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Suitability of the available interatomic potentials for the modeling of 2D materials

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ABSTRACT

Most interatomic potentials, both classical and machine learning-based (MLPs), are parameterized for 3D structures. The question naturally arises whether they are suitable for modeling their 2D allotropes. In the present study, using *ab initio* calculations, I determined the structural and mechanical properties of 2D phases of materials such as MoS2, Si, Ge and Sn and then investigated whether the available potentials are able to reproduce these properties.

2D Molybdenum Disulphide (MoS_2)

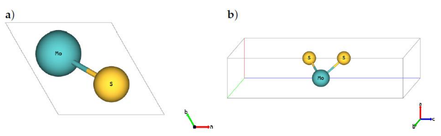

Figure 1. Single-layer (SL) 1H-MoS_2. (a) Top and (b) 3D view.

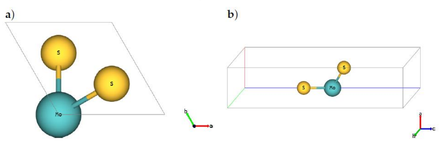

Figure 2. SL 1T-MoS_2. (a) Top and (b) 3D view.

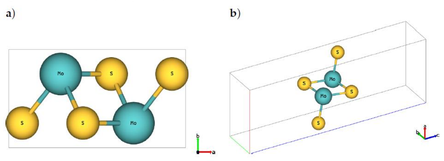

Figure 3. SL 1T′-MoS_2. (a) Top and (b) 3D view.

Table 1. Structural and mechanical properties of SL MoS_2 phases from density functional theory (DFT) calculations: lattice parameters a,b (Å), average cohesive energy E_c (eV/atom), average bond length d (Å), average height h (Å), 2D elastic constants C_{ij} (N/m) and 2D Kelvin moduli K_i (N/m).

Polymorph	1H			1T			1T′		
Source	Present	Exp.	DFT	Present	Exp.	DFT	Present	Exp.	DFT
a	3.165	3.157 [a]	3.183 [b]	3.194		3.179 [b]	5.751		5.717 [b]
b	3.165	3.157 [a]	3.183 [b]	3.194		3.176 [b]	3.177		3.179 [b]
$-E_c$	5.64		5.35 [a]	5.52			5.56		
d_{Mo-S}	2.403	2.38 [a]	2.43 [a]	2.422		2.430 [c]	2.415 ‡		
h_{S-S}	3.120	3.116 [a]	3.11 [a]	3.142		3.184 [c]	3.364		
C_{11}	126.5		127.2 [d]	84.1		103.8 [d]	68.1		94.0 [d]
C_{22}	126.5		127.2 [d]	84.1		103.8 [d]	78.9		119.2 [d]
C_{12}	28.5		25.8 [d]	5.0		−2.5 [d]	18.2		17.2 [d]
C_{44}	49.0		51.0 [d]	39.6		52.8 [d]	43.2		37.5 [d]
K_I	155.0			89.1			90.9		
K_{II}	98.0			79.1			56.1		
K_{III}	98.0			79.1			86.4		

[a] Ref. [50], [b] Ref. [55], [c] Ref. [56], [d] Ref. [57]. ‡ average first-neighbour bond lengths calculated with *cutoff* radius = 3.5 and number of histogram bins = 50.

Table 4. Structural and mechanical properties of SL 1T′-MoS_2 from molecular calculations: lattice parameters a,b (Å), average cohesive energy E_c (eV/atom), average bond lengths d (Å), average height h (Å), 2D elastic constants C_{ij} (N/m), 2D Kelvin moduli K_i (N/m), mean absolute percentage error (MAPE) (%).

