Supporting Information: An approximation to vibrational coupled cluster method for CH-stretching of large molecules: Application to naphthalene and anthracene

Nivedhitha Palanisamy[1], Subrata Banik*[1a]
[1] *Department of Chemistry, School of Chemical and Biotechnology, SASTRA Deemed University, Thanjavur 613401, Tamil Nadu, India*
(Dated: June 10, 2023)

[a] subratachem@gmail.com

Table S1. The harmonic oscillator description for vibrations of naphthalene, calculated by using B3P86 functional with 6-311G(2d,2p) basis set. The numbers in the parenthesis are obtained using B3LYP functional and the same basis set.

Mode No.	Symmetry	Frequency	Intensity	Approx. Description
1	a_g	3206.0 (3190.3)	ia	CH str
2	a_g	3180.4 (3165.7)	ia	CH str
3	a_g	1629.6 (1612.1)	ia	CCC bend
4	a_g	1497.1 (1498.5)	ia	HCC bend
5	a_g	1419.1 (1390.6)	ia	CC str+CCC bend
6	a_g	1182.5 (1188.2)	ia	HCC bend
7	a_g	1051.8 (1045.5)	ia	CC str
8	a_g	779.8 (773.6)	ia	CCC bend
9	a_g	520.9 (520.5)	ia	CCC bend
10	a_u	996.7 (995.3)	ia	Tors
11	a_u	849.4 (849.6)	ia	Tors
12	a_u	638.7 (637.1)	ia	Tors
13	a_u	186.1 (186.6)	ia	Tors
14	b_{1g}	963.1 (962.2)	ia	Tors
15	b_{1g}	727.7 (728.9)	ia	Tors
16	b_{1g}	395.6 (396.7)	ia	Tors
17	b_{1u}	3192.9 (3177.4)	49.1 (63.3)	CH str
18	b_{1u}	3174.7 (3159.7)	6.2 (6.2)	CH str
19	b_{1u}	1653.9 (1641.6)	4.6 (4.8)	CC str
20	b_{1u}	1422.2 (1426.5)	2.0 (4.3)	HCC bend
21	b_{1u}	1290.9 (1291.3)	6.4 (6.4)	CCC bend+HCC bend
22	b_{1u}	1153.0 (1154.5)	5.0 (3.9)	CCC bend
23	b_{1u}	810.6 (811.1)	0.2 (0.3)	CC str+CCC bend
24	b_{1u}	363.3 (366.5)	1.3 (1.4)	CCC bend
25	b_{2g}	1002.7 (1001.4)	ia	Tors
26	b_{2g}	900.3 900.1)	ia	Out plane bending
27	b_{2g}	788.5 (788.0)	ia	Tors+Out plane bending
28	b_{2g}	478.4 (481.2)	ia	Tors+Out plane bending
29	b_{2u}	3205.1 (3189.1)	37.8 (49.7)	CH str
30	b_{2u}	3177.9 (3162.0)	0.5 (0.9)	CH str
31	b_{2u}	1560.6 (1551.0)	9.7 (9.1)	CC str
32	b_{2u}	1406.5 (1389.7)	1.3 (1.0)	CC str
33	b_{2u}	1240.4 (1231.7)	1.2 (0.6)	CC str
34	b_{2u}	1168.7 (1167.8)	0.8 (0.8)	HCC bend
35	b_{2u}	1043.4 (1034.6)	7.5 (6.6)	CC str
36	b_{2u}	633.6 (638.1)	3.5 (3.4)	CCC bend
37	b_{3g}	3192.1 (3176.0)	ia	CH str
38	b_{3g}	3173.6 (3157.7)	ia	CH str
39	b_{3g}	1686.0 (1668.6)	ia	CC str+CCC bend
40	b_{3g}	1499.5 (1496.9)	ia	HCC bend
41	b_{3g}	1271.9 (1275.0)	ia	HCC bend
42	b_{3g}	1172.7 (1173.9)	ia	CC str
43	b_{3g}	953.8 (956.6)	ia	CCC bend
44	b_{3g}	518.1 (520.1)	ia	CCC bend
45	b_{3u}	982.1 (980.5)	3.5 (3.0)	Tors
46	b_{3u}	800.1 (799.7)	105.9 (98.9)	Tors
47	b_{3u}	485.3 (487.2)	19.2 (17.4)	Tors+Out plane bending
48	b_{3u}	171.7 (172.7)	2.1 (1.9)	Tors+Out plane bending

