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A SEARCH FOR INTERSTELLAR CH_3D: LIMITS TO THE METHANE ABUNDANCE IN ORION-KL

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ABSTRACT

A search has been performed for interstellar CH_3D via its $J_K = 1_0\text{–}0_0$ transition at 232 GHz and its $J_K = 2_0\text{–}1_0$ and $J_K = 2_1\text{–}1_1$ lines at 465 GHz using the NRAO 12 m and CSO 10 m telescopes toward Orion-KL. This search was done in conjunction with laboratory measurements of all three transitions of CH_3D using millimeter/submillimeter direct absorption spectroscopy. The molecule was not detected down to a 3 σ level of $T_A^* < 0.05$ K for the $J_K = 2_1\text{–}1_1$ line toward Orion, which indicates an upper limit to the CH_3D column density of $N < 6 \times 10^{18}$ cm^{-2} in the hot core region and a fractional abundance (with respect to H_2) of $<6 \times 10^{-6}$. These measurements suggest that the methane abundance in the Orion hot core is $f < 6 \times 10^{-4}$, assuming D/H ~ 0.01. Such findings are in agreement with recent hot core chemical models, which predict $CH_4/H_2 \sim 10^{-4}$.

Subject headings: ISM: abundances — ISM: clouds — ISM: individual (Orion-KL) — ISM: molecules — molecular processes

1. INTRODUCTION

Chemical models predict CH_4, the simplest stable organic molecule, to be one of the most abundant polyatomic species in interstellar dense clouds, since it only involves one carbon atom bonded to four hydrogen atoms (e.g., Mitchell 1977). In addition, methane is thought to have an enhanced abundance in hot, dense gas, such as the Orion hot core region, because of evaporation of this molecule from grain mantles (e.g., Brown, Charnley, & Millar 1988). However, CH_4 is completely symmetric, having the geometry of a perfect tetrahedron, and hence it does not possess a permanent electric dipole moment. Thus, the molecule does not have a normal pure rotational spectrum and consequently cannot be readily observed in cooler, dense gas typical of molecular clouds.

Methane, on the other hand, does possess a rovibrational spectrum in the infrared which has been detected toward IRC +10216 (Hall & Ridgway 1978) and possibly in a few star-forming regions (Lacy et al. 1991). However, the kinetic temperatures found in molecular clouds, even toward star-forming objects, are typically only $T \sim 10\text{–}100$ K. Therefore, vibrational excitation is not commonly found in interstellar molecules, especially for regions of extended gas, the dominant constituent of molecular clouds. Hence, it is difficult to test models of interstellar chemistry using CH_4 infrared observations.

An alternative method to obtain interstellar methane abundances is possible by observing its singly deuterated form, CH_3D, which has a pure rotational spectrum. Because CH_3D is only slightly asymmetric, however, its dipole moment is small: $\mu \sim 0.006$ D (Wofsy, Muenter, & Klemperer 1970). Moreover, given the normal cosmic D/H ratio of 10^{-5}, detection of CH_3D should prove difficult. A search was carried out for CH_3D via its $J_K = 1_0\text{–}0_0$ transition at 232 GHz by Pickett, Cohen, & Phillips (1980), but given the receiver technology at the time, not very sensitive limits ($T_A^* \lesssim 0.2$ K) were obtained in Orion-KL. However, recent observations of interstellar molecules have clearly demonstrated that significant deuterium fractionation may occur in dense clouds and that the cosmic D/H ratio has little to do with abundances of deuterated species in comparison with their parent molecules. For example, considerable enrichment is found in several deuterated molecules toward Orion-KL, with $HDCO/H_2CO \sim 0.01\text{–}0.03$, $CH_3OD/CH_3OH \sim 0.01\text{–}0.06$, and $NH_2D/NH_3 \sim 0.003\text{–}0.05$ (Loren & Wootten 1985; Mauersberger et al. 1988; Walmsley et al. 1987). These high D/H ratios are believed to indicate that these species evaporated from grain surfaces (e.g., Plambeck & Wright 1987).

Here we present results of a renewed search for interstellar CH_3D toward Orion-KL. We have conducted measurements of the $J_K = 1_0\text{–}0_0$ transition at 232 GHz and the $J_K = 2_1\text{–}1_1$ and $2_0\text{–}1_0$ lines at 465 GHz of this isotopomer. We also have carried out laboratory measurements of all three CH_3D transitions. Although spectral lines were detected at the $J_K = 1_0\text{–}0_0$ and $J_K = 2_0\text{–}1_0$ frequencies in Orion, no feature was present at the $J_K = 2_1\text{–}1_1$ line, hence making our search negative. In this paper both the astronomical and laboratory data are summarized. We also present upper limits for the methane abundance and discuss its implications for interstellar chemistry.

