Kylin-V: An Open-source Package Calculating the Dynamic and Spectroscopic Properties of Large Systems - Supplementary Material

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S1. ETHYLENE

TABLE S1: 100 lowest-energy states of ethylene computed with excitation energies given relative to the ZPE. The number of variantional configurations of the last VHCI iteration $N_{\mathscr{V}}$, threshold for both VHCI and ENPT2 $\varepsilon_{\mathrm{VHCI}}, \varepsilon_{\mathrm{ENPT2}}$ and the time cost are shown below. The time of ENPT2 includes the necessary variational step. For simulation with Coriolis term (labelled with "+Coriolis"), Only those states observed by experiments[1] are listed. All energies are in cm^{-1}.

Method	VHCI[Kylin-V]		VHCI[Berkelbach]	VHCI[Kylin-V]+Coriolis	Exp.
$N_{\mathscr{V}}$	275563		161338	277342	
$\varepsilon_{\mathrm{VHCI}}$	0.5		0.5	0.5	-
	Var.	$\varepsilon_{\mathrm{ENPT2}}=0.01$	$\varepsilon_{\mathrm{ENPT2}}=0.01$	$\varepsilon_{\mathrm{ENPT2}}=0.01$	-
ZPE	11011.61	11011.61	11011.61	11016.94	-
ω_{10}	820.00	819.98	819.99	823.53	825.93
ω_8	926.34	926.32	926.33	935.18	939.86
ω_7	941.66	941.64	941.65	950.60	948.77
ω_4	1017.47	1017.45	1017.45	1025.80	1025.59
ω_6	1222.18	1222.15	1222.16	1224.79	1222.0
ω_3	1341.96	1341.94	1341.95	1342.78	1343.54
ω_{12}	1438.33	1438.31	1438.31	1441.75	1442.48
ω_2	1622.82	1622.78	1622.78	1625.25	1625.4
$2\omega_{10}$	1655.28	1655.21	1655.21	1661.16	1662.2
$\omega_{10}+\omega_8$	1748.16	1748.03	1748.05	1760.28	1766.71
$\omega_{10}+\omega_7$	1766.33	1766.17	1766.19	1781.00	1781.01
$\omega_{10}+\omega_4$	1837.81	1837.65	1837.68	1851.02	1853.96
$2\omega_8$	1858.45	1858.34	1858.36	1875.59	1880.9
$\omega_8+\omega_7$	1871.54	1871.40	1871.42	1888.41	1888.97
$2\omega_7$	1887.13	1887.01	1887.03	1904.53	1899.75
$\omega_8+\omega_4$	1939.61	1939.45	1939.47	1956.35	1958.27
$\omega_7+\omega_4$	1953.19	1953.03	1953.05	1969.92	1965.44
$2\omega_4$	2032.18	2032.07	2032.09	2048.59	2046.4
$\omega_{10}+\omega_6$	2037.59	2037.40	2037.42	2043.93	2047.76
$\omega_8+\omega_6$	2150.36	2150.20	2150.22		-
$\omega_{10}+\omega_3$	2164.65	2164.50	2164.52	2169.00	2172.5
$\omega_7+\omega_6$	2165.84	2165.68	2165.70		-
$\omega_4+\omega_6$	2239.92	2239.76	2239.78	2250.65	2251.5
$\omega_{10}+\omega_{12}$	2256.31	2256.12	2256.14		-
$\omega_8+\omega_3$	2265.41	2265.27	2265.29		-
$\omega_7+\omega_3$	2281.74	2281.59	2281.61	2291.5	2291.5
$\omega_4+\omega_3$	2355.72	2355.58	2355.60		-
$\omega_8+\omega_{12}$	2362.19	2362.03	2362.05		-
$\omega_7+\omega_{12}$	2375.52	2375.35	2375.37		-
$\omega_{10}+\omega_2$	2433.56	2433.37	2433.40	2439.26	2439.0
$2\omega_6$	2443.36	2443.23	2443.25		-
$\omega_4+\omega_{12}$	2454.69	2454.53	2454.55		-
$3\omega_{10}$	2497.06	2496.81	2496.84		-
$\omega_8+\omega_2$	2543.36	2543.18	2543.21		-
$\omega_7+\omega_2$	2560.21	2560.03	2560.06	2571.61	2571.0
$\omega_6+\omega_3$	2561.88	2561.75	2561.77		-
$2\omega_{10}+\omega_8$	2583.87	2583.57	2583.60		-
$2\omega_{10}+\omega_7$	2604.43	2604.12	2604.16		-
$\omega_4+\omega_2$	2637.19	2637.01	2637.04		-
$\omega_6+\omega_{12}$	2654.83	2654.67	2654.69		-
$2\omega_{10}+\omega_4$	2672.84	2672.55	2672.59		-
$2\omega_3$	2682.17	2682.08	2682.09	2683.73	2685.3
$\omega_{10}+2\omega_8$	2682.49	2682.22	2682.25		-
$\omega_{10}+\omega_8+\omega_7$	2698.50	2698.13	2698.18		-
$\omega_{10}+2\omega_7$	2716.79	2716.48	2716.52		-
$\omega_{10}+\omega_8+\omega_4$	2762.35	2762.03	2762.07		-
$\omega_3+\omega_{12}$	2774.94	2774.79	2774.81		-
$\omega_{10}+\omega_7+\omega_4$	2778.78	2778.42	2778.47		-
$3\omega_8$	2795.90	2795.71	2795.74		-

