Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting information for:

$Si_2C_5H_2$ isomers - Search Algorithms Versus Chemical Intuition

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Table S1: Computed energies of $Si_2C_5H_2$ isomers in their singlet ground electronic states calculated at the RHF-fc-CCSD(T)/cc-pVTZ level of theory.

Isomer	Point Group	E a.u	$ZPVE$ kcal mol^{-1}	E+$ZPVE$ a.u	μ Debye	ΔE kcal mol^{-1}	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	C_{2v}	-769.34455300	31.1338	-769.29493818	0.39	0.00	0.00	0
2	C_s	-769.30700346	28.9567	-769.26085806	0.55	23.56	21.39	0
3	C_s	-769.30622036	29.0254	-769.25996548	0.86	24.05	21.95	0
4	C_s	-769.30650635	30.0193	-769.25866759	1.98	23.87	22.76	0
5	C_s	-769.30080521	28.4226	-769.25551095	1.92	27.45	24.74	0
6	C_s	-769.30228227	29.9493	-769.25455507	3.47	26.53	25.34	0
7	C_s	-769.30051910	29.1418	-769.25407873	1.57	27.63	25.64	0
8	C_s	-769.29942591	28.6063	-769.25383891	0.84	28.32	25.79	0
9	C_s	-769.29751788	28.8234	-769.25158491	0.73	29.51	27.20	0
10	C_{2v}	-769.29225878	26.9136	-769.24936927	1.11	32.82	28.59	0
11	C_{2v}	-769.29227928	27.4920	-769.24846803	0.87	32.80	29.16	0
12	C_s	-769.29198611	28.7788	-769.24612421	1.95	32.99	30.63	0
13	C_s	-769.28807432	27.3746	-769.24445016	2.88	35.44	31.68	0
14	C_{2v}	-769.28676187	26.9674	-769.24378662	2.19	36.26	32.10	0
15	C_s	-769.28745482	28.2094	-769.24250032	0.66	35.83	32.91	0
16	C_s	-769.28988467	30.0366	-769.24201834	2.01	34.30	33.21	0
17	C_s	-769.28351058	27.4983	-769.23968929	1.45	38.30	34.67	0
18	C_s	-769.27766241	28.9700	-769.23149582	1.67	41.97	39.81	0

Table S2: Computed energies of $Si_2C_5H_2$ isomers in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer	Point Group	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	C_{2v}	-770.6783795	0.049725	-770.628654	0.00	0
2	C_s	-770.6473552	0.046621	-770.600734	17.52	0
3	C_s	-770.6467098	0.046777	-770.599932	18.02	0
4	C_s	-770.6468126	0.048113	-770.598699	18.80	0
5	C_s	-770.6405389	0.045596	-770.594943	21.15	0
6	C_s	-770.6366832	0.047858	-770.588825	24.99	0
7	C_s	-770.6434194	0.046796	-770.596623	20.10	0
8	C_s	-770.6366782	0.045878	-770.590800	23.75	0
9	C_s	-770.6459363	0.046502	-770.599435	18.34	0
10	C_{2v}	-770.6408517	0.043746	-770.597106	19.80	0
11	C_{2v}	-770.6388129	0.044781	-770.594032	21.73	0
12	C_s	-770.6323326	0.046074	-770.586259	26.60	0
13	C_s	-770.6281327	0.044012	-770.584120	27.95	0
14	C_{2v}	-770.6303640	0.043823	-770.586541	26.43	0
15	C_s	-770.6297575	0.045459	-770.584299	27.83	0
16	C_s	-770.6300475	0.048269	-770.581779	29.41	0
17	C_s	-770.6253429	0.044247	-770.581096	29.84	0
18	C_s	-770.6290167	0.046505	-770.582512	28.95	0

Table S3: Computed energies of $Si_2C_5H_2$ isomers in their triplet ground electronic states calculated at the UB3LYP/6-311++G(2D,2P) level of theory.

Isomer	Point Group	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	C_{2v}	-770.5593138	0.045813	-770.513501	26.61	3
2	C_s	-770.5919917	0.044217	-770.547775	5.10	0
3	C_s	-770.5908066	0.044335	-770.546471	5.92	0
4	C_s	-770.5998711	0.048020	-770.551851	2.54	0
5	C_s	-770.5493823	0.043089	-770.506293	31.13	0
6	C_s	-770.5901774	0.047850	-770.542327	8.52	1
7	C_s	-770.6028184	0.047091	-770.555728	0.11	0
8	C_s	-770.5458270	0.043819	-770.502008	33.82	0
9	C_s	-770.5973847	0.043930	-770.553455	1.53	1
10	C_{2v}	-770.5654589	0.040228	-770.525231	19.24	1
11	C_{2v}	-770.5879971	0.042225	-770.545772	6.35	2
12	C_s	-770.6031721	0.047273	-770.555899	0.00	0
13	C_s	-770.5530911	0.042905	-770.510186	28.69	0
14	C_{2v}	-770.5734208	0.043028	-770.530392	16.01	0
15	C_s	-770.5775491	0.042831	-770.534718	13.29	1
16	C_s	-770.5800925	0.046639	-770.533453	14.09	0
17	C_s	-770.5485300	0.041919	-770.506611	30.93	1
18	C_s	-770.5896180	0.044982	-770.544636	7.07	1

Table S4: Computed energies of $Si_2C_5H_2$ isomers in their triplet ground electronic states calculated at the UCAM-B3LYP/6-311++G(2D,2P) level of theory.

Isomer	Point Group	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	C_{2v}	-770.4042262	0.045765	-770.358461	30.33	2
2	C_s	-770.4420825	0.044907	-770.397176	6.03	0
3	C_s	-770.4409137	0.045017	-770.395897	6.83	1
4	C_s	-770.4528250	0.049153	-770.403672	1.96	0
5	C_s	-770.3971970	0.044715	-770.352482	34.08	0
6	C_s	-770.4282405	0.047652	-770.380588	16.44	0
7	C_s	-770.4259617	0.045128	-770.380834	16.29	1
8	C_s	-770.3967635	0.045561	-770.351202	34.88	0
9	C_s	-770.4461641	0.044684	-770.401480	3.33	1
10	C_{2v}	-770.4027952	0.040792	-770.362003	28.10	1
11	C_{2v}	-770.4329533	0.043536	-770.389417	10.90	0
12	C_s	-770.4552664	0.048479	-770.406788	0.00	0
13	C_s	-770.4056914	0.044241	-770.361451	28.45	0
14	C_{2v}	-770.3968412	0.039765	-770.357077	31.19	3
15	C_s	-770.4106335	0.043386	-770.367247	24.81	1
16	C_s	-770.4270231	0.047675	-770.379348	17.22	0
17	C_s	-770.3982595	0.043319	-770.354941	32.53	1
18	C_s	-770.4415882	0.045810	-770.395778	6.91	1

Table S5: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **1** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a_2	191.7	0.0
2	b_1	215.1	14.8
3	a_1	229.4	2.1
4	b_2	420.1	104.2
5	b_2	509.6	17.6
6	a_2	570.6	0.0
7	b_1	600.0	0.0
8	a_1	610.4	42.5
9	a_1	781.9	1.0
10	b_1	825.4	20.7
11	b_2	880.6	0.2
12	a_2	909.4	0.0
13	a_1	984.0	28.7
14	b_2	997.4	16.5
15	a_1	1099.6	2.4
16	b_2	1266.6	12.4
17	a_1	1331.9	47.8
18	a_1	1454.7	4.3
19	b_2	1476.3	9.3
20	b_2	3201.7	1.9
21	a_1	3221.7	21.4

Table S6: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **2** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a''	88.3	0.3
2	a'	90.6	0.4
3	a''	229.9	0.2
4	a'	249.0	5.2
5	a'	324.6	0.6
6	a''	421.2	2.3
7	a'	440.5	7.9
8	a''	463.0	1.4
9	a'	502.2	1.0
10	a''	575.1	51.5
11	a'	633.3	31.2
12	a'	656.1	82.6
13	a'	794.3	7.3
14	a''	797.4	17.2
15	a'	983.4	5.9
16	a'	1188.1	1.9
17	a'	1360.8	24.1
18	a'	1694.8	4.7
19	a'	2141.4	0.7
20	a'	3159.9	3.2
21	a'	3461.4	84.1

Table S7: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **3** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a'	87.7	0.7
2	a''	119.5	1.6
3	a''	186.6	1.0
4	a'	246.3	0.2
5	a'	324.1	0.7
6	a''	389.4	3.1
7	a'	390.2	11.1
8	a''	518.4	2.9
9	a''	580.3	45.6
10	a'	581.3	52.4
11	a'	630.3	25.5
12	a'	702.8	5.5
13	a'	725.3	31.4
14	a''	790.3	9.9
15	a'	995.4	11.2
16	a'	1192.4	20.4
17	a'	1368.3	8.5
18	a'	1689.0	3.0
19	a'	2138.8	0.6
20	a'	3186.0	2.3
21	a'	3461.1	79.0

Table S8: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **4** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a''	138.7	2.7
2	a'	262.9	12.3
3	a''	280.2	0.0
4	a'	383.7	1.5
5	a''	408.7	0.8
6	a'	465.6	7.0
7	a''	526.8	8.3
8	a'	549.4	39.6
9	a'	610.0	2.7
10	a'	715.2	60.9
11	a''	790.5	17.9
12	a'	813.6	0.9
13	a'	891.1	9.3
14	a''	942.9	0.8
15	a'	1150.4	2.2
16	a'	1267.9	74.7
17	a'	1365.4	15.3
18	a'	1489.0	73.2
19	a'	1701.0	5.3
20	a'	3092.9	11.0
21	a'	3153.1	7.9

Table S9: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **5** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a''	146.1	2.0
2	a''	284.6	4.4
3	a'	309.4	14.6
4	a'	366.2	9.5
5	a''	454.0	3.8
6	a''	486.8	14.2
7	a'	494.2	33.5
8	a''	603.8	0.0
9	a'	613.4	10.5
10	a'	715.0	37.4
11	a''	765.5	19.3
12	a'	771.4	51.0
13	a'	821.2	8.6
14	a'	862.7	15.2
15	a'	958.1	19.8
16	a'	1200.8	2.1
17	a'	1309.0	47.0
18	a'	1387.5	70.0
19	a'	1868.7	278.6
20	a'	2275.8	112.0
21	a'	3187.9	0.8

Table S10: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **6** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a''	118.2	0.2
2	a''	213.7	1.6
3	a'	221.4	14.0
4	a'	274.3	18.8
5	a'	336.2	13.2
6	a''	362.6	11.2
7	a'	539.4	4.5
8	a''	561.8	1.5
9	a'	632.6	2.4
10	a'	662.4	41.9
11	a''	788.2	24.8
12	a'	807.7	6.1
13	a''	934.0	0.5
14	a'	967.5	0.3
15	a'	1127.4	0.7
16	a'	1273.2	24.9
17	a'	1404.9	20.7
18	a'	1513.6	3.8
19	a'	1847.3	4.7
20	a'	3153.1	29.7
21	a'	3210.6	3.7

Table S11: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **7** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a''	138.6	1.3
2	a'	236.6	21.6
3	a''	253.7	1.5
4	a'	352.7	6.9
5	a''	427.9	1.0
6	a'	477.4	15.2
7	a'	497.6	12.5
8	a''	530.9	0.2
9	a'	640.7	21.5
10	a''	662.4	14.9
11	a'	696.1	4.9
12	a'	722.5	31.8
13	a''	791.3	28.9
14	a'	877.2	13.0
15	a'	967.5	170.8
16	a'	1191.2	11.1
17	a'	1316.3	23.0
18	a'	1420.1	35.4
19	a'	1824.1	461.2
20	a'	3102.9	3.4
21	a'	3257.2	25.9

Table S12: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **8** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a''	149.5	0.0
2	a''	268.4	18.9
3	a'	306.0	5.9
4	a'	316.1	24.7
5	a''	405.2	2.7
6	a''	489.1	0.2
7	a'	500.3	6.5
8	a'	512.2	25.0
9	a''	606.0	0.3
10	a''	746.9	25.1
11	a'	757.8	25.0
12	a'	794.0	18.8
13	a'	860.2	42.6
14	a'	950.5	127.6
15	a'	1103.4	3.8
16	a'	1138.6	29.4
17	a'	1320.7	6.2
18	a'	1375.6	36.4
19	a'	1876.7	77.3
20	a'	2291.2	81.4
21	a'	3242.1	0.4

Table S13: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **9** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a'	85.7	1.6
2	a''	91.9	1.8
3	a''	225.1	0.1
4	a'	257.0	2.3
5	a'	308.4	5.4
6	a'	364.5	3.0
7	a''	400.9	1.1
8	a'	439.2	6.7
9	a''	480.6	0.7
10	a''	509.0	1.4
11	a'	569.9	20.7
12	a'	691.0	45.1
13	a''	834.5	46.7
14	a'	932.2	4.7
15	a'	1022.8	0.4
16	a'	1307.6	6.4
17	a'	1470.6	0.2
18	a'	1727.0	4.4
19	a'	2088.4	6.4
20	a'	3135.0	1.9
21	a'	3221.2	1.1

Table S14: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **10** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	b_2	71.5	1.2
2	b_1	92.7	2.4
3	b_2	188.4	2.1
4	b_1	246.5	0.1
5	a_1	351.0	0.0
6	b_2	431.5	0.5
7	b_1	432.8	2.5
8	b_2	481.8	0.1
9	a_1	487.7	2.2
10	b_1	507.4	0.1
11	b_2	591.2	44.9
12	b_1	625.8	37.7
13	b_1	653.9	2.1
14	b_2	786.6	14.2
15	a_1	895.7	0.1
16	b_2	1287.5	283.2
17	a_1	1340.0	5.1
18	a_1	1562.4	8.0
19	a_1	2096.0	0.4
20	a_1	2238.0	33.6
21	a_1	3457.9	119.6

Table S15: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **11** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	b_1	60.4	1.0
2	b_2	73.5	2.1
3	b_1	148.6	2.7
4	b_2	250.3	6.4
5	a_1	321.5	23.5
6	b_1	321.7	0.7
7	b_2	333.8	2.9
8	a_1	398.1	0.0
9	b_2	502.9	0.2
10	b_1	515.1	1.2
11	b_2	595.6	40.3
12	b_1	625.1	42.2
13	b_1	744.3	20.2
14	b_2	792.4	0.5
15	a_1	868.7	18.0
16	b_2	906.7	135.1
17	a_1	966.6	10.9
18	a_1	1964.0	8.9
19	a_1	2136.2	2.5
20	a_1	3254.7	0.9
21	a_1	3451.0	136.7

Table S16: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **12** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a''	113.9	0.2
2	a''	182.6	4.3
3	a'	276.0	0.9
4	a'	287.0	11.8
5	a''	300.0	6.2
6	a'	411.4	15.9
7	a''	422.0	0.3
8	a'	474.1	15.6
9	a'	527.3	26.7
10	a''	594.8	10.6
11	a''	687.6	51.9
12	a'	783.7	24.4
13	a'	871.3	38.4
14	a'	892.9	95.5
15	a'	1060.6	1.4
16	a'	1151.1	28.1
17	a'	1322.4	26.5
18	a'	1404.2	35.8
19	a'	1814.6	101.8
20	a'	3224.5	3.3
21	a'	3329.1	83.3

Table S17: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **13** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a''	143.5	4.7
2	a''	244.7	0.9
3	a'	261.5	6.9
4	a'	299.6	11.4
5	a''	345.9	2.2
6	a'	360.5	32.0
7	a''	476.0	8.5
8	a'	485.2	50.5
9	a''	512.5	0.0
10	a'	517.1	5.9
11	a'	627.1	78.5
12	a''	691.2	23.1
13	a'	745.7	27.8
14	a'	813.6	33.4
15	a'	880.4	13.7
16	a'	1115.9	22.4
17	a'	1244.1	32.8
18	a'	1906.7	78.0
19	a'	1992.7	274.4
20	a'	2297.9	85.4
21	a'	3187.1	1.3

Table S18: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **14** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	b_1	36.6	1.0
2	b_2	107.8	0.2
3	b_1	163.8	2.4
4	b_2	221.9	6.2
5	b_2	311.0	42.7
6	b_1	312.2	7.5
7	a_1	339.0	5.0
8	a_1	438.3	28.1
9	b_2	451.3	0.2
10	b_1	481.9	0.3
11	b_2	542.3	59.3
12	a_1	555.6	11.3
13	b_1	598.6	39.6
14	b_2	607.2	18.9
15	b_1	660.2	32.2
16	b_2	887.0	110.2
17	a_1	1239.8	0.4
18	a_1	1901.8	6.7
19	a_1	2121.3	24.3
20	a_1	3424.1	97.1
21	a_1	3462.5	94.1

Table S19: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **15** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a'	89.4	0.2
2	a''	89.8	0.1
3	a''	161.5	3.0
4	a'	191.6	0.7
5	a'	294.9	25.7
6	a''	391.0	1.4
7	a'	403.3	1.5
8	a''	526.1	0.3
9	a'	552.3	9.3
10	a'	594.2	51.0
11	a''	600.8	45.7
12	a'	685.8	15.7
13	a''	730.3	35.1
14	a'	838.4	30.1
15	a'	928.8	9.6
16	a'	1058.1	62.7
17	a'	1250.7	27.8
18	a'	1511.8	195.7
19	a'	2143.0	23.8
20	a'	3232.3	0.3
21	a'	3458.7	92.6

Table S20: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **16** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a''	117.1	0.6
2	a'	221.3	21.0
3	a''	288.1	0.1
4	a'	351.8	15.5
5	a''	378.0	0.8
6	a'	452.2	73.6
7	a'	481.3	54.6
8	a''	506.7	8.6
9	a'	580.7	24.4
10	a'	647.2	62.0
11	a'	742.9	7.5
12	a''	772.6	25.4
13	a''	970.3	0.7
14	a'	1051.6	15.5
15	a'	1159.6	17.8
16	a'	1327.2	12.9
17	a'	1382.4	0.8
18	a'	1442.6	28.4
19	a'	1846.6	201.1
20	a'	3134.3	9.2
21	a'	3156.5	26.6

Table S21: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **17** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a''	163.9	5.6
2	a''	215.8	3.2
3	a'	225.1	2.1
4	a'	299.5	28.7
5	a''	332.8	2.7
6	a'	406.1	23.2
7	a''	452.6	13.4
8	a'	479.9	10.1
9	a''	550.4	0.7
10	a'	562.0	25.5
11	a'	675.6	43.1
12	a''	684.6	15.4
13	a'	802.5	9.5
14	a'	825.9	17.2
15	a'	875.5	42.8
16	a'	1104.3	44.4
17	a'	1253.1	78.5
18	a'	1894.8	462.8
19	a'	1940.5	77.5
20	a'	2326.5	58.1
21	a'	3164.1	3.6

Table S22: Harmonic vibrational frequencies (cm^{-1}) and IR intensities (km mol^{-1}) of isomer **18** of $Si_2C_5H_2$ calculated at the fc-CCSD(T)/cc-pVTZ level of theory.

