

Supplementary Material to

Millimeter- and Submillimeter Wave Spectroscopy of ^{13}C and ^{15}N Isotopomers of Cyanoacetylen, HCCCN, in the Ground and Vibrationally Excited States

Sven Thorwirth, Holger S. P. Müller, and Gisbert Winnewisser

I. Physikalisches Institut, Universität zu Köln, Zùlpicher Str. 77, 50937 Köln, Germany

Table S1 Measured Transition Frequencies (MHz) of HCC¹³CN and Residuals (kHz).

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
(0, 0, 0, 0)							
14 – 13		126 839.589 (10)	–3	50 – 49		452 749.041 (10)	4
15 – 14		135 898.628 (10)	7	51 – 50		461 792.862 (10)	–2
16 – 15		144 957.454 (10)	–1	57 – 56		516 041.468 (10)	0
17 – 16		154 016.078 (10)	–2	58 – 57		525 080.387 (10)	0
18 – 17		163 074.479 (10)	–5	65 – 64		588 331.005 (15)	–11
19 – 18		172 132.657 (10)	2	66 – 65		597 363.528 (15)	–21
39 – 38		353 226.937 (10)	6	67 – 66		606 395.235 (10)	9
40 – 39		362 277.192 (10)	5	68 – 67		615 426.048 (20)	15
41 – 40		371 326.919 (10)	–6				
(0, 0, 0, 1)							
15 – 14	1e	136 226.214 (10)	1	50 – 49	1e	453 834.067 (20)	8
15 – 14	1f	136 421.848 (10)	0	50 – 49	1f	454 478.946 (15)	3
16 – 15	1e	145 306.857 (10)	2	51 – 50	1e	462 899.276 (15)	3
16 – 15	1f	145 515.502 (10)	0	51 – 50	1f	463 556.738 (15)	5
17 – 16	1e	—		57 – 56	1e	517 275.809 (15)	5
17 – 16	1f	154 608.933 (10)	–1	57 – 56	1f	518 008.313 (15)	9
18 – 17	1e	163 467.486 (10)	1	58 – 57	1e	526 335.973 (15)	–3
18 – 17	1f	163 702.130 (10)	–3	58 – 57	1f	527 080.886 (15)	–25
19 – 18	1e	172 547.445 (10)	–2	64 – 63	1e	580 680.272 (15)	–18
19 – 18	1f	172 795.082 (10)	–3	64 – 63	1f	581 499.401 (15)	14
39 – 38	1e	354 075.574 (20)	1	65 – 64	1e	589 734.778 (15)	12
39 – 38	1f	354 580.997 (15)	4	65 – 64	1f	590 566.138 (15)	–7
40 – 39	1e	363 147.392 (15)	–5	66 – 65	1e	598 788.362 (15)	–14
40 – 39	1f	363 665.576 (15)	–1	66 – 65	1f	599 632.010 (15)	–3
41 – 40	1e	—		67 – 66	1e	607 841.120 (15)	14
41 – 40	1f	372 749.609 (15)	–1	67 – 66	1f	608 696.982 (15)	4
(0, 0, 0, 2)							
13 – 12	0e	118 512.117 (15)	6	41 – 40	0e	373 470.937 (15)	–4
13 – 12	2f	118 516.319 (15)	1	41 – 40	2f	373 636.238 (15)	1
13 – 12	2e	118 522.058 (15)	–2	41 – 40	2e	373 806.361 (15)	0
14 – 13	0e	127 626.546 (15)	1	50 – 49	0e	455 273.719 (15)	–5
14 – 13	2f	127 632.063 (15)	–4	50 – 49	2f	455 557.588 (15)	–3
14 – 13	2e	127 639.236 (15)	–7	50 – 49	2e	455 847.343 (15)	7
15 – 14	0e	136 740.565 (15)	3	51 – 50	0e	464 357.820 (15)	–5
15 – 14	2f	136 747.614 (15)	–4	51 – 50	2f	464 656.629 (15)	2

Table S1—Continued

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
15 – 14	2e	136 756.440 (15)	–6	51 – 50	2e	464 961.430 (15)	7
16 – 15	0e	145 854.138 (15)	5	58 – 57	0e	527 918.308 (15)	–2
16 – 15	2f	145 862.956 (15)	–1	58 – 57	2f	528 328.966 (15)	–1
16 – 15	2e	145 873.668 (15)	–4	58 – 57	2e	528 746.431 (15)	2
17 – 16	0e	154 967.234 (15)	6	64 – 63	0e	582 360.235 (20)	6
17 – 16	2f	154 978.066 (15)	–4	64 – 63	2f	582 873.598 (20)	7
17 – 16	2e	154 990.915 (15)	–5	64 – 63	2e	583 394.445 (20)	–4
18 – 17	0e	164 079.827 (15)	8	65 – 64	0e	591 430.499 (20)	6
18 – 17	2f	164 092.942 (15)	–1	65 – 64	2f	591 961.294 (20)	3
18 – 17	2e	164 108.189 (15)	–3	65 – 64	2e	592 499.687 (20)	–13
19 – 18	0e	173 191.881 (15)	3	66 – 65	0e	600 499.797 (20)	0
19 – 18	2f	173 207.554 (15)	–7	66 – 65	2f	601 048.086 (20)	8
19 – 18	2e	173 225.483 (15)	–6	66 – 65	2e	601 604.079 (20)	–6
39 – 38	0e	355 281.428 (15)	–4	67 – 66	0e	609 568.149 (20)	9
39 – 38	2f	355 424.767 (15)	0	67 – 66	2f	610 133.929 (20)	–7
39 – 38	2e	355 572.702 (15)	4	67 – 66	2e	610 707.567 (20)	–6
40 – 39	0e	364 376.679 (15)	–3				
40 – 39	2f	364 530.777 (15)	–7				
40 – 39	2e	364 689.601 (15)	3				
(0, 0, 1, 0)							
14 – 13	1e	127 040.334 (10)	–4	51 – 50	1e	462 519.964 (15)	–3
14 – 13	1f	127 141.896 (10)	–2	51 – 50	1f	462 888.866 (15)	–8
15 – 14	1e	136 113.689 (10)	–2	57 – 56	1e	516 852.858 (20)	–9
15 – 14	1f	136 222.501 (10)	0	57 – 56	1f	517 264.845 (20)	–11
16 – 15	1e	145 186.842 (10)	–3	58 – 57	1e	525 905.787 (20)	–10
16 – 15	1f	145 302.904 (10)	0	58 – 57	1f	526 324.948 (20)	–8
18 – 17	1e	163 332.505 (10)	–2	64 – 63	1e	580 206.878 (15)	12
18 – 17	1f	163 463.066 (10)	3	64 – 63	1f	580 668.990 (15)	9
19 – 18	1e	172 404.983 (10)	–5	65 – 64	1e	589 254.166 (15)	–3
19 – 18	1f	172 542.788 (10)	–3	65 – 64	1f	589 723.435 (15)	3
40 – 39	1e	362 848.811 (15)	–13	66 – 65	1e	598 300.614 (15)	–3
40 – 39	1f	363 138.508 (15)	0	66 – 65	1f	598 777.026 (15)	2
41 – 40	1e	371 912.773 (15)	34	67 – 66	1e	607 346.193 (15)	–1
41 – 40	1f	372 209.643 (15)	6	67 – 66	1f	607 829.737 (15)	–6
50 – 49	1e	453 462.052 (15)	–2				
50 – 49	1f	453 823.773 (15)	2				

