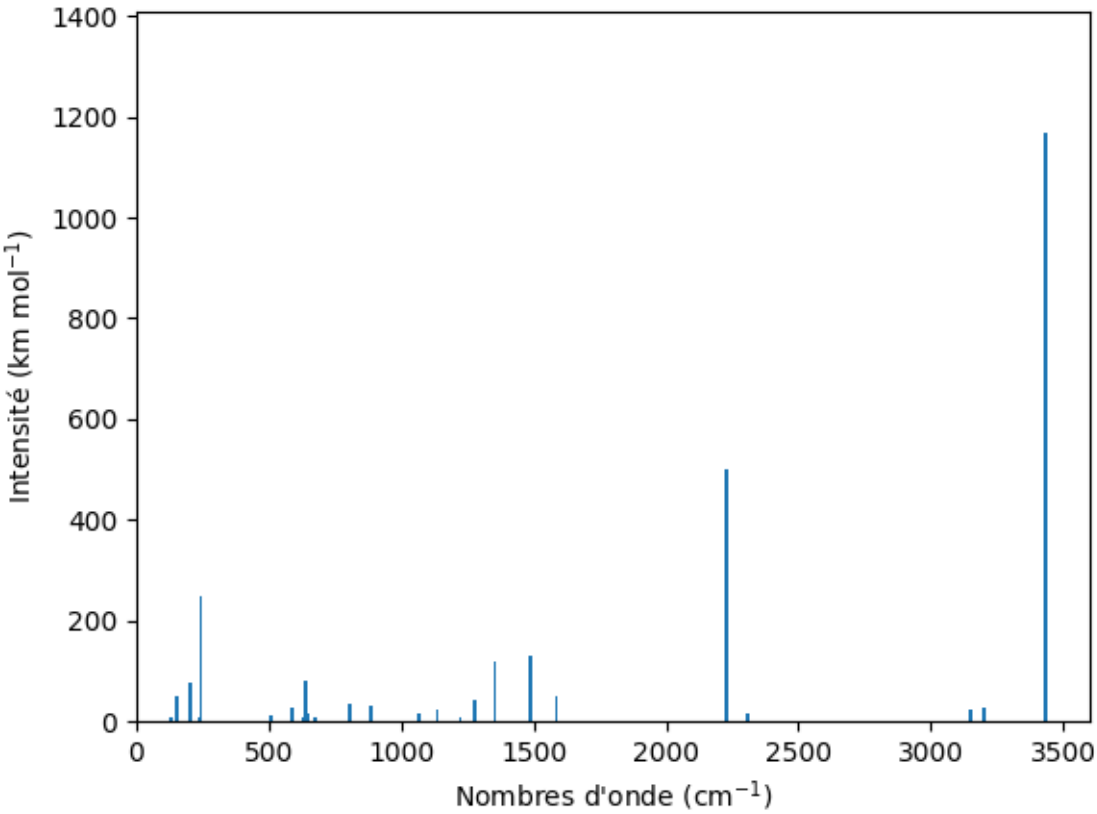
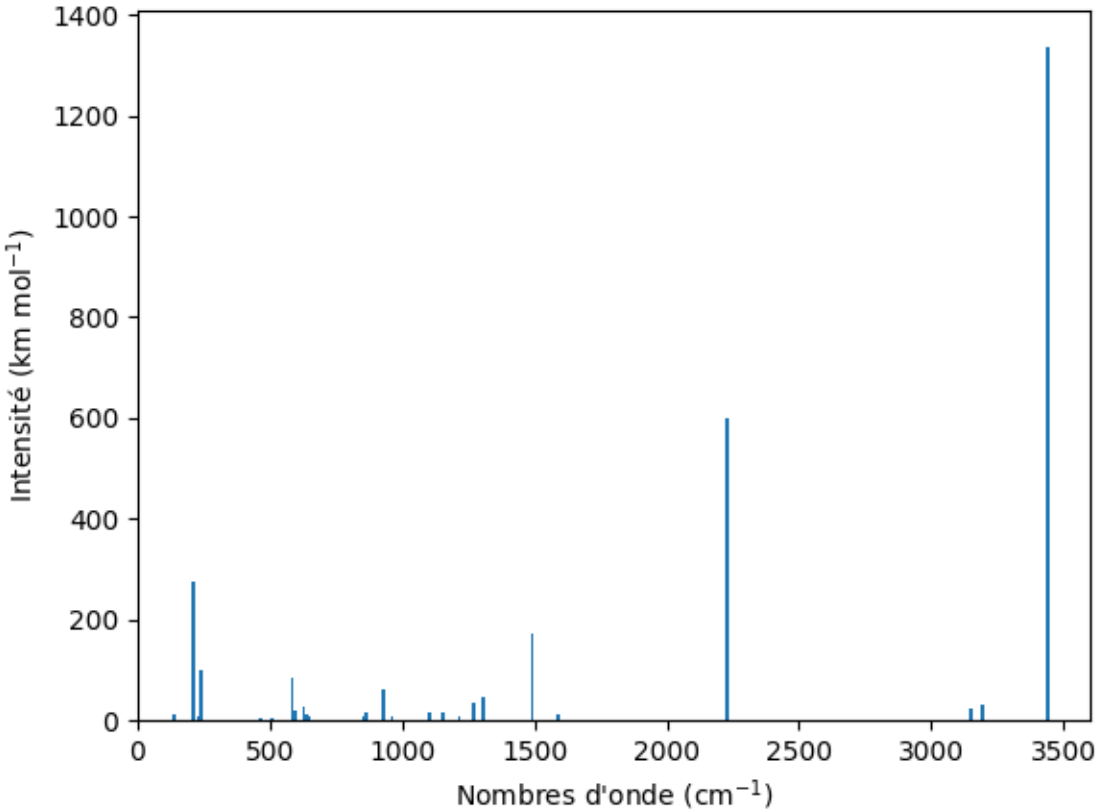
Electronic Supplementary Material (ESI) for New Journal of Chemistry.

Spontaneous formation and protonation of dicyanofuran isomers under interstellar space physical conditions: quantum chemical insights into thermodynamics and spectroscopy

René Simbizi,[a,b] Sahar Abdalla,[c,d] Thierry Bukuru,[a,b] Pierre Claver Mpawenayo,[b,e] Manassé Nihombere [b,e] and Godefroid Gahungu [b,e,+]

Electronic Supplementary Information


Intensité (km mol^{-1})
Nombres d'onde (cm^{-1})
0
200
400
600
800
1000
1200
1400
0
500
1000
1500
2000
2500
3000
3500


Intensité (km mol^{-1})
Nombres d'onde (cm^{-1})
0
200
400
600
800
1000
1200
1400
0
500
1000
1500
2000
2500
3000
3500