2. OBSERVATIONS

The fundamental transition $J_K = 1_0\text{–}0_0$ of CH_3D at 232,644.3 MHz (see Picket et al. 1980) was originally searched for using the NRAO[1] 12 m telescope at Kitt Peak, Arizona in 1991 April. At this frequency, the beam size was 28″, and the beam efficiency was $\eta_C = 0.4$. The temperature scale was determined by the chopper wheel method and corrected for forward spillover losses, and it is given in terms of T_R^*. Conversion to radiation temperature is then

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[1] NRAO is operated by the Associated Universities, Inc., under contract with the National Science Foundation.

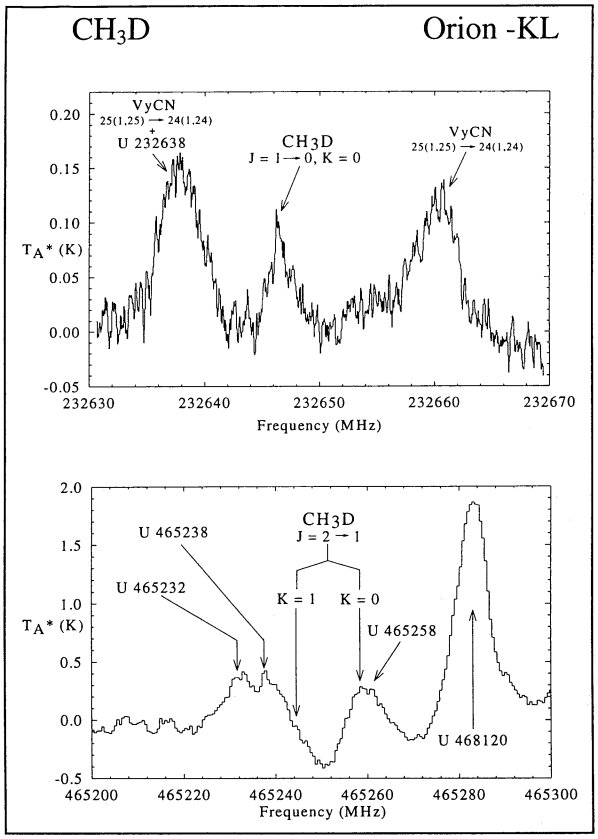

FIG. 1.—Spectra taken at the frequencies of the CH_3D $J = 1 \rightarrow 0$ and $J = 2 \rightarrow 1$ transitions at 232 and 465 GHz, respectively, using the CSO. The frequency scale assumes $V_{\rm LSR} = 9$ km s^{-1} in all cases. *Top:* The spectrum at 232 GHz, where a spectral line occurs near the CH_3D $J_K = 1_0$–0_0 frequency. A vinyl cyanide line is present in the lower sideband in these data, which appears twice due to a local oscillator shift. For one local oscillator setting, it is blended with a *U* line from the upper sideband. *Bottom:* The data taken at 465 GHz. Although a spectral feature occurs near the $J_K = 2_0$–1_0 frequency of CH_3D, there is not a line corresponding to the $J_K = 2_1$–1_1 transition.

$T_R = T_R^*/\eta_C$. The receiver used was a dual channel SIS mixer operated in a double-sideband (DSB) mode. The back ends used were two 256 channel filter banks with 1 MHz resolution, one for each receiver channel. Data were taken by position switching.

Additional observations were done of the $J_K = 1_0$–0_0 line using the Caltech Submillimeter Observatory (CSO) 10 m telescope in 1993 September. At 232 GHz, $\theta_b \sim 40''$ and $\eta_B = 0.8$. The receiver used was a single channel SIS mixer, again operated in DSB mode. In addition measurements of the two *K* components of the $J = 2 \rightarrow 1$ transition of CH_3D near 465 GHz were conducted in 1992 March and 1995 February using the CSO. The two *K* components were observed simultaneously. The beam size at this frequency is $\theta_b \sim 15''$, and $\eta_B = 0.5$. Again, the receiver used was a single channel double-sideband SIS mixer. The temperature scale for all CSO observations is in terms of T_A^*, such that $T_R = T_A^*/\eta_B$. The back end used was a 1024 channel AOS spectrometer with 500 kHz resolution. The CSO measurements were also carried out by position switching.

