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

The prediction of far-infrared spectra for planetary nitrile ices using periodic density functional theory with comparison to thin film experiments.

Supplementary Information

C. Ennis,[1*] R. Auchettl,[1,2] D. R. T. Appadoo,[2] and E. G. Robertson[1]

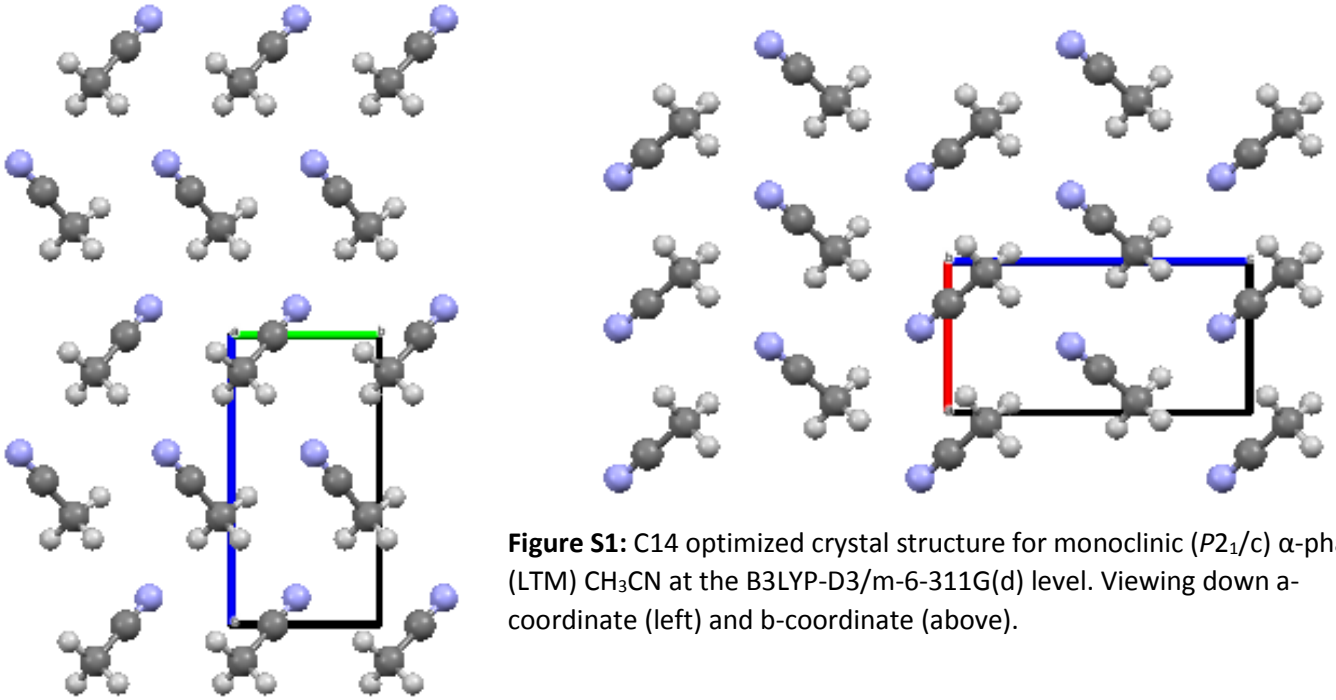

Figure S1: C14 optimized crystal structure for monoclinic ($P2_1/c$) α-phase (LTM) CH_3CN at the B3LYP-D3/m-6-311G(d) level. Viewing down a-coordinate (left) and b-coordinate (above).

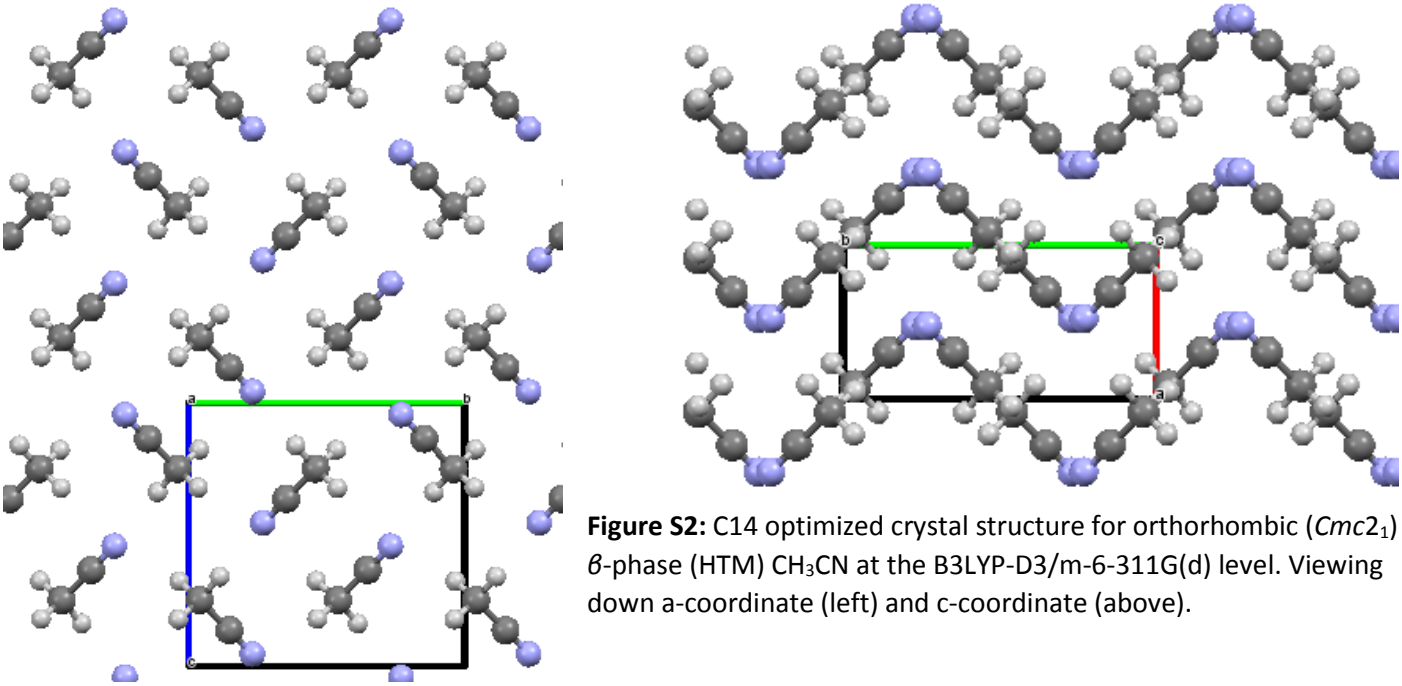

Figure S2: C14 optimized crystal structure for orthorhombic ($Cmc2_1$) *β*-phase (HTM) CH_3CN at the B3LYP-D3/m-6-311G(d) level. Viewing down a-coordinate (left) and c-coordinate (above).