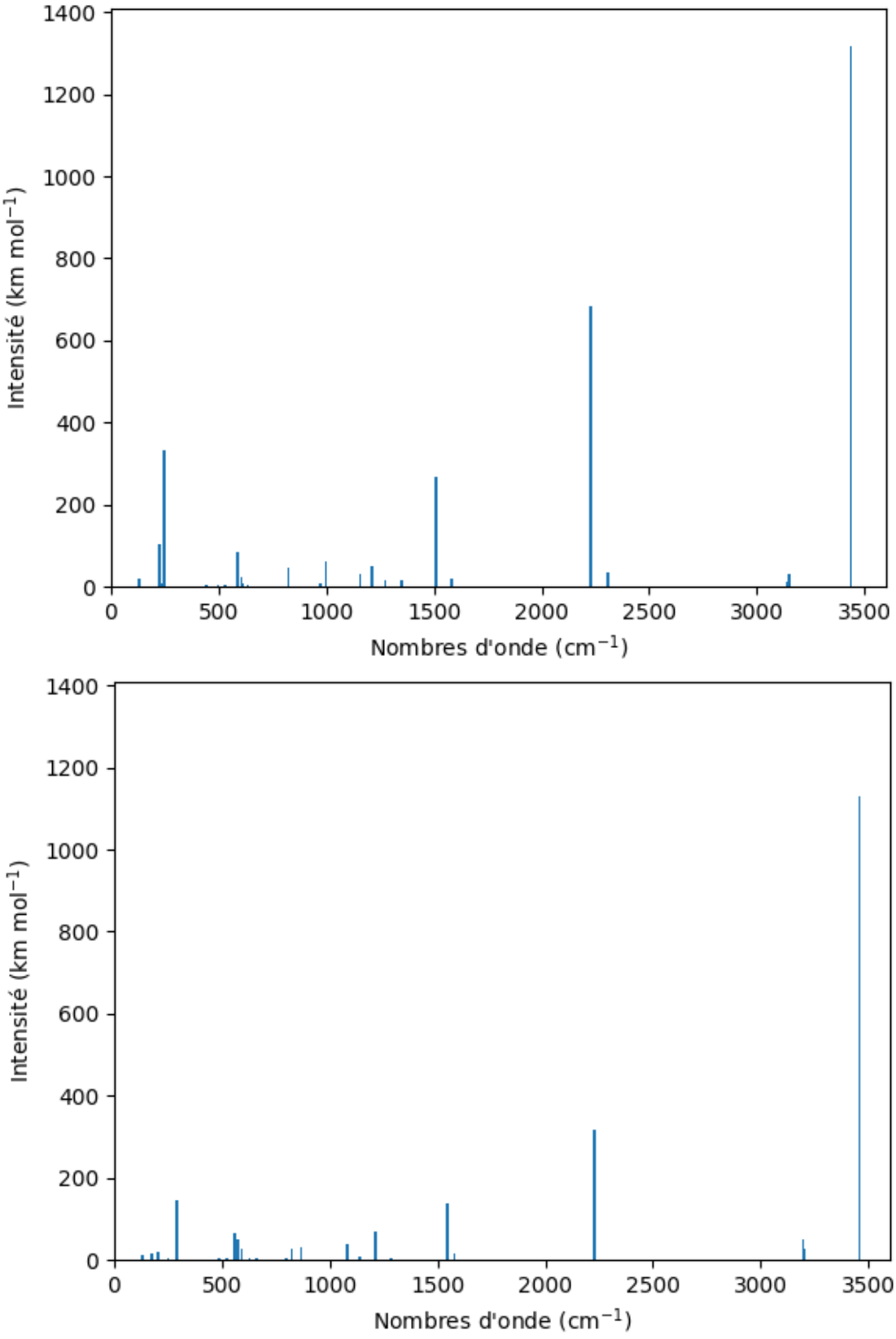
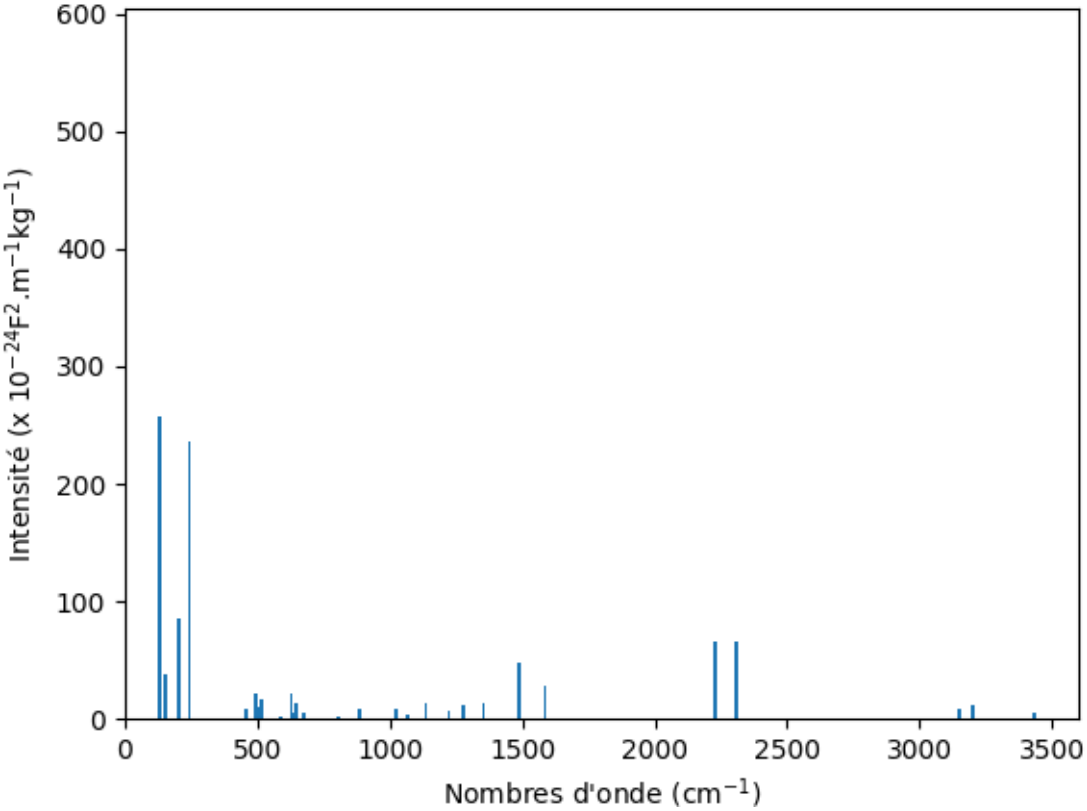
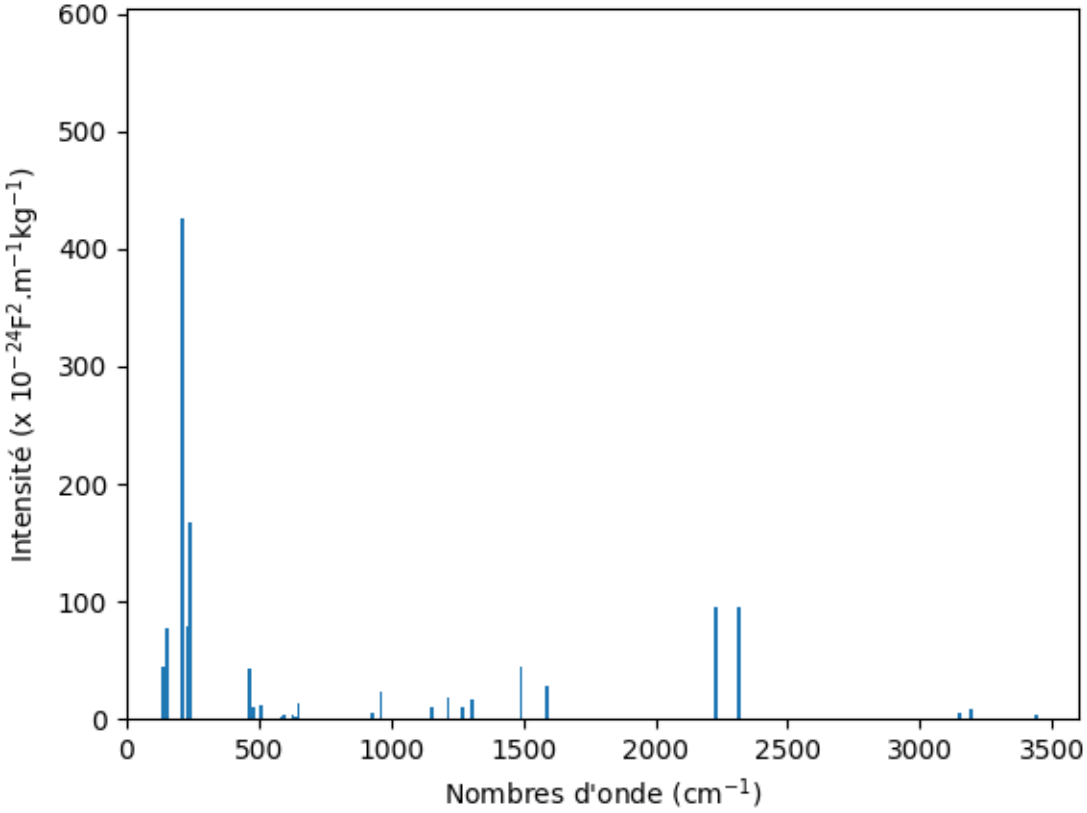


Figure S1. The IR spectra The Raman spectra or 23DCFH$^+$, 24DCFH$^+$, 25DCFH$^+$ and 34DCFH$^+$ (from up to down).


Intensité (x $10^{-24}F^2.m^{-1}kg^{-1}$)
Nombres d'onde (cm^{-1})
0
100
200
300
400
500
600
0
500
1000
1500
2000
2500
3000
3500


Intensité (x $10^{-24}F^2.m^{-1}kg^{-1}$)
Nombres d'onde (cm^{-1})
0
100
200
300
400
500
600
0
500
1000
1500
2000
2500
3000
3500

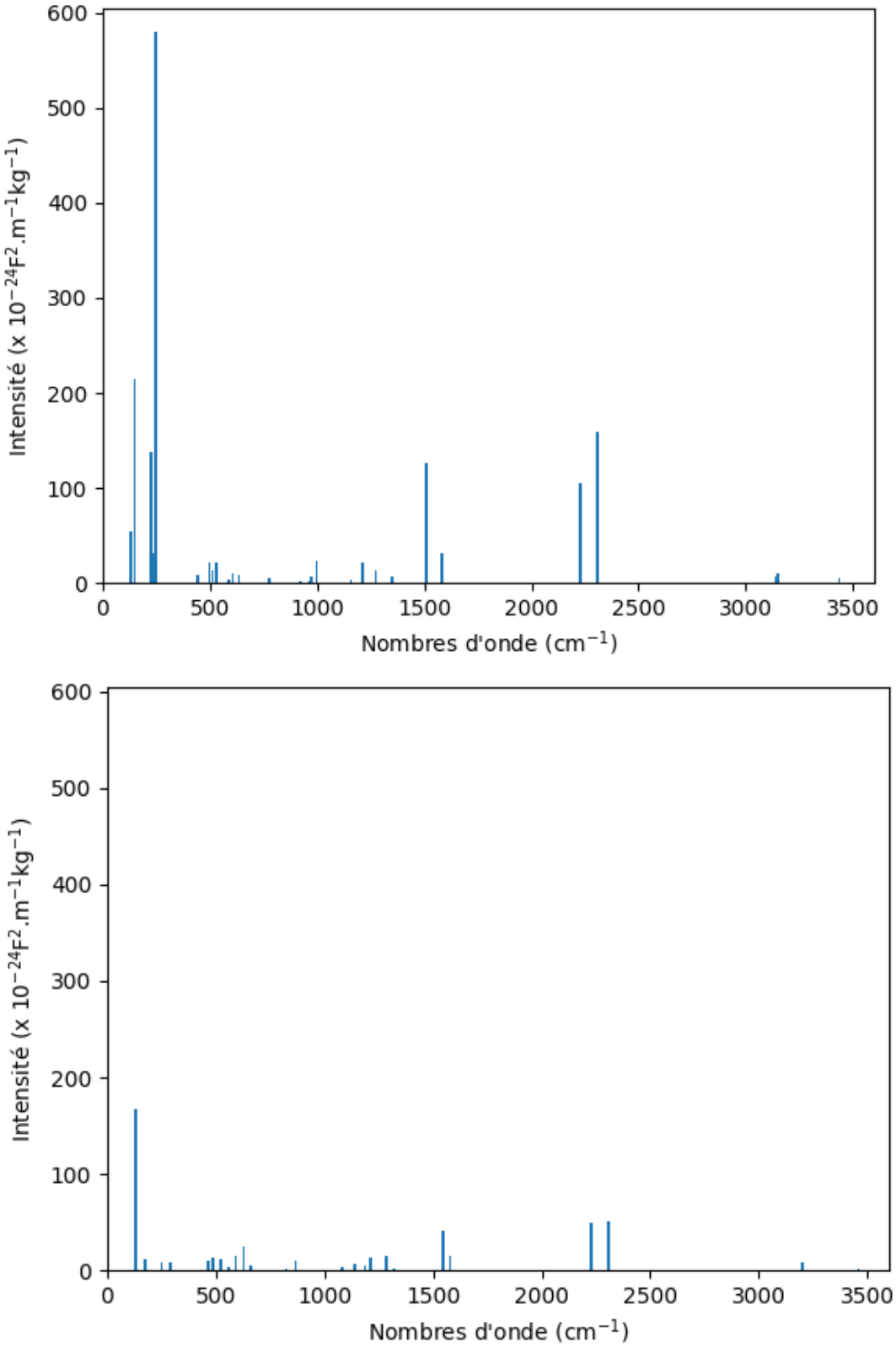


Figure S2. The Raman spectra or23DCFH$^+$, 24DCFH$^+$, 25DCFH$^+$ and 34DCFH$^+$ (from top to bottom).

