Supplementary Information for the manuscript:

Rotational Conformers and Nuclear Spin Isomers of Carbonyl Diisothiocyanate

Eva Gougoula[1,*], Jonathan Pfeiffer[2], Melanie Schnell[1, 3, *], and Frank Tambornino[2,*]

Affiliations:

[1] Deutsches-Elektronen Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany

[2] Fachbereich Chemie, Philipps-Universität Marburg, Hans-Meerwein-Straße 4, 35043 Marburg, Germany

[3] Institut für Physikalische Chemie, Christian-Albrechts-Universität zu Kiel, D-24118 Kiel, Germany

* Corresponding authors: melanie.schnell@desy.de, eva.gougoula@desy.de, tamborni@chemie.uni-marburg.de

ORCIDs:

Eva Gougoula: https://orcid.org/0000-0002-2037-1314

Melanie Schnell: https://orcid.org/0000-0001-7801-7134

Frank Tambornino: https://orcid.org/0000-0003-3538-6049

Keywords: Nuclear Spin Isomers, Chirped-Pulse Fourier Transform Microwave Spectroscopy, Carbonyl Diisothiocyanate, Quantum-Chemical Calculations, Reactive Compounds

1. Methods

1.1 Experimental Methods

Carbonyl diisothiocyanate (CDIT), a clear and viscous liquid at room temperature, was synthesized according to the procedure described by Pfeiffer and co-workers[1]. In brief, a reaction between phosgene and ammonium isothiocyanate at –78 ºC under an inert atmosphere (Schlenk technique) yields CDIT. The above conditions allow for isolating and storing CDIT for a sufficiently long time.

The rotational spectrum of CDIT was recorded in the 2-8 and 8-12 GHz frequency ranges using the COMPACT spectrometer in Hamburg[2,3]. The experiment requires that the molecules of interest are in the gas phase. Due to the air and moisture sensitivity of CDIT, the sample was handled under the atmosphere of a glove box (O_2 < 1.2 ppm, H_2O < 0.5 ppm). CDIT was loaded into the reservoir of a modified pulsed valve, which was then assembled and attached to the solenoid. The valve assembly was placed in an airtight container, equilibrated under glove box atmosphere, and transported to the vacuum chamber where it was connected to the neon gas line. The sample was then pressurized by neon at 3 bar stagnation pressure and heated to 75 ºC while pulsed into the vacuum chamber with an 8 Hz repetition rate.

The sequence of events following the incorporation of the gas pulse generating a molecular ensemble is described briefly. A chirped pulse spanning between 2-8 GHz with 4 μs duration is synthesized by an arbitrary waveform generator (AWG) and amplified by a 300 W traveling wave-tube amplifier. The pulse is broadcast into the vacuum chamber by a horn antenna and excites molecular rotational transitions in resonance with the transmitted frequencies in the fast passage regime. It is noted that the gas pulse and the chirped pulse propagate perpendicular to each other. After the halt of the polarizing pulse, the relaxation of the induced polarization of the molecular ensemble is detected by another horn antenna in the form of a free induction decay (FID) for 40 μs, followed by digitization and Fourier transformation to the frequency domain spectrum by a 100 GS/s oscilloscope. The spectra in the 8-12 GHz region are recorded following the same principles. However, a chirped pulse between 4-6 GHz is generated, which is then frequency doubled to the desired frequencies and amplified by a 50 W solid state amplifier. For both frequency ranges, the duration of the gas pulse, the timescale of the experiment, and the oscilloscope's fast-frame mode allow for eight sequential polarizations of the molecular ensemble, each followed by a FID recording, which are then co-added in the time domain. With this method, spectra containing 1 and 3.2 million averages were recorded in the 2-8 and 8-12 GHz ranges, respectively.

2.1 Quantum Mechanical Calculations

Geometry optimizations of the three possible conformers of CDIT, *syn-syn*, *syn-anti*, and *anti-anti*, were performed with the ORCA 5.0 package[4]. The hybrid functional of Becke-Lee-Yang-Parr (B3LYP)[5] was employed with Grimme's dispersion correction[6–8] (D3BJ) along with the augmented triple-ζ (aug-cc-pVTZ) basis set[9]. The optimized geometries and calculated molecular parameters are summarized in Table 1. Second-order Møller-Plesset perturbation theory[10] (MP2) and the augmented double-ζ (aug-cc-pVDZ) basis set[9] was also used to test the performance of the two methods in the spectral analysis. The MP2 calculated parameters are summarized in Table S1 in the Supplementary Material.

Nudged elastic band calculations[11] (NEB) were performed to get an insight into the barriers to interconversion between the three possible conformers. The method of Perdew-Burke-Ernzerhoff[12] (PBEh-3c) was used to optimize the geometry of the CDIT conformers, followed by NEB calculations of the energy barriers to interconversion at the same level of theory. The calculated barriers to interconversion are summarized in Table 1. Visualization of all structures is achieved with the software Chimera 1.16[13].

2. Supporting Tables and Figures

Table S1. A summary of the theoretically calculated rotational constants of *syn-syn*- and *syn-anti*-CDIT at the B3LYP-D3(BJ)/aug-cc-pVTZ and MP2/aug-cc-pVDZ levels of theory. The numbers in brackets indicate the percentage deviation of the theoretical constants from the experimentally determined ones.

	syn-syn	
	B3LYP-D3(BJ)/aug-cc-pVTZ	MP2-aug-cc-pVDZ
A_e **(MHz)**	10335.6 (–6.4%)‡	10645.8 (–3.6%)
B_e **(MHz)**	443.7 (–1.4%)	437.8 (–2.7%)
C_e **(MHz)**	425.4 (–1.6%)	420.5 (–2.7%)
	syn-anti	
	B3LYP-D3(BJ)/aug-cc-pVTZ	MP2-aug-cc-pVDZ
A_e **(MHz)**	2901.6 (–1.4%)	2819.4 (–4.1%)
B_e **(MHz)**	612.0 (–2.6%)	615.0 (–1.8%)
C_e **(MHz)**	505.4 (–2.3%)	504.9 (–2.2%)

‡The percentage deviations are calculated by means of the formula: $\sigma = \frac{A_e - A_0}{A_0} \times 100$, adapted for each rotational constant.

Table S2. Experimentally determined spectroscopic parameters yielded by Fit 1 and Fit 2 of *syn-syn*-CDIT using Watson's S-reduction as implemented in PGOPHER.

	Fit 1 (*para*)	**Fit 2 (*ortho*)**
A_0 (MHz)	11046.3560(36)[a]	11046.3586(41)
B_0 (MHz)	449.80163(53)	449.80145(53)
C_0 (MHz)	432.27053(70)	432.27030(80)
D_{JK} (kHz)	1.222(39)	1.353(50)
$\chi_{aa}(N_3)$ (MHz)	–1.9176(99) [b]	1.9240(65) [c]
$\chi_{bb}(N_3) - \chi_{cc}(N_3)$ (MHz)	0.772(21)	–0.756(14) [c]
$\chi_{aa}(N_6)$ (MHz)	–	1.9240(65) [c]
$\chi_{bb}(N_6) - \chi_{cc}(N_6)$ (MHz)	–	–0.756(14) [c]
N [c]	42	50
σ_{RMS} [d] (kHz)	13.0	13.2
Q-Branch, $\Delta J = 0, \Delta K_a = 1$	odd[e]	even
P-Branch, $\Delta J = -1, \Delta K_a = 1$	odd	even
R-Branch, $\Delta J = 1, \Delta K_a = 1$	odd	even

[a] Numbers in parentheses are one standard deviation in units of the last significant figures.

[b] The reversed signs of χ_{aa} and $\chi_{bb} - \chi_{cc}$ of Fit 1 when compared to Fit2 arise from the inclusion of only one N nucleus in Fit 1.

[c] The two nitrogen atoms are assumed as equivalent in Fit 2.

[d] Number of hyperfine transitions included in the fit.

[e] Root mean square deviation of the fit.