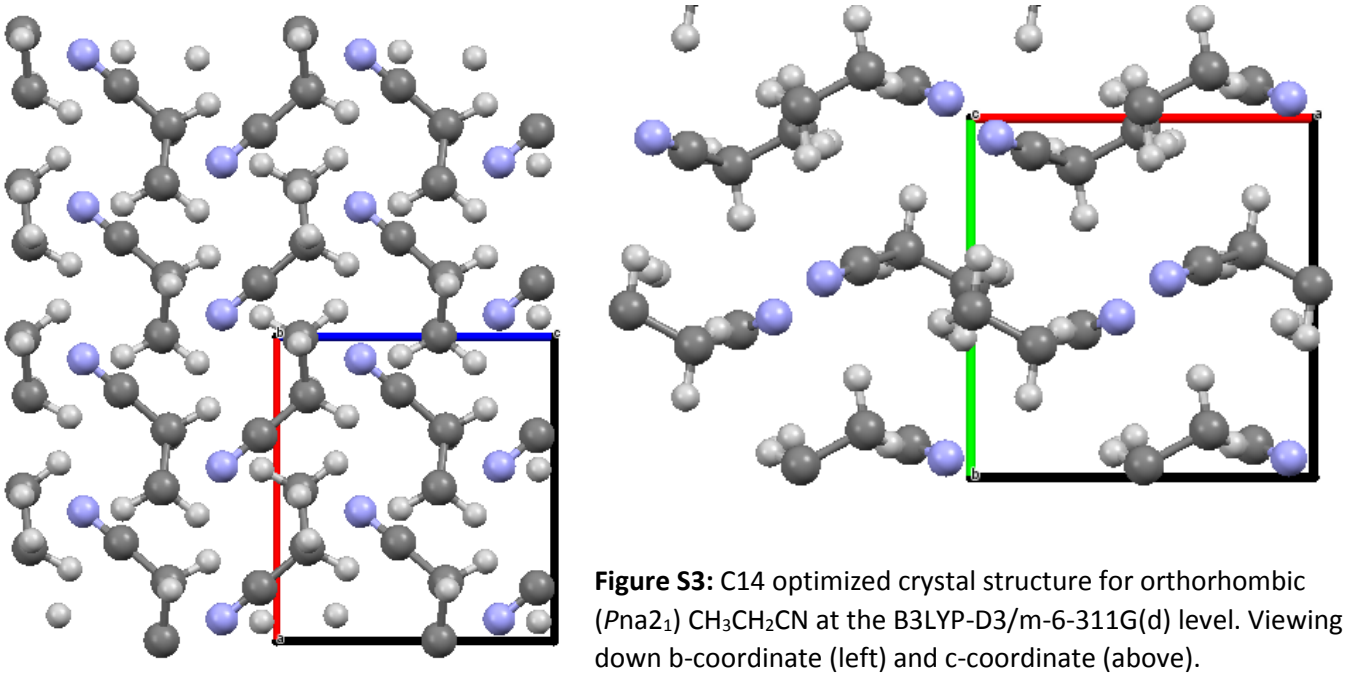

Figure S3: C14 optimized crystal structure for orthorhombic (*P*na2$_1$) CH_3CH_2CN at the B3LYP-D3/m-6-311G(d) level. Viewing down b-coordinate (left) and c-coordinate (above).

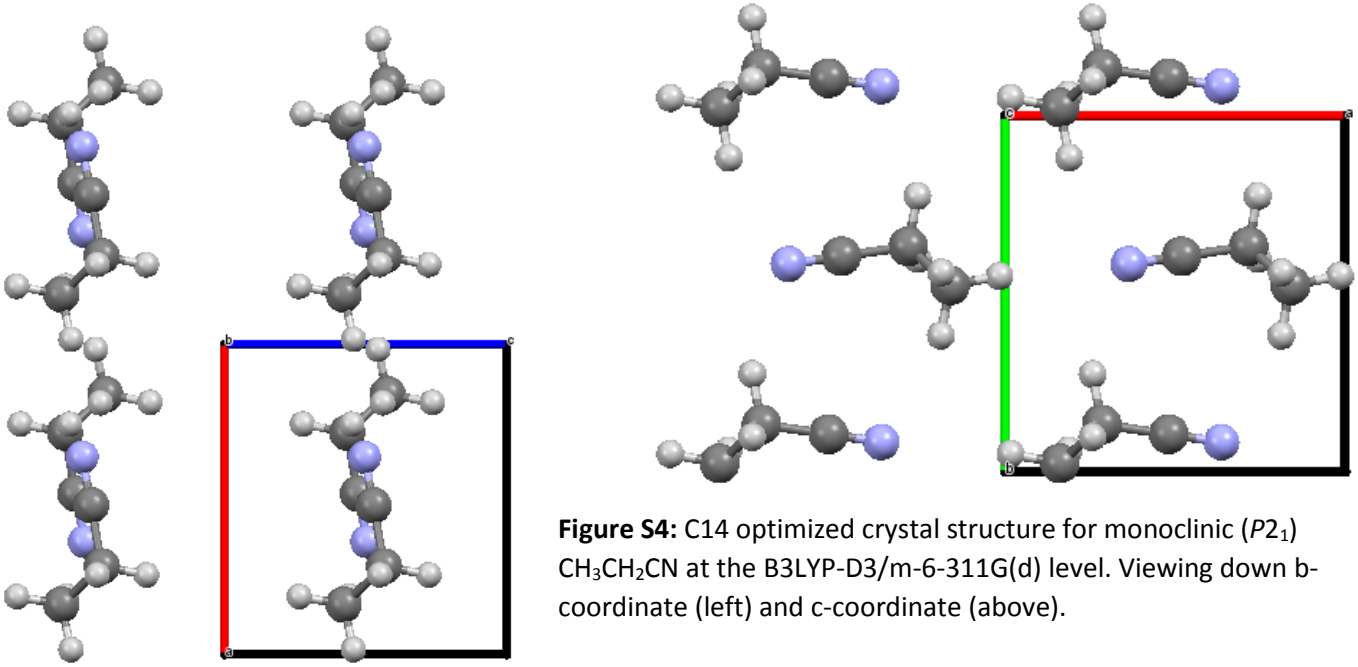

Figure S4: C14 optimized crystal structure for monoclinic (*P*2$_1$) CH_3CH_2CN at the B3LYP-D3/m-6-311G(d) level. Viewing down b-coordinate (left) and c-coordinate (above).