[a] ia=inactive modes

Table S2. The harmonic oscillator description for vibrations of anthracene, calculated by using B3P86 functional and 6-311G(2d,2p) basis set. The numbers in the parenthesis are with B3LYP functional and 6-311G(2d,2p) basis set.

Mode No.	Symmetry	Frequency	Intensity	Approx. Description
1	a_g	3206.2 (3190.4)	ia	CH str
2	a_g	3181.0 (3166.3)	ia	CH str
3	a_g	3173.6 (3158.4)	ia	CH str
4	a_g	1610.3 (1592.3)	ia	CC str+CCC bend
5	a_g	1527.1 (1520.8)	ia	CC str+CCC bend
6	a_g	1447.1 (1422.4)	ia	CC str+CCC bend
7	a_g	1299.4 (1287.4)	ia	CC str
8	a_g	1187.0 (1193.4)	ia	HCC-bend
9	a_g	1037.9 (1029.8)	ia	CC str
10	a_g	770.1 (763.8)	ia	CCC bend
11	a_g	639.6 (644.5)	ia	CCC bend
12	a_g	399.7 (398.3)	ia	CCC bend
13	a_u	997.8 (996.6)	ia	Tors
14	a_u	868.6 (868.5)	ia	Tors
15	a_u	761.3 (760.9)	ia	Tors+out plane bending
16	a_u	505.0 (507.6)	ia	Tors
17	a_u	120.9 (121.2)	ia	CCC bend+HCC bend+Tors
18	b_{1g}	974.8 (973.6)	ia	Tors
19	b_{1g}	770.6 (770.4)	ia	Tors
20	b_{1g}	483.4 (484.4)	ia	Tors+out plane bending
21	b_{1g}	234.3 (235.4)	ia	CCC bend+Tors+out plane bending
22	b_{1u}	3193.6 (3177.9)	51.6 (68.2)	CH str
23	b_{1u}	3175.6 (3160.8)	10.0 (15.6)	CH str
24	b_{1u}	3171.4 (3156.2)	10.4 (7.0)	CH str
25	b_{1u}	1684.3 (1669.5)	6.3 (6.4)	CC str
26	b_{1u}	1496.3 (1491.2)	1.9 (1.7)	HCC-bend
27	b_{1u}	1347.4 (1342.9)	6.5 (4.6)	CC str
28	b_{1u}	1295.0 (1296.8)	4.8 (5.7)	HCC-bend
29	b_{1u}	1173.9 (1176.6)	7.0 (5.5)	HCC-bend
30	b_{1u}	921.6 (924.1)	1.7 (1.8)	CCC bend
31	b_{1u}	663.4 (663.1)	0.7 (0.7)	CCC bend
32	b_{1u}	234.9 (236.2)	1.3 (1.3)	Tors
33	b_{2g}	998.3 (997.1)	ia	Tors
34	b_{2g}	919.3 (920.4)	ia	Tors
35	b_{2g}	845.2 (845.7)	ia	Tors
36	b_{2g}	787.5 (787.9)	ia	Tors+out plane bending
37	b_{2g}	595.0 (594.0)	ia	Tors
38	b_{2g}	269.2 (269.8)	ia	Tors+out plane bending
39	b_{2u}	3205.8 (3189.9)	48.9 (64.3)	CH str
40	b_{2u}	3180.0 (3164.6)	0.02 (0.02)	CH str
41	b_{2u}	1593.1 (1578.6)	6.5 (5.8)	CC str
42	b_{2u}	1488.7 (1489.5)	2.4 (2.3)	HCC bend
43	b_{2u}	1431.6 (1410.4)	1.7 (0.7)	CC str
44	b_{2u}	1392.1 (1374.6)	4.2 (4.7)	CC str+CCC bend
45	b_{2u}	1190.4 (1190.5)	2.1 (1.3)	CC str+CCC bend
46	b_{2u}	1160.4 (1156.3)	1.6 (1.6)	CC str+CCC bend
47	b_{2u}	1032.7 (1024.5)	6.8 (5.9)	CC str
48	b_{2u}	827.3 (820.1)	0.0 (0.1)	CC str
49	b_{2u}	616.6 (619.0)	8.5 (8.3)	CCC bend
50	b_{3g}	3193.4 (3177.6)	ia	CH str
51	b_{3g}	3175.2 (3159.7)	ia	CH str
52	b_{3g}	1683.4 (1665.4)	ia	CC str
53	b_{3g}	1636.9 (1624.6)	ia	CC str
54	b_{3g}	1418.4 (1422.8)	ia	HCC bend
55	b_{3g}	1294.0 (1299.5)	ia	HCC bend
56	b_{3g}	1209.4 (1213.9)	ia	HCC bend
57	b_{3g}	1132.5 (1128.5)	ia	HCC bend
58	b_{3g}	931.8 (933.7)	ia	CCC bend
59	b_{3g}	533.3 (537.2)	ia	CCC bend
60	b_{3g}	393.0 (397.0)	ia	CCC bend
61	b_{3u}	980.2 (978.9)	6.6 (5.8)	Tors
62	b_{3u}	902.5 (902.6)	55.6 (50.9)	Tors
63	b_{3u}	740.0 (741.0)	74.8 (70.5)	Tors
64	b_{3u}	477.2 (479.1)	22.0 (20.0)	Out plane bend
65	b_{3u}	388.2 (388.6)	0.0 (0.0)	Tors
66	b_{3u}	89.8 (90.5)	1.1 (0.9)	Tors