$2\omega_8+\omega_7$	2807.66	2807.38	2807.43	2832.20	2829.8
$\omega_8+2\omega_7$	2823.07	2822.77	2822.82		-
$\omega_6+\omega_2$	2830.77	2830.59	2830.62		-
$3\omega_7$	2837.27	2837.03	2837.07	2862.66	2854.0
$\omega_{10}+2\omega_4$	2853.14	2852.85	2852.89		-
$2\omega_{10}+\omega_6$	2867.29	2866.96	2867.00		-
$2\omega_8+\omega_4$	2868.55	2868.29	2868.34		-
$2\omega_{12}$	2869.61	2869.47	2869.49		-
$\omega_8+\omega_7+\omega_4$	2882.68	2882.28	2882.36		-
$2\omega_7+\omega_4$	2894.99	2894.69	2894.74		-
$\omega_8+2\omega_4$	2951.44	2951.15	2951.20		-
$\omega_3+\omega_2$	2957.48	2957.34	2957.37	2960.26	2961.6
$\omega_7+2\omega_4$	2963.47	2963.18	2963.24		-
$\omega_{10}+\omega_8+\omega_6$	2968.22	2967.88	2967.92	2986.09	2993.29
ω_{11}	2978.08	2977.99	2978.02	2985.26	2988.63
$\omega_{10}+\omega_7+\omega_6$	2986.42	2986.07	2986.11		-
$2\omega_{10}+\omega_3$	2999.88	2999.62	2999.66		-
ω_1	3016.77	3016.71	3016.73	3018.64	3021.85
$3\omega_4$	3044.29	3044.08	3044.12		-
$\omega_{10}+\omega_4+\omega_6$	3055.85	3055.49	3055.54		-
$\omega_{12}+\omega_2$	3070.39	3070.11	3070.16	3077.71	3078.46
ω_5	3071.24	3071.15	3071.18		3082.36
$2\omega_8+\omega_6$	3085.21	3084.96	3084.99		-
$\omega_{10}+\omega_8+\omega_3$	3090.23	3089.91	3089.95	3103.35	3109.32
ω_9	3091.51	3091.37	3091.40	3101.05	3104.87
$2\omega_{10}+\omega_{12}$	3096.00	3095.68	3095.72	3104.28	3104.33
$\omega_8+\omega_7+\omega_6$	3099.73	3099.45	3099.48		-
$\omega_{10}+\omega_7+\omega_3$	3109.37	3109.00	3109.05		-
$2\omega_7+\omega_6$	3113.42	3113.15	3113.19		-
$\omega_8+\omega_4+\omega_6$	3163.96	3163.63	3163.68		-
$\omega_7+\omega_4+\omega_6$	3177.82	3177.51	3177.55		-
$\omega_{10}+\omega_4+\omega_3$	3179.00	3178.65	3178.70		-
$\omega_{10}+\omega_8+\omega_{12}$	3182.49	3182.10	3182.15		-
$2\omega_8+\omega_3$	3194.64	3194.40	3194.44		-
$\omega_{10}+\omega_7+\omega_{12}$	3198.87	3198.49	3198.54		-
$\omega_8+\omega_7+\omega_3$	3209.16	3208.82	3208.87		-
$2\omega_7+\omega_3$	3225.55	3225.29	3225.33		-
$2\omega_2$	3236.04	3235.77	3235.82	3241.51	3239.0
$\omega_{10}+2\omega_6$	3254.81	3254.49	3254.54		-
$2\omega_4+\omega_6$	3254.86	3254.60	3254.64		-
$2\omega_{10}+\omega_2$	3269.90	3269.58	3269.63	3276.28	3276.2
$\omega_{10}+\omega_4+\omega_{12}$	3273.43	3273.07	3273.12		-
$\omega_8+\omega_4+\omega_3$	3274.99	3274.65	3274.71		-
$\omega_7+\omega_4+\omega_3$	3289.71	3289.36	3289.42		-
$2\omega_8+\omega_{12}$	3292.13	3291.84	3291.89		-
$\omega_8+\omega_7+\omega_{12}$	3304.39	3304.02	3304.09	3326.12	3327.3
$2\omega_7+\omega_{12}$	3317.45	3317.12	3317.19		-
$4\omega_{10}$	3349.38	3348.92	3348.99		-
$\omega_{10}+\omega_8+\omega_2$	3356.18	3355.75	3355.81		-
$2\omega_4+\omega_3$	3366.59	3366.35	3366.39		-

