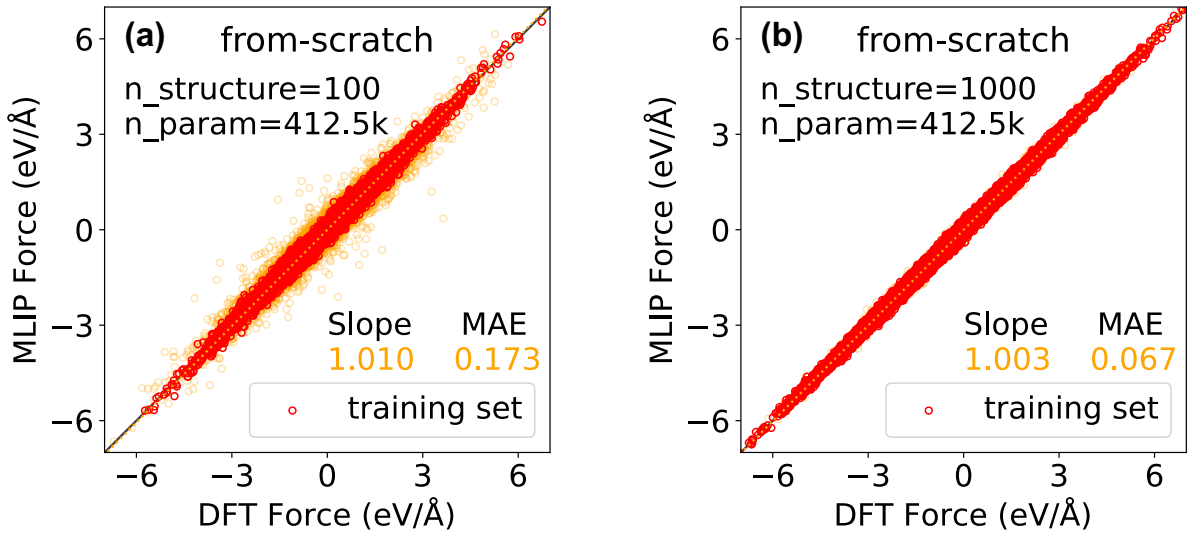
structure label to fine-tune the pre-trained CHGNet by three different strategies: optimizing one 64-dimensional-linear layer (a), one non-linear layer(b), and all model parameters(c). The red points denote training force labels, and the orange points denote test force labels. The width of the red and orange distribution indicates the magnitude of training error and test error. Compared to the linear correction shown in Fig. 6(a) in the main text, we show the MAEs can be further reduced when an increased fraction of model parameters are allowed to be optimized, until reaching the overfitting case shown in Supplementary Fig.



Supplementary Figure 3. **Finetuing part of CHGNet *vs.* training a CHGNet from scratch.** (a-c) Fine-tuning the CHGNet by optimizing only one 64-dimensional linear layer (a), a full readout non-linear layer(b), and all the model weights (c) with 1 training structure.

3(c), where all the 0.4 million CHGNet parameters are fine-tuned with only 1 structure. The number of trainable parameters in the uMLIPs determines the trade-off between model expressivity and the magnitude of over-fitting. When all model parameters are allowed to be optimized, the uMLIP can be largely adjusted to accommodate the PES regions described by training atomic configurations, at the cost of possible degradation in the PES regions OOD from training. Since solving the systematic errors requires optimizing only a limited fraction of the model, fine-tuning can be performed highly effectively by freezing a large fraction of uMLIP parameters, therefore achieving considerable error reduction while avoiding over-fitting issues.

Supplementary Fig. 4(a) shows the performance of randomly initialized CHGNets trained with 100 high-energy structural configurations. The fine-tuned model with only 1 structure label, as shown in Supplementary Fig. 3(a-b), is shown to outperform a trained-from-scratch model with 100 more times data points. Such observation aligns with the previously observed robustness of uMLIPs [2]. When a substantial amount of data is available, as shown in the case of Supplementary Fif. 3(b) with 1000 structural configurations, the training distribution (red) overlaps with the test distribution (orange), and therefore the test set is no longer out-of-distribution. In such cases, training all the uMLUP parameters from scratch does not suffer from the overfitting issue, and the effect of parameter-freezing is minimized.