Mode	symmetry	frequency	intensity
1	a'	47.5	0.5
2	a''	72.6	1.4
3	a'	138.7	2.3
4	a''	175.9	5.1
5	a'	311.8	4.0
6	a''	340.0	14.9
7	a'	456.0	1.3
8	a''	497.6	2.7
9	a'	623.0	3.9
10	a'	705.2	7.6
11	a''	706.4	4.9
12	a'	751.1	41.1
13	a''	853.2	33.6
14	a'	988.7	22.5
15	a'	1133.7	84.0
16	a'	1221.5	20.4
17	a'	1397.3	30.5
18	a'	1558.1	186.5
19	a'	1960.5	412.5
20	a'	3141.6	8.2
21	a'	3184.4	8.3

Table S23: A-reduced Centrifugal Distortion Parameters ($\perp$ representation; in MHz) of $Si_2C_5H_2$ Isomers Calculated at the fc-CCSD(T)/cc-pVTZ Level of Theory.

Isomer	Δ_J	Δ_K	Δ_{JK}	δ_J	δ_K
1	0.3137 $\times10^{-3}$	0.8557 $\times10^{-3}$	-0.8079 $\times10^{-3}$	0.1375 $\times10^{-3}$	0.6925 $\times10^{-4}$
1[a]	0.3175 $\times10^{-3}$	0.8546 $\times10^{-3}$	-0.8098 $\times10^{-3}$	0.1391 $\times10^{-3}$	0.7092 $\times10^{-4}$
2	0.7591 $\times10^{-4}$	0.1109 $\times10^{-1}$	-0.4504 $\times10^{-3}$	0.1204 $\times10^{-4}$	0.4233 $\times10^{-3}$
3	0.1489 $\times10^{-3}$	0.1275 $\times10^{-1}$	-0.1834 $\times10^{-2}$	0.4431 $\times10^{-4}$	0.4224 $\times10^{-3}$
4	0.3831 $\times10^{-4}$	0.1584 $\times10^{-2}$	0.2652 $\times10^{-3}$	0.8572 $\times10^{-5}$	0.2391 $\times10^{-3}$
5	0.4867 $\times10^{-4}$	0.1391 $\times10^{-2}$	0.1694 $\times10^{-3}$	0.1174 $\times10^{-4}$	0.2375 $\times10^{-3}$
6	0.1289 $\times10^{-3}$	0.4401 $\times10^{-3}$	0.3848 $\times10^{-3}$	0.4368 $\times10^{-4}$	0.4021 $\times10^{-3}$
7	0.4320 $\times10^{-4}$	0.2064 $\times10^{-2}$	0.3080 $\times10^{-3}$	0.1045 $\times10^{-4}$	0.2881 $\times10^{-3}$
8	0.8094 $\times10^{-4}$	0.1356 $\times10^{-2}$	0.1131 $\times10^{-4}$	0.2763 $\times10^{-4}$	0.1999 $\times10^{-3}$
9	0.1109 $\times10^{-4}$	0.2980 $\times10^{-1}$	0.1201 $\times10^{-2}$	0.1072 $\times10^{-5}$	0.6325 $\times10^{-3}$
10	0.9392 $\times10^{-5}$	0.1219 $\times10^{-2}$	0.2343 $\times10^{-2}$	0.1281 $\times10^{-5}$	0.1213 $\times10^{-2}$
11	0.2397 $\times10^{-4}$	0.1117 $\times10^{-2}$	0.2475 $\times10^{-2}$	0.3606 $\times10^{-5}$	0.1275 $\times10^{-2}$
12	0.8925 $\times10^{-4}$	0.2015 $\times10^{-2}$	-0.8771 $\times10^{-4}$	0.3094 $\times10^{-4}$	0.1963 $\times10^{-3}$
13	0.7496 $\times10^{-4}$	0.1427 $\times10^{-2}$	0.1888 $\times10^{-3}$	0.2463 $\times10^{-4}$	0.3245 $\times10^{-3}$
14	0.3271 $\times10^{-4}$	0.2534 $\times10^{-2}$	0.1007 $\times10^{-2}$	0.7211 $\times10^{-5}$	0.6182 $\times10^{-3}$
15	0.1350 $\times10^{-3}$	0.8027 $\times10^{-2}$	-0.7320 $\times10^{-3}$	0.5031 $\times10^{-4}$	0.4000 $\times10^{-3}$
16	0.5725 $\times10^{-4}$	0.2745 $\times10^{-2}$	0.3126 $\times10^{-3}$	0.1407 $\times10^{-4}$	0.2489 $\times10^{-3}$
17	0.9314 $\times10^{-4}$	0.9847 $\times10^{-3}$	0.4422 $\times10^{-3}$	0.3186 $\times10^{-4}$	0.4942 $\times10^{-3}$
18	0.1886 $\times10^{-3}$	0.7346 $\times10^{-1}$	-0.5815 $\times10^{-2}$	0.4059 $\times10^{-4}$	0.1030 $\times10^{-2}$

[a] Calculated at the ae-CCSD(T)/cc-pwCVTZ level of theory.

Table S24: S-reduced Centrifugal Distortion Parameters ($\perp$ representation; in MHz) of $Si_2C_5H_2$ Isomers Calculated at the fc-CCSD(T)/cc-pVTZ Level of Theory.

Isomer	D_J	D_K	D_{JK}	D_1	D_2
1	0.2976 $\times10^{-3}$	0.7752 $\times10^{-3}$	-0.7112 $\times10^{-3}$	-0.1375 $\times10^{-3}$	-0.8054 $\times10^{-5}$
1[a]	0.3008 $\times10^{-3}$	0.7713 $\times10^{-3}$	-0.7098 $\times10^{-3}$	-0.1391 $\times10^{-3}$	-0.8331 $\times10^{-5}$
2	0.7412 $\times10^{-4}$	0.1109 $\times10^{-1}$	-0.4397 $\times10^{-3}$	-0.1204 $\times10^{-4}$	-0.8940 $\times10^{-6}$
3	0.1437 $\times10^{-3}$	0.1272 $\times10^{-1}$	-0.1803 $\times10^{-2}$	-0.4431 $\times10^{-4}$	-0.2598 $\times10^{-5}$
4	0.3430 $\times10^{-4}$	0.1564 $\times10^{-2}$	0.2893 $\times10^{-3}$	-0.8572 $\times10^{-5}$	-0.2006 $\times10^{-5}$
5	0.4429 $\times10^{-4}$	0.1370 $\times10^{-2}$	0.1957 $\times10^{-3}$	-0.1174 $\times10^{-4}$	-0.2190 $\times10^{-5}$
6	0.9786 $\times10^{-4}$	0.2850 $\times10^{-3}$	0.5709 $\times10^{-3}$	-0.4368 $\times10^{-4}$	-0.1552 $\times10^{-4}$
7	0.3781 $\times10^{-4}$	0.2038 $\times10^{-2}$	0.3403 $\times10^{-3}$	-0.1045 $\times10^{-4}$	-0.2694 $\times10^{-5}$
8	0.7011 $\times10^{-4}$	0.1302 $\times10^{-2}$	0.7628 $\times10^{-4}$	-0.2763 $\times10^{-4}$	-0.5414 $\times10^{-5}$
9	0.9438 $\times10^{-5}$	0.2980 $\times10^{-1}$	0.1211 $\times10^{-2}$	-0.1072 $\times10^{-5}$	-0.8261 $\times10^{-6}$
10	0.3860 $\times10^{-5}$	0.1191 $\times10^{-2}$	0.2376 $\times10^{-2}$	-0.1281 $\times10^{-5}$	-0.2766 $\times10^{-5}$
11	0.1328 $\times10^{-4}$	0.1063 $\times10^{-2}$	0.2540 $\times10^{-2}$	-0.3606 $\times10^{-5}$	-0.5347 $\times10^{-5}$
12	0.7933 $\times10^{-4}$	0.1965 $\times10^{-2}$	-0.2819 $\times10^{-4}$	-0.3094 $\times10^{-4}$	-0.4960 $\times10^{-5}$
13	0.6232 $\times10^{-4}$	0.1364 $\times10^{-2}$	0.2647 $\times10^{-3}$	-0.2463 $\times10^{-4}$	-0.6321 $\times10^{-5}$
14	0.2404 $\times10^{-4}$	0.2491 $\times10^{-2}$	0.1059 $\times10^{-2}$	-0.7211 $\times10^{-5}$	-0.4335 $\times10^{-5}$
15	0.1235 $\times10^{-3}$	0.7969 $\times10^{-2}$	-0.6628 $\times10^{-3}$	-0.5031 $\times10^{-4}$	-0.5759 $\times10^{-5}$
16	0.4993 $\times10^{-4}$	0.2709 $\times10^{-2}$	0.3566 $\times10^{-3}$	-0.1407 $\times10^{-4}$	-0.3662 $\times10^{-5}$
17	0.7093 $\times10^{-4}$	0.8736 $\times10^{-3}$	0.5755 $\times10^{-3}$	-0.3186 $\times10^{-4}$	-0.1111 $\times10^{-4}$
18	0.1849 $\times10^{-3}$	0.7344 $\times10^{-1}$	-0.5793 $\times10^{-2}$	-0.4059 $\times10^{-4}$	-0.1849 $\times10^{-5}$

[a] Calculated at the ae-CCSD(T)/cc-pwCVTZ level of theory.

Table S25: Optimized geometries of the singlet and triplet ground electronic states of isomer **1** (C_{2v}) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A_1$; fc-CCSD(T)/cc-pVTZ				$\widetilde{X}^1A_1$; ae-CCSD(T)/cc-pwCVTZ			
C	0.000000	0.000000	0.343912	C	0.000000	0.000000	0.345282
C	0.000000	-1.165613	-0.565033	C	0.000000	-1.161309	-0.560329
C	0.000000	1.165613	-0.565033	C	0.000000	1.161309	-0.560329
Si	0.000000	-1.774151	1.087720	Si	0.000000	-1.760440	1.081244
Si	0.000000	1.774151	1.087720	Si	0.000000	1.760440	1.081244
C	0.000000	-0.693784	-1.908991	C	0.000000	-0.691618	-1.900122
C	0.000000	0.693784	-1.908991	C	0.000000	0.691618	-1.900122
H	0.000000	-1.329561	-2.784451	H	0.000000	-1.326630	-2.774469
H	0.000000	1.329561	-2.784451	H	0.000000	1.326630	-2.774469
$\widetilde{X}^1A_1$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A_2$; UB3LYP/6-311++G(2d,2p)			
C	-0.000000	0.000000	0.392572	C	-0.000000	-0.000000	0.439321
C	0.000000	1.156669	-0.511157	C	0.000000	1.199222	-0.552584
C	-0.000000	-1.156669	-0.511157	C	-0.000000	-1.199222	-0.552584
Si	0.000000	1.785540	1.122541	Si	0.000000	1.767620	1.115511
Si	-0.000000	-1.785540	1.122541	Si	-0.000000	-1.767620	1.115511
C	0.000000	0.690661	-1.850277	C	0.000000	0.716137	-1.829525
C	-0.000000	-0.690661	-1.850277	C	-0.000000	-0.716137	-1.829525
H	0.000000	1.324601	-2.724685	H	0.000000	1.326047	-2.720355
H	-0.000000	-1.324601	-2.724685	H	-0.000000	-1.326047	-2.720355

Table S26: Optimized geometries of the singlet and triplet ground electronic states of isomer **2** (C_s) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
Si	0.691439	1.158966	0.000000
C	-0.316895	-0.487673	0.000000
C	1.030031	-0.785741	0.000000
Si	2.688790	-0.246784	0.000000
C	-1.563408	-1.011548	0.000000
H	-1.687093	-2.089800	0.000000
C	-2.739355	-0.207591	0.000000
C	-3.759169	0.454013	0.000000
H	-4.646218	1.040465	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
Si	-0.059076	-1.407903	-0.000000	Si	0.064264	-1.523339	0.000000
C	0.000000	0.522200	0.000000	C	0.096033	0.294258	-0.000000
C	-1.298502	0.111115	-0.000000	C	-1.229855	0.029821	-0.000000
Si	-2.507932	-1.126585	-0.000000	Si	-2.788754	-0.771086	0.000000
C	0.817467	1.591817	0.000000	C	0.851719	1.517148	-0.000000
H	0.376779	2.583437	0.000000	H	0.313015	2.457266	-0.000000
C	2.227129	1.517307	0.000000	C	2.226338	1.550117	-0.000000
C	3.432049	1.495888	0.000000	C	3.443428	1.585626	-0.000000
H	4.492480	1.469432	0.000000	H	4.504205	1.616897	-0.000000

Table S27: Optimized geometries of the singlet and triplet ground electronic states of isomer **3** (C_s) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
Si	1.451954	1.188180	0.000000
C	-0.357869	0.525696	0.000000
C	0.515780	-0.545230	0.000000
Si	2.147121	-1.156508	0.000000
C	-1.663620	0.879436	0.000000
H	-1.932956	1.928289	0.000000
C	-2.718514	-0.080498	0.000000
C	-3.632745	-0.881556	0.000000
H	-4.424743	-1.591158	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
Si	-0.026850	-0.000000	0.006411	Si	-1.954476	0.402339	0.000000
C	0.016549	-0.000000	1.931044	C	-0.159490	0.693224	-0.000000
C	1.293286	0.000000	1.449556	C	-0.239535	-0.656087	0.000000
Si	2.439634	0.000000	0.156481	Si	-0.807493	-2.313970	0.000000
C	-0.751818	-0.000000	3.037683	C	0.923664	1.641385	-0.000000
H	-1.829096	-0.000000	2.932200	H	0.682948	2.696442	-0.000000
C	-0.232708	-0.000000	4.352170	C	2.250090	1.278002	-0.000000
C	0.183957	-0.000000	5.482956	C	3.428098	0.968624	-0.000000
H	0.557705	-0.000000	6.475708	H	4.452634	0.692189	-0.000000

Table S28: Optimized geometries of the singlet and triplet ground electronic states of isomer **4** (C_s) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
C	-0.743469	0.860211	0.000000
C	-0.947782	-0.640011	0.000000
C	0.128042	-1.548469	0.000000
C	1.458712	-1.178618	0.000000
Si	2.252163	0.567258	0.000000
C	0.530523	1.069350	0.000000
Si	-2.508735	0.215107	0.000000
H	-0.131110	-2.604105	0.000000
H	2.180881	-1.997591	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A$; UB3LYP/6-311++G(2d,2p)			
C	0.019428	-0.000000	0.014500	C	-1.044434	0.125910	-0.002794
C	0.002567	-0.000000	1.512543	C	0.030767	1.065273	0.000601
C	1.184959	-0.000000	2.266316	C	1.395203	0.718997	0.002361
C	2.445403	0.000000	1.725020	C	1.828732	-0.591092	0.000989
Si	2.997594	0.000000	-0.111805	Si	0.575783	-1.922145	-0.002359
C	1.234286	0.000000	-0.385972	C	-1.028957	-1.217283	-0.005806
Si	-1.650346	-0.000000	0.880961	Si	-1.536357	1.946380	-0.000272
H	1.073693	-0.000000	3.346122	H	2.137320	1.513505	0.004682
H	3.275455	0.000000	2.429785	H	2.890797	-0.800291	0.002597