All observations were taken toward the position $\alpha = 5^h32^m46\overset{s}{.}8$, $\delta = -5°24'23\overset{''}{.}0$ (1950.0) in Orion.

3. EXPERIMENTAL

Spectra CH_3D were obtained in the laboratory using a millimeter/submillimeter wavelength direct absorption spectrometer, which is described in detail elsewhere (Ziurys et al. 1994). Briefly, the instrument consists of a tunable source of millimeter/submillimeter radiation, a gas absorption cell, and a detector. The sources for the spectrometer are phase-locked Gunn oscillators (65–140 GHz), used in conjunction with frequency multipliers to cover the range 115–520 GHz. The radiation is quasi-optically propagated through the absorption cell, which is a double-pass system. The detector is a helium-cooled InSb bolometer. Phase-sensitive detection is employed by FM modulation of the Gunn oscillators.

For these experiments, CH_3D gas, purchased from a commercial source (Icon), was contained in a quartz tube about 6 cm in diameter and 40 cm in length with quartz windows sealing both ends of the tube. CH_3D was added to this cell at the desired pressure ($\sim$50 mtorr), and the cell was then sealed. The tube was then inserted into the spectrometer and data taken, allowing for long signal averaging without having to free-flow CH_3D gas.

4. RESULTS

In the initial search for CH_3D using the NRAO 12 m telescope, a weak feature was found at the frequency of the $J_K = 1_0$–0_0 line at 232 GHz toward Orion-KL. This line was subsequently confirmed using the CSO, and this spectrum is shown in the top panel of Figure 1. As can be calculated from Table 1, this feature has a line width of $\sim$4 km s^{-1} and an LSR velocity near 5 km s^{-1}, if it arises from the $J_K = 1_0$–0_0 line of CH_3D. The frequency of this transition of CH_3D had been previously measured in the laboratory by Pickett, Cohen, & Phillips (1980) to be 232,644.327(18) MHz, and hence there was no question as to its exact value. Such line parameters suggested that the observed feature, if due to CH_3D, had an origin in the Orion hot core because of its slightly lower velocity (5 vs. 9 km s^{-1}). Because large enhancements for deuterated species are commonly found in the hot core (e.g., Plambeck & Wright 1987), this result was not unexpected. A feature was

TABLE 1

LINES OBSERVED IN THE BANDPASS

Frequency (MHz)	Identification	T_A^* (K)	$V_{\rm LSR}$ (km s^{-1})	$\Delta V_{1/2}$ (km s^{-1})	Comments
229,647.7..........	VyCN	0.12	5.3	6.5	LSB line; appears twice in spectrum
U232,638	...	~0.1	9.0	~5	blended with VyCN
U232,647	...	0.1	9.0	3.8	near CH_3D $J_K = 1_0$–0_0 transition
U465,232	...	0.5	9.0	~6	blended with U465,238
U465,238	...	0.6	9.0	~6	blended with U465,232
U465,258	...	0.5	9.0	5.8	near CH_3D $J_K = 2_0$–1_0 transition
U468,120	...	1.8	9.0	4.5	...

TABLE 2

OBSERVED FREQUENCIES AND REVISED CONSTANTS FOR CH$_3$D

A. OBSERVED FREQUENCIES

Transition (J_K)	Frequency (MHz)
1_0–0_0	232,644.301 (0.075)
2_0–1_0	465,250.691 (0.075)
2_1–1_1	465,235.540 (0.075)

B. REVISED CONSTANTS

Constant	ν (MHz)[a]	Previous Work (MHz)[a]
B_0	116,325.309(12)	116,323(2),[b] 116,325.309(27)[c]
D_J	1.5796(21)	1.572(7)[b]
D_{JK}	3.79(3)	3.78(3)[b]

[a] Errors listed are 3 σ.

[b] From Tarrago et al. 1976.

[c] From Pickett et al. 1980.

also detected near the CH$_3$D frequency in Sgr B2 and W51.