[f] Odd/even refers to the sum of $K_a'' + K_c'' = K_{sum}''$

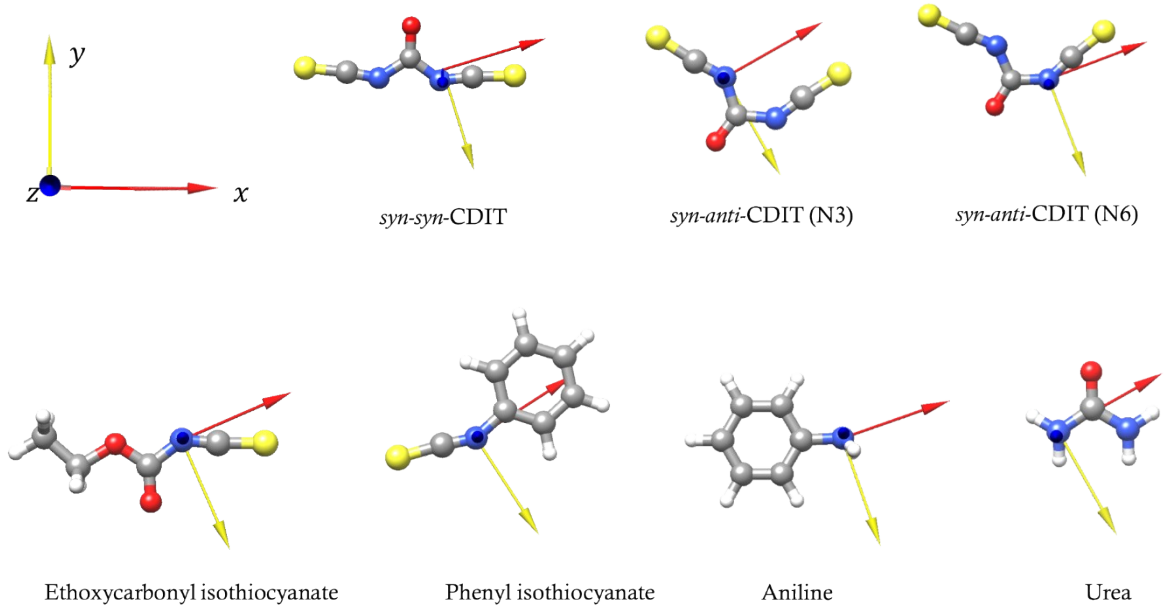

Figure S1. Positioning of the principal nuclear axes , , on each N nucleus of the molecules that are included in Table 6 of the manuscript. The axes are orthogonal and follow a common orientation, regardless of the inertial axes of each molecule.

Table S3. Calculated r_e atomic coordinates of *syn-syn*-CDIT.

***syn-syn*-CDIT @ B3LYP-D3(BJ)/aug-cc-pVTZ**			
	a (Å)	b (Å)	c (Å)
O1	0	1.553	0
C2	0	0.352	0
N3	1.143	–0.441	0
C4	2.337	–0.264	0
S5	3.892	–0.162	0
N6	–1.143	–0.441	0
C7	–2.337	–0.264	0
S8	–3.892	–0.162	0
***syn-syn*-CDIT @ MP2/aug-cc-pVDZ**			
	a (Å)	b (Å)	c (Å)
O1	0	1.509	0
C2	0	0.292	0
N3	2.353	–0.265	0
C4	1.146	–0.523	0
S5	3.919	–0.104	0
N6	–1.146	–0.527	0
C7	–2.353	–0.265	0
S8	–3.919	–0.104	0

Table S4. Calculated r_e atomic coordinates of *syn-anti*-CDIT.

***syn-anti*-CDIT @ B3LYP-D3(BJ)/aug-cc-pVTZ**			
	a (Å)	b (Å)	c (Å)
O1	–0.539	–2.270	0
C2	–0.018	–1.192	0
N3	1.359	–1.010	0
C4	2.137	–0.087	0
S5	3.235	1.019	0
N6	–0.714	0.027	0
C7	–1.878	0.354	0
S8	–3.339	0.895	0
***syn-anti*-CDIT @ MP2/aug-cc-pVDZ**			
	a (Å)	b (Å)	c (Å)
O1	–0.594	–2.276	0
C2	–0.019	–1.207	0
N3	1.379	–1.083	0
C4	2.131	–0.103	0
S5	3.216	1.037	0
N6	–0.655	0.062	0
C7	–1.854	0.364	0
S8	–3.332	0.904	0

Table S5. Calculated r_e atomic coordinates of *anti-anti*-CDIT.

***anti-anti*-CDIT @ B3LYP-D3(BJ)/aug-cc-pVTZ**			
	a (Å)	b (Å)	c (Å)
O1	–0.020	2.981	0
C2	–0.016	1.786	0
N3	–1.185	1.026	0
C4	–1.604	–0.101	0
S5	–2.266	–1.516	0
N6	1.158	1.036	0
C7	1.602	–0.081	0
S8	2.295	–1.481	0
***anti-anti*-CDIT @ MP2/aug-cc-pVDZ**			
	a (Å)	b (Å)	c (Å)
O1	–0.042	3.055	0
C2	–0.026	1.844	0
N3	–1.203	1.070	0
C4	–1.531	–0.118	0
S5	–2.127	–1.579	0
N6	1.170	1.101	0
C7	1.532	–0.076	0
S8	2.17	–1.519	0

3. List of fits with SPFIT/SPCAT

3.1 *syn-syn*-CDIT ($I_{N_3} + I_{N_6} = I_{tot}$, and $I_{tot} + J = F$)

PARAMETERS IN FIT (values truncated and Nlines statistics):

10000	A / /MHz	11046.3581(24)	1
20000	B / /MHz	449.80178(41)	2
30000	C / /MHz	432.27083(56)	3
1100	DJK / /kHz	1.281(38)	4
110010000	3/2(Xaa) /MHz	2.8903(84)	5
-220010000	3/2(Xaa) /MHz	2.8903(84)	= 1.00000 * 5
110040000	1/4(Xbb-Xcc) /MHz	-0.1856(26)	6
-220040000	1/4(Xbb-Xcc) /MHz	-0.1856(26)	= 1.00000 * 6

MICROWAVE AVG = -0.000232 MHz, IR AVG = 0.00000

MICROWAVE RMS = 0.013214 MHz, IR RMS = 0.00000

END OF ITERATION 2 OLD, NEW RMS ERROR= 0.88092 0.88092

distinct frequency lines in fit: 116

distinct parameters of fit: 6

	upper state	lower state	overall
limits of quantum number 1:	1 17	0 16	0 17
limits of quantum number 2:	0 1	0 1	0 1
limits of quantum number 3:	0 17	0 16	0 17
limits of quantum number 4:	0 2	0 2	0 2
limits of quantum number 5:	0 18	0 18	0 18

frequency range: 2357 11847

Linelist L1. *syn-syn*-CDIT

	J''	K_a''	K_c''	I''	F''	J'	K_a'	K_c'	I'	F'	ν **(MHz)**	$\nu_{obs} - \nu_{calc}$
para	8	1	8	1	9	9	0	9	1	10	2357.0905	–0.0078
para	8	1	8	1	7	9	0	9	1	8	2357.0905	0.0025
para	8	1	8	1	8	9	0	9	1	9	2357.2583	–0.0002
orth o	14	0	14	2	16	13	1	13	2	15	2509.0840	0.0057
orth o	14	0	14	2	14	13	1	13	2	13	2509.3532	–0.0013
orth o	7	1	7	2	8	8	0	8	2	8	3306.5238	0.0041
orth o	7	1	7	2	9	8	0	8	2	9	3306.5238	–0.0149
orth o	7	1	7	2	7	8	0	8	2	8	3306.9559	–0.0018
orth o	7	1	7	2	8	8	0	8	2	9	3307.0910	–0.0236
orth o	7	1	7	2	6	8	0	8	2	7	3307.0910	–0.0593
orth o	7	1	7	2	9	8	0	8	2	10	3307.3019	0.0068
orth o	7	1	7	0	7	8	0	8	0	8	3307.3019	–0.0050
orth o	7	1	7	2	5	8	0	8	2	6	3307.3019	–0.0170
para	15	0	15	1	15	14	1	14	1	14	3504.1449	–0.0027
para	15	0	15	1	16	14	1	14	1	15	3504.2813	–0.0019
para	15	0	15	1	14	14	1	14	1	13	3504.2813	–0.0083
para	6	1	6	1	7	7	0	7	1	8	4249.0362	–0.0059
para	6	1	6	1	5	7	0	7	1	6	4249.0362	0.0079
para	6	1	6	1	6	7	0	7	1	7	4249.2236	0.0001
orth o	16	0	16	2	18	15	1	15	2	17	4505.9554	0.0006
orth o	16	0	16	0	16	15	1	15	0	15	4505.9554	0.0066
orth o	16	0	16	2	14	15	1	15	2	13	4505.9554	0.0126