[a] ia=inactive modes

Table S3. Comparison of effective frequencies from EHO against VSCF and VCCM for pyridine molecule.

S.No	VSCF[a]	EHO[b]	VCCM
1	3094.6	3127.3	3043.4 , 3039.0[c]
2	3036.3	3102.9	3075.6
3	3030.6	3077.6	2994.7
4	1616.8	1621.1	1593.5
5	1501.3	1503.1	1486.3
6	1234.7	1236.5	1223.3
7	1092.1	1094.0	1075.3
8	1045.3	1046.0	1038.1
9	1006.8	1007.3	1001.4
10	611.9	612.1	606.8
11	1021.2	1027.6	976.9
12	922.5	933.0	864.9
13	388.5	390.5	365.5
14	1033.9	1040.5	981.6
15	979.9	987.9	926.5
16	772.8	774.3	750.2
17	753.3	761.1	688.1
18	417.3	418.6	404.0
19	3046.0	3118.7	3016.3, 3089.6[d]
20	3001.8	3074.8	3044.5
21	1611.9	1617.2	1589.0
22	1462.9	1464.8	1446.8
23	1369.2	1371.4	1351.7
24	1288.4	1296.1	1245.6
25	1167.0	1170.1	1150.0
26	1076.6	1078.6	1064.9
27	665.4	665.6	661.5

[a] VSCF results were obtained using 8 harmonic oscillator basis for each mode.
[b] Effective frequencies from the ground state EHO calculations
[c] Near equal contributions from 1_1.
[d] Near equal contributions from 19_1.

Table S4: Some of the intense transitions for different target set of pyridine in the frequency range 2900 to 3100 cm^{-1}.

