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DISCOVERY OF INTERSTELLAR METHANE: OBSERVATIONS OF GASEOUS AND SOLID CH_4 ABSORPTION TOWARD YOUNG STARS IN MOLECULAR CLOUDS

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ABSTRACT

We have searched in several molecular clouds for absorption at 7.6 μm due to gaseous and solid methane. Gaseous CH_4 was detected toward NGC 7538 IRS 9 and probably OMC-1 IRc2 and W33 A. The abundance of gaseous CH_4 is typically 10^{-3} that of CO. Solid CH_4 was probably detected toward NGC 7538 IRS 9 and possibly detected toward W33 A and NGC 7538 IRS 1. The abundance of solid CH_4 is comparable to that of solid CO. The total CH_4 abundance (predominantly in the solid phase) is 1%–4% of the total CO abundance (predominantly gaseous). The high fraction of CH_4 in the solid state suggests that it is made in the grain mantles.

Subject headings: infrared: spectra — interstellar: abundances — interstellar: molecules

1. INTRODUCTION

The chemistry of molecular clouds has been studied extensively during the 20 years since the first detection of polyatomic interstellar molecules. Over 70 molecules have been detected, and detailed models have been constructed of the reaction chains which lead to their formation (see, e.g., Prasad et al. 1987). Nevertheless, it is not yet clear whether gas-phase ion-molecule reactions can explain the abundances of interstellar molecules other than H_2 (as is usually assumed), or if reactions on surfaces of dust grains or in icy grain mantles are also important. This question has been difficult to attack through radio astronomy, the dominant technique for study of interstellar molecules, because many of the simplest hydrocarbons, for which grain-surface reactions are likely to be most important, have no rotational transitions. In contrast, several of these molecules are observable in the infrared, both in the gas phase and while still frozen on grains.

Our first attempt to use infrared spectroscopy to study interstellar molecules whose abundances are likely to have been altered by the presence of dust grains resulted in the detection of acetylene (C_2H_2; Lacy et al. 1989b). We found the abundance of C_2H_2 to be substantially higher than expected in a chemically evolved molecular cloud in which the chemistry is controlled by ion-molecular reactions, and we suggested that the C_2H_2 formed by gas-phase reactions when the clouds were chemically young, condensed onto grains, and was released when newly formed stars heated the grains.

Although C_2H_2 may be preserved on dust grains, most studies do not predict that it is formed in significant quantities there. In contrast, CH_4 is thought to form on grains through reactions of hydrogen with carbon atoms (d'Hendecourt, Allamandola, & Greenberg 1985; Brown, Charnley, & Millar 1988). Consequently, observations of a high abundance of CH_4, either frozen on grains or as a gas, would provide strong evidence that reactions on dust grains play an important role in interstellar chemistry.

Unfortunately, CH_4 is substantially more difficult to observe than is C_2H_2. Its observable vibrational bands (ν_3 at 3.3 μm and ν_4 at 7.6 μm) are weaker than the observed band of C_2H_2, and CH_4 is sufficiently abundant in the telluric atmosphere that its strongest lines are opaque over several 0.1 cm^{-1}, requiring a favorable Doppler shift for its observation. Lines of the ν_3 band of CH_4 were searched for by Knacke et al. (1985) toward OMC-1 BN and GL 490 but not detected.[6] Doppler-broadened lines of the ν_3 and ν_4 bands have about equal depths. The ν_4 band has the advantage, however, that it is at a minimum in the interstellar extinction curve, so that deeply embedded sources are observable. Interstellar gaseous CH_4 lines begin to emerge from the telluric lines in the 7.6 μm region at Doppler shifts of about 30 km s^{-1}. They are generally well removed from the telluric CH_4 lines with 50 km s^{-1} shifts, although in some cases telluric H_2O and N_2O lines interfere. The ν_4 band is also a relatively promising band for a search for solid CH_4 in grain mantles. It is relatively strong and is separated from the bands of other known hydrocarbon solids, although close to the wavelength of the 7.7 μm interstellar emission feature which has been attributed to C-C stretching vibrations in aromatic hydrocarbons. We report on a search for both gaseous and solid CH_4 in absorption in the ν_4 band.