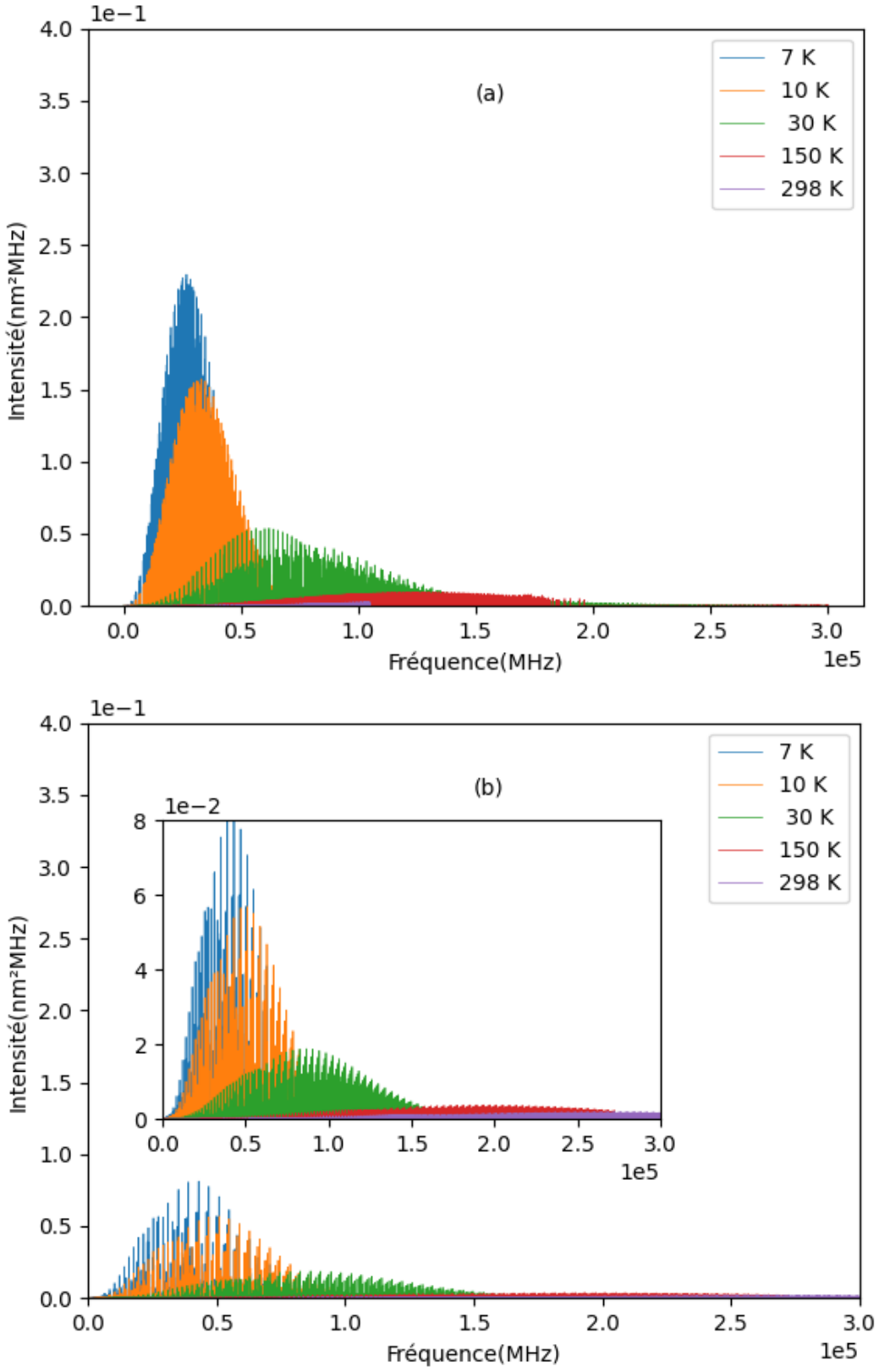


Figure S3. Plots of rotational line transition intensities (including ^{14}N hyperfine quadrupole couplings) for 25DCFH$^+$ (up) and (b) 34DCFH$^+$ (down).

Table S1. B3LYP/6-31G(d,p)optimized structures for furan, 23DCF, 24DCF, 25DCF and 34DCF23CFH$^+$, 24CFH$^+$, 25CFH$^+$, 34CFH$^+$and HCNH$^+$.

Parameter	Furan[a]	23CF	24CF	25CF	34CF	23CFH$^+$	24CFH$^+$	25CFH$^+$	34CFH$^+$	HCNH^{+b}
Bond length(in Å)										
O1-C2	1.3583 (1.362)	1.3607	1.3755	1.3645	1.3551	1.3550	1.3698	1.3589	1.3494	
O1-C5	1.3583 (1.362)	1.3595	1.3450	1.3645	1.3551	1.3538	1.3392	1.3589	1.3494	
C2-C3	1.3621 (1.361)	1.3817	1.3602	1.3737	1.3817	1.3667	1.3679	1.3753	1.3684	
C3-C4	1.4520 (1.431)	1.4342	1.4343	11.418	1.4524	1.4508	1.4510	1.4354	1.4690	
C4-C5	1.3621 (1.361)	1.3602	1.3740	1.3737	1.3667	1.3619	1.3757	1.3753	1.3684	
C5-H/C9		1.0785	1.0787	1.4160	1.0782	1.0723	1.0727	1.4099	1.0722	1.08464 (1.0779)
C2-H/C8		1.4137	1.4154	1.4159	1.0782	1.4099	1.4099	1.4099	1.0722	
C4-H/C9		1.0791	1.4221	1.0796	1.4202	1.0767	1.4099	1.0772	1.4099	
C3-C9/H6		1.4204	1.0791	1.0796	1.4202	1.4099	1.0767	1.0772	1.4099	
C≡N		1.1637	1.1630	1.1636	1.1627	1.0723	1.0727	1.4099	1.0722	1.1397 (1.1368)
N7-H						1.0094	1.0091	1.0113	1.0096	1.0171 (1.0091)
N9-H						1.0114	1.0110	1.0113	1.0096	
Bond angle(in^o)										
O1-C2=C3	110.81 (110.7)	109.74	110.28	110.68	110.37	108.27	110.68	112.09	108.71	
C2=C3-C4	105.62 (106.0)	106.11	105.79	106.16	105.68	107.41	104.17	104.91	107.14	
C5=C4-C3	105.62 (106.0)	105.89	106.17	106.16	105.68	104.24	107.52	105.88	103.94	
O1-C5=C4	110.81 (110.7)	111.00	110.59	110.68	110.37	111.47	109.00	111.44	110.84	
C5-O1-C2	107.12 (106.5)	107.24	107.14	106.28	107.89	108.60	108.61	105.66	109.35	
O1-C2-H6/C8	114.91 (115.9)	118.15	117.60	117.61	116.91	119.87	117.68	117.11	117.39	
O1-C5-H9/C9	114.91 (115.9)	115.67	116.76	117.61	116.91	115.84	117.36	117.08	116.90	
O1-C2-C8/H6		118.15	117.60	117.61	116.91	176.91	176.21	175.80	177.37	
O1-C5-H7/C9		115.67	116.76	117.61	116.91	124.44	128.32	127.50	126.58	
C2/C4-C8-N		179.30	178.79	178.49	179.78	179.69	179.56	178.68	178.16	
C2-C3-C9/H		126.64	126.90	126.03	126.63	132.67	132.06	126.73	132.25	
C4/C5/C3-C9-N		178.88	179.67	178.48	179.77	108.27	110.68	112.09	108.71	

*The numbering scheme used is shown on figure 1.[a] Experimental data for furan from … .;[b] Experimental data for HCNH$^+$ from Amano and Keiichi Amano and Tanaka [32]

Table S2. Gas phase G2(MP2), G3B3 and G4 enthalpy and Gibbs free energy (Δ_rH and Δ_rG in kJ/mol) of the reaction producing cyanofuran and their protonated forms at 150K and 5K, at P=10^{-5} atm.

Equation		T=150 K			T=5 K			
		$\Delta_r H$	$\Delta_r G$	$\Delta_r S$	$\Delta_r H$	$\Delta_r G$	$\Delta_r S$	Methods
$C_4H_4O^+ + CN^- \rightarrow 2CNF + H$		- 566.333	- 557.829	- 0.057	- 533.423	- 533.239	- 0.037	G2MP2
	1	- 571.855	- 563.343	- 0.057	- 565.853	- 594.560	- 0.293	G3B3
		- 563.212	- 554.597	- 0.057	- 563.015	- 562.818	- 0.039	G4
$C_4H_4O^+ + CN^- \rightarrow 3CNF + H$		- 573.212	- 564.727	- 0.057	- 540.286	- 540.102	- 0.037	G2MP2
	2	- 578.277	- 569.765	- 0.057	- 572.257	- 600.964	- 0.039	G3B3
		- 570.369	- 561.786	- 0.057	- 570.167	- 569.972	- 0.039	G4
$C_4H_3OCN^+ + CN^- \rightarrow 23CNF + H$		- 617.864	- 608.922	- 0.060	- 584.872	- 584.670	- 0.040	G2MP2
	3	- 619.663	- 610.920	- 0.058	- 613.726	- 642.420	- 0.042	G3B3
		- 610.972	- 602.151	- 0.059	- 610.854	- 610.641	- 0.043	G4
$C_4H_3OCN^+ + CN^- \rightarrow 24CNF + H$		- 622.580	- 613.655	- 0.059	- 589.637	- 589.433	- 0.041	G2MP2
	4	- 623.688	- 614.947	- 0.058	- 617.793	- 646.485	- 0.043	G3B3
		- 615.013	- 606.191	- 0.059	- 614.939	- 614.721	- 0.044	G4
$C_4H_3OCN^+ + CN^- \rightarrow 25CNF + H$		- 614.430	- 605.480	- 0.060	- 581.462	- 581.257	- 0.042	G2MP2
	5	- 616.520	- 607.766	- 0.058	- 610.605	- 639.299	- 0.042	G3B3
		- 607.483	- 598.638	- 0.059	- 607.381	- 607.168	- 0.043	G4
$C_4H_3OCN^+ + CN^- \rightarrow 34CNF + H$		- 625.289	- 616.389	- 0.059	- 592.289	- 592.090	- 0.039	G2MP2
	6	- 625.980	- 617.239	- 0.058	- 620.020	- 648.719	- 0.041	G3B3
		- 621.658	- 612.878	- 0.059	- 621.56	- 621.35	- 0.042	G4
$23CNF + H_3^+ \rightarrow 23DCNFH^+ + H_2$		- 351.008	- 317.533	- 0.223	- 346.976	- 346.332	- 0.129	G2MP2
	7	- 346.577	- 345.177	- 0.009	- 346.361	- 346.366	0.001	G3B3
		- 344.975	- 343.376	- 0.011	- 344.652	- 344.655	0.001	G4
$24CNF + H_3^+ \rightarrow 24DCNFH^+ + H_2$		- 347.645	- 314.144	- 0.223	- 343.602	- 342.959	- 0.129	G2MP2
	8	- 343.077	- 341.704	- 0.009	- 342.882	- 342.888	- 0.001	G3B3
		- 341.620	- 340.021	- 0.011	- 341.302	- 341.307	- 0.001	G4
$25CNF + H_3^+ \rightarrow 25DCNFH^+ + H_2$		- 371.58	- 307.18	- 0.43	- 363.36	- 362.18	- 0.235	G2MP2
	9	- 336.68	- 334.88	- 0.00	- 333.04	- 333.05	0.001	G3B3
		- 333.95	- 333.49	- 0.01	- 334.20	- 334.20	0.001	G4
$34CNF + H_3^+ \rightarrow 34DCNFH^+ + H_2$		- 385.30	- 320.93	- 0.43	- 377.13	- 375.95	- 0.236	G2MP2
	10	- 349.30	- 334.88	- 0.01	- 351.29	- 351.29	0.001	G3B3
		- 347.871	- 346.29	- 0.01	- 347.55	- 347.54	- 0.001	G4

Table S3. B3LYP/6-31G(d,p) frequencies (cm^{-1}), Infrared intensities ($km.mol^{-1}$), Raman activities ($Å^4/amu$) and intensities ($\times 10^{-24}\ F^2.m^{-1}.kg^{-1}$) for 23CNFH$^+$.