The results of the search for the $J_K = 2_1$–1_1 and 2_0–1_0 transition near 465 GHz were not as clear. In the 1992 March measurements, the data showed two spectral features which possibly could correspond to the $J_K = 2_1$–1_1 and $J_K = 2_0$–1_0 components of CH$_3$D, provided they had velocities near 5–6 km s^{-1}. However, the candidate $J_K = 2_1$–1_1 feature appeared as a shoulder on a stronger line, which is unidentified, and the signal-to-noise ratio was not particularly good. Moreover, the frequencies of these two K components of the $J = 2 \rightarrow 1$ transition of CH$_3$D had never been measured directly before in the laboratory, and they could only be predicted from the ro-vibrational data of Tarrago et al. (1976). The absolute accuracy of such calculations was not clear.

Because of these uncertainties, we decided to measure directly the $J = 2 \rightarrow 1$ CH$_3$D transitions in the laboratory and remeasure the $J = 1 \rightarrow 0$ line frequency as well. The details of these measurements have been described in the experimental section of this paper. The resulting rest frequencies are listed in Table 2 and have an accuracy of better than ± 75 kHz. A sample laboratory spectrum of the $J = 2 \rightarrow 1$ transitions, observed in absorption, is shown in Figure 2.

With these new laboratory frequencies, we repeated our search for the $J = 2 \rightarrow 1$ lines of CH$_3$D in 1995 February. For these observations, a spectrum with much better sensitivity was obtained toward Orion because of an improved receiver. These data are shown in the bottom panel of Figure 1. As this figure illustrates, a line with $T_A^* \sim 0.5$ K is present at the frequency of the $J_K = 2_0$–1_0 transition, for a velocity of $V_{\rm LSR} \simeq 5$ km s^{-1}, the same LSR velocity as the $J = 1 \rightarrow 0$ candidate line. However, there is no obvious feature present at the frequency of the $J_K = 2_1$–1_1 transition, which is 15.15 MHz lower in frequency (or to the left) of the $J_K = 2_0$–1_0 component. There are lines near 465,238 MHz and 465,232 MHz, either of which could correspond to the $J_K = 2_1$–1_1 line, but neither have a matching $J_K = 2_0$–1_0 component at $\sim$15 MHz to higher frequency.

The obvious failure to detect two distinct K components of CH$_3$D is clear evidence for its absence in Orion. The components are separated in energy by only 2 K, so both should be detectable in the high-excitation gas of the Orion hot core. An upper limit to the CH$_3$D antenna temperature can be derived from the $J_K = 2_1$–1_1 component, which is $T_A^* \sim 0.05$ K.

The frequencies, intensities, and line widths of the spectral lines detected in Figure 1 are listed in Table 1. In addition, revised rotational constants for CH$_3$D are given in Table 2. As the table shows, they are in good agreement with past estimates from Pickett et al. (1980) and Tarrago et al. (1976).

5. DISCUSSION

5.1. *Column Density and Fractional Abundance for* CH$_3$D

The upper limit to the CH$_3$D column density was calculated using the following formula, which assumes low optical depth, for $J_K \rightarrow (J-1)_K$:

$$N_{\rm tot} \le \frac{3k10^5 T_R \Delta V_{1/2} J e^{h\nu/kT_{\rm ex}} Q_{\rm rot}}{8\pi^3 \nu \mu_0^2 (J^2 - K^2) e^{-\Delta E/kT_{\rm rot}} S_{I,K}} . \qquad (1)$$

In this equation, which does not assume the Rayleigh-Jeans approximation, ν is the frequency of the transition, μ_0 is the dipole moment, ΔE is the energy of the $(J-1)$ level above ground state, and $T_{\rm rot}$ and $T_{\rm ex}$ are the rotational and excitation temperatures, respectively. T_R is the upper limit to the measured line temperature, and $\Delta V_{1/2}$ is the assumed line width. The term $Q_{\rm rot}$ is the rotational partition function, which for a symmetric top molecule can be approximated by (e.g., Townes & Schawlow 1975)

$$Q_{\rm rot} \sim (B^2 A / \pi T_{\rm rot}^3)^{-1/2} , \qquad (2)$$

where B and A are the rotational constants of the molecule. $S_{I,K}$ defines the statistical weight factor due to the presence of three equivalent nuclei with $I = \frac{1}{2}$, i.e., the three protons (also see Townes & Schawlow 1975):

$$S_{I,K} = \frac{2(4I^2 + 4I)}{4I^2 + 4I + 1} . \qquad (3)$$

In this formula, the 2.7 K microwave background was neglected (see Ziurys, Hollis, & Snyder 1994). Also because the dipole moment of CH$_3$D is so small, it was assumed that