2. OBSERVATIONS

The observations were made with IRSHELL, the University of Texas infrared echelle spectrograph (Lacy et al. 1989a) on the NASA Infrared Telescope Facility. IRSHELL measures a 64 point spectrum at each of 10 positions along a slit with 10" length. For observations of gaseous CH_4, the instrument was used with a dispersion of 0.06 cm^{-1} $pixel^{-1}$ and a slit width of 1.5 pixels, or 1".5. The resolution was about 0.12 cm^{-1}. The sources were placed at the center of the slit, splitting the light between columns 5 and 6 of the infrared detector array. These

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[6] A detection of a rotational transition of CH_4, weakly allowed by centrifugal distortion of the molecule, has been claimed by Fox & Jennings (1978), but refuted by Elldér et al. (1980) and Wilson & Snyder (1985); the observations reported here require a column density of CH_4 five orders of magnitude less than that claimed by Fox and Jennings.

two columns are offset spectrally by 0.5 pixel, resulting in 128 point Nyquist-sampled spectra when observations are made in this way. The spectra were flat-fielded with a dome-temperature blackbody spectrum and divided by a comparison source spectrum to remove telluric absorption lines.

For observations of solid CH_4, the echelle grating was replaced with a first-order grating, resulting in a dispersion of 0.002 μm, or approximately 0.34 cm^{-1}, $pixel^{-1}$. The spectral coverage of the detector array was 0.13 μm. To cover the 1 μm spectral region desired to obtain an adequate baseline for the solid CH_4 feature, a number of grating settings were required. We stepped the grating by one resolution element (2 pixel) steps, so that each pixel of the array independently scanned the spectrum. Two low-resolution observations of the comparison source, Vega, were made and were found to be consistent within the noise. As with the high-resolution spectra, the spectra were first flat-fielded, then divided by the flat-fielded spectrum of the comparison source.

Six young stars embedded in molecular clouds were observed for gaseous CH_4 absorption: W3 IRS 5 (on UT 1989 July 27), OMC-1 IRc2 (1990 February 15), W33 A (1990 July 11), GL 2591 (1990 July 4), and NGC 7538 IRS 1 and IRS 9 (1990 July 11). Only the $\nu_4 R(0)$ (1311.43 cm^{-1}) line was observed toward the first four of these sources. The $R(0)$ line and two blended $R(2)$[7] lines (1322.08, 1322.16 cm^{-1}) were observed toward the two sources in NGC 7538. The $R(0)$ spectra of OMC-1 IRc2, W33 A, NGC 7538 IRS 1, and NGC 7538 IRS 9 are shown in Figure 1. The $R(2)$ spectra of NGC 7538 IRS 1 and IRS 9 are shown in Figure 2. In each figure, a lunar spectrum is shown before correction for telluric absorption to show the positions and depths of the atmospheric lines. Note that the noise is much larger in the vicinity of telluric absorption lines, so that for the NGC 7538 sources, which have large Doppler shifts, the noise at the wavelengths of the interstellar lines is considerably less than it is in several other regions of the spectra, whereas the opposite is true for OMC-1 IRc2 and W33 A.

The equivalent widths and Doppler shifts of the observed lines are presented in Table 1. The primary uncertainty in determining the equivalent widths was the determination of the continuum level, as all lines were on the wings of telluric absorption lines. Statistical errors are small in all cases. The quoted uncertainties are estimated, based on the flatness of the nearby continuum and the proximity of telluric absorption features. In all cases where lines were detected, the observed velocity is within 7 km s^{-1} (one quarter the resolution) of the velocity of CO absorption. The detection of $R(0)$ and $R(2)$ absorption toward NGC 7538 IRS 9, at the expected Doppler shifts, and the sparsity of detectable interstellar absorption lines in the infrared, argues strongly that CH_4 has been detected toward this source. CH_4 has probably been detected toward OMC-1 IRc2 and W33 A as well, although telluric interference and the observation of only one line make these observations less certain. The $R(2)$ line may have been detected toward NGC 7538 IRS 1, but the failure to detect the $R(0)$ line makes this questionable (see below).

Three sources were observed on 1990 July 19 for solid CH_4 absorption: W33 A, NGC 7538 IRS 1, and NGC 7538 IRS 9.

[7] Because CH_4 is a spherical top molecule with four identical nuclei, it has a somewhat more complicated spectrum than diatomic or linear molecules. In particular, the $R(0)$ line ($\nu_4 = 0, J = 0 \rightarrow \nu_4 = 1, J = 1$) is single, whereas there are two $R(2)$ lines ($\nu_4 = 0, J = 2 \rightarrow \nu_4 = 1, J = 3$; see Herzberg (1945), pp. 37ff., 100ff., and 446ff.).