S2. NAPHTHALENE

TABLE S2: 128 lowest-energy states of naphthalene computed with excitation energies given relative to the ZPE, computed with VHCI and VHCI+ENPT2. The reference data from VHCI[Brorsen][2] are listed for comparison. All energies are in cm^{-1}.

Method	VHCI[Kylin-V]	VHCI[Brorsen]	VHCI[Kylin-V]	VHCI[Brorsen]
$N_{\mathcal{V}}$	2787046	2447654	7496009	6791503
$\varepsilon_{\mathrm{VHCI}}$	1.0	1.0	0.5	0.5
Cores	64	40	64	40

ZPE	31769.18	31763.78	31772.27	31769.41	31765.81	31761.15	31766.97	31763.17
ω_{48}	166.07	164.14	172.28	170.99	164.93	163.21	166.36	164.97
ω_{13}	179.82	177.98	185.99	184.36	178.73	177.02	180.34	178.82
$2\omega_{48}$	332.51	328.32	335.52	332.96	329.61	326.45	331.11	328.68
$\omega_{13}+\omega_{48}$	345.70	341.29	351.42	348.53	342.63	339.11	344.86	342.03
ω_{24}	357.37	355.55	366.35	364.43	356.49	354.60	360.55	358.69
$2\omega_{13}$	359.82	356.02	364.89	362.15	357.23	354.37	359.75	357.28
ω_{16}	390.32	388.15	397.04	395.10	389.23	386.99	392.48	390.51
ω_{28}	465.89	463.33	473.47	471.50	464.46	461.86	466.29	464.21
ω_{47}	474.50	471.76	483.15	480.96	472.97	470.26	475.48	473.19
$3\omega_{48}$	497.41	491.39	503.76	499.66	493.33	488.76	495.88	492.03
ω_{44}	508.31	506.68	519.44	517.87	507.48	505.60	511.21	509.43
$\omega_{13}+2\omega_{48}$	510.45	503.71	522.01	516.77	505.81	500.34	509.84	505.19
ω_{9}	514.97	513.59	519.44	517.87	514.20	512.51	516.06	514.50
$\omega_{24}+\omega_{48}$	523.20	519.72	531.01	528.20	521.09	518.08	524.89	522.20
$2\omega_{13}+\omega_{48}$	524.42	517.86	535.22	530.00	519.99	514.76	524.19	519.66
$\omega_{13}+\omega_{24}$	537.71	533.68	543.84	540.94	534.83	531.62	539.05	536.32
$3\omega_{13}$	540.01	534.63	549.92	545.22	535.35	531.21	539.31	535.52
$\omega_{16}+\omega_{48}$	556.63	552.49	563.53	559.95	554.23	550.65	558.81	555.43
$\omega_{13}+\omega_{16}$	570.97	566.46	574.63	571.52	568.12	564.14	571.17	568.08
ω_{12}	624.40	622.11	634.55	631.74	623.06	620.69	628.55	626.03
ω_{36}	624.87	622.97	637.36	634.71	623.95	621.85	630.50	628.34
$\omega_{28}+\omega_{48}$	632.55	626.91	641.92	637.91	628.71	623.54	632.21	628.46
$\omega_{47}+\omega_{48}$	642.05	636.72	648.58	644.93	638.71	634.06	643.65	640.08
$\omega_{13}+\omega_{28}$	646.82	641.34	659.20	654.75	643.15	638.12	648.00	643.58
$\omega_{13}+\omega_{47}$	655.56	649.77	663.29	658.75	651.88	646.70	656.94	652.51
$4\omega_{48}$	664.60	657.23	667.74	663.57	659.17	654.86	659.51	656.58
$\omega_{44}+\omega_{48}$	676.14	671.95	685.53	681.79	673.66	669.89	679.00	675.59
$\omega_{13}+3\omega_{48}$	677.62	668.72	684.70	678.94	670.91	665.20	673.09	668.71
$\omega_{9}+\omega_{48}$	684.01	679.90	692.24	688.71	681.66	677.92	684.27	681.00
