Dataset S1. Optimized Cartesian coordinates and vibrational frequencies for all intermediates, transition states, reactants and products involved in the reactions of the methylidyne radical (CH) with diacetylene (C_4H_2).

C_4H_2
E(B2PLYPD3/CC-PVTZ) = -153.41210303613
E(CCSD(T)-F12/cc-pVTZ-f12) = -153.25751210
T1 diagnostic: 0.01430063

C	0.000000	0.000000	-0.682678
C	0.000000	0.000000	0.682678
C	0.000000	0.000000	1.892449
C	0.000000	0.000000	-1.892449
H	0.000000	0.000000	2.952882
H	0.000000	0.000000	-2.952882

Frequencies

235.0765	235.0765	514.8336
514.8336	647.0760	647.0760
652.8027	652.8027	907.6757
2068.6848	2239.9944	3481.5268
3482.3818		

IR intensities

7.2503	7.2503	0.0000
0.0000	0.0000	0.0000
86.8944	86.8944	0.0000
1.2166	0.0000	176.9049
0.0000		

CH
E(B2PLYPD3/CC-PVTZ) = -38.457258836327
E(CCSD(T)-F12/cc-pVTZ-f12) = -38.30811780
T1 diagnostic: 0.01287831

C	0.000000	0.000000	0.159796
H	0.000000	0.000000	-0.958776

Frequencies
2874.2920
IR intensities
149.9344

i1
E(B3LYP/6-311G**) = -192.110106922
E(RS2C) = -191.69195525

C	1.322691	-0.016997	0.000001
C	0.023844	-0.046391	0.000001
C	-1.284122	0.350266	0.000001
C	2.564880	-0.075707	0.000001
C	-2.412647	-0.406033	-0.000099
H	-1.447518	1.438575	0.000201
H	3.626370	-0.102066	-0.000599
H	-3.466726	-0.167338	0.000974

Frequencies

80.2613	145.4110	333.6182
355.3682	432.3169	435.2609
485.8203	577.0782	752.2755
788.3778	949.1610	1211.8915
1354.3197	1582.0728	1863.0350
2980.1574	3229.4710	3466.6726

IR intensities

5.9329	11.6042	45.5110
5.7281	5.2517	11.2072
36.2545	37.1977	11.6378
3.5578	13.3336	5.1211
9.3863	0.0516	0.4718
6.0786	1.2803	114.7589

i2
E(B2PLYPD3/CC-PVTZ) = -192.00932663068
E(CCSD(T)-F12/cc-pVTZ-f12) = -191.81833437
T1 diagnostic: 0.01687103

C	1.090194	-0.000041	0.019270
C	-0.256669	-0.000055	0.267263
C	-1.508527	0.657265	-0.057496
C	2.305350	0.000013	-0.072245
C	-1.508587	-0.657212	-0.057528
H	-2.044703	1.575689	-0.207651
H	3.359029	0.000056	-0.180267
H	-2.044895	-1.575564	-0.207669

Frequencies

139.4565	198.2912	413.8151
437.7457	513.8196	610.3686
652.3500	711.1039	803.2934
867.2183	974.7166	1047.7174
1427.4097	1664.8367	2090.7653
3266.4494	3311.1407	3491.1139

IR intensities

0.0547	0.0820	0.6234
0.1380	0.7423	0.2721
0.3117	1.2862	1.9735
0.6411	0.6661	0.9873
11.1327	9.1205	17.4168
6.8105	7.3595	8.1999

i3
E(B2PLYPD3/CC-PVTZ) = -192.05794990518
E(CCSD(T)-F12/cc-pVTZ-f12) = -191.86563854
T1 diagnostic: 0.01881318

C	1.220824	0.093840	0.000000
C	0.000000	0.759519	0.000003
C	-1.220822	0.093836	0.000001
C	2.297009	-0.466448	0.000000
C	-2.297007	-0.466453	-0.000007
H	-0.000002	1.841152	0.000044
H	3.233404	-0.963477	0.000016
H	-3.233422	-0.963445	-0.000044

Frequencies

140.8273	348.2556	357.8692
386.0024	574.6556	576.8031
598.2996	664.1855	665.9787
670.4516	959.3926	1127.7089
1379.6936	2075.6455	2142.0440
3185.2369	3483.7414	3487.2243