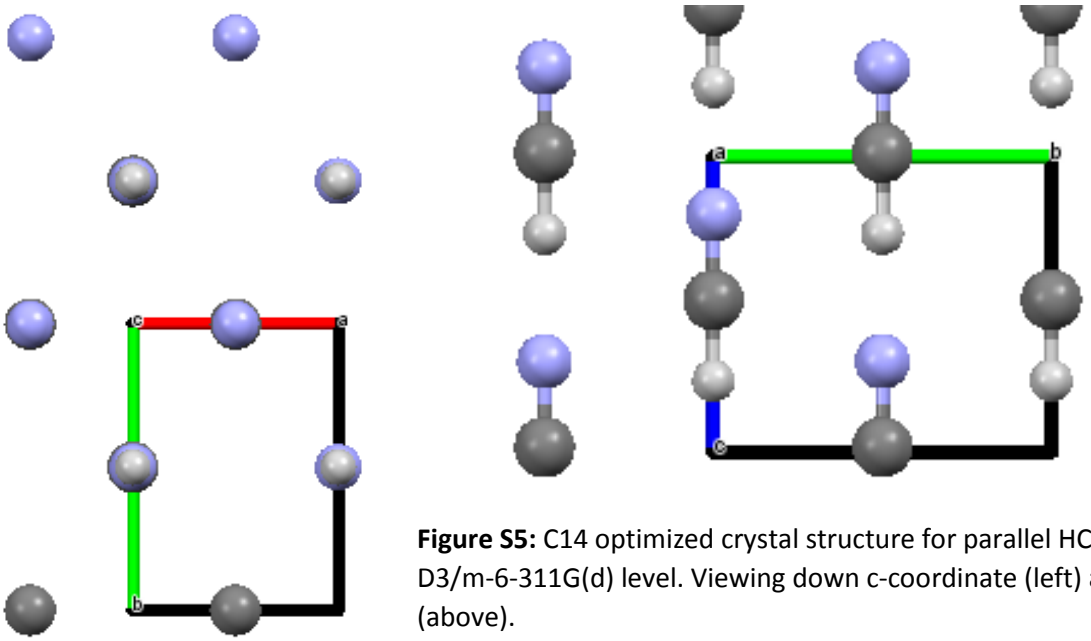

Figure S5: C14 optimized crystal structure for parallel HCN at the B3LYP-D3/m-6-311G(d) level. Viewing down c-coordinate (left) and a-coordinate (above).

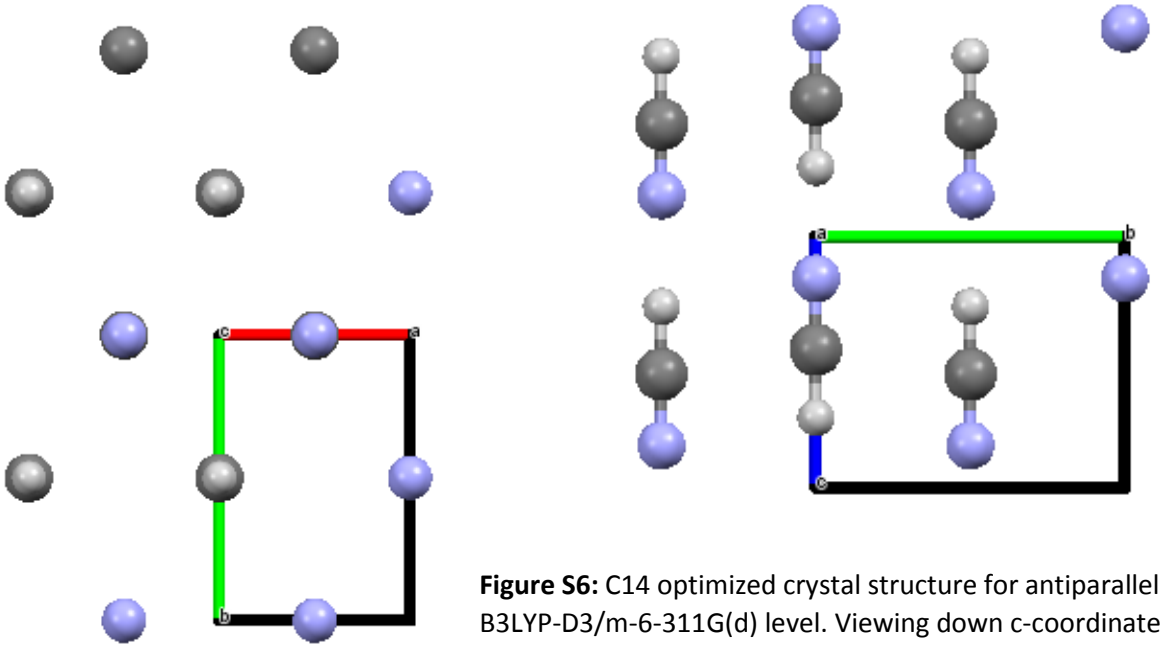

Figure S6: C14 optimized crystal structure for antiparallel HCN at the B3LYP-D3/m-6-311G(d) level. Viewing down c-coordinate (left) and a-coordinate (above).