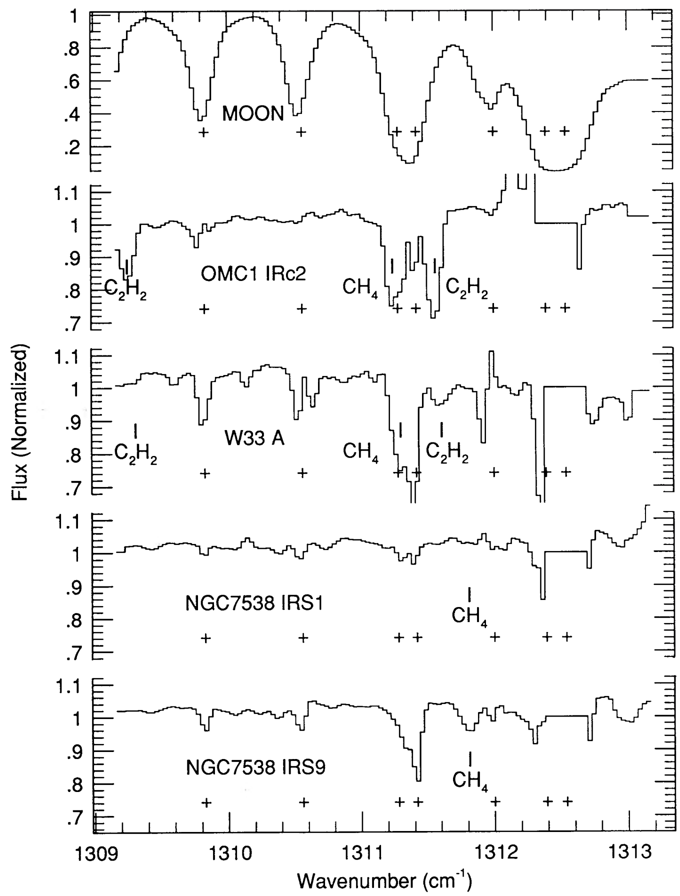

FIG. 1.—CH_4 $R(0)$ spectra. The top spectrum is of the Moon is flat-fielded, but not divided by a comparison spectrum. The other spectra are flat-fielded and divided by the spectrum of the Moon to remove atmospheric features. Atmospheric lines are labeled with a cross. Interstellar lines are labeled "I" and are shown at their expected Doppler-shifted positions. The two C_2H_2 lines, seen toward IRc2 and perhaps W33 A, are the $\nu_4 + \nu_5$ $P(8)$ (1309.48 cm^{-1}) and $P(7)$ (1311.77 cm^{-1}) lines. If unsaturated, the $P(7)$ line should be about 3 times stronger than the $P(8)$. Note the presence of excess noise and residual absorption on the telluric features, especially the CH_4 1311.43 cm^{-1} line.

These sources have been previously observed at lower resolution by Soifer et al. (1979), Willner et al. (1982), and Tielens & Allamandola (1987); they show strong 9.7 μm silicate absorption as well as 3.1, 6.0, and 6.8 μm features due to ices. The spectra, flat-fielded and divided by the spectrum of α Lyr, are shown, along with fits and an atmospheric transmission spectrum, in Figure 3. The spectra were fitted with the following function:

$$F_\lambda = F_0 \exp(-\tau) ,$$

where

$$\tau = \tau_a(\lambda - \lambda_a)^2 + \tau_{CH_4} \exp[-4(\ln 2)(\lambda - \lambda_{CH_4})^2/\Delta\lambda^2_{CH_4}] .$$

The first term in τ was used to approximate the falloff at long wavelengths due to the silicate feature and at short wavelengths due to the 6.8 μm feature, the short wavelength increase in dust extinction, and the cool temperatures of the sources. The second term represents the solid CH_4 absorption, with $\Delta\lambda_{CH_4}$ the FWHM of the feature. For W33 A and NGC 7538 IRS 1, an additional term representing a second absorption