Non Scaled frequencies	Scaled frequencies	IR Intensity	Raman Activity	Raman Intensity
3685.38	3434.99	1169.58	61.75	4.54
3374.54	3199.56	27.80	135.91	12.52
3321.89	3149.68	22.66	84.11	8.12
2309.18	2305.75	17.58	298.63	65.40
2233.97	2226.57	499.03	278.22	65.96
1610.11	1585.93	51.58	59.83	28.06
1517.67	1485.62	130.95	89.39	47.11
1384.65	1352.68	119.09	21.60	13.37
1291.82	1278.39	41.66	17.45	11.87
1272.15	1222.61	9.39	8.98	6.57
1161.22	1135.13	22.85	16.72	13.76
1101.74	1065.72	17.13	3.71	3.37
1065.48	1021.14	1.83	9.30	9.01
904.45	887.85	0.77	0.18	0.22
900.61	883.88	29.73	7.35	8.84
806.81	804.81	36.80	0.94	1.30
677.46	674.83	9.31	2.65	4.72
657.69	646.12	15.69	6.98	13.23
644.19	639.79	79.71	2.40	4.62
633.42	627.51	6.78	10.95	21.66
590.61	585.79	28.11	0.98	2.14
531.23	514.70	0.87	6.05	15.95
521.59	507.43	13.66	3.65	9.82
495.53	492.17	1.13	7.86	22.11
465.78	458.37	2.49	2.86	8.93
239.63	242.56	247.17	28.25	236.79
230.82	237.57	9.23	0.13	1.10
206.92	204.17	78.62	7.73	86.19
156.49	153.19	48.60	2.08	37.86
133.06	129.68	8.66	10.51	256.60

Table S4. B3LYP/6-31G(d,p) frequencies (cm^{-1}), Infrared intensities ($km.mol^{-1}$), Raman activities ($Å^4$/amu) and intensities ($\times 10^{-24}$ $F^2.m^{-1}.kg^{-1}$) for 24DCFH$^+$.

Non Scaled frequencies	Scaled frequencies	IR Intensity	Raman Activity	Raman Intensity
3690.62	3439.97	1335.01	45.32	3.31
3368.79	3194.24	30.86	86.18	7.98
3323.50	3151.17	22.67	46.53	4.49
2318.45	2313.63	2.52	436.40	94.83
2238.00	2229.88	601.07	400.07	94.54
1625.86	1591.10	11.93	61.65	28.74
1526.11	1492.39	173.33	85.95	44.93
1337.89	1304.39	48.34	24.18	15.90
1303.64	1269.53	34.15	15.61	10.73
1255.29	1214.69	8.38	24.36	18.00
1198.04	1152.53	16.34	12.70	10.20
1148.55	1101.22	16.36	1.14	0.98
980.00	962.30	8.43	21.36	22.65
950.35	927.40	61.22	4.60	5.15
874.49	864.15	15.89	0.48	0.60
860.45	852.60	6.90	0.73	0.93
659.27	650.13	7.59	7.52	14.13
646.35	638.39	14.01	1.35	2.60
624.32	628.76	28.15	1.87	3.68
615.14	597.30	21.54	1.51	3.20
605.57	583.40	83.34	0.96	2.12
543.51	534.19	0.51	0.27	0.66
530.69	508.86	4.98	4.22	11.30
484.79	479.16	0.91	3.40	9.94
474.52	466.34	4.57	14.00	42.63
238.89	240.74	98.21	19.79	167.92
237.39	233.63	8.88	8.82	78.62
215.19	209.65	276.59	40.00	426.42
157.60	152.12	2.02	4.23	77.96
133.71	138.84	11.44	2.08	45.11

Table S5. B3LYP/6-31G(d,p) frequencies (cm^{-1}), Infrared intensities ($km.mol^{-1}$), Raman activities ($Å^4/amu$) and intensities ($\times 10^{-24}$ $F^2.m^{-1}.kg^{-1}$) forfor25DCFH$^+$.

Non Scaled frequencies	**Scaled frequencies**	**IR Intensity**	**Raman Activity**	**Raman Intensity**
3686.37	3436.19	1317.78	68.57	5.03
3322.11	3150.29	31.25	101.42	9.79
3313.67	3141.32	12.03	64.35	6.27
2310.35	2306.09	36.40	723.93	158.49
2237.35	2228.75	684.93	445.90	105.49
1601.76	1582.07	21.47	66.34	31.24
1518.44	1511.71	268.99	248.61	127.04
1379.73	1347.73	16.35	11.54	7.19
1284.61	1274.80	15.95	20.11	13.73
1232.37	1211.42	50.12	28.67	21.28
1209.41	1158.70	32.63	5.38	4.29
1034.81	998.31	61.64	23.71	23.78
990.66	971.02	9.00	7.14	7.47
987.43	965.21	0.50	2.03	2.14
918.21	920.24	0.05	1.06	1.20
817.69	824.13	47.33	0.72	0.96
691.40	777.31	2.31	3.53	5.12
646.72	635.90	5.12	4.48	8.69
638.65	610.50	9.15	0.45	0.93
611.72	605.55	23.73	5.14	10.69
608.39	587.09	82.91	1.57	3.41
545.95	528.13	4.06	8.82	22.38
522.91	511.40	0.69	5.04	13.40
504.27	495.92	3.03	7.95	22.11
465.40	440.99	5.32	2.78	9.20
240.53	248.17	332.42	71.72	579.09
238.52	231.72	7.48	3.42	30.88
230.09	224.91	105.08	14.46	137.17
153.47	148.89	1.66	11.19	214.21
131.54	132.74	20.78	2.34	54.84

Table S6. B3LYP/6-31G(d,p) frequencies (cm^{-1}), Infrared intensities ($km.mol^{-1}$), Raman activities ($Å^4/amu$) and intensities ($\times 10^{-24}$ $F^2.m^{-1}.kg^{-1}$) for 34DCFH$^+$.

Non Scaled frequencies	Scaled frequencies	IR Intensity	Raman Activity	Raman Intensity
3710.26	3458.46	1129.24	18.78	1.35
3376.02	3201.13	26.41	97.43	8.96
3371.55	3196.63	49.27	70.61	6.52
2312.13	2308.05	0.19	235.43	51.44
2237.67	2229.42	316.70	212.56	50.25
1596.89	1578.02	17.65	30.52	14.44
1565.79	1545.96	138.07	84.64	41.56
1338.79	1319.71	2.00	2.17	1.40
1309.61	1286.12	4.59	22.72	15.30
1237.29	1211.03	70.22	17.90	13.29
1208.56	1186.73	1.28	5.92	4.54
1155.43	1137.68	9.02	8.97	7.35
1101.88	1082.57	40.05	4.91	4.35
881.69	866.70	32.55	7.67	9.50
827.27	823.03	26.52	1.90	2.54
808.20	798.05	5.42	0.42	0.59
662.61	662.58	2.73	3.31	6.05
648.02	641.12	1.46	0.19	0.36
633.22	627.96	6.08	12.46	24.61
596.73	592.27	25.86	7.11	15.28
583.52	571.82	51.84	0.22	0.50
583.28	557.79	67.21	1.23	2.89
522.15	523.38	5.13	4.59	11.80
491.69	487.49	6.20	4.84	13.80
481.50	464.87	0.15	3.23	9.88
269.33	291.36	144.29	1.26	7.88
249.21	249.20	5.26	1.16	9.27
214.33	205.10	18.39	0.04	0.40
168.73	173.34	15.51	0.76	11.24
134.15	132.66	11.71	7.10	166.49

Table S7. Different types of vibration modes and the corresponding numerical values of frequencies for 23DCFH$^+$.

Frequencies	**PED**	**Mode**	**Atoms**				**PED**	**Mode**	**Atoms**			
(cm^{-1})	**(%)**		**h**	**k**	**l**	**m**	**(%)**		**h**	**k**	**l**	**m**
In plane vibration modes												
3458.46	96%	νNH	11	12	0	0						
3201.13	51%	νCH	2	6	0	0	48%	νCH	5	7	0	0
3196.63	51%	νCH	5	7	0	0	48%	νCH	2	6	0	0
2308.05	91%	νC≡N	8	10	0	0	7%	νCC	3	8	0	0
2229.42	88%	νC≡N	9	11	0	0	7%	νCC	4	9	0	0
1578.02	36%	νC=C	4	5	0	0	16%	νC=C	2	3	0	0
	9%	δCCC	4	3	8	0	6%	δCCC	3	4	5	0
	6%	δCCC	3	4	9	0						
1545.96	43%	νC=C	2	3	0	0	14%	νC=C	4	5	0	0
	8%	δOCH	1	2	6	0	5%	δCCH	3	2	6	0
1319.71	45%	νCC	3	4	0	0	8%	νCC	4	9	0	0
	7%	νCC	3	8	0	0	6%	νC=C	4	5	0	0
	5%	δC=CC	5	4	9	0						
1286.12	16%	δOCH	1	5	7	0	14%	νCC	1	2	0	0
	13%	δOCH	1	2	6	0	12%	νCC	3	4	0	0
	12%	νOC	1	5	0	0	9%	δCCH	4	5	7	0
1211.03	36%	νOC	1	5	0	0	33%	νOC	1	2	0	0
	8%	δOCH	1	5	7	0						
1186.73	20%	δOCH	1	2	6	0	19%	δ0CH	1	5	7	0
	18%	δC=CH	4	5	7	0	17%	δC=CH	3	2	6	0
	9%	νOC	1	5	0	0	8%	νOC	1	2	0	0
1137.68	13%	νCC	3	8	0	0	12%	δC=CC	2	3	8	0
	11%	δC=CC	5	4	9	0	8%	νCC	4	9	0	0
	7%	δCC=C	3	4	5	0	7%	δOC=C	1	5	4	0
1082.57	30%	νOC	1	2	0	0	15%	νOC	1	5	0	0
	11%	δOCH	1	2	6	0	11%	δOCH	1	5	7	0
	8%	δC=CC	2	3	8	0	6%	δC=CH	4	5	7	0
866.70	28%	δCOC	5	1	2	0	18%	νCC	3	4	0	0
	9%	νCC	3	8	0	0	8%	δOCC	1	2	3	0
	8%	νOC=C	1	5	4	0	8%	δC=CH	3	2	6	0
662.58	37%	δCC≡N	4	9	11	3	27%	δCC≡N	4	9	11	4
	15%	νCC	3	8	0	0	8%	νCC	4	9	0	0
	6%	δCCC	3	4	9	0						
641.12	17%	δCC≡N	4	9	11	4	7%	δCC≡N	4	9	11	3
627.96	58%	νCC	3	8	0	0	10%	δCCC	3	4	8	0
	9%	νCC	3	4	0	0						