$\omega_{24}+2\omega_{48}$	689.91	683.70	700.10	694.99	685.99	680.85	691.25	686.62
$\omega_{13}+\omega_{44}$	689.95	685.62	701.46	697.50	687.42	683.47	692.25	688.68
$2\omega_{13}+2\omega_{48}$	690.81	681.84	697.94	692.70	684.05	678.24	687.40	682.64
$\omega_{9}+\omega_{13}$	696.95	692.82	704.42	700.68	694.62	690.77	697.49	694.02
ω_{15}	698.10	692.28	710.45	706.13	694.12	688.58	697.00	692.45
$\omega_{13}+\omega_{24}+\omega_{48}$	703.75	697.22	718.87	713.34	699.44	693.64	706.81	701.55
$3\omega_{13}+\omega_{48}$	705.30	696.99	716.46	709.95	699.06	693.84	703.11	698.46
$2\omega_{24}$	714.17	711.07	726.64	723.51	712.47	709.58	718.89	716.24
$2\omega_{13}+\omega_{24}$	717.12	711.13	726.84	721.87	713.40	708.26	719.30	714.75
$4\omega_{13}$	719.43	713.00	729.66	724.23	714.69	711.12	718.80	715.57
$\omega_{16}+2\omega_{48}$	724.61	717.14	735.29	729.38	719.86	713.51	725.49	719.96
$\omega_{13}+\omega_{16}+\omega_{48}$	738.38	730.32	751.08	744.89	733.06	726.04	739.63	733.66
$\omega_{16}+\omega_{24}$	746.62	743.08	757.28	753.66	744.66	741.16	751.95	748.59
$2\omega_{13}+\omega_{16}$	751.40	744.32	762.33	756.72	746.80	740.78	751.79	746.55
ω_{8}	758.82	756.28	759.90	757.56	757.47	754.72	757.99	755.65
ω_{46}	767.43	762.59	776.02	772.35	764.17	759.55	767.11	763.25
ω_{27}	771.40	768.09	781.20	778.30	769.51	766.00	773.43	770.49
$2\omega_{16}$	781.49	777.40	791.86	788.22	779.18	775.39	785.74	782.35
$\omega_{12}+\omega_{48}$	791.58	786.13	799.14	794.82	787.82	783.00	792.13	788.07
$\omega_{36}+\omega_{48}$	792.17	787.79	805.03	801.14	789.54	785.54	796.70	793.08
ω_{23}	795.14	792.60	806.27	803.52	793.86	791.02	800.32	797.69
$\omega_{28}+2\omega_{48}$	798.23	789.63	807.28	801.21	792.21	784.60	797.11	790.83
$\omega_{12}+\omega_{13}$	805.27	800.22	813.47	809.28	801.87	797.42	806.31	802.48
$\omega_{13}+\omega_{36}$	806.27	802.02	819.31	815.42	803.79	799.86	809.18	805.60
$\omega_{47}+2\omega_{48}$	808.03	800.48	818.38	812.80	803.19	796.81	808.03	802.59
$\omega_{13}+\omega_{28}+\omega_{48}$	812.58	803.49	826.33	820.00	806.12	797.81	811.04	804.13
ω_{11}	820.54	814.49	833.52	828.98	819.91	814.70	821.24	816.05
$\omega_{24}+\omega_{28}$	823.15	818.86	834.37	830.55	820.58	816.46	826.72	823.05
$\omega_{13}+\omega_{47}+\omega_{48}$	823.78	817.36	836.54	831.29	815.65	808.04	824.87	819.83
$2\omega_{13}+\omega_{28}$	825.92	817.95	832.77	826.82	820.54	813.49	825.49	819.50
$\omega_{24}+\omega_{47}$	830.72	826.30	840.69	836.80	828.21	824.06	833.46	829.71
$2\omega_{13}+\omega_{47}$	834.55	826.54	844.69	838.59	829.43	822.42	833.86	827.81
$\omega_{44}+2\omega_{48}$	842.10	835.94	851.29	846.35	838.21	832.97	843.02	838.49