Method	DFT	SW2013	SW2015	SW2016	SW2017	REBO	SNAP	ReaxFF
a	5.751	4.944	5.757	5.263	5.728 †	5.563	5.321 †	5.609
b	3.177	3.062	3.148	3.172	3.307 †	3.245	3.072 †	3.209
$-E_c$	5.56	3.02	0.55	1.87	4.96	6.93	2.31	4.83
d_{Mo-S} ‡	2.415	2.399	2.406	2.504	2.42	2.468	2.476	2.490
h_{S-S}	3.364	4.641	5.173	4.142	2.973	3.781	3.454	3.399
C_{11}	68.1	1.1	0.0	60.4	121.8	56.8	437.1	120.1
C_{22}	78.9	100.5	37.6	94.6	121.8	113.0	437.1	255.7
C_{12}	18.2	1.1	0.0	20.3	28.6	23.1	6.1	68.1
C_{44}	43.2	27.1	0.0	26.9	46.6	70.5	215.5	6.4
K_I	90.9	100.5	37.6	88.4	150.4	121.3	443.2	194.3
K_{II}	56.1	1.1	0.0	66.6	93.2	48.5	431.0	181.5
K_{III}	86.4	54.2	0.0	53.8	93.2	141.0	431.0	12.8
MAPE $_{1T'}$		42.070	63.020	20.110	30.399	25.395	249.177	91.913
∑MAPE		127.830	151.074	102.187	100.522	104.840	483.573	325.504

† Input 1T′ converges to 1T. ‡ average first-neighbour bond lengths calculated with *cutoff* radius = 3.5 and number of histogram bins = 50.

$$\mathrm{MAPE} = \frac{100\%}{n}\sum_{t=1}^{n}\left|\frac{\mathrm{Value}^{\mathrm{DFT}} - \mathrm{Value}^{\mathrm{potential}}}{\mathrm{Value}^{\mathrm{DFT}}}\right|$$

1. SW2013: the Stillinger–Weber potential
2. SW2015
3. SW2016
4. SW2017
5. REBO: the reactive many-body potential
6. ReaxFF: the reactive force-field
7. SNAP: the machine-learning-based spectral neighbour analysis potential

[1] M. Maździarz - Transferability of interatomic potentials for silicene, Materials 14, 519 (2021)

Silicene - 2D Silicon (Si)

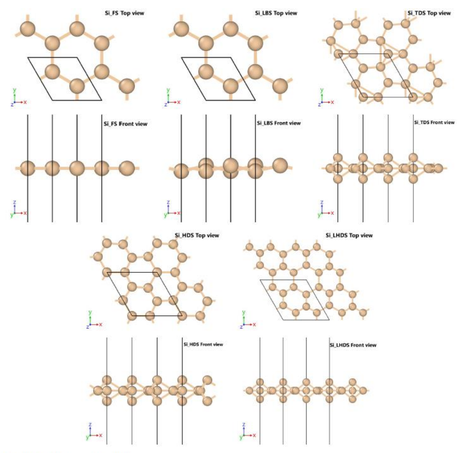

Figure 1: Polymorphs of silicene.

Table 1: Structural and mechanical properties of flat (FS), low-buckled (LBS), trigonal dumbbell (TDS), honeycomb dumbbell (HDS) and large honeycomb dumbbell (LHDS) silicene phases from density functional theory (DFT) calculations: lattice parameters a,b (Å), average cohesive energy E_c (eV/atom), average bond length d (Å), average height h (Å), 2D elastic constants C_{ij} (N/m), 2D Young's modulus E (N/m), Poisson's ratio ν and 2D Kelvin moduli K_i (N/m).