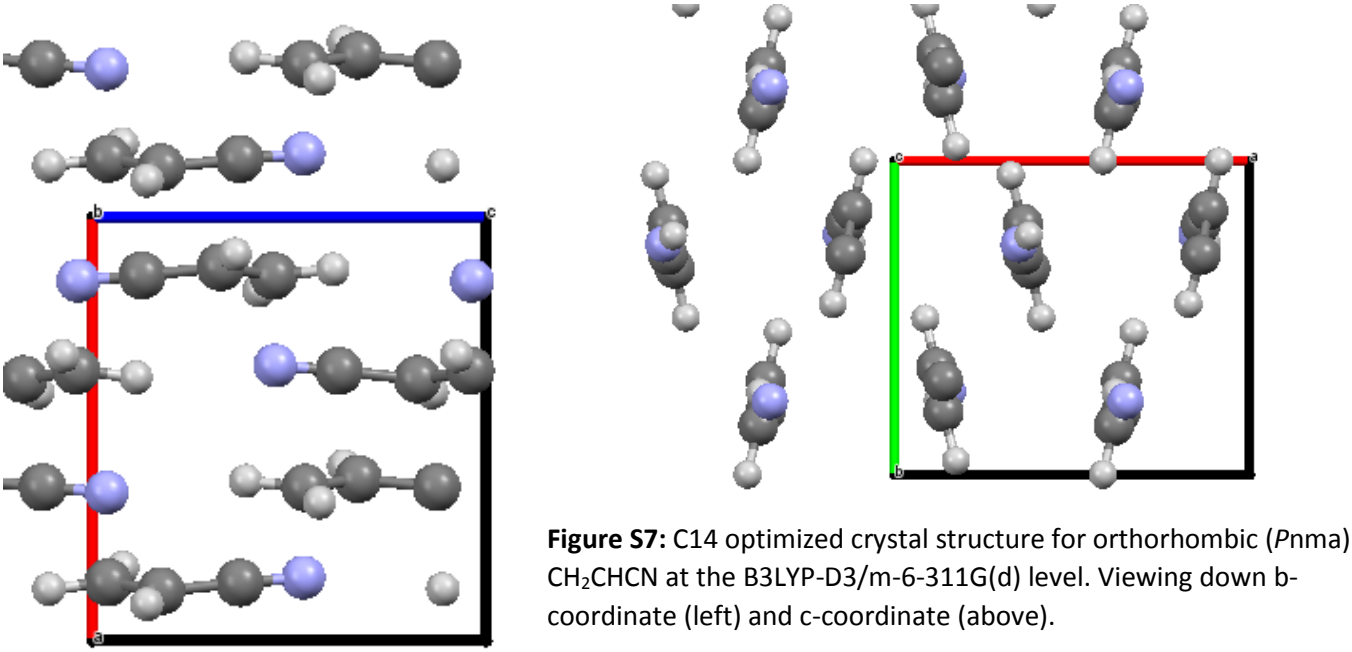

Figure S7: C14 optimized crystal structure for orthorhombic (*P*nma) CH_2CHCN at the B3LYP-D3/m-6-311G(d) level. Viewing down b-coordinate (left) and c-coordinate (above).

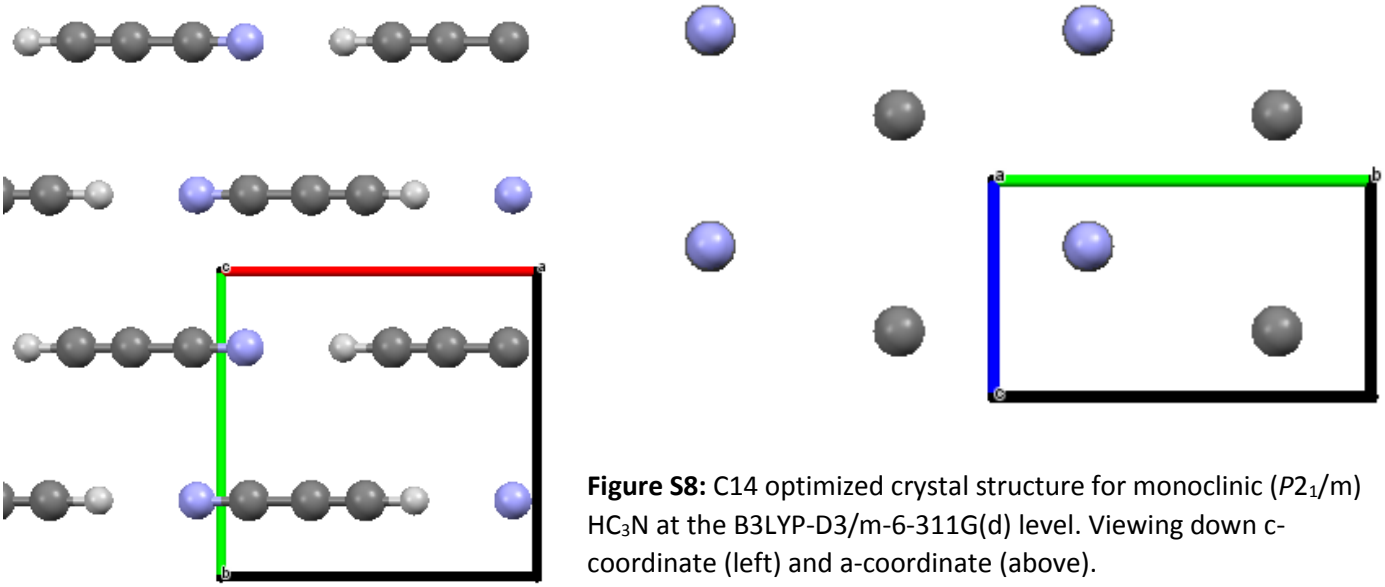

Figure S8: C14 optimized crystal structure for monoclinic ($P2_1/m$) HC_3N at the B3LYP-D3/m-6-311G(d) level. Viewing down c-coordinate (left) and a-coordinate (above).

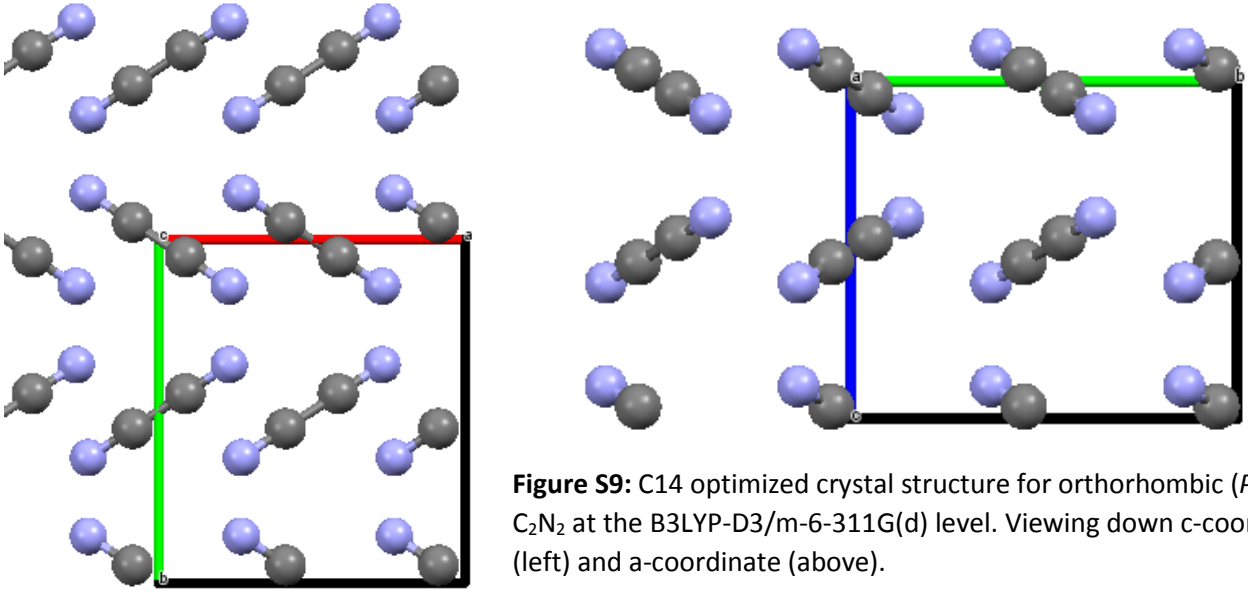

Figure S9: C14 optimized crystal structure for orthorhombic (*P*cab) C_2N_2 at the B3LYP-D3/m-6-311G(d) level. Viewing down c-coordinate (left) and a-coordinate (above).

