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Supplementary Material

Characterization of the simplest sulfenyl thiocyanate: isomers, spectroscopy and implications of astrophysical and biological relevance

Aicha Benabdelkrim, Ala Eddine Tourchi, Dalila Hammoutène, Saida Ben Yaghlane, Hassan H. Abdallah, Roberto Linguerri, and Majdi Hochlaf

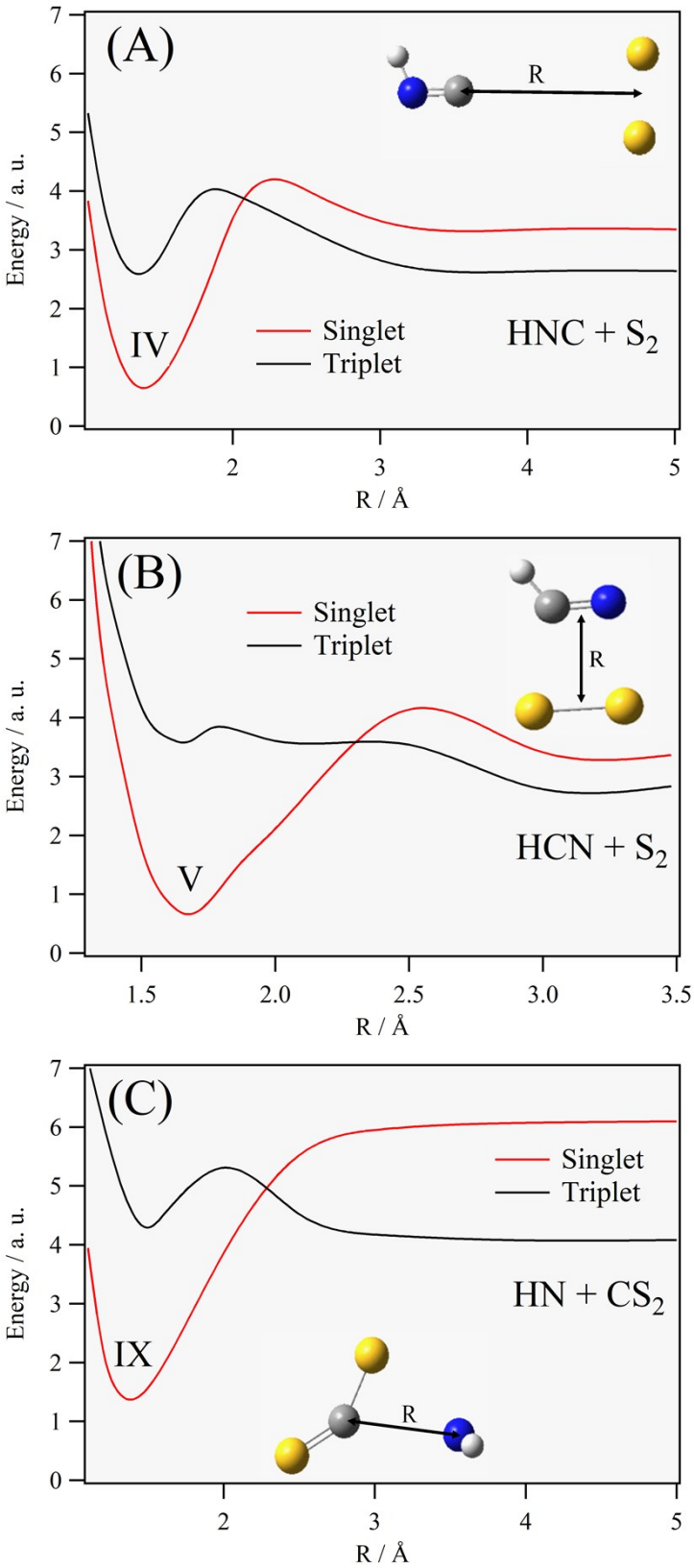


Figure S1: (R)CCSD(T)/aug-cc-pVTZ one-dimensional cuts of the lowest singlet (in red) and triplet (in black) potential energy surfaces of the HC_2NS_2 system along the R coordinate as defined in each panel. The remaining internal coordinates were kept fixed at their equilibrium values in **IV** (in (A)), in **V** (in (B)) and in **IX** (in (C)) as given in Table 1. The reference energy is the energy of compound **I** at equilibrium.

Table S1: Optimized equilibrium geometries (R_i in Å, angles in degrees) and harmonic frequencies (ω_i, in cm^{-1}) of all the molecules calculated with different methods. The total energy (E, in Hartree) of each structure is also given. For planar structures, ω_{1-7} are of a' symmetry and ω_{8-9} are of a" symmetry. For C_1 species all frequencies are of a symmetry. See Figure I for the numbering of the structures. aVXZ denotes aug-cc-pVXZ basis set and aV(X+d)Z is for aug-cc-pV(X+d)Z.

Method	Base	R_1	R_2	R_3	R_4	θ_1	θ_2	θ_3	τ_1	τ_2	ω_1	ω_2	ω_3	ω_4	ω_5	ω_6	ω_7	ω_8	ω_9	E
I																				
MP2	aVDZ	1.350	2.100	1.720	1.190	96.1	101.0	176.9	-88	176	2716	2079	887	682	506	418	396	273	147	-888.523628
	aVTZ	1.341	2.067	1.699	1.178	96.6	101.1	177.1	-88	174	2728	2100	901	692	528	431	407	277	149	-888.702213
CCSD(T)	aV(D+d)Z	1.350	2.100	1.720	1.180	96.3	101.5	176.9	-88	176	2669	2159	889	666	489	409	404	279	146	-888.594424
	aV(T+d)Z	1.340	2.070	1.710	1.170	96.7	101.6	176.9	-88	176	2675	2187	899	673	512	422	412	282	148	-888.773544
	CBS	1.336	2.057	1.706	1.166	96.9	101.6	176.9	-88	175										-888.848963
II																				
MP2	aVDZ	1.340	1.710	1.230	1.580	94.2	132.6	173.8	180	-180	2750	2010	1078	945	600	481	130	416	177	-888.528219
	aVTZ	1.338	1.687	1.215	1.572	94.6	134.9	174.5	180	-180	2757	2041	1099	972	604	487	124	434	188	-888.692915
CCSD(T)	aV(D+d)Z	1.350	1.720	1.230	1.590	93.9	129.9	174.1	180	-180	2705	1976	1071	924	597	482	131	416	187	-888.584210
	aV(T+d)Z	1.340	1.690	1.210	1.580	94.5	133.5	174.9	180	-180	2708	2015	1087	950	602	489	126	435	203	-888.762279
	CBS	1.336	1.677	1.202	1.576	94.8	135.0	175.2	180	-180										-888.837256
III																				
MP2	aVDZ	1.360	1.720	1.230	1.590	98.0	131.7	175.2	0	180	2694	2009	1006	974	583	486	114	408	136	-888.512060
	aVTZ	1.344	1.685	1.214	1.572	97.9	138.0	175.4	0	180	2702	2042	1030	1001	586	490	103	425	149	-888.690242
CCSD(T)	aV(D+d)Z	1.360	1.720	1.230	1.590	98.0	131.7	175.2	0	180	2647	1975	1001	953	582	489	118	408	160	-888.581647
	aV(T+d)Z	1.350	1.690	1.210	1.580	98.1	135.8	175.8	0	180	2652	2013	1016	982	586	492	107	426	168	-888.759717
	CBS	1.346	1.677	1.202	1.576	98.1	137.5	176.1	0	180										-888.834694
IV																				
MP2	aVDZ	1.030	1.270	1.740	1.770	111.1	145.3	77.1	0	180	3490	1693	1109	681	610	425	359	770	447	-888.515086
	aVTZ	1.019	1.256	1.738	1.764	111.7	145.5	76.5	0	180	3507	1708	1109	693	605	452	366	785	464	-888.683373
CCSD(T)	aV(D+d)Z	1.030	1.270	1.750	1.780	111.0	145.2	77.2	0	180	3461	1698	1148	677	625	410	354	780	434	-888.572431
	aV(T+d)Z	1.020	1.260	1.740	1.770	111.7	145.5	76.3	0	180	3487	1719	1145	686	620	441	363	798	453	-888.752763
	CBS	1.016	1.256	1.736	1.766	112.0	145.6	75.9	0	180										-888.828692
V																				
MP2	aVDZ	1.090	1.740	1.330	1.730	124.9	108.8	97.7	-180	0	3236	1439	1212	847	722	560	478	777	449	-888.509087
	aVTZ	1.081	1.738	1.313	1.709	125.2	108.5	97.9	180	0	3228	1428	1218	861	753	576	506	780	468	-888.680162
CCSD(T)	aV(D+d)Z	1.100	1.760	1.320	1.750	124.8	109.2	98.3	180	0	3203	1489	1215	831	709	551	459	781	444	-888.568290
	aV(T+d)Z	1.080	1.750	1.300	1.720	125.2	108.6	98.5	180	0	3202	1490	1226	842	738	568	488	794	462	-888.751281
	CBS	1.072	1.746	1.292	1.707	125.4	108.3	98.6	180	0										-888.828329
VI																				

