Pure Rotational Spectra of CS

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The pure rotational spectra of CS and its isotopomers $^{12}C^{33}S$, $^{12}C^{34}S$, $^{13}C^{32}S$, $^{13}C^{33}S$, $^{13}C^{34}S$ including the very rare isotopomer $^{12}C^{36}S$ were observed. The rotational spectra include ground and vibrationally excited transitions up to $v = 16$. The new measurements have been performed with the Cologne terahertz spectrometer covering the frequency region from 259 to 1075 GHz. These newly observed rotational transitions together with earlier data were fitted to a Dunham-type Hamiltonian. The obtained isotopically invariant parameters include vibrational and rotational expansion coefficients.

1. Introduction

Although carbon monosulfid, CS, possesses a closed electronic shell, and thus a $^1\Sigma$ electronic ground state, it is chemically rather unstable. In terms of chemical lifetime it displays features which are known from open shell molecules, i. e. like molecular free radicals. Since its first detection by microwave techniques in 1953 [1], CS has served as a probe-molecule for testing experimental advances, particulary associated with the generation of highly unstable free radicals [2]. We have measured the terahertz spectrum of CS for three reasons: (i) as a preparatory step in the generation of other sulfur bearing unstable species, e. g. SH, SSH, PS, and (ii) to study high vibrationally excited CS together with rare isotopomers. (iii) to enlarge the existing data base to derive isotopically invariant parameters.

Bird and Mockler [1] observed the $J = 1 \leftarrow 0$ rotational transition of $^{12}C^{32}S$ and $^{12}C^{34}S$. The following investigation by the same authors [3] in 1955 resulted in measurements with improved accuracy and, for the first time, the $J = 1 \leftarrow 0$ rotational transition in the first vibrational excited state of $^{12}C^{32}S$ was reported. In addition, they recorded the hyperfine structure of the $^{12}C^{33}S$ isotopomer, and from the observed splittings of the $J = 1 \leftarrow 0$ transition the electric quadrupole hyperfine constant $eQq(^{33}S)$ = 12.835(26) MHz was deduced. The dipole moment of carbon monosulfide in the ground and first excited vibrational state was measured by Winnewisser and Cook [4]. The splitting of the $J = 1 \leftarrow 0$ transition into three Stark components caused by an applied electric field was used in determining the dipole moment to be $\mu_D = 1.958(05)$ D for the vibrational ground state and $\mu_D = 1.936(10)$ D for the first excited vibrational state. Because of the relatively high dipole moment of CS and the low stretching vibration ($\omega = 1285.08\ \mathrm{cm}^{-1}$ [3]), it is possible to measure rotational transitions in highly excited vibrational states, particularly since CS is generated *in situ* in an electric discharge. The early millimeter wave work on the ground state by Kewley et al. [2] was extended by Bogey et al. [5] to include the isotopomers $^{12}C^{32}S$, $^{12}C^{34}S$ and $^{13}C^{32}S$. The frequency coverage was increased up to 538 GHz. Their analysis employed the reduced Dunham coefficients and mass scaling coefficients. In an earlier publication Bogey et al. [6] presented the results of measurements on the rare isotopic species $^{13}C^{34}S$ and $^{12}C^{36}S$ in natural abundance. In addition, the millimeter wave spectra of $^{12}C^{33}S$ ($v \leq 1$) and $^{13}C^{33}S$ ($v = 0$) were observed, improving the accuracy of the hyperfine constant eQq. In passing it might be noted that rotational transitions of interstellar CS have become ubiquitous tracers of high density molecular cloud regions. Interstellar CS has been detected from almost all rare isotopomers, including $C^{36}S$ [7]. The main isotopomer $^{12}C^{32}S$ has also been detected in the first excited vibrational state $v = 1$ in star forming cores [8]. CS emission has also been detected in various comets, including the rotational transition $J = 7 \leftarrow 6$ in the comet HYAKUTAKE (1996 B2) by Wouterloot et al. [9] using the