Target set	**Freq.**	**Final state**	**Intensity**
All modes	3039.0	$0.23*17_1 16_1 4_1-0.16*1_1-0.05*22_1 16_1 12_1+0.05*2_1$	3.3
($10a_1+3a_2+5b_1+9b_2$)	3043.4	$0.50*17_1 16_1 4_1+0.17*1_1-0.03*24_1 16_1 13_1 10_1-0.03*2_1$	2.4
	2916.2	$0.43*22_1 5_1-0.15*23_1 4_1+0.10*26_1 15_2+0.07*20_1$	4.0
	3044.5	$0.21*20_1-0.12*22_1 4_1-0.10*7_1 12_1 5_1+0.08*17_1 13_1 12_1$	6.9
	3069.1	$0.36*21_1 5_1-0.19*14_1 12_1 6_1-0.09*19_1+0.06*22_1 4_1$	5.0
100c-150f	3039.6	$0.26*2_1-0.13*1_1+0.11*5_2+0.11*3_1$	8.3
($7a_1+3a_2+5b_1+7b_2$)	2929.2	$0.50*22_1 5_1-0.19*23_1 4_1+0.11*20_1-0.03*19_1$	7.1
	3052.3	$0.23*20_1-0.16*22_1 4_1-0.16*19_1-0.09*11_1 13_1 12_1 8_1$	19.4
	3085.9	$0.40*22_1 4_1+0.23*21_1 5_1-0.20*19_1+0.02*23_1 16_1 14_1$	4.3
100c-200f		Same as 100c-150f set	
100c-250f	3038.3	$0.27*2_1-0.18*22_1 21_1+0.13*3_1+0.09*5_2$	7.2
($7a_1+3a_2+5b_1+8b_2$)	2924.5	$0.52*22_1 5_1-0.18*23_1 4_1+0.10*20_1+0.02*23_1 5_1$	5.5
	3049.4	$0.28*20_1-0.21*22_1 4_1-0.07*19_1-0.07*17_1 13_1 12_1 11_1$	16.7
	3078.9	$0.40*21_1 5_1+0.23*22_1 4_1-0.21*19_1-0.02*16_1 12_1 5_1$	6.5
100c-300f		Same as 100c-250f set	
150c-150f	3038.9	$0.23*2_1+0.18*5_2-0.17*1_1+0.10*3_1$	8.3
($5a_1+3a_2+5b_1+6b_2$)	2931.9	$0.26*22_1 5_1-0.19*17_1 16_1 15_1 13_1-0.14*16_2 15_1 13_1-0.10*23_1 4_1$	6.0
	3047.9	$0.31*17_1 12_1 5_1-0.13*19_1+0.12*20_1-0.08*18_1 16_1 15_1 12_1$	12.7
	3091.8	$0.57*22_1 4_1-0.14*19_1+0.12*21_1 5_1+0.04*23_1 16_1 14_1$	2.2
150c-200f		Same as 150c-150f set	
150c-250f	3038.6	$0.25*2_1-0.18*1_1+0.14*5_2+0.09*3_1$	8.6
($5a_1+3a_2+5b_1+7b_2$)	2930.7	$0.38*22_1 5_1+0.12*20_1-0.12*23_1 4_1+0.07*24_1 4_1$	6.1
	3050.2	$0.13*19_1-0.13*20_1+0.11*17_1 12_1 5_1+0.11*17_1 13_1 12_1 11_1$	13.4
	3087.2	$0.49*22_1 4_1+0.19*21_1 5_1-0.17*19_1-0.03*16_1 12_1 5_1$	3.2
150c-300f	3039.0	$0.27*2_1-0.13*1_1+0.12*5_2+0.12*3_1$	8.5
($6a_1+3a_2+5b_1+6b_2$)	2930.6	$0.25*22_1 5_1-0.14*17_1 16_1 15_1 13_1-0.09*16_2 15_1 13_1-0.07*23_1 4_1$	3.2