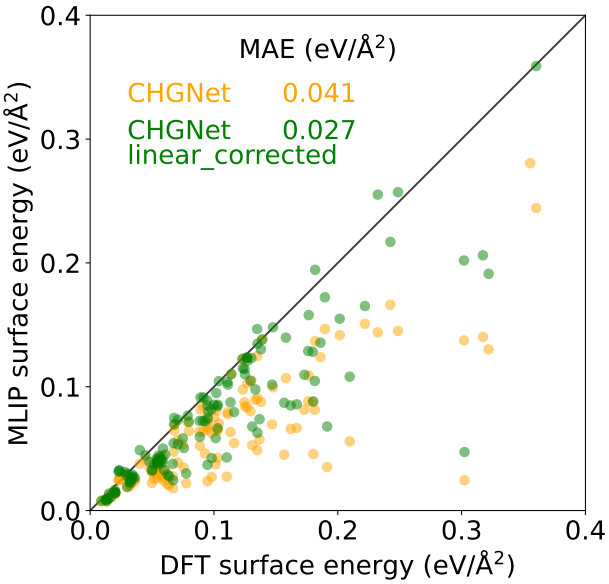
Supplementary Figure 4. Training the randomly initialized CHGNet by optimizing all the model weights with 100 (a), and 1000 (b) training structures.

VII. LINEAR CORRECTED CHGNET FOR SURFACE CALCULATION

In this section, we provide an additional experiment to show that the linear correction not only decreases force MAEs, but can also increase the accuracy in calculating surface energy.

Specifically, we performed high-temperature CHGNet MD to sample the high-energy bulk structure for each of the materials discussed in the surface benchmark section. The corresponding softening scales were extracted for each material separately, and we calculated the surface energy for each material using a customized linearly corrected CHGNet. The resulting surface energy predictions from these linear corrected CHGNet are compared to DFT and pretrained CHGNet. Note that direct extraction of the softening scale from surface configurations can be highly challenging, as the forces in slabs are typically not large enough to show a clear systematic tilting.

As shown in Supplementary Fig. 5, the surface energy error is greatly reduced after the linear correction using one single high-energy bulk structure. A complete resolution of the underestimation in the surface calculations requires more significant effort to also resolve the random errors. A fine-tuning process with a larger dataset would be required.



Supplementary Figure 5. Application of linear corrected CHGNet for surface energy calculation as compared to pretrained CHGNet. Substantial error reduction in surface energy can be achieved with the linear correction with 1 label.

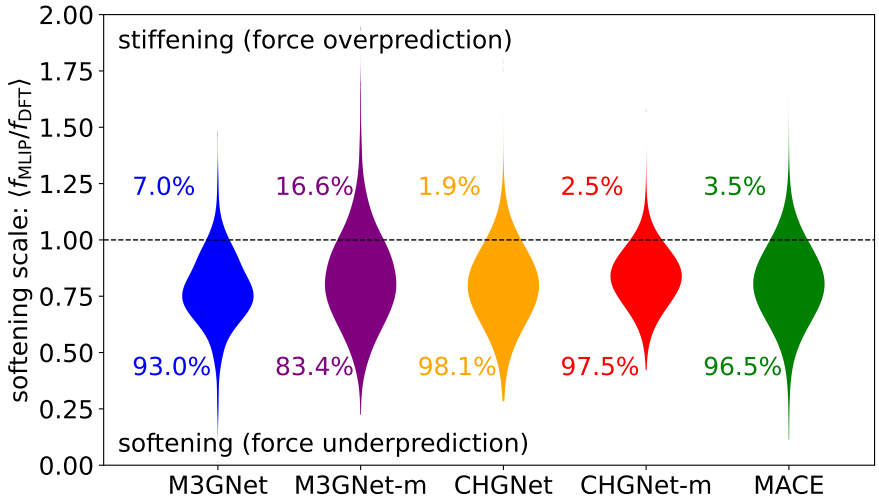
As a result, the proposed linear correction represents a baseline example of typical fine-tuning processes. The observation of systematic error correction therefore explains the commonly observed data-efficiency in fine-tuning uMLIPs.