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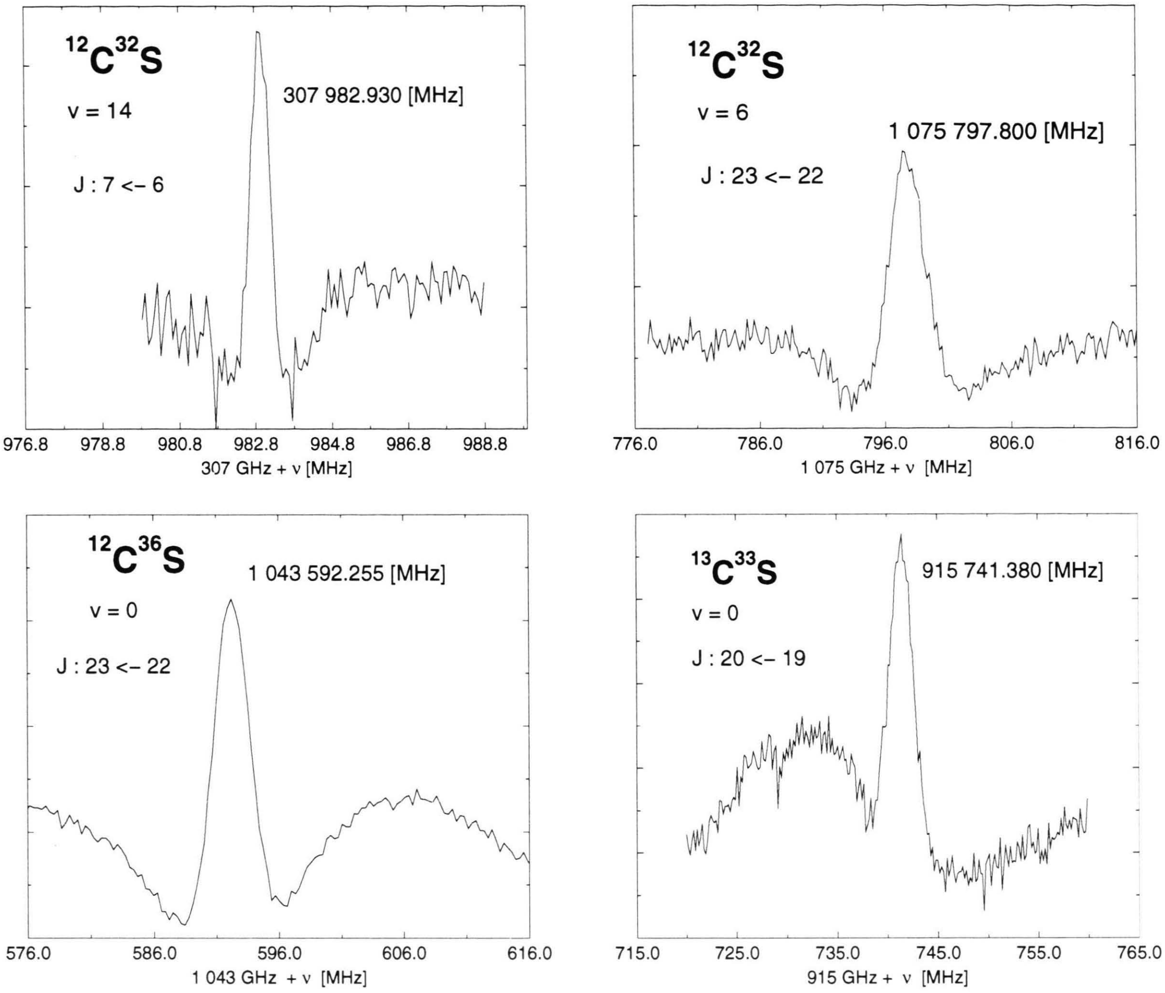

Fig. 1. Sample spectra of measured rotational transitions of the CS molecule.