Table S29: Optimized geometries of the singlet and triplet ground electronic states of isomer **5** (C_s) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
C	-0.860033	0.772173	0.000000
C	-0.735059	-0.672713	0.000000
C	0.462664	-1.396431	0.000000
Si	1.958199	-0.413567	0.000000
C	1.652195	1.353212	0.000000
C	0.378683	1.335784	0.000000
Si	-2.479346	-0.058090	0.000000
H	0.455380	-2.479903	0.000000
H	3.313828	-1.001638	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A''$; UB3LYP/6-311++G(2d,2p)			
C	-0.126697	-0.000000	-0.153140	C	1.248201	0.134149	-0.000000
C	-0.039075	-0.000000	1.291923	C	-0.020774	0.979818	-0.000000
C	1.127923	0.000000	2.041438	C	-1.311774	0.541160	0.000000
Si	2.649640	0.000000	1.109155	Si	-1.580908	-1.245556	0.000000
C	2.371291	0.000000	-0.651895	C	-0.015277	-2.028247	0.000000
C	1.110299	0.000000	-0.687382	C	0.898019	-1.138073	0.000000
Si	-1.760133	-0.000000	0.632386	Si	1.557587	1.927849	-0.000000
H	1.095122	0.000000	3.122377	H	-2.113169	1.268351	0.000000
H	3.984742	0.000000	1.733008	H	-2.896195	-1.897613	0.000000

Table S30: Optimized geometries of the singlet and triplet ground electronic states of isomer **6** (C_s) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
Si	-0.143223	1.138521	0.000000
C	-0.540414	-0.720716	0.000000
C	0.480633	-1.729932	0.000000
C	1.757308	-1.229013	0.000000
C	1.766657	0.190478	0.000000
C	1.742885	1.472321	0.000000
Si	-2.196531	-0.107531	0.000000
H	0.279587	-2.797527	0.000000
H	2.671613	-1.808013	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)			
Si	-0.029738	0.000000	-0.015815
C	0.001581	0.000000	1.873199
C	1.203572	-0.000000	2.636611
C	2.364748	-0.000000	1.910583
C	2.132593	-0.000000	0.527065
C	1.745248	-0.000000	-0.671303
Si	-1.745677	0.000000	1.691907
H	1.226369	-0.000000	3.722199
H	3.360837	-0.000000	2.329278

$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
Si	-0.865779	-0.937859	-0.000000
C	-0.145619	0.746296	-0.000000
C	1.273456	0.827809	0.000000
C	2.116055	-0.273518	0.000000
C	1.417809	-1.469423	0.000000
C	0.413857	-2.214088	0.000000
Si	-1.458239	2.037891	-0.000000
H	1.727383	1.814993	0.000000
H	3.191384	-0.198139	0.000000

Table S31: Optimized geometries of the singlet and triplet ground electronic states of isomer **7** (C_s) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
C	-0.961235	0.825952	0.000000
C	-0.733936	-0.656618	0.000000
C	0.489795	-1.296881	0.000000
Si	2.153160	-0.525772	0.000000
C	1.542564	1.347582	0.000000
C	0.242772	1.367906	0.000000
Si	-2.499287	-0.146876	0.000000
H	0.442506	-2.387270	0.000000
H	2.260405	2.152455	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)			
C	0.018410	-0.000000	0.020649
C	-0.000994	-0.000000	1.503191
C	1.103972	0.000000	2.319565
Si	2.854978	0.000000	1.803193
C	2.545179	0.000000	-0.148973
C	1.275219	0.000000	-0.357251
Si	-1.656142	-0.000000	0.721751
H	0.904831	0.000000	3.390338
H	3.385188	0.000000	-0.821303

$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
C	-1.136573	0.172843	0.000000
C	0.056091	0.961679	0.000000
C	1.380639	0.609942	0.000000
Si	1.603420	-1.178782	0.000000
C	-0.026750	-2.019017	0.000000
C	-1.109646	-1.207120	0.000000
Si	-1.551909	1.919003	0.000000
H	2.195288	1.318854	0.000000
H	-0.117264	-3.099017	0.000000

Table S32: Optimized geometries of the singlet and triplet ground electronic states of isomer **8** (C_s) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
C	-0.765967	0.755058	0.000000
C	-0.525719	-0.638089	0.000000
Si	1.180065	-1.203662	0.000000
C	2.213735	0.220029	0.000000
C	1.428519	1.224128	0.000000
C	0.183697	1.807333	0.000000
Si	-2.324571	-0.250427	0.000000
H	1.643110	-2.603311	0.000000
H	-0.047018	2.860699	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A''$; UB3LYP/6-311++G(2d,2p)			
C	0.381099	-0.000000	0.022527	C	-1.095052	-0.101113	0.000000
C	0.107881	-0.000000	1.396890	C	0.037695	0.849076	0.000000
Si	1.498868	0.000000	2.529710	Si	1.684249	0.188012	0.000000
C	2.953176	0.000000	1.559466	C	1.502760	-1.572993	0.000000
C	2.591216	0.000000	0.352012	C	0.286466	-1.849980	0.000000
C	1.636010	0.000000	-0.623918	C	-1.048305	-1.482542	0.000000
Si	-1.430156	-0.000000	0.400186	Si	-1.591493	1.658029	0.000000
H	1.441939	0.000000	3.998734	H	2.932672	0.955978	0.000000
H	1.789160	0.000000	-1.690116	H	-1.908019	-2.131106	0.000000

Table S33: Optimized geometries of the singlet and triplet ground electronic states of isomer **9** (C_s) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
Si	1.169064	1.235648	0.000000
C	-0.301725	-0.056748	0.000000
C	0.909490	-0.713240	0.000000
Si	2.655794	-0.693076	0.000000
C	-1.617907	-0.136811	0.000000
C	-2.888372	-0.143724	0.000000
C	-4.216949	-0.181335	0.000000
H	-4.747452	-1.126390	0.000000
H	-4.800065	0.732258	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
Si	-0.844952	-1.546098	-0.000000	Si	-0.969417	-1.577662	0.000000
C	0.000000	0.237295	0.000000	C	-0.223463	0.093153	0.000000
C	0.935139	-0.748927	0.000000	C	0.839337	-0.742609	0.000000
Si	1.452705	-2.403518	-0.000000	Si	1.811660	-2.204097	0.000000
C	-0.308492	1.507195	0.000000	C	-0.444082	1.466502	0.000000
C	-0.655119	2.719757	0.000000	C	-0.687467	2.667543	0.000000
C	-1.004785	3.989690	0.000000	C	-0.949298	4.008802	0.000000
H	-0.262807	4.778201	0.000000	H	-0.145902	4.731940	0.000000
H	-2.046195	4.286367	0.000000	H	-1.965874	4.376389	0.000000

Table S34: Optimized geometries of the singlet and triplet ground electronic states of isomer **10** (C_{2v}) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
C	0.000000	0.000000	0.778391
C	0.000000	0.000000	-0.619049
Si	0.000000	-1.284841	2.044500
Si	0.000000	1.284841	2.044500
H	0.000000	0.000000	3.131377
C	0.000000	0.000000	-1.844027
C	0.000000	0.000000	-3.216068
C	0.000000	0.000000	-4.433679
H	0.000000	0.000000	-5.497310

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
C	-0.026977	0.000000	1.000000	C	0.042226	0.000000	1.000000
C	1.350532	-0.000000	1.000000	C	1.338449	-0.000000	1.000000
Si	-1.289967	0.000000	2.283106	Si	-1.352000	0.000000	2.246173
Si	-1.289967	-0.000000	-0.283106	Si	-1.352000	-0.000000	-0.246173
H	-2.372387	0.000000	1.000000	H	-2.393715	0.000000	1.000000
C	2.570216	-0.000000	1.000000	C	2.611915	-0.000000	1.000000
C	3.926495	-0.000000	1.000000	C	3.926479	-0.000000	1.000000
C	5.135052	-0.000000	1.000000	C	5.158252	-0.000000	1.000000
H	6.195738	-0.000000	1.000000	H	6.219129	-0.000000	1.000000

Table S35: Optimized geometries of the singlet and triplet ground electronic states of isomer **11** (C_{2v}) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
C	0.000000	0.000000	-0.223235
C	0.000000	0.000000	-1.470293
C	0.000000	0.000000	-2.835014
C	0.000000	0.000000	-4.054860
Si	0.000000	-1.317636	1.325584
Si	0.000000	1.317636	1.325584
C	0.000000	0.000000	2.529340
H	0.000000	0.000000	3.608102
H	0.000000	0.000000	-5.119075

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
C	-0.006949	-0.000000	1.000000	C	-0.094000	-0.000000	1.000000
C	1.231265	-0.000000	1.000000	C	1.210891	-0.000000	1.000000
C	2.579903	-0.000000	1.000000	C	2.521208	-0.000000	1.000000
C	3.790849	-0.000000	1.000000	C	3.756194	-0.000000	1.000000
Si	-1.545559	0.000000	2.310676	Si	-1.466786	0.000000	2.307506
Si	-1.545559	-0.000000	-0.310676	Si	-1.466786	-0.000000	-0.307506
C	-2.741106	0.000000	1.000000	C	-2.701439	0.000000	1.000000
H	-3.817127	0.000000	1.000000	H	-3.778469	0.000000	1.000000
H	4.851935	-0.000000	1.000000	H	4.816839	-0.000000	1.000000

Table S36: Optimized geometries of the singlet and triplet ground electronic states of isomer **12** (C_{2v}) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
C	-0.537753	0.578102	0.000000
C	-0.804350	-0.804659	0.000000
C	0.209080	-1.784215	0.000000
C	1.441659	-1.187514	0.000000
C	2.303213	-0.234157	0.000000
Si	1.106532	1.358773	0.000000
Si	-2.350048	0.217578	0.000000
H	0.063968	-2.854406	0.000000
H	3.356845	-0.035139	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
C	-0.071837	0.000000	0.043245	C	-0.057498	0.000000	-0.073040
C	-0.002149	0.000000	1.436443	C	0.102114	0.000000	1.318558
C	1.202782	-0.000000	2.160810	C	1.208293	-0.000000	2.163325
C	2.254022	-0.000000	1.301511	C	2.427742	-0.000000	1.521086
C	2.860636	-0.000000	0.181776	C	2.795889	-0.000000	0.197241
Si	1.341100	-0.000000	-1.105051	Si	1.479602	-0.000000	-1.026571
Si	-1.742811	0.000000	0.814644	Si	-1.677468	0.000000	0.730951
H	1.299031	-0.000000	3.235407	H	1.115306	-0.000000	3.239567
H	3.837904	-0.000000	-0.254005	H	3.845748	-0.000000	-0.074038

Table S37: Optimized geometries of the singlet and triplet ground electronic states of isomer **13** (C_s) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
Si	-2.299516	0.167156	0.000000
C	0.145859	-1.515898	0.000000
C	1.509642	-1.299187	0.000000
Si	1.919374	0.438863	0.000000
C	0.517818	1.526533	0.000000
C	-0.688624	1.168867	0.000000
C	-1.066128	-1.193528	0.000000
H	2.250482	-2.089879	0.000000
H	3.318355	0.903176	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)			
Si	-0.686703	0.000000	-0.870736
C	0.099128	0.000000	1.981935
C	1.272966	-0.000000	2.681540
Si	2.702508	-0.000000	1.616117
C	2.331932	-0.000000	-0.106467
C	1.188594	-0.000000	-0.599768
C	-0.603619	0.000000	0.957498
H	1.343841	-0.000000	3.761203
H	4.066658	-0.000000	2.164209

$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
Si	0.103836	-0.000000	0.191295
C	-0.032617	-0.000000	2.923810
C	0.956720	-0.000000	3.903619
Si	2.602597	0.000000	3.295186
C	2.729301	0.000000	1.524157
C	1.856541	0.000000	0.647411
C	-0.624066	-0.000000	1.842844
H	0.736441	-0.000000	4.961719
H	3.766103	0.000000	4.187351

Table S38: Optimized geometries of the singlet and triplet ground electronic states of isomer **14** (C_{2v}) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
C	0.000000	0.000000	-0.862419
C	0.000000	0.000000	-2.270101
C	0.000000	0.000000	-3.488455
H	0.000000	0.000000	-4.551348
Si	0.000000	-1.329243	0.347438
Si	0.000000	1.329243	0.347438
C	0.000000	0.000000	1.896780
C	0.000000	0.000000	3.133634
H	0.000000	0.000000	4.200338

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
Si	1.871386	0.796348	1.001197	Si	0.395394	-1.412866	-0.089340
Si	2.042813	-1.844166	0.997939	Si	0.488885	1.211377	-0.082961
C	3.162856	-0.445633	0.999936	C	-0.826672	-0.063061	0.008202
C	4.550271	-0.355290	1.000081	C	-2.202825	0.005355	0.011830
C	5.756896	-0.276886	1.000171	C	-3.413553	0.067256	0.025358
H	6.814865	-0.208009	1.000336	H	-4.472787	0.120336	0.036783
C	-0.806270	-0.703587	1.000109	C	3.096492	-0.229912	0.029142
C	0.414640	-0.624216	0.999795	C	1.794345	-0.158111	0.007026
H	-1.867188	-0.772531	1.000442	H	3.930423	0.459598	0.055535

Table S39: Optimized geometries of the singlet and triplet ground electronic states of isomer **15** (C_s) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
Si	0.141314	1.662813	0.000000
C	-0.969719	0.283997	0.000000
C	0.271573	-0.369496	0.000000
C	-2.264282	-0.276001	0.000000
C	-3.388080	-0.740726	0.000000
H	-4.369058	-1.151207	0.000000
C	0.983910	-1.564024	0.000000
H	0.594006	-2.570637	0.000000
Si	2.296544	-0.385118	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
Si	-0.029022	0.000000	0.024034	Si	0.144050	0.000000	0.093227
C	0.011744	0.000000	1.788884	C	0.142924	0.000000	2.031841
C	1.375243	-0.000000	1.498810	C	1.494095	-0.000000	1.939607
C	-0.647526	0.000000	3.020268	C	-0.736227	0.000000	3.085344
C	-1.233849	0.000000	4.074056	C	-1.566121	0.000000	3.972105
H	-1.750123	0.000000	5.000387	H	-2.284795	0.000000	4.752898
C	2.669503	-0.000000	1.976594	C	2.810182	-0.000000	1.932754
H	3.004106	-0.000000	3.001263	H	3.548252	-0.000000	2.721260
Si	2.985876	-0.000000	0.252192	Si	2.621461	-0.000000	0.039203

Table S40: Optimized geometries of the singlet and triplet ground electronic states of isomer **16** (C_s) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
C	-0.664105	0.872165	0.000000
Si	-0.318731	-1.073408	0.000000
C	1.519565	-1.163280	0.000000
C	2.377375	-0.058519	0.000000
C	1.849571	1.268628	0.000000
C	0.577637	1.347097	0.000000
Si	-2.304444	0.187692	0.000000
H	1.976624	-2.150158	0.000000
H	3.448778	-0.244587	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
C	-0.145076	-0.000000	0.099404	C	-1.014204	-0.696996	-0.000000
Si	-0.110507	-0.000000	2.071770	Si	0.023532	0.954240	-0.000000
C	1.683990	0.000000	2.434448	C	1.781900	0.633637	0.000000
C	2.688750	0.000000	1.471483	C	2.264939	-0.704917	0.000000
C	2.376418	0.000000	0.083591	C	1.312319	-1.693389	0.000000
C	1.138088	0.000000	-0.179908	C	0.017242	-1.449398	0.000000
Si	-1.869927	-0.000000	0.495449	Si	-2.327320	0.648575	-0.000000
H	1.984929	0.000000	3.476619	H	2.487587	1.453217	0.000000
H	3.719600	0.000000	1.814365	H	3.326647	-0.918934	0.000000

Table S41: Optimized geometries of the singlet and triplet ground electronic states of isomer **17** (C_s) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
C	-0.485880	1.252938	0.000000
Si	-2.145665	0.406106	0.000000
C	-1.325857	-1.295400	0.000000
C	0.014322	-1.446154	0.000000
C	1.289202	-1.358074	0.000000
Si	1.992442	0.229631	0.000000
C	0.743502	1.502077	0.000000
H	-1.978803	-2.163113	0.000000
H	3.430678	0.525310	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
C	0.391344	0.000000	0.579925	C	-0.200052	-0.000000	-0.061983
Si	-0.455863	0.000000	2.231267	Si	0.079814	-0.000000	1.696724
C	1.196056	0.000000	3.148187	C	1.979853	0.000000	1.989246
C	2.368914	-0.000000	2.509345	C	2.665214	0.000000	0.863616
C	3.327753	-0.000000	1.683278	C	3.069375	0.000000	-0.346100
Si	2.993658	-0.000000	-0.010274	Si	2.051302	-0.000000	-1.722086
C	1.245290	-0.000000	-0.317955	C	0.361986	-0.000000	-1.169106
H	1.141214	0.000000	4.230320	H	2.372165	0.000000	2.993203
H	3.992876	-0.000000	-1.079321	H	2.441602	-0.000000	-3.130886

Table S42: Optimized geometries of the singlet and triplet ground electronic states of isomer **18** (C_s) of $Si_2C_5H_2$ in Cartesian coordinates (in Ångström units) at different levels.