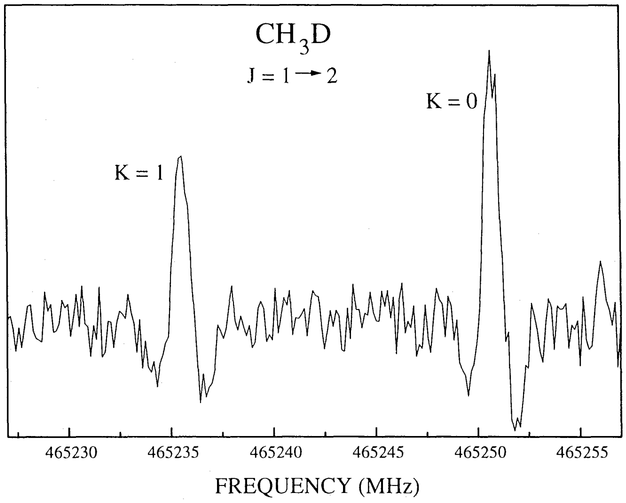

FIG. 2.—Laboratory spectrum of the CH$_3$D $J_K = 2_0$–1_0 and $J_K = 2_1$–1_1 transitions observed in absorption near 465 GHz. These measurements confirm the 15 MHz splitting of the two K components of this transition. This spectrum represents one 3 minute scan. The lines appear in emission because of the phase-sensitive detection scheme employed.

$T_{ex} = T_{rot}$, i.e., the excitation temperature within the 2_1–1_1 transition equals that governing the population in the rotational ladder.

To actually calculate the limit to the column density, it was assumed that CH_3D would arise primarily from the Orion hot core region. Simple, heavily saturated species such as NH_3 appear to have high abundances in the hot core, and CH_4 is likely to follow this pattern (e.g., Brown et al. 1988). For this region, the approximate kinetic temperature is ~200 K (e.g., Blake et al. 1987), and hence it was assumed that $T_{ex} = T_{rot} = 200$ K. Also $\Delta V_{1/2}$ was estimated to be 10 km s^{-1}, the canonical "hot core" line width. The upper limit to the line radiation temperature was $T_R = T_A^*/\eta_B = 0.1$ K, where $T_A^* < 0.05$ was the upper limit for the $J_K = 2_1$–1_1 transition.

Using these assumptions, the upper limit to the CH_3D column density in the Orion hot core was calculated to be $N_{tot} \lesssim 2 \times 10^{18}$ cm^{-2}. This number is based solely on the limit obtained for the $J_K = 2_1$–1_1 transition (assuming $V_{LSR} = 5$ km s^{-1}), which was not contaminated by other lines as the other two transitions were. This limit assumes that the source fills the 15″ beam at 465 GHz. Correcting for the hot core size of 10″ (Masson et al. 1985), however, increases the upper limit by a factor of 3, or $N_{tot} < 6 \times 10^{18}$ cm^{-2}. If the total hydrogen column density is assumed to be $N(H_2) \sim 10^{24}$ cm^{-2} (e.g., Masson et al. 1985), as is appropriate for the hot core, then the fractional abundance of CH_3D in this region is $f \lesssim 6 \times 10^{-6}$.

5.2. *Implications for the* CH_4 *Abundance*

Estimating the methane concentration from CH_3D depends critically on the D/H ratio, which fortunately can be estimated from other molecules in Orion. Large deuterium enhancements have been found in several clumps toward the KL/IRc2 region, including the hot core and the so-called compact ridge, and in the "northern condensation" (e.g., Mangum, Plambeck, & Wootten 1991). In the hot core, measurements of NH_2D/NH_3 by Walmsley et al. (1987) yield D/H ~ 0.003, while DCN/HCN suggest D/H ~ 0.005 (Mangum et al. 1991). Observations of CH_3OH and CH_3OD by Mauersberger et al. (1988) indicate higher ratios of D/H ~ 0.01–0.06. However, it is not entirely clear from what region the CH_3OD lines actually arise. If a D/H ratio of 0.01 is assumed, the upper limit to the fractional abundance of methane in the Orion hot core is $f \lesssim 6 \times 10^{-4}$, with a corresponding column density limit of $N_{tot}(CH_4) \lesssim 6 \times 10^{20}$ cm^{-2}.