Table S1—Continued

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
(0, 0, 1, 1)							
13 – 12	0e	118 375.360 (10)	1	60 – 59	0e	545 443.105 (15)	6
13 – 12	0f	118 378.777 (10)	3	60 – 59	0f	545 761.448 (15)	–7
13 – 12	2f	118 392.321 (10)	5	60 – 59	2f	546 083.642 (15)	–8
13 – 12	2e	118 394.956 (10)	0	60 – 59	2e	546 397.952 (20)	12
14 – 13	0e	127 478.935 (10)	–4	61 – 60	0e	554 511.177 (15)	–5
14 – 13	0f	127 483.092 (10)	–4	61 – 60	0f	554 840.228 (15)	0
14 – 13	2f	127 499.419 (10)	1	61 – 60	2f	555 168.462 (15)	–4
14 – 13	2e	127 502.739 (10)	–1	61 – 60	2e	555 493.364 (20)	3
15 – 14	0e	136 582.028 (10)	–1	62 – 61	0e	563 578.518 (15)	4
15 – 14	0f	136 587.046 (15)	–1	62 – 61	0f	563 918.208 (20)	15
15 – 14	2f	136 606.507 (10)	1	62 – 61	2f	564 252.419 (15)	5
15 – 14	2e	136 610.627 (10)	–1	62 – 61	2e	564 587.858 (15)	2
16 – 15	0e	145 684.597 (10)	–1	65 – 64	0e	590 775.904 (20)	–8
16 – 15	0f	145 690.608 (10)	1	65 – 64	0f	591 147.063 (25)	–22
16 – 15	2f	145 713.573 (10)	1	65 – 64	2f	591 498.960 (15)	20
16 – 15	2e	145 718.624 (10)	0	65 – 64	2e	591 865.588 (20)	–23
17 – 16	0e	154 786.612 (10)	0	66 – 65	0e	599 840.143 (15)	–3
17 – 16	0f	154 793.758 (15)	1	66 – 65	0f	—	
17 – 16	2f	154 820.607 (10)	1	66 – 65	2f	600 579.276 (15)	–22
17 – 16	2e	154 826.732 (10)	0	66 – 65	2e	600 956.240 (20)	11
18 – 17	0e	163 888.044 (10)	1	67 – 66	0e	608 903.582 (15)	5
18 – 17	0f	163 896.483 (15)	1	67 – 66	0f	609 295.375 (30)	6
18 – 17	2f	163 927.599 (10)	0	67 – 66	2f	609 658.737 (20)	10
18 – 17	2e	163 934.956 (10)	–1	67 – 66	2e	610 045.835 (15)	–2
19 – 18	0e	172 988.860 (10)	2	68 – 67	0e	617 966.191 (20)	–2
19 – 18	0f	172 998.768 (10)	1	68 – 67	0f	618 368.211 (50)	38
19 – 18	2f	173 034.531 (10)	–2	68 – 67	2f	618 737.217 (20)	3
19 – 18	2e	173 043.296 (10)	–3	68 – 67	2e	619 134.423 (20)	1
(0, 1, 0, 0)							
14 – 13	1e	126 847.899 (15)	–7	50 – 49	1e	452 782.570 (20)	2
14 – 13	1f	126 918.457 (15)	–5	50 – 49	1f	—	
15 – 14	1e	135 907.526 (15)	–2	55 – 54	1e	498 056.202 (30)	16
15 – 14	1f	135 983.123 (15)	0	55 – 54	1f	—	
16 – 15	1e	144 966.954 (15)	–1	56 – 55	1e	505 067.089 (50)	8
16 – 15	1f	145 047.588 (15)	–1	56 – 55	1f	507 320.692 (30)	7
17 – 16	1e	154 026.170 (15)	–5	57 – 56	1e	517 596.064 (30)	–4
17 – 16	1f	154 111.846 (15)	–1	57 – 56	1f	—	

Table S1—Continued

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
18 – 17	1e	163 085.172 (15)	–1	58 – 57	1e	525 281.089 (30)	17
18 – 17	1f	—		58 – 57	1f	—	
19 – 18	1e	172 143.937 (15)	–1	59 – 58	1e	534 199.337 (30)	–5
19 – 18	1f	172 239.688 (15)	0	59 – 58	1f	—	
41 – 40	1e	371 351.671 (20)	4	60 – 59	1e	543 212.131 (30)	2
41 – 40	1f	371 559.667 (20)	1	60 – 59	1f	—	
42 – 41	1e	380 401.562 (20)	1	61 – 60	1e	552 240.115 (30)	6
42 – 41	1f	380 615.192 (20)	10	61 – 60	1f	552 535.430 (30)	8
43 – 42	1e	389 450.935 (20)	0	66 – 65	1e	597 403.947 (30)	–32
43 – 42	1f	389 670.483 (20)	–13	66 – 65	1f	597 733.115 (30)	–20
46 – 45	1e	—		67 – 66	1e	606 435.719 (30)	–6
46 – 45	1f	416 839.747 (20)	–3	67 – 66	1f	606 770.255 (30)	23
47 – 46	1e	—		68 – 67	1e	615 466.720 (30)	–3
47 – 46	1f	425 904.036 (20)	6	68 – 67	1f	615 806.461 (30)	–10
48 – 47	1e	—					
48 – 47	1f	434 994.773 (20)	–1				
(0, 0, 0, 3)							
13 – 12	1e	118 712.128 (15)	8	49 – 48	1e	—	
13 – 12	3e	118 883.939 (15)	–10	49 – 48	3e	—	
13 – 12	3f	118 883.939 (15)	–10	49 – 48	3f	447 962.221 (20)	27
13 – 12	1f	119 053.599 (15)	4	49 – 48	1f	—	
14 – 13	1e	127 842.553 (15)	4	50 – 49	1e	456 209.453 (20)	–7
14 – 13	3e	128 028.309 (15)	–10	50 – 49	3e	457 053.248 (20)	–2
14 – 13	3f	128 028.309 (15)	–10	50 – 49	3f	457 098.761 (20)	17
14 – 13	1f	128 210.220 (15)	1	50 – 49	1f	—	
15 – 14	1e	136 972.705 (15)	8	55 – 54	1e	501 685.959 (30)	–15
15 – 14	3e	137 172.554 (15)	–12	55 – 54	3e	—	
15 – 14	3f	137 172.554 (15)	–12	55 – 54	3f	—	
15 – 14	1f	137 366.544 (15)	2	55 – 54	1f	—	
16 – 15	1e	146 102.555 (15)	9	56 – 55	1e	512 820.351 (50)	5
16 – 15	3e	—		56 – 55	3e	511 833.917 (30)	3
16 – 15	3f	—		56 – 55	3f	511 912.149 (30)	24
16 – 15	1f	146 522.547 (15)	5	56 – 55	1f	512 219.005 (30)	5
17 – 16	1e	155 232.083 (15)	8	57 – 56	1e	518 434.990 (30)	5
17 – 16	3e	155 460.650 (15)	–10	57 – 56	3e	—	
17 – 16	3f	155 460.650 (15)	–10	57 – 56	3f	—	
17 – 16	1f	155 678.200 (15)	4	57 – 56	1f	—	
18 – 17	1e	164 361.276 (15)	10	58 – 57	1e	528 891.941 (30)	1
18 – 17	3e	164 604.478 (15)	–10	58 – 57	3e	—	
18 – 17	3f	164 604.478 (15)	–10	58 – 57	3f	—	

