Supplemental Information: ED Model for Density-Functional Prediction of Vibrational Free Energies of Molecular Crystals

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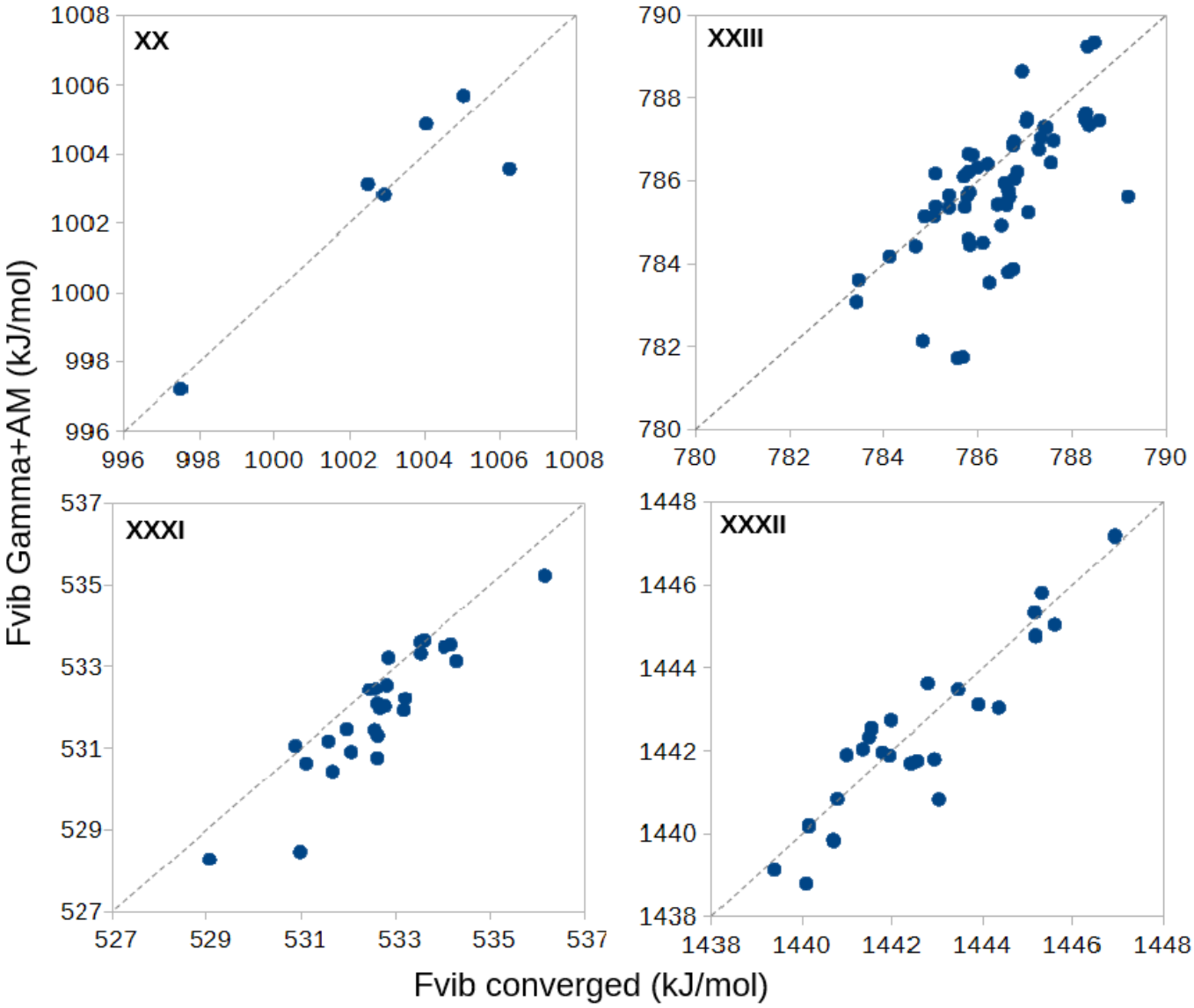

FIG. 1. Comparison of F_{vib} values between Einstein-Debye and converged phonons for each of the blind test molecules.