Table S-1: α-CH_3CN CRYSTAL14 Output File Data: B3LYP-D3/m-6-311G(d)

Mode	Frequencies	Symmetry	Infrared	Intensity	Raman	Assignment
	(cm^{-1})			(km/mol)		
1	0.00	B2	A	0	A	3x Translation
2	0.00	B1	A	0	A	
3	0.00	A1	A	0	A	
4	24.67	A2	I	0	A	Lattice
5	63.95	A1	A	0.47	A	
6	88.88	A2	I	0	A	
7	93.05	A1	A	35.18	A	
8	94.09	B1	A	0.07	A	
9	97.21	B2	A	61.60	A	
10	119.34	B1	A	37.30	A	
11	167.10	A2	I	0	A	
12	178.25	B2	A	1.29	A	
13	407.04	A1	A	3.85	A	ν_8(CCN bend)
14	411.33	B1	A	4.99	A	
15	414.41	A2	I	0	A	
16	416.86	B2	A	5.79	A	
17	912.34	B1	A	4.77	A	ν_4(CC stretch)
18	914.26	A1	A	3.19	A	
19	1064.98	B1	A	8.19	A	ν_7(CH_3 rock)
20	1065.91	A1	A	8.76	A	
21	1070.35	B2	A	16.73	A	
22	1076.45	A2	I	0	A	
23	1414.98	A1	A	5.60	A	ν_3(CH_3 deform)
24	1416.65	B1	A	15.85	A	
25	1468.90	B1	A	5.41	A	ν_6(CH_3 deform)
26	1475.39	A1	A	33.54	A	
27	1476.90	B2	A	44.89	A	
28	1479.35	A2	I	0	A	
29	2343.44	A1	A	73.24	A	ν_2(CN stretch)
30	2346.52	B1	A	75.29	A	
31	3012.24	A1	A	7.82	A	ν_1(CH stretch)
32	3014.85	B1	A	5.86	A	
33	3096.19	B2	A	14.14	A	ν_5(CH stretch)
34	3096.79	A2	I	0	A	
35	3106.15	A1	A	6.18	A	
36	3106.28	B1	A	21.03	A	

Table S-2: β-CH_3CN CRYSTAL14 Output File Data: B3LYP-D3/m-6-311G(d)

Mode	Frequencies	Irrep	Infrared	Intensity	Raman	Assignment
	(cm^{-1})			(km/mol)		
1	0.00	Bu	A	0	I	3x Translation
2	0.00	Bu	A	0	I	
3	0.00	Au	A	0	I	
4	20.18	Bg	I	0	A	Lattice
5	37.26	Au	A	3.50	I	
6	39.64	Ag	I	0	A	
7	52.29	Ag	I	0	A	
8	58.07	Bu	A	20.13	I	
9	67.72	Ag	I	0	A	
10	74.69	Bg	I	0	A	
11	79.30	Au	A	2.13	I	
12	86.06	Bu	A	86.46	I	
13	88.68	Ag	I	0	A	
14	93.21	Ag	I	0	A	
15	93.40	Bg	I	0	A	
16	100.41	Au	A	36.66	I	
17	108.56	Au	A	14.77	I	
18	108.94	Bg	I	0	A	
19	115.64	Ag	I	0	A	
20	119.39	Bu	A	33.38	I	
21	120.08	Bg	I	0	A	
22	124.45	Au	A	36.04	I	
23	128.09	Bg	I	0	A	
24	141.98	Bu	A	37.89	I	
25	403.55	Au	A	7.55	I	ν_8(CCN bend)
26	404.01	Ag	I	0	A	
27	407.20	Bu	A	10.14	I	
28	409.13	Bg	I	0	A	
29	412.76	Bu	A	7.24	I	
30	413.58	Au	A	0.84	I	
31	414.35	Bg	I	0	A	
32	414.47	Ag	I	0	A	
33	911.14	Bg	I	0	A	ν_4(CC stretch)
34	911.93	Au	A	7.72	I	
35	912.69	Bu	A	12.27	I	
36	916.03	Ag	I	0	A	
37	1063.82	Bu	A	20.53	I	ν_7(CH_3 rock)
38	1065.78	Ag	I	0	A	
39	1065.96	Bg	I	0	A	
40	1066.57	Au	A	17.31	I	
41	1070.38	Bg	I	0	A	
42	1071.86	Ag	I	0	A	
43	1076.38	Bu	A	25.93	I	
44	1078.16	Au	A	2.32	I	
45	1418.91	Bu	A	22.31	I	ν_3(CH_3 deform)
46	1422.58	Ag	I	0	A	

47	1422.90	Bg	I	0	A	
48	1423.72	Au	A	32.57	I	
49	1454.91	Bu	A	49.60	I	ν_6(CH_3 deform)
50	1466.00	Bg	I	0	A	
51	1467.90	Au	A	36.6	I	
52	1471.27	Bu	A	79.24	I	
53	1473.97	Ag	I	0	A	
54	1475.01	Au	A	8.60	I	
55	1478.82	Bg	I	0	A	
56	1481.73	Ag	I	0	A	
57	2346.36	Ag	I	0	A	ν_2(CN stretch)
58	2348.77	Au	A	86.16	I	
59	2349.58	Bu	A	160.84	I	
60	2349.58	Bg	I	0	A	
61	3017.58	Bu	A	52.01	I	ν_1(CH stretch)
62	3017.93	Au	A	0.94	I	
63	3019.40	Ag	I	0	A	
64	3021.40	Bg	I	0	A	
65	3102.50	Bg	I	0	A	ν_5(CH stretch)
66	3102.74	Ag	I	0	A	
67	3103.75	Bu	A	26.13	I	
68	3104.42	Au	A	33.25	I	
69	3109.31	Bu	A	76.91	I	
70	3109.76	Ag	I	0	A	
71	3110.01	Au	A	30.56	I	
72	3110.07	Bg	I	0	A	

Table S-3: C_2H_5CN ($Pna2_1$) CRYSTAL14 Output File Data: B3LYP-D3/m-6-311G(d)