MP2	aVDZ	1.350	2.050	1.690	1.200	97.5	103.2	175.0	88	-177	2706	2003	894	637	513	391	326	224	130	-888.486528
	aVTZ	1.342	2.039	1.673	1.189	97.4	103.0	175.3	87	-176	2716	2030	908	664	535	407	348	244	140	-888.654274
CCSD(T)	aV(D+d)Z	1.360	2.070	1.700	1.200	97.3	103.1	174.6	88	-176	2656	2030	895	623	497	392	316	216	122	-888.551374
	aV(T+d)Z	1.340	2.050	1.680	1.190	97.6	103.4	174.6	87	-178	2660	2058	903	653	519	402	336	238	133	-888.732277
	CBS	1.332	2.042	1.672	1.186	97.7	103.5	174.6	87	-178										-888.808447
VII																				
MP2	aVDZ	1.350	1.710	1.190	1.620	95.6	155.6	176.3	180	-180	2711	2129	986	924	522	380	111	356	148	-888.484614
	aVTZ	1.342	1.684	1.190	1.587	95.7	160.3	176.9	-180	180	2720	2135	1005	948	523	399	108	385	156	-888.664055
CCSD(T)	aV(D+d)Z	1.350	1.710	1.190	1.620	95.6	155.6	176.3	180	-180	2672	2103	988	889	511	375	90	347	181	-888.551644
	aV(T+d)Z	1.340	1.690	1.180	1.610	95.7	162.5	177.5	180	-180	2672	2144	991	915	499	394	89	380	170	-888.731108
	CBS	1.336	1.682	1.176	1.606	95.7	165.4	178.0	180	-180										-888.806672
VIII																				
MP2	aVDZ	1.020	1.230	1.630	2.000	123.9	170.3	109.2	91	-180	3557	1958	799	711	592	495	474	336	122	-888.487130
	aVTZ	1.014	1.209	1.618	1.994	126.6	171.4	107.8	89	-178	3570	1974	798	703	605	528	490	349	124	-888.654950
CCSD(T)	aV(D+d)Z	1.030	1.230	1.640	2.020	120.9	170.9	109.0	90	-179	3487	1903	837	724	596	500	462	305	129	-888.545537
	aV(T+d)Z	1.020	1.210	1.620	2.000	122.9	171.9	108.6	91	-180	3522	1935	806	724	605	521	477	329	129	-888.727268
	CBS	1.016	1.202	1.612	1.992	123.7	172.3	108.4	91	-180										-888.803786
IX																				
MP2	aVDZ	1.750	1.610	1.400	1.030	45.0	73.0	141.6	-80	101	3410	1435	1079	992	782	607	512	391	309	-888.494469
	aVTZ	1.747	1.597	1.386	1.024	45.0	72.1	142.1	-79	102	3436	1449	1077	1005	773	610	528	406	315	-888.658922
CCSD(T)	aV(D+d)Z	1.750	1.610	1.410	1.030	45.1	73.1	141.0	-79	101	3395	1397	1101	997	784	601	495	381	306	-888.550655
	aV(T+d)Z	1.740	1.600	1.390	1.020	45.3	71.9	141.7	-78	101	3431	1417	1097	1007	776	604	520	396	312	-888.727185
	CBS	1.736	1.596	1.382	1.016	45.4	71.4	142.0	-78	102										-888.801514
X																				
MP2	aVDZ	1.350	1.730	1.750	1.270	39.3	79.9	139.2	0.0	180.0	2743	1728	1001	868	598	478	248	387	159	-888.485861
	aVTZ	1.339	1.718	1.748	1.258	39.3	79.1	139.5	0.0	180.0	2759	1724	1009	873	601	485	250	408	179	-888.650348
CCSD(T)	aV(D+d)Z	1.350	1.740	1.760	1.270	38.4	82.3	137.7	0.0	180.0	2706	1679	1006	865	540	417	239	392	189	-888.544399
	aV(T+d)Z	1.340	1.730	1.750	1.250	38.8	80.3	138.6	0.0	180.0	2714	1705	1012	869	547	443	243	412	210	-888.720383
	CBS	1.336	1.726	1.746	1.242	38.9	79.5	139.0	0.0	180.0										-888.794482
XI																				
MP2	aVDZ	1.090	1.720	1.690	1.720	134.0	92.0	87.0	-180	0	3286	1153	946	879	796	752	595	497	263	-888.484051
	aVTZ	1.077	1.706	1.674	1.706	134.3	91.5	87.4	180	0	3277	1139	955	883	812	774	600	505	314	-888.654432
CCSD(T)	aV(D+d)Z	1.090	1.710	1.710	1.710	133.3	93.4	86.6	180	0	3258	1165	944	868	742	680	557	531	372	-888.531459
	aV(T+d)Z	1.080	1.700	1.680	1.700	133.9	92.1	87.2	180	0	3262	1149	968	881	778	706	583	556	405	-888.716417
	CBS	1.076	1.696	1.667	1.696	134.2	91.6	87.5	180	0										-888.794294
XII																				
MP2	aVDZ	1.800	1.020	1.270	1.560	122.2	121.3	176.8	0	180	3524	1892	1235	861	625	389	122	505	368	-888.44554
	aVTZ	1.760	1.015	1.262	1.545	123.5	122.5	177.5	0	180	3529	1903	1240	865	652	362	111	529	384	-888.642551
CCSD(T)	aV(D+d)Z	1.800	1.020	1.270	1.560	121.7	120.3	177.4	0	180	3506	1841	1254	847	595	390	129	514	355	-888.536788
	aV(T+d)Z	1.770	1.010	1.260	1.550	122.2	121.0	177.5	0	180	3518	1853	1254	855	618	371	126	541	387	-888.713509
	CBS	1.757	1.006	1.256	1.546	122.4	121.3	177.5	0	180										-888.787918