KOSMA-3m-radiotelescope. Now we are able to extend the existing laboratory CS measurements into the terahertz region including transitions as high as $J = 23 \leftarrow 22$, and for the isotopically more common species towards highly excited vibrational states up to $v = 16$. Some samples spectra are shown in Figure 1.

2. Experimental Details

For more detailed descriptions of the Cologne submillimeter wave spectrometer system, the reader is referred to some recent publications [10], [11]. Several backward wave oscillators (BWO) were used as microwave radiation sources covering a total frequency range from 250 to 1100 GHz. The phase-locked radiation of the various BWO's was focused through the absorption cell and subsequently detected with a He-cooled InSb hot electron bolometer. The experimental uncertainties were estimated from the obtained signal-to-noise ratios. For strong, well-isolated lines the uncertainity in the frequency determination is estimated to be 10 kHz or less, whereas for weak, blended lines, e. g. for transitions in high excited vibrational states, the uncertainty can be as high as 500 kHz. The CS molecule was produced in a dc discharge of carbon disulfide (CS_2) and argon. For establishing the optimum discharge conditions, the partial pressure of argon was stabilized at 80 μbar, while less than 5 μbar of CS_2 was used. The optimum discharge current was about 335 mA. As a result, the colour of the discharge was a light grey. It was not necessary to cool the absorption cell during the measurements.

Table 1. Isotopically Invariant Parameters of CS.

Parameter	This work	Bogey et al. (1982)	Unit
U^e_{01}	214502.750(18)	214502.646(19)	MHz amu
U^e_{02}	−3.0597(20)	−3.0496[a]	MHz amu^2
U^e_{11}	−4572.945(46)	−4572.712(54)	MHz amu$^{3/2}$
U^e_{21}	−2.126(45)	−2.314(42)	MHz amu^2
U^e_{12}	−0.0046(22)	−0.009[b]	MHz amu$^{5/2}$
U^e_{22}	−0.00145(46)	—	MHz amu^3
U^e_{31}	−0.138(21)	−0.067(11)	MHz amu$^{5/2}$
U^e_{41}	−0.0113(44)	−0.02330(92)	MHz amu^3
Δ^C_{01}	−2.5055(53)	−2.586(25)	MHz amu$^{1/2}$
Δ^S_{01}	−2.342(14)	−2.525(64)	MHz amu$^{1/2}$
eq_{00}	−1.9807(12)[c]	−2.0052(56)[c]	MHz
$C_{00}(^{33}S)$	0.751(71)[c]	1.1385(32)[c]	MHz

[a] Calculated from $U^e_{02} = -4U^3_{01}/U^2_{10}$, (Bogey et al. [5]); [b] calculated from $U^e_{12} = (192U_{20}U^3_{01} + U^2_{11}U^2_{10} + 120U_{11}U_{10}U^2_{01})/6U^3_{10}$, (Bogey et al. [5]); [c] constant determined in a global fit together with data obtained by Bogey et al. [6].

3. Analysis

The fundamental relation between energy levels of a vibrating rotor can be written in the form

$$E(v, J) = \sum_{i,j} Y_{i,j}(v + \frac{1}{2})^i [J(J + 1)]^j, \qquad (1)$$

where i and j are summation indicies and the Y_{ij} are observable constants known as Dunham coefficients [12] and [13]. For two reasons it is very advantageous to use the Dunham expansion method. (i) the number of constants that have to be fitted for obtaining predictions of transition frequencies for different isotopomeric species can be reduced via the introduction of the isotopic invariant parameters U_{ij}. Because of the relation (Watson [14], Bunker [15])

$$Y_{ij} = U_{ij}\mu^{-(\frac{i}{2}+j)}[1 + \frac{m_e}{M_A}\Delta^A_{ij} + \frac{m_e}{M_B}\Delta^B_{ij} + \mathcal{O}(\frac{m_e^2}{M^2_{A,B}})] \qquad (2)$$