Mode	Frequencies	Irrep	Infrared	Intensity	Raman	Assignment
	(cm^{-1})			(km/mol)		
1	0.00	A1	A	0	A	3x Translation
2	0.00	B2	A	0	A	
3	0.00	B1	A	0	A	
4	15.54	A2	I	0	A	Lattice
5	28.67	A1	A	16.32	A	
6	37.91	A2	I	0	A	
7	72.72	B2	A	0.14	A	
8	74.30	B1	A	13.12	A	
9	83.97	A1	A	0.29	A	
10	88.78	A2	I	0	A	
11	97.68	A1	A	21.83	A	
12	103.21	B1	A	2.97	A	
13	103.76	A2	I	0	A	
14	119.76	A1	A	1.34	A	
15	124.03	B1	A	61.13	A	
16	126.90	B2	A	11.31	A	
17	131.61	B2	A	2.68	A	
18	147.19	A2	I	0	A	
19	162.11	B1	A	5.47	A	
20	163.67	A2	I	0	A	
21	165.14	B2	A	70.14	A	
22	190.87	B1	A	0.27	A	
23	194.63	B2	A	0.21	A	
24	198.64	A1	A	2.76	A	
25	227.08	A1	A	1.20	A	ν_{13}, ν_{21}(C-C≡N bend/torsion)
26	236.65	A2	I	0	A	
27	258.46	B1	A	24.62	A	
28	259.50	B2	A	2.88	A	
29	351.17	A1	A	1.24	A	
30	356.36	A2	I	0	A	
31	399.14	B1	A	6.24	A	
32	401.13	B2	A	0	A	
33	420.14	A2	I	0	A	ν_{20}(C-C≡N bend)
34	423.19	A1	A	1.40	A	
35	517.17	B2	A	0.19	A	
36	534.06	B1	A	0.64	A	
37	568.92	A1	A	14.11	A	ν_{12}(C-C-C deform)
38	569.01	A2	I	0	A	
39	591.47	B1	A	4.40	A	
40	604.89	B2	A	0.08	A	
41	801.33	B2	A	21.14	A	ν_{19}(CH_2 rock)
42	804.02	A1	A	25.83	A	
43	811.27	B1	A	22.74	A	
44	814.37	A2	I	0	A	
45	835.25	B1	A	0	A	ν_{11}(C-C stretch)
46	836.33	A2	I	0	A	

47	840.26	B2	A	1.68	A	
48	843.24	A1	A	1.96	A	
49	1004.63	A2	I	0	A	ν_{10}(C-C stretch)
50	1006.39	A1	A	0.05	A	
51	1010.38	B1	A	0.23	A	
52	1014.03	B2	A	13.67	A	
53	1092.66	A1	A	27.98	A	ν_9 (CH_3 rock)
54	1093.35	B2	A	4.15	A	
55	1097.02	B1	A	3.35	A	
56	1099.11	A2	I	0	A	
57	1119.25	B1	A	8.56	A	ν_{18}(CH_2 rock)
58	1124.38	B2	A	6.08	A	
59	1126.66	A2	I	0	A	
60	1129.41	A1	A	0.79	A	
61	1295.89	A2	I	0	A	ν_{17}(CH_2 twist)
62	1298.02	A1	A	1.55	A	
63	1305.86	B1	A	1.54	A	
64	1306.42	B2	A	1.93	A	
65	1360.42	A1	A	14.64	A	ν_8(CH_2 wag)
66	1364.14	B2	A	26.28	A	
67	1365.18	A2	I	0	A	
68	1366.40	B1	A	4.50	A	
69	1415.33	B2	A	2.46	A	ν_7(CH_3 sym. deform)
70	1416.49	B1	A	0	A	
71	1423.02	A1	A	0.32	A	
72	1432.02	A2	I	0	A	
73	1468.31	A2	I	0	A	ν_6(CH_2 bending)
74	1470.98	B2	A	13.94	A	
75	1476.87	A1	A	4.39	A	
76	1478.39	B1	A	53.21	A	
77	1511.12	B2	A	2.92	A	ν_5, ν_{16}(CH_3 deform)
78	1511.77	B1	A	3.85	A	
79	1516.09	B2	A	3.01	A	
80	1517.47	A2	I	0	A	
81	1518.21	B1	A	84.59	A	
82	1518.55	A1	A	138.95	A	
83	1578.86	A1	A	0.42	A	
84	1585.34	A2	I	0	A	
85	2344.90	A1	A	137.96	A	ν_4(CN stretch)
86	2346.42	B1	A	5.14	A	
87	2346.94	B2	A	87.31	A	
88	2347.30	A2	I	0	A	
89	3033.38	A2	I	0	A	ν_3(CH_2 sym. stretch)
90	3034.36	B1	A	5.72	A	
91	3035.00	B2	A	13.27	A	
92	3035.11	A1	A	2.78	A	
93	3040.84	A1	A	3.12	A	ν_2(CH_3 sym. stretch)
94	3041.86	A2	I	0	A	
95	3042.05	B2	A	27.07	A	

96	3042.37	B1	A	8.21	A	
97	3095.77	A1	A	5.44	A	ν_{15}(CH_2 asym. stretch)
98	3096.63	B2	A	13.59	A	
99	3097.50	B1	A	16.20	A	
100	3098.43	A2	I	0	A	
101	3143.17	B1	A	26.80	A	ν_{14}(CH_3 asym. stretch)
102	3143.91	B2	A	26.04	A	
103	3150.21	A2	I	0	A	
104	3150.60	A1	A	0.73	A	
105	3173.78	A1	A	54.49	A	ν_1(CH_3 asym. stretch)
106	3173.86	A2	I	0	A	
107	3181.81	B2	A	4.01	A	
108	3182.03	B1	A	0.07	A	

Table S-4: C_2H_5CN ($P2_1$) CRYSTAL14 Output File Data: B3LYP-D3/m-6-311G(d)

Mode	Frequencies	Irrep	Infrared	Intensity	Raman	Assignment
	(cm^{-1})			(km/mol)		
1	0.00	B	A	0	A	3x Translation
2	0.00	B	A	0	A	
3	0.00	A	A	0.01	A	
4	26.48	A	A	0.07	A	Lattice
5	38.64	A	A	0.08	A	
6	47.56	A	A	0.82	A	
7	54.05	B	A	0.45	A	
8	56.72	B	A	8.33	A	
9	80.42	B	A	4.50	A	
10	80.79	A	A	0.01	A	
11	105.43	B	A	1.06	A	
12	138.25	A	A	48.84	A	
13	218.87	A	A	8.69	A	ν_{13}, ν_{21}(C-C≡N bend)
14	225.07	B	A	10.60	A	
15	249.21	B	A	1.56	A	
16	252.32	A	A	1.05	A	
17	409.21	A	A	2.03	A	ν_{20}(C-C≡N bend)
18	411.29	B	A	1.56	A	
19	563.35	A	A	1.44	A	ν_{12}(C-C-C deform)
20	565.63	B	A	6.07	A	
21	794.11	A	A	6.93	A	ν_{19}(CH_2 rock)
22	795.69	B	A	3.51	A	
23	833.29	B	A	3.33	A	ν_{11}(C-C stretch)
24	836.00	A	A	0.14	A	
25	992.37	A	A	0.53	A	ν_{10}(C-C stretch)
26	995.11	B	A	1.17	A	
27	1097.70	A	A	2.03	A	ν_9 (CH_3 rock)
28	1097.98	B	A	7.27	A	
29	1125.80	A	A	0.02	A	ν_{18}(CH_2 rock)
30	1126.86	B	A	1.82	A	
31	1291.37	B	A	3.45	A	ν_{17}(CH_2 twist)