IR intensities

1.5725	2.3255	18.5164
0.0000	0.0000	95.0185
7.7116	19.0869	65.6691
5.6771	8.0750	19.5345
19.6990	23.3081	1.0771
0.0665	115.0005	22.9139

i4

E(B2PLYPD3/CC-PVTZ) = -191.99221199717
E(CCSD(T)-F12/cc-pVTZ-f12) = -191.80084939
T1 diagnostic: 0.01716206

C	1.091943	-0.080660	0.000013
C	-0.264339	-0.297377	0.000012
C	-1.613842	0.625583	-0.000002
C	2.281062	0.148589	0.000007
C	-1.434917	-0.836762	-0.000017
H	-1.841921	1.152892	-0.919336
H	3.324453	0.338007	-0.000086
H	-1.841969	1.152862	0.919337

Frequencies

185.8838	211.4855	449.3688
484.0509	594.9601	623.6346
664.1405	737.7260	899.3040
1003.6337	1070.9876	1078.6998
1509.2671	1839.4401	2165.7312
3098.8660	3188.7832	3482.5418

IR intensities

5.0294	1.0103	10.3366
0.3890	10.9563	45.1192
34.4954	6.9116	6.0617
43.7167	0.2522	7.7803
7.1995	20.0267	11.5351
32.5538	12.3162	93.4515

i5
E(B2PLYPD3/CC-PVTZ) = -192.06351184552
E(CCSD(T)-F12/cc-pVTZ-f12) = -191.86799853
T1 diagnostic: 0.02195857

C	-1.414858	-0.000824	0.000514
C	-0.062114	-0.001104	-0.000207
C	1.168462	-0.000336	0.000528
C	-2.626170	0.000667	-0.001328
C	2.523785	0.000754	0.000399
H	3.075466	0.928693	-0.000284
H	-3.686386	0.003066	0.001032
H	3.076295	-0.926699	-0.000183

Frequencies

144.2293	151.2137	347.2602
402.3956	497.8122	517.5519
635.5252	648.2695	725.6545
727.8090	1034.0586	1298.5536
1479.3573	2059.7111	2224.7792
3165.522	3259.3455	3488.4061

IR intensities

5.0956	5.2196	0.0671
0.0467	0.7650	0.1455
46.2001	47.4352	0.4254
48.0986	0.1885	0.0012
0.1550	7.6842	0.5772
1.2187	0.0595	116.2756

i1_i2
E(B3LYP/6-311G**) = -192.108978455
E(RS2C) = -191.69547112

C	-1.184146	0.066359	-0.000009
C	0.060585	0.426440	-0.000029
C	1.464972	0.524862	0.000014
C	-2.394900	-0.213395	0.000011
C	1.846984	-0.728191	-0.000006
H	2.023873	1.456150	0.000056
H	-3.424669	-0.463066	0.000040
H	2.639830	-1.449530	0.000028

Frequencies

-472.6012	108.5402	133.5652
298.5672	392.5643	409.0912
424.1732	441.0349	596.1251
819.8441	872.9355	1242.1861
1535.9916	1615.2910	1762.6569
3115.4077	3323.4549	3476.7306

IR intensities

0.7424	6.9264	14.1338
38.1787	35.6770	54.4989
11.1647	29.9351	86.4923
4.2932	8.3787	35.1180
2.5416	5.9465	10.4541
56.1387	22.5408	112.0235

i1_i3
E(B2PLYPD3/CC-PVTZ) = -191.93182471288
E(CCSD(T)-F12/cc-pVTZ-f12) = -191.73607578
T1 diagnostic: 0.05405081

C	-1.348851	-0.031724	-0.000006
C	-0.029778	-0.166744	0.000005
C	1.280390	0.023838	0.000006
C	-2.572655	0.035909	-0.000015
C	2.573549	-0.155551	0.000007
H	0.843696	1.101332	0.000030
H	-3.631183	0.097444	-0.000023
H	3.371557	0.566854	0.000007

Frequencies

-974.7224	119.7741	147.4160
417.8837	425.2943	435.5328
490.1249	542.5702	543.1616
693.5253	778.8189	850.0363
1432.6426	1746.4697	2023.9339
2597.8489	3271.7508	3478.3577

IR intensities

261.2150	1.0887	0.4233
0.0191	7.4026	5.5669
20.9576	52.2803	50.5401
386.5423	257.2039	17.2159
16.7769	50.4167	39.1879
53.2443	4.5795	165.3838

i2_i4
E(B2PLYPD3/CC-PVTZ) = -19190796367347
E(CCSD(T)-F12/cc-pVTZ-f12) = -191.71848001
T1 diagnostic: 0.01939159