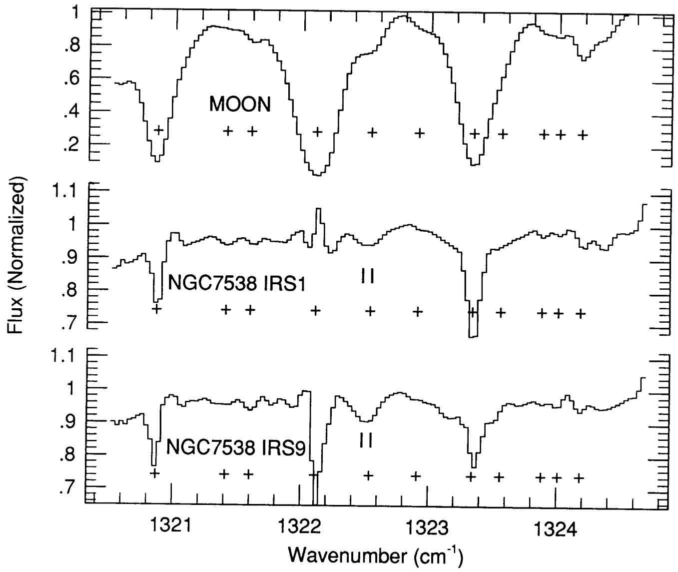

FIG. 2.—CH_4 $R(2)$ spectra. The two unresolved CH_4 lines are labeled "I."

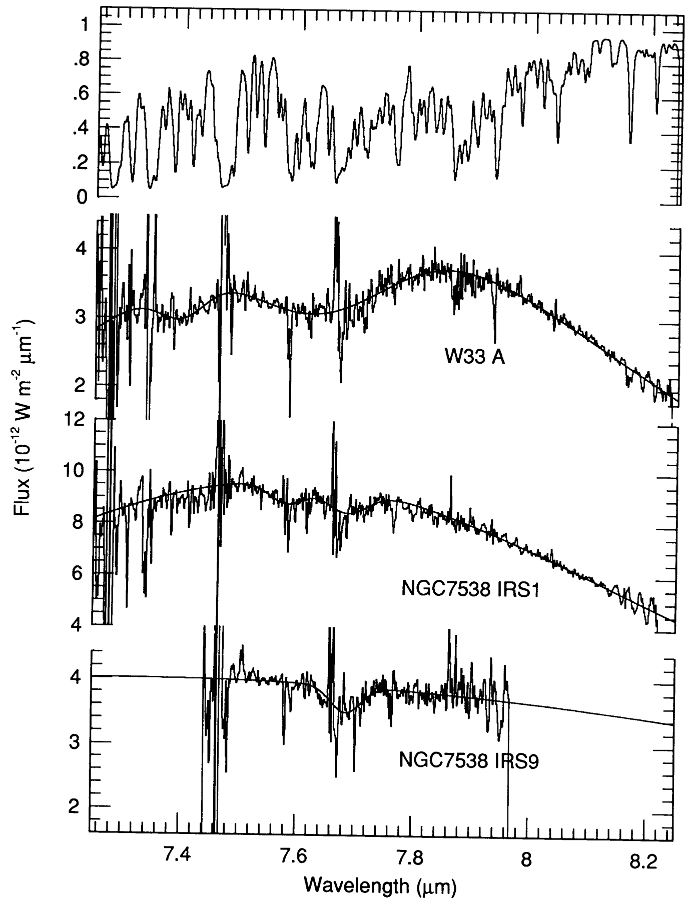

FIG. 3.—Solid CH_4 spectra. The top spectrum shows the measured atmospheric transmission. The next three spectra are of W33 A and NGC 7538 IRS 1 and IRS 9, each flat-fielded and flux calibrated by dividing by a spectrum of Vega. The smooth lines through the spectra are the fits described in the text.

feature was included. The fits were weighted by the atmospheric transmission at each wavelength. The results of the fitting are given in Table 2.

The band center of gaseous CH_4 is at 1306 cm^{-1} (7.66 μm). Laboratory measurements of CH_4 frozen in various ices give a band center between 1297 and 1306 cm^{-1} (7.71 and 7.66 μm), and a width of 8–15 cm^{-1} (0.05–0.09 μm) (d'Hendecourt & Allamandola (1986; d'Hendecourt 1984). The feature seen toward NGC 7538 IRS 9 has the center wavelength and width expected for CH_4 frozen in an ice mixture which broadens the transition only slightly. As we were unable to find another molecule with a solid state band at the observed wavelength, and essentially all solids have considerably broader bands than does CH_4, we identify the observed band with solid CH_4. Unfortunately the absorption by the telluric CH_4 Q-branch is severe over much of the feature, making its equivalent width rather uncertain. In addition, it is possible that gaseous interstellar CH_4 contributes to the feature. The fact that the observed feature extends shortward of the gaseous Q-branch edge argues that it is not purely due to CH_4 gas, but some contamination cannot be ruled out.

