

Structure, stability, and generation of CH₃CNS

Supporting Material

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Table S1: Calculated^a Equilibrium Structure of CH₃CNS

level/ basis set	B3LYP/ cc-pVTZ	B3LYP/ aug-cc-pVTZ ^b	CCSD(T)(fc)/ cc-pVTZ	CCSD(T)(ae)/ cc-pCVTZ	CCSD(T)(fc)/ aug-cc-pVTZ ^b
$r_e(\text{H-C})$	1.092	1.091	1.091	1.089	1.091
$r_e(\text{C-C})$	1.449	1.449	1.462	1.459	1.462
$r_e(\text{C}\equiv\text{N})$	1.159	1.159	1.166	1.164	1.166
$r_e(\text{N}\rightarrow\text{S})$	1.618	1.612	1.630	1.620	1.627
$\alpha_e(\text{HCC})$	110.5	110.5	109.9	110.0	109.9
$\alpha_e(\text{CCN})$	180.0	180.0	180.0	180.0	180.0
$\alpha_e(\text{CNS})$	180.0	180.0	180.0	180.0	180.0
Tot. energy	-530.997790	-531.001956	-530.221358	-530.713316	-530.238372
μ_e^c	5.27	5.35	6.95	2.15	--- ^e
A^d	159.9795	160.0047	159.0590	159.5449	158.7705
B^d	2.4068	2.4146	2.3732	2.3908	2.3783
C^d	2.4068	2.4146	2.3732	2.3908	2.3783

^a Bond angles in degrees, bond lengths in angstrom, total energies in a.u.^b aug-cc-pV(T+d)Z basis set used on S atom.^c Dipole moment in Debye.^d Rotational constants in GHz, Isotopes: C-12, H-1, N-14, S-32.^e Not calculated.**Table S2:** Total energies (in atomic units) and relative Gibbs free energies ($\Delta G^\circ_{0\text{K}}$, in kJ mol⁻¹) of the minima and transition states of the bimolecular reaction routes of CH₃CNS^a

	Total energy	$\Delta G^\circ_{0\text{K}}$		Total energy	$\Delta G^\circ_{0\text{K}}$
2 CH ₃ CNS	2(-530.17886)	0	TS6	-1060.35735	9
TS1	-1060.33846	53	tct	-1060.36203	-2
ctc	-1060.36446	-7	TS7	-1060.35630	11
TS2	-1060.36227	-3	TSs1	-1060.33777	55
ttc	-1060.36733	-15	SP1	-1060.44692	-218
TS5	-1060.35805	7	TSs2	-1060.33722	57
tfur	-1060.43066	-176	SP2	-1060.42932	-177
TS4	-1060.35881	6	TS-t	-1060.33827	50
SS	-1060.42366	-161			
TS3	-1060.36397	-8	2 CH ₃ CN +	2(-132.49828)	
ttt	-1060.37050	-25	¹ S ₂	+ -795.40710	-127

^a Calculated at the MR-AQCC(2,2)//UB3LYP/cc-pVTZ level. Total energies of CH₃CNS and CH₃CN are calculated at the SR-AQCC//B3LYP/cc-pVTZ level.