Polymorph	FS		LBS		TDS		HDS		LHDS	
Source	This work	Refs.	This work	Refs.	This work	Refs.	This work	Refs.	This work	Refs.
a	3.855	3.90[a]	3.828	3.87[a], 3.83[b]	6.434	6.52[b]	6.297	6.38[b]	7.334	7.425[b]
b	3.855	3.90[a]	3.828	3.87[a], 3.83[b]	6.434	6.52[b]	6.297	6.38[b]	7.334	7.425[b]
$-E_c$	4.562	4.764[a]	4.577	4.784[a], 5.16[b]	4.679		4.679		4.769	
$d^\dagger$	2.225		2.249	2.25[b]	2.331		2.399		2.357	
h	0.0	0.0[a]	0.421[a]	0.45[a], 0.44[b]	2.734		2.635		2.683	
C_{11}	84.8		69.2		100.5		141.6		104.5	
C_{22}	84.8		69.2		100.5		141.6		104.5	
C_{12}	40.6		22.1		52.3		96.4		52.7	
C_{33}	22.1		23.6		24.1		22.6		25.9	
E	65.4		62.2	61.8[a]	73.3		76.0		77.9	
ν	0.48		0.32	0.31[a]	0.52		0.68		0.50	
K_I	125.4		91.3		152.8		238.0		157.2	
K_{II}	44.3		47.1		48.3		45.2		51.8	
K_{III}	44.3		47.1		48.3		45.2		51.8	

[a] Ref. [47], [b] Ref. [7].
† An average bond lengths calculated using radial pair distribution function with a *cut-off* radius = 3.0 Å and a number of histogram bins = 1000 [31].

Table 6: Structural and mechanical properties of large honeycomb dumbbell silicene (LHDS) from molecular calculations: lattice parameters a, b (Å), average cohesive energy E_c (eV/atom), average bond length d (Å), average height h (Å), 2D elastic constants C_{ij} (N/m), 2D Kelvin moduli K_i (N/m), mean absolute percentage error (MAPE) (%), relative performance measured as normalized timesteps/second in molecular dynamics (MD) simulation.

Method	DFT	Tersoff 1988	Tersoff 2007	Tersoff 2017	MEAM 2007	MEAM 2011	SW 1985	SW 2014	EDIP	ReaxFF	COMB	ML-IAP SNAP	ML-IAP qSNAP	ML-IAP SO(3)	ML-IAP ACE
a	7.334	7.000†	7.249	7.236	7.900†	7.363	7.403	7.062	7.705	7.167	7.422	7.648	7.741	7.427	7.393
b	7.334	6.978†	7.249	7.236	7.560†	7.363	7.403	7.062	7.705	7.167	7.422	7.648	7.741	7.427	7.393
$-E_c$	4.769	4.468	3.897	4.004	3.505	3.911	3.399	2.602	4.113	3.623	3.646	4.804	4.794	4.698	0.370
d	2.357	2.381	2.387	2.369	2.627	2.407	2.456	2.345	2.438	2.370	2.454	2.436	2.403	2.385	2.515
h	2.683	2.692	3.109	3.050	3.050	2.857	3.250	3.100	2.637	3.112	2.994	2.538	2.562	2.700	2.712
C_{11}	104.5	2.3	78.5	68.6	45.6	88.0	84.5	73.0	53.7	99.0	53.1	44.6	43.8	25.9	59.8
C_{22}	104.5	7.3	78.5	68.6	56.2	88.0	84.5	73.0	53.7	99.0	53.1	44.6	43.8	25.9	59.8
C_{12}	52.7	-13.1	42.3	25.5	22.0	29.9	46.1	38.9	36.0	40.4	31.7	16.1	19.1	0.4	15.2
C_{33}	25.9	9.0	18.1	21.6	14.0	29.0	19.2	17.1	8.8	29.3	10.7	14.2	12.3	12.7	22.3
K_I	157.2	18.5	120.8	94.1	73.6	117.9	130.6	111.9	89.7	139.4	84.8	60.6	62.8	26.4	75.0
K_{II}	51.8	17.8	36.2	43.1	28.8	58.1	38.4	34.1	17.6	58.6	21.5	28.5	24.7	25.5	44.7
K_{III}	51.8	-8.5*	36.2	43.1	27.4	58.1	38.4	34.1	17.6	58.6	21.5	28.5	24.7	25.5	44.7
$\mathrm{MAPE}_{\mathrm{LHDS}}$		55.693	18.373	20.287	34.499	13.636	16.741	23.855	33.228	10.809	33.466	33.219	34.603	40.928	29.310
∑MAPE		223.826	129.306	150.256	164.364	82.553	130.721	155.483	165.722	78.185	137.365	181.422	205.224	201.861	124.310
timesteps/s		387.2	382.7	355.2	505.7	416.9	904.9	1753.8	2032.1	23.6	26.4	7.9	4.7	4.2	1.0

† Potential does not reproduce the correct symmetry of the structure (a≠b),
* Negative Kelvin moduli K_i indicating a lack of mechanical stability.