592.27	44%	δCC≡N	4	9	11	4	19%	δCC≡N	4	9	11	3
	9%	δCC≡N	3	8	2	10	9%	δCC≡N	3	8	1	10
571.82	40%	δCC≡N	4	9	11	4	27%	δCC≡N	4	9	11	3
	23%	δCNH	9	11	12	2						
557.79	56%	δCNH	9	11	12	2	13%	δCC≡N	4	9	11	3
	10%	δCC≡N	3	8	2	10	8%	δCNH	9	11	12	1
	8%	δCC≡N	3	8	1	10						
523.38	36%	δCC≡N	4	9	11	3	30%	δCC≡N	4	9	11	4
	19%	νCC	3	8	0	0						
487.49	29%	δC=CC	2	3	8	0	27%	νCC	4	9	0	0
	8%	δC=CC	2	3	8	0	7%	δC=CC	5	4	9	0
	6%	δCC≡N	3	8	2	10	5%	δCC=C	3	4	5	0
464.87	37%	δCC≡N	3	8	1	10	36%	δCC≡N	3	8	2	10
	18%	δCC≡N	4	9	11	4						
291.36	55%	δCNH	9	11	12	1	21%	δCC≡N	4	9	11	4
	14%	δCNH	9	11	12	2						
249.20	44%	δCC≡N	4	9	11	3	37%	δCC≡N	4	9	11	4
205.10	32%	δCC≡N	3	8	1	10	19%	δCNH	9	11	12	1
	9%	δCC≡N	3	8	2	10						
173.34	38%	δCC≡N	3	8	2	10	31%	δCC≡N	3	8	1	10
	15%	δCC≡N	4	9	11	4						
132.66	36%	δCC≡N	4	9	11	3	21%	δCC≡N	4	9	11	4
	14%	νCC	3	8	0	0	11%	νCC	4	9	0	0
Out of plane vibration modes												
823.03	28%	γCC=CH	9	4	5	7	26%	γCC=CH	3	4	5	7
	20%	γCOCH	2	1	5	7						
798.05	26%	γCC=CH	8	3	2	6	25%	γCC=CH	4	3	2	6
	21%	γCOCH	5	1	2	6						
641.12	18%	γCCCC	9	4	3	8	16%	γC=CCC	2	3	4	9
	13%	γCCC=C	8	3	4	5						
205.10	13%	γC=CCC	2	3	4	9	7%	γCCC=C	8	3	4	5
173.34	5%	γCCCC	9	4	3	8						

Table S8. Different types of vibration modes and the corresponding numerical values of frequencies for 24DCFH$^+$.

Frequencies	**PED**	**Mode**	**Atoms**				**PED**	**Mode**	**Atoms**			
(cm−1)	**(%)**		**h**	**k**	**l**	**m**	**(%)**		**h**	**k**	**l**	**m**
In plane vibration modes												
3439.97	97%	νNH	10	12	0	0						
3194.24	99%	νCH	5	7	0	0						
3151.17	99%	νCH	3	6	0	0						
2313.63	85%	νC≡N	9	11	0	0	15%	νCC	4	9	0	0
2229.88	84%	νC≡N	8	10	0	0	13%	νCC	2	8	0	0
1591.10	27%	νC=C	4	5	0	0	24%	νC=C	2	3	0	0
	11%	νCC	4	9	0	0	6%	δCCH	4	3	6	0
1492.39	38%	νC=C	2	3	0	0	16%	νC=C	4	5	0	0
	10%	νCC	2	8	0	0	9%	νCC	4	9	0	0
	5%	νCC	3	7	0	0						
1304.39	21%	νCC	3	4	0	0	13	νOC	1	5	0	0
	13%	νCC	4	5	0	0	7%	δCCH	4	5	7	0
	6%	νOC	1	2	0	0	6%	δCCH	2	3	6	0
1269.53	19%	νCC	3	4	0	0	18%	νCC	1	2	0	0
	16%	νCCH	4	9	0	0	11%	δOCH	1	5	7	0
	10%	νCC	2	8	0	0						
1214.69	30%	νOC	1	5	0	0	19%	νOC	1	2	0	0
	8%	νCC	2	8	0	0	7%	δCCH	2	3	6	0
	7%	δCCH	4	5	7	0	7%	δOCH	1	5	7	0
1152.53	32%	δOCH	1	5	7	0	25%	νOC	1	5	0	0
	20%	δCCH	4	5	7	0	8%	νCC	4	5	0	0
1101.22	33%	δCCH	4	3	6	0	32%	δCCH	2	3	6	0
	11%	νCC	2	8	0	0	10%	νCC	4	9	0	0
962.30	20%	νCC	3	4	0	0	14%	νOC	1	2	0	0
	13%	δC=CC	2	3	4	0	9%	δOC=C	1	2	3	0
	8%	δCCC	3	4	5	0	7%	νCC	4	9	0	0
927.40	19%	νOC	1	2	0	0	18%	δCOC	5	1	2	0
	16%	νCC	3	4	0	0	12%	δCCH	4	5	7	0
	9%	δOCC	1	5	4	0	8%	νC=C	2	3	0	0
650.13	26%	νCC≡N	2	8	0	0	23%	νCC≡N	4	9	0	0
	9%	δCOC	5	1	2	0	6%	δOC=C	1	2	3	0
	6%	δOCC	1	5	4	0						
638.39	22%	δCC≡N	2	8	2	10	19%	δCNH	8	10	12	5
628.76	31%	δCC≡N	4	9	11	3	10%	δCC≡N	4	9	11	4
	10%	δCNH	8	10	12	6						
597.30	38%	δCC≡N	2	8	1	10	20%	δCC≡N	2	8	2	10

	10%	δCNH	8	10	12	6	7%	δC=CC	3	2	8	0
	6%	δOCC	1	2	8	0						
583.40	69%	δCNH	8	10	12	6	9%	δCC≡N	4	9	11	4
534.19	26%	δCC≡N	4	9	11	4	26%	δCC≡N	2	8	1	10
	18%	δCC≡N	2	8	2	10	7%	δC=CC	5	4	9	0
508.86	33%	δCC≡N	4	9	11	3	29%	δCC≡N	2	8	2	10
	9%	δCC≡N	4	9	11	4						
479.16	34%	δCC≡N	2	8	2	10	29%	δCC≡N	4	9	11	3
	13%	δCC≡N	4	9	11	4						
466.34	23%	νCC	2	8	0	0	20%	νCC	4	9	0	0
	7%	δCC≡N	2	8	1	10	6%	δCCC	3	4	5	0
	6%	δCC	3	4	0	0	6%	δC=CC	2	3	4	0
240.74	46%	δCNH	8	10	12	5	11%	δCC≡N	2	8	2	10
	9%	νC≡N	8	10	0	0	6%	δCC≡N	4	9	11	3
	6%	δCNN	2	8	1	10						
233.63	25%	δCC≡N	2	8	1	10	23%	δCC≡N	2	8	2	10
	17%	δCC≡N	4	9	11	4	8%	δCCC	3	4	9	0
	8%	δCCC	5	4	9	0	6%	δC=CC	3	2	8	0
209.65	59%	δCNH	8	10	12	5	12%	νC≡N	8	10	0	0
	5%	δCC≡N	2	8	2	10						
152.12	29%	δCC≡N	2	8	1	10	22%	δCC≡N	2	8	2	10
	12%	δOCC	1	2	8	0	11%	δCCC	3	2	8	0
	8%	δCCC	3	4	9	0	8%	δC=CC	5	4	9	0
138.84	20%	δCNH	8	10	12	5						
Out of plane vibration modes												
864.15	26%	γCCCH	8	2	3	6	25%	γHCCC	6	3	4	9
	21%	γOC=CH	1	2	3	6	17%	γCCCH	5	4	3	6
852.60	42%	γCCCH	9	4	5	7	25%	γCCCH	3	4	5	7
	13%	γCOCH	2	1	5	7						
638.39	14%	γCOC=C	5	1	2	3	12%	γOC=CC	1	2	3	4
	7%	γCOC=C	2	1	5	4	6%	γOC=CC	1	2	3	4
	6%	γOCCH	1	2	3	6						
628.76	9%	γCCCH	3	4	5	7	5%	γCCCO	3	4	5	1
	5%	γCCCH	5	4	3	6						
138.84	24%	γCCCC	8	2	3	4	12%	γCOCC	5	1	2	8
	9%	γCC=CH	8	2	3	6	8%	γCC=CO	9	4	5	1
	8%	γC=CCC	2	3	4	9						

Table S9. Different types of vibration modes and the corresponding numerical values of frequencies for 25DCFH$^+$.