TABLE I. Vibrational free energies and DFT lattice energies, in kJ/mol per molecule, for the PV17 set of polymorph pairs. Supercell dimensions represent the number of times the unit cell is replicated along the a, b, and c lattice vectors.

Refcode	Γ	ED	Converged	E_{DFT}	Supercell dimensions	# of atoms in supercell	# of displacements
CUMMIG01	288.0	282.4	284.1	-613114.4	3 × 1 × 3	252	42
CUMMIG02	286.9	280.9	282.6	-613109.5	2 × 4 × 2	448	42
DAVVUR	1173.4	1160.4	1160.2	-3438863.1	1 × 2 × 3	348	174
DAVVUR01	1177.6	1164.5	1163.4	-3438866.7	2 × 2 × 2	464	174
EFUMAU	316.4	313.2	314.5	-558625.7	2 × 3 × 1	336	84
EFUMAU03	316.0	312.8	314.1	-558624.7	2 × 3 × 2	672	168
EFURIH	494.8	490.1	491.1	-1806486.8	3 × 2 × 1	288	72
EFURIH04	496.0	490.7	491.4	-1806493.6	3 × 2 × 2	576	144
ETDIAM16	279.7	274.9	275.0	-500732.6	2 × 2 × 2	192	36
ETDIAM18	281.0	276.6	276.4	-500733.0	2 × 2 × 2	192	36
FUMAAC	191.4	188.4	190.5	-1198070.3	3 × 2 × 2	576	144
FUMAAC01	200.8	192.9	192.7	-1198077.7	3 × 2 × 3	216	36
GICTIV	240.6	234.0	237.3	-2633078.5	4 × 1 × 4	640	120
GICTIV01	245.1	237.6	238.8	-2633086.1	3 × 3 × 1	360	60
MALEHY10	209.1	204.3	204.4	-1090256.6	3 × 2 × 2	288	72
MALEHY12	205.7	202.8	204.6	-1090256.8	3 × 3 × 2	864	72
MALIAC12	192.5	189.6	191.0	-1198058.0	3 × 1 × 2	288	72
MALIAC13	195.0	190.0	190.6	-1198057.7	4 × 2 × 2	384	72
OXALAC03	114.8	112.1	112.7	-994659.9	2 × 2 × 3	384	24
OXALAC04	114.5	110.2	112.1	-994662.5	3 × 2 × 2	192	24
QQQCIV01	107.3	104.6	104.9	-348883.8	3 × 2 × 2	288	36
QQQCIV08	109.7	104.8	105.7	-348884.1	3 × 3 × 2	216	36
REKBUE	384.9	379.1	379.3	-2088217.1	3 × 2 × 2	528	66
REKBUE01	387.9	378.5	378.6	-2088220.9	4 × 3 × 1	264	66
SUCACB02	255.1	250.4	251.7	-1201305.4	2 × 1 × 3	168	42
SUCACB07	253.6	248.8	250.4	-1201305.9	3 × 2 × 3	504	84
THHYDT	186.8	183.8	183.5	-1842418.6	3 × 2 × 3	792	66
THHYDT02	187.7	182.5	182.4	-1842421.0	3 × 2 × 2	264	66
TRDMPP01	406.6	401.1	401.2	-1300118.3	2 × 3 × 2	480	60
TRDMPP02	406.4	400.8	401.6	-1300118.8	2 × 2 × 2	320	60
TRITAN03	204.5	201.3	202.0	-3459800.9	3 × 2 × 1	288	40
TRITAN10	207.6	201.8	202.2	-3459801.3	2 × 2 × 3	288	40
XOCJEE	211.8	206.4	207.2	-2085230.8	3 × 2 × 4	624	78
XOCJEE01	212.1	206.8	206.7	-2085230.2	2 × 2 × 3	312	78

TABLE II. Vibrational free energies and DFT lattice energies, in kJ/mol per molecule, for the CSP blind-test compound XX. Supercell dimensions represent the number of times the unit cell is replicated along the a, b, and c lattice vectors.