C	1.094924	0.006891	0.008308
C	-0.257748	-0.030055	-0.095970
C	-1.517243	0.589061	-0.016309
C	2.312175	0.035202	0.031126
C	-1.492219	-0.837499	-0.065626
H	-2.127135	1.469941	-0.138158
H	3.371153	0.055619	0.064901
H	-2.083345	-0.107161	0.904084

Frequencies

-1368.1370	112.6002	197.0474
397.0871	507.2546	513.0915
618.6755	639.2961	783.1151
830.7212	907.7862	1139.823
1276.0615	1532.5522	1885.7348
2100.0964	3233.7323	3486.3401

IR intensities

154.0053	11.4495	7.3475
4.1409	36.6258	19.7493
5.1528	45.2586	31.6715
4.0226	11.8443	5.6984
5.0990	22.2650	138.7258
1.2535	5.3475	102.5232

i4_p1
E(B2PLYPD3/CC-PVTZ) = -191.90207131219
E(CCSD(T)-F12/cc-pVTZ-f12) = -191.71061426
T1 diagnostic: 0.01679626

C	1.086388	-0.000231	-0.006864
C	-0.296445	-0.091744	-0.031929
C	-1.499756	0.490901	-0.191230
C	2.292696	0.069715	0.022022
C	-1.463789	-0.908833	0.065142
H	-2.044335	1.322180	-0.605312
H	-2.021906	1.191596	1.414855
H	3.351672	0.127377	0.047615

Frequencies

-864.9663	177.2796	201.9100
255.0359	442.9485	521.7791
588.7313	614.3306	698.5064
735.3925	911.3040	990.7756
1096.0268	1254.9223	1669.7730
2182.9900	3249.9710	3481.3415

IR intensities

13.1437	2.8829	3.9289
0.1948	12.5716	3.5437
1.7834	52.2805	1.7631
30.3300	4.1586	4.6675
8.7993	33.7856	7.1235
9.4371	3.9102	87.2292

i4_i5
E(B2PLYPD3/CC-PVTZ) = -191.97846408840
E(CCSD(T)-F12/cc-pVTZ-f12) = -191.78509141
T1 diagnostic: 0.03063652

C	-1.190491	-0.060618	0.000000
C	0.124867	-0.403177	-0.000001
C	1.956942	0.562388	0.000000
C	-2.363449	0.239728	0.000000
C	1.326177	-0.778984	0.000000
H	2.133466	1.070643	0.933693
H	2.133457	1.070649	-0.933692
H	-3.391195	0.502691	0.000001

Frequencies

-657.0437	85.3021	177.5780
212.9758	467.9518	489.4260
590.5824	595.9483	735.2708
860.2750	970.8480	1093.2353
1428.7721	1891.1007	2223.5809
3170.2965	3292.2819	3482.7502

IR intensities

2.7026	0.0046	0.0678
0.1357	0.3101	1.4941
0.3760	0.2540	0.4110
3.6578	0.6912	4.7919
1.3386	20.4619	19.8104
6.1875	7.1666	8.2529

i5_p5
E(B2PLYPD3/CC-PVTZ) = -191.90557350489
E(CCSD(T)-F12/cc-pVTZ-f12) = -191.70963447
T1 diagnostic: 0.02767635

C	-1.5853510446	-0.0360752145	-0.0025352884
C	-0.2387452145	-0.0642135867	-0.0100224070
C	0.9994391138	-0.0935167428	-0.0170219062
C	-2.7961436142	-0.0124782124	0.0041394714
C	2.3194571771	0.1627059192	-0.0148278733
H	-3.8564515529	0.0100943672	0.0100487797
H	3.1955235392	-0.9670465344	-0.0570332536
H	2.6945985962	-1.6006059959	-0.0758815187

Frequencies

-800.5088	136.0688	142.6280
316.0508	367.6681	411.8683
478.3998	550.2294	600.5778
612.6627	721.1971	754.8321
1174.2524	1357.0406	2079.9567
2318.9303	3382.4005	3490.3364

IR intensities

975.1114	1.1106	3.4890
1.0693	0.2300	4.4132
0.0072	8.4773	50.2851
3.7092	37.4192	20.4998
9.2045	55.9415	148.5706
98.2876	125.9638	123.4410

p1
E(B2PLYPD3/CC-PVTZ) = -191.40936971150
E(CCSD(T)-F12/cc-pVTZ-f12) = -191.21845018
T1 diagnostic: 0.01371840