1. Tersoff1988: the Tersoff potential
2. Tersoff2007
3. Tersoff2017
4. MEAM2007: the modified embedded-atom method potential
5. MEAM2011
6. SW1985: the Stillinger–Weber potential
7. SW2014
8. EDIP: the environment-dependent interatomic potential
9. ReaxFF: the reactive force-field
10. COMB: the charge optimized many-body potential
11. SNAP: the machine-learning-based spectral neighbour analysis potential
12. qSNAP
13. SO(3): the smooth power spectrum potential
14. ACE: the atomic cluster expansion potential

[2] M. Maździarz - Transferability of interatomic potentials for silicene, Beilstein J. Nanotechnol. 14, 574–585 (2023)

Germanene - 2D Germanium (Ge)

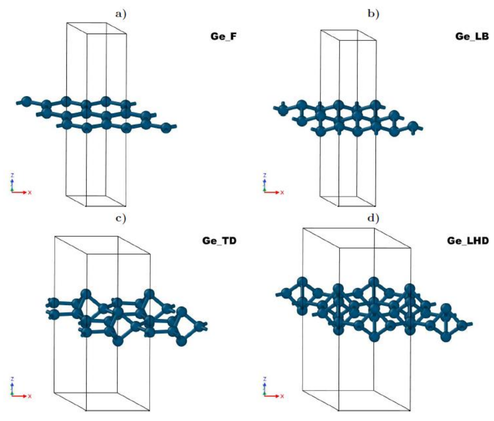

FIG. 1. Polymorphs of germanene: a) flat (F), b) low-buckled (LB), c) trigonal dumbbell (TD), d) large honeycomb dumbbell (LHD).

TABLE I. Structural and mechanical properties of flat (F), low-buckled (LB), trigonal dumbbell (TD) and large honeycomb dumbbell (LHD) germanene phases from density functional theory (DFT) calculations: lattice parameters a,b (Å), average cohesive energy E_c (eV/atom), average bond length d (Å), average height h (Å), 2D elastic constants C_{ij} (N/m), 2D Young's modulus E (N/m), Poisson's ratio ν and 2D Kelvin moduli K_i (N/m).

Polymorph	F		LB		TD		LHD	
Source	This work	Refs.	This work	Refs.	This work	Refs.	This work	Refs.
a	4.066		3.955	4.06[a], 4.03[b], 3.97[c]	6.698	6.772[a]	7.667	7.88[a]
b	4.066		3.955	4.06[a], 4.03[b], 3.97[c]	6.698	6.772[a]	7.667	7.88[a]
$-E_c$	3.566		3.687	3.19[a], 4.15[c]	3.793	3.30[a]	3.863	3.35[a]
d	2.349†		2.370	2.47[b], 2.38[c]	2.449		2.485	
h	0.0		0.636	0.69[a], 0.68[b], 0.64[c]	2.977	2.90[a]	2.875	2.96[a]
C_{11}	70.34		49.86		53.56		57.72	48.9[d]
C_{22}	70.34		49.86		53.56		57.72	48.9[d]
C_{12}	29.64		15.56		17.20		20.68	
C_{33}	20.35		17.15		18.18		18.52	
E	57.85		45.00	42.05[b]	48.04		50.31	
ν	0.42		0.31	0.33[b]	0.32		0.36	
K_I	99.98		65.42		70.76		78.40	
K_{II}	40.70		34.30		36.36		37.04	
K_{III}	40.70		34.30		36.36		37.04	

[a] Ref. [7], [b] Ref. [32], [c] Ref. [33], [d] Ref. [34].
† An average bond lengths calculated using radial pair distribution function with a *cut-off* radius = 3.5 Å and a number of histogram bins = 1000[20].