Table S1—Continued

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
18 – 17	1 <i>f</i>	164 833.489 (15)	6	58 – 57	1 <i>f</i>	—	
19 – 18	1 <i>e</i>	—		59 – 58	1 <i>e</i>	538 113.950 (30)	13
19 – 18	3 <i>e</i>	173 748.012 (50)	40	59 – 58	3 <i>e</i>	—	
19 – 18	3 <i>f</i>	173 748.282 (50)	–65	59 – 58	3 <i>f</i>	—	
19 – 18	1 <i>f</i>	173 988.384 (15)	4	59 – 58	1 <i>f</i>	—	
41 – 40	1 <i>e</i>	374 197.966 (20)	–9	60 – 59	1 <i>e</i>	547 239.719 (30)	14
41 – 40	3 <i>e</i>	374 842.700 (20)	–2	60 – 59	3 <i>e</i>	—	
41 – 40	3 <i>f</i>	374 859.979 (20)	–4	60 – 59	3 <i>f</i>	—	
41 – 40	1 <i>f</i>	375 247.793 (25)	–35	60 – 59	1 <i>f</i>	—	
42 – 41	1 <i>e</i>	383 313.756 (20)	–1	61 – 60	1 <i>e</i>	556 348.538 (30)	–4
42 – 41	3 <i>e</i>	383 979.238 (20)	–6	61 – 60	3 <i>e</i>	557 465.194 (30)	–4
42 – 41	3 <i>f</i>	383 998.696 (20)	–1	61 – 60	3 <i>f</i>	557 581.915 (30)	20
42 – 41	1 <i>f</i>	384 386.431 (20)	–4	61 – 60	1 <i>f</i>	557 816.664 (30)	–1
43 – 42	1 <i>e</i>	392 428.764 (20)	–3	66 – 65	1 <i>e</i>	601 841.861 (30)	–13
43 – 42	3 <i>e</i>	393 115.304 (20)	–9	66 – 65	3 <i>e</i>	603 076.582 (30)	4
43 – 42	3 <i>f</i>	393 137.158 (20)	11	66 – 65	3 <i>f</i>	603 243.680 (30)	6
43 – 42	1 <i>f</i>	393 523.707 (20)	–2	66 – 65	1 <i>f</i>	603 374.235 (30)	23
46 – 45	1 <i>e</i>	—		67 – 66	1 <i>e</i>	610 936.057 (30)	29
46 – 45	3 <i>e</i>	—		67 – 66	3 <i>e</i>	612 196.245 (30)	–21
46 – 45	3 <i>f</i>	—		67 – 66	3 <i>f</i>	612 374.962 (30)	–15
46 – 45	1 <i>f</i>	420 922.601 (20)	–5	67 – 66	1 <i>f</i>	612 480.946 (30)	5
47 – 46	1 <i>e</i>	—		68 – 67	1 <i>e</i>	620 029.019 (30)	–11
47 – 46	3 <i>e</i>	—		68 – 67	3 <i>e</i>	621 315.033 (30)	–15
47 – 46	3 <i>f</i>	—		68 – 67	3 <i>f</i>	621 505.875 (30)	–25
47 – 46	1 <i>f</i>	430 044.364 (20)	–6	68 – 67	1 <i>f</i>	621 586.065 (30)	16
48 – 47	1 <i>e</i>	—					
48 – 47	3 <i>e</i>	—					
48 – 47	3 <i>f</i>	—					
48 – 47	1 <i>f</i>	439 137.929 (20)	–3				
Cross-ladder transitions : (0, 0, 0, 3) $\leftarrow$ (0, 1, 0, 0)							
49 – 48	1 <i>f</i>	444 332.790 (20)	–7	57 – 56	1 <i>e</i>	520 700.724 (30)	–11
56 – 55	1 <i>e</i>	507 332.843 (30)	12	58 – 57	1 <i>e</i>	531 996.609 (30)	3
Cross-ladder transitions: (0, 1, 0, 0) $\leftarrow$ (0, 0, 0, 3)							
49 – 48	1 <i>f</i>	447 982.429 (20)	–6	57 – 56	1 <i>e</i>	515 330.312 (30)	–6
56 – 55	1 <i>e</i>	510 554.583 (30)	–14	58 – 57	1 <i>e</i>	522 176.419 (30)	14

Table S1—Continued

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
(1, 0, 0, 0)							
14 – 13		126 667.275 (20)	–2	17 – 16		153 806.540 (20)	–2
15 – 14		135 713.917 (20)	1	18 – 17		162 852.503 (20)	1
16 – 15		144 760.343 (20)	1	19 – 18		171 898.209 (20)	0
(1, 0, 0, 1)							
14 – 13	1e	126 984.858 (25)	–2	17 – 16	1e	154 191.919 (25)	–1
14 – 13	1f	127 172.340 (25)	1	17 – 16	1f	154 419.351 (25)	–4
15 – 14	1e	136 054.115 (25)	3	18 – 17	1e	163 260.447 (25)	2
15 – 14	1f	136 254.923 (25)	1	18 – 17	1f	163 501.173 (25)	0
16 – 15	1e	145 123.136 (25)	–1	19 – 18	1e	172 328.698 (25)	–1
16 – 15	1f	145 337.266 (25)	0	19 – 18	1f	172 582.705 (25)	2

Table S2. Measured Transition Frequencies (MHz) of HC¹³CCN and Residuals (kHz).

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
(0, 0, 0, 0)							
14 – 13		126 827.349 (10)	–3	50 – 49		452 705.425 (20)	5
15 – 14		135 885.508 (10)	1	51 – 50		461 748.386 (20)	5
16 – 15		144 943.467 (10)	0	57 – 56		515 991.775 (20)	–9
17 – 16		154 001.217 (10)	–1	58 – 57		525 029.839 (20)	0
18 – 17		163 058.742 (10)	–7	65 – 64		588 274.408 (20)	–12
19 – 18		172 116.047 (10)	1	66 – 65		597 306.086 (20)	–5
39 – 38		353 192.876 (20)	–1	67 – 66		606 336.916 (20)	11
40 – 39		362 242.262 (20)	–1	68 – 67		615 366.855 (20)	4
41 – 40		371 291.139 (20)	9				
(0, 0, 0, 1)							
13 – 12	1e	—		50 – 49	1e	453 771.452 (20)	26
13 – 12	1f	118 218.811 (15)	0	50 – 49	1f	454 421.003 (20)	0
14 – 13	1e	127 127.753 (15)	–1	51 – 50	1e	462 835.392 (20)	–8
14 – 13	1f	127 311.695 (15)	–5	51 – 50	1f	463 497.659 (20)	14
15 – 14	1e	136 207.340 (15)	1	57 – 56	1e	—	
15 – 14	1f	136 404.396 (15)	–1	57 – 56	1f	517 942.352 (20)	9
16 – 15	1e	145 286.727 (15)	2	58 – 57	1e	526 263.428 (20)	–3
16 – 15	1f	145 496.886 (15)	–1	58 – 57	1f	527 013.769 (20)	–38
17 – 16	1e	154 365.897 (15)	0	64 – 63	1e	580 600.295 (30)	–40
17 – 16	1f	—		64 – 63	1f	581 425.465 (30)	22
18 – 17	1e	163 444.844 (15)	2	65 – 64	1e	589 653.596 (50)	18
18 – 17	1f	163 681.192 (15)	–3	65 – 64	1f	590 491.053 (20)	–11
19 – 18	1e	172 523.548 (15)	2	66 – 65	1e	598 705.957 (20)	1
19 – 18	1f	—		66 – 65	1f	599 555.812 (20)	16
39 – 38	1e	354 026.626 (20)	–5	67 – 66	1e	607 757.463 (20)	10
39 – 38	1f	354 535.725 (20)	4	67 – 66	1f	608 619.610 (30)	–16
40 – 39	1e	363 097.209 (20)	2	68 – 67	1e	616 808.056 (30)	–2
40 – 39	1f	363 619.160 (20)	11	68 – 67	1f	617 682.551 (35)	11
41 – 40	1e	372 167.241 (20)	–9				
41 – 40	1f	372 702.028 (20)	–1				
(0, 0, 0, 2)							
13 – 12	0e	118 491.746 (25)	3	41 – 40	0e	373 399.284 (20)	4
13 – 12	2f	118 496.439 (25)	0	41 – 40	2f	—	
13 – 12	2e	118 502.507 (25)	2	41 – 40	2e	—	
14 – 13	0e	127 604.558 (25)	2	50 – 49	0e	455 183.760 (20)	–11