	3050.9	$0.21*20_1-0.14*19_1-0.13*22_1 4_1-0.10*17_1 13_1 12_1 11_1$	17.8
	3085.9	$0.45*22_1 4_1+0.23*21_1 5_1-0.19*19_1-0.016*16_1 12_1 5_1$	3.9
200c-150f	3039.7	$0.36*2_1+0.13*5_2+0.11*3_1+0.11*23_1 21_1$	7.6
($5a_1+3a_2+4b_1+6b_2$)	2931.1	$0.53*22_1 5_1+0.16*20_1-0.16*23_1 4_1-0.05*19_1$	10.5
	3048.0	$0.32*23_1 16_1 15_1-0.12*19_1+0.11*20_1+0.07*17_1 12_1 5_1$	11.6
	3090.9	$0.61*22_1 4_1-0.16*19_1+0.14*21_1 5_1+0.01*22_1 17_1 15_1$	2.6
200c-200f		Same as 200c-150f set	
200c-250f	3039.5	$0.35*2_1-0.13*22_1 21_1+0.11*5_2+0.10*3_1$	7.7
($5a_1+3a_2+4b_1+7b_2$)	2929.0	$0.52*22_1 5_1+0.14*20_1-0.13*23_1 4_1+0.04*24_1 4_1$	7.6
	3047.3	$0.19*23_1 16_1 15_1+0.19*17_1 12_1 5_1+0.12*20_1-0.11*19_1$	11.5
	3086.1	$0.49*22_1 4_1+0.21*21_1 5_1-0.18*19_1+0.03*21_1 17_1 15_1$	3.4
200c-300f	3039.3	$0.35*2_1-0.19*22_1 21_1+0.13*3_1+0.09*5_2$	6.7
($6a_1+3a_2+4b_1+7b_2$)	2929.2	$0.58*22_1 5_1-0.13*23_1 4_1+0.11*20_1+0.04*24_1 4_1$	6.3
	3048.4	$0.42*17_1 12_1 5_1+0.17*20_1-0.10*19_1-0.07*22_1 4_1$	13.7
	3084.7	$0.39*22_1 4_1+0.24*21_1 5_1-0.16*19_1+0.07*15_1 12_1 6_1$	3.6
250c-150f	3047.2	$0.72*1_1-0.10*5_2-0.04*22_1 21_1-0.03*22_2$	6.2
($5a_1+2a_2+4b_1+5b_2$)	2931.5	$0.53*22_1 5_1+0.17*20_1-0.17*23_1 4_1-0.06*19_1$	11.6
	3044.6	$0.30*20_1-0.20*19_1+0.14*23_1 4_1-0.13*23_1 17_1 14_1$	25.0
	3092.7	$0.41*22_1 4_1+0.34*22_1 17_1 15_1-0.09*19_1+0.08*21_1 5_1$	1.4
250c-200f		Same as 250c-150f set	
250c-250f	3047.2	$0.72*1_1-0.10*5_2-0.04*22_1 21_1-0.03*22_2$	6.2
($5a_1+2a_2+4b_1+6b_2$)	2931.5	$0.53*22_1 5_1+0.17*20_1-0.17*23_1 4_1-0.06*19_1$	11.6
	3044.6	$0.30*20_1-0.20*19_1+0.14*23_1 4_1-0.13*23_1 17_1 14_1$	25.0
	3085.0	$0.39*22_1 4_1-0.29*22_1 17_1 15_1-0.14*19_1+0.11*21_1 5_1$	2.5
250c-300f	3048.3	$0.68*1_1-0.07*5_2-0.06*22_1 21_1-0.03*5_1 4_1$	7.4
($6a_1+2a_2+4b_1+4b_2$)	2929.5	$0.60*22_1 5_1-0.14*23_1 4_1+0.12*20_1-0.03*19_1$	7.4
	3046.5	$0.42*20_1-0.18*19_1+0.14*23_1 4_1-0.08*22_1 4_1$	29.4
	3083.4	$0.34*22_1 4_1-0.32*22_1 17_1 15_1-0.14*19_1+0.13*21_1 5_1$	2.7