it is not necessary to calculate rotational constants for each isotopomer but it is possible to evaluate data from each isotopomer individually in only one fitting procedure. In (2) μ is the reduced mass of the molecule, Δ^A_{ij} and Δ^B_{ij} are correction coefficients which are important if the breakdown of Born-Oppenheimer approximation is taken into account. The correction term $\mathcal{O}(m_e^2/M^2_{A,B})$ is too small to be experimentally determined. (ii) the second advantage stems from the relation between Dunham coefficients of different isotopomers Y^ρ_{ij} and Y^σ_{ij}:

$$Y^\rho_{ij} = Y^\sigma_{ij}(\frac{\mu_\rho}{\mu_\sigma})^{-(\frac{i}{2}+j)}. \qquad (3)$$

From the knowledge of the reduced mass, e.g. of a rare isotopomer, the corresponding Dunham coefficient can be determined with the aid of (3). For the breakdown of the Born-Oppenheimer approximation the U_{ij} also have to be corrected. The correction for the Dunham coefficient U_{01} was worked out by Tiemann [16] :

$$U_{01} = U^e_{01} + U^A_{01}(1 - \frac{M^0_A}{M_A}) + U^B_{01}(1 - \frac{M^0_B}{M_B}). \qquad (4)$$

In (4) U^e_{01} is the rotational constant in equilibrium, M^0_A and M^0_B are the atomic masses of the reference istopomer (here $^{12}C^{32}S$). The U^A_{01}, U^B_{01} are related to the parameters Δ^A_{ij} and Δ^B_{ij} by the expression

$$\Delta^{A,B}_{01} = -\frac{M^0_{A,B}}{m_e U_{01}} U^{A,B}_{01}. \qquad (5)$$

The Dunham coefficients U_{ij} and the mass scaling coefficients $\Delta^{C,S}_{ij}$ are presented and compared with molecular parameters calculated by Bogey et al. [5] in Table 1.

The hyperfine constants C_I and eQq had been determined from a global fit with help of the program using the isotopic invariant formulation of the Dunham coefficients written by Saleck and Klaus. Ryzlewicz et al. [17] introduced a Dunham-type expansion of the nuclear electric quadrupole moment

$$eQq = Q\sum_{ij} eq_{ij}\mu^{-\frac{i}{2}-j}(v + \frac{1}{2})^i J^j(J + 1)^j \qquad (6)$$

in order to make use of the advantages of Dunham-type series mentioned above. For the same reason the interaction between nuclear spin and rotation as described by the nuclear magnetic coupling parameter C_I can formally be treated by a Dunham-type expansion, as has been introduced by Saleck et al. [18] for the HBr molecule and its isotopomers. Klaus et al. [19] have applied this expansion for fitting the rotational spectra of the SO molecule [19] and those of HCl and DCl [20]. We have made use of it in fitting the complete CS data set

$$C_I = \frac{\mu_I}{\mu_N I}\sum_{i,j} C_{ij}\mu^{-\frac{i}{2}-j-1}(v + \frac{1}{2})^i J^j(J + 1)^j. \qquad (7)$$

Table 2. Derived molecular parameters for CS in MHz (μ = reduced mass).

Isotopomer	μ	B_e	D_e	α_e	$\beta_e \cdot 10^{-5}$	γ_e
$^{12}C^{32}S$	8.72519	24584.3070(20)	–0.040191(26)	–177.4326(17)	–2.04(97)	–0.02792(59)
$^{12}C^{34}S$	8.86738	24190.0933(20)	–0.038912(25)	–173.1820(17)	–1.96(93)	–0.02703(57)
$^{13}C^{32}S$	9.424381	22760.4097(19)	–0.034448(22)	–158.0581(16)	–1.68(80)	–0.02393(50)
$^{12}C^{33}S$	8.79797	24380.9367(20)	–0.039528(26)	–175.2355(16)	–2.00(86)	–0.02746(58)
$^{13}C^{33}S$	9.32553	23001.6685(19)	–0.035182(26)	–160.5778(16)	–1.73(82)	–0.02445(51)
$^{13}C^{33}S$	9.40355	22810.8267(19)	–0.034601(26)	–158.5835(15)	–1.69(81)	–0.02404(51)
$^{12}C^{36}S$	8.99794	23839.0953(20)	–0.037791(25)	–169.4264(17)	–1.89(91)	–0.02625(55)

Table 3. Observed rotational frequencies for $^{12}C^{32}S$ and $^{12}C^{34}S$; [a] previous measurements by Bogey et al. [5].