VIII. EFFECT OF MODEL SIZE

Supplementary Fig. 6 shows the distribution of softening scales including `M3GNet-matgl` (denoted as `M3GNet-m`) and `CHGNet-matgl` (denoted as `CHGNet-m`). `CHGNet-matgl` is trained with the same MPtrj dataset [3], but is increased to 11 million parameters in contrast to the original CHGNet with 0.4 million parameters as shown in the main text. `M3GNet-matgl` has 1.1 million parameters and is trained with sampled data from the MPF dataset [4].

A clear decrease in softening is observed for `CHGNet-matgl`, which has a significant increase in the model parameters compared to the previous CHGNet. A similar trend is also observed for `M3GNet-matgl`, which shows a smaller enhancement compared to the previous version.

The force MAEs of the two new models on WBM compounds are presented in Supplementary Fig. 7. The `CHGNet-matgl` with 11 million parameters shows a clearly improved



Supplementary Figure 6. Distribution of softening scales sampled from 1000 compounds in the WBM dataset, extended from Fig. 5(b) of main text with `M3GNet-matgl` model and `CHGNet-matgl` model.

force prediction performance.

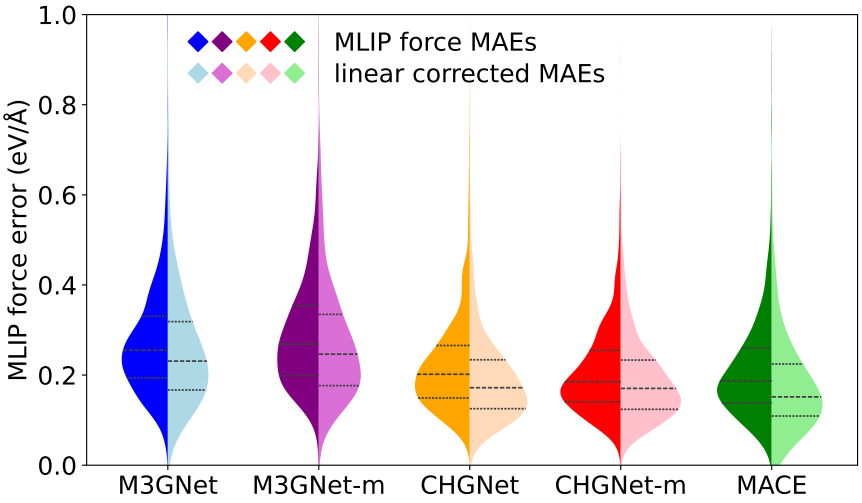
---

[1] K. Jun, Y. Sun, Y. Xiao, Y. Zeng, R. Kim, H. Kim, L. J. Miara, D. Im, Y. Wang, and G. Ceder, Lithium superionic conductors with corner-sharing frameworks, Nature Materials **21**, 924–931 (2022).

[2] A. Merchant, S. Batzner, S. S. Schoenholz, M. Aykol, G. Cheon, and E. D. Cubuk, Scaling deep learning for materials discovery, Nature , 1–6 (2023).

[3] B. Deng, P. Zhong, K. Jun, J. Riebesell, K. Han, C. J. Bartel, and G. Ceder, Chgnet as a pretrained universal neural network potential for charge-informed atomistic modelling, Nature Machine Intelligence **5**, 1031 (2023).

[4] C. Chen and S. P. Ong, A universal graph deep learning interatomic potential for the periodic table, Nature Computational Science **2**, 718–728 (2022).



Supplementary Figure 7. Distribution of force MAEs and linear corrected MAEs (cMAEs) for the 1000 WBM compounds, extended from Fig. 6(e) of main text with `M3GNet-matgl` model and `CHGNet-matgl` model. Quartiles are labeled by dashed lines.