Table S2—Continued

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
14 – 13	2f	127 610.656 (25)	–5	50 – 49	2f	455 481.659 (20)	11
14 – 13	2e	127 618.238 (25)	–3	50 – 49	2e	455 784.701 (20)	7
15 – 14	0e	136 716.940 (25)	0	51 – 50	0e	464 265.864 (50)	–5
15 – 14	2f	136 724.684 (25)	0	51 – 50	2f	464 579.182 (20)	–5
15 – 14	2e	136 734.006 (25)	–4	51 – 50	2e	464 897.772 (20)	0
16 – 15	0e	145 828.865 (25)	0	58 – 57	0e	527 812.760 (20)	–15
16 – 15	2f	145 838.501 (25)	4	58 – 57	2f	528 241.093 (20)	0
16 – 15	2e	145 849.808 (25)	–7	58 – 57	2e	528 675.368 (20)	7
17 – 16	0e	154 940.304 (25)	3	64 – 63	0e	582 243.852 (25)	7
17 – 16	2f	154 952.082 (25)	–2	64 – 63	2f	582 776.816 (60)	–16
17 – 16	2e	—		64 – 63	2e	583 316.341 (60)	0
18 – 17	0e	164 051.219 (25)	0	65 – 64	0e	591 312.360 (60)	–20
18 – 17	2f	164 065.430 (25)	–1	65 – 64	2f	591 863.075 (60)	18
18 – 17	2e	—		65 – 64	2e	592 420.340 (60)	–13
19 – 18	0e	173 161.591 (25)	2	66 – 65	0e	—	
19 – 18	2f	173 178.520 (25)	–3	66 – 65	2f	600 948.370 (25)	0
19 – 18	2e	173 197.452 (25)	–7	66 – 65	2e	601 523.414 (60)	–63
39 – 38	0e	355 213.833 (20)	12	67 – 66	0e	609 446.649 (25)	17
39 – 38	2f	355 365.421 (60)	58	67 – 66	2f	610 032.753 (25)	–4
39 – 38	2e	355 520.962 (20)	–9	67 – 66	2e	610 625.689 (25)	2
40 – 39	0e	364 307.050 (20)	0				
40 – 39	2f	364 469.871 (20)	1				
40 – 39	2e	364 636.854 (20)	–4				
(0, 0, 1, 0)							
14 – 13	1e	127 031.155 (15)	0	51 – 50	1e	462 486.448 (20)	10
14 – 13	1f	127 131.854 (15)	1	51 – 50	1f	462 852.245 (20)	4
15 – 14	1e	136 103.854 (15)	3	57 – 56	1e	516 815.359 (20)	–8
15 – 14	1f	136 211.738 (15)	–1	57 – 56	1f	517 223.888 (20)	–8
16 – 15	1e	145 176.356 (15)	6	58 – 57	1e	525 867.608 (20)	–26
16 – 15	1f	145 291.424 (15)	–2	58 – 57	1f	526 283.293 (20)	18
17 – 16	1e	154 248.636 (15)	–1	64 – 63	1e	580 164.755 (20)	36
17 – 16	1f	154 370.901 (15)	1	64 – 63	1f	580 622.963 (20)	–1
18 – 17	1e	163 320.701 (15)	2	65 – 64	1e	589 211.359 (20)	2
18 – 17	1f	163 450.148 (15)	–1	65 – 64	1f	589 676.684 (20)	–9
19 – 18	1e	172 392.525 (15)	1	66 – 65	1e	598 257.129 (20)	–10
19 – 18	1f	172 529.157 (15)	–2	66 – 65	1f	598 729.554 (20)	–7
40 – 39	1e	362 822.551 (20)	–2	67 – 66	1e	607 302.044 (20)	–6
40 – 39	1f	363 109.793 (20)	1	67 – 66	1f	607 781.559 (20)	4
41 – 40	1e	371 885.812 (20)	3	68 – 67	1e	616 346.079 (30)	–1
41 – 40	1f	372 180.204 (20)	2	68 – 67	1f	616 832.673 (30)	8

Table S2—Continued

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
50 – 49	1e	453 429.187 (20)	1				
50 – 49	1f	453 787.855 (20)	–3				
(0, 0, 1, 1)							
13 – 12	0e	118 360.786 (20)	6	17 – 16	0e	154 766.259 (20)	1
13 – 12	0f	118 366.098 (20)	7	17 – 16	0f	154 777.634 (30)	1
13 – 12	2f	118 377.835 (20)	–4	17 – 16	2f	154 801.205 (20)	–2
13 – 12	2e	118 382.508 (20)	–1	17 – 16	2e	154 811.745 (20)	1
14 – 13	0e	127 462.996 (20)	1	18 – 17	0e	163 866.104 (20)	–1
14 – 13	0f	127 469.519 (20)	–11	18 – 17	0f	163 879.533 (30)	2
14 – 13	2f	—		18 – 17	2f	163 906.930 (20)	–4
14 – 13	2e	127 489.577 (20)	–2	18 – 17	2e	163 919.480 (20)	7
15 – 14	0e	136 564.664 (20)	–5	19 – 18	0e	172 965.278 (20)	0
15 – 14	0f	136 572.605 (30)	–10	19 – 18	0f	172 980.999 (20)	0
15 – 14	2f	136 589.606 (20)	9	19 – 18	2f	173 012.588 (20)	–6
15 – 14	2e	136 596.794 (20)	–8	19 – 18	2e	173 027.380 (20)	1
16 – 15	0e	145 665.767 (20)	–2				
16 – 15	0f	145 675.329 (20)	7				
16 – 15	2f	145 695.438 (20)	14				
16 – 15	2e	145 704.183 (20)	–5				
(0, 1, 0, 0)							
14 – 13	1e	126 835.609 (20)	–11	17 – 16	1e	154 011.254 (20)	5
14 – 13	1f	126 906.224 (20)	0	17 – 16	1f	154 096.989 (20)	7
15 – 14	1e	135 894.363 (20)	0	18 – 17	1e	163 069.370 (20)	4
15 – 14	1f	135 970.014 (20)	5	18 – 17	1f	—	
16 – 15	1e	144 952.905 (20)	–6	19 – 18	1e	172 127.253 (20)	4
16 – 15	1f	145 033.587 (20)	–13	19 – 18	1f	172 223.070 (20)	2

Table S3. Measured Transition Frequencies (MHz) of HCCC¹⁵N and Residuals (kHz).