C	-1.026899	0.005928	-0.001241
C	0.360688	-0.039392	-0.000456
C	1.543481	0.575429	0.000272
C	-2.235196	0.030787	0.000645
C	1.558643	-0.831152	0.000409
H	2.091579	1.501344	0.000557
H	-3.295880	0.049055	0.001671

Frequencies

199.8608	219.8484	518.7340
543.4556	613.2289	704.1204
740.5472	901.9127	953.8552
1108.1315	1280.1540	1713.3055
2169.4049	3268.3179	3480.7066

IR intensities

6.2011	0.8064	2.9449
3.4445	53.2909	1.3820
29.1389	15.8882	3.0117
5.0817	34.5127	4.4530
6.7371	0.7003	83.9460

p2
E(B2PLYPD3/CC-PVTZ) = -191.40939833364
E(CCSD(T)-F12/cc-pVTZ-f12) = -191.21105952
T1 diagnostic: 0.02367040

C	1.305740	-0.001257	0.000008
C	0.000000	-0.002389	0.000000
C	-1.305740	-0.001259	-0.000008
C	2.538765	0.001731	0.000016
C	-2.538765	0.001736	-0.000016
H	3.598993	0.004318	0.000022
H	-3.598993	0.004301	-0.000024

Frequencies

117.2187	415.0558	415.0564
417.4006	428.1324	485.7109
485.7117	489.4374	490.9390
763.0047	1560.9630	1662.1454
1999.7392	3469.9206	3480.1092

IR intensities

4.7661	0.0001	0.0002
3.0075	3.0899	0.0004
0.0021	91.1144	92.7685
0.0000	12.2154	1.2286
0.0000	225.0383	0.0007

p3
E(B2PLYPD3/CC-PVTZ) = -191.39311245639
E(CCSD(T)-F12/cc-pVTZ-f12) = -191.19516299
T1 diagnostic: 0.02081073

C	-1.489173	0.000023	0.000494
C	-0.189833	0.000065	-0.000397
C	2.392217	-0.000024	0.000182
C	-2.771587	-0.000048	-0.000245
C	1.073062	-0.000007	-0.000093
H	2.955958	0.925583	0.000093
H	2.955930	-0.925640	0.000263

Frequencies

130.4536	141.6705	262.6791
287.7117	483.2834	608.8989
761.5467	980.6449	1040.9444
1368.5555	1516.3477	1965.4078
2203.8700	3126.7423	3208.5567

IR intensities

3.0436	0.0078	7.7409
10.1749	1.0758	3.0364
0.1174	30.2247	0.3906
11.3521	7.7707	154.7890
875.9303	1.8988	0.0011

p4
E(B2PLYPD3/CC-PVTZ) = -191.38181767278
E(CCSD(T)-F12/cc-pVTZ-f12) = -191.18794451
T1 diagnostic: 0.01618328

C	-1.175844	-0.000040	-0.000310
C	0.157457	-0.000067	-0.000128
C	1.402477	-0.666179	0.000082
C	-2.440923	0.000015	0.000212
C	1.402385	0.666238	0.000133
H	1.963509	-1.583811	0.000439
H	1.963178	1.584012	-0.000378

Frequencies

166.6848	172.3304	486.8356
497.3169	736.8380	781.9520
938.3998	957.3481	977.6678
1129.7659	1461.0628	1666.0068
2084.6977	3261.7128	3299.5490

IR intensities

1.8613	0.5850	12.1290
0.0812	0.3495	42.6186
10.9069	0.0001	1.3113
15.1644	145.0826	37.9012
940.2794	7.7509	12.6408

p5
E(B2PLYPD3/CC-PVTZ) = -190.75023926044
E(CCSD(T)-F12/cc-pVTZ-f12) = -190.54782176
T1 diagnostic: 0.03400174

C	1.305740	-0.001257	0.000008
C	0.000000	-0.002389	0.000000
C	-1.305740	-0.001259	-0.000008
C	2.538765	0.001731	0.000016
C	-2.538765	0.001736	-0.000016
H	-3.598993	0.004301	-0.000024

Frequencies

137.9794	280.3587	301.2332
413.5590	489.9967	609.6496
768.3278	794.1687	1461.4267
2000.2722	2239.1155	3481.9397

IR intensities

2.0602	5.0532	8.9848
4.3044	56.1985	4.9615
5.3892	23.2411	72.6765
580.3107	258.6792	96.4224