

Table 1: Experimental conditions of the recorded spectra.

#	$P_{\text{CH}_3\text{I}}$ (in mbar)	Temperature (in K)	Number of scans	Bruker resolution (in cm^{-1})
1	0.980	293.8	366	0.002
2	1.865	293.7	416	0.002
3	4.918	293.2	760	0.004
4	9.134	293.8	700	0.004

Table 2: Sample of measured line parameters. The column ‘Assignment’ refers firstly to the type of sub-branch ($\Delta K, \Delta J$) and then for the upper and lower states to the rotational quantum numbers (J and K) as well as to the symmetry level. A star has been added at the end of ‘Assignment’ field when the observed transition corresponds to one or several hyperfine components (see text for details). The line positions σ are given in cm^{-1} , the line intensities S are for 296 K in $\text{cm}^{-1} (\text{molecule cm}^{-2})^{-1}$ and for natural abundance of $^{12}\text{CH}_3\text{I}$, the transition dipole moments squared noted R^2 are in debye^2 , and the self-broadening coefficients are given in $\text{cm}^{-1} \text{atm}^{-1}$ for a temperature close to 293-294 K. The columns $\Delta\sigma$ (in cm^{-1}), ΔS (in %) and $\Delta\gamma$ (in $\text{cm}^{-1} \text{atm}^{-1}$) correspond to the statistical RMS of the fit of the positions, line intensities and the self-broadening coefficients respectively. The whole set of measurements is available as supplementary material.

Branch type	Assignment						σ	$\Delta\sigma$	S	ΔS	R^2	γ_{self}	$\Delta\gamma$	E''
	Upper state	Lower state												
RQ	38	3	E	38	2	E	900.114678	0.000013	0.611E-21	0.6	0.3963E-02	0.2897	0.0032	389.53282
RR	14	2	E	13	1	E	900.286667	0.000028	0.830E-21	1.2	0.4633E-02	0.4468	0.0054	50.44001
RQ	34	3	E	34	2	E	900.344436	0.000016	0.795E-21	0.7	0.4043E-02	0.3269	0.0042	316.73621
RP	31	5	E	32	4	E	900.381812	0.000025	0.263E-21	1.2	0.4385E-02	0.3639	0.0053	341.27450
RQ	32	3	E	32	2	E	900.449843	0.000016	0.928E-21	0.6	0.4264E-02	0.3657	0.0033	283.31751
RP	16	4	A2	17	3	A	*	900.461297	0.000157			0.4711	0.0228	120.61216
PR	49	0	E	48	1	E	900.482280	0.000046	0.152E-21	2.2	0.4221E-02	0.2293	0.0083	592.05996
RQ	31	3	E	31	2	E	900.500130	0.000016	0.928E-21	0.7	0.4071E-02	0.3541	0.0041	267.35367
RQ	30	3	E	30	2	E	900.548875	0.000015	0.976E-21	0.6	0.4102E-02	0.3662	0.0032	251.88705
RP	45	6	E	46	5	E	900.615777	0.000040	0.845E-22	2.6	0.4353E-02	0.2326	0.0080	658.29976
RQ	28	3	E	28	2	E	900.641703	0.000019	0.908E-21	1.7	0.3541E-02	0.3400	0.0046	222.44611
RP	29	5	E	30	4	E	901.473531	0.000021	0.286E-21	0.9	0.4455E-02	0.3872	0.0050	309.97609
RP	14	4	A2	15	3	A	*	901.509461	0.000058			0.4097	0.0090	104.15739
RP	14	4	A2	15	3	A	*	901.511053	0.000035			0.3609	0.0052	104.15739
RR	17	2	E	16	1	E	901.710895	0.000014	0.659E-21	0.7	0.3489E-02	0.3809	0.0024	72.94296
PR	52	0	E	51	1	E	901.720827	0.000026	0.102E-21	1.3	0.3804E-02	0.1888	0.0051	666.78194
RP	57	7	A1	58	6	A	901.815729	0.000038	0.335E-22	2.4	0.3898E-02	0.1576	0.0058	1020.15235
RR	18	2	E	17	1	E	902.182244	0.000010	0.692E-21	0.5	0.3640E-02	0.3849	0.0021	81.44318
RP	56	7	A1	57	6	A	902.397760	0.000032	0.445E-22	1.9	0.4611E-02	0.1537	0.0049	991.65665
RR	36	1	A1	35	0	A2	902.483967	0.000013	0.847E-21	0.6	0.3950E-02	0.3413	0.0030	314.93742
RP	27	5	E	28	4	E	902.559500	0.000019	0.281E-21	0.8	0.4166E-02	0.3763	0.0037	280.65881
RR	19	2	E	18	1	E	902.651926	0.000016	0.812E-21	0.5	0.4259E-02	0.4506	0.0034	90.44289
RR	37	1	A2	36	0	A1	902.915364	0.000015	0.765E-21	0.6	0.3789E-02	0.3176	0.0034	332.91364
RP	55	7	A1	56	6	A	902.978612	0.000034	0.438E-22	2.1	0.4040E-02	0.1643	0.0062	963.64671
RP	11	4	A2	12	3	A	*	903.072483	0.000068			0.3721	0.0137	83.21230
RR	20	2	E	19	1	E	903.119967	0.000018	0.776E-21	0.6	0.4075E-02	0.4289	0.0042	99.94199
RP	54	7	A1	55	6	A	903.557973	0.000038	0.511E-22	2.2	0.4218E-02	0.1852	0.0068	936.12282
RP	10	4	A2	11	3	A	*	903.586610	0.000065			0.3395	0.0041	77.22745
RP	10	4	A2	11	3	A	*	903.590196	0.000057			0.2949	0.0074	77.22745
RP	25	5	E	26	4	E	903.639688	0.000020	0.294E-21	0.8	0.4228E-02	0.4040	0.0043	253.32386
PR	57	0	E	56	1	E	903.746982	0.000060	0.568E-22	3.3	0.3720E-02	0.1626	0.0115	801.18566
RP	39	6	E	40	5	E	903.991142	0.000034	0.132E-21	1.6	0.4344E-02	0.2967	0.0066	529.24506