	J''	K_a''	K_c''	I''	F''	J'	K_a'	K_c'	I'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
orth o	16	0	16	2	16	15	1	15	2	15	4506.1948	–0.0259
orth o	5	1	5	2	7	6	0	6	2	7	5182.0838	–0.0052
orth o	5	1	5	2	5	6	0	6	2	6	5182.4594	0.0034
orth o	5	1	5	2	4	6	0	6	2	5	5182.6626	–0.0289
orth o	5	1	5	2	6	6	0	6	2	7	5182.6626	0.0234
orth o	5	1	5	2	3	6	0	6	2	4	5182.8604	–0.0319
orth o	5	1	5	0	5	6	0	6	0	6	5182.8604	–0.0143
orth o	5	1	5	2	7	6	0	6	2	8	5182.8604	0.0030
orth o	5	1	5	2	5	6	0	6	2	5	5183.3455	–0.0098
orth o	5	1	5	2	4	6	0	6	2	4	5183.3455	–0.0007
orth o	5	1	5	2	6	6	0	6	0	6	5183.5171	–0.0039
para	17	0	17	1	18	16	1	16	1	17	5514.3537	0.0122
para	17	0	17	1	16	16	1	16	1	15	5514.3537	0.0066
para	4	1	4	1	5	5	0	5	1	6	6107.9157	0.0033
para	4	1	4	1	3	5	0	5	1	4	6107.9157	0.0278
para	4	1	4	1	4	5	0	5	1	5	6108.1534	0.0070
orth o	3	1	3	2	3	4	0	4	2	4	7024.3186	0.0058
orth o	3	1	3	2	4	4	0	4	2	5	7024.5712	0.0043
orth o	3	1	3	2	2	4	0	4	2	3	7024.7011	0.0037
orth o	3	1	3	0	3	4	0	4	0	4	7024.8967	–0.0527
orth	3	1	3	2	5	4	0	4	2	6	7024.8967	–0.0097

	J''	K_a''	K_c''	I''	F''	J'	K_a'	K_c'	I'	F'	ν_{obs} (MHz)	$\nu_{obs} - \nu_{calc}$
ortho	3	1	3	2	3	4	0	4	2	3	7025.3167	0.0097
ortho	3	1	3	2	4	4	0	4	0	4	7025.5289	0.0054
para	2	1	2	1	2	3	0	3	1	2	7932.3855	0.0100
para	2	1	2	1	1	3	0	3	1	2	7932.6771	0.0052
para	2	1	2	1	3	3	0	3	1	4	7932.8022	0.0118
para	2	1	2	1	2	3	0	3	1	3	7933.2517	0.0097
para	2	1	2	1	3	3	0	3	1	3	7933.4420	0.0098
ortho	1	1	1	0	1	2	0	2	2	2	8831.1725	–0.0112
ortho	1	1	1	2	2	2	0	2	2	2	8831.1725	0.0481
ortho	1	1	1	2	2	2	0	2	2	3	8831.4185	0.0059
ortho	1	1	1	2	3	2	0	2	2	3	8832.0166	0.0034
ortho	1	1	1	0	1	2	0	2	2	1	8832.4680	–0.0358
ortho	1	1	1	2	2	2	0	2	2	1	8832.4680	0.0236
ortho	1	1	1	2	2	2	0	2	0	2	8832.6237	0.0289
ortho	1	1	1	0	1	2	0	2	0	2	8832.6237	–0.0304
ortho	1	1	1	2	3	2	0	2	2	4	8832.8413	0.0024
ortho	1	1	1	2	1	2	0	2	2	1	8833.7897	0.0033
ortho	1	1	1	2	1	2	0	2	0	2	8833.9383	0.0016
ortho	1	1	1	2	1	2	0	2	2	0	8834.2689	0.0017
para	1	1	0	1	1	1	0	1	1	0	10613.2682	–0.0007
para	1	1	0	1	2	1	0	1	1	2	10613.9585	–0.0003

	J''	K_a''	K_c''	I''	F''	J'	K_a'	K_c'	I'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
para	1	1	0	1	1	1	0	1	1	2	10614.1329	–0.0032
para	1	1	0	1	0	1	0	1	1	1	10614.2609	–0.0095
para	1	1	0	1	2	1	0	1	1	1	10614.5359	–0.0008
para	1	1	0	1	1	1	0	1	1	1	10614.7113	–0.0028
orth o	2	1	1	0	2	2	0	2	2	2	10630.3586	0.0025
orth o	2	1	1	2	1	2	0	2	2	2	10630.4648	0.0041
orth o	2	1	1	0	2	2	0	2	2	3	10630.6460	0.0016
orth o	2	1	1	2	4	2	0	2	2	3	10630.8931	0.0019
orth o	2	1	1	2	3	2	0	2	2	2	10631.1753	0.0002
orth o	2	1	1	2	3	2	0	2	2	3	10631.4341	–0.0291
orth o	2	1	1	2	0	2	0	2	2	1	10631.4341	–0.0124
orth o	2	1	1	2	2	2	0	2	2	2	10631.4341	0.0596
orth o	2	1	1	2	4	2	0	2	2	4	10631.7142	–0.0026
orth o	2	1	1	0	2	2	0	2	2	1	10631.7142	0.0380
orth o	2	1	1	2	2	2	0	2	2	3	10631.7142	0.0514
orth o	2	1	1	0	2	2	0	2	0	2	10631.8188	–0.0076
orth o	2	1	1	2	1	2	0	2	0	2	10631.9591	0.0279
orth o	2	1	1	2	3	2	0	2	2	4	10632.2876	–0.0013
orth o	2	1	1	2	1	2	0	2	2	0	10632.2876	0.0260
orth o	2	1	1	2	2	2	0	2	2	1	10632.6919	–0.0026

	J''	K_a''	K_c''	I''	F''	J'	K_a'	K_c'	I'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
orth o	2	1	1	2	3	2	0	2	0	2	10632.6919	0.0464
orth o	2	1	1	2	2	2	0	2	0	2	10632.8336	–0.0113
para	3	1	2	1	3	3	0	3	1	2	10657.1508	–0.0043
para	3	1	2	1	3	3	0	3	1	4	10657.3730	–0.0069
para	3	1	2	1	2	3	0	3	1	2	10657.9545	–0.0181
para	3	1	2	1	4	3	0	3	1	4	10657.9545	–0.0308
para	3	1	2	1	3	3	0	3	1	3	10657.9545	–0.0672
para	3	1	2	1	4	3	0	3	1	3	10658.6183	–0.0087
para	3	1	2	1	2	3	0	3	1	3	10658.8305	–0.0085
orth o	4	1	3	0	4	4	0	4	2	5	10692.2249	–0.0154
orth o	4	1	3	2	3	4	0	4	2	4	10692.2249	0.0022
orth o	4	1	3	2	6	4	0	4	2	5	10692.4247	0.0073
orth o	4	1	3	2	2	4	0	4	2	3	10692.5626	–0.0087
orth o	4	1	3	2	5	4	0	4	2	4	10692.7467	–0.0040
orth o	4	1	3	2	3	4	0	4	2	3	10693.1881	–0.0288
orth o	4	1	3	2	2	4	0	4	2	2	10693.1883	–0.0014
orth o	4	1	3	0	4	4	0	4	0	4	10693.1883	–0.0088
orth o	4	1	3	2	6	4	0	4	2	6	10693.1883	–0.0162
orth o	4	1	3	2	3	4	0	4	2	2	10693.7849	–0.0503
orth o	4	1	3	2	4	4	0	4	2	5	10693.7849	0.0361
orth o	4	1	3	2	3	4	0	4	0	4	10693.7849	0.1172
orth	4	1	3	2	5	4	0	4	2	6	10694.0262	0.0000

	J''	K_a''	K_c''	I''	F''	J'	K_a'	K_c'	I'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
o												
orth												
o	4	1	3	2	5	4	0	4	0	4	10694.2403	0.0445
orth												
o	4	1	3	2	4	4	0	4	2	3	10694.2403	–0.0143
para	5	1	4	1	5	5	0	5	1	4	10736.5237	–0.0035
para	5	1	4	1	5	5	0	5	1	6	10736.6564	–0.0065
para	5	1	4	1	5	5	0	5	1	5	10737.3746	0.0462
para	5	1	4	1	6	5	0	5	1	6	10737.3746	–0.0168
para	5	1	4	1	4	5	0	5	1	4	10737.3746	–0.0296
para	5	1	4	1	6	5	0	5	1	5	10738.0682	0.0113
para	5	1	4	1	4	5	0	5	1	5	10738.1929	–0.0124
orth												
o	6	1	5	2	5	6	0	6	2	6	10789.7150	0.0712
orth												
o	6	1	5	2	8	6	0	6	2	7	10789.7150	–0.0066
orth												
o	6	1	5	2	7	6	0	6	2	6	10790.0055	–0.0208
orth												
o	6	1	5	2	8	6	0	6	2	8	10790.4700	–0.0200
orth												
o	6	1	5	0	6	6	0	6	0	6	10790.4700	–0.0058
orth												
o	6	1	5	2	4	6	0	6	2	4	10790.4700	0.0081
orth												
o	6	1	5	2	6	6	0	6	2	6	10790.6237	–0.0310
orth												
o	6	1	5	2	7	6	0	6	2	7	10790.6237	0.0383
orth												
o	6	1	5	2	5	6	0	6	0	6	10791.0611	–0.0234
orth												
o	6	1	5	2	6	6	0	6	2	7	10791.2064	–0.0073
orth												
o	6	1	5	2	5	6	0	6	2	4	10791.2064	0.0086
orth												
o	6	1	5	2	7	6	0	6	0	6	10791.5301	0.0628