$\widetilde{X}^1A'$; fc-CCSD(T)/cc-pVTZ			
C	-2.923384	0.649573	0.000000
Si	-2.504456	-1.133042	0.000000
C	-1.575362	0.465096	0.000000
C	-0.401696	1.308356	0.000000
C	0.828795	0.779693	0.000000
C	2.000416	0.261197	0.000000
Si	3.539687	-0.493412	0.000000
H	-3.575487	1.514871	0.000000
H	-0.500407	2.390756	0.000000

$\widetilde{X}^1A'$; B3LYP/6-311++G(2d,2p)				$\widetilde{X}^3A'$; UB3LYP/6-311++G(2d,2p)			
Si	-2.905346	0.000000	0.887836	Si	-2.683731	2.622144	-0.000000
C	-0.031880	0.000000	1.642476	C	-0.038995	1.107344	-0.000000
C	1.258862	-0.000000	1.965611	C	1.162605	0.454914	0.000000
C	2.308513	-0.000000	0.988390	C	1.293360	-0.968710	0.000000
C	3.660537	-0.000000	1.009445	C	2.324888	-1.845283	0.000000
Si	3.048902	-0.000000	-0.703089	Si	0.711854	-2.721082	0.000000
C	-1.264190	0.000000	1.337280	C	-1.118756	1.728501	-0.000000
H	1.507577	-0.000000	3.024392	H	2.071085	1.055287	0.000000
H	4.407601	-0.000000	1.792496	H	3.403168	-1.749738	0.000000

1 Various Other Isomers of $Si_2C_5H_2$

On the basis of the systematic approach explained in the main part of the article, we had obtained several geometries of $Si_2C_5H_2$. Here below, these other possibilities are given as a further information.

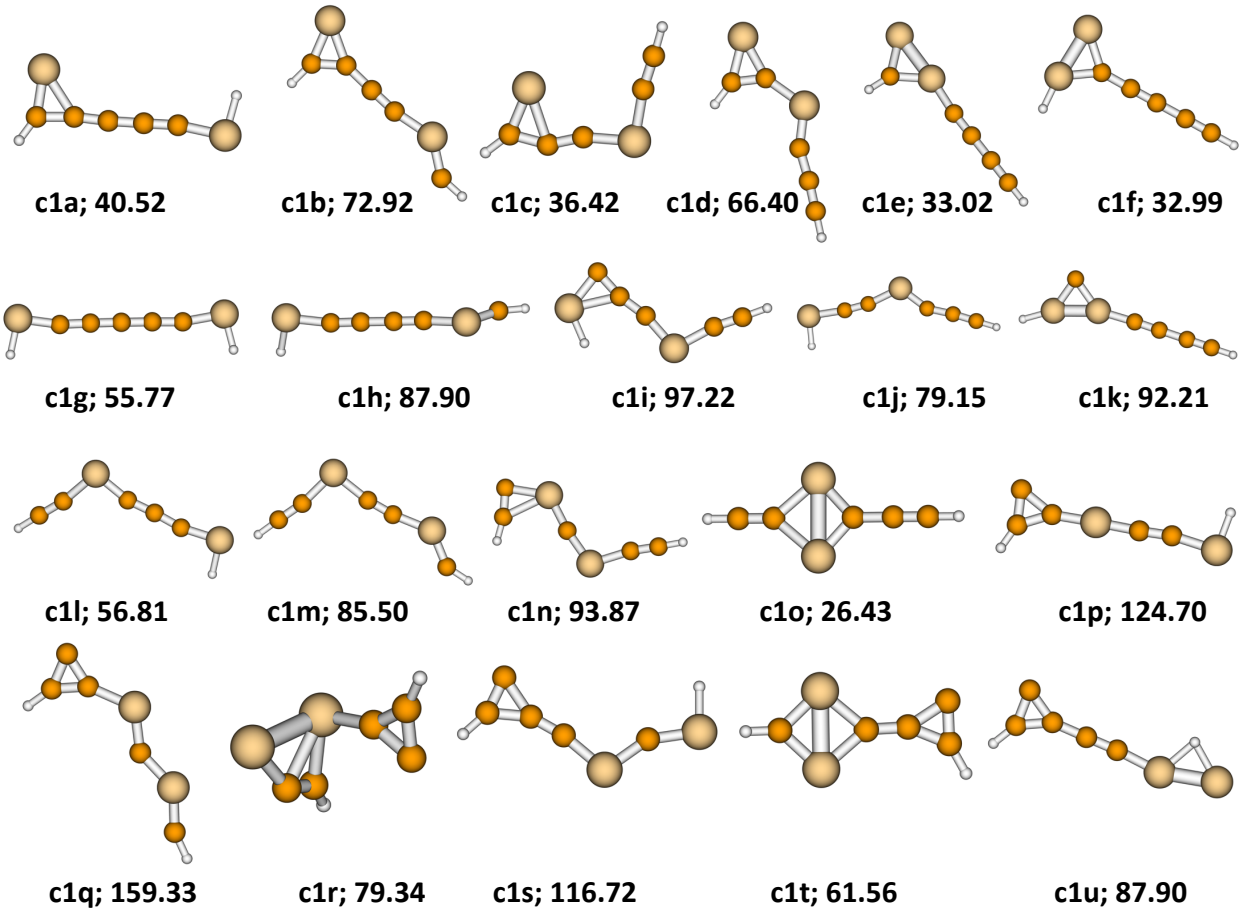

Figure S1: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, 1-(buta-1,3-diynyl)cyclopropenylidene.

Table S43: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S1 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c1a	-770.6052473	0.041162	-770.564086	40.52	0
c1b	-770.5562236	0.043777	-770.512446	72.92	1
c1c	-770.6144366	0.043819	-770.570617	36.42	0
c1d	-770.5662696	0.043438	-770.522832	66.40	1
c1e	-770.6211584	0.045127	-770.576031	33.02	0
c1f	-770.6190106	0.042928	-770.576083	32.99	0
c1g	-770.5773429	0.037567	-770.539776	55.77	0
c1h	-770.5281384	0.039559	-770.488580	87.90	0
c1i	-770.5121510	0.038420	-770.473731	97.22	1
c1j	-770.5414740	0.038962	-770.502513	79.15	1
c1k	-770.5230602	0.041348	-770.481712	92.21	0
c1l	-770.5778578	0.039736	-770.538122	56.81	0
c1m	-770.5339309	0.041526	-770.492405	85.50	1
c1n	-770.5200808	0.041016	-770.479065	93.87	0
14; c1o	-770.6303640	0.043823	-770.586541	26.43	0
c1p	-770.4685134	0.038588	-770.429925	124.70	1
c1q	-770.4153651	0.040622	-770.374743	159.33	2
c1r	-770.5454824	0.043270	-770.502213	79.34	0
c1s	-770.4806785	0.038031	-770.442648	116.72	0
c1t	-770.5741102	0.043563	-770.530547	61.56	0
c1u	-770.5293502	0.040779	-770.488571	87.90	0

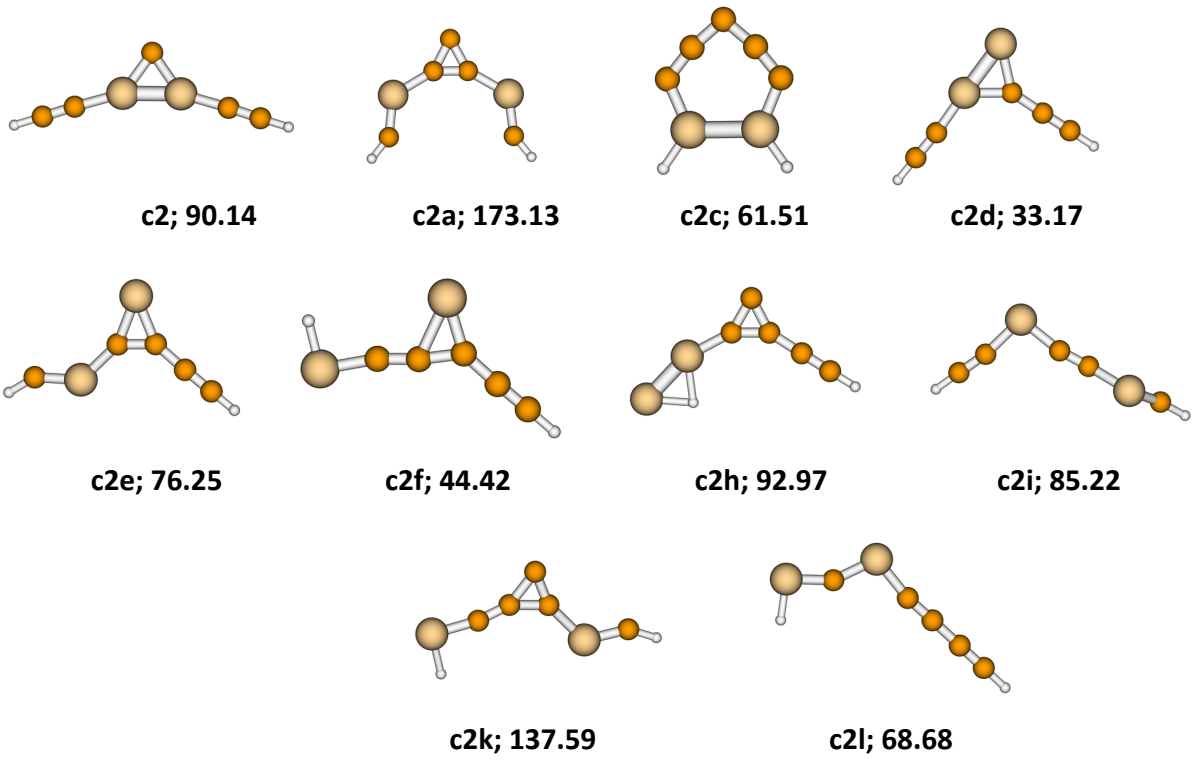

Figure S2: Various structural possibilities obtained for $Si_2C_5H_2$ using the base geometry, 1,2-diethynylcyclopropenylidene (C_7H_2)

Table S44: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S2 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c2	-770.5278997	0.042889	-770.485011	90.14	0
c2a	-770.3927341	0.039985	-770.352749	173.13	2
c2c	-770.5719557	0.041318	-770.530637	61.51	0
c2d	-770.6203285	0.044535	-770.575793	33.17	0
c2e	-770.5505321	0.043393	-770.507139	76.25	0
c2f	-770.5993434	0.041472	-770.557871	44.42	0
c2h	-770.5209474	0.040456	-770.480491	92.97	1
c2i	-770.5343586	0.041515	-770.492843	85.22	0
c2k	-770.4466510	0.037267	-770.409384	137.59	1
c2l	-770.5595397	0.040334	-770.519206	68.68	0

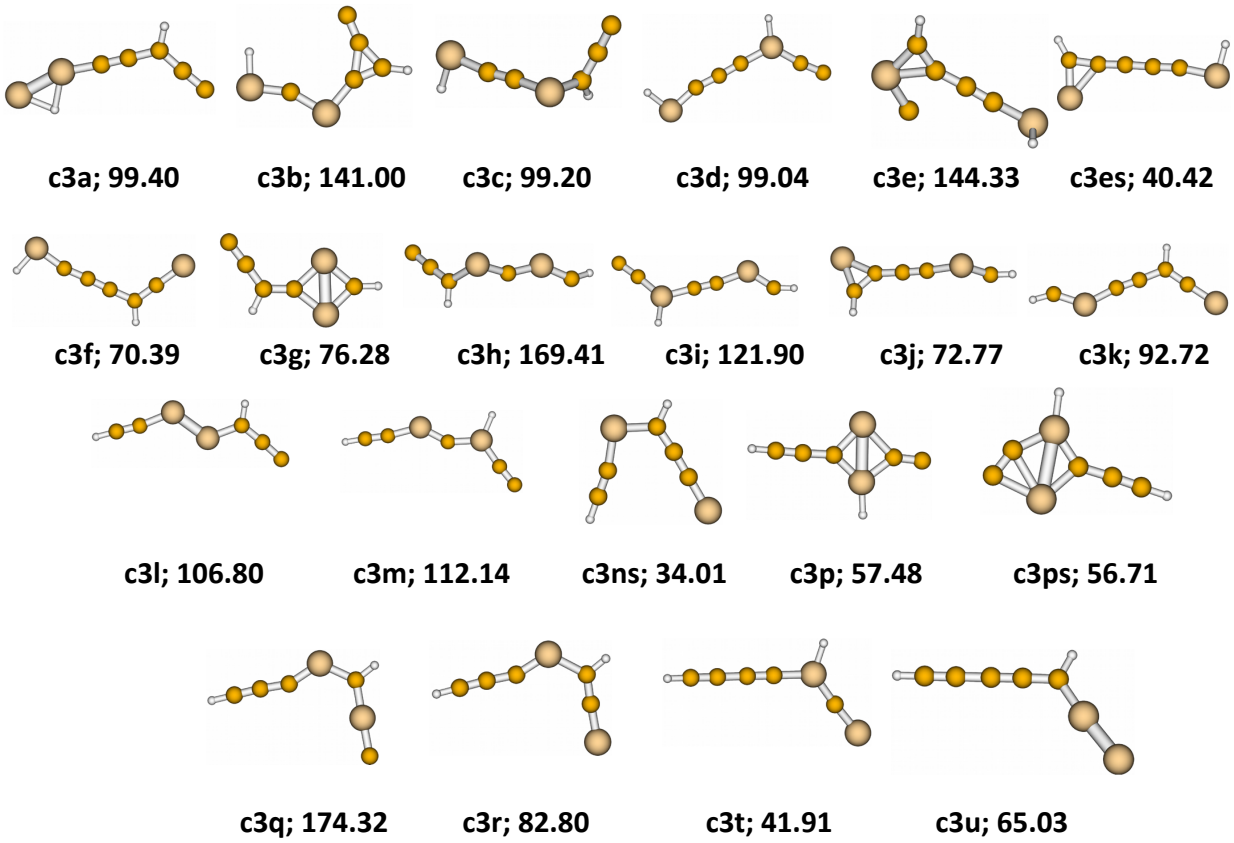

Figure S3: Various structural possibilities obtained for $Si_2C_5H_2$ using the base geometry, 1-(buta-1,3-diynyl)propadienylidene (C_7H_2)

Table S45: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S3 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c3a	-770.5107931	0.040537	-770.470256	99.40	0
c3b	-770.4417465	0.037792	-770.403955	141.00	0
c3c	-770.5090026	0.038426	-770.470576	99.20	0
c3d	-770.5073715	0.036547	-770.470824	99.04	0
c3e	-770.4368208	0.038176	-770.398645	144.33	1
c3es	-770.6052339	0.040992	-770.564242	40.42	0
c3f	-770.5574940	0.041020	-770.516474	70.39	0
c3g	-770.5513028	0.044204	-770.507098	76.28	0
c3h	-770.3984663	0.039778	-770.358688	169.41	0
c3i	-770.4729829	0.038583	-770.434400	121.90	1
c3j	-770.5564731	0.043792	-770.512682	72.77	0
c3k	-770.5245284	0.043631	-770.480898	92.72	0
c3l	-770.5005668	0.042102	-770.458465	106.80	1
c3m	-770.4892907	0.039343	-770.449948	112.14	0
c3ns	-770.6188970	0.044444	-770.574453	34.01	0
c3p	-770.5778815	0.040831	-770.537051	57.48	0
c3ps	-770.5790670	0.040788	-770.538279	56.71	1
c3q	-770.3912325	0.040381	-770.350852	174.32	1
c3r	-770.5396965	0.042986	-770.496710	82.80	1
c3t	-770.6045943	0.042720	-770.561874	41.91	0
c3u	-770.5701562	0.045129	-770.525027	65.03	0

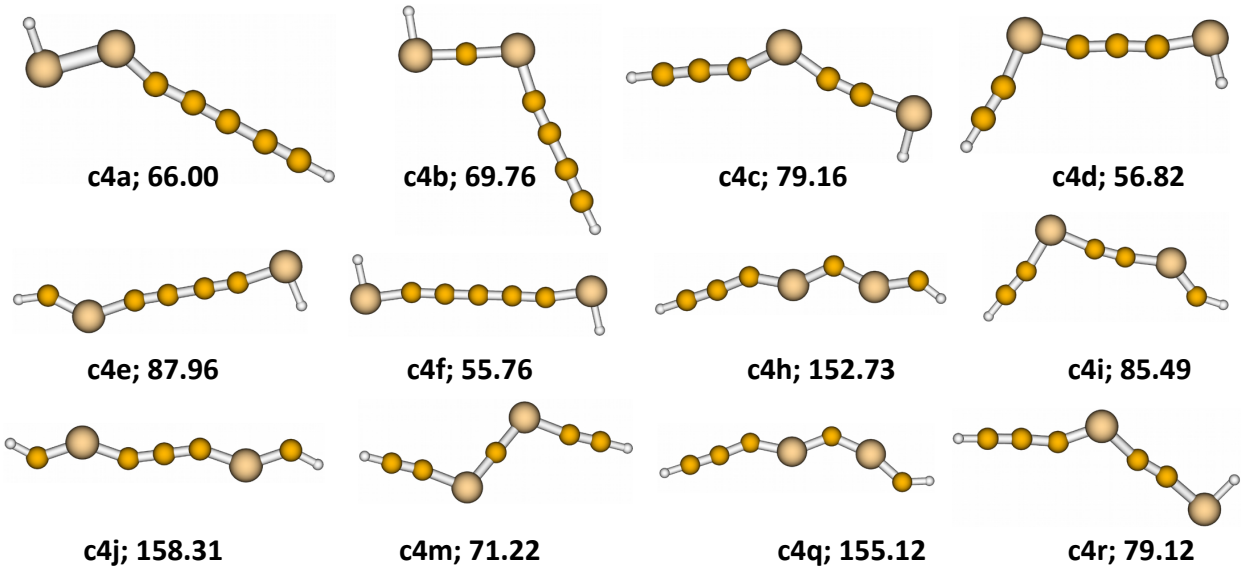

Figure S4: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, buta-1,3-diynylethynylcarbene.