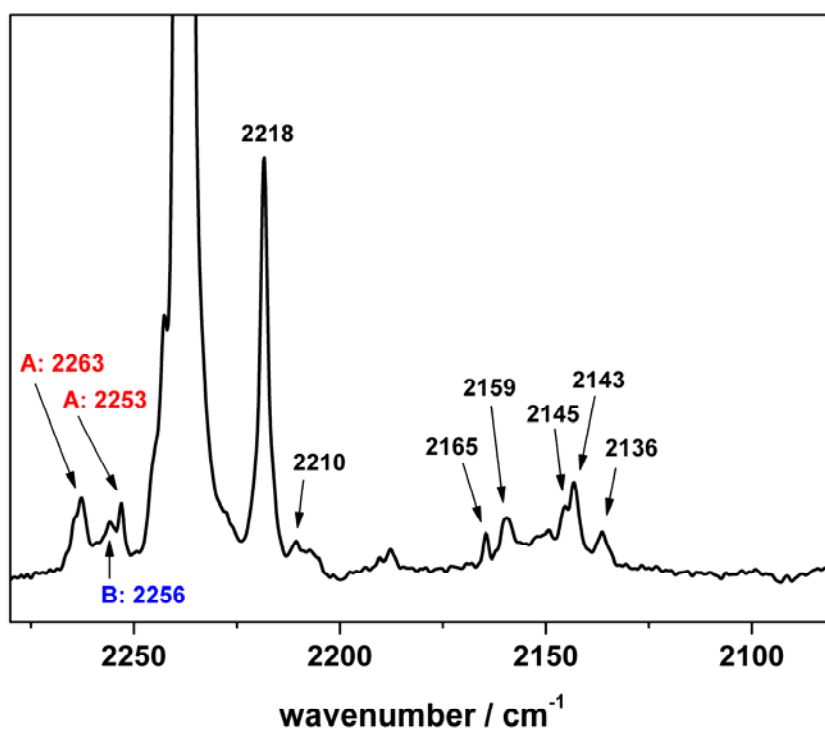


Figure S1. Enlarged experimental IR spectrum obtained as the difference of the spectra of 3,4-dimethyl-1,2,5-thiadiazole in Ar matrix after and before 254 nm UV photolysis. A: CH₃CN in the weak CH₃CN·CH₃CNS van der Waals complex; B: CH₃CN dimer (CH₃CN·CH₃CN van der Waals complex).

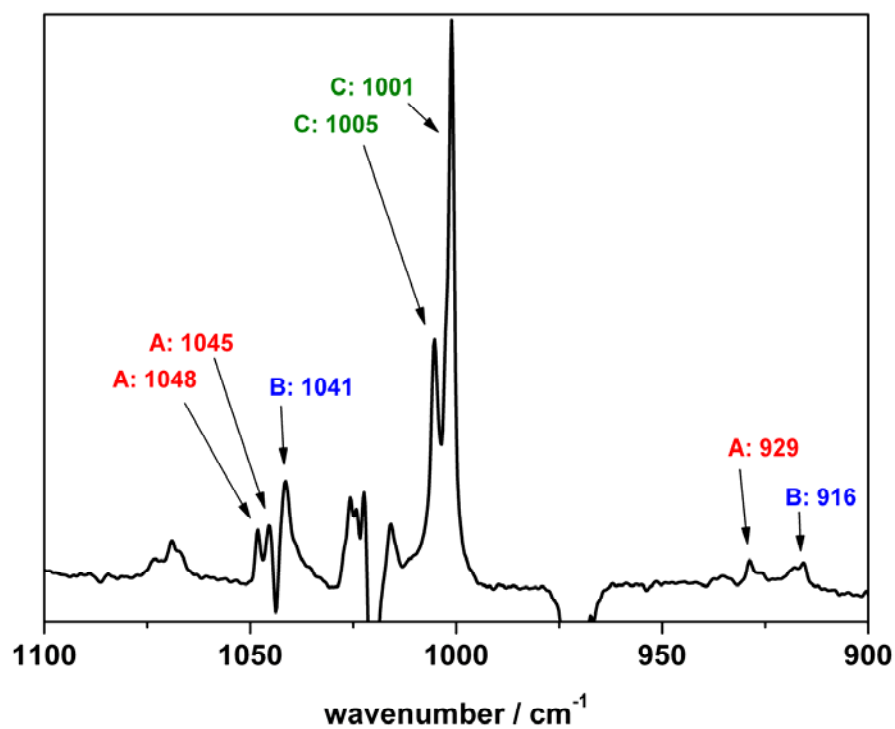


Figure S2. Enlarged experimental IR spectrum obtained as the difference of the spectra of 3,4-dimethyl-1,2,5-thiadiazole in Ar matrix after and before 254 nm UV photolysis. A: CH₃CN in the weak CH₃CN·CH₃CNS van der Waals complex; B: CH₃CN dimer (CH₃CN·CH₃CN van der Waals complex); C: CH₃CNS and/or CH₃CNS in the weak CH₃CNS·CH₃CN van der Waals complex.