TABLE V. Structural and mechanical properties of large honeycomb dumbbell (LHD) germanene from molecular calculations: lattice parameters a, b (Å), average cohesive energy E_c (eV/atom), average bond length d (Å), average height h (Å), 2D elastic constants C_{ij} (N/m), 2D Kelvin moduli K_i (N/m), mean absolute percentage error (MAPE) (%), relative performance measured as normalized timesteps/second in molecular dynamics (MD) simulation.

Method	DFT	Tersoff 1989	Tersoff 2017	MEAM 2008	SW 1986	SW 2009	EDIP	ReaxFF	qSNAP	SNAP 2020	SNAP 2020	SNAP 2023	ACE	POD
a	7.667	7.763	7.527	7.517	7.791	7.243	9.018	7.660	8.702	11.746	8.319	7.879	8.184	
b	7.667	7.763	7.527	7.517	7.791	7.243	9.018	7.660	8.702	11.746	8.319	7.879	8.184	
$-E_c$	3.863	2.960	3.374	2.817	2.844	3.277	5.272	3.152	4.544	5.682	4.070	4.117	4.583	
d	2.485	2.560	2.617	2.700	2.642	2.617	2.838	2.664	2.682	2.220	2.615	2.746	2.600	
h	2.875	3.581	3.100	3.831	3.449	3.852	2.966	3.229	2.741	3.636	2.708	3.273	2.787	
C_{11}	57.72	55.39	54.55	78.50	76.29	46.06	24.56	56.43†	24.17	0.00*	19.34	53.99†	48.20	
C_{22}	57.72	55.39	54.55	78.50	76.29	46.06	24.56	51.68†	24.17	0.00*	19.34	22.30†	48.20	
C_{12}	20.68	18.62	25.44	11.54	35.40	32.05	17.71	14.90†	9.84	0.00*	10.85	33.11†	28.04	
C_{33}	18.52	18.39	14.56	33.48	20.44	7.01	3.44	16.29†	7.17	0.00*	4.24	13.59†	10.08	
K_I	78.40	74.01	79.99	90.04	111.69	78.10	42.27	68.77	34.01	0.00	30.19	67.22	76.25	
K_{II}	37.04	36.77	29.11	66.95	40.89	14.01	6.86	39.35	14.34	0.00	8.49	9.07	20.16	
K_{III}	37.04	36.77	29.11	66.95	40.89	14.01	6.86	32.58	14.34	0.00	8.49	27.17	20.16	
$\mathrm{MAPE}_{\mathrm{LHD}}$		26.98	10.80	27.02	22.09	28.98	42.40	10.11	38.87	74.22	42.23	25.62	20.66	
∑MAPE		139.88	138.42	139.14	84.13	140.02	122.76	75.22	154.41	171.68	182.63	154.73	113.89	
timesteps/s		120.05	120.07	194.94	167.50	150.19	524.37	6.75	2.00	1.00	3.35	6.94	1.79	

† Potential does not reproduce the isotropy of the elasticity tensor ($C_{11} \neq C_{22}$ and $2 \cdot C_{33} \neq C_{11} - C_{12}$),
* Nonsensical zero 2D elastic constants C_{ij}.