XIII																				
MP2	aVDZ	1.920	1.820	1.270	1.100	109.7	93.4	128.2	105	-70	3182	1604	1117	793	730	606	293	232	198	-888.475461
	aVTZ	1.910	1.806	1.253	1.085	109.9	90.0	130.2	104	-70	3185	1608	1077	775	749	641	294	232	182	-888.641544
CCSD(T)	aV(D+d)Z	1.970	1.830	1.260	1.100	106.1	96.8	127.0	110	-66	3146	1630	1041	770	621	586	281	181	145	-888.530916
	aV(T+d)Z	1.940	1.800	1.240	1.090	109.1	87.2	133.0	106	-69	3171	1665	1032	777	656	637	295	220	150	-888.709839
	CBS	1.927	1.787	1.232	1.086	110.4	83.2	135.5	104	-70										-888.785175
XIV																				
MP2	aVDZ	1.102	1.639	1.405	1.616	122.8	124.7	119.0	60	-127	3137	1287	1160	1025	850	809	467	319	123	-888.442299
	aVTZ	1.090	1.623	1.389	1.588	123.2	124.3	120.4	61	-126	3131	1292	1185	1040	860	827	461	328	120	-888.636718
CCSD(T)	aV(D+d)Z	1.100	1.640	1.410	1.600	122.9	124.9	120.2	60	-128	3113	1292	1152	1010	838	806	461	309	120	-888.536981
	aV(T+d)Z	1.090	1.630	1.390	1.580	123.0	124.6	121.6	60	-127	3114	1296	1186	1019	848	822	454	314	116	-888.710911
	CBS	1.086	1.626	1.382	1.572	123.0	124.5	122.2	60	-127										-888.784145
XV																				
MP2	aVDZ	1.100	1.640	1.420	1.640	122.9	119.9	113.7	-180	0	3096	1313	1090	1016	909	404	291	867	44	-888.460714
	aVTZ	1.093	1.622	1.410	1.606	123.2	120.3	114.2	-180	0	3085	1321	1102	1029	939	408	291	881	34	-888.633084
CCSD(T)	aV(D+d)Z	1.110	1.640	1.430	1.620	123.0	120.2	114.3	-180	0	3080	1318	1064	1007	928	402	290	858	61	-888.535890
	aV(T+d)Z	1.090	1.630	1.410	1.600	123.1	120.5	114.7	-180	0	3080	1328	1073	1030	949	405	290	875	57	-888.708802
	CBS	1.082	1.626	1.402	1.592	123.1	120.6	114.9	-180	0										-888.781607
XVI																				
MP2	aVDZ	1.020	1.340	1.760	1.740	123.6	125.8	96.3	-180	0	3571	1409	1258	800	732	548	436	597	379	-888.449681
	aVTZ	1.012	1.332	1.754	1.726	124.3	125.8	96.2	180	0	3576	1402	1256	826	758	564	467	614	405	-888.620001
CCSD(T)	aV(D+d)Z	1.020	1.350	1.780	1.760	123.8	125.7	96.3	180	0	3569	1391	1233	792	707	535	418	563	355	-888.516506
	aV(T+d)Z	1.010	1.340	1.770	1.730	124.3	125.6	96.1	180	0	3582	1390	1239	817	731	548	450	584	383	-888.698239
	CBS	1.006	1.336	1.766	1.717	124.5	125.6	96.0	180	0										-888.774758
XVII																				
MP2	aVDZ	1.030	1.630	1.670	1.590	110.0	113.8	161.5	-78	23	3417	1430	1128	954	588	461	421	194	93	-888.433746
	aVTZ	1.023	1.601	1.624	1.564	111.6	115.8	179.1	-77	1	3473	1482	1117	931	604	472	382	135	106	-888.617364
CCSD(T)	aV(D+d)Z	1.030	1.620	1.650	1.580	110.6	114.6	164.9	-83	22	3416	1386	1131	829	585	453	388	192	107	-888.507450
	aV(T+d)Z	1.020	1.590	1.620	1.560	112.2	116.1	179.9	-50	0	3474	1443	1119	862	601	471	371	141	99	-888.687897
	CBS	1.016	1.577	1.607	1.552	112.9	116.7	186.2	-36	-9										-888.763875
XVIII																				
MP2	aVDZ	1.110	1.890	1.510	1.620	113.4	120.0	111.8	-180	0	3101	1266	1182	1067	650	290	175	717	67	-888.414292
	aVTZ	1.092	1.859	1.482	1.610	114.1	119.2	112.5	180	0	3103	1315	1183	1066	673	307	176	718	66	-888.595508
CCSD(T)	aV(D+d)Z	1.110	1.870	1.520	1.630	113.2	120.8	110.6	-180	0	3081	1186	1138	1041	651	304	182	717	66	-888.505450
	aV(T+d)Z	1.090	1.850	1.500	1.610	113.6	120.4	111.1	-180	0	3090	1192	1171	1046	673	318	183	724	76	-888.682057
	CBS	1.082	1.842	1.492	1.602	113.7	120.2	111.3	-180	0										-888.756418
XIX																				
MP2	aVDZ	1.030	1.700	1.780	1.695	112.7	93.7	89.0	119	-5	3404	1150	905	855	773	641	559	548	360	-888.430231
	aVTZ	1.023	1.686	1.756	1.685	114.4	92.9	89.3	-121	5	3455	1159	924	878	747	672	569	567	391	-888.599100
CCSD(T)	aV(D+d)Z	1.030	1.720	1.800	1.718	111.4	93.6	88.9	117	-5	3399	1159	891	826	758	630	554	537	353	-888.496779
	aV(T+d)Z	1.020	1.700	1.760	1.699	113.9	92.6	89.3	120	-5	3454	1169	900	841	740	663	580	552	377	-888.678247

	CBS	1.020	1.690	1.740	1.691	115.0	92.2	89.5	121	-4										-888.754655
XX																				
MP2	aVDZ	1.100	1.970	1.500	1.620	115.3	119.4	110.7	0	180	3098	1309	1159	1080	470	361	166	648	91	-888.410067
	aVTZ	1.092	1.922	1.475	1.602	114.8	120.0	111.4	0	-180	3096	1348	1174	1085	493	404	173	659	82	-888.591009
CCSD(T)	aV(D+d)Z	1.110	1.930	1.510	1.620	113.7	122.0	110.8	0	180	3068	1183	1150	1062	489	400	173	652	84	-888.500861
	aV(T+d)Z	1.090	1.900	1.500	1.610	113.5	122.2	111.2	0	180	3078	1196	1183	1068	511	425	178	666	80	-888.677425
	CBS	1.080	1.890	1.500	1.610	113.4	122.3	111.4	0	180										-888.751768
XXI																				
MP2	aVDZ	1.350	1.740	1.360	1.915	98.4	117.6	62.5	60	-13	2678	1240	972	678	605	467	302	261	201	-888.407828
	aVTZ	1.346	1.720	1.336	1.881	98.1	119.2	63.9	60	-16	2690	1287	996	685	630	475	301	264	188	-888.572151
CCSD(T)	aV(D+d)Z	1.360	1.750	1.370	1.913	98.3	117.4	63.3	59	-15	2631	1200	976	650	575	449	307	265	202	-888.474746
	aV(T+d)Z	1.350	1.720	1.340	1.872	98.3	120.0	64.8	59	-19	2640	1265	994	667	596	465	298	259	186	-888.650119
	CBS	1.346	1.707	1.327	1.854	98.3	121.1	65.4	60	-20										-888.723960
XXII																				
MP2	aVDZ	1.360	1.730	1.270	1.810	92.9	146.4	80.1	180	0	2708	1646	1000	743	550	395	199	229	88	-888.392433
	aVTZ	1.344	1.699	1.262	1.789	93.2	147.8	78.9	180	0	2715	1661	1020	765	570	416	202	247	123	-888.570581
CCSD(T)	aV(D+d)Z	1.360	1.730	1.280	1.800	93.2	144.0	82.9	180	0	2667	1615	1005	718	505	352	197	227	94	-888.470975
	aV(T+d)Z	1.350	1.710	1.260	1.790	93.5	145.8	81.0	180	0	2670	1635	1020	746	527	381	201	240	113	-888.648635
	CBS	1.346	1.702	1.252	1.786	93.6	146.6	80.2	180	0										-888.723439
XXIII																				
MP2	aVDZ	1.110	1.610	1.810	1.600	105.9	101.1	112.4	180	180	3042	1017	954	894	514	343	223	790	75	-888.386431
	aVTZ	1.100	1.605	1.781	1.591	106.0	101.4	112.5	180	180	3071	1044	987	875	540	368	232	810	78	-888.54826
CCSD(T)	aV(D+d)Z	1.120	1.640	1.830	1.600	105.3	100.1	113.4	180	180	3001	997	954	881	502	338	222	760	78	-888.457240
	aV(T+d)Z	1.100	1.620	1.780	1.580	105.5	101.1	113.7	180	180	3029	1026	972	882	539	366	231	781	84	-888.632049
	CBS	1.092	1.612	1.759	1.572	105.6	101.5	113.8	180	180										-888.705653

Table S2: Harmonic (ω, cm^{-1}) and anharmonic (ν, cm^{-1}) frequencies of HSSCN species as computed using the MP2 approach. Δν (= ν – ω, cm^{-1}) corresponds to the anharmonicity.

Structure	Symmetry	aug-cc-pVDZ			aug-cc-pVTZ		
		ω	ν	Δν	ω	ν	Δν
I	a	2715.6	2612.7	-103	2728.3	2624.4	-103.9
		2079.1	2051.4	-27.7	2100.0	2071.0	-29
		886.8	868.4	-18.4	901.1	882.9	-18.2
		682.1	674.6	-7.5	691.6	684.7	-6.9
		506.3	496.4	-10	528.0	516.8	-11.2
		417.9	415.2	-2.7	430.6	427.8	-2.7
		396.0	386.0	-10	407.4	397.6	-9.8
		273.3	264.6	-8.7	277.0	268.0	-8.9
		146.8	145.9	-0.9	148.9	148.0	-0.9
II	a'	2749.5	2645.4	-104.1	2756.7	2653.3	-103.4
		2009.8	1986.8	-23	2041.4	2000.2	-41.2
		1078.1	1059.2	-18.9	1098.6	1079.6	-19
		945.1	917.6	-27.5	972	949.5	-22.5
		599.7	584.8	-14.9	603.5	587.1	-16.4
		480.8	468.8	-12	487.4	482.8	-4.6
		130	125.8	-4.2	124	119.5	-4.5
	a''	415.5	412.4	-3.1	434.3	431.8	-2.5
		177	160.2	-16.8	188.4	171.3	-17.1
III	a'	2694.1	2588.9	-105.2	2701.5	2595.8	-105.6
		2009.1	1970.1	-39	2042.3	2011.2	-31.1
		1006.1	991.5	-14.6	1030.1	1016.7	-13.4
		974.1	964.0	-10.1	1001.0	984.4	-16.5
		582.9	568.3	-14.6	586.2	569.3	-16.9
		486.3	480.4	-5.9	489.8	484.1	-5.7
		114.3	110.1	-4.2	102.8	97.8	-5
	a''	407.5	406.0	-1.4	425.1	422.6	-2.5