$5\omega_{48}$	845.61	842.61	851.31	848.32	843.51	842.26	845.37	843.83
$\omega_9+2\omega_{48}$	851.05	844.97	859.63	848.32	847.24	842.06	851.33	847.08
$\omega_{24}+3\omega_{48}$	855.11	848.13	865.48	860.10	850.16	845.78	854.96	851.20
$\omega_{13}+\omega_{44}+\omega_{48}$	856.22	849.44	866.79	861.35	851.88	845.91	856.76	851.66
$\omega_{16}+\omega_{28}$	857.19	852.20	868.66	864.39	854.08	849.47	859.85	855.79
$\omega_{13}+4\omega_{48}$	858.48	854.08	867.98	863.70	855.26	852.58	858.82	855.98
$\omega_{15}+\omega_{48}$	861.11	851.91	881.34	875.13	853.36	844.50	857.28	849.97
$\omega_9+\omega_{13}+\omega_{48}$	864.18	857.62	872.30	867.14	860.08	854.23	864.43	859.50
$\omega_{24}+\omega_{44}$	864.73	861.59	876.73	873.84	863.00	859.95	869.98	867.11
$\omega_{16}+\omega_{47}$	866.39	860.34	878.00	872.99	862.42	857.15	867.90	863.44
$\omega_{13}+\omega_{24}+2\omega_{48}$	868.87	860.76	882.12	875.96	863.00	857.44	867.96	863.18
$2\omega_{13}+\omega_{44}$	869.10	863.14	880.37	875.48	865.42	860.27	870.74	866.29
$2\omega_{13}+3\omega_{48}$	870.82	866.30	881.01	876.67	867.44	864.74	871.80	868.90
$\omega_9+\omega_{24}$	871.87	868.79	881.77	878.77	870.31	867.22	874.50	871.72
ω_{26}	872.39	868.36	881.57	878.37	869.98	865.87	872.97	869.54
$\omega_{13}+\omega_{15}$	875.29	865.96	888.17	881.85	868.07	859.43	871.48	864.43
$\omega_9+2\omega_{13}$	876.44	870.73	884.52	879.92	873.00	868.04	877.26	872.95
$2\omega_{24}+\omega_{48}$	880.42	875.19	894.11	889.74	877.24	872.67	883.70	879.68
$2\omega_{13}+\omega_{24}+\omega_{48}$	882.24	874.41	894.36	888.12	876.95	871.57	882.99	878.25
$3\omega_{13}+2\omega_{48}$	884.87	880.53	N/A	N/A	881.36	879.05	885.05	882.53
$\omega_{16}+3\omega_{48}$	890.16	881.89	900.61	894.06	884.63	879.12	889.90	885.13
$\omega_{13}+2\omega_{24}$	893.30	888.25	906.76	902.35	890.26	885.75	896.99	892.97
$\omega_{16}+\omega_{44}$	898.66	894.97	910.66	907.06	896.61	893.08	903.03	899.76
$4\omega_{13}+\omega_{48}$	899.67	895.97	N/A	N/A	897.18	895.66	900.37	898.47
$\omega_{13}+\omega_{16}+2\omega_{48}$	904.30	894.73	917.06	909.89	897.31	890.58	901.41	895.71
$\omega_9+\omega_{16}$	905.52	901.85	914.02	910.60	903.54	899.98	907.95	904.77
$\omega_{16}+\omega_{24}+\omega_{48}$	913.62	907.32	927.67	922.41	909.72	904.17	917.95	913.06
$2\omega_{13}+\omega_{16}+\omega_{48}$	918.82	909.70	N/A	N/A	914.92	909.09	N/A	N/A
$\omega_{13}+\omega_{16}+\omega_{24}$	N/A	N/A	N/A	N/A	922.86	915.93	N/A	N/A
$\omega_8+\omega_{48}$	925.76	920.94	932.49	928.78	923.02	918.50	924.79	920.99