1. Tersoff1989: the Tersoff potential
2. Tersoff2017
3. MEAM2008: the modified embedded-atom method potential
4. SW1986: the Stillinger–Weber potential
5. SW2009
6. EDIP: the environment-dependent interatomic potential
7. ReaxFF: the reactive force-field
8. SNAP2020: the machine-learning-based spectral neighbour analysis potential
9. qSNAP2020
10. SNAP2023
11. ACE: the atomic cluster expansion potential
12. POD: the proper orthogonal descriptors based potential

[3] M. Maździarz - Transferability of interatomic potentials for germanene (2D germanium), J. Appl. Phys. 134, 184303 (2023)

Stanene - 2D Tin (Sn)

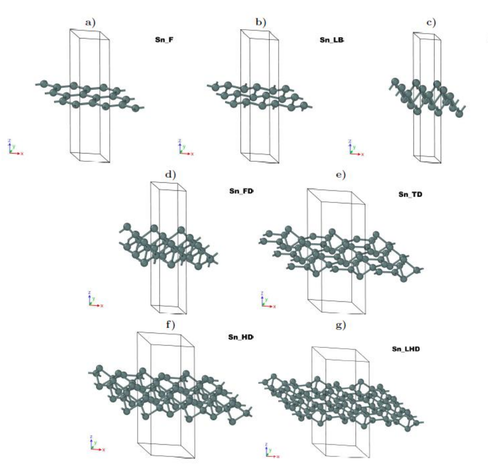

TABLE I. Structural and mechanical properties of flat (F), low-buckled (LB), high-buckled (HB), full dumbbell (FD), trigonal dumbbell (TD), honeycomb dumbbell (HD) and large honeycomb dumbbell (LHD) stanene phases from density functional theory (DFT) calculations: lattice parameters a,b (Å), average cohesive energy E_c (eV/atom), average bond length d (Å), average height h (Å), 2D elastic constants C_{ij} (N/m), 2D Young's modulus E (N/m), Poisson's ratio ν and 2D Kelvin moduli K_i (N/m).

Polymorph	F		LB		HB		FD		TD		HD		LHD	
Source	This work	Refs.	This work	Refs.	This work	Refs.	This work	Refs.	This work	Refs.	This work	Refs.	This work	Refs.
a	4.753		4.594	4.66[b], 4.67[c]	3.329	3.413[d]	4.309		7.821		7.646	7.74[c]	8.890	9.05[c]
b	4.753		4.594	4.66[b], 4.67[c]	3.329	3.413[d]	4.309		7.821		7.646	7.74[c]	8.890	9.05[c]
$-E_c$	2.719		2.844	2.68[c]	3.179		3.043		2.933		3.040	2.84[c]	3.062	2.86[c]
d	2.746†		2.791	2.83[a], 2.83[b]	3.266		3.012		2.830		2.945		2.897	
h	0.0		0.875	0.85[a], 0.9[b]	2.497		3.267		3.514		3.290	3.34[c]	3.404	3.42[c]
C_{11}	31.335		23.426	28.6[a]	44.046		15.251		27.650		27.923		33.028	
C_{22}	31.335		23.426	28.6[a]	44.046		15.251		27.650		27.923		33.028	
C_{12}	17.704		8.082	11.3[a]	28.644		8.710		11.031		13.078		15.753	
C_{33}	6.815		7.672	8.65[a]	7.701		3.270		8.310		7.422		8.637	
E	21.332		20.638	24.14[a], 24.46[b]	25.417		10.276		23.250		21.797		25.514	
ν	0.565		0.345	0.395[a], 0.390[b]	0.650		0.571		0.399		0.468		0.477	
K_I	49.039		31.508		72.690		23.961		38.681		41.001		48.782	
K_{II}	13.630		15.345		15.401		6.540		16.620		14.845		17.275	
K_{III}	13.630		15.345		15.401		6.540		16.620		14.845		17.275	

[a] Ref. [9], [b] Ref. [8], [c] Ref. [26] [d] Ref. [3].
† An average bond lengths calculated using radial pair distribution function with a *cut-off* radius = 3.5 Å and a number of histogram bins = 1000[38].

