Supplementary Information

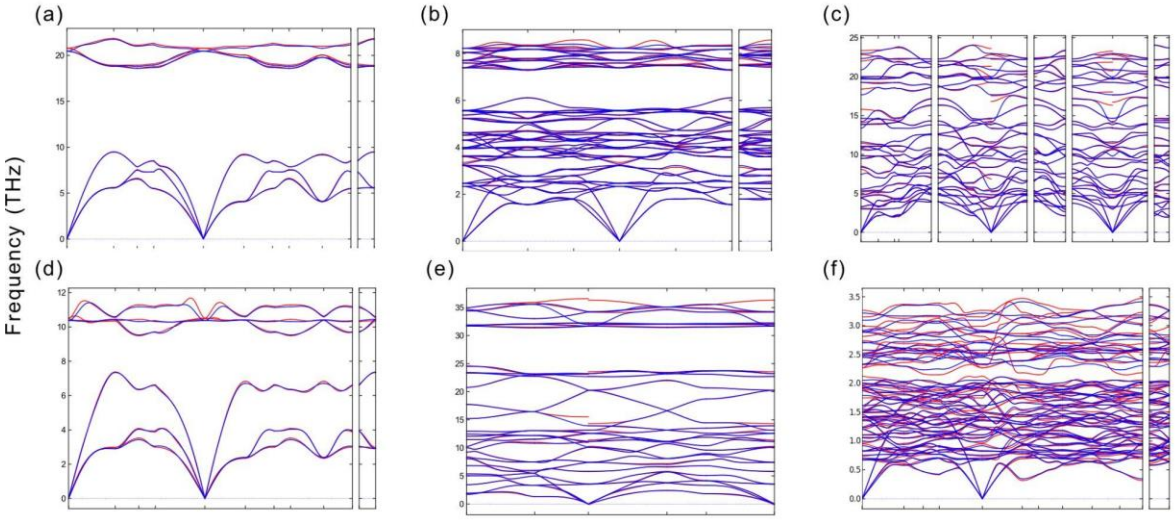

Fig. S1. Phonon dispersions of DFT (red) and NNP (blue) of the crystals considered in Fig. 1, (a) BAs, (b) $CoSb_3$, (c) β-Ga_2O_3, (d) GaP, (e) α-SiO_2, and (f) Tl_9BiTe_6. Non-analytic corrections are applied to (c) and (e). We recall that LO–TO splitting is not considered in the calculation of κ_l ; the computed phonon dispersions deviate from the experiment for polar materials.

Table S1. Detailed information of the computational settings and the κ_l values calculated by DFT, NNP-AIMD-400 structures, and NNP-AIMD-50 structures.

Compound	Space group number of unit cell	Number of atoms in supercell for calculating 2nd-order force constant	Number of atoms in supercell for calculating 3rd-order force constant	Plane wave energy cutoff (eV)	k-point grids for unit-cell optimization	Number of atoms in supercell for AIMD	Cutoff distance for 3rd-order force constant (n^{th} neighbor atom)	Number of structures with atomic displacements for calculating 3rd-order force constant	$\kappa_{l,DFT}$	$\kappa_{l,NNP-400}$	$\kappa_{l,NNP-50}$
SiO_2	154	1944	72	520	6×6×6	72	11th	624	3.93/7.96	4.17/7.94	4.01/8.14
$RbCaF_3$	221	135	135	650	3×3×3	135	4th	388	1.61	1.67	1.71
$TlSbTe_2$	166	108	108	250	7×7×1	108	4th	364	0.804/0.80	0.67/0.74	0.73/1.03
$PbSnS_3$	62	120	120	350	6×3×2	120	7th	824	0.73/3.09/0.77	0.74/2.87/0.52	0.95/3.01/0.77
Tl_9BiTe_6	87	576	128	230	6×6×4	128	4th	912	0.168/0.164	0.174/0.162	-
Ba_2BiAu	225	128	128	350	4×4×4	128	3rd	224	0.45	0.44	0.41
As_2Ga_2Sr	10	120	120	250	8×3×3	120	4th	1300	4.14/3.58/2.72	4.18/3.79.3.14	3.94/3.47/2.8
$K_2Bi_8Se_{13}$	2	460	69	250	8×3×2	69	8th	2916	1.29/0.63/1.02	0.98/0.48/0.57	0.8/0.39/0.47
CoGeTe	61	96	96	450	5×5×3	96	30th	1944	9.53/8.27/6.94	9.12/7.88/7.08	8.21/7.08/6.23
KCuS	33	384	96	400	4×3×2	96	8th	3024	1.7/1.31/1.56	1.59/1.27/1.51	1.37/1.07/1.29
$TlAgI_2$	140	432	128	350	3×3×3	128	6th	472	0.071/0.12	0.076/0.11	0.072/0.107
SnS	62	512	64	300	8×7×3	64	13th	596	2.80/0.975	2.38/0.983	2.43/1.32
GaP	216	576	192	300	16×16×16	64	5th	376	108.94	95	92.8