	J''	K_a''	K_c''	I''	F''	J'	K_a'	K_c'	I'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
orth o	6	1	5	2	6	6	0	6	2	5	10791.5301	–0.0240
para	7	1	6	1	7	7	0	7	1	7	10852.7560	0.0158
para	7	1	6	1	8	7	0	7	1	8	10852.8481	0.0110
para	7	1	6	1	6	7	0	7	1	6	10852.8481	–0.0029
orth o	8	1	7	2	7	8	0	8	2	8	10923.4792	0.0157
orth o	8	1	7	2	10	8	0	8	2	9	10923.4792	–0.0020
orth o	8	1	7	2	10	8	0	8	2	10	10924.2309	–0.0067
orth o	8	1	7	0	8	8	0	8	0	8	10924.2309	0.0067
orth o	8	1	7	2	6	8	0	8	2	6	10924.2309	0.0200
orth o	8	1	7	2	8	8	0	8	2	8	10924.4307	–0.0179
orth o	8	1	7	2	9	8	0	8	2	9	10924.4307	0.0749
orth o	8	1	7	2	8	8	0	8	2	7	10925.3010	0.0001
para	9	1	8	1	9	9	0	9	1	9	11005.0958	0.0322
para	9	1	8	1	10	9	0	9	1	10	11005.2084	0.0314
para	9	1	8	1	8	9	0	9	1	8	11005.2084	0.0188
orth o	10	1	9	2	12	10	0	10	2	12	11095.3965	–0.0122
orth o	10	1	9	0	10	10	0	10	0	10	11095.3965	–0.0003
orth o	10	1	9	2	8	10	0	10	2	8	11095.3965	0.0116
orth o	10	1	9	2	10	10	0	10	2	10	11095.6485	0.0031
para	11	1	10	1	11	11	0	11	1	11	11195.3669	–0.0229
para	11	1	10	1	12	11	0	11	1	12	11195.5196	0.0064
para	11	1	10	1	10	11	0	11	1	10	11195.5196	–0.0048
orth	12	1	11	2	14	12	0	12	2	14	11305.1829	–0.0423

	J''	K_a''	K_c''	I''	F''	J'	K_a'	K_c'	I'	F'	ν_{obs} (MHz)	$\nu_{obs} - \nu_{calc}$
o												
orth												
o	12	1	11	0	12	12	0	12	0	12	11305.1829	–0.0316
orth												
o	12	1	11	2	10	12	0	12	2	10	11305.1829	–0.0210
orth												
o	12	1	11	2	13	12	0	12	2	13	11305.3097	–0.0524
orth												
o	12	1	11	2	11	12	0	12	2	11	11305.3097	–0.0205
orth												
o	12	1	11	2	12	12	0	12	2	12	11305.4636	–0.0150
para	13	1	12	1	13	13	0	13	1	13	11425.0734	0.0070
para	13	1	12	1	14	13	0	13	1	14	11425.2159	0.0190
para	13	1	12	1	12	13	0	13	1	12	11425.2159	0.0089
orth												
o	1	1	1	2	2	0	0	0	2	2	11478.1597	0.0004
orth												
o	1	1	1	0	1	0	0	0	0	0	11478.1597	–0.0592
orth												
o	1	1	1	2	3	0	0	0	2	2	11478.7532	–0.0068
orth												
o	1	1	1	2	1	0	0	0	2	2	11479.4960	–0.0052
orth												
o	14	1	13	2	16	14	0	14	2	16	11555.1411	–0.0170
orth												
o	14	1	13	0	14	14	0	14	0	14	11555.1411	–0.0075
orth												
o	14	1	13	2	12	14	0	14	2	12	11555.1411	0.0020
orth												
o	14	1	13	2	13	14	0	14	2	13	11555.2917	0.0197
orth												
o	14	1	13	2	15	14	0	14	2	15	11555.2917	–0.0089
orth												
o	14	1	13	2	14	14	0	14	2	14	11555.4344	0.0099
para	15	1	14	1	15	15	0	15	1	15	11695.6946	0.0091
para	15	1	14	1	16	15	0	15	1	16	11695.8496	0.0278

	J''	K_a''	K_c''	I''	F''	J'	K_a'	K_c'	I'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
para	15	1	14	1	14	15	0	15	1	14	11695.8496	0.0187
orth *o*	16	1	15	2	18	16	0	16	2	18	11846.8990	–0.0194
orth *o*	16	1	15	0	16	16	0	16	0	16	11846.8990	–0.0107
orth *o*	16	1	15	2	14	16	0	16	2	14	11846.8990	–0.0020
orth *o*	16	1	15	2	15	16	0	16	2	15	11847.0453	0.0059
orth *o*	16	1	15	2	17	16	0	16	2	17	11847.0454	–0.0200
orth *o*	16	1	15	2	16	16	0	16	2	16	11847.2036	0.0080

3.2 ^{34}S-*syn*-*syn*-CDIT ($J + I_{N_3} = F_1$ and $F_1 + I_{N_6} = F$)

PARAMETERS IN FIT (values truncated and Nlines statistics):

10000	A / /MHz	11042.9488(27)		1
20000	B / /MHz	438.30798(69)		2
30000	C / /MHz	421.6377(14)		3
110010000	3/2(Xaa)/MHz	2.878(12)		4
-220010000	3/2(Xaa) /MHz	2.878(12)	= 1.00000 *	4
110040000	1/4(Xbb-Xcc)/MHz	-0.1878(42)		5
-220040000	1/4(Xbb-Xcc)/MHz	-0.1878(42)	= 1.00000 *	5
1100	DJK / /kHz	1.86(24)		6

MICROWAVE AVG = -0.001051 MHz, IR AVG = 0.00000

MICROWAVE RMS = 0.014575 MHz, IR RMS = 0.00000

END OF ITERATION 1 OLD, NEW RMS ERROR= 0.97169 0.96117

distinct frequency lines in fit: 74

distinct parameters of fit: 6

	upper state	lower state	overall
limits of quantum number 1:	1 11	0 11	0 11
limits of quantum number 2:	1 1	0 0	0 1
limits of quantum number 3:	0 10	0 11	0 11
limits of quantum number 4:	0 12	0 12	0 12
limits of quantum number 5:	0 13	0 13	0 13

Linelist L2. ^{34}S-*syn*-*syn*-CDIT

J''	K_a''	K_c''	F_1''	F''	J'	K_a'	K_c'	F_1'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
6	1	6	7	8	7	0	7	8	9	4420.5455	0.0114
6	1	6	7	6	7	0	7	8	7	4420.5455	–0.0030
6	1	6	5	4	7	0	7	6	5	4420.5455	–0.0181
5	1	5	5	5	6	0	6	6	6	5329.1825	0.0112
5	1	5	6	5	6	0	6	7	6	5329.5930	–0.0055
5	1	5	6	7	6	0	6	7	8	5329.5930	0.0127
2	1	2	2	2	3	0	3	3	3	8007.5807	0.0055
2	1	2	2	3	3	0	3	3	4	8007.9188	0.0121
2	1	2	2	1	3	0	3	3	2	8007.9188	–0.0631
2	1	2	3	3	3	0	3	4	4	8008.0805	–0.0218
2	1	2	1	1	3	0	3	2	2	8008.2703	0.0004
2	1	2	3	4	3	0	3	4	5	8008.4472	–0.0101
1	1	1	1	1	2	0	2	2	2	8883.4933	–0.0307
1	1	1	1	2	2	0	2	2	2	8883.4933	0.0289
1	1	1	1	2	2	0	2	2	3	8883.7420	–0.0083
1	1	1	2	3	2	0	2	2	3	8884.3505	–0.0030
1	1	1	2	2	2	0	2	3	3	8884.5603	–0.0020
1	1	1	2	1	2	0	2	2	1	8884.5603	–0.0624
1	1	1	1	2	2	0	2	1	2	8884.9488	0.0252
1	1	1	1	1	2	0	2	1	2	8884.9488	–0.0344
1	1	1	2	3	2	0	2	3	4	8885.1715	–0.0013
1	1	1	2	1	2	0	2	1	1	8885.5758	–0.0028
1	1	1	0	1	2	0	2	1	1	8886.1085	–0.0133
1	1	1	0	1	2	0	2	1	2	8886.2710	0.0000
1	1	1	0	1	2	0	2	1	0	8886.6110	0.0119
1	1	0	2	1	1	0	1	1	0	10620.4679	–0.0273
1	1	0	1	1	1	0	1	1	2	10620.4679	0.0041
1	1	0	2	3	1	0	1	1	2	10620.7005	0.0041
1	1	0	2	2	1	0	1	2	2	10621.1640	–0.0204
1	1	0	0	1	1	0	1	1	1	10621.1640	0.0641
1	1	0	2	1	1	0	1	2	2	10621.3272	–0.0286