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
(0, 0, 0, 0)							
14 – 13		123 663.453 (15)	0	50 – 49		441 419.576 (30)	–9
15 – 14		132 495.664 (15)	–3	51 – 50		450 237.454 (15)	10
16 – 15		141 327.698 (15)	1	59 – 58		520 756.785 (15)	6
17 – 16		150 159.532 (15)	1	60 – 59		529 568.570 (15)	0
18 – 17		158 991.154 (15)	–2	65 – 64		573 616.264 (20)	18
19 – 18		167 822.563 (15)	4	66 – 65		582 423.437 (20)	–4
20 – 19		176 653.730 (15)	0	67 – 66		591 229.791 (20)	–35
41 – 40		362 032.724 (15)	1	68 – 67		600 035.398 (20)	8
42 – 41		370 855.633 (15)	–28	69 – 68		608 840.130 (20)	8
(0, 0, 0, 1)							
14 – 13	1e	123 968.616 (30)	–3	50 – 49	1e	442 502.530 (25)	–20
14 – 13	1f	124 142.276 (30)	3	50 – 49	1f	—	
15 – 14	1e	132 822.598 (30)	–6	51 – 50	1e	451 341.775 (20)	12
15 – 14	1f	133 008.625 (30)	–11	51 – 50	1f	451 967.069 (20)	–8
16 – 15	1e	141 676.397 (30)	–3	59 – 58	1e	522 031.240 (25)	–5
16 – 15	1f	141 874.803 (30)	0	59 – 58	1f	522 751.643 (25)	24
17 – 16	1e	150 529.993 (30)	0	60 – 59	1e	530 864.186 (25)	–29
17 – 16	1f	150 740.758 (30)	–6	60 – 59	1f	—	
18 – 17	1e	159 383.358 (30)	–14	66 – 65	1e	583 845.748 (25)	40
18 – 17	1f	159 606.526 (30)	24	66 – 65	1f	584 648.209 (25)	–3
41 – 40	1e	362 922.775 (20)	15	67 – 66	1e	592 673.112 (25)	–7
41 – 40	1f	363 427.690 (20)	5	67 – 66	1f	593 487.243 (25)	–27
42 – 41	1e	371 767.199 (60)	2	68 – 67	1e	601 499.678 (25)	–8
42 – 41	1f	372 284.241 (20)	9	68 – 67	1f	602 325.476 (25)	15
43 – 42	1e	380 611.090 (20)	–14				
43 – 42	1f	381 140.232 (15)	–1				
(0, 0, 0, 2)							
14 – 13	0e	124 441.180 (40)	30	18 – 17	0e	159 985.504 (40)	–26
14 – 13	2f	124 446.127 (40)	3	18 – 17	2f	159 997.023 (40)	–8
14 – 13	2e	124 452.212 (40)	10	18 – 17	2e	160 009.877 (40)	–70
15 – 14	0e	133 327.886 (40)	16	42 – 41	0e	373 039.338 (40)	–23
15 – 14	2f	133 334.168 (40)	3	42 – 41	2f	373 191.766 (40)	23
15 – 14	2e	133 341.641 (40)	0	42 – 41	2e	373 347.432 (40)	3
16 – 15	0e	142 214.201 (40)	11	43 – 42	0e	381 906.268 (40)	13
16 – 15	2f	142 222.017 (40)	12	43 – 42	2f	382 069.114 (40)	–8

Table S3—Continued

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
16 – 15	2e	142 231.091 (40)	13	43 – 42	2e	382 235.366 (40)	–6
17 – 16	0e	—					
17 – 16	2f	151 109.639 (40)	8				
17 – 16	2e	151 120.522 (40)	8				
(0, 0, 1, 0)							
14 – 13	1e	—		45 – 44	1e	397 978.922 (40)	–10
14 – 13	1f	123 963.750 (40)	–16	45 – 44	1f	—	
15 – 14	1e	—		46 – 45	1e	406 814.151 (40)	–50
15 – 14	1f	132 817.351 (40)	–61	46 – 45	1f	407 125.061 (50)	31
16 – 15	1e	141 562.505 (40)	–29	60 – 59	1e	530 442.100 (50)	13
16 – 15	1f	141 670.839 (40)	–30	60 – 59	1f	530 846.940 (50)	82
17 – 16	1e	150 408.991 (40)	–33	61 – 60	1e	539 267.473 (50)	23
17 – 16	1f	150 524.152 (40)	26	61 – 60	1f	—	
41 – 40	1e	—		62 – 61	1e	548 092.066 (50)	13
41 – 40	1f	362 909.544 (40)	–2	62 – 61	1f	548 510.107 (50)	–98
42 – 41	1e	371 469.802 (40)	–7	66 – 65	1e	583 382.609 (50)	–18
42 – 41	1f	—		66 – 65	1f	—	
43 – 42	1e	380 306.713 (40)	–11	67 – 66	1e	—	
43 – 42	1f	380 597.367 (40)	0	67 – 66	1f	592 654.818 (50)	15
44 – 43	1e	389 143.126 (40)	24				
44 – 43	1f	389 440.457 (40)	–20				
(0, 0, 1, 1)							
16 – 15	0e	142 051.762 (50)	21	17 – 16	0e	150 926.848 (50)	–19
16 – 15	0f	142 058.297 (50)	–2	17 – 16	0f	150 934.681 (50)	2
16 – 15	2f	142 076.948 (50)	–18	17 – 16	2f	150 956.588 (50)	16
16 – 15	2e	142 082.887 (50)	–1	17 – 16	2e	—	
(0, 1, 0, 0)							
14 – 13	1e	123 671.676 (50)	0	16 – 15	1e	141 337.113 (50)	0
14 – 13	1f	123 738.883 (300)	86	16 – 15	1f	141 413.797 (200)	–22
15 – 14	1e	132 504.484 (50)	–1				
15 – 14	1f	132 576.371 (300)	–28				

Table S4. Measured Transition Frequencies (MHz) of H¹³CCCN and Residuals (kHz).

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
(0, 0, 0, 0)							
14 – 13		123 430.763 (10)	–5	50 – 49		440 589.111 (20)	–1
15 – 14		132 246.364 (10)	0	51 – 50		449 390.387 (20)	0
16 – 15		141 061.778 (10)	3	59 – 58		519 777.099 (20)	–6
17 – 16		149 876.990 (10)	–1	60 – 59		528 572.327 (20)	0
18 – 17		158 691.994 (10)	–5	66 – 65		581 327.835 (20)	14
19 – 18		167 506.787 (10)	1	67 – 66		590 117.648 (20)	–2
20 – 19		176 321.340 (10)	–1	68 – 67		598 906.641 (20)	–20
41 – 40		361 351.579 (20)	4	69 – 68		607 694.859 (20)	18
42 – 41		370 157.918 (20)	3	70 – 69		616 482.172 (20)	–6
(0, 0, 0, 1)							
14 – 13	1e	123 737.972 (15)	–9	50 – 49	1e	441 679.309 (20)	–16
14 – 13	1f	123 910.874 (15)	–3	50 – 49	1f	442 289.989 (15)	5
15 – 14	1e	132 575.486 (15)	–8	51 – 50	1e	450 502.098 (15)	4
15 – 14	1f	132 760.714 (15)	0	51 – 50	1f	451 124.668 (15)	3
16 – 15	1e	141 412.816 (15)	–2	59 – 58	1e	—	
16 – 15	1f	141 610.356 (15)	0	59 – 58	1f	521 777.269 (25)	–14
17 – 16	1e	150 249.939 (15)	0	60 – 59	1e	529 876.622 (30)	9
17 – 16	1f	150 459.790 (15)	0	60 – 59	1f	530 605.577 (20)	13
18 – 17	1e	159 086.840 (15)	–7	66 – 65	1e	582 759.538 (20)	–8
18 – 17	1f	159 309.002 (15)	–2	66 – 65	1f	583 558.539 (20)	19
19 – 18	1e	167 923.534 (15)	6	67 – 66	1e	591 570.542 (20)	5
19 – 18	1f	168 157.988 (15)	3	67 – 66	1f	592 381.085 (20)	–20
20 – 19	1e	176 759.971 (15)	2	68 – 67	1e	—	
20 – 19	1f	177 006.725 (15)	5	68 – 67	1f	601 202.828 (20)	3
40 – 39	1e	353 419.075 (15)	–1	69 – 68	1e	609 189.969 (20)	–6
40 – 39	1f	353 909.724 (15)	3	69 – 68	1f	610 023.654 (25)	–13
41 – 40	1e	362 247.584 (15)	7	70 – 69	1e	617 998.427 (40)	27
41 – 40	1f	362 750.292 (15)	1	70 – 69	1f	618 843.629 (30)	9
42 – 41	1e	371 075.559 (15)	–1				
42 – 41	1f	371 590.311 (15)	–20				
(0, 0, 0, 2)							
14 – 13	0e	124 211.919 (30)	–11	42 – 41	0e	372 354.855 (20)	2
14 – 13	2f	124 216.853 (30)	0	42 – 41	2f	372 504.151 (20)	3
14 – 13	2e	124 222.790 (30)	4	42 – 41	2e	372 656.371 (20)	–14
15 – 14	0e	133 082.297 (15)	–3	50 – 49	0e	443 133.868 (20)	9