Fréquence	PED	Mode	Atoms				PED	Mode	Atoms			
(cm−1)	(%)		h	k	l	m	(%)		h	k	l	m
In plane vibration modes												
3436.19	97%	νNH	10	12	0	0						
3150.29	50%	νCH	4	7	0	0	49%	νCH	3	6	0	0
3141.32	50%	νCH	3	6	0	0	49%	νCH	4	7	0	0
2306.09	84%	νC≡N	9	11	0	0	15%	νCC	5	9	0	0
2228.75	83%	νC≡N	8	10	0	0	14%	νCC	2	8	0	0
1582.07	40%	νC=C	4	5	0	0	27%	νC=C	2	3	0	0
	7%	νCC	5	9	0	0	7%	δCCH	3	4	7	0
	5%	δCCH	4	3	6	0						
1511.71	36%	νC=C	2	3	0	0	19%	νC=C	4	5	0	0
	15%	δCOC	5	1	2	0	11%	νCC	5	9	0	0
	8%	νCC	2	8	0	0						
1347.73	19%	νOC	1	2	0	0	15	νOC	1	5	0	0
	13%	νCC	3	4	0	0	11%	δCCH	2	3	6	0
	8%	δCCH	5	4	7	0	6%	δOCC	1	2	8	0
1274.80	36%	νOC	1	2	0	0	20%	νCC	3	4	0	0
	16%	νCC	2	8	0	0	7%	νOC	1	5	0	0
	7%	δCCH	5	4	7	0						
1211.42	31%	νOC	1	2	0	0	22%	νCC	3	4	0	0
	11%	δCCH	2	3	6	0	8%	νCC	5	9	0	0
	8%	νC=C	4	5	0	0	6%	νCCH	4	3	6	0
1158.70	28%	δCCH	3	4	7	0	21%	δC=CH	5	4	7	0
	20%	δCCH	4	3	6	0	13%	δC=CH	2	3	6	0
	6%	νCC	2	8	0	0	6%	νCC	5	9	0	0
998.31	14%	δC=CH	5	4	7	0	13%	δCCH	3	4	7	0
	12%	δCCH	4	3	6	0	11%	δCCH	5	1	2	0
	10%	νCC	1	5	0	0	8%	νCC	3	4	0	0
971.02	26%	δCOC	5	1	2	0	14%	δCCH	3	4	7	0
	13%	δC=CC	4	3	6	0	13%	δCCH	5	4	7	0
	9%	νCC	3	4	0	0	9%	νCC	2	8	0	0
965.21	24%	δCCH	2	3	6	0	13%	δOCC	1	2	3	0
	13%	νCC	3	4	0	0	12%	δOC	1	5	0	0
	8%	νCC	5	9	0	0	7%	δCCH	4	3	6	0
920.24	33%	δHCCH	6	3	4	7	13%	δCCCH	5	4	3	6
	11%	δCCCH	8	2	3	6	10%	δOC=CH	1	5	4	7
	8%	δOCC	9	5	4	7						
824.13	29%	δCCCH	9	5	4	7	16%	δC=CCH	2	3	4	7

	12%	δOC=CH	1	5	4	7						
777.31	19%	δC=CCH	5	4	3	6	16%	δOC=CH	1	2	3	6
	12%	δOC=CH	1	5	4	7	6%	δCC≡N	2	8	10	1
	5%	δCC≡N	2	8	10	2						
635.90	34%	δCC≡N	2	8	10	2	18%	δOCC	1	2	8	0
	13%	δOC=C	1	2	3	0	7%	δC=CC	4	5	9	0
	6%	νCC	2	8	0	0	5%	δCNH	8	10	6	12
610.50	32%	δCC≡N	2	8	10	2	21%	δCNH	8	10	6	12
	8%	δOCC	1	2	8	0	6%	νCC	5	9	0	0
	6%	δCC≡N	2	8	10	1	δOC=C	1	2	3	0	
605.55	37%	δCC≡N	5	9	11	3	27%	δCC≡N	5	9	11	4
	6%	δCC≡N	2	8	10	1						
587.09	76%	δCNH	8	10	6	12	6%	νOCC	1	2	8	0
528.13	23%	δCC≡N	2	8	10	2	19%	δCC≡N	5	9	11	4
	15%	δCOC	5	1	2	0	13%	δC=CC	4	5	9	0
	11%	δCC≡N	2	8	10	1	6%	νCC	5	9	0	0
511.40	64%	δCC≡N	2	8	10	1	27%	δCC≡N	2	8	10	2
495.92	30%	δCC≡N	2	8	10	2	24%	δCC≡N	2	8	10	1
	9%	δCC≡N	5	9	11	4	8%	δCOC	5	1	2	0
	8%	νCC	2	8	0	0	6%	νCC	5	9	0	0
440.99	55%	δCC≡N	5	9	11	3	34%	δCC≡N	5	9	11	4
248.17	34%	δCC≡N	2	8	10	1	33%	δCNH	8	10	5	12
	18%	δCC≡N	2	8	10	2	7%	νC≡N	8	10	0	0
231.72	35%	δCC≡N	2	8	10	2	24%	δOCC	1	2	8	0
	19%	δCC≡N	5	9	11	4	18%	δC=CC	4	5	9	0
224.91	24%	δCNH	8	10	5	12	19%	δCC≡N	5	9	11	3
	14%	δCC≡N	5	9	11	4	10%	δCC=CC	4	3	2	8
	7%	δCC=CC	3	4	5	9	6%	δCC=CH	9	5	4	7
148.89	31%	δC=CC	4	5	9	0	28%	δCC≡N	2	8	10	2
	27%	δOCC	1	2	8	0	7%	δCC≡N	5	9	11	4
132.74	27%	δCCCC	4	3	2	8	13%	δCNH	8	10	5	12
	12%	δCC=CC	3	4	5	9	12%	δCOCC	5	1	2	9
	11%	δCC=CH	8	2	3	6	7%	δCC=CH	9	5	4	7
Out of plane vibration modes												
920.24	7%	γC=COC	4	5	1	2						
824.13	13%	γC=COC	4	5	1	2						
777.31	20%	γC=COC	4	5	1	2						
605.55	5%	γC=COC	4	5	1	2						

Table S10. Different types of vibration modes and the corresponding numerical values of frequencies for 34DCFH$^+$.

Fréquence	PED	Mode	Atoms				PED	Mode	Atoms			
(cm−1)	(%)		h	k	l	m	(%)		h	k	l	m
in-plane vibrational mode												
3458.46	96%	νNH	11	12	0	0						
3201.13	51%	νCH	2	6	0	0	48%	νCH	5	7	0	0
3196.63	51%	νCH	5	7	0	0	48%	νCH	2	6	0	0
2308.05	91%	νC≡N	8	10	0	0	7%	νCC	3	8	0	0
2229.42	88%	νC≡N	9	11	0	0	7%	νCC	4	9	0	0
1578.02	36%	νC=C	4	5	0	0	16%	νC=C	2	3	0	0
	9%	δCCC	4	3	8	0	6%	δCCC	3	4	5	0
	6%	δCCC	3	4	9	0						
1545.96	43%	νC=C	2	3	0	0	14%	νC=C	4	5	0	0
	8%	δOCH	1	2	6	0	5%	δCCH	3	2	6	0
1319.71	45%	νCC	3	4	0	0	8%	νCC	4	9	0	0
	7%	νCC	3	8	0	0	6%	νC=C	4	5	0	0
	5%	δC=CC	5	4	9	0						
1286.12	16%	δOCH	1	5	7	0	14%	νCC	1	2	0	0
	13%	δOCH	1	2	6	0	12%	νCC	3	4	0	0
	12%	νOC	1	5	0	0	9%	δCCH	4	5	7	0
1211.03	36%	νOC	1	5	0	0	33%	νOC	1	2	0	0
	8%	δOCH	1	5	7	0						
1186.73	20%	δOCH	1	2	6	0	19%	δ0CH	1	5	7	0
	18%	δC=CH	4	5	7	0	17%	δC=CH	3	2	6	0
	9%	νOC	1	5	0	0	8%	νOC	1	2	0	0
1137.68	13%	νCC	3	8	0	0	12%	δC=CC	2	3	8	0
	11%	δC=CC	5	4	9	0	8%	νCC	4	9	0	0
	7%	δCC=C	3	4	5	0	7%	δOC=C	1	5	4	0
1082.57	30%	νOC	1	2	0	0	15%	νOC	1	5	0	0
	11%	δOCH	1	2	6	0	11%	δOCH	1	5	7	0
	8%	δC=CC	2	3	8	0	6%	δC=CH	4	5	7	0
866.70	28%	δCOC	5	1	2	0	18%	νCC	3	4	0	0
	9%	νCC	3	8	0	0	8%	δOCC	1	2	3	0
	8%	νOC=C	1	5	4	0	8%	δC=CH	3	2	6	0
662.58	37%	δCC≡N	4	9	11	3	27%	δCC≡N	4	9	11	4
	15%	νCC	3	8	0	0	8%	νCC	4	9	0	0
	6%	δCCC	3	4	9	0						
641.12	17%	δCC≡N	4	9	11	4	7%	δCC≡N	4	9	11	3
627.96	58%	νCC	3	8	0	0	10%	δCCC	3	4	8	0
	9%	νCC	3	4	0	0						

592.27	44%	δCC≡N	4	9	11	4	19%	δCC≡N	4	9	11	3
	9%	δCC≡N	3	8	2	10	9%	δCC≡N	3	8	1	10
571.82	40%	δCC≡N	4	9	11	4	27%	δCC≡N	4	9	11	3
	23%	δCNH	9	11	12	2						
557.79	56%	δCNH	9	11	12	2	13%	δCC≡N	4	9	11	3
	10%	δCC≡N	3	8	2	10	8%	δCNH	9	11	12	1
	8%	δCC≡N	3	8	1	10						
523.38	36%	δCC≡N	4	9	11	3	30%	δCC≡N	4	9	11	4
	19%	νCC	3	8	0	0						
487.49	29%	δC=CC	2	3	8	0	27%	νCC	4	9	0	0
	8%	δC=CC	2	3	8	0	7%	δC=CC	5	4	9	0
	6%	δCC≡N	3	8	2	10	5%	δCC=C	3	4	5	0
464.87	37%	δCC≡N	3	8	1	10	36%	δCC≡N	3	8	2	10
	18%	δCC≡N	4	9	11	4						
291.36	55%	δCNH	9	11	12	1	21%	δCC≡N	4	9	11	4
	14%	δCNH	9	11	12	2						
249.20	44%	δCC≡N	4	9	11	3	37%	δCC≡N	4	9	11	4
205.10	32%	δCC≡N	3	8	1	10	19%	δCNH	9	11	12	1
	9%	δCC≡N	3	8	2	10						
173.34	38%	δCC≡N	3	8	2	10	31%	δCC≡N	3	8	1	10
	15%	δCC≡N	4	9	11	4						
132.66	36%	δCC≡N	4	9	11	3	21%	δCC≡N	4	9	11	4
	14%	νCC	3	8	0	0	11%	νCC	4	9	0	0
Modes	hors	plan										
823.03	28%	γCC=CH	9	4	5	7	26%	γCC=CH	3	4	5	7
	20%	γC0CH	2	1	5	7						
798.05	26%	γCC=CH	8	3	2	6	25%	γCC=CH	4	3	2	6
	21%	γCOCH	5	1	2	6						
641.12	18%	γCCCC	9	4	3	8	16%	γC=CCC	2	3	4	9
	13%	γCCC=C	8	3	4	5						
205.10	13%	γC=CCC	2	3	4	9	7%	γCCC=C	8	3	4	5
173.34	5%	γCCCC	9	4	3	8						

Table S11. Different types of vibration modes and the corresponding numerical values of frequencies for 23DCFH$^+$.