Table S46: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S4 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c4a	-770.5640886	0.040620	-770.523469	66.00	1
c4b	-770.5576392	0.040158	-770.517481	69.76	0
c4c	-770.5414727	0.038970	-770.502503	79.16	1
c4d	-770.5778563	0.039750	-770.538107	56.82	0
c4e	-770.5279890	0.039504	-770.488485	87.96	1
c4f	-770.5773429	0.037553	-770.539790	55.76	0
c4h	-770.4250760	0.039819	-770.385257	152.73	0
c4i	-770.5339620	0.041550	-770.492412	85.49	1
c4j	-770.4153896	0.039021	-770.376368	158.31	0
c4m	-770.5574488	0.042297	-770.515152	71.22	0
c4q	-770.4209934	0.039546	-770.381448	155.12	0
c4r	-770.5414245	0.038947	-770.502477	79.18	1

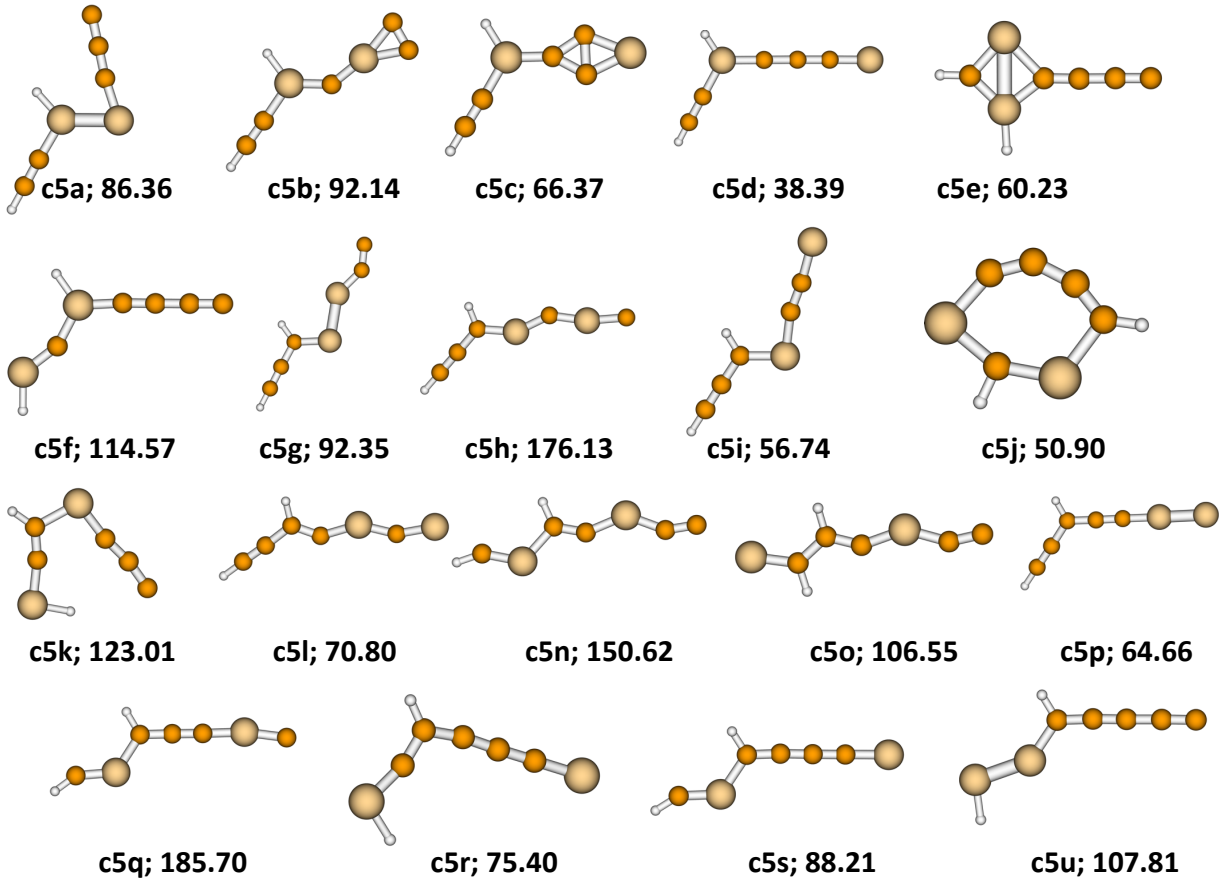

Figure S5: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, 1-(ethynyl)pentatetraenylidene.

Table S47: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S5 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c5a	-770.5314552	0.040420	-770.491035	86.36	0
c5b	-770.5218127	0.040000	-770.481813	92.14	0
c5c	-770.5640983	0.041211	-770.522888	66.37	0
c5d	-770.6101195	0.042637	-770.567482	38.39	0
c5e	-770.5745464	0.041873	-770.532673	60.23	0
c5f	-770.4833193	0.037239	-770.446080	114.57	0
c5g	-770.5237135	0.042234	-770.481479	92.35	0
c5h	-770.3884152	0.040447	-770.347968	176.13	1
c5i	-770.5820796	0.043843	-770.538236	56.74	0
c5j	-770.5920304	0.044491	-770.547540	50.90	0
c5k	-770.4704804	0.037862	-770.432619	123.01	1
c5l	-770.5601783	0.044351	-770.515827	70.80	0
c5n	-770.4287059	0.040073	-770.388633	150.62	0
c5o	-770.5019054	0.043043	-770.458862	106.55	0
c5p	-770.5710211	0.045409	-770.525612	64.66	1
c5q	-770.3730294	0.040308	-770.332721	185.70	1
c5r	-770.5489934	0.040500	-770.508493	75.40	1
c5s	-770.5314477	0.043360	-770.488088	88.21	0
c5u	-770.4963145	0.039469	-770.456845	107.81	0

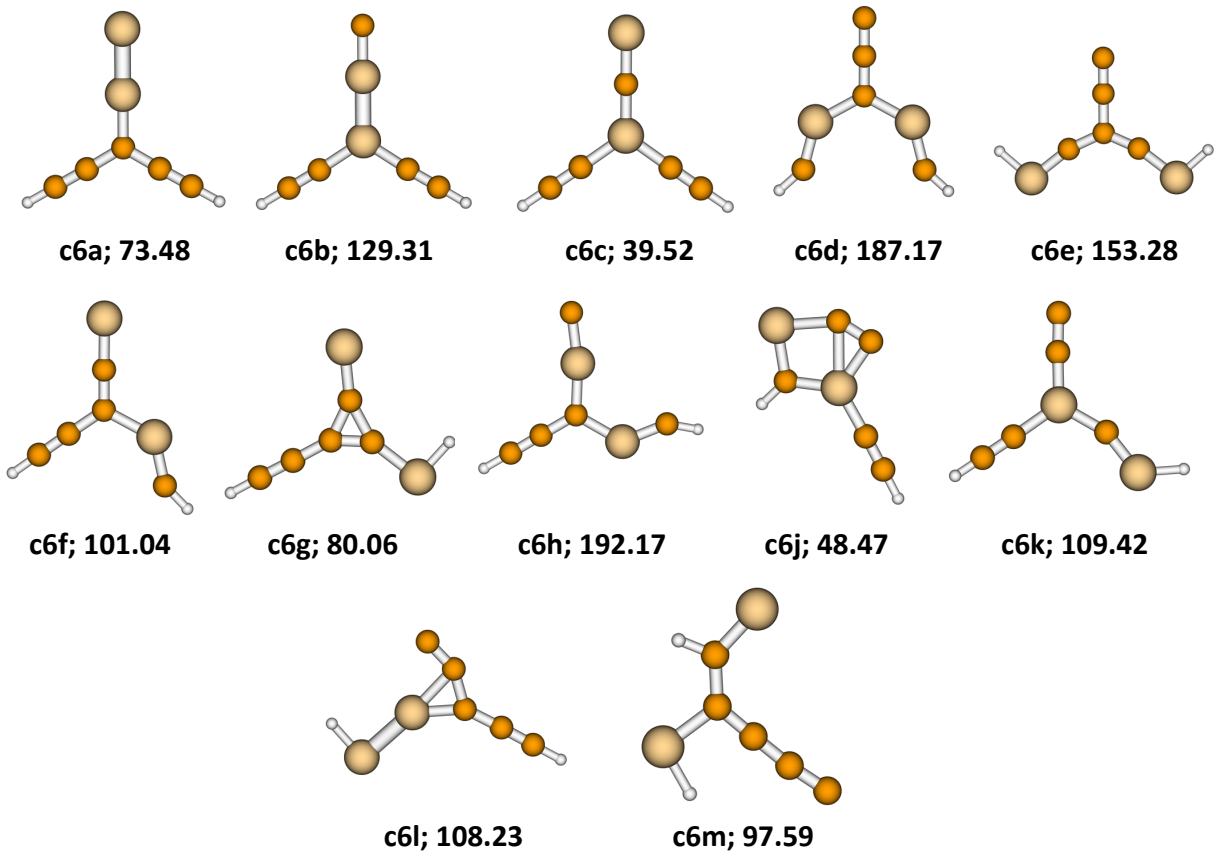

Figure S6: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, 1,1-(diethynyl)propadienylidene.

Table S48: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S6 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	$E+ZPVE$ a.u	$\Delta E+ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c6a	-770.5562017	0.044645	-770.511557	73.48	1
c6b	-770.4641999	0.041612	-770.422588	129.31	2
c6c	-770.6094398	0.043761	-770.565679	39.52	0
c6d	-770.3695251	0.039146	-770.330379	187.17	1
c6e	-770.4190760	0.034694	-770.384382	153.28	0
c6f	-770.5103313	0.042694	-770.467637	101.04	1
c6g	-770.5417496	0.040684	-770.501065	80.06	0
c6h	-770.3624469	0.040036	-770.322410	192.17	1
c6j	-770.5950156	0.043607	-770.551408	48.47	1
c6k	-770.4926116	0.038323	-770.454288	109.42	0
c6l	-770.4955756	0.039391	-770.456185	108.23	0
c6m	-770.5135895	0.040454	-770.473135	97.59	0

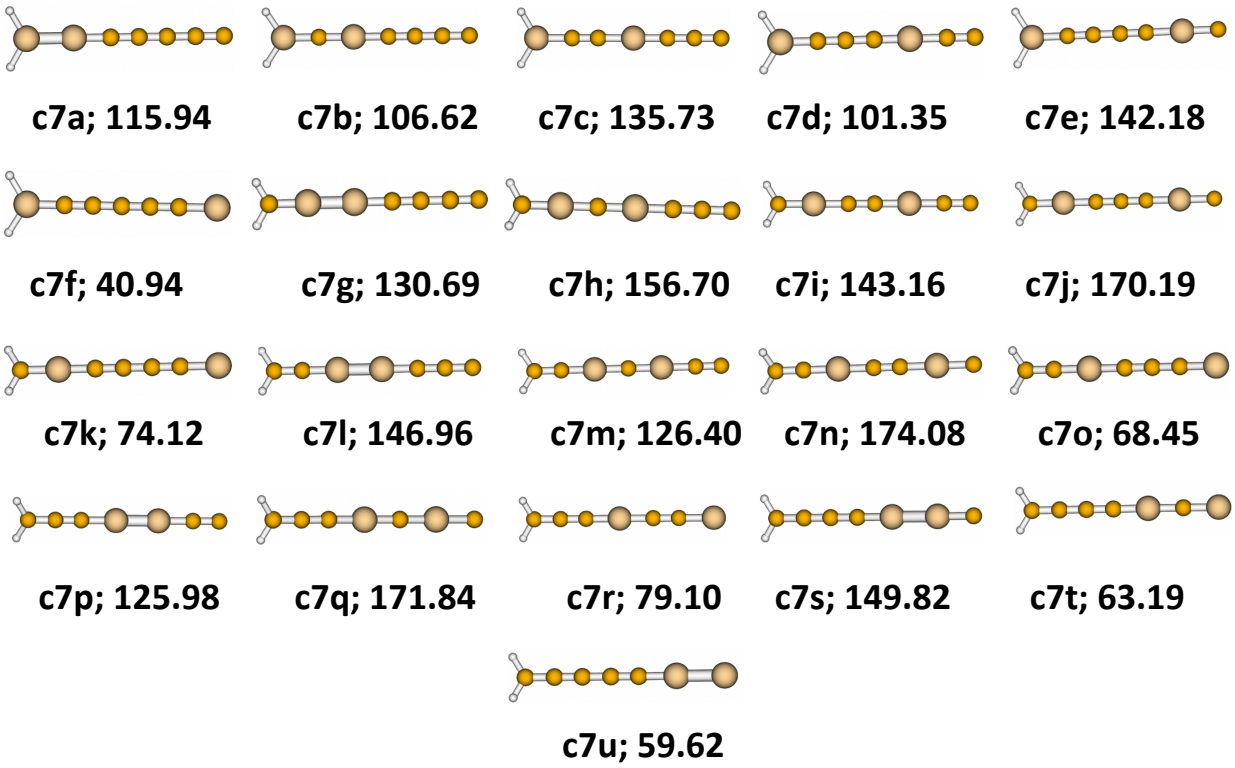

Figure S7: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, heptahexaenylidene.

