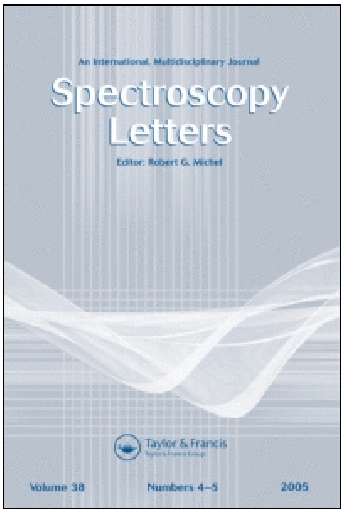

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Vibrational Transition Moment for the v_2 Band of CH_3D from Spectra Between 2100 and 2350 cm^{-1}

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Vibrational Transition Moment for the ν_2 Band of CH_3D from Spectra between 2100 and 2350 cm^{-1}.

Key Words: CH_3D; Transition Moment; Intensities; Planetary Atmospheres

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ABSTRACT: Absolute intensities of the ν_2 band of monodeuterated methane in the range of 2100-2350 cm^{-1} have been measured at 296.3 K by means of a Fourier Transform spectrometer at 0.006 cm^{-1} resolution. Together with data obtained earlier at 98.3 K we determined the rotationless transition moment $|R|^2=(9.33x10^{-4}\pm3\%)$ Debye^2 and the Herman-Wallis factor C=(0.0136±0.0003). We report the intensity measurements and the recalculated intensity values for both temperatures.

INTRODUCTION

IR line profile measurements appear as a basic tool in order to provide data usable by atmosphere chemist and modelers. The 5 μm region is of particular interest for being almost completely free of strong molecular absorptions. Also, in view of the increased level of accuracy required for the remote sensing of temperature it is of interest to perform detailed investigation of the associated parameters, together with the need for understanding the composition and dynamical structures of planetary atmospheres , which is part of an overall goal of planetary science, i.e. to learn about the current state of the solar system and to try to understand its formation and evolution.

The absorption spectrum of $^{12}CH_3D$ in the range 2000-2400 cm^{-1} is due to transitions of the parallel-type $\nu_2(A_1)$ band because of the C-D stretching[1]. The enormous amount of methane observed in the spectra of the outer planets is second only to the presence of molecular hydrogen. Its monodeuteride is an abundant minor constituent and thus an ideal probe of the deuterium in the related atmosphere.

Since CH_3D have been detected in the IR spectra of the atmospheres of Jupiter[2] (by an identification of the lines P(2) through P(12) in the ν_2), Saturn and Titan[3], absolute intensities and half-widths for individual rotation-vibration lines are needed for determining the CH_3D abundance and the D/H ratio.

In previous articles we have reported line intensities[4] and self-broadened half-widths[5] for several transitions of this parallel band from data recorded with a FT spectrometer. In the present study, similar measurements were performed but with emphasis on intensities.

All measurements reported in this work were obtained at room temperature, but references and recalculations are given also for T=98.3 K throughout the text.

Intensities were determined for about 250 transitions belonging to R, P and Q branches. The line intensity coefficients, the squared vibrational transition dipole-moment matrix elements, and the coefficient of the Herman-Wallis factor deduced in this work are reported.

Studies for this particular transition have been published, notably by Chackerian and Guelachvili [6], where previous works are referenced.

EXPERIMENTAL CONDITIONS

Several sets of spectra were recorded under different conditions related with different pressures and abundances of CH_3D, for further use in a fitting series of our spectral lines model. The experimental set up is essentially the same as before, but in short the spectra analyzed here were recorded with a Bomem DA 3.002 Fourier Transform spectrometer with actual resolution of 0.006 cm^{-1}. We used absorption cells of 2 , 5 and 57 mm in length, at pressures going from 50 to 150 torr, 15 to 100 torr, and 5 to 10 torr, respectively.