The only interstellar source in which methane has been conclusively detected is in the circumstellar shell of IRC +10216 (Hall & Ridgway 1978). These authors observed the 3.3 μm v_3 vibrational band of CH_4 and derived a corresponding column density of $N_{tot} = 3 \times 10^{17}$ cm^{-2}. They also measured a CO column depth of $N_{tot} = 10^{20}$ cm^{-2} in IRC +10216. If $CO/H_2 \sim 10^{-4}$, then the fractional abundance of methane, relative to H_2, is $f \sim 3 \times 10^{-7}$ in this object. This value is several orders of magnitude below our upper limit of $f \lesssim 6 \times 10^{-4}$ in Orion. However, the chemistry in the envelope of IRC +10216, a late-type carbon star, is quite different from that of the Orion hot core, so methane abundances are likely to vary between the two sources.

Lacy et al. (1991) searched for the 7.6 μm v_4 band of CH_4 toward several molecular clouds. There authors appeared to detect the R(0) and R(2) lines of this band toward NGC 7538 IRS 9, although the spectra were highly contaminated by telluric features. From these two lines, Lacy et al. derived a CH_4 column density of $N_{tot} \sim 2 \times 10^{16}$ cm^{-2} toward NGC 7538. Using their quoted CO column depth of 1.5×10^{19} cm^{-2}, and assuming $CO/H_2 \sim 10^{-4}$, the fractional abundance of CH_4 in this molecular cloud is $f \sim 10^{-7}$, relative to H_2. A similar fractional abundance was possibly found for W33A. Again, these CH_4 abundances are several orders of magnitude lower than our upper limit.

Lacy et al. also suggest a possible detection of the R(0) line of methane in Orion/IRc2. However, they do not derive a column density for CH_4 from this measurement, although a comparison of methane abundances may have been useful. Lacy et al. determined an LSR velocity of the possible R(0) feature of $V_{LSR} = -1$ km s^{-1}. To our knowledge, there is no known molecular emission in Orion that occurs at that velocity. Because of these uncertainties, the upper limit derived from CH_3D is probably the best estimate of the CH_4 abundance in Orion-KL.

5.3. *Constraints for Chemical Models*

The "hot core" cloud model by Brown et al. (1988) predicts high abundances of simple saturated molecules such as NH_3 and CH_4. These calculations considered gas-phase and grain-surface (hydrogenation) reactions, as well as grain mantle evaporation, in dense gas heated by a young star with $T \sim 150$ K. In this model, these authors estimate $f(CH_4) \sim 10^{-4}$, relative to H_2. A newer set of calculations by Brown & Millar (1989) considers the same model as Brown et al. (1988), but it also considers deuterium chemistry, along with a corresponding set of deuterium reactions on grains. Once again, the estimated abundance of methane for hot core-type gas is $f \sim 10^{-4}$.

The upper limit for methane in the Orion hot core region determined from the CH_3D search suggests $f \lesssim 6 \times 10^{-4}$. This value certainly does not contradict model predictions; however, this limit is not stringent enough to put the chemical calculations to the test. Given the contaminating spectral features at two of the three easily accessible CH_3D rotational transitions, obtaining more sensitive limits will be difficult.

6. CONCLUSIONS

A search has been carried out for the $J_K = 1_0$–0_0, 2_0–1_0, and 2_1–1_1, millimeter-submillimeter transitions of CH_3D at 232.64, 465.25, and 465.24 GHz. Laboratory measurements of these CH_3D frequencies were also performed to substantiate the astronomical work. Although spectral lines were clearly present at the 1_0–0_0 and 2_0–1_0 frequencies for CH_3D in Orion, assuming the hot core velocity, there was no obvious feature at the 2_1–1_1 frequency. Hence, CH_3D was clearly not detected in this source. The upper limit to the column density and fractional abundance of CH_3D on the Orion hot core, relative to H_2, is $N_{tot} \lesssim 6 \times 10^{18}$ cm^{-2} and $f \lesssim 6 \times 10^{-6}$. These values imply that the methane abundance in the hot core is $f < 6 \times 10^{-4}$, consistent with current chemical models, which predict $CH_4/H_2 \sim 10^{-4}$.

This search for CH_3D also illustrates the need for careful and objective measurements in the detection of new interstellar molecules. Based on two lines alone (the $J_K = 1_0$–0_0 and 2_0–1_0 transitions), we could have claimed an interesting discovery of interstellar CH$_3$D. However, failure to observe a consistent third transition, namely the $J_K = 2_1 - 1_1$ line, showed beyond any doubt that such a claim would have been premature and incorrect.

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