32	1291.46	A	A	2.61	A	
33	1350.70	A	A	6.89	A	ν_8(CH_2 wag)
34	1354.76	B	A	0.33	A	
35	1431.67	A	A	0.05	A	ν_7(CH_3 sym. deform)
36	1432.99	B	A	0.51	A	
37	1477.02	A	A	1.77	A	ν_6(CH_2 bending)
38	1477.81	B	A	7.97	A	
39	1509.29	A	A	9.18	A	ν_5, ν_{16}(CH_3 deform)
40	1511.37	B	A	2.48	A	
41	1516.70	A	A	0.02	A	
42	1522.73	B	A	19.50	A	
43	2348.31	A	A	1.51	A	ν_4(CN stretch)
44	2350.20	B	A	78.82	A	
45	3013.17	B	A	11.98	A	ν_3(CH_2 sym. stretch)
46	3014.49	A	A	1.43	A	
47	3024.85	A	A	2.25	A	ν_2(CH_3 sym. stretch)
48	3025.02	B	A	15.17	A	
49	3077.62	A	A	16.25	A	ν_{15}(CH_2 asym. stretch)
50	3078.60	B	A	0.33	A	
51	3101.29	A	A	0	A	ν_{14}(CH_3 asym. stretch)
52	3101.67	B	A	13.58	A	
53	3108.08	A	A	1.39	A	ν_1(CH_3 asym. stretch)
54	3108.90	B	A	24.50	A	

Table S-5: HCN (parallel) CRYSTAL14 Output File Data: B3LYP-D3/m-6-311G(d)

Mode	Frequencies	Symmetry	Infrared	Intensity	Raman	Assignment
	(cm^{-1})			(km/mol)		
1	0.00	A	A	0	A	3x Translation
2	0.00	A	A	0	A	
3	0.00	A	A	0	A	
4	132.80	A	A	0.02	A	Lattice T
5	139.54	A	A	0	A	T
6	195.15	A	A	3.07	A	L
7	195.35	A	A	0	A	T
8	205.79	A	A	104.78	A	L
9	237.73	A	A	115.83	A	L
10	257.73	A	A	0.55	A	L
11	904.59	A	A	0.02	A	v_2(HCN bend)
12	905.26	A	A	79.50	A	
13	919.68	A	A	0.12	A	
14	920.44	A	A	90.97	A	
15	2200.77	A	A	700.24	A	v_3(CN stretch)
16	2210.41	A	A	51.29	A	
17	3157.70	A	A	1151.89	A	v_1(CH stretch)
18	3183.15	A	A	796.20	A	

Table S-6: HCN (antiparallel) CRYSTAL14 Output File Data: B3LYP-D3/m-6-311G(d)

Mode	Frequencies	Symmetry	Infrared	Intensity	Raman	Assignment
	(cm^{-1})			(km/mol)		
1	0.00	A	A	0	A	3x Translation
2	0.00	A	A	0	A	
3	0.00	A	A	0	A	
4	76.01	A	A	0.08	A	Lattice
5	123.25	A	A	0	A	
6	175.22	A	A	85.42	A	
7	181.26	A	A	1.98	A	
8	183.41	A	A	91.14	A	
9	224.15	A	A	0.05	A	
10	306.95	A	A	0.01	A	
11	952.36	A	A	0.01	A	v_2(HCN bend)
12	958.70	A	A	92.61	A	
13	962.55	A	A	0.24	A	
14	964.86	A	A	102.21	A	
15	2194.81	A	A	31.83	A	v_3(CN stretch)
16	2199.99	A	A	996.87	A	
17	3042.79	A	A	2200.75	A	v_1(CH stretch)
18	3044.62	A	A	507.42	A	

Table S-7: C_2H_3CN (*P*nma) CRYSTAL14 Output File Data: B3LYP-D3/m-6-311G(d)

Mode	Frequencies	Irrep	Infrared	Intensity	Raman	Assignment
	(cm^{-1})			(km/mol)		
1	0.00	B2	A	0	A	3x Translation
2	0.00	B3	A	0	A	
3	0.00	B1	A	0.01	A	
4	8.05	B2	A	5.27	A	Lattice
5	15.72	B3	A	0.32	A	
6	26.94	A	I	0	A	
7	43.94	A	I	0	A	
8	51.65	B1	A	0	A	
9	65.35	A	I	0	A	
10	68.61	B2	A	0	A	
11	75.27	B1	A	0.92	A	
12	84.97	B3	A	0.06	A	
13	91.99	A	I	0	A	
14	101.18	B1	A	0.09	A	
15	105.20	B3	A	0.06	A	
16	107.30	B2	A	43.10	A	
17	113.87	A	I	0	A	
18	126.34	B2	A	6.46	A	
19	128.46	B3	A	51.37	A	
20	149.57	B1	A	0.82	A	
21	150.30	B3	A	4.35	A	
22	158.65	B1	A	0	A	
23	168.73	A	I	0	A	
24	169.24	B2	A	20.63	A	
25	252.12	A	I	0	A	ν_{15}(CCN in-plane bend)
26	257.15	B1	A	6.51	A	
27	257.42	B3	A	60.14	A	
28	259.53	B2	A	1.22	A	
29	378.88	B3	A	0.06	A	ν_{14}(CCN out-plane bend)
30	380.59	A	I	0	A	
31	384.55	B2	A	2.30	A	
32	387.03	B1	A	3.32	A	
33	601.58	B2	A	0.01	A	ν_{13}(C-C=C bend)
34	602.60	B1	A	1.20	A	
35	607.81	A	I	0	A	
36	607.85	B3	A	0.74	A	
37	721.94	B2	A	125.59	A	ν_{12}(C=C torsion)
38	722.17	A	I	0	A	
39	724.63	B3	A	5.12	A	
40	729.38	B1	A	7.87	A	
41	888.20	B1	A	14.95	A	ν_{10}(CH_2 wag)
42	888.62	B3	A	1.89	A	
43	888.84	A	I	0	A	
44	889.10	B2	A	0.07	A	
45	1015.34	B2	A	269.54	A	ν_{11}(C-C stretch)
46	1015.46	A	I	0	A	