The spectral parameters were obtained by means of non-linear least-squares fitting techniques of each individual lines or multiplets, as appropriate, as described in ref.6. The isotopic composition of the sample was taken into account.

The quality of the fits can be estimated by calculating the standard deviations of the "observed-line/calculated-line" ratio point by point for the set of the points that each line or spectral segment under consideration spanned. This deviation was only a fraction of 1%.

RESULTS AND DISCUSSION

The line intensities are related to the squared vibrational transition dipole matrix element and can be expressed for a symmetric top molecule as follows:

$$S(J,K)=\frac{8\pi^3}{3hc}\frac{N(T)}{Q(T)}H_L.\nu(J,K).G_{JK}.\exp(-E_R^{''}.hc/kT).$$
$$\exp(-E_V^{''}.hc/kT).[1-\exp(-hc\nu/kT)]\left|R\right|^2.F$$

N(T) is the total number of molecules per unit volume and per unit pressure of the absorbing gas at temperature T. Q(T) is the total partition function; H_L is the Höln-London factor; ν(J,K) is the wavenumber of the rotation-vibration transition; G_{JK} is the corresponding spin-rotation statistical weight for a given J,K level; $E_R^{''}$ and $E_V^{''}$ are the lower state rotational and vibrational energies, respectively.

For the ν_2 fundamental band $E_0^{''}$=0, and the [1-exp(-hcν/kT))] factor is the induced transition term and practically unity, but it was also included.

The partition function is $Q(T)=Q_V\,Q_R$, where the first factor is taken as 1.0109 and Q_R =1599.314 at T=296.3 K and 308.513 for T=98.3 K that we have calculated using the newer constants obtained by Chackerian et al.[7]

|R| is the vibrational transition moment to be determined, independent of the temperature, and F is the Herman-Wallis factor, a function of the quantum numbers J and K, and accounts for vibration-rotation interactions.

The lower state energy $E_R^{''}$ for each transition was calculated with the ground-state constants given in ref.7. The rotational term value expression for a symmetric top molecule is:

$$E_R^{''}=B_0J(J+1)+(A_0-B_0)K^2-D_0^JJ^2(J+1)^2-D_0^{JK}(J+1)K^2$$
$$-D_0^KK^4+H_0^JJ^3(J+1)^3+H_0^{JK}J^2(J+1)^2K^2+H_0^{KJ}J(J+1)K^4+H_0^KK^6$$

For the Höln-London factors we have:

$$H_L=\frac{J^2-K^2}{J(2J+1)};\qquad \frac{K^2}{J(J+1)};\qquad \text{and}\qquad \frac{(J+1)^2-K^2}{(J+1)(2J+1)}$$

for P, Q and R branches, respectively. For the spin-rotation statistical weight:

$$G_{JK}=4(2J+1)(2-\delta_{0,K})\qquad \text{or}\qquad 4(2J+1)$$

for K=3n and K≠3n, respectively, with n=0,1,2...; δ is the Kronecker delta.

According to Hanson and Nielsen[8] for a parallel band: $F(m)=1+Cm$ where $m=-J$; 0; and J+1 for P, Q, and R branches, and C is the Herman-Wallis factor to be also determined.

After some manipulation we can write:

$$\mathcal{F}(m)=|R|^2\times(1+Cm)$$

and using the experimental data and the appropriate parameters the left side of the equation can be evaluated for each line. A plot and a linear least-squares fit of the function $\mathcal{F}(m)$ give us the $|R|^2$ and the C values, obtaining:

$$|R|^2=(9.33\mathrm{x}10^{-4}\pm3\%)\mathrm{Debye}^2 \quad \text{and} \quad C=0.0136\pm0.0003.$$

Figure 1 shows a plot of all the data and the line has been drawn according to least-square routines.