Table S49: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S7 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c7a	-770.4818971	0.038003	-770.443894	115.94	2
c7b	-770.4969221	0.038172	-770.458750	106.62	1
c7c	-770.4488082	0.036447	-770.412362	135.73	2
c7d	-770.5048258	0.037686	-770.467140	101.35	1
c7e	-770.4395925	0.037511	-770.402081	142.18	2
c7f	-770.6048300	0.041418	-770.563412	40.94	0
c7g	-770.4626383	0.042254	-770.420385	130.69	2
c7h	-770.4185428	0.039610	-770.378933	156.70	3
c7i	-770.4404315	0.039918	-770.400514	143.16	2
c7j	-770.3974557	0.040023	-770.357433	170.19	2
c7k	-770.5538651	0.043321	-770.510544	74.12	1
c7l	-770.4363644	0.041900	-770.394465	146.96	3
c7m	-770.4685836	0.041367	-770.427216	126.40	3
c7n	-770.3917528	0.040509	-770.351244	174.08	4
c7o	-770.5639069	0.044334	-770.519573	68.45	1
c7p	-770.4702053	0.042310	-770.427895	125.98	2
c7q	-770.3953198	0.040516	-770.354804	171.84	3
c7r	-770.5459019	0.043308	-770.502594	79.10	2
c7s	-770.4321491	0.042255	-770.389894	149.82	3
c7t	-770.5718824	0.043933	-770.527950	63.19	1
c7u	-770.5792553	0.045606	-770.533649	59.62	1

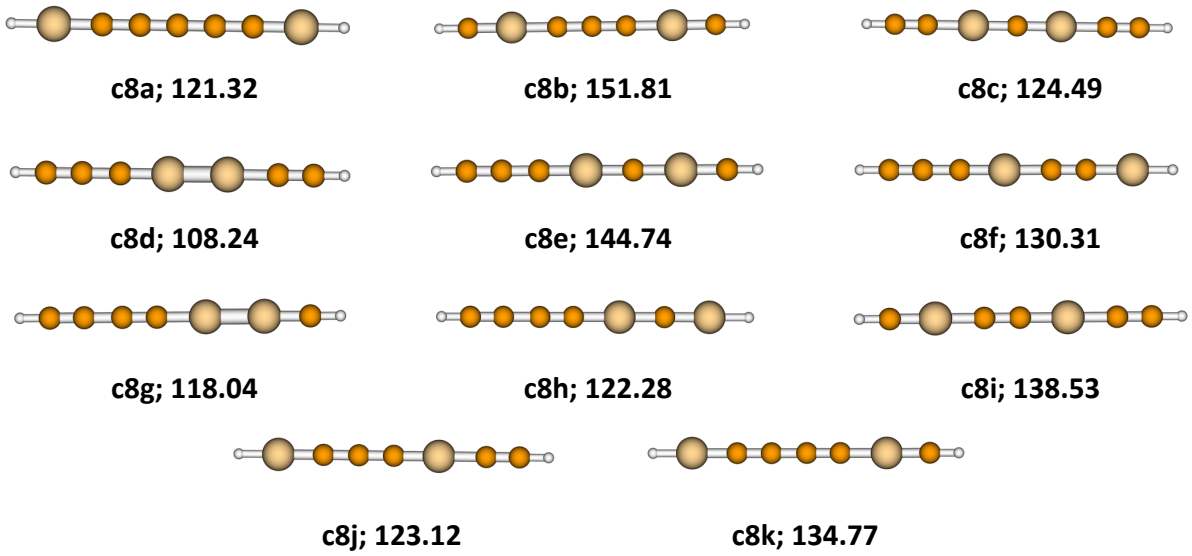

Figure S8: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, hexatriynylidene.

Table S50: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S8 in their triplet ground electronic states calculated at the UB3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c8a	-770.4702368	0.034912	-770.435324	121.32	4
c8b	-770.4248521	0.038127	-770.386725	151.81	4
c8c	-770.4705217	0.040249	-770.430273	124.49	4
c8d	-770.4972891	0.041130	-770.456159	108.24	4
c8e	-770.4365091	0.038515	-770.397994	144.74	4
c8f	-770.4583353	0.037340	-770.420996	130.31	4
c8g	-770.4816449	0.041096	-770.440549	118.04	4
c8h	-770.4718362	0.038040	-770.433796	122.28	4
c8i	-770.4472557	0.039362	-770.407894	138.53	4
c8j	-770.4701865	0.037739	-770.432448	123.12	4
c8k	-770.4511470	0.037266	-770.413882	134.77	4

Table S51: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S8 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c8a	-770.4468455	0.036357	-770.410488	136.90	2
c8b	-770.3991762	0.039064	-770.360112	168.51	4
c8c	-770.4494617	0.041149	-770.408313	138.27	2
c8d	-770.4732788	0.041796	-770.431483	123.73	3
c8e	-770.4119794	0.039128	-770.372852	160.52	5
c8f	-770.4336879	0.038256	-770.395432	146.35	3
c8g	-770.4558674	0.042330	-770.413538	134.99	3
c8h	-770.4504333	0.039453	-770.410981	136.59	2
c8i	-770.4235990	0.040195	-770.383404	153.90	3
c8j	-770.4479685	0.039106	-770.408863	137.92	2
c8k	-770.4265339	0.038582	-770.387952	151.04	3

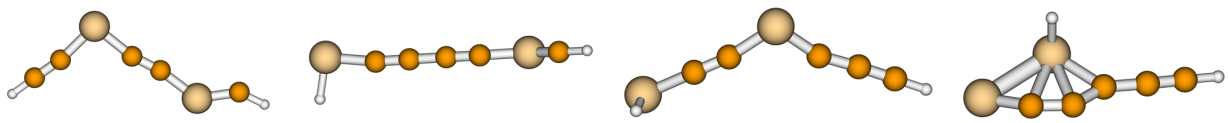

Figure S9: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, *bent*-1,3,5-hexatriynylcarbene.

Table S52: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S9 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c10i	-770.5340330	0.041449	-770.492584	85.39	1
c10k	-770.5281384	0.039564	-770.488574	87.90	0
c10r	-770.5451901	0.039043	-770.506147	76.87	0
c10u	-770.5970300	0.042729	-770.554302	46.66	0

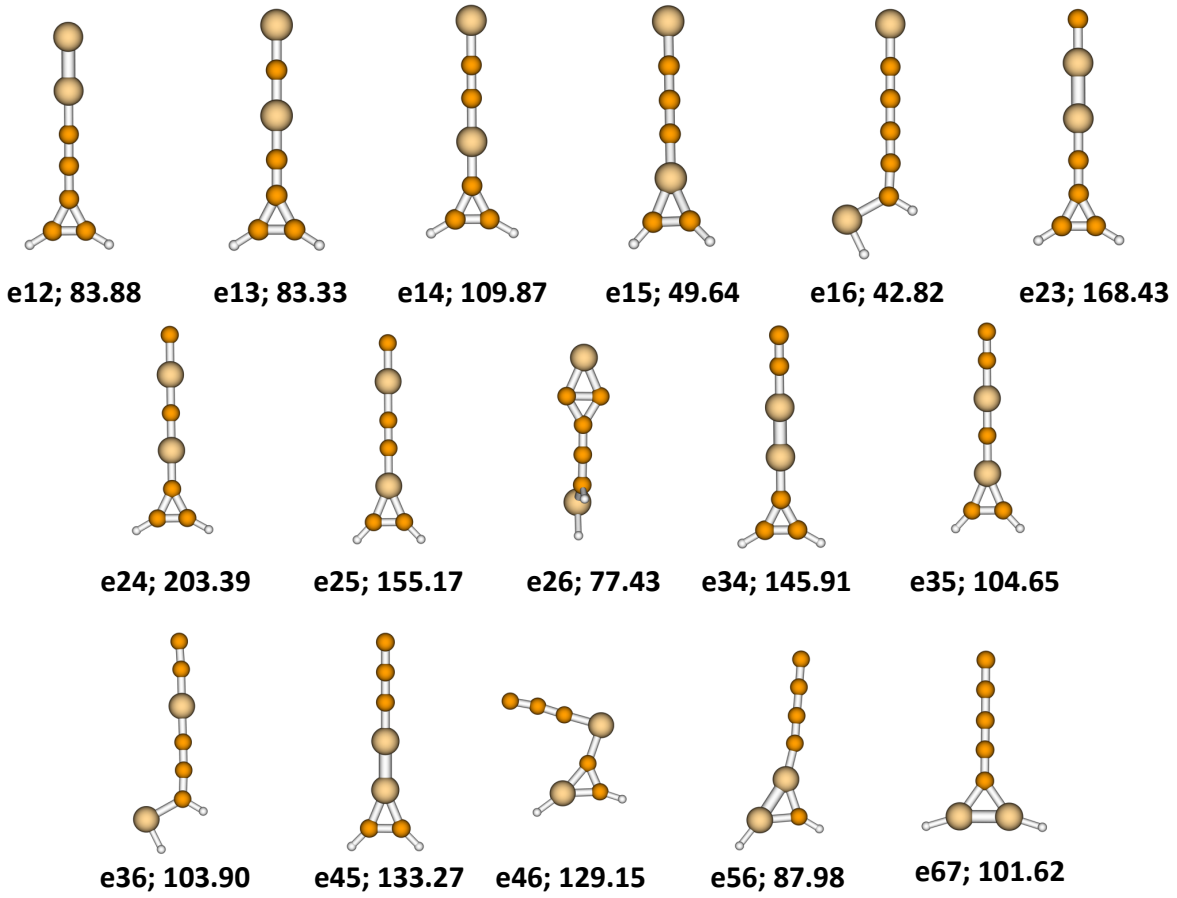

Figure S10: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, 3-(didehydrobutatrienylidene)cyclopropene.

Table S53: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S10 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
e12	-770.5406767	0.045696	-770.494980	83.88	1
e13	-770.5405104	0.044655	-770.495856	83.33	1
e14	-770.4972920	0.043724	-770.453568	109.87	2
e15	-770.5956438	0.046103	-770.549541	49.64	0
e16	-770.6029841	0.042565	-770.560420	42.82	0
e23	-770.4030670	0.042816	-770.360251	168.43	3
e24	-770.3454156	0.040880	-770.304535	203.39	3
e25	-770.4237944	0.042427	-770.381367	155.17	2
e26	-770.5467107	0.041449	-770.505262	77.43	0
e34	-770.4392737	0.043150	-770.396124	145.91	2
e35	-770.5050515	0.043173	-770.461879	104.65	2
e36	-770.5021255	0.039041	-770.463085	103.90	1
e45	-770.4597961	0.043529	-770.416267	133.27	2
e46	-770.4620627	0.039219	-770.422843	129.15	1
e56	-770.5297548	0.041313	-770.488441	87.98	0
e67	-770.5041868	0.037475	-770.466712	101.62	0

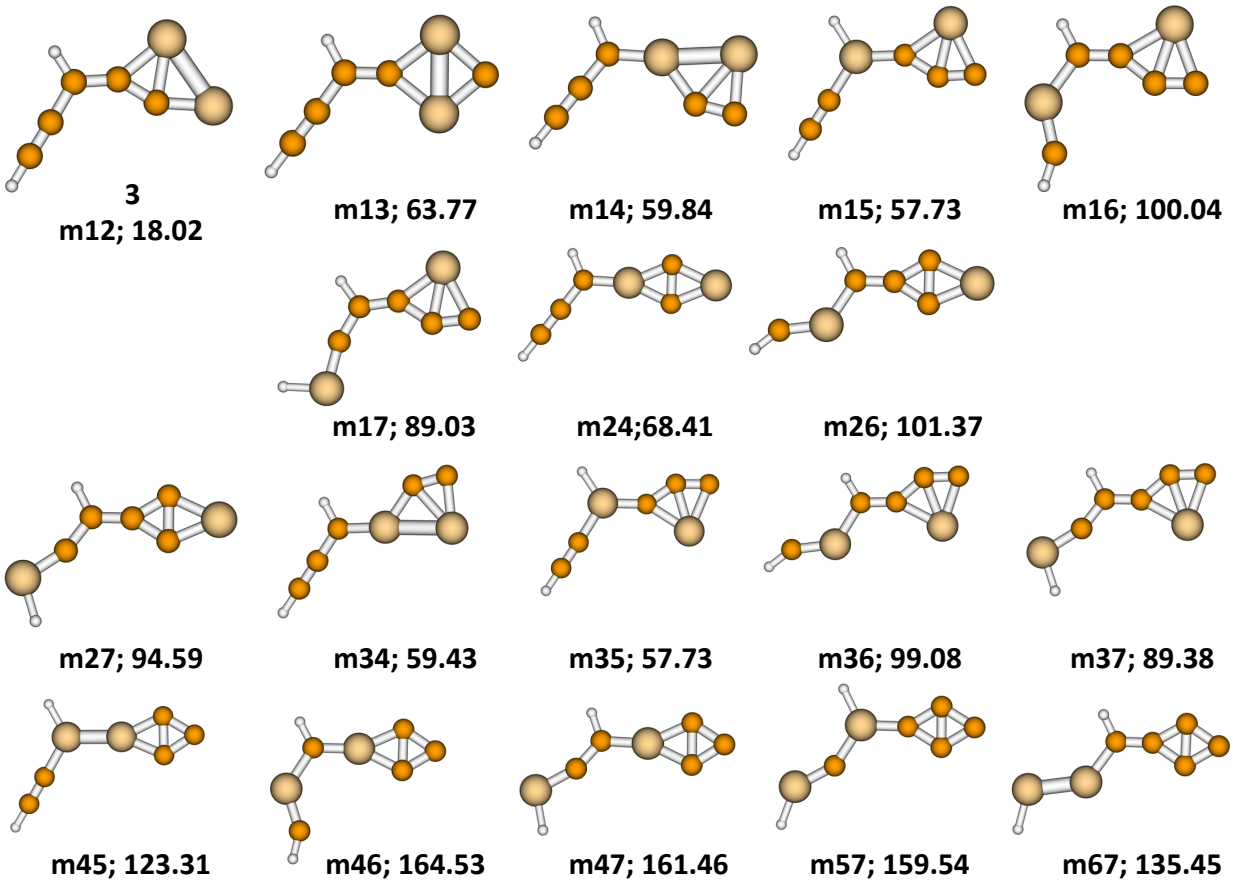

Figure S11: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, 2-(ethynyl)methylenebicyclo[1.1.0]-but-1(3)-ene-4-ylidene.

Table S54: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S11 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
3; m12	-770.6467098	0.046777	-770.599932	18.02	0
m13	-770.5723890	0.045355	-770.527034	63.77	0
m14	-770.5771674	0.043881	-770.533287	59.84	0
m15	-770.5782172	0.041556	-770.536661	57.73	0
m16	-770.5128009	0.043568	-770.469233	100.04	0
m17	-770.5271398	0.040358	-770.486781	89.03	1
m24	-770.5640365	0.044406	-770.519631	68.41	0
m26	-770.5108442	0.043733	-770.467112	101.37	0
m27	-770.5185454	0.040623	-770.477923	94.59	1
m34	-770.5776603	0.043707	-770.533953	59.43	0
m35	-770.5781353	0.041472	-770.536663	57.73	0
m36	-770.5141219	0.043363	-770.470759	99.08	0
m37	-770.5264812	0.040263	-770.486218	89.38	1
m45	-770.4713730	0.039222	-770.432151	123.31	1
m46	-770.4067756	0.040316	-770.366460	164.53	0
m47	-770.4080964	0.036743	-770.371354	161.46	2
m57	-770.4096247	0.035207	-770.374418	159.54	1
m67	-770.4521073	0.039311	-770.412796	135.45	0

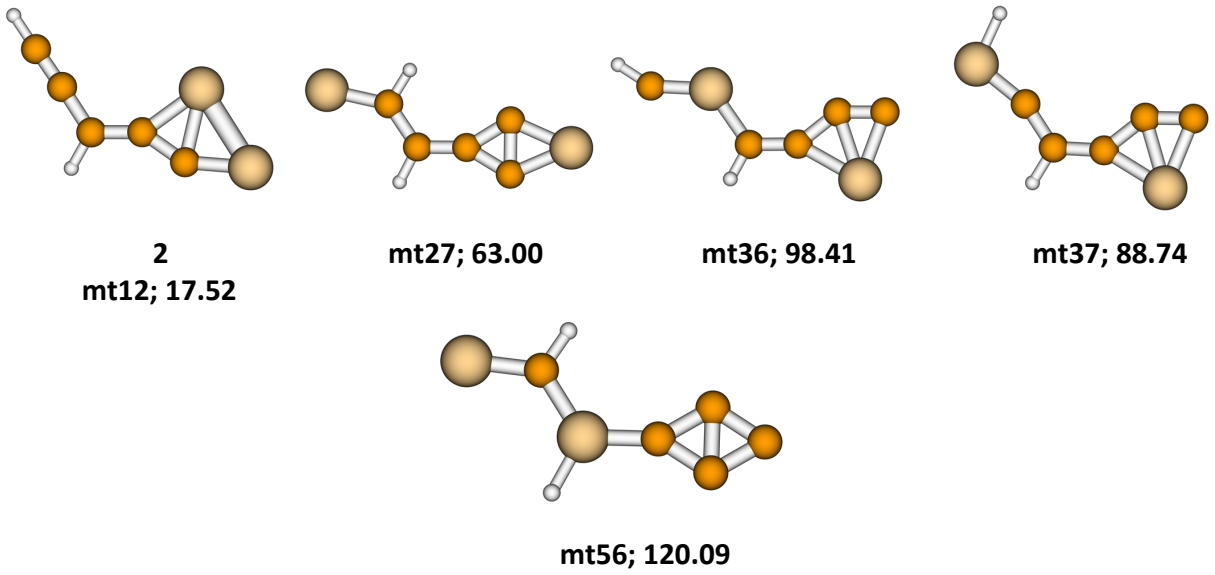

Figure S12: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, 2-(ethynyl)methylenebicyclo[1.1.0]-but-1(3)-ene-4-ylidene.