Continued on next page

Target set	Freq.	Final state	Intensity
300c-150f	3035.9	$0.33 * 2_1 + 0.12 * 3_1 + 0.10 * 23_1 21_1 - 0.09 * 22_1 21_1$	3.6
$(5a_1 + 2a_2 + 3b_1 + 5b_2)$	2931.0	$0.53 * 22_1 5_1 + 0.18 * 20_1 - 0.17 * 23_1 4_1 - 0.07 * 19_1$	12.2
	3042.4	$0.39 * 20_1 - 0.22 * 19_1 + 0.18 * 23_1 4_1 - 0.12 * 22_1 5_1$	30.4
	3090.5	$0.68 * 22_1 4_1 - 0.15 * 19_1 + 0.09 * 21_1 5_1 - 0.05 * 22_1 17_1 15_1$	2.0
300c-200f		Same as 300c-150f set	
300c-250f	3034.5	$0.36 * 22_1 17_1 12_1 - 0.18 * 2_1 - 0.08 * 17_2 4_1 - 0.08 * 23_1 17_1 11_1$	2.7
$(5a_1 + 2a_2 + 3b_1 + 6b_2)$	2929.2	$0.52 * 22_1 5_1 0.14 * 20_1 - 0.14 * 23_1 4_1 + 0.07 * 24_1 17_1 15_1$	8.7
	3042.2	$0.37 * 20_1 - 0.23 * 19_1 + 0.16 * 23_1 4_1 - 0.09 * 22_1 5_1$	30.2
	3085.5	$0.61 * 22_1 4_1 - 0.19 * 19_1 + 0.13 * 21_1 5_1 - 0.03 * 22_1 17_1 15_1$	3.0
300c-300f	3048.2	$0.68 * 1_1 - 0.08 * 5_2 - 0.05 * 22_1 21_1 - 0.03 * 5_1 4_1$	7.7
$(6a_1 + 2a_2 + 3b_1 + 6b_2)$	2929.5	$0.59 * 22_1 5_1 - 0.14 * 23_1 4_1 + 0.12 * 20_1 - 0.04 * 19_1$	7.6
	3044.8	$0.38 * 20_1 - 0.15 * 24_1 12_2 - 0.14 * 19_1 + 0.13 * 23_1 4_1$	25.6
	3084.0	$0.56 * 15_1 4_1 - 0.19 * 19_1 + 0.16 * 21_1 5_1 - 0.03 * 22_1 17_1 15_1$	3.4

Table S5. The vibrational modes included in the target sets for naphthalene

Sr.No	Target Set	Modes included
1	250c-200f	1 2 3 4 5 10 11 14 15 17 18 19 20 25 26 29 30 31 32 37 38 39 40 45 46
2	200c-200f	1 2 3 4 5 10 11 14 15 17 18 19 20 25 26 29 30 31 32 37 38 39 40 45 46 47
3	200c-250f	1 2 3 4 5 10 11 14 15 17 18 19 20 21 25 26 29 30 31 32 33 37 38 39 40 41 45 46 47

Table S6. Comparison of VCCM results against VPT2 and experimental values for naphthalene. The VCCM calculations used 200c-200f target set. The VPT2 and experimental values are taken from Mackie et. al.[1]