For this fitting we have used the intensity data obtained previously for the Q and P branches at T=98.3 K, and the present values for the P and R branches at T=296.3 K. For better accuracy, we have chosen not to use for this purpose the strong CH_3D absorption peaks in the Q branch measured at T=296.3 K, although they are mostly resolved at the pressure we run our spectra in this work.

Using the quantities determined, calculated intensities, Scalc, were computed for each line with the expression for S(J,K) given in the first equation, with the corresponding J and K values, and they are listed in Tables 1 to 5 along with the experimental values for each branch at both temperatures, including the Q at T=296.3 K. The present experimental data show to be an average 5% higher than those in ref.6. The same difference holds for $|R|^2$ and there is good agreement for the coefficient C.

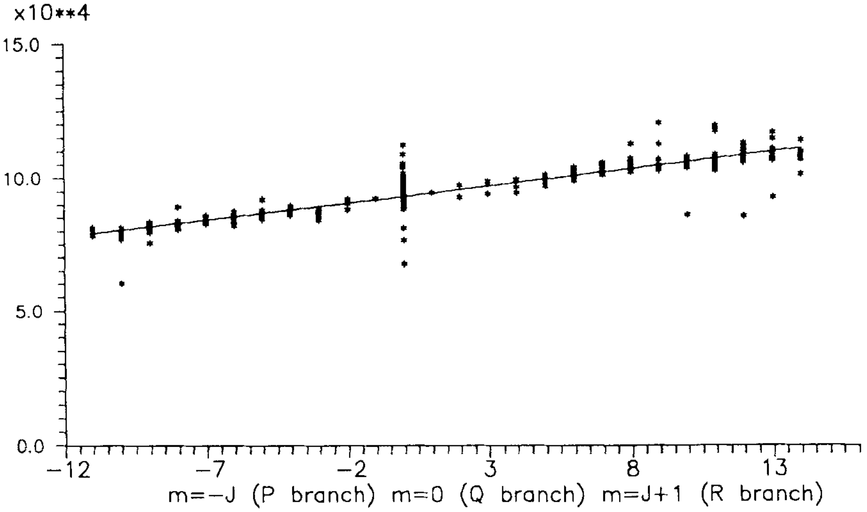

Figure 1: Plot and least-squared fitting of the function $\mathcal{F}(m)\times 10^4$ for the P, Q, and R branches of CH_3D from spectra at 98.3 K and 296.3 K.

In our case, the ratio Sexp/Scalc was evaluated for each line and they are reported in each case. For the P branch at 98.3 K the agreement is very good, with a standard deviation (S.D.) of 0.023. For the Q branch at 98.3 K the agreement is also good (S.D.=0.062), because it is very close to unity for most of the lines, except for some J rather high, as for instance the cases of the Q(9,4), Q(9,7), Q(8,7) and Q(6,5) lines. Probably they are not well isolated lines. For the P branch at 296.3 K the agreement is again very good, with S.D.=0.023. For Q lines at 296.3 K we find discrepancies as high as 10% lines for Q(5,3) and Q(5,4). One reason is notably that this portion of the spectrum is much crowded at this temperature, with more overlapping and wing effects. However, for most lines the comparison is much better, with a final S.D.=0.058 for this branch. For 108 lines of the R branch at 296.3 K the