		135.9	116.8	-19.1	149.1	126.7	-22.4
IV	a'	3489.7	3317	-172.7	3507	3341.5	-165.5
		1693	1677.6	-15.4	1708.2	1688.2	-20
		1108.5	1064.5	-44	1109.1	1063.5	-45.6
		680.5	665.7	-14.8	692.6	677.9	-14.7
		609.5	591.7	-17.8	604.7	585.5	-19.2
		424.6	420.2	-4.4	451.9	447.3	-4.6
		358.7	355.4	-3.3	366.4	362.4	-4
	a''	769.8	748.3	-21.5	785.2	762.3	-22.9
		446.8	444.1	-2.7	464	459.8	-4.2
V	a'	3236.3	3099.1	-137.2	3228	3097.2	-130.8
		1439.0	1402.0	-37	1427.6	1384.1	-43.5
		1212.0	1183.1	-28.9	1218.2	1194	-24.2
		846.8	830.7	-16.1	860.9	846.3	-14.6
		721.8	710.8	-11	752.6	741.2	-11.4
		559.9	549.3	-10.6	576.3	565.7	-10.6
		478.0	472.2	-5.8	505.7	500.3	-5.4
	a''	776.6	753.6	-23	780.2	761.6	-18.6
		448.5	442.0	-6.5	468.4	462.0	-6.4
VI	a	2705.6	2600.7	-104.9	2716.1	2610.0	-106.2
		2003.3	1968.8	-34.5	2029.6	1995.4	-34.1
		894.0	875.6	-18.4	908.2	889.7	-18.4
		636.6	620.7	-16	663.5	656.1	-7.4
		513.3	503.2	-10.1	535.0	525.1	-9.9
		391.3	377.9	-13.4	406.6	392.0	-14.6
		326.0	318.3	-7.7	347.5	344.6	-2.9
		223.5	224.0	0.6	244.4	242.4	-2
		129.7	131.0	1.3	139.5	140.1	0.6
VII	a'	2711.2	2596.5	-114.7	2720.0	2604.3	-115.7
		2128.7	2112.5	-16.2	2134.8	2126.1	-8.8

		986.5	951.4	-35	1005.2	971.0	-34.2
		924.0	910.0	-14	948.3	934.3	-14
		521.8	492.2	-29.6	523.1	486.7	-36.4
		379.8	371.7	-8	398.9	397.1	-1.8
		110.6	94.7	-15.9	108.4	98.3	-10.1
	a"	355.7	354.8	-0.9	384.6	383.8	-0.8
		148.4	119.4	-29.1	155.9	129.2	-26.7
VIII	a	3556.5	3382.1	-174.5	3569.8	3404.8	-165
		1958.3	1918.5	-39.9	1974.1	1935.2	-38.9
		799.4	768.3	-31.1	798.4	774.9	-23.5
		711.4	666.3	-45.1	702.6	650.7	-51.9
		592.1	572.0	-20	604.5	585.0	-19.5
		494.8	481.9	-12.9	527.8	511.6	-16.2
		473.8	463.3	-10.5	489.5	479.7	-9.8
		336.0	329.9	-6.2	349.3	344.4	-4.9
		122.3	118.9	-3.3	123.7	121.0	-2.7
IX	a	3410.1	3229.9	-180.2	3435.7	3264.3	-171.4
		1434.5	1421.4	-13.1	1449.3	1434.4	-14.9
		1079.4	1036.4	-43	1076.6	1032.8	-43.8
		991.8	957.5	-34.3	1004.8	971.9	-32.9
		782.2	750.9	-31.3	773.3	745.2	-28.1
		606.5	591.9	-14.6	609.6	594.7	-14.9
		512.3	501.4	-10.9	527.9	515.6	-12.3
		390.8	389.7	-1.1	405.8	404.0	-1.8
		309.1	305.5	-3.6	315.3	311.9	-3.4
X	a'	2742.9	2637.5	-105.4	2758.7	2652.9	-105.8
		1727.8	1719.1	-8.7	1724.2	1710.7	-13.5
		1000.6	976.3	-24.3	1009.4	986.4	-23
		867.8	852.3	-15.5	873.3	859.6	-13.7

		597.7	596.7	-1	601.1	601.3	0.2
		478.2	471.3	-6.9	485.4	479.5	-5.9
		248.1	245.7	-2.4	249.7	247.6	-2.1
	a"	386.6	377.3	-9.2	408.0	396.2	-11.8
		159.1	159.6	0.5	179.3	176.5	-2.8
XI	a'	a)	a)		3277.4	3140.4	-137
					1138.6	1113.9	-24.7
					955.1	943.6	-11.5
					883.1	870.3	-12.8
					812.3	803.4	-8.9
					774.1	760.3	-13.8
					599.5	591.2	-8.3
	a"				505.1	500.3	-4.8
					314.0	332.0	18
XII	a'	3523.7	3344.8	-178.9	3528.9	3357.7	-171.2
		1891.6	1864.4	-27.2	1903.1	1878.1	-24.9
		1235.4	1187.7	-47.7	1240.1	1197.7	-42.3
		861.3	856.3	-5	864.5	855.3	-9.2
		625.2	606.4	-18.8	651.8	639.0	-12.7
		389.3	363.4	-25.9	361.9	292.6	-69.3
		121.5	113.5	-7.9	111.0	87.6	-23.4
	a"	505.2	487.1	-18.1	529.1	497.2	-31.9
		367.5	359.1	-8.5	384.4	380.9	-3.5
XIII	a	3181.5	3018.2	-163.3	3185.2	3047.4	-137.8
		1603.5	1569.0	-34.5	1608.2	1585.4	-22.8
		1116.8	1075.6	-41.2	1076.8	1051.5	-25.3
		792.6	780.0	-12.6	774.5	761.6	-12.9
		729.7	702.9	-26.8	748.5	733.7	-14.8
		605.9	559.4	-46.5	640.8	611.7	-29.1

		293.1	283.3	-9.8	293.6	289.8	-3.8
		231.8	229.7	-2.1	232.2	234.0	1.8
		192.2	189.9	-2.3	181.6	185.2	3.6
XIV	a	3136.9	2989.8	-147.1	3131.3	2990.8	-140.5
		1286.9	1257.5	-29.4	1291.9	1254.4	-37.5
		1159.6	1122.6	-37	1184.7	1145.7	-39
		1025.4	1012.0	-13.4	1040.2	1027.5	-12.7
		850.4	837.0	-13.4	859.5	847.0	-12.5
		808.5	795.6	-12.9	826.9	812.2	-14.7
		467.3	456.6	-10.7	461.0	450.7	-10.3
		319.1	306.8	-12.3	327.6	314.0	-13.6
		122.6	115.7	-6.9	119.9	113.6	-6.3
XV	a'	3096.0	2949.2	-146.8	3085.3	2947.3	-138
		1312.9	1280.9	-32	1321.3	1289.6	-31.7
		1090.2	1066.5	-23.7	1101.7	1078.1	-23.6
		1015.6	986.0	-29.6	1028.9	1001.6	-27.3
		909.4	901.2	-8.2	938.5	924.5	-14
		403.5	397.6	-5.9	407.7	401.0	-6.7
		290.8	291.0	0.2	291.4	292.6	1.2
	a"	867.3	844.5	-22.8	881.2	858.0	-23.2
		44.2	39.3	-4.9	34.3	27.9	-6.4
XVI	a'	3571.1	3401.1	-170	3576.2	3409.9	-166.3
		1409.3	1370.9	-38.4	1401.5	1372.5	-29
		1257.6	1223.2	-34.4	1256.1	1226.4	-29.7
		800.0	784.3	-15.7	825.7	815.2	-10.5
		732.1	715.9	-16.2	758.3	743.3	-15
		547.8	533.9	-13.9	563.5	550.9	-12.6
		435.7	427.5	-8.2	466.9	459	-7.9
	a"	597.2	571.4	-25.8	614.3	594.2	-20.1