Table S55: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S12 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
2; mt12	-770.6473552	0.046621	-770.600734	17.52	0
mt27	-770.5740532	0.045794	-770.528259	63.00	0
mt36	-770.5153110	0.043477	-770.471834	98.41	0
mt37	-770.5276882	0.040447	-770.487241	88.74	1
mt56	-770.4761786	0.038907	-770.437272	120.09	0

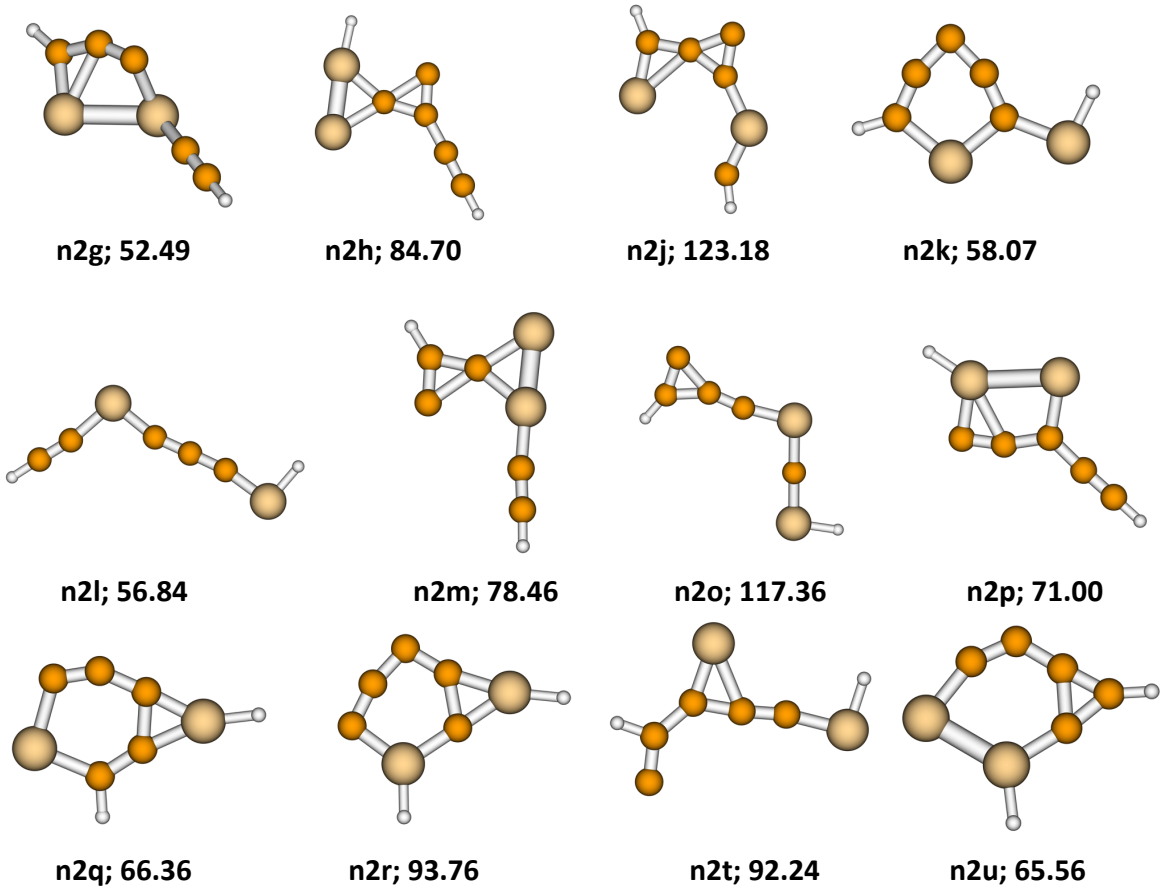

Figure S13: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, 2,5-(didehydro)-1-ethynylspiro[2.2]penta-1,4-diene.

Table S56: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S13 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
n2g	-770.5889593	0.043952	-770.545007	52.49	0
n2h	-770.5342756	0.040606	-770.493670	84.70	0
n2j	-770.4744081	0.042061	-770.432347	123.18	0
n2k	-770.5785420	0.042421	-770.536121	58.07	0
n2l	-770.5779026	0.039831	-770.538072	56.84	0
n2m	-770.5465314	0.042911	-770.503620	78.46	0
n2o	-770.4793759	0.037755	-770.441621	117.36	0
n2p	-770.5568807	0.041370	-770.515510	71.00	1
n2q	-770.5653788	0.042476	-770.522903	66.36	0
n2r	-770.5190269	0.039782	-770.479245	93.76	1
n2t	-770.5206625	0.039000	-770.481662	92.24	0
n2u	-770.5672881	0.043103	-770.524185	65.56	0

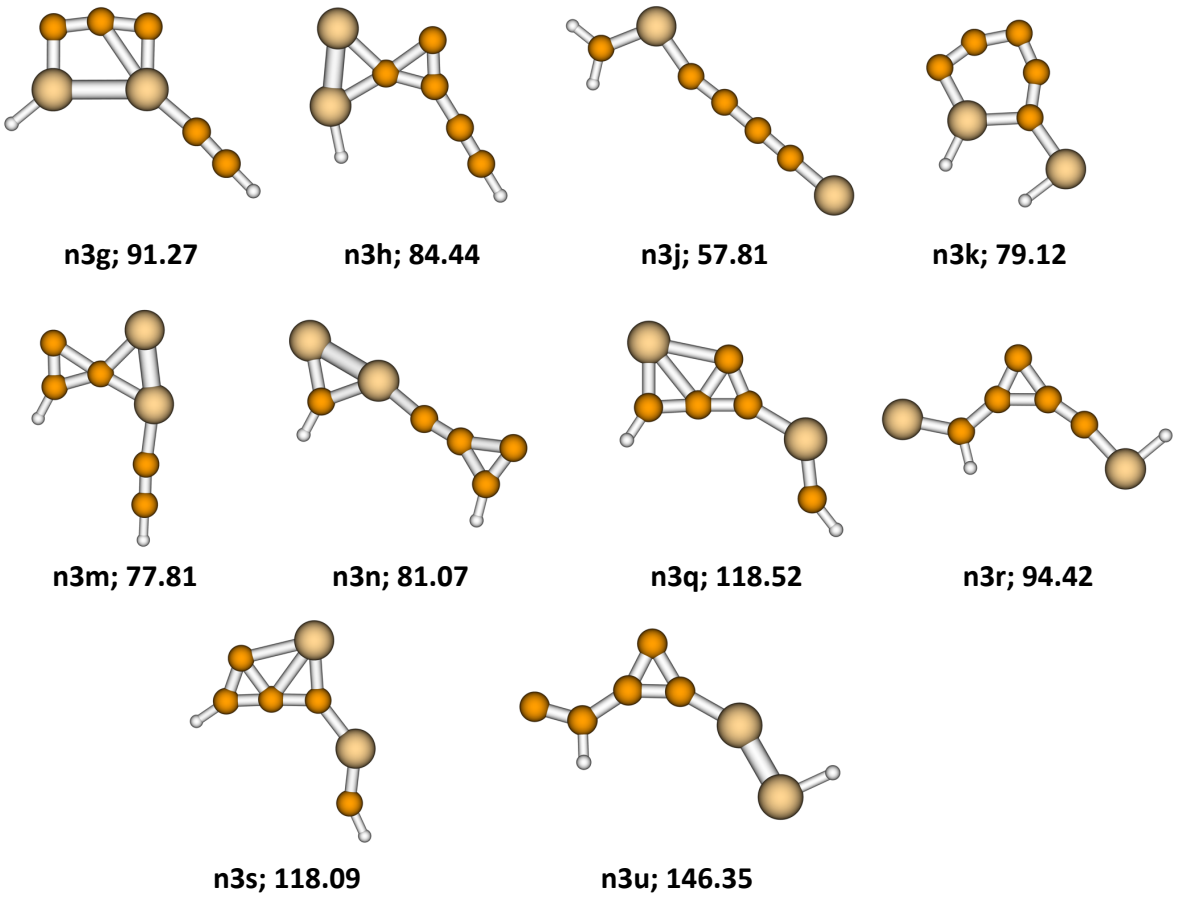

Figure S14: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, 2,4-(didehydro)-1-ethynylspiro[2.2]penta-1,4-diene.

Table S57: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S14 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
n3g	-770.5243537	0.041142	-770.483212	91.27	0
n3h	-770.5342560	0.040163	-770.494093	84.44	0
n3j	-770.5809808	0.044460	-770.536521	57.81	0
n3k	-770.5433660	0.040796	-770.502570	79.12	0
n3m	-770.5472010	0.042553	-770.504648	77.81	0
n3n	-770.5418273	0.042363	-770.499464	81.07	0
n3q	-770.4819715	0.042192	-770.439780	118.52	0
n3r	-770.5177352	0.039555	-770.478181	94.42	0
n3s	-770.4824358	0.041976	-770.440460	118.09	1
n3u	-770.4329822	0.037554	-770.395428	146.35	1

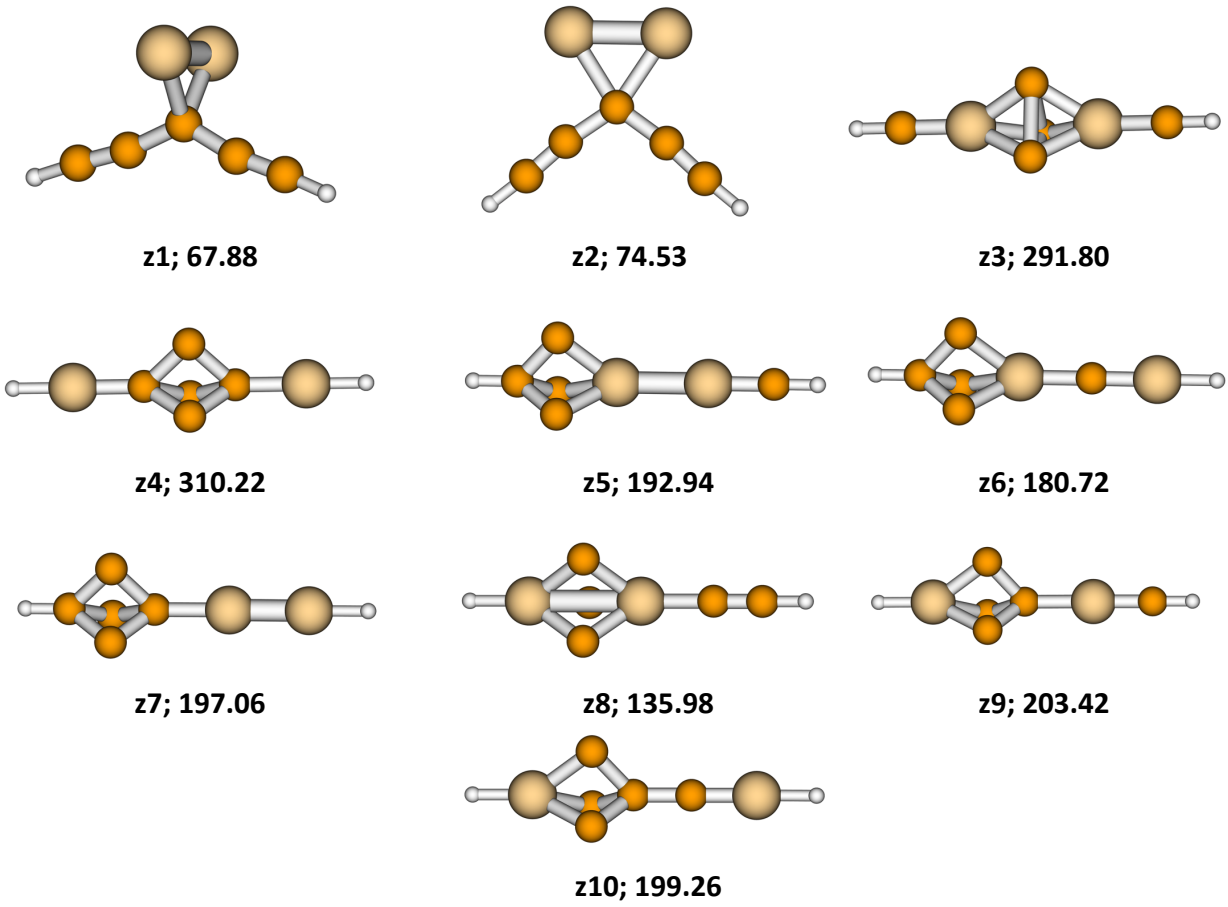

Figure S15: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_5H_2 geometry, pentacyclo[2.1.0.0^{1,3}.0^{2,5}.0^{3,5}]pentane

Table S58: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S15 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
z1	-770.5642055	0.043722	-770.520484	67.88	2
z2	-770.5533472	0.043471	-770.509876	74.53	1
z3	-770.1980051	0.034365	-770.163640	291.80	4
z4	-770.1640080	0.029714	-770.134294	310.22	4
z5	-770.3623391	0.041149	-770.321190	192.94	2
z6	-770.3796120	0.038953	-770.340659	180.72	2
z7	-770.3545323	0.039915	-770.314617	197.06	2
z8	-770.4524442	0.040486	-770.411958	135.98	0
z9	-770.3422288	0.037750	-770.304479	203.42	2
z10	-770.3470475	0.035933	-770.311114	199.26	2

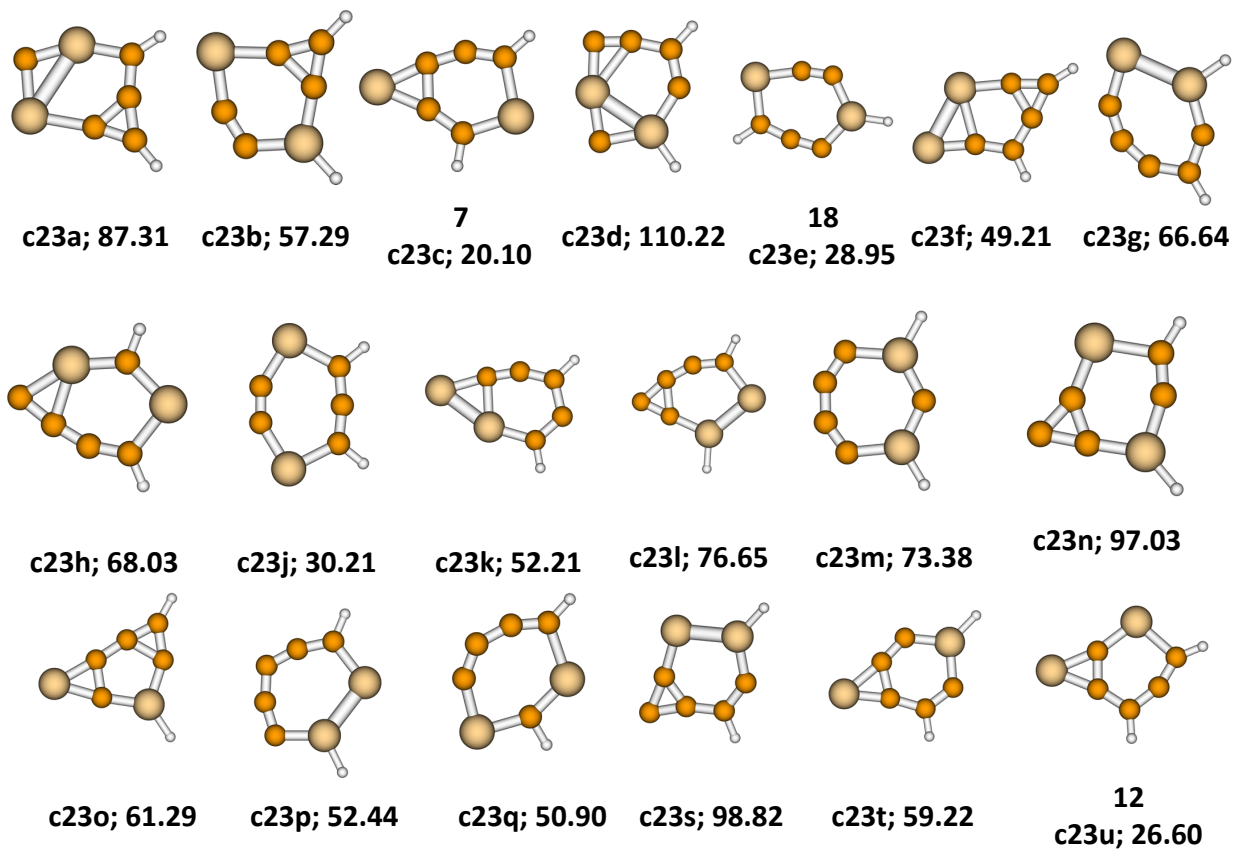

Figure S16: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, 3,5-(didehydro)bicyclo[4.1.0]hepta-1(6),2,4-triene-7-ylidene.