$\omega_{13}+\omega_{16}+\omega_{24}$	926.53	920.47	N/A	N/A	922.86	915.93	N/A	N/A
$\omega_{46}+\omega_{48}$	929.32	920.81	934.87	929.72	N/A	N/A	924.76	918.63
ω_{14}	929.47	924.61	939.73	935.93	926.28	921.49	928.59	924.55
$2\omega_{28}$	933.24	928.14	944.12	940.16	929.92	925.26	935.20	931.46
$\omega_{27}+\omega_{48}$	936.79	931.15	950.08	946.21	933.05	927.61	939.19	934.74
$\omega_8+\omega_{13}$	940.24	935.50	946.85	942.90	937.55	933.07	939.85	936.03
$\omega_{28}+\omega_{47}$	941.09	935.51	952.80	948.74	937.70	932.45	944.23	939.78
$\omega_{13}+\omega_{46}$	941.34	935.73	952.10	947.36	936.43	929.18	940.35	934.46
$2\omega_{16}+\omega_{48}$	948.54	941.75	959.87	954.68	944.36	938.39	951.31	946.29
$2\omega_{47}$	949.74	944.32	958.77	954.47	946.42	941.59	950.96	946.78
$\omega_{13}+\omega_{27}$	950.44	944.87	962.79	958.26	946.92	941.45	951.57	946.73
ω_{43}	950.70	945.72	962.04	958.66	947.59	943.25	953.45	950.44
ω_{45}	950.84	946.08	959.09	955.62	947.59	942.82	951.52	947.73
$\omega_{12}+2\omega_{48}$	956.44	949.61	N/A	N/A	950.85	943.22	N/A	N/A
$\omega_{36}+2\omega_{48}$	956.93	950.73	N/A	N/A	953.07	947.76	958.64	953.98
ω_{10}	959.66	954.46	967.86	963.93	956.06	950.76	958.62	954.32
$\omega_{13}+2\omega_{16}$	961.92	955.57	N/A	N/A	958.10	952.26	N/A	N/A
$\omega_{23}+\omega_{48}$	962.95	958.31	974.18	970.24	960.50	955.38	966.45	962.65
ω_{25}	964.02	958.95	976.64	972.85	961.15	956.01	963.68	959.30
$\omega_{12}+\omega_{13}+\omega_{48}$	971.12	963.03	N/A	N/A	965.50	958.33	N/A	N/A
$\omega_{13}+\omega_{36}+\omega_{48}$	971.63	964.95	N/A	N/A	967.38	961.43	N/A	N/A
$\omega_{28}+\omega_{44}$	974.37	970.33	985.82	982.00	971.96	967.95	977.97	974.49
$\omega_{13}+\omega_{23}$	975.50	970.98	987.12	983.12	972.95	968.66	977.74	974.05
$\omega_{24}+\omega_{36}$	980.35	977.26	992.73	989.59	978.72	975.60	986.20	983.25
$\omega_{12}+\omega_{24}$	981.17	977.51	991.92	988.43	978.97	975.51	984.18	981.02
$\omega_9+\omega_{28}$	981.52	977.47	991.96	988.41	979.14	975.18	982.10	978.66
$\omega_{44}+\omega_{47}$	982.85	978.47	993.34	989.39	980.43	976.26	986.50	982.75
$\omega_{12}+2\omega_{13}$	984.26	977.33	N/A	N/A	979.90	974.10	N/A	N/A
$2\omega_{13}+\omega_{36}$	985.31	979.55	N/A	N/A	981.77	976.76	N/A	N/A
$\omega_{11}+\omega_{48}$	988.02	980.43	1000.18	994.89	982.68	975.77	987.55	981.99
$\omega_{24}+\omega_{28}+\omega_{48}$	989.26	982.14	N/A	N/A	984.56	977.90	990.01	984.33
$\omega_9+\omega_{47}$	990.08	985.77	999.56	995.86	987.78	983.63	991.09	987.45
$\omega_{24}+\omega_{47}+\omega_{48}$	997.05	990.37	N/A	N/A	992.97	987.05	N/A	N/A