Table 1: Q branch T=98.3 K CH_3D ν_2

J	K	FREQ.	EXP.	CALC.	RATIO
9	4	2196.814	0.01378	0.01230	1.120
8	3	2197.319	0.05392	0.04935	1.092
9	6	2197.575	0.03787	0.03716	1.019
7	2	2197.813	0.03591	0.03406	1.054
8	5	2197.928	0.05328	0.04982	1.070
7	3	2198.003	0.14630	0.13871	1.055
9	7	2198.066	0.02181	0.01950	1.118
7	4	2198.269	0.11540	0.10721	1.076
6	1	2198.296	0.02463	0.02312	1.065
8	6	2198.345	0.12065	0.11516	1.048
6	2	2198.410	0.08991	0.08709	1.032
6	3	2198.600	0.38118	0.35463	1.075
9	8	2198.628	0.01964	0.01887	1.041
5	1	2198.806	0.05811	0.05411	1.074
8	7	2198.835	0.06733	0.06042	1.114
6	4	2198.865	0.28601	0.27405	1.044
5	2	2198.920	0.21030	0.20384	1.032
5	3	2199.110	0.83447	0.82998	1.005
6	5	2199.205	0.40024	0.35762	1.119
4	1	2199.231	0.11712	0.11716	1.000
4	2	2199.344	0.45308	0.44135	1.027
5	4	2199.374	0.65152	0.64130	1.016
3	1	2199.569	0.23454	0.23922	0.980
6	6	2199.620	0.81576	0.82622	0.987
3	2	2199.683	0.89481	0.90109	0.993
5	5	2199.714	0.82313	0.83669	0.984
4	4	2199.797	1.33255	1.38819	0.960
2	1	2199.823	0.48581	0.48051	1.011
1	1	2199.992	1.04730	1.08558	0.965

Tables 1-5: Experimental and calculated intensities of rotation-vibration lines of P, Q and R branches of CH_3D at T=98.3 K and T=296.3 K.

In each table the last column corresponds to the ratio of experimental/calculated values.

Table 2: P branch T=98.3 K CH_3D ν_2

J	K	FREQ.	EXP.	CALC.	RATIO
6	0	2152.252	0.40952	0.41097	0.996
6	1	2152.291	0.38402	0.39166	0.981
6	2	2152.410	0.33576	0.33726	0.996
6	3	2152.607	0.50422	0.51502	0.979
6	4	2152.882	0.16520	0.16583	0.996
6	5	2153.235	0.07786	0.07617	1.022
5	0	2160.417	0.69755	0.68925	1.012
5	1	2160.456	0.63987	0.64858	0.987
5	2	2160.574	0.52485	0.53447	0.982
5	3	2160.769	0.73963	0.73691	1.004
5	4	2161.042	0.19084	0.18016	1.059
4	0	2168.504	1.00603	0.99057	1.016
4	1	2168.542	0.89723	0.91026	0.986
4	2	2168.659	0.69018	0.68577	1.006
4	3	2168.853	0.72034	0.72385	0.995
3	0	2176.510	1.15960	1.19125	0.973
3	1	2176.548	0.97730	1.03789	0.942
3	2	2176.664	0.59792	0.61086	0.979
2	0	2184.435	1.14790	1.13652	1.010
2	1	2184.473	0.83455	0.83548	0.999
1	0	2192.278	0.72903	0.72577	1.004

Table 3: Q branch T=296.3 K CH_3D ν_2

J	K	FREQ.	EXP.	CALC.	RATIO
10	6	2196.715	0.07352	0.07232	1.017
8	2	2197.128	0.01346	0.01254	1.073
9	5	2197.157	0.04352	0.04348	1.001
8	3	2197.319	0.05461	0.05460	1.000
10	8	2197.768	0.05726	0.05342	1.072
8	6	2198.345	0.18882	0.18268	1.034
5	1	2198.806	0.01142	0.01095	1.043
5	3	2199.110	0.20551	0.18698	1.099
5	4	2199.374	0.17388	0.15867	1.096
8	8	2199.398	0.12986	0.13486	0.963
7	7	2199.515	0.18396	0.17481	1.052
4	3	2199.534	0.27937	0.27706	1.008
6	6	2199.620	0.40145	0.42108	0.953
5	5	2199.714	0.24337	0.23354	1.042
4	4	2199.797	0.24649	0.23510	1.048
3	3	2199.872	0.39108	0.41760	0.936
2	2	2199.936	0.16644	0.15346	1.085