Structure	Γ	ED	Converged	E_{DFT}	Supercell dimensions	# of atoms in supercell	# of displacements
xx-day-2	1013.8	1005.7	1005.0	-5716928.9	2 × 1 × 2	440	330
xx-expt	1001.7	997.2	997.5	-5716931.1	2 × 4 × 1	1760	330
xx-kendrick-1	1007.2	1003.6	1006.2	-5716929.3	3 × 1 × 1	660	660
xx-kendrick-2	1009.2	1004.9	1004.0	-5716935.4	4 × 1 × 1	880	330
xx-kendrick-3	1011.4	1002.8	1002.9	-5716935.5	2 × 3 × 2	1320	330
xx-price-2	1007.7	1003.1	1002.5	-5716937.7	2 × 2 × 2	1760	330

TABLE III. Vibrational free energies and DFT lattice energies, in kJ/mol per molecule, for the CSP blind-test compound XXIII. Supercell dimensions represent the number of times the unit cell is replicated along the a, b, and c lattice vectors.

Structure	Γ	ED	Converged	E_{DFT}	Supercell dimensions	# of atoms in supercell	# of displacements
xxiii-003	787.5	783.9	786.8	-5097443.1	3 × 1 × 1	516	516
xxiii-005	791.7	787.6	788.3	-5097443.6	3 × 2 × 1	1032	516
xxiii-007	790.3	786.2	785.8	-5097442.4	3 × 2 × 1	1032	516
xxiii-008	788.1	785.6	786.7	-5097442.5	3 × 2 × 1	2064	516
xxiii-010	787.9	785.4	786.4	-5097442.2	3 × 2 × 1	2064	516
xxiii-011	791.8	787.5	788.6	-5097443.1	2 × 2 × 1	688	516
xxiii-013	787.0	784.6	785.8	-5097442.0	3 × 2 × 1	2064	516
xxiii-018	794.7	786.8	787.3	-5097442.9	2 × 2 × 2	688	258
xxiii-020	783.6	781.7	785.6	-5097442.1	4 × 1 × 1	1376	516
xxiii-021	788.1	785.8	786.7	-5097441.7	3 × 2 × 1	2064	516
xxiii-022	791.2	787.0	786.8	-5097442.2	3 × 2 × 1	1032	516
xxiii-023	783.7	781.8	785.7	-5097442.0	4 × 1 × 1	1376	516
xxiii-024	788.5	786.2	786.8	-5097442.6	3 × 2 × 1	2064	516
xxiii-025	791.4	787.4	788.4	-5097441.3	2 × 2 × 1	688	516
xxiii-026	785.8	783.8	786.6	-5097442.4	3 × 2 × 1	2064	516
xxiii-029	788.4	786.1	786.8	-5097440.8	4 × 1 × 1	1376	516
xxiii-030	791.6	787.5	788.3	-5097441.0	2 × 2 × 1	688	516
xxiii-033	789.9	785.7	785.8	-5097441.0	3 × 2 × 1	1032	516
xxiii-036	789.8	785.7	785.4	-5097441.0	3 × 2 × 1	1032	516
xxiii-038	790.1	785.6	785.8	-5097441.8	3 × 2 × 1	1032	258
xxiii-040	789.7	785.4	785.1	-5097440.1	3 × 2 × 1	1032	516
xxiii-042	794.9	787.0	787.3	-5097441.0	3 × 2 × 1	516	258
xxiii-043	791.5	787.3	787.5	-5097440.5	2 × 2 × 2	1376	258
xxiii-044	793.2	788.7	786.9	-5097440.8	3 × 2 × 1	1032	516