Table 3: Statistics of comparisons for line positions (σ), line intensities (S), and self-broadening coefficients (γ).

Comparisons with literature [12,13]		
	Number of comparisons	Average (SD)
$\sigma_{calc [12]} - \sigma_{obs \text{ This work}}$ (in 10^{-3} cm^{-1})	989	0.80(20)
$\sigma_{calc [13]} - \sigma_{obs \text{ This work}}$ (in 10^{-3} cm^{-1})	402	0.13(6)
$S_{obs [13]} / S_{obs \text{ This work}}$	402	0.98(11)
Comparisons of measurements, model and calculations from this work		
$\sigma_{calc \text{ This work}} - \sigma_{obs \text{ This work}}^{\dagger}$ (in 10^{-3} cm^{-1})	989	0.00(20)
$S_{calc \text{ This work}} / S_{obs \text{ This work}}$	958	1.004(70)
$\gamma_{emp \text{ This work}} / \gamma_{obs \text{ This work}}$	1225	1.001(74)
$\gamma_{SC \text{ This work}} / \gamma_{obs \text{ This work}}$	390 ($^R R$)	1.14(9)
$\gamma_{SE \text{ This work}} / \gamma_{obs \text{ This work}}$	390 ($^R R$)	1.02(9)

[†] Line positions $\sigma_{calc \text{ This work}}$ are the one published in Ref. [12] corrected by $-0.8 \times 10^{-3} \text{ cm}^{-1}$ (see text). Since Ref. [12] does not include hyperfine structure, the comparison includes only 989 common transitions for which no hyperfine structure is experimentally measured (excluding transitions noted with a star in Table 2, see text).