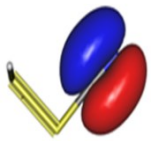
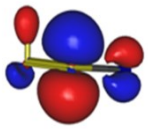
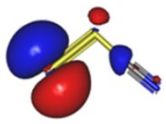
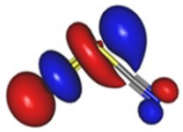
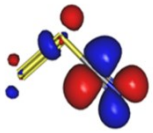
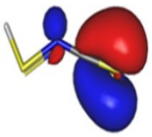
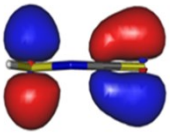
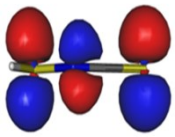
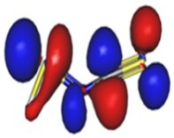
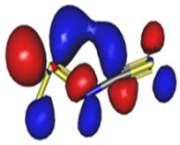
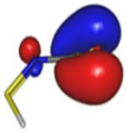
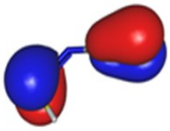
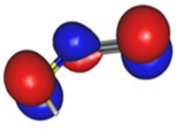
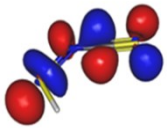
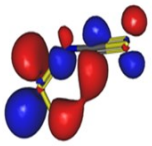
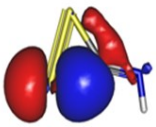
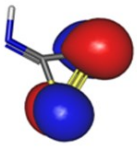
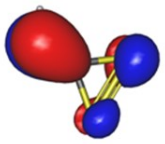
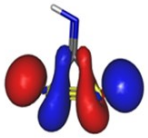
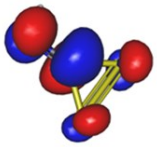
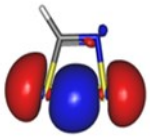
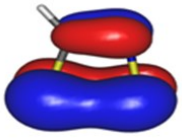
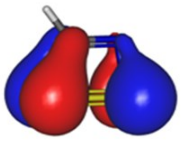
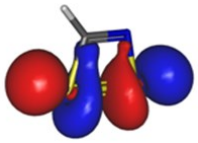
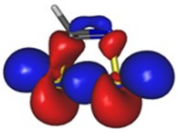
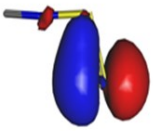
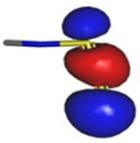
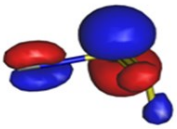
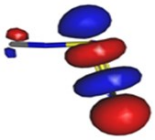
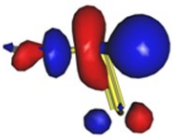
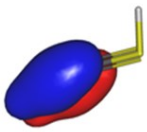
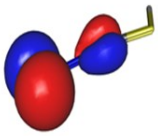
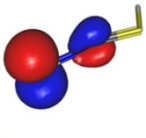
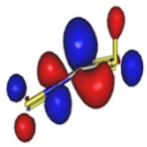
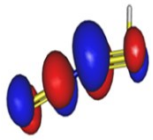
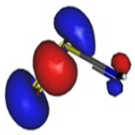
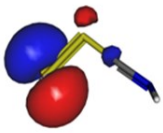
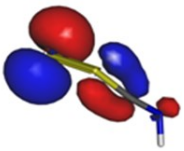
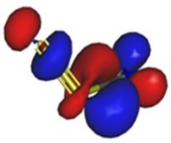
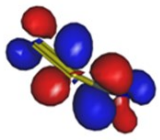
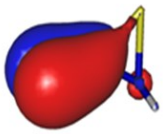
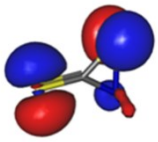
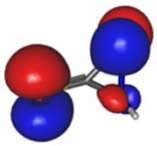
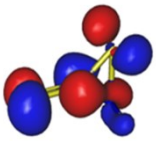
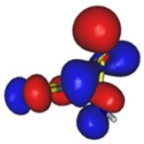
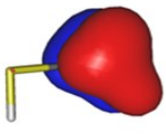
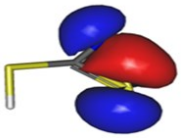
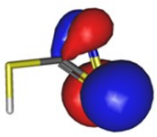
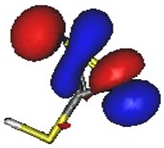
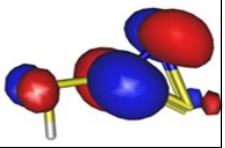
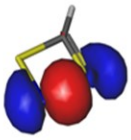
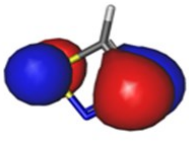
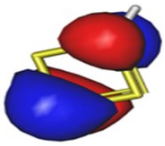
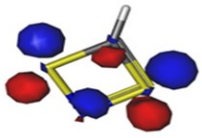
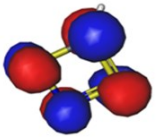
		378.6	373.1	-5.5	405.1	400.5	-4.6
XVII	a	3417.0	3252.6	-164.3	3473.4	3312.9	-160.4
		1430.4	1416.1	-14.3	1482.1	1452.7	-29.5
		1128.0	1106.0	-22	1116.5	1095.6	-20.9
		954.4	937.1	-17.4	931.2	924.3	-6.9
		587.7	577.2	-10.5	604.0	607.4	3.4
		461.4	440.9	-20.5	471.6	463.9	-7.7
		420.7	418.0	-2.6	381.5	372.1	-9.4
		194.3	177.8	-16.5	134.7	122.8	-12
		93.3	89.2	-4.2	106.2	167.3	61.1
XVIII	a'	3101.4	2952.4	-149	3102.5	2962.5	-140
		1266.1	1256	-10.1	1315.4	1303.6	-11.8
		1181.8	1155.1	-26.7	1182.5	1159.6	-22.9
		1066.9	1052.4	-14.5	1065.7	1054.4	-11.3
		650.2	628.7	-21.5	673.0	653.5	-19.5
		290.1	280.3	-9.8	307.4	299.6	-7.8
		175.3	172.3	-3	175.9	174.8	-1.1
	a''	716.5	707.3	-9.2	718.2	696.7	-21.5
		66.7	64.2	-2.5	65.8	64.5	-1.3
XIX	a	3404.1	3223.5	-180.6	3454.6	3283.2	-171.4
		1149.5	1106.8	-42.7	1158.7	1117.4	-41.3
		905.1	877.6	-27.5	923.5	899.5	-24
		855.0	833.0	-22	878.0	856.8	-21.2
		773.3	747.9	-25.4	746.7	716.3	-30.4
		640.6	622.1	-18.5	671.7	653.7	-18
		558.7	543.9	-14.8	569.1	555.1	-14
		547.8	534.6	-13.2	567.0	548.3	-18.7
		359.7	354.3	-5.4	390.8	385.4	-5.4
XX	a'	3097.9	2949.5	-148.4	3095.9	2955.5	-140.4

		1309.2	1305.2	-4	1348.3	1337.2	-11.1
		1158.8	1130.2	-28.6	1173.8	1147.6	-26.2
		1079.7	1064	-15.7	1084.7	1072.4	-12.3
		470.1	454.1	-16	492.6	475.9	-16.7
		360.8	344.2	-16.6	404.2	391.3	-12.9
		165.8	149.5	-16.3	173	162.2	-10.8
	a"	647.5	642.4	-5.1	659.2	658.5	-0.7
		90.7	86.3	-4.4	82.3	86.4	4.1
XXI	a	2677.8	2570.1	-107.7	2689.9	2581.1	-108.8
		1240.2	1180.2	-60	1286.8	1233.5	-53.3
		972.O	951.4	-20.6	996.0	974.8	-21.2
		677.9	664.2	-13.7	684.5	669.4	-15.1
		604.5	592.3	-12.2	629.7	612.0	-17.7
		467.3	453.7	-13.6	474.7	477.7	3
		302.1	287.8	-14.3	301.0	285.7	-15.3
		261.1	249.8	-11.3	264.2	247.5	-16.7
		201.3	197.0	-4.3	188.4	177.9	-10.5
XXII	a'	2708.3	2607.6	-100.7	2715.3	2614.0	-101.3
		1645.8	1618.6	-27.2	1660.8	1633.8	-27
		1000.1	974.6	-25.5	1019.6	994.6	-25
		743.4	717.8	-25.6	765.3	741.1	-24.2
		549.6	542.5	-7.1	569.7	562.3	-7.4
		395.0	388.6	-6.4	415.7	410.5	-5.2
		199.4	203.6	18.7	201.7	208.1	6.4
	a"	228.7	247.4	4.2	247.3	225.5	-21.8
		88.2	60.8	-27.4	123.0	76.5	-46.5
XXIII	a'	3042.1	2896.9	-145.2	3071.0	2934.4	-136.5
		1017.4	996.7	-20.7	1044.0	1025.3	-18.8
		954.1	943.8	-10.3	986.6	974.5	-12.1

		893.5	853.8	-39.6	874.8	826.0	-48.7
		514.3	475.6	-38.6	540.0	505.5	-34.5
		342.5	321.8	-20.8	367.7	351.9	-15.7
		223.3	218.8	-4.6	231.9	228.3	-3.6
	a"	789.9	773.6	-16.2	810.4	795.3	-15.1
		74.8	70.8	-4	77.6	73.5	-4

a) Convergence problems.