TABLE VIII. Structural and mechanical properties of large honeycomb dumbbell (LHD) stanene from molecular calculations: lattice parameters a, b (Å), average cohesive energy E_c (eV/atom), average bond length d (Å), average height h (Å), 2D elastic constants C_{ij} (N/m), 2D Kelvin moduli K_i (N/m), mean absolute percentage error (MAPE) (%), relative performance measured as normalized timesteps/second in molecular dynamics (MD) simulation.

Method	DFT	Tersoff 2016	MEAM 1997	MEAM 2017	MEAM 2018a	MEAM 2018b	RANN 2023	DP-PBE 2023	DP-SCAN 2023	POLY 2023	MTP 2024
a	8.890	9.175	9.090	8.788	8.977	8.907	9.467	9.103	8.220	9.017	8.724†
b	8.890	9.175	9.090	8.788	8.977	8.907	9.467	9.103	8.220	9.017	8.821†
$-E_c$	3.062	3.128	2.459	2.548	2.322	2.830	2.792	3.399	2.981	2.771	1762.220•
d	2.897	3.037	3.046	2.938	3.130	2.894	7.508	2.968	3.066	2.922	1.570
h	3.404	3.976	4.153	3.968	4.853	3.617	2.932	3.492	5.058	3.638	2.515
C_{11}	33.028	62.603	21.657	28.337	23.896	16.664	23.408‡	22.768	148.381‡	7.968‡	7739.930•
C_{22}	33.028	62.603	21.657	28.337	23.896	16.664	22.444‡	22.768	139.851‡	9.575‡	9143.533•
C_{12}	15.753	25.854	10.222	11.978	7.496	7.277	18.709‡	14.050	106.473‡	15.109‡	4278.919•
C_{33}	8.637	18.374	5.718	8.180	8.200	4.693	1.940‡	4.359	16.008‡	-1.213‡	2009.998•
K_I	48.782	88.457	31.878	40.315	31.393	23.941	41.630	36.818	250.503	23.859	12662.705
K_{II}	17.275	36.748	11.435	16.360	16.400	9.387	4.223	8.718	37.729	-6.315*	4220.758
K_{III}	17.275	36.748	11.435	16.360	16.400	9.387	3.879	8.718	32.016	-2.427*	4019.996
$\mathrm{MAPE}_{\mathrm{LHD}}$		57.744	24.280	10.221	19.605	29.589	43.341	22.240	168.606	48.901	19337.268
∑MAPE		209.382	130.073	63.928	149.014	124.659	223.611	204.086	710.469	175.759	19450.445
timesteps/s		4989.047	2159.590	4552.066	1980.937	5539.323	96.654	1.000	5.987	6.232	105.317

† Potential does not reproduce the correct symmetry of the structure (a≠b),
‡ Potential does not reproduce the isotropy of the elasticity tensor ($C_{11} \neq C_{22}$ and $2 \cdot C_{33} \neq C_{11} - C_{12}$),
* Negative Kelvin moduli K_i indicating a lack of mechanical stability,
• Highly overstated value.

1. Tersoff2016: the Tersoff potential
2. MEAM1997
3. MEAM2017: the modified embedded-atom method potential
4. MEAM2018a
5. MEAM2018b
6. RANN: combination of embedded atom method and rapid artificial neural network potential
7. DP-PBE: the machine-learning-based potential
8. DP-SCAN
9. POLY: the polynomial machine-learning-based potential
10. MTP: the moment tensor machine-learning interatomic potential

[4] M. Maździarz - Suitability of available interatomic potentials for Sn to model its 2D allotropes, arXiv (2024)

CONCLUSION

In general, classical, physics-based potentials show better transferability/universality than purely interpolative potentials based on machine learning. According to the methodology used here, it can be concluded that taking into account the performance and cost of computation, classical potentials such as Tersoff, SW and MEAM seem to be the best choice here. The cost of computing MLPs potentials is up to 3 orders of magnitude higher than classical ones, and they often exhibit improper behavior, such as violating the Neumann's principle, which states that the symmetry elements of any physical property of a crystal must include the symmetry elements of the crystal's point group.

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