J''	K_a''	K_c''	F_1''	F''	J'	K_a'	K_c'	F_1'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
1	1	0	1	2	1	0	1	2	3	10621.3272	0.0278
1	1	0	2	3	1	0	1	2	3	10621.5611	0.0040
1	1	0	1	0	1	0	1	2	1	10621.5611	0.0603
1	1	0	2	2	1	0	1	2	1	10621.7690	0.0110
1	1	0	2	1	1	0	1	2	1	10621.9401	0.0107
1	1	0	1	1	1	0	1	0	1	10622.3788	–0.0071
1	1	0	1	2	1	0	1	0	1	10622.3788	0.0178
2	1	1	1	2	2	0	2	2	2	10636.7189	–0.0004
2	1	1	1	1	2	0	2	2	2	10636.8307	0.0063
2	1	1	1	2	2	0	2	2	3	10637.0133	0.0079
2	1	1	3	4	2	0	2	2	3	10637.2415	–0.0117
2	1	1	3	2	2	0	2	3	3	10637.5370	0.0175
2	1	1	2	3	2	0	2	2	2	10637.5370	–0.0047
2	1	1	1	0	2	0	2	1	1	10637.7922	–0.0066
2	1	1	2	3	2	0	2	2	3	10637.7922	–0.0355
2	1	1	2	1	2	0	2	2	1	10637.7922	–0.0560
2	1	1	3	3	2	0	2	3	3	10637.9732	0.0229
2	1	1	3	4	2	0	2	3	4	10638.0907	0.0181
2	1	1	3	2	2	0	2	3	2	10638.0907	–0.0430
2	1	1	1	1	2	0	2	1	0	10638.6326	0.0209
2	1	1	2	3	2	0	2	3	4	10638.6326	–0.0145
2	1	1	2	1	2	0	2	3	2	10638.8009	–0.0033
2	1	1	2	2	2	0	2	1	1	10639.0667	0.0147
3	1	2	2	3	3	0	3	3	4	10662.0114	–0.0446
3	1	2	2	2	3	0	3	3	3	10662.0114	0.0362
3	1	2	4	5	3	0	3	3	4	10662.2666	–0.0039
3	1	2	4	3	3	0	3	3	2	10662.2666	0.0520
3	1	2	3	4	3	0	3	3	3	10662.5352	–0.0761
3	1	2	3	2	3	0	3	3	3	10662.5352	0.0145
3	1	2	2	1	3	0	3	2	2	10662.5352	0.0198
3	1	2	2	3	3	0	3	2	2	10662.7468	0.0222
3	1	2	2	2	3	0	3	2	3	10663.4203	0.0065
3	1	2	3	3	3	0	3	3	4	10663.4203	–0.0044

J''	K_a''	K_c''	F_1''	F''	J'	K_a'	K_c'	F_1'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
3	1	2	3	3	3	0	3	2	2	10664.1168	0.0235
4	1	3	3	3	4	0	4	4	4	10695.5454	0.0018
4	1	3	3	4	4	0	4	4	5	10695.5454	–0.0120
4	1	3	5	6	4	0	4	4	5	10695.7512	0.0167
4	1	3	3	2	4	0	4	3	3	10695.9136	0.0290
4	1	3	4	5	4	0	4	4	4	10696.0775	0.0057
4	1	3	3	4	4	0	4	3	3	10696.0775	0.0179
4	1	3	5	6	4	0	4	5	6	10696.4969	–0.0188
4	1	3	5	4	4	0	4	5	4	10696.4969	–0.0120
4	1	3	3	4	4	0	4	3	4	10696.4969	–0.0100
4	1	3	3	2	4	0	4	3	2	10696.4969	–0.0013
4	1	3	4	4	4	0	4	4	5	10697.0643	–0.0018
4	1	3	3	3	4	0	4	3	2	10697.0643	–0.0795
4	1	3	4	5	4	0	4	5	6	10697.3199	–0.0177
4	1	3	4	3	4	0	4	5	4	10697.3199	–0.0501
4	1	3	4	4	4	0	4	3	3	10697.5663	–0.0019
5	1	4	4	3	5	0	5	4	4	10737.7980	0.0021
5	1	4	6	5	5	0	5	6	6	10737.7980	0.0102
5	1	4	4	3	5	0	5	4	3	10738.4797	0.0479
5	1	4	4	5	5	0	5	4	5	10738.4797	0.0341
5	1	4	6	5	5	0	5	6	5	10738.4797	0.0315
5	1	4	6	7	5	0	5	6	7	10738.4797	0.0202
5	1	4	4	4	5	0	5	4	4	10738.4797	–0.0175
5	1	4	6	6	5	0	5	6	6	10738.4797	–0.0363
5	1	4	5	4	5	0	5	5	4	10738.4797	–0.0502
5	1	4	5	6	5	0	5	5	6	10738.4797	–0.0590
5	1	4	4	4	5	0	5	4	3	10739.1187	–0.0144
5	1	4	5	5	5	0	5	5	6	10739.1187	–0.0005
5	1	4	5	4	5	0	5	6	5	10739.3144	–0.0105
5	1	4	5	6	5	0	5	6	7	10739.3144	0.0051
5	1	4	5	5	5	0	5	4	4	10739.5479	0.0251
6	1	5	7	8	6	0	6	7	8	10788.9690	0.0043
6	1	5	7	6	6	0	6	7	6	10788.9690	0.0169

J''	K_a''	K_c''	F_1''	F''	J'	K_a'	K_c'	F_1'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
6	1	5	5	6	6	0	6	5	6	10788.9690	0.0192
6	1	5	5	4	6	0	6	5	4	10788.9690	0.0341
7	1	6	8	9	7	0	7	8	9	10848.1277	0.0114
7	1	6	8	7	7	0	7	8	7	10848.1277	0.0241
7	1	6	6	7	7	0	7	6	7	10848.1277	0.0261
7	1	6	6	5	7	0	7	6	5	10848.1277	0.0407
8	1	7	9	10	8	0	8	9	10	10916.0058	–0.0095
8	1	7	9	8	8	0	8	9	8	10916.0058	0.0028
8	1	7	7	8	8	0	8	7	8	10916.0058	0.0044
8	1	7	7	6	8	0	8	7	6	10916.0058	0.0184
8	1	7	8	7	8	0	8	8	7	10916.1251	–0.0029
8	1	7	8	9	8	0	8	8	9	10916.1251	–0.0136
9	1	8	8	7	9	0	9	8	7	10992.7671	0.0153
9	1	8	8	9	9	0	9	8	9	10992.7671	0.0021
9	1	8	10	9	9	0	9	10	9	10992.7671	0.0008
9	1	8	8	8	9	0	9	8	8	10992.9007	0.0322
9	1	8	10	10	9	0	9	10	10	10992.9007	0.0162
9	1	8	10	10	9	0	9	10	10	10992.9007	0.0162
9	1	8	9	10	9	0	9	9	10	10992.9007	–0.0073
10	1	9	11	12	10	0	10	11	12	11078.5223	–0.0133
10	1	9	11	10	10	0	10	11	10	11078.5223	–0.0020
10	1	9	9	8	10	0	10	9	8	11078.5223	0.0115
10	1	9	10	11	10	0	10	10	11	11078.6608	–0.0096
10	1	9	9	9	10	0	10	9	9	11078.6612	0.0281
10	1	9	11	11	10	0	10	11	11	11078.6612	0.0133
10	1	9	10	9	10	0	10	10	9	11078.6612	0.0009
10	1	9	10	10	10	0	10	10	10	11078.7865	0.0050
11	1	10	12	13	11	0	11	12	13	11173.3975	–0.0355
11	1	10	12	11	11	0	11	12	11	11173.3975	–0.0248
11	1	10	10	9	11	0	11	10	9	11173.3975	–0.0121
11	1	10	10	10	11	0	11	10	10	11173.5415	0.0048
11	1	10	12	12	11	0	11	12	12	11173.5415	–0.0088
11	1	10	11	10	11	0	11	11	10	11173.5415	–0.0205

J''	K_a''	K_c''	F_1''	F''	J'	K_a'	K_c'	F_1'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
11	1	10	11	12	11	0	11	11	12	11173.5415	–0.0302
1	1	1	1	0	0	0	0	1	1	11463.9059	–0.0089
1	1	1	1	1	0	0	0	1	0	11464.1184	–0.0572
1	1	1	1	2	0	0	0	1	2	11464.1184	0.0027
1	1	1	2	2	0	0	0	1	1	11464.4970	–0.0207
1	1	1	2	3	0	0	0	1	2	11464.7003	–0.0184
1	1	1	2	1	0	0	0	1	1	11464.8961	–0.0236
1	1	1	0	1	0	0	0	1	2	11465.4479	–0.0152