			— $^{12}C^{32}S$ —		— $^{12}C^{34}S$ —	
J'	J''	v	ν [MHz]	o-c [kHz]	ν [MHz]	o-c [kHz]
6	5	0	293 912.244(80)[a]	81	289 209.230(80)[a]	102
6	5	1	291 782.294(80)[a]	42	287 130.314(120)[a]	70
6	5	2	289 651.693(80)[a]	99	285 050.631(200)[a]	07
6	5	3	287 520.206(150)[a]	71	282 970.285(200)	26
6	5	4	285 388.071(200)[a]	260	280 889.065(80)	20
6	5	5	283 254.857(200)[a]	30	–	
6	5	6	281 120.322(30)	20	–	–
6	5	7	278 984.961(50)	08	–	–
6	5	8	276 848.413(50)	11	–	–
6	5	9	274 710.590(50)	19	–	–
6	5	10	272 571.398(80)	07	–	–
6	5	11	270 430.679(50)	13	–	–
6	5	12	268 288.335(80)	03	–	–
6	5	13	266 144.209(80)	08	–	–
6	5	14	263 998.128(100)	01	–	–
6	5	15	261 850.009(100)	53	–	–
6	5	16	259 699.481(500)	27	–	–
7	6	0	342 883.600(200)[a]	85	–	–
7	6	1	340 398.080(200)[a]	74	–	–
7	6	4	–	–	327 698.645(80)	42
7	6	5	–	–	325 260.475(100)	63
7	6	6	–	–	322 830.205(200)	50
7	6	7	325 467.675(50)	42	320 398.750(200)	04
7	6	8	322 975.020(50)	55	317 965.795(200)	111
7	6	9	320 895.895(50)	42	–	–
7	6	10	317 985.120(50)	53	–	–
7	6	11	315 487.600(50)	44	–	–
7	6	12	312 988.180(50)	19	–	–
7	6	13	310 486.660(50)	12	–	–
7	6	14	307 982.930(50)	44	–	–
7	6	15	305 476.670(80)	23	–	–
7	6	16	302 967.825(100)	76	–	–
12	11	0	587 616.485(20)	19	578 217.069(20)	28
12	11	1	583 356.495(20	07	574 059.122(30)	33
12	11	2	579 094.972(20)	13	569 899.705(20)	31
12	11	3	574 831.822(20)	14	–	–
13	12	0	636 532.460(30)	16	626 351.384(20)	03
13	12	1	631 917.410(30)	05	621 846.900(20)	19
13	12	2	627 300.695(30)	04	617 340.853(20)	60
13	12	3	622 682.230(30)	15	612 832.820(20)	180
13	12	4	618 061.868(30)	26	608 323.260(20)	117
13	12	5	613 439.475(30)	51	603 811.950(30)	169
14	13	1	–	–	669 622.570(50)	04
14	13	2	–	–	664 769.770(50)	20
14	13	3	670 520.100(50)	13	659 915.400(200)	223
14	13	4	665 544.240(50)	15	655 058.665(200)	79
14	13	5	660 566.170(100)	15	650 199.950(200)	86
19	18	0	929 732.125(80)	41	914 870.705(50)	01
19	18	1	922 986.485(30)	40	908 286.590(30)	22
19	18	2	916 238.255(50)	36	901 700.160(30)	10
19	18	3	909 487.545(30)	13	895 111.105(60)	102
19	18	4	902 733.975(50)	10	–	–
19	18	5	895 977.415(50)	41	–	–
20	19	0	–	–	962 900.785(50)	06
20	19	1	971 439.540(50)	08	955 970.050(80)	29
20	19	2	964 336.105(80)	20	949 036.760(50)	24
20	19	3	957 229.885(50)	13	942 100.855(50)	37
20	19	4	950 120.740(80)	38	935 162.085(80)	58
20	19	5	943 008.345(50)	02	928 220.365(100)	48
20	19	6	935 892.530(50)	11	921 275.095(500)	72
20	19	7	928 773.030(80)	15	–	–
20	19	8	921 649.450(50)	04	–	–
20	19	9	914 521.540(100)	18	900 416.885(50)	84
21	20	0	–	–	1 010 912.265(50)	17
21	20	1	1 019 873.435(80)	09	1 003 634.835(50)	09
21	20	2	1 012 414.650(50)	15	996 354.750(50)	14
21	20	3	1 004 952.895(80)	40	989 071.790(500)	120
21	20	4	–	–	981 786.075(100)	20
21	20	6	982 548.215(80)	22	967 204	
21	20	7	975 072.460(80)	39	959 908	
21	20	9	960 108.075(100)	39	–	–
21	20	10	952 618.536(50)	15	–	–
22	21	0	1 076 098.525(200)	98	1 058 904.175(50)	182
22	21	1	1 068 287.100(50)	138	–	–
22	21	2	1 060 473.065(80)	31	1 043 653.215(50)	08
22	21	3	1 052 655.795(50)	05	1 036 023.355(100)	10
22	21	4	1 044 835.375(50)	65	–	–
22	21	5	1 037 011.375(100)	69	–	–
22	21	6	1 029 183.570(80)	62	1 013 113.935(200)	29
22	21	7	1 021 351.625(80)	21	–	–
22	21	8	1 013 515.275(100)	20	–	–
22	21	9	1 005 674.075(150)	09	–	–
23	22	6	1 075 797.800(500)	266	–	–
23	22	8	1 059 416.690(500)	160	–	–