Table S3: Outermost molecular orbitals (HOMO-2, HOMO-1, HOMO, LUMO, LUMO+1) of the [H, C, N, S, S] isomers as computed at the CASSCF/aug-cc-pV(T+d)Z.

	HOMO-2	HOMO-1	HOMO	LUMO	LUMO+1
I					
II					
III					
IV					
V					
VI					
VII					
VIII					
IX					
X					
XI					

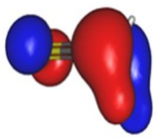
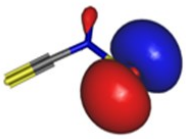
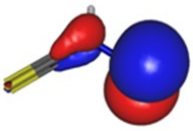
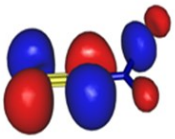
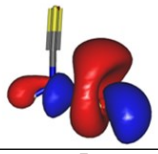
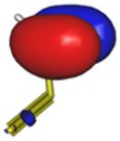
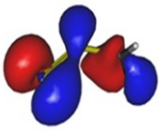
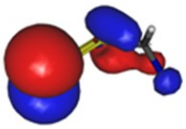
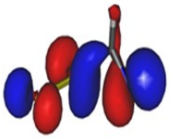
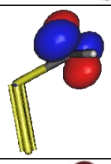
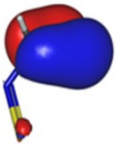
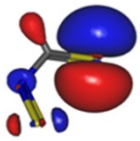
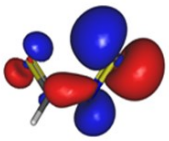
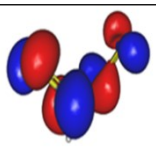
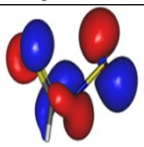
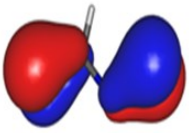
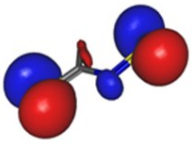
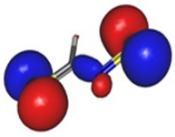
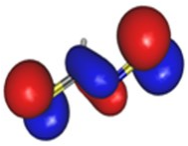
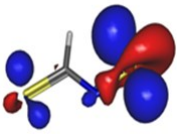
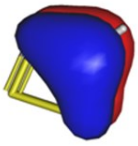
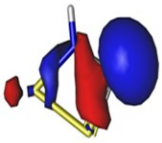
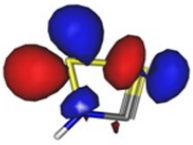
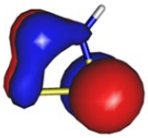
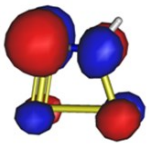
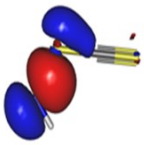
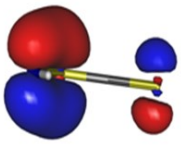
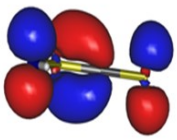
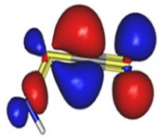
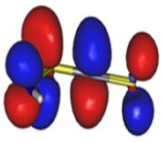
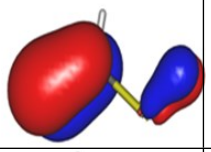
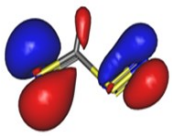
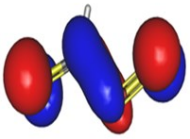
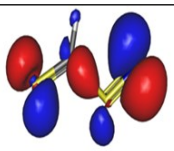
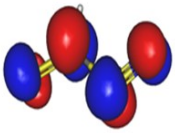
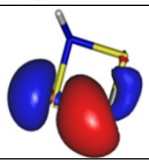
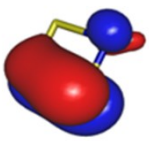
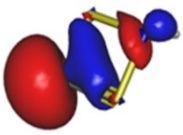
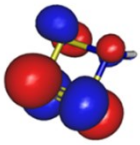
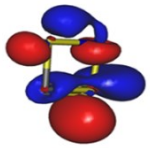
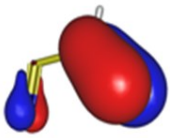
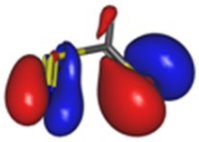
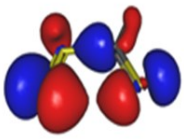
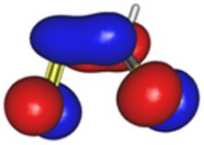
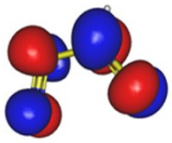
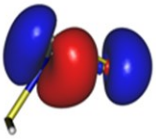
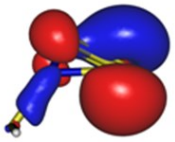
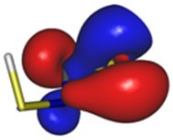
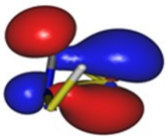
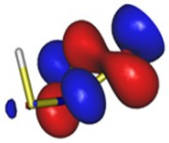
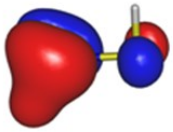
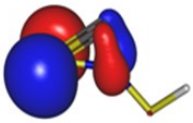
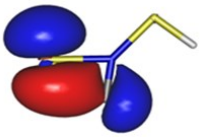
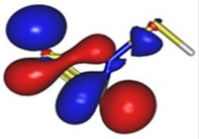
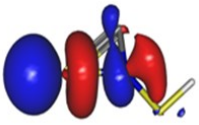
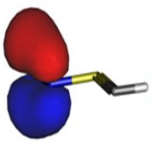
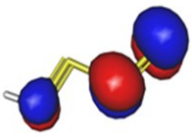
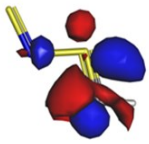
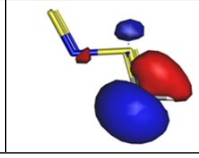
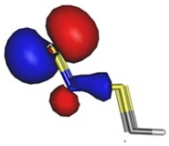
XII					
XIII					
XIV					
XV					
XVI					
XVII					
XVIII					
XIX					
XX					
XXI					
XXII					
XXIII					

Table S4: Dipole moment components (μ_i, debye) of all twenty three stable $HCNS_2$ structures, calculated with the MP2/aug-cc-pVTZ method. For the designation of S1 and S2 atoms, see Figure 1.