Table 4: Parameters $a_{J \text{ exp}}^0$ (in $\text{cm}^{-1} \text{atm}^{-1}$) and $a_{J \text{ exp}}^2$ (in $10^{-3} \text{cm}^{-1} \text{atm}^{-1}$) retrieved for each set of self-broadening coefficients with same value of J . N is the number of measured self-broadening coefficients used to fit the $a_{J \text{ exp}}^0$ and $a_{J \text{ exp}}^2$ parameters. $a_{J \text{ smooth}}^0$ and $a_{J \text{ smooth}}^2$ are smoothed or extrapolated values of the fitted $a_{J \text{ exp}}^0$ and $a_{J \text{ exp}}^2$ parameters. Between parentheses are 1SD for the $a_{J \text{ exp}}^0$ and $a_{J \text{ exp}}^2$ coefficients retrieved using Eq. (5). The 1SD is expressed in the last unit of the digit: 0.4052(134) meaning 0.4052 ± 0.0134 .

J	$a_{J \text{ exp}}^0$	$a_{J \text{ exp}}^2$	N	$a_{J \text{ smooth}}^0$	$a_{J \text{ smooth}}^2$
0	0.493 (7)	--	3	0.520	--
1	0.503 (8)	--	2	0.466	-70E-3
2	0.4213 (78)	--	3	0.424	-20E-3
3	0.3837 (434)	--	3	0.392	-13E-3
4	0.4052 (134)	-1.06E-2 (23)	8	0.370	-9.70E-3
5	0.3465 (104)	-6.05E-3 (60)	8	0.355	-7.50E-3
6	0.3099 (102)	-3.13E-3 (98)	8	0.346	-5.80E-3
7	0.3352 (75)	-5.76E-3 (75)	12	0.343	-4.50E-3
8	0.3450 (119)	-2.47E-3 (57)	13	0.344	-3.50E-3
9	0.3519 (71)	-3.35E-3 (59)	15	0.348	-2.70E-3
10	0.3728 (97)	-2.61E-3 (96)	16	0.354	-2.15E-3
11	0.3489 (96)	-1.48E-3 (36)	29	0.362	-1.80E-3
12	0.3716 (104)	-1.32E-3 (48)	21	0.371	-1.50E-3
13	0.3886 (74)	-1.05E-3 (15)	27	0.380	-1.20E-3
14	0.3933 (126)	-7.88E-4 (385)	19	0.389	-1.00E-3
15	0.3843 (80)	-1.55E-3 (33)	16	0.397	-8.20E-4
16	0.4174 (86)	-1.03E-3 (21)	25	0.404	-6.80E-4
17	0.4126 (91)	-8.15E-4 (239)	29	0.410	-5.70E-4
18	0.4170 (86)	-8.77E-4 (376)	23	0.415	-4.80E-4
19	0.4134 (61)	-4.80E-4 (185)	21	0.417	-3.90E-4
20	0.4198 (57)	-9.66E-4 (227)	19	0.418	-3.30E-4
21	0.4143 (87)	-9.32E-4 (559)	21	0.418	-3.10E-4
22	0.4063 (56)	-3.21E-4 (119)	26	0.416	-2.98E-4
23	0.4147 (48)	-3.20E-4 (123)	28	0.413	-2.95E-4
24	0.4168 (41)	-2.87E-4 (102)	26	0.410	-2.94E-4
25	0.4098 (51)	-3.05E-4 (162)	24	0.406	-2.93E-4
26	0.3999 (46)	-8.95E-5 (895)	32	0.402	-2.91E-4
27	0.3993 (35)	-3.96E-4 (107)	26	0.399	-2.90E-4
28	0.3841 (63)	-3.84E-4 (173)	36	0.391	-2.89E-4
29	0.3843 (53)	-8.44E-4 (390)	21	0.388	-2.88E-4
30	0.3901 (50)	-3.03E-4 (110)	23	0.380	-2.86E-4
31	0.3731 (54)	-3.13E-4 (127)	22	0.369	-2.85E-4
32	0.3633 (40)	-2.99E-4 (135)	23	0.359	-2.84E-4
33	0.3510 (53)	-3.63E-4 (207)	26	0.348	-2.83E-4
34	0.3394 (64)	-4.16E-4 (272)	26	0.338	-2.82E-4
35	0.3304 (35)	-3.10E-4 (88)	21	0.328	-2.80E-4
36	0.3214 (62)	-4.83E-4 (326)	25	0.319	-2.79E-4
37	0.3162 (46)	-3.07E-4 (99)	25	0.309	-2.78E-4
38	0.3012 (26)	-2.48E-4 (59)	23	0.300	-2.77E-4
39	0.2913 (27)	-2.30E-4 (74)	29	0.291	-2.76E-4
40	0.2879 (42)	-2.35E-4 (88)	26	0.282	-2.74E-4
41	0.2811 (41)	-2.87E-4 (103)	28	0.274	-2.73E-4
42	0.2694 (38)	-4.06E-4 (161)	20	0.265	-2.72E-4
43	0.2576 (41)	-2.17E-4 (77)	24	0.257	-2.71E-4
44	0.2475 (37)	-3.50E-4 (147)	15	0.249	-2.69E-4