Table S59: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S16 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c23a	-770.5347469	0.045229	-770.489518	87.31	0
c23b	-770.5802125	0.042859	-770.537353	57.29	1
7; c23c	-770.6434198	0.046791	-770.596629	20.10	0
c23d	-770.4954423	0.042432	-770.453010	110.22	1
18; c23e	-770.6290167	0.046505	-770.582512	28.95	0
c23f	-770.5972604	0.047023	-770.550237	49.21	0
c23g	-770.5652507	0.042790	-770.522460	66.64	0
c23h	-770.5644984	0.044264	-770.520235	68.03	0
c23j	-770.6241664	0.043647	-770.580519	30.21	1
c23k	-770.5915527	0.046106	-770.545447	52.21	0
c23l	-770.5483746	0.041871	-770.506504	76.65	0
c23m	-770.5527726	0.041052	-770.511720	73.38	0
c23n	-770.5137820	0.039748	-770.474034	97.03	1
c23o	-770.5743518	0.043367	-770.530984	61.29	0
c23p	-770.5887595	0.043678	-770.545082	52.44	0
c23q	-770.5920290	0.044491	-770.547538	50.90	0
c23s	-770.5128938	0.041721	-770.471173	98.82	1
c23t	-770.5784321	0.044156	-770.534276	59.22	0
12; c23u	-770.6323326	0.046074	-770.586259	26.60	0

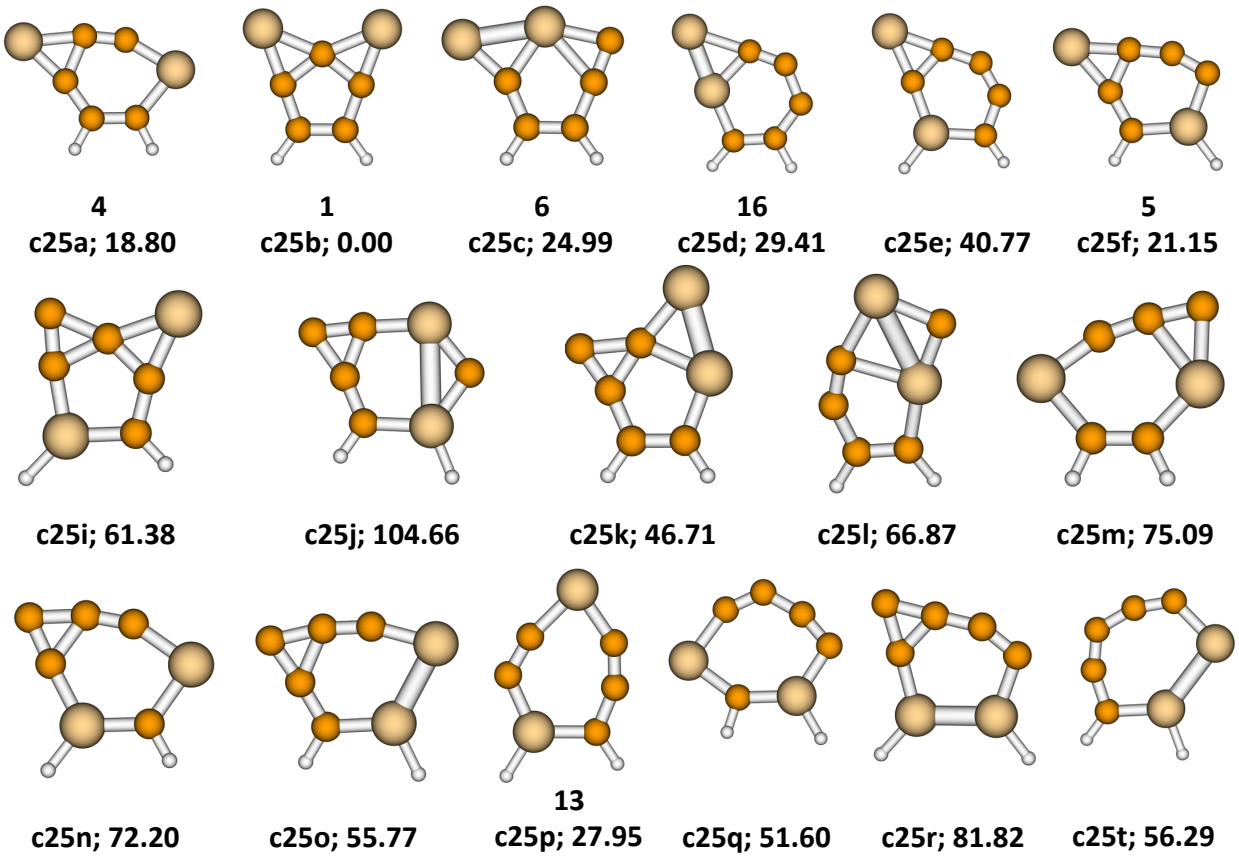

Figure S17: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, bicyclo[4.1.0]hepta-1(6),4(5)-diene-2(3)-yne-7-ylidene.

Table S60: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S17 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
4; c25a	-770.6468126	0.048113	-770.598699	18.80	0
1; c25b	-770.6783795	0.049725	-770.628654	0.00	0
6; c25c	-770.6366832	0.047858	-770.588825	24.99	0
16; c25d	-770.6300475	0.048269	-770.581779	29.41	0
c25e	-770.6082254	0.044550	-770.563675	40.77	0
5; c25f	-770.6405389	0.045596	-770.594943	21.15	0
c25i	-770.5737679	0.042933	-770.530835	61.38	0
c25j	-770.5034721	0.041605	-770.461867	104.66	1
c25k	-770.6013334	0.047116	-770.554218	46.71	0
c25l	-770.5684716	0.046385	-770.522087	66.87	0
c25m	-770.5540906	0.045107	-770.508984	75.09	0
c25n	-770.5555843	0.041989	-770.513595	72.20	0
c25o	-770.5828171	0.043042	-770.539775	55.77	0
13; c25p	-770.6281327	0.044012	-770.584120	27.95	0
c25q	-770.5896321	0.043206	-770.546426	51.60	0
c25r	-770.5381368	0.039875	-770.498262	81.82	0
c25t	-770.5816653	0.042711	-770.538954	56.29	0

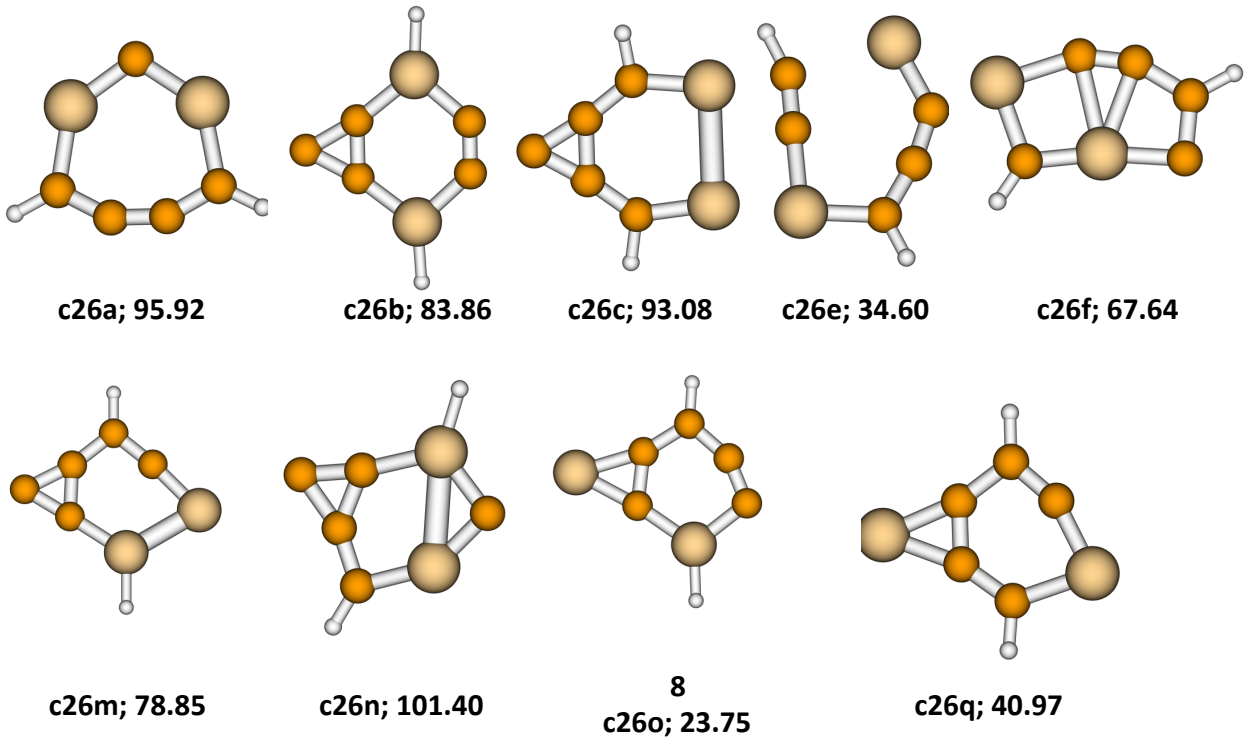

Figure S18: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, bicyclo[4.1.0]hepta-1,5-diene-3-yne-7-ylidene.

Table S61: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S18 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c26a	-770.5195310	0.043742	-770.475789	95.92	2
c26b	-770.5342798	0.039272	-770.495007	83.86	0
c26c	-770.5204216	0.040101	-770.480320	93.08	3
c26e	-770.6182454	0.044732	-770.573514	34.60	0
c26f	-770.5664597	0.045589	-770.520870	67.64	0
c26m	-770.5456798	0.042677	-770.503003	78.85	0
c26n	-770.5091319	0.042065	-770.467067	101.40	0
8; c26o	-770.6366782	0.045878	-770.590800	23.75	0
c26q	-770.6101113	0.046742	-770.563369	40.97	0

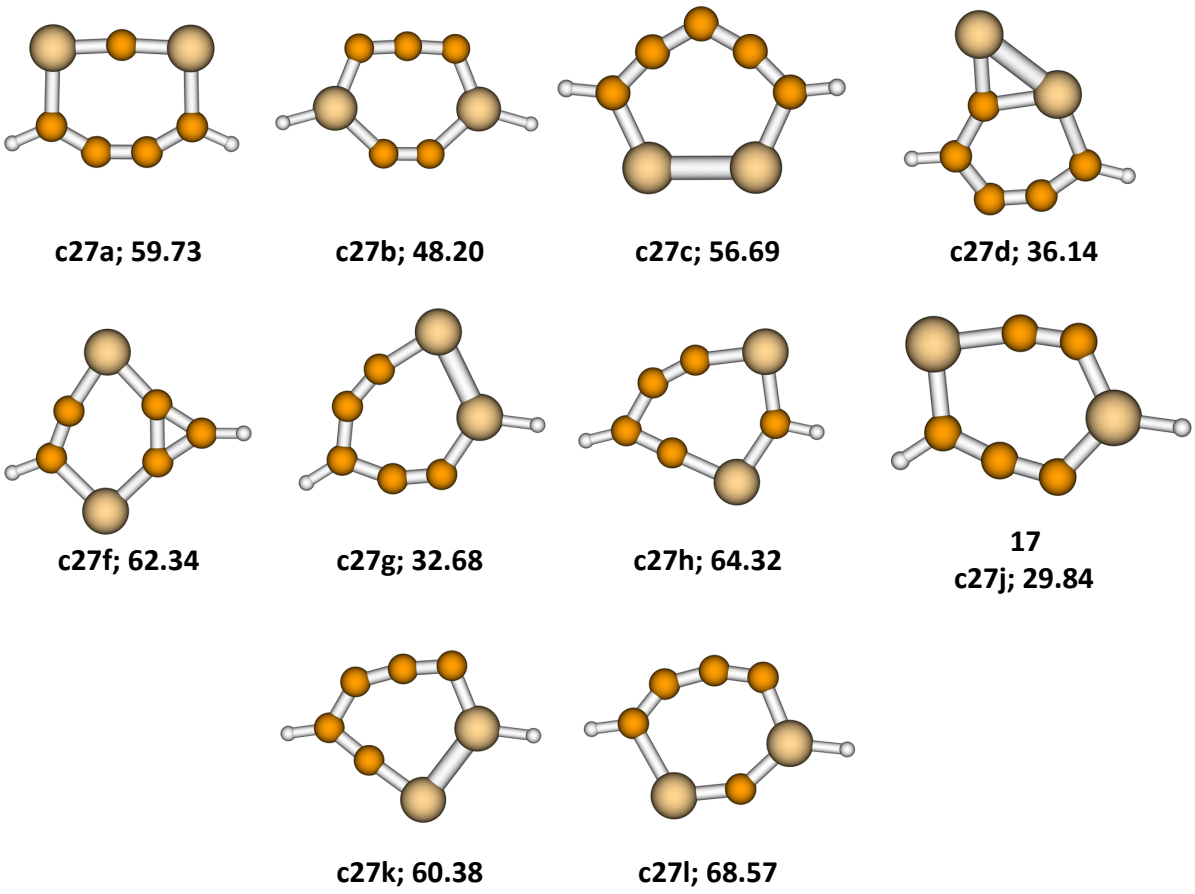

Figure S19: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, cyclohepta-1,2,3,4-tetraene-6-yne.

Table S62: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S19 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c27a	-770.5791516	0.045680	-770.533472	59.73	0
c27b	-770.5937085	0.041860	-770.551849	48.20	0
c27c	-770.5834950	0.045181	-770.538314	56.69	0
c27d	-770.6187115	0.047646	-770.571065	36.14	0
c27f	-770.5735532	0.044248	-770.529305	62.34	0
c27g	-770.6213075	0.044733	-770.576574	32.68	0
c27h	-770.5711212	0.044963	-770.526158	64.32	0
17; c27j	-770.6253429	0.044247	-770.581096	29.84	0
c27k	-770.5761218	0.043684	-770.532438	60.38	0
c27l	-770.5621512	0.042766	-770.519385	68.57	0

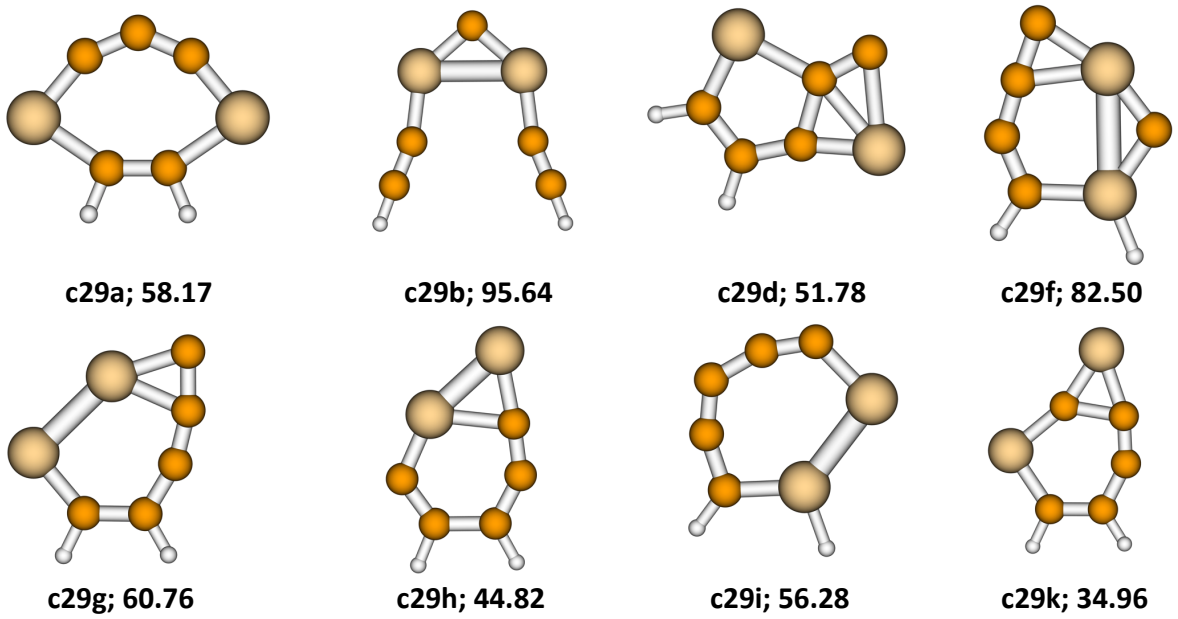

Figure S20: Various structural possibilities obtained for $Si_2C_5H_2$ using the base C_7H_2 geometry, bicyclo[4.1.0]hepta-1,2,4,5-tetraene-7-ylidene.

Table S63: Computed energies of $Si_2C_5H_2$ isomers shown in Figure S20 in their singlet ground electronic states calculated at the B3LYP/6-311++G(2D,2P) level of theory.

Isomer or Label	E a.u	$ZPVE$ a.u	E+$ZPVE$ a.u	ΔE+$ZPVE$ kcal mol^{-1}	NImag
1	-770.6783795	0.049725	-770.628654	0.00	0
c29a	-770.5814188	0.045467	-770.535952	58.17	0
c29b	-770.5176415	0.041398	-770.476243	95.64	2
c29d	-770.5917338	0.045595	-770.546138	51.78	0
c29f	-770.5400690	0.042891	-770.497178	82.50	0
c29g	-770.5781069	0.046277	-770.531829	60.76	0
c29h	-770.6045876	0.047360	-770.557228	44.82	0
c29i	-770.5816669	0.042706	-770.538961	56.28	0
c29k	-770.6197288	0.046794	-770.572935	34.96	0