Table 4: P branch T=296.3 K CH_3D ν_2 band

J	K	FREQ.	EXP.	CALC.	RATIO
11	4	2110.909	0.03127	0.03098	1.009
11	5	2111.278	0.02685	0.02670	1.006
11	6	2111.728	0.04374	0.04398	0.995
11	7	2112.256	0.01756	0.01710	1.027
11	8	2112.858	0.01212	0.01226	0.989
10	0	2118.806	0.05615	0.05559	1.010
10	1	2118.847	0.05413	0.05468	0.990
10	2	2118.970	0.05159	0.05198	0.992
10	3	2119.174	0.09379	0.09537	0.983
10	4	2119.459	0.04099	0.04203	0.975
10	5	2119.825	0.03481	0.03537	0.984
10	6	2120.270	0.05494	0.05614	0.979
10	7	2120.794	0.02005	0.02053	0.977
10	8	2121.394	0.01253	0.01312	0.955
9	0	2127.285	0.07533	0.07430	1.014
9	1	2127.325	0.07093	0.07290	0.973
9	2	2127.447	0.06821	0.06880	0.991
9	3	2127.649	0.12420	0.12449	0.998
9	4	2127.932	0.05473	0.05366	1.020
9	5	2128.294	0.04422	0.04357	1.015
9	6	2128.735	0.06464	0.06511	0.993
9	7	2129.255	0.02074	0.02124	0.976
9	8	2129.851	0.00944	0.01022	0.924
8	0	2135.685	0.09545	0.09446	1.011
8	1	2135.725	0.08985	0.09237	0.973
8	2	2135.846	0.08547	0.08625	0.991
8	3	2136.046	0.14854	0.15299	0.971
8	4	2136.326	0.06187	0.06374	0.971
8	5	2136.685	0.04885	0.04880	1.001
8	6	2137.123	0.06504	0.06515	0.998
8	7	2137.638	0.01720	0.01601	1.074
7	0	2144.008	0.11600	0.11382	1.019
7	1	2144.047	0.10914	0.11077	0.985
7	2	2144.167	0.10096	0.10181	0.992
7	3	2144.365	0.17393	0.17511	0.993
7	4	2144.643	0.06862	0.06896	0.995
7	5	2144.999	0.04773	0.04725	1.010
7	6	2145.433	0.04679	0.04759	0.983
6	0	2152.252	0.12997	0.12938	1.005
6	1	2152.291	0.12196	0.12495	0.976
6	2	2152.410	0.11118	0.11200	0.993
6	3	2152.607	0.17732	0.18285	0.970
6	4	2152.882	0.06424	0.06465	0.994
6	5	2153.235	0.03216	0.03350	0.960
5	0	2160.417	0.13883	0.13766	1.008
5	1	2160.457	0.12779	0.13128	0.973
5	2	2160.574	0.11226	0.11261	0.997
5	3	2160.769	0.16509	0.16601	0.994
5	4	2161.042	0.04466	0.04457	1.002
4	0	2168.504	0.13557	0.13540	1.001
4	1	2168.543	0.12326	0.12610	0.978
4	2	2168.659	0.09769	0.09889	0.988
4	3	2168.853	0.11037	0.11161	0.989
3	0	2176.510	0.11943	0.12021	0.994
3	1	2176.549	0.10219	0.10614	0.963
3	2	2176.664	0.06350	0.06503	0.976
2	0	2184.435	0.09284	0.09133	1.017
2	1	2184.473	0.06622	0.06805	0.973

S.D. value is only 0.046, even in spite of a few cases, like the R(8,8); R(11,11); and R(12,3) lines with discrepancies of around 15%.