Table S4—Continued

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
15 – 14	2f	133 088.516 (15)	–2	50 – 49	2f	443 375.581 (20)	1
15 – 14	2e	133 095.808 (15)	–9	50 – 49	2e	443 620.747 (20)	–10
16 – 15	0e	141 952.266 (20)	–10	51 – 50	0e	451 976.889 (50)	38
16 – 15	2f	141 959.985 (15)	3	51 – 50	2f	452 231.660 (20)	6
16 – 15	2e	141 968.844 (15)	3	51 – 50	2e	452 489.968 (20)	–8
17 – 16	0e	150 821.822 (15)	–10	59 – 58	0e	522 685.313 (60)	–17
17 – 16	2f	150 831.237 (15)	4	59 – 58	2f	523 054.712 (20)	–2
17 – 16	2e	150 841.856 (15)	–3	59 – 58	2e	523 428.101 (30)	–1
18 – 17	0e	159 690.946 (30)	4	66 – 65	0e	584 503.931 (30)	–4
18 – 17	2f	159 702.260 (30)	3	66 – 65	2f	584 983.972 (30)	8
18 – 17	2e	159 714.869 (30)	0	66 – 65	2e	585 468.390 (40)	–6
19 – 18	0e	168 559.574 (15)	–7	67 – 66	0e	593 331.278 (30)	–20
19 – 18	2f	168 573.047 (15)	5	67 – 66	2f	593 827.570 (30)	–32
19 – 18	2e	168 587.866 (15)	–2	67 – 66	2e	594 328.366 (30)	4
20 – 19	0e	177 427.733 (15)	9	68 – 67	0e	602 157.690 (40)	–9
20 – 19	2f	177 443.577 (15)	5	68 – 67	2f	602 670.343 (40)	–6
20 – 19	2e	177 460.859 (15)	2	68 – 67	2e	603 187.548 (35)	35
40 – 39	0e	354 650.575 (20)	0	69 – 68	0e	610 983.132 (25)	–6
40 – 39	2f	354 780.449 (20)	18	69 – 68	2f	611 512.184 (30)	–10
40 – 39	2e	354 913.092 (20)	–7	69 – 68	2e	612 045.838 (30)	20
41 – 40	0e	363 503.182 (20)	4	70 – 69	0e	619 807.645 (50)	30
41 – 40	2f	363 642.566 (20)	4	70 – 69	2f	620 353.155 (50)	30
41 – 40	2e	—		70 – 69	2e	620 903.258 (50)	3
(0, 0, 1, 0)							
14 – 13	1e	123 634.414 (15)	–6	50 – 49	1e	441 312.490 (20)	9
14 – 13	1f	123 728.683 (15)	13	50 – 49	1f	441 648.240 (20)	7
15 – 14	1e	132 464.546 (15)	–1	51 – 50	1e	450 128.053 (20)	7
15 – 14	1f	132 565.525 (15)	–1	51 – 50	1f	450 470.471 (20)	–5
16 – 15	1e	141 294.487 (15)	0	59 – 58	1e	—	
16 – 15	1f	141 402.193 (15)	–2	59 – 58	1f	521 024.449 (60)	–4
17 – 16	1e	150 124.228 (15)	–1	60 – 59	1e	529 438.104 (20)	0
17 – 16	1f	150 238.663 (15)	0	60 – 59	1f	529 840.522 (20)	4
18 – 17	1e	158 953.755 (15)	–3	66 – 65	1e	582 278.471 (30)	5
18 – 17	1f	159 074.920 (15)	1	66 – 65	1f	582 720.746 (30)	–6
19 – 18	1e	167 783.073 (15)	10	67 – 66	1e	—	
19 – 18	1f	167 910.948 (15)	–3	67 – 66	1f	591 531.320 (30)	5
20 – 19	1e	176 612.135 (15)	3	68 – 67	1e	599 885.481 (30)	–9
20 – 19	1f	176 746.746 (15)	1	68 – 67	1f	600 341.046 (25)	3
41 – 40	1e	361 945.879 (20)	–11	69 – 68	1e	608 687.748 (25)	7
41 – 40	1f	362 221.454 (20)	–1	69 – 68	1f	609 149.922 (25)	–1

Table S4—Continued

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
42 – 41	1e	370 766.588 (20)	–18	70 – 69	1e	617 489.116 (40)	–18
42 – 41	1f	371 048.861 (20)	–7	70 – 69	1f	617 957.935 (40)	–8
(0, 0, 1, 1)							
14 – 13	0e	124 066.967 (20)	–1	18 – 17	0e	159 502.274 (20)	4
14 – 13	0f	124 072.144 (20)	–3	18 – 17	0f	159 512.741 (30)	–10
14 – 13	2f	124 083.545 (20)	4	18 – 17	2f	159 534.575 (20)	3
14 – 13	2e	124 088.006 (20)	–2	18 – 17	2e	159 544.133 (20)	–5
15 – 14	0e	132 926.535 (20)	–9	19 – 18	0e	168 359.744 (20)	–2
15 – 14	0f	132 932.814 (30)	1	19 – 18	0f	168 371.985 (20)	0
15 – 14	2f	132 946.398 (20)	2	19 – 18	2f	168 397.136 (20)	–3
15 – 14	2e	132 951.905 (20)	5	19 – 18	2e	168 408.417 (20)	3
16 – 15	0e	141 785.639 (20)	–2	20 – 19	0e	177 216.628 (20)	3
16 – 15	0f	141 793.160 (20)	13	20 – 19	0f	177 230.814 (20)	–5
16 – 15	2f	141 809.186 (20)	–5	20 – 19	2f	—	
16 – 15	2e	141 815.881 (20)	–2	20 – 19	2e	177 272.795 (20)	4
17 – 16	0e	150 644.231 (20)	5				
17 – 16	0f	150 653.135 (30)	3				
17 – 16	2f	150 671.915 (20)	–3				
17 – 16	2e	150 679.959 (20)	–3				
(0, 1, 0, 0)							
14 – 13	1e	123 434.128 (30)	–9	18 – 17	1e	158 695.667 (30)	–3
14 – 13	1f	123 501.663 (30)	–6	18 – 17	1f	158 782.314 (30)	–3
15 – 14	1e	132 249.819 (30)	–6	19 – 18	1e	167 510.519 (30)	7
15 – 14	1f	132 322.136 (30)	1	19 – 18	1f	167 601.941 (30)	0
16 – 15	1e	141 065.315 (30)	–1	20 – 19	1e	176 325.120 (30)	4
16 – 15	1f	141 142.407 (30)	3	20 – 19	1f	176 421.335 (30)	7
17 – 16	1e	149 880.601 (30)	0				
17 – 16	1f	149 962.464 (30)	–4				

Table S5. Measured Transition Frequencies (MHz) of $\text{HC}^{13}\text{C}^{13}\text{CN}$ and Residuals (kHz).