45	0.2391 (26)	-1.67E-4 (46)	23	0.241	-2.68E-4
46	0.2296 (30)	-1.81E-4 (57)	28	0.234	-2.67E-4
47	0.2232 (32)	-2.37E-4 (65)	29	0.227	-2.66E-4
48	0.2127 (30)	-1.78E-4 (85)	16	0.220	-2.65E-4
49	0.2106 (49)	-1.00E-4 (84)	19	0.213	-2.63E-4
50	0.1976 (40)	-1.11E-4 (88)	22	0.206	-2.62E-4
51	0.1950 (24)	-1.00E-4 (54)	21	0.200	-2.61E-4
52	0.1911 (48)	-1.14E-4 (94)	18	0.193	-2.60E-4
53	0.1879 (61)	-2.23E-4 (356)	13	0.187	-2.59E-4
54	0.1842 (28)	-2.54E-4 (201)	14	0.182	-2.57E-4
55	0.1807 (67)	-2.92E-4 (183)	14	0.176	-2.56E-4
56	0.1669 (38)	-2.37E-4 (97)	16	0.171	-2.55E-4
57	0.1603 (36)	-4.44E-5 (916)	18	0.166	-2.54E-4
58	0.1582 (94)	-1.43E-4 (387)	5	0.161	-2.52E-4
59	0.1566 (75)	--	6	0.156	-2.51E-4
60	0.1547 (58)	-7.37E-5 (3749)	9	0.152	-2.50E-4
61	0.1498 (43)	-2.69E-4 (105)	10	0.148	-2.49E-4
62	0.1482 (28)	-4.47E-4 (156)	9	0.144	-2.48E-4
63	0.1513 (83)		7	0.140	-2.46E-4
64	0.1379 (53)		2	0.136	-2.45E-4
65	0.1382 (--)		1	0.133	-2.44E-4
66	0.1243 (--)		1	0.130	-2.41E-4
67	0.1298 (76)		4	0.127	-2.40E-4
68	0.1165 (81)		2	0.124	-2.39E-4
69	0.1215 (--)		1	0.122	-2.38E-4
70				0.119	-2.37E-4
71	--			0.117	-2.35E-4
72	--			0.115	-2.34E-4
73	--			0.114	-2.33E-4
74	--			0.112	-2.32E-4
75	--			0.111	-2.31E-4
76	--			0.110	-2.29E-4
77	--			0.1095	-2.28E-4
78	--			0.109	-2.27E-4
79	--			0.1085	-2.26E-4
80	--			0.108	-2.25E-4

Table 5: Rotational constants used in semi-classical computations (data of Ref. [11] converted to cm^{-1}).

Rotational constants	Ground state	$\nu_6 = 1$ vibrational state
A (cm^{-1})	5.17394986	5.20864873
B (cm^{-1})	0.25021630	0.24942867

Table 6: Semi-empirical fitting parameters with 1SD-uncerainties deduced from experimental values for $0 \leq K \leq 10$ (Q -branch) and $0 \leq K \leq 7$ (P - and R -branches) and extrapolated, on the basis of experimental trends, for higher K .