3.3 ^{13}C-*syn*-*syn*-CDIT ($J + I_{N_3} = F_1$ and $F_1 + I_{N_6} = F$)

PARAMETERS IN FIT (values truncated and Nlines statistics):

10000	A / /MHz	11029.0588(35)		1
20000	B / /MHz	447.6461(18)		2
30000	C / /MHz	430.2687(18)		3
110010000	3/2(Xaa) /MHz	2.943(19)		4
-220010000	3/2(Xaa) /MHz	2.943(19)	= 1.00000 *	4
110040000	1/4(Xbb-Xcc)/MHz	-0.1731(60)		5
-220040000	1/4(Xbb-Xcc) /MHz	-0.1731(60)	= 1.00000 *	5
1100	DJK / /kHz	[-1.861]		6

MICROWAVE AVG = -0.000000 MHz, IR AVG = 0.00000

MICROWAVE RMS = 0.014865 MHz, IR RMS = 0.00000

END OF ITERATION 1 OLD, NEW RMS ERROR= 0.99100 0.99100

distinct frequency lines in fit: 32

distinct parameters of fit: 5

	upper state		lower state		overall	
limits of quantum number 1:	1	9	0	9	0	9
limits of quantum number 2:	1	1	0	0	0	1
limits of quantum number 3:	0	8	0	9	0	9
limits of quantum number 4:	0	9	0	9	0	9
limits of quantum number 5:	0	9	0	9	0	9

frequency range: 8824 11460

Linelist L3. ^{13}C-*syn*-*syn*-CDIT

J''	K_a''	K_c''	F_1''	F''	J'	K_a'	K_c'	F_1'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
1	1	1	1	2	2	0	2	2	3	8824.5748	–0.0076
1	1	1	2	2	2	0	2	3	3	8825.3893	–0.0120
1	1	1	2	3	2	0	2	3	4	8826.0094	–0.0114
1	1	1	2	2	2	0	2	3	2	8826.0094	–0.0225
1	1	0	1	1	1	0	1	1	1	10597.9722	–0.0277
1	1	0	2	1	1	0	1	1	0	10597.9722	0.0012
1	1	0	1	2	1	0	1	1	1	10597.9722	0.0001
1	1	0	2	3	1	0	1	1	2	10598.1834	0.0132
1	1	0	2	2	1	0	1	2	2	10598.6469	–0.0169
1	1	0	0	1	1	0	1	1	1	10598.6469	0.0360
1	1	0	1	2	1	0	1	2	3	10598.8155	0.0481
1	1	0	2	1	1	0	1	2	2	10598.8155	–0.0386
1	1	0	2	3	1	0	1	2	3	10599.0432	–0.0101
1	1	0	2	2	1	0	1	2	1	10599.2611	0.0086
1	1	0	2	1	1	0	1	2	1	10599.4365	–0.0062
1	1	0	1	1	1	0	1	0	1	10599.8587	–0.0260
1	1	0	1	2	1	0	1	0	1	10599.8587	0.0018
2	1	1	3	4	2	0	2	2	3	10615.4451	–0.0047
2	1	1	3	2	2	0	2	2	1	10615.4451	0.0675
2	1	1	3	2	2	0	2	3	3	10615.7403	0.0121
2	1	1	2	3	2	0	2	2	2	10615.7403	0.0150
2	1	1	2	1	2	0	2	2	1	10615.9785	–0.0630
2	1	1	1	0	2	0	2	1	1	10615.9785	–0.0483
2	1	1	2	3	2	0	2	2	3	10615.9785	–0.0403
2	1	1	2	2	2	0	2	2	2	10615.9785	0.0548
2	1	1	3	3	2	0	2	3	3	10616.1806	0.0258
2	1	1	3	4	2	0	2	3	4	10616.3016	0.0108
2	1	1	2	3	2	0	2	3	4	10616.8485	–0.0112
2	1	1	1	1	2	0	2	1	0	10616.8485	–0.0005
2	1	1	2	2	2	0	2	1	1	10617.2794	0.0113

3	1	2	2	1	3	0	3	2	1	10642.4125	0.0057
J''	K_a''	K_c''	F_1''	F''	J'	K_a'	K_c'	F_1'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
3	1	2	2	3	3	0	3	2	3	10642.4125	0.0221
3	1	2	4	3	3	0	3	4	3	10642.4125	0.0271
3	1	2	4	5	3	0	3	4	5	10642.4125	0.0390
3	1	2	2	2	3	0	3	2	2	10642.4125	0.0485
3	1	2	2	2	3	0	3	2	3	10642.7185	–0.0079
3	1	2	3	3	3	0	3	3	4	10642.7185	0.0080
5	1	4	5	5	5	0	5	5	5	10721.1279	–0.0336
5	1	4	5	6	5	0	5	5	6	10721.1279	0.0085
5	1	4	5	4	5	0	5	5	4	10721.1279	0.0154
5	1	4	6	6	5	0	5	6	6	10721.1279	0.0262
5	1	4	4	4	5	0	5	4	4	10721.1279	0.0410
6	1	5	5	4	6	0	6	5	4	10773.7903	0.0387
6	1	5	5	6	6	0	6	5	6	10773.7903	0.0263
6	1	5	7	6	6	0	6	7	6	10773.7903	0.0244
6	1	5	7	8	6	0	6	7	8	10773.7903	0.0138
7	1	6	7	8	7	0	7	7	8	10835.5808	–0.0481
7	1	6	7	6	7	0	7	7	6	10835.5808	–0.0388
7	1	6	8	8	7	0	7	8	8	10835.5808	–0.0263
7	1	6	6	6	7	0	7	6	6	10835.5808	–0.0103
7	1	6	8	9	7	0	7	8	9	10835.5808	0.0500
7	1	6	8	7	7	0	7	8	7	10835.5808	0.0609
7	1	6	6	5	7	0	7	6	5	10835.5808	0.0751
7	1	6	7	7	7	0	7	7	7	10835.7093	0.0058
8	1	7	7	7	8	0	8	7	7	10906.4933	–0.0079
8	1	7	9	9	8	0	8	9	9	10906.4933	–0.0230
8	1	7	8	7	8	0	8	8	7	10906.4933	–0.0352
8	1	7	8	9	8	0	8	8	9	10906.4933	–0.0445
9	1	8	9	9	9	0	9	9	9	10986.7980	–0.0101
1	1	1	1	0	0	0	0	1	1	11458.6422	–0.0254
1	1	1	1	1	0	0	0	1	0	11458.8639	–0.0621
1	1	1	1	2	0	0	0	1	2	11458.8639	–0.0027
1	1	1	2	2	0	0	0	1	1	11459.2558	–0.0090

1	1	1	2	3	0	0	0	1	2	11459.4591	–0.0049
J''	K_a''	K_c''	F_1''	F''	J'	K_a'	K_c'	F_1'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
1	1	1	2	1	0	0	0	1	1	11459.6598	–0.0031
1	1	1	0	1	0	0	0	1	2	11460.1987	–0.0025

3.4 ^{15}N-*syn*-*syn*-CDIT ($J + I_N = F$)

PARAMETERS IN FIT (values truncated and Nlines statistics):

10000	A / /MHz	11002.9064(90)	1
20000	B / /MHz	449.3188(98)	2
30000	C / /MHz	431.7588(90)	3
110010000	1.5 Chi.aa) /MHz	[2.9805]	4
110040000	0.25 (Chi.bb-Chi.cc) /MHz	[-0.18775]	5
1100	DJK / /kHz	[-1.861]	6

MICROWAVE AVG = -0.000000 MHz, IR AVG = 0.00000

MICROWAVE RMS = 0.010786 MHz, IR RMS = 0.00000

END OF ITERATION 2 OLD, NEW RMS ERROR= 0.71908 0.71908

distinct frequency lines in fit: 6

distinct parameters of fit: 3

	upper state		lower state		overall	
limits of quantum number 1:	1	6	0	6	0	6
limits of quantum number 2:	1	1	0	0	0	1
limits of quantum number 3:	0	5	0	6	0	6
limits of quantum number 4:	1	6	1	6	1	6

frequency range: 10571 11434

Linelist L4. ^{15}N-*syn*-*syn*-CDIT

J''	K_a''	K_c''	F''	J'	K_a'	K_c'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
1	1	0	2	1	0	1	2	10571.2864	0.0048
2	1	1	1	2	0	2	1	10588.8764	–0.0185
3	1	2	4	3	0	3	4	10615.1932	0.0175
5	1	4	4	5	0	5	4	10694.7015	–0.0044
6	1	5	6	6	0	6	6	10748.0891	0.0006
1	1	1	2	0	0	0	1	11434.7375	0.0000