Overall, the standard deviation of the ratio Sexp/Scalc amounts to 0.034, having included 233 lines in this calculation. The largest deviations were noted for the P(J,K=9) lines, which were not included for the final fit. In fact, for the P(11,9) we measured Sexp=0.00876 (at T=296.3 K), but the ratio was only 0.56. For the P(10,9), Sexp=0.00928, and the ratio 0.74. At the lower

Table 5: R branch T=296.3 CH_3D ν_2 band

J	K	FREQ.	EXP.	CALC.	RATIO
0	0	2207.714	0.05396	0.05385	1.002
1	0	2215.304	0.10731	0.10547	1.017
1	1	2215.342	0.07625	0.07858	0.970
2	0	2222.807	0.15018	0.14917	1.007
2	1	2222.844	0.12774	0.13172	0.970
2	2	2222.955	0.08212	0.08070	1.018
3	0	2230.221	0.18213	0.18056	1.009
3	1	2230.256	0.16174	0.16816	0.962
3	2	2230.369	0.13333	0.13188	1.011
3	3	2230.552	0.14631	0.14884	0.983
4	0	2237.544	0.20026	0.19729	1.015
4	1	2237.581	0.18332	0.18815	0.974
4	2	2237.691	0.16220	0.16139	1.005
4	3	2237.874	0.23506	0.23790	0.988
4	4	2238.130	0.06310	0.06387	0.988
5	0	2244.775	0.20542	0.19926	1.031
5	1	2244.811	0.18908	0.19245	0.983
5	2	2244.921	0.17706	0.17249	1.026
5	3	2245.103	0.28133	0.28159	0.999
5	4	2245.358	0.09991	0.09956	1.003
5	5	2245.685	0.05214	0.05158	1.011
6	0	2251.911	0.19486	0.18840	1.034
6	1	2251.947	0.18154	0.18334	0.990
6	2	2252.056	0.17300	0.16850	1.027
6	3	2252.238	0.28888	0.28981	0.997
6	4	2252.492	0.11489	0.11413	1.007
6	5	2252.817	0.07882	0.07820	1.008
6	6	2253.213	0.08039	0.07875	1.021
7	0	2258.950	0.17221	0.16803	1.025
7	1	2258.987	0.16232	0.16432	0.988
7	2	2259.095	0.15673	0.15342	1.022
7	3	2259.276	0.29628	0.27213	1.089
7	5	2259.853	0.08801	0.08679	1.014
7	6	2260.248	0.11580	0.11585	1.000
7	7	2260.712	0.02950	0.02847	1.036
8	0	2265.891	0.14504	0.14206	1.021
8	1	2265.927	0.13736	0.13939	0.985
8	2	2266.035	0.13419	0.13153	1.020
8	3	2266.216	0.23428	0.23799	0.984
8	4	2266.468	0.10073	0.10257	0.982
8	5	2266.791	0.08338	0.08326	1.001
8	6	2267.184	0.12337	0.12442	0.992
8	7	2267.647	0.04363	0.04059	1.075
8	8	2268.175	0.02245	0.01952	1.150
9	0	2272.729	0.11621	0.11424	1.017
9	1	2272.765	0.11005	0.11235	0.980
9	2	2272.873	0.10788	0.10682	1.010
9	3	2273.053	0.19239	0.19594	0.982
9	4	2273.305	0.08496	0.08636	0.984
9	5	2273.627	0.07384	0.07266	1.016
9	6	2274.019	0.11396	0.11531	0.988
9	7	2274.479	0.04228	0.04215	1.003
9	8	2275.003	0.02670	0.02694	0.991
10	0	2279.463	0.09746	0.08759	1.113
10	1	2279.499	0.09444	0.08630	1.094
10	2	2279.607	0.08358	0.08249	1.013
10	3	2279.787	0.14956	0.15283	0.979
10	4	2280.037	0.06586	0.06841	0.963
10	5	2280.358	0.05793	0.05895	0.983
10	6	2280.749	0.09575	0.09708	0.986
10	7	2281.208	0.03677	0.03773	0.974
10	8	2281.725	0.02715	0.02705	1.004
10	10	2283.037	0.00752	0.00785	0.958
11	0	2286.090	0.06464	0.06415	1.008
11	1	2286.126	0.06444	0.06328	1.018
11	2	2286.233	0.06114	0.06075	1.006
11	3	2286.412	0.11197	0.11338	0.988
11	4	2286.662	0.05040	0.05133	0.982
11	5	2286.982	0.04680	0.04498	1.040
11	6	2287.372	0.07580	0.07594	0.998
11	7	2287.828	0.03146	0.03066	1.026
11	8	2288.338	0.02278	0.02338	0.974
11	10	2289.675	0.01025	0.01013	1.012
11	11	2290.385	0.00526	0.00461	1.142
12	0	2292.606	0.04513	0.04494	1.004
12	2	2292.748	0.04216	0.04274	0.986
12	3	2292.927	0.09326	0.08022	1.163
12	4	2293.176	0.03690	0.03663	1.007
12	5	2293.495	0.03262	0.03250	1.004
12	6	2293.883	0.05568	0.05585	0.997
12	7	2294.336	0.02411	0.02313	1.042
12	8	2294.836	0.01778	0.01833	0.970
12	10	2296.202	0.00930	0.00950	0.979
12	11	2296.911	0.00611	0.00575	1.063
12	12	2297.694	0.00500	0.00514	0.973
13	0	2299.006	0.02976	0.03014	0.987
13	1	2299.042	0.02869	0.02980	0.963
13	2	2299.149	0.02823	0.02877	0.981
13	3	2299.326	0.05565	0.05423	1.026
13	4	2299.575	0.02404	0.02493	0.964
13	5	2299.893	0.02146	0.02233	0.961
13	6	2300.279	0.03752	0.03888	0.965
13	7	2300.709	0.01496	0.01640	0.912
14	0	2305.288	0.01959	0.01938	1.011
14	1	2305.324	0.01810	0.01917	0.944
14	2	2305.430	0.01830	0.01855	0.987
14	3	2305.607	0.03537	0.03509	1.008
14	4	2305.854	0.01602	0.01622	0.988
14	5	2306.170	0.01500	0.01464	1.025
14	6	2306.554	0.02646	0.02574	1.028
14	7	2306.998	0.01173	0.01101	1.066
14	8	2307.461	0.00939	0.00913	1.029
14	10	2308.909	0.00512	0.00559	0.916
15	0	2311.446	0.01189	0.01195	0.995
15	1	2311.482	0.01145	0.01183	0.968
15	2	2311.587	0.01132	0.01146	0.987
15	3	2311.764	0.02330	0.02175	1.071
15	6	2312.703	0.01674	0.01625	1.030