xxiii-046	789.8	785.4	785.4	-5097440.6	2 × 2 × 2	1376	516
xxiii-047	790.8	786.6	785.9	-5097440.6	3 × 2 × 1	1032	516
xxiii-048	789.5	785.6	789.2	-5097441.0	4 × 1 × 1	688	258
xxiii-049	795.8	787.5	787.0	-5097440.2	2 × 2 × 2	688	258
xxiii-052	790.7	786.7	785.8	-5097440.6	3 × 2 × 1	1032	516
xxiii-053	793.7	786.2	785.1	-5097441.5	3 × 2 × 1	516	258
xxiii-057	789.6	785.4	785.7	-5097440.5	3 × 2 × 1	1032	516
xxiii-059	790.2	786.1	785.7	-5097440.6	3 × 2 × 1	1032	516
xxiii-060	790.5	786.3	786.0	-5097440.6	3 × 2 × 1	1032	516
xxiii-063	788.9	785.3	787.1	-5097440.6	3 × 2 × 1	1032	516
xxiii-065	789.4	785.4	786.6	-5097440.5	3 × 2 × 1	1032	516
xxiii-067	788.1	784.5	785.8	-5097440.8	3 × 2 × 1	1032	516
xxiii-068	790.6	786.4	787.6	-5097440.4	3 × 2 × 1	1032	258
xxiii-070	788.5	784.2	784.1	-5097443.4	2 × 2 × 1	688	258
xxiii-080	788.5	784.5	786.1	-5097440.8	3 × 2 × 1	1032	258
xxiii-083	787.2	783.6	786.3	-5097439.6	4 × 1 × 1	688	516
xxiii-091	793.8	786.0	786.6	-5097440.2	3 × 2 × 1	516	258
xxiii-095	793.7	786.4	786.2	-5097440.4	3 × 2 × 1	516	258
xxiii-097	788.6	784.4	784.7	-5097440.5	3 × 2 × 1	1032	516
xxiii-100	789.1	784.9	786.5	-5097440.4	3 × 2 × 1	1032	258
xxiii-boese-hofmann1-1	797.0	789.4	788.5	-5097441.1	2 × 2 × 2	688	258
xxiii-expt-a	787.7	783.1	783.4	-5097443.1	2 × 2 × 1	688	258
xxiii-expt-b	794.6	787.4	787.0	-5097444.0	3 × 2 × 1	516	258
xxiii-expt-c	791.7	787.6	788.3	-5097443.6	2 × 2 × 1	688	516
xxiii-expt-d	788.1	783.6	783.5	-5097443.0	1 × 2 × 2	688	516
xxiii-expt-e	789.3	785.2	785.1	-5097442.9	3 × 1 × 1	516	516
xxiii-neumann1-3	795.3	787.3	787.4	-5097443.2	2 × 2 × 2	688	258
xxiii-neumann2-1	787.8	785.5	786.6	-5097443.5	1 × 3 × 1	1032	516
xxiii-neumann2-2	791.0	786.9	786.8	-5097444.9	3 × 2 × 1	1032	516
xxiii-poly59-23-1	794.5	787.0	787.6	-5097441.1	1 × 2 × 3	516	258
xxiii-poly59-23-2	795.4	789.3	788.3	-5097440.7	3 × 1 × 2	516	258
xxiii-tkatchenko2-1	792.5	785.2	784.9	-5097441.8	3 × 2 × 1	516	258
xxiii-vaneijck1-1	789.1	782.1	784.8	-5097441.8	4 × 1 × 1	344	258

TABLE IV. Vibrational free energies and DFT lattice energies, in kJ/mol per molecule, for the CSP blind-test compound XXXI. Supercell dimensions represent the number of times the unit cell is replicated along the a, b, and c lattice vectors.