	Q -branch	R -branch	P -branch	P -, Q -, R -branches
K	c_1	c_1	c_1	c_2
0	10.5(1)	8.0(2)	9.0(2)	1.2(2)
1	3.5(1)	3.5(2)	2.2(2)	0.4(1) ¹
2	1.6(1)	1.6(1)	1.6(1)	0.10(5)
3	1.8(2)	1.0(2)	1.55(17)	0.095(8) ²
4	1.50(18)	1.50(17)	1.50(18)	0.09(7)
5	1.45(19)	1.45(18)	1.45(23)	0.085(5)
6	1.4(14)	1.4(12)	1.25(27)	0.08(3)
7	1.05(16)	1.35(13)	1.24(12)	0.075(3)
8	1.28(13)	1.28(8)	1.22(14)	0.07(5)
9	1.22(13)	1.22(17)	1.20(14)	0.065(6)
10-20	1.76 - 0.06 K (13)		0.110 - 0.005 K (7)	

¹ $c_2=0.2(2)$ for P -branch

² $c_2=0.0(2)$ for R -branch

Table 7: Sample of line list in HITRAN format (see Ref. [51] for details). The whole line list is available as supplementary material. Note that instead of Einstein-A coefficients, transition dipole moments squared R^2 are given in debye².

501	910.0132	0.783E-21	0.397E-02	0.09840.408	96.13250.75-	.010000	nu6	Ground 18	3	E	17	2	E	455522	1	1	1	1	1	1
501	910.0492	0.202E-21	0.408E-02	0.08820.381	338.66070.75-	.010000	nu6	Ground 28	6	E	29	5	E	455522	1	1	1	1	1	1
501	910.3388	0.160E-21	0.388E-02	0.07960.182	741.28150.75-	.010000	nu6	Ground 55	1	A2	54	0	A1	455522	1	1	1	1	1	1
501	910.3780	0.438E-21	0.392E-02	0.08690.328	319.75060.75-	.010000	nu6	Ground 36	2	E	35	1	E	455522	1	1	1	1	1	1
501	910.4057	0.753E-22	0.411E-02	0.08310.247	643.82660.75-	.010000	nu6	Ground 42	7	A2	43	6	A1	455522	1	1	1	1	1	1
501	910.4057	0.753E-22	0.411E-02	0.08310.247	643.82660.75-	.010000	nu6	Ground 42	7	A1	43	6	A2	455522	1	1	1	1	1	1
501	910.4829	0.781E-21	0.397E-02	0.09700.413	105.12270.75-	.010000	nu6	Ground 19	3	E	18	2	E	455522	1	1	1	1	1	1
501	910.5191	0.165E-21	0.404E-02	0.10270.361	124.03420.75-	.010000	nu6	Ground 12	5	E	13	4	E	455522	1	1	1	1	1	1
501	910.5571	0.145E-22	0.414E-02	0.07870.154	1051.76830.75-	.010000	nu6	Ground 56	8	E	57	7	E	455522	1	1	1	1	1	1
501	910.5915	0.206E-21	0.408E-02	0.08850.384	324.29580.75-	.010000	nu6	Ground 27	6	E	28	5	E	455522	1	1	1	1	1	1
501	910.7317	0.142E-21	0.388E-02	0.07940.176	768.66480.75-	.010000	nu6	Ground 56	1	A1	55	0	A2	455522	1	1	1	1	1	1
501	910.8168	0.411E-21	0.392E-02	0.08660.319	337.72060.75-	.010000	nu6	Ground 37	2	E	36	1	E	455522	1	1	1	1	1	1
501	910.9510	0.776E-21	0.396E-02	0.09590.415	114.61180.75-	.010000	nu6	Ground 20	3	E	19	2	E	455522	1	1	1	1	1	1
501	910.9676	0.808E-22	0.411E-02	0.08360.255	622.64630.75-	.010000	nu6	Ground 41	7	A1	42	6	A2	455522	1	1	1	1	1	1