$\omega_{11}+\omega_{13}$	999.87	992.64	1006.99	1002.09	994.75	988.19	997.28	991.96

S3. EXAMPLES FOR QUICK START OF KYLIN-V

Here we provide the detailed input and command lines for three examples in this paper. Through this section, we expect users can become familiar with Kylin-V and get start for their own models immediately.

A. Calculation of spectra of PBI-I trimer using SS-TD-DMRG

The PBI-trimer has 6 excitonic states and 15 vibrational modes. One should prepare a Hamiltonian input file `pbi.input` before calculation with following contents:

```
# electronic part
6 14               // number of excitonic states, non-zero excitonic Hamiltonian terms
1 1 2.13
1 3 -0.06203345
3 1 -0.06203345    // i j \epsilon_{ij}
...

# vibrational part
15                 // number of vibrational modes
1 0.036847869299999995 10 // K, \omega_{K}, maximal occupation number
2 0.17567873039999998 10
...

# 1st-order coupling part
18                 // number of terms
1 1 1 0.043909303913508266 // K i j g_{ij}^{K}
2 1 1 0.16480125714678628
...

# 2nd-order coupling part
0                  // number of terms

#reordering
0 1 6 7 8 9 10 2 3 11 12 13 14 15 4 5 16 17 18 19 20
// default: 0~N_{ex}-1 = ex DoFs; N_{ex}~N_{ex}+1 = vib DoFs
```

All comments labelled with `#` are required and those with `//` are deleted in pratice. Then we can construct the necessary operators and states by running the `kln-holstein.a` command:

```
kln-holstein.a -i pbi.input -t 1e-8
# -i (input file)
# -t (truncation threshold for MPO compression)
```

Once the construction program is finished, two new directories exist: `State/, Op/` which save the MPSs and MPOs respectively. The newest version builds not only the total Hamiltonian `Op/H_tot` but also all the density single operators $\{\hat{b}_j^\dagger \hat{b}_j\}$ named as `Op/Density_j` and pure excitonic and vibrational Hamiltonian `Op/H_e,Op/H_v`. Some other operators except raising/lowering operators $(\hat{b}_j^\dagger, \hat{b}_j)$ which break the U(1)-symmetry are not built. Despite this, one may need some special operators due to their problems. Kylin-V supports the addition and multiplication of operators by `kln-add-mpo.a` commands. One can always build their own operators in this way unless they do not consist of fundamental operators.

```
kln-add-mpo.a -x o1 -y o2 -a c1 -b c2 -o res
# -x (MPO file 1)
# -y (MPO file 2)
# -a (coefficient for MPO 1)
# -b (coefficient for MPO 2)
# -o (result MPO file)
```

This created a resulting operator of $\hat{O}_{res} = c1\hat{O}_1 + c2\hat{O}_2$.

Similarly for MPS wave functions, Kylin-V only builds N_{ex} states: `State/Ele-*.mps` without vibrational excitation. But one can still obtain assigned state using `kln-add-mps.a` command.

```
kln-add-mps.a -x s1 -y s2 -a c1 -b c2 -o res
# -x (MPS file 1)
# -y (MPS file 2)
# -a (coefficient for MPS 1)
# -b (coefficient for MPS 2)
# -o (result MPS file)
```

This created a resulting operator of $|res\rangle = c1\,|s1\rangle + c2\,|s2\rangle$. For example, the PBI-1 model need linear combination of three S_1 states: $\frac{1}{\sqrt{3}}(\left|S_1^1\right\rangle + \left|S_1^2\right\rangle + \left|S_1^1\right\rangle)$:

```
kln-add-mps.a -x State/Ele-1.mps -y State/Ele-3.mps -a 0.5773502691896258 -b 0.5773502691896258
-o State/med.mps
kln-add-mps.a -x State/med.mps -y State/Ele-5.mps -a 0.5773502691896258 -b 0.5773502691896258
-o State/init_s1.mps
```