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
(0, 0, 0, 0)							
14 – 13		126 283.158 (10)	0	42 – 41		378 707.570 (15)	–5
15 – 14		135 302.456 (10)	10	43 – 42		387 716.552 (15)	4
16 – 15		144 321.540 (10)	0	50 – 49		450 763.085 (15)	4
17 – 16		153 340.423 (10)	–4	51 – 50		459 767.261 (15)	12
18 – 17		162 359.094 (10)	–1	52 – 51		468 770.760 (15)	3
19 – 18		171 377.528 (10)	–1	53 – 52		477 773.590 (15)	–4
41 – 40		369 698.045 (15)	–14	59 – 58		531 775.784 (15)	–4
(0, 0, 0, 1)							
14 – 13	1e	126 577.048 (10)	–1	17 – 16	1e	153 697.200 (10)	–2
14 – 13	1f	126 760.517 (10)	–2	17 – 16	1f	153 919.888 (10)	0
15 – 14	1e	135 617.305 (10)	1	18 – 17	1e	162 736.820 (10)	1
15 – 14	1f	135 813.852 (10)	1	18 – 17	1f	162 972.563 (10)	–1
16 – 15	1e	144 657.361 (10)	2	19 – 18	1e	171 776.197 (10)	–1
16 – 15	1f	144 866.980 (10)	1	19 – 18	1f	172 024.994 (10)	0
(0, 0, 0, 2)							
14 – 13	0e	127 046.617 (20)	2	17 – 16	0e	154 262.664 (20)	–1
14 – 13	2f	127 052.696 (20)	1	17 – 16	2f	154 274.578 (20)	2
14 – 13	2e	127 060.555 (20)	2	17 – 16	2e	154 288.643 (20)	–3
15 – 14	0e	136 119.108 (20)	–1	18 – 17	0e	—	
15 – 14	2f	136 126.871 (20)	1	18 – 17	2f	163 348.073 (20)	–5
15 – 14	2e	136 136.534 (20)	–3	18 – 17	2e	163 364.775 (20)	1
16 – 15	0e	145 191.135 (20)	0	19 – 18	0e	—	
16 – 15	2f	145 200.833 (20)	–2	19 – 18	2f	172 421.330 (20)	3
16 – 15	2e	145 212.568 (20)	1	19 – 18	2e	172 440.955 (20)	1
(0, 0, 1, 0)							
14 – 13	1e	126 478.137 (15)	–1	17 – 16	1e	153 577.139 (15)	6
14 – 13	1f	126 580.165 (15)	–2	17 – 16	1f	153 701.010 (15)	–2
15 – 14	1e	135 511.336 (15)	–2	18 – 17	1e	162 609.701 (15)	0
15 – 14	1f	135 620.653 (15)	2	18 – 17	1f	162 740.865 (15)	2
16 – 15	1e	144 544.341 (15)	0	19 – 18	1e	171 642.031 (15)	–3
16 – 15	1f	144 660.937 (15)	0	19 – 18	1f	171 780.476 (15)	–1

Table S5—Continued

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
(0, 0, 1, 1)							
14 – 13	0e	126 904.027 (20)	4	17 – 16	0e	154 087.613 (20)	–37
14 – 13	0f	—		17 – 16	0f	154 097.615 (30)	33
14 – 13	2f	126 925.758 (20)	–7	17 – 16	2f	154 124.028 (20)	13
14 – 13	2e	126 930.744 (20)	16	17 – 16	2e	154 133.035 (20)	–7
15 – 14	0e	135 965.807 (20)	–4	18 – 17	0e	163 147.633 (20)	–2
15 – 14	0f	135 972.726 (30)	–16	18 – 17	0f	163 159.366 (30)	–10
15 – 14	2f	135 991.863 (20)	–11	18 – 17	2f	163 190.026 (20)	–1
15 – 14	2e	135 998.004 (20)	–3	18 – 17	2e	163 200.809 (20)	0
16 – 15	0e	145 027.064 (20)	32	19 – 18	0e	172 206.962 (20)	8
16 – 15	0f	145 035.361 (20)	–5	19 – 18	0f	172 220.733 (20)	1
16 – 15	2f	145 057.958 (20)	–2	19 – 18	2f	172 255.983 (20)	1
16 – 15	2e	145 065.444 (20)	1	19 – 18	2e	172 268.748 (20)	–2
(0, 1, 0, 0)							
14 – 13	1e	—		16 – 15	1e	—	
14 – 13	1f	126 361.278 (30)	–7	16 – 15	1f	144 410.818 (30)	–6
15 – 14	1e	135 311.117 (30)	0				
15 – 14	1f	135 386.165 (30)	13				

Table S6. Measured Transition Frequencies (MHz) of $\text{H}^{13}\text{CC}^{13}\text{CN}$ and Residuals (kHz).

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
(0, 0, 0, 0)							
14 – 13		122 880.470 (10)	1	42 – 41		368 507.710 (15)	–10
15 – 14		131 656.755 (10)	–6	43 – 42		377 274.294 (15)	0
16 – 15		140 432.879 (10)	7	44 – 43		386 040.346 (15)	–1
17 – 16		149 208.786 (10)	–2	50 – 49		438 624.982 (15)	–1
18 – 17		157 984.491 (10)	–5	51 – 50		447 387.030 (15)	0
19 – 18		166 759.987 (10)	2	52 – 51		456 148.458 (15)	2
20 – 19		175 535.248 (10)	5	53 – 52		464 909.266 (20)	15
41 – 40		359 740.635 (15)	2	60 – 59		526 216.063 (15)	–3
(0, 0, 0, 1)							
14 – 13	1e	123 180.485 (10)	2	18 – 17	1e	158 370.097 (10)	–3
14 – 13	1f	123 353.001 (10)	–3	18 – 17	1f	158 591.773 (10)	–1
15 – 14	1e	131 978.178 (10)	–3	19 – 18	1e	167 166.972 (10)	0
15 – 14	1f	132 163.002 (10)	3	19 – 18	1f	167 400.920 (10)	1
16 – 15	1e	140 775.695 (10)	3	20 – 19	1e	175 963.608 (10)	1
16 – 15	1f	140 972.801 (10)	0	20 – 19	1f	176 209.817 (10)	–1
17 – 16	1e	149 573.003 (10)	1				
17 – 16	1f	149 782.398 (10)	1				
(0, 0, 0, 2)							
14 – 13	0e	123 646.699 (20)	6	18 – 17	0e	158 964.046 (20)	–1
14 – 13	2f	123 651.575 (20)	6	18 – 17	2f	158 975.491 (20)	–4
14 – 13	2e	123 657.742 (20)	4	18 – 17	2e	158 988.605 (20)	–3
15 – 14	0e	—		19 – 18	0e	167 792.254 (20)	0
15 – 14	2f	132 482.854 (20)	–8	19 – 18	2f	167 805.914 (20)	0
15 – 14	2e	—		19 – 18	2e	167 821.343 (50)	13
16 – 15	0e	141 306.216 (20)	–7	20 – 19	0e	176 619.960 (20)	4
16 – 15	2f	141 313.957 (20)	0	20 – 19	2f	176 636.082 (20)	2
16 – 15	2e	141 323.167 (20)	–1	20 – 19	2e	—	
17 – 16	0e	150 135.360 (20)	–1				
17 – 16	2f	150 144.842 (20)	3				
17 – 16	2e	150 155.886 (20)	–1				
(0, 0, 1, 0)							
14 – 13	1e	123 076.326 (15)	–1	18 – 17	1e	158 236.242 (15)	1
14 – 13	1f	123 171.736 (15)	7	18 – 17	1f	158 358.875 (15)	–9