Table 4. Observed rotational frequencies for $^{12}C^{33}S$ and $^{12}C^{36}S$.

			— $^{12}C^{33}S$ —		— $^{12}C^{36}S$ —	
J'	J''	v	ν [MHz]	o-c [kHz]	ν [MHz]	o-c [kHz]
6	5	0	291 485.935(30)	86	285 021.055(50)	53
6	5	1	289 382.425(30)	70	–	–
12	11	0	582 767.500(70)	94	–	–
13	12	0	631 280.360(120)	12	617 285.160(100)	90
19	18	0	922 065.565(30)	44	901 636.275(60)	21
19	18	1	915 403.465(50)	32	–	–
19	18	2	908 738.935(70)	68	–	–
20	19	0	970 472.470(50)	22	948 973.240(80)	20
20	19	1	963 459.465(100)	25	–	–
20	19	2	956 444.090(100)	73	–	–
21	20	0	1 018 860.405(50)	20	996 292.305(50)	77
21	20	1	1 011 496.695(80)	39	–	–
21	20	2	1 004 130.300(50)	61	–	–
22	21	0	1 067 228.500(80)	296	1 043 592.255(500)	57
22	21	1	1 059 514.030(100)	11	1 036 133.250(500)	254
22	21	2	1 051 796.315(200)	300	–	–

However, since for CS only the low J rotational lines contain information on the electric quadrupole and the nuclear spin-rotation interaction, only the first term of each expansion is of significance and quoted in Table 1.