And the inital linearly combined S_2 state can be obtained in the same way. Thereafter, we can carry out the time evolution using `kln-tdvp.a` command:

```
kln-tdvp.a -w State/init_s1.mps -o Op/H_tot -s [3,50,2447,0] -t 1e-6 -v 0.2 -D 6000
# -w inital wave function
# -o Hamiltonian operator
# -t SVD threshold
# -v time step
# -D maximal bond dimension
# -s four stages of time evolution :
[global Krylov, 2TDVP, SS, 1TDVP].
```

Note that running SS needs at least one 2TDVP sweep. The resulting wave function at step j is saved as `State/T-j.mps` for subsequent analyzation. To simulate the final absorption spectra, one has to obtain the so-called time-correlation function. This needs the `kln-join.a` command:

```
kln-join.a -a State/T-0.mps -b State/T-j.mps -o Op/Eye
```

The line above calculates the expectation $\langle\Psi_0|\hat{I}\left|\Psi_j\right\rangle$. Then we can use Fourier transform to obtain the spectra.

B. Calculation of population of spin-boson model using HM-TD-DMRG

The only difference between TD-DMRG and HM-TD-DMRG lies on the MPO construction. HM-TD-DMRG uses different input format and command `kln-holstein-map.a`. The HM input file is much more complicated to write because we use the projected-purification in Kylin-V. Therefore, we recommend users to generate HM input file by the python script, `bin/hm.py` together a regular input file `no_mapping.input` in section S3 A. Beform performing HM, one should make sure that the number of effective modes (per block) N_{eff} and blocks N_b are well-defined in `bin/hm.py`:

```
# number of direct modes
Neff = 6

# number of INDIRECT blocks
Nbk = 14
```

Copy the file into your working directory and run:

```
python3 hm.py no_mapping.input
```

The HM input file `map.input` then exists. Remember to change the `NumTerm` in the first line into realistic number of terms in the file. After these preparation, one can build the HM Hamiltonian and perform time-evolution by a series of commands:

```
kln-holstein-map.a -i map.input -t 1e-8
kln-tdvp.a -w State/Ele-1.mps -o Op/H_tot -s [0,160,160,0] -v 0.05 -D 6000 -t 1e-4
for((i=0;i<320;++i)); do kln-join.a -a State/T-$i.mps -b State/T-$i.mps -o Op/Density_1; done
```

Then the population dynamics of spin-up state is obtained.

C. Calculation of anharmonic frequencies of ethylene using VHCI

Kylin-V currently supports VHCI Watson models with Coriolis couplings thus accepts special input file form:

```
1  1  0  0  0  0 5.927529690151226  \\ \omega_{i} Q_{i} Q_{i}
-1  -1  0  0  0  0 -5.927529690151226 \\ \omega_{i} P_{i} P_{i}
2  2  0  0  0  0 6.339926014913254
-2  -2  0  0  0  0 -6.339926014913254
...
1  -2  1  -2  0  0  -0.02773628575631299 \\ \zeta_{ijkl} Q_{i} P_{j} Q_{k} P_{l}
...
1  1  1  1  0  0  0.0635927789210315 \\ F_{ijkl} Q_{i} Q_{j} Q_{k} Q_{l}
1  1  1  1  1  0  -0.002136909096219944 \\ F_{ijklm} Q_{i} Q_{j} Q_{k} Q_{l} Q_{m}
1  1  1  1  2  6  0.0010678391467589675 \\ \\ F_{ijklmn} Q_{i} Q_{j} Q_{k} Q_{l} Q_{m} Q_{n}
...
```

Kylin-V treats Watson Hamiltonian terms equally because all of them can be represented by $\hat{Q}, \hat{P}$ and scalars. After that one can run the `kln-watson-hci.a` command:

```
kln-watson-hci.a ethylene.inp 100 10 0.5 0.01
# [input file] [number of targetted states] [maximal total quanta]
[threshold for VHCI] [threshold for ENPT2]
```

Then one can easily find all the information like variational and perturbed energies, dominating configurations and time cost in the output.

[1]M. B. R. GEORGES and M. HERMAN, Molecular Physics **97**, 279 (1999).
[2]A. U. Bhatty and K. R. Brorsen, Molecular Physics **119**, e1936250 (2021).