47	1018.59	B1	A	19.31	A	
48	1025.34	B3	A	1.57	A	
49	1047.25	A	I	0	A	ν_9(CH_2 wag)
50	1047.37	B2	A	119.28	A	
51	1051.65	B1	A	18.32	A	
52	1052.33	B3	A	10.85	A	
53	1110.63	B3	A	1.08	A	ν_8(CH_2 rock)
54	1111.71	A	I	0	A	
55	1111.97	B1	A	5.94	A	
56	1115.20	B2	A	1.29	A	
57	1319.63	A	I	0	A	ν_7(CH rock)
58	1322.63	B1	A	15.13	A	
59	1324.29	B3	A	4.30	A	
60	1329.62	B2	A	0.01	A	
61	1461.50	B1	A	78.13	A	ν_6(CH_2 deform)
62	1462.82	A	I	0	A	
63	1467.24	B2	A	5.36	A	
64	1468.81	B3	A	29.43	A	
65	1670.59	A	I	0	A	ν_5(C=C stretch)
66	1671.43	B1	A	27.83	A	
67	1672.98	B3	A	1.36	A	
68	1674.63	B2	A	0.10	A	
69	2325.30	B1	A	482.42	A	ν_4(CN stretch)
70	2325.88	A	I	0	A	
71	2326.03	B3	A	0.19	A	
72	2331.23	B2	A	0.91	A	
73	3137.45	B3	A	9.86	A	ν_3(CH sym. stretch)
74	3138.19	A	I	0	A	
75	3138.53	B2	A	3.91	A	
76	3138.65	B1	A	175.15	A	
77	3215.51	B1	A	2.21	A	ν_2(CH sym. stretch)
78	3216.03	B2	A	3.02	A	
79	3216.55	B3	A	37.39	A	
80	3216.70	A	I	0	A	
81	3241.49	A	I	0	A	ν_1(CH asym. stretch)
82	3242.13	B1	A	39.79	A	
83	3242.39	B2	A	7.38	A	
84	3242.64	B3	A	19.50	A	

Table S-8: HC_3N CRYSTAL14 Output File Data: B3LYP-D3/m-6-311G(d)

Mode	Frequencies	Irrep	Infrared	Intensity	Raman	Assignment
	(cm^{-1})			(km/mol)		
1	0.00	Bu	A	0	I	3x Translation
2	0.00	Au	A	0	I	
3	0.00	Bu	A	0	I	
4	20.34	Ag	I	0	A	Lattice
5	43.79	Ag	I	0	A	
6	62.95	Bg	I	0	A	
7	85.60	Bu	A	17.27	I	
8	94.70	Ag	I	0	A	
9	99.52	Au	A	19.31	I	
10	114.01	Bg	I	0	A	
11	250.09	Au	A	1.01	I	ν_7(C-C≡C bend)
12	253.78	Bu	A	1.99	I	
13	253.91	Ag	I	0	A	
14	266.80	Bg	I	0	A	
15	583.61	Au	A	36.33	I	ν_6(C-C≡N bend)
16	588.47	Bu	A	35.13	I	
17	590.21	Ag	I	0	A	
18	595.82	Bg	I	0	A	
19	772.31	Au	A	118.33	I	ν_5(H-C≡C bend)
20	778.89	Bg	I	0	A	
21	790.50	Bu	A	98.21	I	
22	790.62	Ag	I	0	A	
23	914.30	Bu	A	21.25	I	ν_4(C-C stretch)
24	916.61	Ag	I	0	A	
25	2158.35	Bu	A	157.19	I	ν_3(C≡C stretch)
26	2158.71	Ag	I	0	A	
27	2368.07	Bu	A	729.64	I	ν_2(CN stretch)
28	2372.96	Ag	I	0	A	
29	3309.00	Bu	A	1606.84	I	ν_1(CH stretch)
30	3309.81	Ag	I	0	A	

Table S-9: C_2N_2 CRYSTAL14 Output File Data: B3LYP-D3/m-6-311G(d)

Mode	Frequencies	Irrep	Infrared	Intensity	Raman	Assignment
	(cm^{-1})			(km/mol)		
1	0.00	B3u	A	0	I	3x Translation
2	0.00	B1u	A	0	I	
3	0.00	B2u	A	0.02	I	
4	37.73	B1u	A	0.03	I	Lattice
5	48.37	B2g	I	0	A	
6	51.51	Ag	I	0	A	
7	56.30	Au	I	0	I	
8	59.91	B3u	A	0.91	I	
9	71.03	Ag	I	0	A	
10	71.56	Au	I	0	I	
11	84.48	B3g	I	0	A	
12	86.86	B2u	A	0.64	I	
13	98.87	B3u	A	0.58	I	
14	100.78	B1g	I	0	A	
15	103.40	B1u	A	1.67	I	
16	104.89	B2u	A	0.95	I	
17	107.50	Au	I	0	I	
18	113.51	B1g	I	0	A	
19	115.70	B3g	I	0	A	
20	120.02	B2g	I	0	A	
21	252.37	B1u	A	172.53	I	v_5(NCCN asym. bend)
22	254.28	B3u	A	152.73	I	
23	256.59	B2u	A	85.98	I	
24	258.54	Au	I	0	I	
25	260.47	B1u	A	0.16	I	
26	260.91	Au	I	0	I	
27	267.83	B2u	A	51.76	I	
28	276.79	B3u	A	8.15	I	
29	563.91	B3g	I	0	A	v_2(NCCN sym. bend)
30	565.07	B1g	I	0	A	
31	566.07	Ag	I	0	A	
32	566.56	B2g	I	0	A	
33	568.74	B1g	I	0	A	
34	570.13	Ag	I	0	A	
35	570.16	B3g	I	0	A	
36	571.35	B2g	I	0	A	
37	877.71	Ag	I	0	A	v_1(C-C sym. stretch)
38	878.04	B2g	I	0	A	
39	878.54	B1g	I	0	A	
40	878.89	B3g	I	0	A	
41	2279.78	B3u	A	8.20	I	v_3(C≡N asym. stretch)
42	2279.90	B2u	A	17.87	I	
43	2281.83	Au	I	0	I	
44	2282.44	B1u	A	0.75	I	
45	2449.81	Ag	I	0	A	
46	2450.81	B2g	I	0	A	

47	2451.49	B1g	I	0	A	
48	2451.84	B3g	I	0	A	