T = 10,0 K											
$J'_{K_{-1}K_{+1}} \leftarrow J''_{K_{-1}K_{+1}}$	$F'_1 \leftarrow F''_1$	$F'_2 \leftarrow F''_2$	ν_{cal}	Err	Int	$J'_{K_{-1}K_{+1}} \leftarrow J''_{K_{-1}K_{+1}}$	$F'_1 \leftarrow F''_1$	$F'_2 \leftarrow F''_2$	ν_{cal}	Err	Int
	14 ← 13	14 ← 14	44088.0058	0.0019	0.1878		21 ← 21	22 ← 22	43009.2082	0.3998	0.0004
	12 ← 11	12 ← 12	44088.0238	0.0019	0.2202		21 ← 21	20 ← 20	43009.2082	0.3998	0.0597
$13_{9\,4} \leftarrow 12_{9\,3}$	13 ← 12	12 ← 11	44088.0752	0.0019	30.6600	$22_{0\,22} \leftarrow 21_{1\,21}$	21 ← 21	21 ← 21	43009.2103	0.3998	0.0035
	13 ← 12	14 ← 13	44088.0777	0.0019	35.8040		21 ← 22	22 ← 23	43009.8637	0.0000	0.0000
	12 ← 11	11 ← 10	44088.2027	0.0019	28.1670		21 ← 20	20 ← 19	43009.8950	0.3998	0.4670
	22 ← 24	23 ← 23	41101.0056	0.0000	0.0000		20 ← 20	21 ← 21	41099.1943	0.3466	0.0967
	24 ← 24	25 ← 25	41101.0141	0.3994	0.0001		20 ← 20	19 ← 19	41099.1943	0.3466	0.0664
$23_{2\,21} \leftarrow 23_{1\,22}$	22 ← 22	21 ← 21	41101.0188	0.3994	0.0224	$21_{1\,21} \leftarrow 20_{1\,20}$	20 ← 20	20 ← 20	41099.1967	0.3466	0.1168
	23 ← 24	24 ← 24	41101.2130	0.0000	0.0000		20 ← 21	21 ← 22	41099.8477	0.0000	0.0001
	23 ← 22	22 ← 22	41101.2264	0.0000	0.0000		20 ← 19	19 ← 18	41099.8804	0.3466	0.4949
	17 ← 18	18 ← 18	39193.2204	0.0000	0.0000		18 ← 19	19 ← 19	39191.1474	0.0000	0.0000
	17 ← 16	16 ← 16	39193.2343	0.1500	0.0007		18 ← 17	17 ← 17	39191.1615	0.2157	0.1396
$18_{3\,16} \leftarrow 17_{3\,15}$	18 ← 17	17 ← 17	39193.2445	0.1500	0.0001	$19_{1\,18} \leftarrow 18_{2\,17}$	19 ← 18	18 ← 18	39191.1677	0.2157	0.0018
	19 ← 18	20 ← 19	39193.4946	0.1500	40.6350		18 ← 19	19 ← 20	39191.4638	0.0000	0.0000
	17 ← 16	16 ← 15	39193.4959	0.1500	31.4570		18 ← 17	17 ← 16	39191.4953	0.2157	34.1230
	22 ← 23	22 ← 23	39189.0722	0.0000	0.0015		19 ← 17	18 ← 17	37280.0122	0.0000	0.0000
	22 ← 21	22 ← 21	39189.0991	0.0000	0.0003		17 ← 17	16 ← 16	37280.3073	0.1808	0.0008
$22_{2\,20} \leftarrow 22_{1\,21}$	22 ← 23	23 ← 24	39189.1240	0.0000	0.0001	$18_{1\,17} \leftarrow 17_{1\,16}$	17 ← 17	18 ← 18	37280.3075	0.1808	0.0000
	22 ← 23	21 ← 22	39189.1261	0.0000	0.0033		17 ← 18	18 ← 19	37280.8708	0.0000	0.0001
	22 ← 21	21 ← 20	39189.1532	0.0000	0.0032		17 ← 16	16 ← 15	37280.9037	0.1808	34.7520

Table S12. Different types of vibration modes and the corresponding numerical values of frequencies for 24DCFH$^+$.

T = 10,0 K												
$J'_{K_{-1}K_{+1}} \leftarrow J''_{K_{-1}K_{+1}}$	$F'_1 \leftarrow F''_1$	$F'_2 \leftarrow F''_2$	ν_{cal}	*Err*	*Int*	$J'_{K_{-1}K_{+1}} \leftarrow J''_{K_{-1}K_{+1}}$	$F'_1 \leftarrow F''_1$	$F'_2 \leftarrow F''_2$	ν_{cal}	*Err*	*Int*
	24 ← 23	23 ← 23	40597.2888	0.0009	0.3216		24 ← 23	23 ← 22	40222.1067	0.0010	192.7600
	26 ← 25	25 ← 25	40597.2914	0.0009	0.2965		23 ← 22	22 ← 21	40222.1067	0.0010	184.5100
$25_{1\,24} \leftarrow 24_{1\,23}$	25 ← 24	24 ← 24	40597.2938	0.0009	0.3108	$24_{2\,22} \leftarrow 23_{2\,21}$	24 ← 23	25 ← 24	40222.1070	0.0010	209.5300
	24 ← 23	23 ← 22	40597.3301	0.0009	177.7300		23 ← 22	24 ← 23	40222.1071	0.0010	201.3100
	26 ← 25	25 ← 26	40597.3310	0.0000	0.0001		25 ← 24	24 ← 23	40222.1093	0.0010	201.3300
	24 ← 24	25 ← 25	39114.0044	0.0008	0.0969		23 ← 22	23 ← 23	38567.1130	0.0009	0.4104
	24 ← 24	24 ← 24	39114.0044	0.0008	0.0933		22 ← 21	22 ← 22	38567.1135	0.0009	0.4209
$24_{7\,18} \leftarrow 23_{7\,17}$	24 ← 24	23 ← 23	39114.0044	0.0008	0.0892	$23_{2\,21} \leftarrow 22_{2\,20}$	24 ← 23	24 ← 24	38567.1160	0.0009	0.3853
	23 ← 22	22 ← 22	39114.6801	0.0008	0.3775		23 ← 22	22 ← 21	38567.2308	0.0009	204.6000
	23 ← 22	22 ← 21	39114.7048	0.0008	191.0500		22 ← 21	21 ← 20	38567.2314	0.0009	195.4500
	23 ← 22	22 ← 21	38471.5809	0.0000	0.0000		26 ← 26	25 ← 25	37615.4561	0.0378	0.0015
	21 ← 20	20 ← 19	38471.5810	0.0000	0.0000		26 ← 26	27 ← 27	37615.4562	0.0378	0.0017
$22_{10\,13} \leftarrow 21_{11\,11}$	23 ← 22	24 ← 23	38471.5830	0.0000	0.0000	$25_{25\,1} \leftarrow 25_{23\,2}$	26 ← 26	26 ← 26	37615.4595	0.0378	0.0016
	22 ← 21	21 ← 20	38471.5872	0.0000	0.0000		24 ← 24	23 ← 23	37615.5098	0.0378	0.0014
	22 ← 21	23 ← 22	38471.5895	0.0000	0.0000		24 ← 24	24 ← 24	37615.5132	0.0378	0.0015
	23 ← 24	24 ← 24	37082.0299	0.0000	0.0000		23 ← 22	22 ← 17	37006.1699	0.0009	0.3834
	23 ← 22	22 ← 23	37082.2418	0.0000	0.0002		24 ← 23	23 ← 16	37006.1729	0.0009	0.3867
$24_{0\,24} \leftarrow 23_{0\,23}$	25 ← 24	24 ← 25	37082.2456	0.0000	0.0001	$24_{1\,24} \leftarrow 23_{1\,23}$	25 ← 24	24 ← 18	37006.1730	0.0009	0.3495
	23 ← 22	22 ← 21	37082.2549	0.0009	197.8200		23 ← 22	21 ← 19	37006.4807	0.0009	196.6600
	24 ← 23	23 ← 22	37082.2574	0.0009	206.6500		25 ← 24	25 ← 15	37006.4829	0.0009	232.5000

Table S13. Different types of vibration modes and the corresponding numerical values of frequencies for 25DCFH$^+$.

T = 10,0 K												
$J'_{K_{-1}K_{+1}} \leftarrow J''_{K_{-1}K_{+1}}$	$F'_1 \leftarrow F''_1$	$F'_2 \leftarrow F''_2$	ν_{cal}	*Err*	*Int*	$J'_{K_{-1}K_{+1}} \leftarrow J''_{K_{-1}K_{+1}}$	$F'_1 \leftarrow F''_1$	$F'_2 \leftarrow F''_2$	ν_{cal}	*Err*	*Int*
	23 ← 22	23 ← 23	42445.0131	0.0885	0.1864		22 ← 23	23 ← 22	41602.1340	0.0000	0.0009
	22 ← 21	22 ← 22	42445.0153	0.0885	0.2081		22 ← 21	22 ← 21	41602.1491	0.0748	111.4300
$23_{3\,20} \leftarrow 22_{3\,19}$	24 ← 23	24 ← 24	42445.0156	0.0885	0.2374	$23_{2\,21} \leftarrow 22_{2\,20}$	24 ← 23	24 ← 23	41602.1507	0.0748	121.5500
	22 ← 22	22 ← 21	42445.3928	0.0000	0.0003		22 ← 21	21 ← 20	41602.1580	0.0748	106.4200
	22 ← 21	21 ← 20	42445.8158	0.0885	101.8400		24 ← 23	25 ← 24	41602.1609	0.0748	126.7500
	24 ← 24	24 ← 23	41082.0008	0.0000	0.0001		26 ← 24	25 ← 25	40804.0365	0.0000	0.0000
	23 ← 23	23 ← 24	41082.0027	0.0000	0.0003		24 ← 25	23 ← 24	40804.2370	0.0000	0.0001
$24_{1\,25} \leftarrow 23_{1\,22}$	25 ← 24	25 ← 25	41082.0144	0.0825	0.1981	$25_{0\,25} \leftarrow 24_{0\,24}$	24 ← 25	25 ← 26	40804.2398	0.0000	0.0001
	23 ← 23	22 ← 22	41082.9817	0.0825	0.1990		24 ← 23	24 ← 23	40804.2471	0.1013	106.1400
	23 ← 22	22 ← 21	41082.9927	0.0825	100.7000		24 ← 23	23 ← 22	40804.2483	0.1013	101.7800
	25 ← 25	25 ← 26	40798.0073	0.0000	0.0003		21 ← 22	22 ← 21	40575.2699	0.0000	0.0000
	25 ← 23	25 ← 24	40798.0175	0.0000	0.0004		22 ← 21	21 ← 20	40575.2807	0.0783	113.2200
$25_{1\,25} \leftarrow 24_{1\,24}$	24 ← 24	23 ← 23	40798.5649	0.1013	0.1817	$22_{3\,19} \leftarrow 21_{3\,18}$	22 ← 21	23 ← 22	40575.2824	0.0783	124.1100
	24 ← 24	25 ← 25	40798.5671	0.1013	0.1658		21 ← 20	20 ← 19	40575.2877	0.0783	107.9600
	24 ← 23	23 ← 22	40798.8340	0.1013	101.7600		23 ← 22	24 ← 23	40575.2898	0.0783	129.6100
	21 ← 22	21 ← 21	39966.0181	0.0000	0.0001		22 ← 23	23 ← 22	39529.3394	0.0000	0.0001
	23 ← 22	23 ← 23	39966.0442	0.0667	0.2822		22 ← 21	22 ← 21	39529.3738	0.0724	112.9600
$22_{2\,20} \leftarrow 21_{2\,19}$	22 ← 21	22 ← 22	39966.0679	0.0667	0.2377	$23_{1\,22} \leftarrow 22_{1\,21}$	22 ← 23	22 ← 23	39529.3743	0.0000	0.0001
	21 ← 21	21 ← 20	39966.3362	0.0000	0.0026		22 ← 21	21 ← 20	39529.3813	0.0724	107.9100
	21 ← 20	20 ← 19	39966.8923	0.0667	113.7700		22 ← 23	23 ← 24	39529.3833	0.0000	0.0001

Table S14. Different types of vibration modes and the corresponding numerical values of frequencies for 34DCFH$^+$.