Ga_2O_3	12	720	120	600	5×5×3	120	19th	1936	23.63/22.9/18.26	20.08/21.95/16.56	-
GaN	186	192	72	500	6×6×4	72	4th	200	310.2/340	293.3/347.3	-
InAs	216	216	216	300	10×10×10	144	4th	172	21.8	22.3	-
Si	227	216	216	500	8×8×8	144	4th	92	134.4	125.3	-
$KZnF_3$	221	320	135	500	3×3×3	135	4th	384	2.7	2.75	-
$FeSi_2$	64	192	192	350	4×4×4	96	4th	492	25.5/26.4	27.4/28.2	-
$Ca_5Al_2Sb_6$	55	208	208	350	7×3×3	104	4th	856	1.68/1.46/1.47	2.01/1.33/1.39	-
$KAlSb_4$	62	192	192	300	8×3×2	120	4th	928	2.74/0.55/0.55	3.13/0.68/0.61	2.35/0.57/0.47
SnSe	62	400	72	300	8×8×4	72	14th	552	4.97/3.43/1.75	4.36/3.40/1.47	-
$CoSb_3$	204	128	128	350	4×4×4	128	4th	312	10.06	9.76	-
BAs	216	512	216	400	16×16×16	144	6th	320	1201	864	689
$CdGeAs_2$	122	512	64	300	8×8×5	64	7th	1200	5.72/3.18	5.16/6.13	5.48/7.15

Table S2. List of hyperparameters of ACSFs. Atomic type index of 3 is used only for ternary materials.

G^2			G^4														
Atomic type index of neighbor atoms	η(Å^{-2})	R_s	Atomic type index of neighbor atoms		η(Å^{-2})	ζ	λ	Atomic type index of neighbor atoms		η(Å^{-2})	ζ	λ	Atomic type index of neighbor atoms		η(Å^{-2})	ζ	λ
1	0.003214	0	1	1	0.000357	1	-1	1	2	0.000357	4	-1	1	3	0.000357	2	1
1	0.035711	0	1	1	0.028569	1	-1	1	2	0.028569	4	-1	1	3	0.028569	2	1
1	0.071421	0	1	1	0.089277	1	-1	1	2	0.089277	4	-1	1	3	0.089277	2	1
1	0.124987	0	1	1	0.000357	2	-1	1	2	0.000357	1	1	1	3	0.000357	4	1
1	0.214264	0	1	1	0.028569	2	-1	1	2	0.028569	1	1	1	3	0.028569	4	1
1	0.357106	0	1	1	0.089277	2	-1	1	2	0.089277	1	1	1	3	0.089277	4	1
1	0.714213	0	1	1	0.000357	4	-1	1	2	0.000357	2	1	2	2	0.000357	1	-1
1	1.428426	0	1	1	0.028569	4	-1	1	2	0.028569	2	1	2	2	0.028569	1	-1
2	0.003214	0	1	1	0.089277	4	-1	1	2	0.089277	2	1	2	2	0.089277	1	-1
2	0.035711	0	1	1	0.000357	1	1	1	2	0.000357	4	1	2	2	0.000357	2	-1
2	0.071421	0	1	1	0.028569	1	1	1	2	0.028569	4	1	2	2	0.028569	2	-1
2	0.124987	0	1	1	0.089277	1	1	1	2	0.089277	4	1	2	2	0.089277	2	-1
2	0.214264	0	1	1	0.000357	2	1	1	3	0.000357	1	-1	2	2	0.000357	4	-1
2	0.357106	0	1	1	0.028569	2	1	1	3	0.028569	1	-1	2	2	0.028569	4	-1
2	0.714213	0	1	1	0.089277	2	1	1	3	0.089277	1	-1	2	2	0.089277	4	-1
2	1.428426	0	1	1	0.000357	4	1	1	3	0.000357	2	-1	2	2	0.000357	1	1
3	0.003214	0	1	1	0.028569	4	1	1	3	0.028569	2	-1	2	2	0.028569	1	1
3	0.035711	0	1	1	0.089277	4	1	1	3	0.089277	2	-1	2	2	0.089277	1	1
3	0.071421	0	1	2	0.000357	1	-1	1	3	0.000357	4	-1	2	2	0.000357	2	1
3	0.124987	0	1	2	0.028569	1	-1	1	3	0.028569	4	-1	2	2	0.028569	2	1

3	0.214264	0	1	2	0.089277	1	-1	1	3	0.089277	4	-1	2	2	0.089277	2	1
3	0.357106	0	1	2	0.000357	2	-1	1	3	0.000357	1	1	2	2	0.000357	4	1
3	0.714213	0	1	2	0.028569	2	-1	1	3	0.028569	1	1	2	2	0.028569	4	1
3	1.428426	0	1	2	0.089277	2	-1	1	3	0.089277	1	1	2	2	0.089277	4	1

G^4														
Atomic type index of neighbor atoms		η(Å^{-2})	ζ	λ	Atomic type index of neighbor atoms		η(Å^{-2})	ζ	λ	Atomic type index of neighbor atoms		η(Å^{-2})	ζ	λ
2	3	0.000357	1	-1	2	3	0.000357	2	1	3	3	0.000357	4	-1
2	3	0.028569	1	-1	2	3	0.028569	2	1	3	3	0.028569	4	-1
2	3	0.089277	1	-1	2	3	0.089277	2	1	3	3	0.089277	4	-1
2	3	0.000357	2	-1	2	3	0.000357	4	1	3	3	0.000357	1	1
2	3	0.028569	2	-1	2	3	0.028569	4	1	3	3	0.028569	1	1
2	3	0.089277	2	-1	2	3	0.089277	4	1	3	3	0.089277	1	1
2	3	0.000357	4	-1	3	3	0.000357	1	-1	3	3	0.000357	2	1
2	3	0.028569	4	-1	3	3	0.028569	1	-1	3	3	0.028569	2	1
2	3	0.089277	4	-1	3	3	0.089277	1	-1	3	3	0.089277	2	1
2	3	0.000357	1	1	3	3	0.000357	2	-1	3	3	0.000357	4	1
2	3	0.028569	1	1	3	3	0.028569	2	-1	3	3	0.028569	4	1
2	3	0.089277	1	1	3	3	0.089277	2	-1	3	3	0.089277	4	1