3.5 *syn-anti*-CDIT ($J + I_{N_3} = F_1$ and $F_1 + I_{N_6} = F$)

PARAMETERS IN FIT (values truncated and Nlines statistics):

10000	A / /MHz	2938.6797(15)	1
20000	B / /MHz	626.51962(71)	2
30000	C / /MHz	516.14194(50)	3
110010000	3/2(Xaa) /MHz	2.618(16)	4
110040000	1/4(Xbb-Xcc) /MHz	-0.1596(45)	5
220010000	3/2(Xaa) /MHz	[0.]	6
220040000	1/4(Xbb-Xcc) /MHz	0.2203(57)	7
1100	DJK / /kHz	-3.099(39)	8
200	DJ / /kHz	0.2008(52)	9
2000	DK /MHz	-0.02074(16)	10
40100	d1 /kHz	-0.0779(19)	11

MICROWAVE AVG = -0.000349 MHz, IR AVG = 0.00000

MICROWAVE RMS = 0.016327 MHz, IR RMS = 0.00000

END OF ITERATION 7 OLD, NEW RMS ERROR= 1.08849 1.08849

distinct frequency lines in fit: 98

distinct parameters of fit: 10

	upper state	lower state	overall
limits of quantum number 1:	1 10	0 9	0 10
limits of quantum number 2:	0 3	0 2	0 3
limits of quantum number 3:	0 10	0 9	0 10
limits of quantum number 4:	0 10	0 9	0 10
limits of quantum number 5:	1 11	0 10	0 11

frequency range: 2422 11963

Linelist L5. *syn-anti*-CDIT

J''	K_a''	K_c''	F_1''	F''	J'	K_a'	K_c'	F_1'	F'	ν (MHz)	$\nu_{obs} - \nu_{calc}$
1	1	0	2	1	1	0	1	1	0	2422.1900	–0.0184
1	1	0	1	1	1	0	1	2	1	2422.4504	–0.0067
1	1	0	1	2	1	0	1	2	3	2422.4504	0.0157
1	1	0	2	2	1	0	1	2	2	2422.5764	–0.0103
1	1	0	0	1	1	0	1	2	1	2422.9420	0.0024
1	1	0	1	2	1	0	1	0	1	2423.2048	–0.0152
1	1	0	2	2	1	0	1	0	1	2423.3865	0.0144
4	0	4	3	4	3	1	3	4	3	2479.0136	–0.0077
4	0	4	3	3	3	1	3	2	2	2479.0136	0.0037
4	0	4	4	4	3	1	3	3	3	2479.2595	–0.0074
2	1	1	3	3	2	0	2	3	2	2536.7151	0.0028
2	1	1	2	1	2	0	2	2	1	2536.7151	0.0104
2	1	1	1	2	2	0	2	1	2	2536.8960	–0.0107
2	1	1	1	2	2	0	2	1	1	2536.8960	–0.0077
2	1	1	3	2	2	0	2	3	2	2536.8960	0.0244
3	1	2	4	4	3	0	3	4	4	2715.1840	–0.0008
3	1	2	3	3	3	0	3	3	3	2715.1840	–0.0040
3	1	2	2	2	3	0	3	2	2	2715.1840	–0.0221
3	1	2	4	5	3	0	3	4	5	2715.3646	0.0263
3	1	2	3	2	3	0	3	3	2	2715.3646	0.0171
3	1	2	4	3	3	0	3	4	3	2715.3646	–0.0075
4	1	3	4	4	4	0	4	4	4	2966.2496	–0.0466
4	1	3	5	5	4	0	4	5	5	2966.2496	0.0107
4	1	3	3	3	4	0	4	3	3	2966.2496	0.0150
4	1	3	4	5	4	0	4	4	5	2966.4576	0.0154
4	1	3	5	4	4	0	4	5	4	2966.4576	0.0172
4	1	3	4	3	4	0	4	4	4	2966.4576	0.0094
4	1	3	3	2	4	0	4	3	2	2966.4576	0.0141
5	1	4	4	4	5	0	5	4	4	3300.1086	–0.0109
5	1	4	6	6	5	0	5	6	6	3300.1086	–0.0237
5	1	4	5	5	5	0	5	5	5	3300.2085	–0.0154

5	1	4	5	6	5	0	5	5	6	3300.3899	–0.0034
1	1	1	1	2	0	0	0	1	2	3454.5288	0.0167
1	1	1	0	1	0	0	0	1	2	3455.4383	0.0197
6	1	5	5	5	6	0	6	5	5	3728.0243	0.0158
6	1	5	7	7	6	0	6	7	7	3728.0243	–0.0001
6	1	5	5	6	6	0	6	5	5	3728.1419	0.0038
6	1	5	6	7	6	0	6	6	6	3728.2667	–0.0008
6	1	5	6	7	6	0	6	6	7	3728.3590	0.0278
5	0	5	4	5	4	1	4	5	6	3774.0417	–0.0207
5	0	5	6	6	4	1	4	5	5	3774.0417	–0.0358
5	0	5	5	5	4	1	4	4	4	3774.2252	–0.0259
7	1	6	8	8	7	0	7	8	8	4260.2846	0.0169
7	1	6	6	7	7	0	7	6	7	4260.4433	–0.0187
7	1	6	7	8	7	0	7	7	8	4260.6200	0.0071
2	1	2	1	2	1	0	1	1	2	4486.5126	0.0148
2	1	2	3	3	1	0	1	1	2	4486.6855	–0.0107
2	1	2	2	2	1	0	1	1	1	4486.8704	–0.0099
2	1	2	3	4	1	0	1	2	3	4487.1317	0.0156
2	1	2	1	2	1	0	1	0	1	4487.8222	0.0153
2	1	2	1	1	1	0	1	0	1	4487.8222	–0.0238
6	0	6	5	5	5	1	5	4	4	5078.4935	–0.0020
6	0	6	6	6	5	1	5	5	5	5078.6394	0.0099
3	1	3	3	4	2	0	2	2	3	5465.5089	0.0216
3	1	3	2	2	2	0	2	3	2	5465.7091	0.0086
3	1	3	4	4	2	0	2	3	3	5465.8525	0.0253
3	1	3	2	1	2	0	2	1	1	5465.8525	0.0149
3	1	3	4	4	2	0	2	3	4	5465.8525	0.0194
3	1	3	2	1	2	0	2	1	0	5465.8525	0.0073
3	1	3	3	4	2	0	2	3	4	5466.0391	–0.0047
9	1	8	9	9	9	0	9	9	9	5662.9499	–0.0069
9	1	8	9	10	9	0	9	9	10	5663.1756	–0.0179
8	2	6	7	8	8	1	7	7	8	5970.9827	–0.0182
8	2	6	7	6	8	1	7	7	6	5970.9827	–0.0142
8	2	6	9	10	8	1	7	9	10	5970.9827	–0.0064

6	2	4	6	6	6	1	5	6	6	6239.0766	–0.0190
6	2	4	7	7	6	1	5	7	7	6239.2488	–0.0320
7	0	7	7	7	6	1	6	6	6	6378.6980	–0.0352
7	0	7	8	7	6	1	6	7	6	6378.6980	–0.0233
7	0	7	8	9	6	1	6	7	8	6378.6980	–0.0097
7	0	7	6	7	6	1	6	5	6	6378.6980	–0.0047
4	1	4	4	5	3	0	3	3	4	6395.3961	0.0156
4	1	4	3	2	3	0	3	2	1	6395.5927	–0.0183
4	1	4	4	5	3	0	3	4	5	6395.9362	–0.0136
5	2	3	5	5	5	1	4	5	5	6424.0787	0.0092
5	2	3	6	6	5	1	4	6	6	6424.3310	0.0061
5	2	3	6	7	5	1	4	6	6	6424.3310	–0.0369
4	2	2	4	4	4	1	3	4	4	6614.8737	0.0168
4	2	2	5	5	4	1	3	5	5	6615.2343	0.0156
3	2	1	4	4	3	1	2	4	4	6792.6605	0.0004
3	2	1	2	2	3	1	2	2	2	6792.8327	–0.0024
2	2	0	3	3	2	1	1	3	3	6940.4017	0.0131
2	2	1	2	3	2	1	2	2	3	7266.7666	–0.0074
2	2	1	2	1	2	1	2	2	1	7266.7666	–0.0254
2	2	1	2	3	2	1	2	3	4	7266.9813	–0.0085
2	2	1	2	1	2	1	2	1	0	7267.1307	–0.0363
2	2	1	3	4	2	1	2	2	3	7267.3212	–0.0138
2	2	1	1	1	2	1	2	1	0	7268.0233	–0.0164
5	1	5	5	6	4	0	4	4	5	7284.4296	0.0036
5	1	5	6	7	4	0	4	5	6	7284.5772	0.0125
5	1	5	4	3	4	0	4	3	2	7284.5772	–0.0223
3	2	2	2	3	3	1	3	2	3	7435.5717	0.0051
4	2	3	4	5	4	1	4	4	5	7660.5124	–0.0209
4	2	3	4	3	4	1	4	4	3	7660.5276	–0.0362
4	2	3	5	5	4	1	4	5	5	7660.5276	–0.0276
4	2	3	3	3	4	1	4	3	3	7660.5755	–0.0289
8	0	8	7	8	7	1	7	6	7	7662.3325	0.0149
8	0	8	9	10	7	1	7	8	9	7662.3325	0.0126
8	0	8	8	8	7	1	7	7	7	7662.3325	–0.0031