501	910.9676	0.808E-22	0.411E-02	0.08360.255	622.64630.75-	.010000	nu6	Ground 41	7	A2	42	6	A1	455522	1	1	1	1	1	1
501	911.0378	0.144E-21	0.404E-02	0.10400.347	117.56690.75-	.010000	nu6	Ground 11	5	E	12	4	E	455522	1	1	1	1	1	1
501	911.1226	0.127E-21	0.388E-02	0.07910.171	796.54080.75-	.010000	nu6	Ground 57	1	A2	56	0	A1	455522	1	1	1	1	1	1
501	911.1323	0.208E-21	0.407E-02	0.08890.392	310.42490.75-	.010000	nu6	Ground 26	6	E	27	5	E	455522	1	1	1	1	1	1
501	911.1377	0.162E-22	0.414E-02	0.07900.159	1023.88940.75-	.010000	nu6	Ground 55	8	E	56	7	E	455522	1	1	1	1	1	1
501	911.1641	0.602E-21	0.400E-02	0.10100.275	47.29950.75-	.010000	nu6	Ground 4	4	A1	3	3	A2	455522	1	1	1	1	1	1
501	911.1641	0.602E-21	0.400E-02	0.10100.275	47.29950.75-	.010000	nu6	Ground 4	4	A2	3	3	A1	455522	1	1	1	1	1	1
501	911.2539	0.385E-21	0.392E-02	0.08610.309	356.18730.75-	.010000	nu6	Ground 38	2	E	37	1	E	455522	1	1	1	1	1	1
501	911.4173	0.768E-21	0.396E-02	0.09470.417	124.59970.75-	.010000	nu6	Ground 21	3	E	20	2	E	455522	1	1	1	1	1	1
501	911.5113	0.112E-21	0.387E-02	0.07880.166	824.90910.75-	.010000	nu6	Ground 58	1	A1	57	0	A2	455522	1	1	1	1	1	1
501	911.5280	0.864E-22	0.411E-02	0.08400.264	601.95560.75-	.010000	nu6	Ground 40	7	A1	41	6	A2	455522	1	1	1	1	1	1
501	911.5280	0.864E-22	0.411E-02	0.08400.264	601.95560.75-	.010000	nu6	Ground 40	7	A2	41	6	A1	455522	1	1	1	1	1	1
501	911.5551	0.121E-21	0.404E-02	0.10520.333	111.59680.75-	.010000	nu6	Ground 10	5	E	11	4	E	455522	1	1	1	1	1	1
501	911.6566	0.613E-21	0.400E-02	0.10550.283	49.29480.75-	.010000	nu6	Ground 5	4	A1	4	3	A2	455522	1	1	1	1	1	1
501	911.6566	0.613E-21	0.400E-02	0.10550.283	49.29480.75-	.010000	nu6	Ground 5	4	A2	4	3	A1	455522	1	1	1	1	1	1
501	911.6716	0.210E-21	0.407E-02	0.08920.395	297.04810.75-	.010000	nu6	Ground 25	6	E	26	5	E	455522	1	1	1	1	1	1
501	911.6893	0.360E-21	0.392E-02	0.08570.300	375.15080.75-	.010000	nu6	Ground 39	2	E	38	1	E	455522	1	1	1	1	1	1
501	911.7170	0.181E-22	0.414E-02	0.07930.163	996.49420.75-	.010000	nu6	Ground 54	8	E	55	7	E	455522	1	1	1	1	1	1
501	911.8820	0.757E-21	0.396E-02	0.09370.417	135.08620.75-	.010000	nu6	Ground 22	3	E	21	2	E	455522	1	1	1	1	1	1
501	911.8979	0.991E-22	0.387E-02	0.07860.161	853.76950.75-	.010000	nu6	Ground 59	1	A2	58	0	A1	455522	1	1	1	1	1	1