	EXP		VPT2				VCCM			
ID	Freq	Rel.I	Freq	Rel.I	Transition	Source	Freq	Transition	Rel.I	Source
A	2963.8	0.20	2934.0	0.12	20_1	17_1	2935.5	$31_1 5_1$	0.03	29_1
					41_1	....	.....	.....	....	
B	2972.4	0.30	2944.6	0.021	$18_1 17_1$	......	2961.4	$47_1 26_1 15_2$	0.52	18_1
					$20_1 3_1$	18_1	2960.2	18_1	0.28	18_1
					$5_1 19_1$	....	....	....	.....	.....
C	2981.3	0.20	2956.4	0.032	29_1	29_1	2966.5	$47_1 26_1 3_1$	0.11	18_1
						30_1	....	....	....	.....
					40_1	....	....	....	....	.....
C	2981.3	0.20	2956.6	0.02	41_1	....	....	....	....	.....
					5_1	....	....	....	.....	...
D	2989.0	0.28	2967.5	0.037	17_1	17_1	2974.6	$32_1 3_1$	0.38	29_1 and 30_1
					18_1		2973.5	$46_1 15_1 4_1$	0.06	.....
					$32_1 41_1$	....	....	....	....	......
E	3014.0	0.30	2987.7	0.04	$30_1 29_1$	.....	2978.9	$32_1 26_1 15_1$	0.36	17_1
				$20_1 39_1$	....	....	....	....	....	.....
F	3029.5	0.19	3016.6	0.074	18_1	18_1	2982.4	17_1	0.56	$17_1$and 18_1
					$40_1 3_1$	....	....	....	....	.....
G	3034.5	0.15	....	....	....	....	....	....	....	.....
H	3039.5	0.19	....	....	....	....	....	....	....	.....
I	3042.3	0.29	....	....	....	....	....	....	....	.....
J	3043.8	0.35	3028.8	0.12	29_1	29_1	3006.3	$47_2 14_1 10_1$	0.02	.....
					30_1	....	3005.4	$31_1 4_1$	0.41	29_1 and 30_1
					$4_1 19_1$	....	....	....	....	.....
					$20_1 39_1$	....	....	....	....	.....
K	3048.2	0.19	......	......	......	....	....	....	....	....
L	3052.2	0.17	......	......	......	....	....	....	....	....
M	3058.1	0.65	3046.5	0.16	29_1	29_1	3039.3	$40_1 31_1$	0.89	17_1
					30_1	....	....	....	....	....
					$4_1 19_1$	....	....	....	....	.....
N	3060.5	0.54	......	......	......	....	....	....	....	.....
O	3065.2	0.99	3053.4	1	17_1	17_1	3037.7	29_1	1	29_1
P	3071.4	0.10	......	......	......	....	3046.0	$47_2 15_1 11_1$	0.02	.....
Q	3076.2	0.37	......	......	......	....	....	....	....	.....
R	3079.2	1.00	3061.9	0.28	29_1	29_1	3054.1	$39_1 32_1$	0.45	17_1
					$31_1 3_1$	....	3056.3	$47_1 15_2 14_1$	0.04	.....
					$4_1 19_1$	....	....	....	....	.....
S	3092.6	0.21	......	......	......	....	3061.3	$39_1 32_1$	0.14	17_1
T	3083.9	0.16	3068.7	0.069	$18_1 17_1$	......	.....	....	.....	.....
					$40_1 19_1 18_1$	....	....	....	....	.....
U	3100.2	0.45	3088.1	0.24	29_2	....	3107.8	$40_1 19_1$	0.06	29_1
V	3102.6	0.21	....	....	$31_1 3_1$	....	....	....	....	.....
W	3109.4	0.45	....	....	....	.....	3144.7	$31_1 3_1$	0.03	30_1

Table S7. Comparison of VCCM results against VPT2 and experimental values for anthracene. The VCCM calculations used 200c-200f target set. The VPT2 and experimental values are taken from Mackie et. al.[1]