T = 10,0 K												
$J'_{K_{-1}K_{+1}} \leftarrow J''_{K_{-1}K_{+1}}$	$F'_1 \leftarrow F''_1$	$F'_2 \leftarrow F''_2$	ν_{cal}	*Err*	*Int*	$J'_{K_{-1}K_{+1}} \leftarrow J''_{K_{-1}K_{+1}}$	$F'_1 \leftarrow F''_1$	$F'_2 \leftarrow F''_2$	ν_{cal}	*Err*	*Int*
	16 ← 16	17 ← 16	62854.0286	0.0000	0.0000		15 ← 15	16 ← 15	58899.0796	0.0000	0.0000
	16 ← 14	16 ← 15	62854.0419	0.0000	0.0000		15 ← 15	14 ← 15	58899.0854	0.0000	0.0000
$16_{160} \leftarrow 15_{150}$	16 ← 16	17 ← 17	62854.0562	0.0000	0.0033	$15_{150} \leftarrow 14_{140}$	14 ← 13	14 ← 14	58899.1042	0.1370	0.2120
	16 ← 16	16 ← 16	62854.0574	0.0000	0.0029		16 ← 15	16 ← 16	58899.1052	0.1370	0.1831
	16 ← 16	16 ← 15	62854.0639	0.0000	0.0003		15 ← 14	16 ← 15	58899.1851	0.1370	48.3690
	15 ← 13	15 ← 14	58061.3355	0.0000	0.0004		15 ← 14	15 ← 15	58032.0627	0.1360	0.2119
	15 ← 15	15 ← 16	58061.3489	0.0000	0.0007		16 ← 15	16 ← 16	58032.0858	0.1360	0.1342
$15_{141} \leftarrow 14_{132}$	15 ← 15	15 ← 15	58061.4652	0.1360	0.1263	$15_{142} \leftarrow 14_{131}$	14 ← 13	14 ← 14	58032.0871	0.1360	0.1719
	15 ← 13	14 ← 13	58061.5725	0.0000	0.0315		15 ← 13	14 ← 13	58032.0967	0.0000	0.0545
	14 ← 13	13 ← 12	58061.6748	0.1360	32.8460		14 ← 13	13 ← 12	58032.1998	0.1360	32.8180
	14 ← 14	15 ← 14	54945.0041	0.0000	0.0024		16 ← 16	15 ← 15	54943.0755	0.0000	0.0001
	13 ← 13	13 ← 12	54945.1979	0.0000	0.0001		16 ← 16	17 ← 17	54943.0769	0.0000	0.0001
$14_{140} \leftarrow 13_{131}$	13 ← 12	13 ← 13	54945.2346	0.1114	0.2585	$17_{116} \leftarrow 16_{115}$	16 ← 16	16 ← 16	54943.1109	0.0000	0.0001
	15 ← 14	15 ← 15	54945.2356	0.1114	0.2207		18 ← 17	18 ← 18	54943.3027	0.0000	0.0001
	13 ← 12	12 ← 11	54945.3219	0.1114	41.1980		14 ← 12	14 ← 13	54943.3575	0.0000	0.0001
	14 ← 12	14 ← 13	54118.4951	0.0000	0.0006		14 ← 12	14 ← 13	54061.4776	0.0000	0.0011
	14 ← 14	14 ← 15	54118.5079	0.0000	0.0010		14 ← 14	14 ← 15	54061.4898	0.0000	0.0010
$14_{131} \leftarrow 13_{122}$	14 ← 14	14 ← 14	54118.6263	0.1104	0.1243	$14_{132} \leftarrow 13_{121}$	14 ← 14	14 ← 14	54061.6098	0.1106	0.1115
	14 ← 12	13 ← 12	54118.7373	0.1104	0.1290		14 ← 12	13 ← 12	54061.7203	0.1106	0.3879
	13 ← 12	12 ← 11	54118.8143	0.1104	33.5020		13 ← 12	12 ← 11	54061.7994	0.1106	33.4510

Cartesian coordinates for the optimized and corrected geometries of:

1. dicyano-2,3-furan (23DCF)

C	-0.62329700	0.13934500	0.00005000
C	0.64265900	-0.41844600	-0.00004700
C	0.43715500	-1.85466000	0.00011600
C	-0.91382000	-2.02705100	0.00000200
O	-1.56150600	-0.83829100	-0.00010500
H	1.20158000	-2.61283400	0.00017200
H	-1.53987000	-2.89755500	-0.00001800
C	-1.05864100	1.48035000	0.00002600
N	-1.40550500	2.59948600	0.00003400
C	1.87318900	0.26974900	-0.00002600
N	2.90732900	0.82050900	-0.00002800

2. dicyano-2,4-furan (24DCF)

C	-1.08201400	-0.06152100	-0.00003800
C	0.02041600	-0.87140200	0.00025000
C	1.15776200	0.02957700	-0.00002000
C	0.62713000	1.29885500	-0.00001900
O	-0.71144500	1.25720600	0.00008400
H	0.03919700	-1.94796900	0.00029300
H	1.08094100	2.27084400	-0.00006000
C	-2.46147200	-0.35291800	-0.00005000
N	-3.60217200	-0.62046800	-0.00009800
C	2.52517100	-0.31395300	-0.00004500
N	3.65978000	-0.60625900	-0.00008100

3. dicyano-2,5-furan (25DCF)

C	1.08970100	0.18578300	0.00005100
C	0.71771300	1.50982200	-0.00011900
C	-0.71770300	1.50983000	0.00009300
C	-1.08970500	0.18579400	0.00001800
O	-0.00000600	-0.62608900	-0.00018500
H	1.37987500	2.35940700	-0.00007700
H	-1.37985900	2.35941900	0.00017600
C	2.36073200	-0.42437800	0.00003800
N	3.43078300	-0.90163200	0.00007100
C	-2.36073400	-0.42437100	0.00002400
N	-3.43078000	-0.90163600	0.00004300

4. dicyano-3,4-furan (34DCF)

C	-1.44574700	1.09341500	-0.00019800
C	-0.12530300	0.73451700	0.00007800
C	-0.12526700	-0.73451200	-0.00005400

C	-1.44569400	-1.09347400	0.00009700
O	-2.23652400	-0.00004900	0.00005600
H	-1.93586500	-2.04702500	0.00018500
C	0.98982300	-1.59729400	-0.00003300
N	1.91477600	-2.31648900	-0.00003400
C	0.98974900	1.59734800	0.00003900
N	1.91469700	2.31655100	0.00003600
H	-1.93596500	2.04694200	-0.00026700

5. Protonated dicyano-2,3-furan (23DCFH)$^+$

O	-1.80590500	0.03443600	-0.00026200
C	-0.51166900	0.43566000	-0.00032900
C	0.29161500	-0.69062900	0.00001400
C	-0.58347200	-1.84784900	0.00036700
C	-1.83848700	-1.31892200	0.00016600
H	-0.29862500	-2.88613500	0.00076800
H	-2.81444600	-1.76301800	0.00011500
C	-0.20295400	1.81134700	-0.00064800
C	1.70127800	-0.66474100	0.00023700
N	0.11470800	2.93911800	0.00069200
N	2.87283400	-0.64939200	-0.00038200
H	3.88183600	-0.62199200	0.00126700

6. Protonated dicyano-2,4-furan (24DCFH)$^+$

O	0.74425000	1.25075400	-0.00012800
C	1.11675200	-0.06742500	-0.00019900
C	0.01656100	-0.88032800	-0.00004000
C	-1.10516600	0.04002100	0.00020300
C	-0.59334100	1.31700800	0.00016800
H	-0.00588000	-1.95682500	0.00022300
H	-1.03863600	2.29292700	0.00013900
C	2.49630200	-0.35838500	-0.00046000
C	-2.47728600	-0.28417600	0.00037900
N	3.63729000	-0.62470700	0.00038300
N	-3.61361300	-0.56972800	-0.00043200
H	-4.59299900	-0.81288200	0.00174200

7. Protonated dicyano-2,5-furan (25DCFH)$^+$

O	0.02974600	-0.62075000	0.01247600
C	-1.04257700	0.21395600	0.01190300
C	-0.66806600	1.53724100	0.00152100
C	0.76714100	1.51483000	-0.00745800
C	1.12291400	0.18636500	-0.00009200
H	-1.31170400	2.40093900	-0.00124300

H	1.43432200	2.36040800	-0.01835500
C	-2.32047100	-0.38157000	0.02430700
C	2.38492700	-0.44223200	-0.00289000
N	-3.38964500	-0.85527200	-0.04817300
N	3.44535600	-0.94049400	-0.00520300
H	-4.27247000	-1.25676100	0.23827800

8. Protonated dicyano-3,4-furan (34DCFH)$^{+}$

O	-2.21391800	-0.49441500	-0.00004900
C	-1.20660800	-1.39236400	0.00013700
C	-0.01648700	-0.71709100	0.00001500
C	-0.33279500	0.71748100	-0.00011800
C	-1.70066800	0.75360900	-0.00005300
H	-1.47170300	-2.43123500	0.00006700
H	-2.40036600	1.56598200	-0.00054800
C	0.59316100	1.78069500	0.00009100
C	1.27308300	-1.28702700	0.00036700
N	1.40224000	2.62814400	0.00003000
N	2.35934100	-1.72614500	-0.00039200
H	3.29806700	-2.09782800	0.00108300