5	2	4	6	6	5	1	5	6	6	7943.9903	0.0226
5	2	4	6	5	5	1	5	6	5	7944.1373	–0.0084
6	1	6	7	8	5	0	5	6	7	8143.3137	0.0015
6	1	6	6	6	5	0	5	5	5	8143.3137	0.0061
6	1	6	5	4	5	0	5	4	3	8143.3137	–0.0201
6	2	5	5	5	6	1	6	5	5	8286.3926	0.0331
6	2	5	7	7	6	1	6	7	7	8286.3926	0.0386
7	2	6	7	7	7	1	7	7	7	8688.3111	0.0119
7	2	6	7	6	7	1	7	7	6	8688.4923	0.0172
9	0	9	9	10	8	1	8	8	9	8920.2199	–0.0121
9	0	9	9	9	8	1	8	8	8	8920.2199	0.0333
7	1	7	7	6	6	0	6	6	5	8984.1318	–0.0037
7	1	7	7	7	6	0	6	6	6	8984.2510	0.0011
7	1	7	6	5	6	0	6	5	6	8984.2510	0.0025
7	1	7	8	9	6	0	6	7	8	8984.2510	0.0042
7	1	7	8	7	6	0	6	7	6	8984.2510	0.0170
7	1	7	6	6	6	0	6	5	5	8984.3432	–0.0343
7	1	7	8	8	6	0	6	7	7	8984.3432	–0.0071
2	2	1	2	1	1	1	0	1	0	9331.7511	0.0066
2	2	1	3	2	1	1	0	0	1	9331.7511	0.0237
2	2	1	3	3	1	1	0	2	2	9332.0921	0.0118
2	2	1	1	1	1	1	0	1	0	9332.5976	–0.0196
8	1	8	9	8	7	0	7	8	7	9820.4055	–0.0115
8	1	8	8	9	7	0	7	7	8	9820.4055	0.0609
3	2	2	3	3	2	1	1	2	2	10364.0055	0.0055
3	2	2	4	4	2	1	1	3	3	10364.4464	–0.0023
3	2	2	2	1	2	1	1	1	0	10364.4464	–0.0066
9	1	9	9	10	8	0	8	8	9	10663.8180	0.0335
9	1	9	10	9	8	0	8	9	8	10663.8180	–0.0259
3	2	1	3	4	2	1	2	2	3	10714.4929	–0.0276
3	2	1	4	3	2	1	2	2	3	10714.4929	–0.0229
3	2	1	2	2	2	1	2	2	1	10714.4929	–0.0120
3	2	1	2	3	2	1	2	2	3	10714.4929	–0.0047
3	2	1	4	4	2	1	2	2	3	10714.4929	0.0020

3	2	1	2	1	2	1	2	1	0	10714.9140	0.0121
4	2	3	4	3	3	1	2	3	3	11340.7267	0.0140
4	2	3	3	4	3	1	2	4	4	11340.9858	0.0008
7	3	4	6	7	7	2	5	6	7	11515.6729	–0.0119
7	3	4	8	8	7	2	5	8	8	11515.6729	–0.0390
7	3	4	6	6	7	2	5	6	6	11515.6729	–0.0692
10	1	10	10	11	9	0	9	9	10	11523.8332	–0.0148
10	1	10	10	9	9	0	9	9	8	11523.8332	–0.0104
6	3	3	6	6	6	2	4	6	6	11649.6927	–0.0077
6	3	3	5	6	6	2	4	5	6	11649.9335	–0.0223
6	3	3	7	7	6	2	4	7	7	11649.9335	–0.0244
3	3	0	4	4	3	2	1	4	3	11818.9758	0.0265
3	3	0	4	3	3	2	1	4	3	11818.9758	0.0239
3	3	0	4	4	3	2	1	4	5	11818.9758	0.0180
3	3	0	4	4	3	2	1	3	3	11818.9758	0.0173
3	3	0	4	5	3	2	1	4	5	11818.9758	0.0160
3	3	0	4	3	3	2	1	3	3	11818.9758	0.0147
3	3	1	4	4	3	2	2	2	3	11838.1285	–0.0044
3	3	1	4	4	3	2	2	4	4	11838.1285	–0.0046
4	3	2	3	2	4	2	3	3	2	11848.7796	–0.0027
4	3	2	3	4	4	2	3	3	4	11848.7796	–0.0006
5	3	3	4	3	5	2	4	4	3	11869.8208	0.0048
5	3	3	4	5	5	2	4	4	5	11869.8208	0.0077
5	3	3	4	4	5	2	4	4	4	11869.8208	0.0190
6	3	4	5	6	6	2	5	5	6	11906.2567	0.0407
6	3	4	5	4	6	2	5	5	4	11906.2567	0.0372
7	3	5	6	7	7	2	6	6	7	11963.3164	0.0232
7	3	5	6	5	7	2	6	6	5	11963.3164	0.0192

4. References

1 J. Pfeiffer, C. Trost, A. Pachkovska and F. Tambornino, A Crystallographic, Spectroscopic, and Computational Investigation of Carbonyl and Oxalyl Diisothiocyanate, *Inorg. Chem.*, 2021, **60**, 10722–10728.

2 D. Schmitz, V. Alvin Shubert, T. Betz and M. Schnell, Multi-resonance effects within a single chirp in broadband rotational spectroscopy: The rapid adiabatic passage regime for benzonitrile, *J. Mol. Spectrosc.*, 2012, **280**, 77–84.

3 C. Pérez, A. Krin, A. L. Steber, J. C. López, Z. Kisiel and M. Schnell, Wetting Camphor: Multi-Isotopic Substitution Identifies the Complementary Roles of Hydrogen Bonding and Dispersive Forces, *J. Phys. Chem. Lett.*, 2016, **7**, 154–160.

4 F. Neese, Software update: The ORCA program system—Version 5.0, *WIREs Comput. Mol. Sci.*, 2022, **12**, e1606.

5 A. D. Becke, Density-functional thermochemistry. III. The role of exact exchange, *J. Chem. Phys.*, 1993, **98**, 5648–5652.

6 S. Grimme and M. Steinmetz, Effects of London dispersion correction in density functional theory on the structures of organic molecules in the gas phase, *Phys. Chem. Chem. Phys.*, 2013, **15**, 16031–16042.

7 S. Grimme, J. Antony, S. Ehrlich and H. Krieg, A consistent and accurate ab initio parametrization of density functional dispersion correction (DFT-D) for the 94 elements H-Pu, *J. Chem. Phys.*, 2010, **132**, 154104.

8 S. Grimme, S. Ehrlich and L. Goerigk, Effect of the damping function in dispersion corrected density functional theory, *J. Comput. Chem.*, 2011, **32**, 1456–1465.

9 R. A. Kendall, T. H. Dunning Jr. and R. J. Harrison, Electron affinities of the first-row atoms revisited. Systematic basis sets and wave functions, *J. Chem. Phys.*, 1992, **96**, 6796–6806.

10 C. Møller and M. S. Plesset, Note on an Approximation Treatment for Many-Electron Systems, *Phys. Rev.*, 1934, **46**, 618–622.

11 V. Ásgeirsson, B. O. Birgisson, R. Bjornsson, U. Becker, F. Neese, C. Riplinger and H. Jónsson, Nudged Elastic Band Method for Molecular Reactions Using Energy-Weighted Springs Combined with Eigenvector Following, *J. Chem. Theory Comput.*, 2021, **17**,

4929–4945.

12 S. Grimme, J. G. Brandenburg, C. Bannwarth and A. Hansen, Consistent structures and interactions by density functional theory with small atomic orbital basis sets, *J. Chem. Phys.*, 2015, **143**, 54107.

13 E. F. Pettersen, T. D. Goddard, C. C. Huang, G. S. Couch, D. M. Greenblatt, E. C. Meng and T. E. Ferrin, UCSF Chimera--a visualization system for exploratory research and analysis., *J. Comput. Chem.*, 2004, **25**, 1605–1612.