Struc.	THE CARTESIAN COORDINATES				Mulliken charges		μ_x	μ_y	μ_z
I	C	-1.39701300	-0.10034700	-0.00258100	C	0.155553	3.3488	1.1002	0.8897
	S1	-0.02662100	0.90350800	0.01116100	S1	0.110239			
	N	-2.38051700	-0.74845800	-0.00346700	N	-0.309359			
	S2	1.49202600	-0.49486700	-0.08590600	S2	-0.155017			
	H	1.59921300	-0.69698200	1.23568500	H	0.198584			
II	C	0.00000000	0.66424300	0.00000000	C	-0.125028	-0.8587	-1.9613	0.0000
	S1	-0.34802800	-1.99880500	0.00000000	S1	0.045468			
	N	-0.67684300	-0.34464900	0.00000000	N	0.002598			
	S2	0.74632000	2.04823900	0.00000000	S2	-0.118336			
	H	-1.63476700	-2.36386900	0.00000000	H	0.195298			
III	H	1.67884400	1.40097800	-0.00062400	H	0.180191	1.3498	1.3216	-0.0002
	S1	-2.16153500	0.13677900	0.00002900	S1	-0.124537			
	C	-0.62956000	-0.21730200	0.00000300	C	-0.142732			
	S2	2.06191100	0.11255300	0.00008100	S2	0.100969			
	N	0.52750200	-0.58378200	-0.00016400	N	-0.013891			
IV	C	-0.72851400	-0.03518000	-0.00000700	C	0.315567	0.5770	1.3437	0.0000
	S1	0.62109200	1.10118500	0.00000000	S1	-0.045464			
	S2	0.67053300	-1.06615500	0.00000100	S2	0.001433			
	N	-1.97849800	-0.15692800	0.00000000	N	-0.519068			
	H	-2.44544600	0.74909600	0.00001900	H	0.247531			
V	H	1.23117100	-1.98187300	0.00000000	H	0.548287	-0.9461	-1.5926	0.0000
	C	0.75090100	-1.01309100	0.00000000	C	-0.160042			
	S1	-0.96064900	-0.71296800	0.00000000	S1	-0.046944			
	N	1.37625800	0.14177000	0.00000000	N	-0.469148			
	S2	0.00000000	1.15472000	0.00000000	S2	0.127847			
VI	H	-1.61531700	-0.67537900	1.23106500	H	0.186620	-2.5308	0.7142	0.8893
	S1	0.07063500	0.85547000	0.01309300	S1	0.292465			
	S2	-1.49031000	-0.45283100	-0.08682700	S2	-0.106245			
	N	1.40737800	-0.15061600	-0.00386300	N	0.057797			
	C	2.41307900	-0.78542100	-0.00404600	C	-0.430637			
VII	H	-2.58576800	1.13420600	-0.00024300	H	0.203177	-5.5496	0.9026	-0.0001
	C	-0.52363400	0.22221300	0.00012600	C	0.066443			
	S1	2.23908300	-0.04436500	-0.00003600	S1	-0.153398			
	S2	-2.16843000	-0.14102300	-0.00002300	S2	-0.071596			
	N	0.65673300	0.07124700	0.00006100	N	-0.044627			
VIII	H	2.94334600	-0.93101700	0.60291100	H	0.310944	5.3349	0.7395	1.3175
	N	2.33646900	-0.56860800	-0.12457100	N	-0.198639			
	S1	-1.57983000	-0.52670000	-0.00028100	S1	-0.452984			
	C	1.29546100	0.02630500	0.03391000	C	0.117751			
	S2	-0.11213200	0.82379100	0.00438300	S2	0.222928			
IX	C	0.17529200	0.15183600	-0.02973700	C	0.288534	-1.5339	0.5762	1.2978
	H	-0.82402100	1.68033700	0.76482600	H	0.209680			
	S1	1.73938800	-0.16659800	0.00959000	S1	-0.248987			
	S2	-1.43612800	-0.52039500	0.01251900	S2	0.064451			
	N	-0.72570000	1.20007700	-0.13430900	N	-0.313679			
X	S1	-1.52994700	-0.46873900	-0.00002300	S1	-0.057969	2.6714	-2.0407	0.0003
	H	1.49924000	-1.43283600	0.00045500	H	0.203525			
	S2	1.72174000	-0.11296100	-0.00004000	S2	-0.142390			
	C	0.04980200	0.28022100	0.00005000	C	0.406805			
	N	-0.69524900	1.29410300	0.00003600	N	-0.409972			
XI	H	0.00001300	-2.23716800	-0.00007100	H	0.503911	0.0000	-3.1083	-0.0005
	S1	-1.22152600	0.03053500	0.00012300	S1	0.431294			
	S2	1.22153300	0.03055800	-0.00018600	S2	0.431260			
	C	0.00001900	-1.16028200	-0.00004500	C	-0.631583			
	N	-0.00003400	1.17448300	0.00019200	N	-0.734882			
XII	H	-1.97600000	0.37697800	0.00000000	H	0.249205	-0.1321	5.3227	0.0000
	N	-1.04453400	-0.02611200	0.00000000	N	0.109970			
	S1	-0.73572400	-1.75843100	0.00000000	S1	-0.444547			
	C	0.00000000	0.68134900	0.00000000	C	0.043081			
	S2	1.31620700	1.49078800	0.00000000	S2	0.042292			
XIII	S1	-1.53454000	-0.34614900	-0.05296000	S1	-0.308920	2.0257	0.3044	2.6273
	S2	0.06714000	0.69076700	0.04282600	S2	0.355158			
	H	1.66794000	-0.83345400	1.48575300	H	0.564635			

	C	1.45125900	-0.36072300	0.53372700	C	-0.240213			
	N	1.87184300	-0.35944300	-0.64656800	N	-0.370659			
XIV	C	0.61506000	0.79712800	0.18520300	C	-0.201487	-0.8837	0.7988	0.9763
	H	0.85733200	1.74791000	0.65916700	H	0.469335			
	S1	-1.69185800	-0.36326000	0.07013900	S1	0.225455			
	S2	1.70201800	-0.38835800	-0.03411600	S2	-0.144289			
	N	-0.67289300	0.78503000	-0.33525100	N	-0.349014			
XV	N	-0.53228400	-0.56104300	0.00016000	N	-0.331994	-1.3269	1.6371	0.0002
	C	0.49489400	0.40534400	0.00018100	C	-0.172927			
	H	0.17981000	1.45221400	0.00030000	H	0.514962			
	S1	2.05118000	-0.05331100	-0.00008100	S1	-0.148302			
	S2	-2.01512900	0.05600000	-0.00007500	S2	0.138262			
XVI	H	-1.36187500	1.79031500	0.00000700	H	0.191552	-2.3039	-0.4649	-0.0002
	N	-0.65308300	1.06738300	0.00002700	N	0.093735			
	S1	-0.99115700	-0.62503000	-0.00018900	S1	0.013947			
	S2	1.11312900	-0.42887700	0.00013700	S2	0.191611			
	C	0.66365300	1.26675100	0.00010500	C	-0.490845			
XVII	H	-1.59356300	1.62968100	-0.00887400	H	0.179251	3.0312	-0.4787	-0.0300
	N	-2.11403900	0.74917300	0.00194600	N	-0.783973			
	S1	1.98512000	0.15238500	-0.00177400	S1	-0.077404			
	S2	-1.13183000	-0.51525900	-0.00237000	S2	0.595315			
	C	0.45653300	-0.17798600	0.01025900	C	0.086812			
XVIII	C	0.46138100	0.62572900	-0.00006200	C	-0.411052	1.3007	0.1985	-0.0003
	H	0.27093300	1.70147700	-0.00016000	H	0.420113			
	S1	1.89959300	-0.09721900	0.00003400	S1	-0.081738			
	S2	-1.07689000	-0.41740300	-0.00004300	S2	0.710134			
	N	-2.31463800	0.39687200	0.00009700	N	-0.637457			
XIX	C	0.00002400	-1.24291500	0.00583500	C	-0.595425	0.0001	2.4728	1.2501
	H	0.00007100	1.85205700	0.63020100	H	0.119680			
	N	-0.00002900	1.17209000	-0.13423700	N	-0.457173			
	S1	-1.22120700	-0.08123200	0.00857800	S1	0.466454			
	S2	1.22120600	-0.08121800	0.00857400	S2	0.466464			
XX	C	-0.56311100	0.83676800	0.00004900	C	-0.425775	-0.1019	3.2379	0.0003
	H	-0.76035000	1.91098300	0.00009900	H	0.437832			
	S1	-1.68495300	-0.30711000	-0.00003600	S1	-0.083391			
	S2	1.29786200	0.35802600	0.00007100	S2	0.644585			
	N	1.47606900	-1.10660500	-0.00013600	N	-0.573251			
XXI	C	0.63711100	1.13334900	0.22742000	C	-0.330593	-1.0904	-0.8755	0.8238
	H	-1.46474600	0.08796700	1.32478200	H	0.214417			
	S1	-1.63425600	-0.20134700	0.02131300	S1	0.098018			
	N	-0.10284100	0.33660400	-0.54797300	N	-0.121572			
	S2	1.53187800	-0.37642100	0.05034400	S2	0.139730			
XXII	N	0.00000000	0.34193700	0.00000000	N	0.045340	3.1259	0.6549	0.0000
	S1	1.66954000	0.02632300	0.00000000	S1	0.131798			
	S2	-1.44720000	-0.70939400	0.00000000	S2	-0.050910			
	C	-0.92497400	1.20076200	0.00000000	C	-0.338337			
	H	1.99239900	1.33101900	0.00000000	H	0.212109			
XXIII	H	2.64557900	-1.64625800	0.00000000	H	0.403429	-1.1600	-2.2050	0.0000
	S1	-1.58952500	0.87345400	0.00000000	S1	0.175581			
	C	2.19085800	-0.64430900	0.00000000	C	-0.732332			
	S2	0.60260500	-0.87794100	0.00000000	S2	0.589586			
	N	0.00000000	0.79770000	0.00000000	N	-0.436264			

Table S5: Tentative assignment of the anharmonic frequencies (ν_i) of HSSCN isomers. For the planar structures, ν_{1-7} are of a' symmetry and ν_{8-9} are of a" symmetry. For non-planar species, all frequencies are of a symmetry.