Structure	Γ	ED	Converged	E_{DFT}	Supercell dimensions	# of atoms in supercell	# of displacements
XXXI-1	535.6	531.5	532.0	-3795199.5	3 × 2 × 1	768	192
XXXI-10	533.5	531.2	531.6	-3795196.6	3 × 2 × 1	1536	192
XXXI-11	536.0	531.9	533.2	-3795199.8	3 × 3 × 1	1152	192
XXXI-17	537.8	533.5	534.0	-3795202.0	4 × 1 × 1	512	192
XXXI-18	534.9	530.6	531.1	-3795197.4	3 × 2 × 1	768	192
XXXI-20	535.4	531.0	530.9	-3795198.3	2 × 2 × 2	1024	192
XXXI-21	535.0	530.9	532.1	-3795196.3	3 × 2 × 1	768	192
XXXI-22	536.6	532.4	532.6	-3795197.2	3 × 2 × 1	768	192
XXXI-23	541.2	533.6	533.6	-3795198.0	3 × 2 × 1	384	192
XXXI-24	532.4	528.5	531.0	-3795197.4	2 × 3 × 1	768	192
XXXI-25	532.7	528.3	529.1	-3795197.9	3 × 2 × 1	768	192
XXXI-28	538.5	530.8	532.6	-3795199.5	3 × 2 × 1	384	192
XXXI-30	541.3	533.5	534.2	-3795198.0	3 × 2 × 1	384	192
XXXI-32	536.0	532.0	532.8	-3795199.0	4 × 2 × 1	1024	192
XXXI-34	540.3	532.4	532.4	-3795199.4	3 × 3 × 1	576	192
XXXI-39	538.1	533.6	533.5	-3795196.8	2 × 2 × 2	1024	192
XXXI-41	543.0	535.2	536.1	-3795197.0	3 × 2 × 1	384	192
XXXI-53	539.7	532.1	532.6	-3795196.4	3 × 2 × 1	384	192
XXXI-57	536.5	532.2	533.2	-3795198.7	3 × 2 × 1	768	192
XXXI-63	537.7	533.2	532.8	-3795197.6	3 × 3 × 2	2304	192
XXXI-70	537.8	533.3	533.5	-3795197.6	3 × 2 × 1	768	192
XXXI-71	537.4	533.1	534.3	-3795201.9	4 × 1 × 1	512	192
XXXI-73	536.0	532.0	532.7	-3795199.2	4 × 2 × 1	1024	192
XXXI-75	538.1	530.4	531.7	-3795197.7	3 × 2 × 1	384	192
XXXI-77	537.0	532.5	532.8	-3795197.4	2 × 2 × 2	1024	192
XXXI-98	535.6	531.4	532.6	-3795200.9	3 × 2 × 1	768	192
XXXI-99	535.3	531.3	532.6	-3795196.9	3 × 2 × 1	768	192

TABLE V. Vibrational free energies and DFT lattice energies, in kJ/mol per molecule, for the CSP blind-test compound XXXII. Supercell dimensions represent the number of times the unit cell is replicated along the a, b, and c lattice vectors.

Structure	Γ	ED	Converged	E_{DFT}	Supercell dimensions	# of atoms in supercell	# of displacements
XXXII-7	1444.2	1439.8	1440.7	-6471784.6	3 × 1 × 1	912	456
XXXII-9	1450.2	1441.9	1441.0	-6471788.6	2 × 2 × 2	1216	456
XXXII-57	1451.0	1442.3	1441.5	-6471783.0	2 × 2 × 1	608	456
XXXII-82	1450.5	1442.0	1441.4	-6471784.5	2 × 2 × 2	1216	456
XXXII-114	1446.4	1441.9	1441.9	-6471782.4	3 × 1 × 1	912	456
XXXII-117	1451.0	1443.0	1444.4	-6471784.1	2 × 2 × 1	608	456
XXXII-130	1444.9	1439.9	1440.7	-6471782.4	2 × 2 × 1	1216	912
XXXII-140	1453.6	1445.8	1445.3	-6471782.2	2 × 2 × 1	608	456
XXXII-165	1447.7	1443.5	1443.5	-6471786.1	2 × 2 × 1	1216	456
XXXII-193	1445.1	1440.8	1443.0	-6471784.2	3 × 2 × 1	1824	456
XXXII-199	1443.2	1438.8	1440.1	-6471782.9	3 × 1 × 1	912	456
XXXII-232	1443.5	1439.1	1439.4	-6471783.0	2 × 2 × 1	1216	912
XXXII-236	1451.7	1443.6	1442.8	-6471783.0	2 × 2 × 1	608	456
XXXII-250	1449.8	1441.7	1442.4	-6471784.0	3 × 2 × 1	912	456
XXXII-270	1445.5	1440.8	1440.8	-6471787.5	2 × 2 × 1	1216	456
XXXII-299	1449.8	1441.8	1442.5	-6471784.8	3 × 2 × 1	912	456
XXXII-317	1449.0	1440.2	1440.1	-6471782.2	2 × 2 × 1	608	456
XXXII-320	1450.0	1445.3	1445.2	-6471786.9	2 × 2 × 1	1216	456
XXXII-331	1450.4	1442.0	1441.8	-6471782.7	2 × 2 × 1	608	456
XXXII-334	1447.7	1443.1	1443.9	-6471783.3	2 × 2 × 1	1216	456
XXXII-342	1451.1	1442.7	1442.0	-6471785.7	2 × 2 × 2	1216	456
XXXII-388	1451.5	1447.2	1446.9	-6471786.1	2 × 2 × 1	1216	456
XXXII-423	1449.5	1445.0	1445.6	-6471788.1	2 × 2 × 1	1216	456
XXXII-448	1449.2	1444.8	1445.2	-6471782.0	2 × 2 × 1	1216	456
XXXII-478	1451.2	1442.5	1441.6	-6471783.0	2 × 1 × 2	608	456
XXXII-500	1449.8	1441.8	1442.9	-6471787.9	3 × 2 × 1	912	456