4. Results and Discussion

The molecular constants in Table 1 were fitted simultaneously with the data published by Bogey et al. [5, 6] covering for all observed isotopomers 216 rotational transitions. Table 2 presents the calculated equilibrium molecular parameters, B_e, D_e, α_e, β_e, γ_e according to the relations

$$\begin{aligned} B_v &= B_e - \alpha_e(v + \frac{1}{2}) + \gamma_e(v + \frac{1}{2})^2 + \ldots, \\ D_v &= D_e + \beta_e(v + \frac{1}{2}) + \ldots. \end{aligned}$$

The α_e and γ_e constants are rotation-vibration interaction constants, whereas β_e is a vibration correction on the centrifugal distortion constant. Our new measurements extend the existing data base by 182 new lines positioned in the frequency region between 259 and 1075 GHz. In detail we added the following data: 67 lines for $^{12}C^{32}S$, 44 lines for $^{12}C^{34}S$, 28 lines for $^{13}C^{32}S$, 16 lines for $^{12}C^{33}S$, 14 lines for $^{13}C^{34}S$, 8 lines for $^{12}C^{36}S$, and 5 lines for $^{13}C^{33}S$. These new data sets are summarized in Tables 3 - 6. Especially rotational transitions in highly excited vibrational states could be measured, e. g. in case of $^{12}C^{32}S$ up to v = 16. Hence the Dunham constants with higher indices corresponding to high vibrational

Table 5. Observed rotational frequencies for $^{13}C^{32}S$ and $^{13}C^{34}S$.

			— $^{13}C^{32}S$ —		— $^{13}C^{34}S$ —	
J'	J''	v	ν [MHz]	o-c [kHz]	ν [MHz]	o-c [kHz]
6	5	0	277 455.405(30)	23	–	–
6	5	1	275 502.230(30)	13	–	–
6	5	2	273 548.395(50)	01	270 847.890(500)	22
7	6	0	323 684.995(30)	09	–	–
7	6	1	321 406.250(50)	10	–	–
7	6	2	319 126.740(50)	20	–	–
7	6	3	316 846.320(50)	110	–	–
13	12	0	600 906.480(30)	05	590 723.060(30)	96
13	12	1	596 674.330(100)	30	586 598.370(100)	135
13	12	2	592 440.510(100)	267	582 471.950(100)	149
13	12	3	588 205.592(100)	41	–	–
14	13	0	647 076.150(50)	42	636 111.130(30)	72
14	13	1	642 518.455(200)	75	631 669.140(200)	150
14	13	2	637 958.925(200)	120	–	–
14	13	3	633 397.980(200)	79	–	–
20	19	0	923 812.235(50)	58	908 167.305(50)	32
20	19	1	917 300.390(60)	73	901 820.760(80)	128
20	19	2	910 786.485(100)	82	–	–
20	19	3	904 269.965(60)	87	–	–
21	20	0	969 879.720(200)	66	953 456.985(80)	117
21	20	1	963 042.375(80)	84	946 793.430(500)	200
21	20	3	949 360.015(200)	209	–	–
21	20	5	935 666.850(200)	30	–	–
22	21	0	1 015 929.415(50)	23	–	–
22	21	1	–	–	991 747.875(100)	51
22	21	5	980 086.750(500)	151	–	
23	22	0	1 061 960.275(500)	268	1 043 983.155(200)	200
23	22	1	1 054 471.425(100)	187	1 036 684.350(900)	151
23	22	2	1 046 979.895(500)	167	–	–
23	22	3	1 039 485.365(200)	341	–	–
23	22	4	1 031 987.950(200)	388	–	–
23	22	5	1 024 487.455(500)	274	–	–

Table 6. Observed rotational frequencies for $^{13}C^{33}S$.

J'	J''	v	ν [MHz]	o-c [kHz]
6	5	0	275028 .795(300)	07
7	6	0	320 854 .215(800)	48
13	12	0	595 653 .140(100)	61
14	13	0	641 419 .630(140)	128
20	19	0	915 741 .380(80)	08

and rotational influence had to be fitted. It is remarkable that only the Dunham coefficient U_{22} had to be added to the set of parameters used by Bogey et al. [5, 6] in order to reproduce the complete data set, i. e. including the newly observed spectra. As can be seen from the comparison of the Dunham constants obtained from our extended data set quoted in Table 1 and the ones derived by Bogey et al. [5] reproduced in Table 1 the two sets are in very good agreement.

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