Struc.	ν_1	ν_2	ν_3	ν_4	ν_5	ν_6	ν_7	ν_8	ν_9
I	SH stretching	CN stretching	In-plane HSS bending	SC stretching	SS stretching	NCS bending	Out-of-plane HSSC torsion	Out-of-plane SSCN torsion	SSC bending
II	SH stretching	CN stretching	CS stretching + HSN bending	CS stretching + HSN bending	SN stretching	NCS bending	CNS bending	Out-of-plane HSNC torsion	Out-of-plane SNCS torsion
III	SH stretching	CN stretching	CS stretching + HSN bending	HSN bending	SN stretching + SNC bending	NCS bending	CNS bending	Out-of-plane HSNC torsion	Out-of-plane SNCS torsion
IV	NH stretching	CN stretching	HNC bending	Symmetric CS2 stretching	Anti Symmetric CS2 stretching	Symmetric in-plane CS2 bending	Antisymmetric CS2 bending	Out-of-plane HNCS torsion	Out-of-plane NCSS torsion
V	CH stretching	CN stretching	HCN bending	SC stretching	SN stretching	NSS bending + CS2 bending	SS stretching	Out-of-plane HCNS torsion	Out-of-plane SCNS torsion
VI	SH stretching	CN stretching	HSS bending	SN stretching	SS stretching	Out-of-plane HSSN torsion	CNS bending	Out-of-plane SSNC torsion	CNS bending
VII	SH stretching	CN stretching	CS stretching+ HSC bending	CS stretching + HSC bending	SN stretching	CNS bending	NCS bending	Out-of-plane HSCN torsion	Out-of-plane SCNS torsion
VIII	NH stretching	CN stretching	CS stretching + HNC bending	HNC bending	Out-of-plane HNCS torsion	SS stretching	NCS bending	Out-of-plane NCSS torsion	CSS bending
IX	NH stretching	CS stretching	HNC bending	Out-of-plane HNCS torsion	CN stretching	CSN bending	NCS bending	Out-of-plane HNCS	SCS bending

								torsion	
X	SH stretching	CN stretching	CS stretching + HSC bending	CS stretching + HSC bending	NCS bending	CS2 bending + NCS bending	SCS bending	Out-of-plane HSCN torsion	Out-of-plane SCSN torsion
XI	CH stretching	HCS bending	Ring symmetric stretching	HCS bending + ring antisymmetric stretchinh	Ring symmetric stretching	Ring antisymmetric stretching	Ring scissoring	Out-of-plane HCSN torsion	Ring wagging
XII	NH stretching	CN stretching	HNS bending	CS stretching	SN stretching	NCS bending	CNS bending	Out-of-plane HNSC torsion	Out-of-plane SCNS torsion
XIII	NH stretching	CN stretching	SS stretching + HNS bending	Out-of-plane HCSS torsion	Out-of-plane NCSS torsion	CS stretching	NCS bending	CSS bending	NCS bending
XIV	CH stretching	HCS bending	CN stretching	SN stretching + CS stretching	Out-of-plane HCNS torsion	Out-of-plane SCNS torsion	NCS bending	CNS bending	SNC bending + NCS bending
XV	CH stretching	HCS bending	CS stretching	CN stretching	NCS bending + CNS bending	NCS bending + CNS bending	NCS bending + CNS bending	Out-of-plane HCNS torsion	Out-of-plane SNCS torsion
XVI	NH stretching	CN stretching	HNC bending	SN stretching	CS stretching	CSS bending	SS stretching	Out-of-plane HNCS torsion	Out-of-plane NCSS torsion
XVII	NH stretching	Anti symmetric CS2 stretching	HNS bending	SN stretching	Symmetric CS2 stretching	Symmetric CS2 bending	Out-of-plane HNSC torsion	Anti Symmetric CS2 bending	Out-of-plane SCSN torsion
XVIII	CH stretching	SN stretching	CS stretching + HCS bending	HCS bending	CS stretching	NSC bending	NSC bending + CS2 bending	Out-of-plane HCSN torsion	Out-of-plane SCSN torsion

XIX	NH stretching	HNS bending	Ring symmetric stretching	Antisymmetric CS2 stretching	Out-of-plane HNSC torsion	Ring antisymmetric stretching	HNS bendig + NS2 symmetric stretching	Ring scissoring	Ring wagging
XX	CH stretching	SN stretching	CS stretching + HCS bending	CS stretching + HCS bending	CS2 bending + CSN bending	CS stretching	CS2 bending + CSN bending	Out-of-plane HCSN torsion	Out-of-plane SCSN torsion
XXI	SH stretching	CN stretching	HSN bending	CS stretching	NSC bending	SN stretching	Out-of-plane HSNC torsion	Out-of-plane SNCS torsion	CNS bending
XXII	SH stretching	CN stretching	HSN bending	CS stretching	NSC bending	SN stretching	Out-of-plane HSNC torsion	Out-of-plane SNCS torsion	CNS bending
XXIII	CH bending	CS stretching	SN stretching	HCS bending	HCS bending + SNS bending	CSN bending	SNS bending+ NSC bendind	Out-of-plane HCSN torsion	Out-of-plane CSNS torsion

Table S6: (R)CCSD(T)/aug-cc-pV(D+d)Z fragmentation energies (E_0, eV) of the twenty three structures of $HCNS_2$ sum formula for the formation of atomic + tetratomic species in addition to those given in Table 4. We also included the ZPE correction All molecular species are taken in their equilibrium positions as optimized at the (R)CCSD(T)/aug-cc-pV(D+d)Z level.

Reactant	Products	E_0
I	H(^{2}S) + SSCN (^{2}A1)	3,11
II	S(^{3}P)+HSNC(^{1}A′)	3.08
III	S(^{3}P)+HSNC(^{1}A′)	3.02
IV	S(^{3}P)+HNCS(^{1}A′)	1.15
V	H(^{2}S)+ CNSS(^{2}A")	3.28
	H(^{2}S)+lin-SNCS(^{2}A$_1$)	2.72
	N(^{4}S)+trans-HCSS(^{2}A')	5.69
	S(^{3}P)+lin-HCNS(^{1}A′)	2.60
VI	C(^{3}P)+HSSN(^{1}A$_1$)	6.37
VII	S(^{3}P)+HSCN(^{1}A′)	0.91
VIII	S(^{3}P)+HNCS(^{1}A′)	1.19
IX	S(^{3}P)+HNCS(^{1}A′)	0.57
	S(^{3}P)+trans-HNSC(^{1}A′)	5.62
X	H(^{2}S)+trans-NSCS(^{4}A$_1$)	5.04
	S(^{3}P)+HSCN(^{1}A′)	0.72
	N(^{4}S)+HSCS(^{2}A')	3.51
XI	S(^{3}P)+HCSN(^{3}A$_1$)	5.38
XII	S(^{3}P)+cyc-H-CNS(^{1}A′)	3.33
	S(^{3}P)+HNCS (^{1}A′)	0.19
XIII	N(^{4}S)+trans-HCSS(^{2}A')	4.76
	S(^{3}P)+H-NCS(^{1}A')	2.04
XIV	S(^{3}P)+lin-HCNS(^{1}A')	1.78
	S(^{3}P)+H-NCS(^{1}A')	2.16
XV	S(^{3}P)+lin-HCNS(^{1}A')	1.76
	S(^{3}P)+H-NCS(^{1}A')	2.14
XVI	H(^{2}S)+lin-SNCS(^{2}A$_1$)	1.32
	H(^{2}S)+CNSS(^{2}A")	1.88
	C(^{3}P) + cis-HNSS (^{1}A′)	4.64
	S(^{3}P)+HNCS(^{1}A′)	-0.37
XVII	S(^{3}P)+trans-HNSC(^{1}A′)	4.51
XVIII	N(^{4}S)+H-SCS(^{2}A′)	2.12
	S(^{3}P)+HCSN(^{3}A$_1$)	4.72
XIX	S(^{3}P)+trans-HNSC(^{1}A′)	4.18
XX	H(^{2}S)+trans-NSCS(^{4}A$_1$)	3.84
	N(^{4}S)+ H-SCS(^{2}A′)	2.00
	S(^{3}P)+HCSN(^{3}A$_1$)	4.60
XXI	S(^{3}P)+HSNC(^{1}A′)	0.18
XXII	S(^{3}P)+HSNC(^{1}A′)	0.07
XXIII	S(^{3}P)+HCSN(^{3}A$_1$)	3.45