Table S6—Continued

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
15 – 14	1e	131 866.592 (15)	–4	19 – 18	1e	167 025.697 (15)	3
15 – 14	1f	131 968.806 (15)	–4	19 – 18	1f	167 155.149 (15)	5
16 – 15	1e	140 656.682 (15)	2	20 – 19	1e	175 814.911 (15)	–2
16 – 15	1f	140 765.704 (15)	1	20 – 19	1f	175 951.170 (15)	1
17 – 16	1e	149 446.567 (15)	1				
17 – 16	1f	149 562.399 (15)	0				
(0, 0, 1, 1)							
14 – 13	0e	—		18 – 17	0e	158 776.746 (20)	–6
14 – 13	0f	123 506.618 (20)	–5	18 – 17	0f	158 785.283 (20)	–11
14 – 13	2f	123 519.565 (20)	0	18 – 17	2f	158 809.887 (20)	–1
14 – 13	2e	123 523.003 (20)	–5	18 – 17	2e	158 817.347 (20)	–2
15 – 14	0e	—		19 – 18	0e	167 594.045 (20)	–4
15 – 14	0f	132 326.817 (20)	–5	19 – 18	0f	167 604.024 (20)	5
15 – 14	2f	132 342.223 (20)	1	19 – 18	2f	167 632.312 (20)	–2
15 – 14	2e	132 346.486 (20)	13	19 – 18	2e	167 641.137 (20)	–3
16 – 15	0e	—		20 – 19	0e	176 410.770 (20)	0
16 – 15	0f	141 146.692 (20)	13	20 – 19	0f	176 422.342 (30)	5
16 – 15	2f	141 164.832 (20)	–1	20 – 19	2f	176 454.661 (20)	7
16 – 15	2e	141 170.018 (20)	1	20 – 19	2e	176 465.019 (20)	0
17 – 16	0e	149 958.914 (20)	11				
17 – 16	0f	149 966.175 (20)	0				
17 – 16	2f	149 987.387 (20)	–5				
17 – 16	2e	149 993.639 (20)	–2				
(0, 1, 0, 0)							
15 – 14	1e	131 658.343 (30)	–10	18 – 17	1e	157 986.391 (30)	–15
15 – 14	1f	131 729.924 (30)	–9	18 – 17	1f	158 072.295 (30)	–8
16 – 15	1e	140 434.564 (30)	–6	19 – 18	1e	166 761.991 (30)	–12
16 – 15	1f	140 510.933 (30)	12	19 – 18	1f	166 852.675 (30)	5
17 – 16	1e	149 210.633 (30)	41	20 – 19	1e	175 537.369 (30)	2
17 – 16	1f	149 291.712 (30)	–4	20 – 19	1f	175 632.809 (30)	2

Table S7. Measured Transition Frequencies (MHz) of $\text{H}^{13}\text{C}^{13}\text{CCN}$ and Residuals (kHz).

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
(0, 0, 0, 0)							
14 – 13		122 974.813 (15)	–4	18 – 17		158 105.799 (15)	–7
15 – 14		131 757.848 (15)	–2	19 – 18		166 888.044 (15)	8
16 – 15		140 540.711 (15)	10	20 – 19		175 670.032 (15)	–4
17 – 16		149 323.355 (15)	–2				
(0, 0, 0, 1)							
14 – 13	1e	123 269.913 (20)	6	18 – 17	1e	158 485.075 (20)	2
14 – 13	1f	123 444.066 (20)	3	18 – 17	1f	158 708.841 (20)	–4
15 – 14	1e	132 073.987 (20)	–6	19 – 18	1e	167 288.332 (20)	0
15 – 14	1f	132 260.562 (20)	1	19 – 18	1f	167 524.490 (20)	–3
16 – 15	1e	140 877.888 (20)	–2	20 – 19	1e	176 091.353 (20)	0
16 – 15	1f	141 076.869 (20)	2	20 – 19	1f	176 339.901 (20)	5
17 – 16	1e	149 681.589 (20)	0				
17 – 16	1f	149 892.963 (20)	–3				
(0, 0, 1, 0)							
14 – 13	1e	123 173.402 (30)	7	18 – 17	1e	158 361.035 (30)	–3
14 – 13	1f	123 268.159 (30)	19	18 – 17	1f	158 482.824 (30)	–9
15 – 14	1e	131 970.593 (30)	–4	19 – 18	1e	167 157.427 (30)	6
15 – 14	1f	132 072.093 (30)	–13	19 – 18	1f	167 285.995 (30)	17
16 – 15	1e	140 767.619 (30)	6	20 – 19	1e	175 953.565 (30)	–5
16 – 15	1f	140 875.871 (30)	–14	20 – 19	1f	176 088.888 (30)	0
17 – 16	1e	149 564.424 (30)	–6				
17 – 16	1f	—					

Table S8. Measured Transition Frequencies (MHz) of $\text{HCC}^{13}\text{C}^{15}\text{N}$ and Residuals (kHz).

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
(0, 0, 0, 0)							
14 – 13		123 215.167 (15)	–2	44 – 43		387 091.736 (15)	4
15 – 14		132 015.359 (15)	–7	50 – 49		439 819.542 (15)	0
16 – 15		140 815.381 (15)	2	51 – 50		448 605.446 (25)	1
17 – 16		149 615.197 (15)	–1	52 – 51		457 390.720 (15)	–4
18 – 17		158 414.811 (15)	2	53 – 52		466 175.368 (15)	0
19 – 18		167 214.201 (15)	2	60 – 59		527 649.074 (15)	1
20 – 19		176 013.361 (15)	3				
(0, 0, 0, 1)							
14 – 13	1e	123 513.340 (20)	6	18 – 17	1e	158 798.025 (20)	–2
14 – 13	1f	123 686.969 (20)	2	18 – 17	1f	159 021.127 (20)	–5
15 – 14	1e	—		19 – 18	1e	167 618.663 (20)	–3
15 – 14	1f	132 520.817 (20)	5	19 – 18	1f	167 854.128 (20)	4
16 – 15	1e	141 156.080 (20)	–3	20 – 19	1e	176 439.072 (20)	6
16 – 15	1f	141 354.461 (20)	–3	20 – 19	1f	176 686.870 (20)	1
17 – 16	1e	149 977.159 (20)	–3				
17 – 16	1f	150 187.905 (20)	–3				
(0, 0, 0, 2)							
14 – 13	0e	123 978.665 (30)	24	18 – 17	0e	159 390.660 (30)	1
14 – 13	2f	123 983.603 (30)	7	18 – 17	2f	159 402.370 (30)	–9
14 – 13	2e	123 989.942 (30)	–25	18 – 17	2e	159 415.910 (30)	–11
15 – 14	0e	132 832.256 (30)	–27	19 – 18	0e	168 242.492 (75)	–20
15 – 14	2f	132 838.631 (30)	27	19 – 18	2f	168 256.514 (30)	3
15 – 14	2e	132 846.448 (30)	6	19 – 18	2e	168 272.433 (30)	1
16 – 15	0e	141 685.519 (30)	2	20 – 19	0e	177 093.857 (30)	4
16 – 15	2f	141 693.418 (30)	5	20 – 19	2f	177 110.385 (30)	–5
16 – 15	2e	141 702.923 (30)	–4	20 – 19	2e	177 128.962 (30)	12
17 – 16	0e	150 538.319 (30)	0				
17 – 16	2f	150 547.996 (30)	–13				
17 – 16	2e	150 559.425 (30)	5				
(0, 0, 1, 0)							
14 – 13	1e	123 412.668 (75)	–18	18 – 17	1e	158 668.689 (25)	–7
14 – 13	1f	123 508.804 (25)	7	18 – 17	1f	158 792.257 (25)	7
15 – 14	1e	132 226.985 (25)	5	19 – 18	1e	167 482.171 (25)	–1

Table S8—Continued

$J' - J''$	ℓ	Frequency	$o - c$	$J' - J''$	ℓ	Frequency	$o - c$
15 – 14	1 <i>f</i>	—		19 – 18	1 <i>f</i>	167 612.582 (25)	–3
16 – 15	1 <i>e</i>	141 041.090 (25)	2	20 – 19	1 <i>e</i>	176 295.405 (25)	–7
16 – 15	1 <i>f</i>	141 150.891 (25)	–31	20 – 19	1 <i>f</i>	176 432.691 (25)	8
17 – 16	1 <i>e</i>	149 855.008 (25)	12				
17 – 16	1 <i>f</i>	149 971.703 (25)	11				