	EXP		VPT2				VCCM			
ID	Freq	Rel.I	Freq	Rel.I	Transition	Source	Freq	Rel.I	Transition	Source
F	2973.2	0.1	2956.2	0.036	$54_1 53_1$	22_1	.....	.....	......	.....
					$44_1 52_1$	....	.....	.....	......	.....
G	2979.6	0.08	2958.6	0.028	$54_1 53_1$	22_1	2931.8	0.03	$25_1 7_1$	23_1
					$44_1 52_1$	24_1	.....	.....	.....	.....
H	2992.5	0.07	2984.8	0.056	40_1	39_1	.....	.....	.....	.....
					$42_1 4_1$	....	.....	.....	.....	.....
					$43_1 52_1$	....	.....	.....	.....	.....
I	3011.8	0.07	2992.4	0.094	22_1	23_1	2943.3	0.02	$25_1 7_1$	23_1
					$26_1 4_1$	....	.....	......	.....	.....
J	3022	0.3	3004.2	0.05	$26_1 4_1$	22_1	2945.3	0.53	$54_1 41_1$	23_1 and 22_1
					$6_1 25_1$	....	....	....	....	....
					$54_1 52_1$	....	....	....	....	....
J	3022	0.3	3005	0.027	$26_1 4_1$	22_1	2949.7	0.01	$63_1 54_1 35_1$	....
					$6_1 25_1$	....	....	....	....	....
					$54_1 52_1$	....	....	....	....	....
L	3030	0.25	3023.6	0.14	24_1	24_1	2968.8	0.09	$26_1 5_1$	....
					$42_1 5_1$	......	....	......	....	....
M	3033.7	0.2	3030.6	0.043	$26_1 53_1$	39_1	2975.9	0.07	$52_1 27_1$	40_1
N	3046.7	0.3	3046.3	0.19	39_1	39_1	2984.7	0.08	$41_1 6_1$	.....
					40_1	....	....	....	......	.....
P	3055.4	0.54	3049.1	0.11	23_1	23_1	2996.7	0.05	$42_1 36_2$	40_1
					$42_1 53_1$	....	2997.4	0.03	$43_1 36_1 35_1$	40_1
Q	3062.3	0.45	.....	.....	....	....	....	.....	.....	.....
R	3065.3	0.72	3054.7	1	22_1	22_1	3012	0.43	$53_1 43_1$	22_1
S	3066.9	0.64	.....	.....	.....	....	......	....	.....	.....
T	3071.9	1	3060.7	0.44	39_1	39_1	3026.8	1.00	22_1	22_1
					$41_1 4_1$	....	3026.7	0.73	39_1	39_1
					$26_1 52_1$	....	....	....	....	.....
					$26_1 53_1$	....	....	....	....	.....
U	3077.8	0.4	3080.5	0.052	$26_1 52_1$	39_1	3037.6	0.05	$63_1 41_1$	.....
V	3081.8	0.24	3080.8	0.088	$42_1 52_1$	22_1	3047.2	0.01	$64_1 63_1 61_1 36_1$	36_1
					.....	....	3047.7	0.01	$35_1 20_1 27_1$	.....
W	3095.9	0.24	.....	.....	.....	....	3055.7	0.11	$26_1 4_1$	.....
						....	3056.1	0.02	$63_1 35_1 5_1$	.....
X	3109.6	0.32	3093.8	0.29	39_1	39_1	3084.8	0.08	$53_1 26_1$	.....
					$41_1 4_1$	....	3084.8	0.04	$53_1 26_1$	.....

Table S8. The vibrational modes included in the target sets for anthracene

Sr.No	Target Set	Modes included
1	250c-200f	1 2 3 4 5 6 7 13 14 18 19 22 23 24 25 26 27 28 33 34 35 36 39 40 41 42 43 44 50 51 52 53 54 55 61 62 63 64
2	200c-200f	1 2 3 4 5 6 7 13 14 18 19 22 23 24 25 26 27 28 33 34 35 36 39 40 41 42 43 44 50 51 52 53 54 55 61 62 63 64
3	200c-250f	1 2 3 4 5 6 7 13 14 18 19 22 23 24 25 26 27 28 32 33 34 35 36 39 40 41 42 43 44 50 51 52 53 54 55 61 62 63 64

[1] C. J. Mackie, A. Candian, X. Huang, E. Maltseva, A. Petrignani, J. Oomens, W. J. Buma, T. J. Lee and A. G. G. M. Tielens, *J. Chem. Phys.*, 2015, **143**, 224314.