temperature these lines do not show up. This effect has been already noted in ref.6 and according to Olson[9] this perturbation is due to a localized resonance of J,K=9 levels and J,K=8, l=+2 levels of $2\nu_6$. This resonance affecting line positions in these bands should be also perturbing the individual line intensities.

CONCLUSION

Summarizing, we have obtained the spectrum of CH_3D in the important 5 μm observational window, between 2150 and 2350 cm^{-1} and measured the intensity of individual lines of the spectrum. Our fitting procedure allowed us to accurate reproducing profiles of individual lines or set of lines within limited segments, providing us intensity parameters of similar quality. From these data, the rotationless transition moment is derived, and this is a very useful quantity that can be used to recalculate intensities at any given temperature. For the three branches of this spectrum we have found an overall agreement within 3.5% for 233 lines included in the corresponding calculation. For the intensities reported we have employed a greater variety of abundances than in past works, extending the data for the R branch and somewhat for the Q, both at 296.3 K. In addition, as the spectra obtained at lower temperature are less blended or overlapped by other lines or wings of neighboring lines, we think that these new data and the related quantities seem to be preferred.

As $^{12}CH_3D$ is known to be present in Jupiter, as well as in Saturn and Titan the investigation of parameters like these ones in the laboratory is hence of importance for astrophysical applications.

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