TABLE VI. Vibrational free energies and DFT lattice energies, in kJ/mol per molecule, for the FP10 set of polymorph pairs.

Refcode	Γ	ED	E_{DFT}
BEDMIG	543.7	539.9	-4167127.0
BEDMIG01	539.0	536.6	-4167124.3
BEDMIG02	547.3	539.4	-4167125.2
BEDMIG03	541.4	539.2	-4167125.1
BEDMIG04	540.8	536.8	-4167124.9
BEDMIG20	542.3	537.9	-4167122.7
CIMETD	646.7	643.0	-2940107.3
CIMETD01	645.8	642.2	-2940107.6
FOGVIG01	627.3	623.5	-5377747.5
FOGVIG02	631.2	627.7	-5377744.1
GICKEM	962.0	953.3	-6337341.8
GICKEM01	962.4	954.9	-6337345.1
GICKEM02	961.6	957.1	-6337347.8
JAKGEH	749.7	743.1	-2499275.0
JAKGEH01	749.0	745.0	-2499274.8
MELFIT01	1135.2	1127.4	-5601204.4
MELFIT02	1131.4	1127.1	-5601207.3
MELFIT05	1139.6	1131.9	-5601209.9
MELFIT07	1140.5	1132.3	-5601210.3
MELFIT09	1132.6	1128.2	-5601207.9
POPFAC	852.8	849.1	-3555132.8
POPFAC01	849.8	847.6	-3555131.8
UGIVAI	596.7	591.0	-2432541.6
UGIVAI01	599.6	596.6	-2432546.5
ZZZPUS01	709.1	705.3	-3161179.8
ZZZPUS06	708.0	703.7	-3161176.5
ZZZPUS07	709.4	705.3	-3161178.6
ZZZPUS10	705.9	703.5	-3161175.1
INDMET02	745.9	743.1	-4077003.3
INDMET03	750.8	743.3	-4077001.0
INDMET07	747.4	743.2	-4077002.6
INDMET09	740.2	738.2	-4076998.0

TABLE VII. Mean absolute errors in absolute and relative free-energy corrections at 0 K, in kJ/mol per molecule, relative to converged supercell results for the PV10 set and blind-test compounds.

	Gamma		Einstein-Debye	
System	F_{vib}	ΔF_{vib}	F_{vib}	ΔF_{vib}
PV17	0.74	0.32	0.28	0.15
XXXI	0.21	0.10	0.07	0.06
XXIII	0.19	0.11	0.07	0.07
XXXII	0.33	0.11	0.18	0.07
XX	0.12	0.16	0.07	0.09