Features centered near the expected wavelength for solid CH_4 were also seen toward W33 A and NGC 7538 IRS 1. The feature seen toward NGC 7538 IRS 1 either is much broader than that seen toward IRS 9, or consists of two neighboring absorptions. If the latter is the case, the longer wavelength absorption matches that attributed to solid CH_4 in the spectrum of IRS 9. There appears to be a broad absorption feature toward W33 A, centered just shortward of the expected wavelength of solid CH_4, and a second feature centered near 7.4 μm. These features have previously been seen as a broad absorption in lower resolution spectra of W33 A, and a similar broad feature has been seen toward NGC 7538 IRS 9 (Tielens & Allamandola 1987). Possible explanations for the broad feature

TABLE 1

CH_4 EQUIVALENT WIDTHS AND VELOCITIES

OBJECT	w_ν (cm^{-1}) $R(0)$	w_ν (cm^{-1}) $R(2)$[a]	v_{LSR} (km s^{-1}) CH_4	v_{LSR} (km s^{-1}) CO[b]
W3 IRS 5	$<5 \times 10^{-3}$	...[c]	...	−42
OMC-1 IRc2	$(2.9 \pm 1) \times 10^{-2}$	...[c]	−1	−6[d]
W33 A	$(3.7 \pm 1) \times 10^{-2}$	...[c]	+27	+33
GL 2591	$<6 \times 10^{-3}$	...[c]	...	−11
7538 IRS 1	$<2 \times 10^{-3}$	$(0.8 \pm 0.4) \times 10^{-2}$[e]	−65	−59
7538 IRS 9	$(1.1 \pm 0.2) \times 10^{-2}$	$(1.5 \pm 0.4) \times 10^{-2}$[e]	−66	−60

[a] Including both unresolved lines.
[b] From Mitchell et al. 1990.
[c] Not observed.
[d] C_2H_2 velocity from Lacy et al. 1989b.
[e] May be affected by poorly corrected telluric absorption; see text.

TABLE 2

SOLID CH_4 FITTED PARAMETERS

Object	τ_{CH_4}	λ_{CH_4} (μm)	$\Delta\lambda_{CH_4}$ (μm)	w_λ (μm)
W33 A	0.33 ± 0.05[a]	7.631	0.27	0.096 ± 0.02[a]
	0.14 ± 0.05[a]	7.397	0.09	0.014 ± 0.01[a]
7538 IRS 1	0.11 ± 0.02	7.684	0.08	0.009 ± 0.003
	0.09 ± 0.02[a]	7.583	0.07	0.007 ± 0.003[a]
7538 IRS 9	0.10 ± 0.02	7.685	0.06	0.007 ± 0.003

[a] May not be due to solid CH_4.

TABLE 3

TEMPERATURES AND COLUMN DENSITIES

Object	T_{CH_4} (K)	$N_{CH_4 gas}$ (cm^{-2})	T_{CO}[a] (K)	$N_{CO gas}$[a] (cm^{-2})	$N_{CH_4 solid}$ (cm^{-2})	$N_{CO solid}$[a] (cm^{-2})	$\frac{CH_{4 tot}}{CO_{tot}}$
W33 A	23[b]	1.8×10^{16}[c]	23	2.0×10^{19}	1.7×10^{18}[d]	7×10^{17}	4.4×10^{-2}[d]
	120[b]	2.1×10^{17}[c]	120	1.9×10^{19}	...	...	...
7538 IRS 1	176[b]	$<2 \times 10^{16}$	25	1.4×10^{19}	2.6×10^{17}[e]	8×10^{16}	1.0×10^{-2}[e]
			176	1.3×10^{19}	4.5×10^{17}[d]	...	1.7×10^{-2}[d]
7538 IRS 9	86	3.8×10^{16}	26	1.5×10^{19}	2.0×10^{17}	1.5×10^{18}	1.3×10^{-2}
	40[f]	1.2×10^{16}[f]	...	...	...	...	...

[a] From Mitchell et al. 1990.
[b] Assumed temperature.
[c] Assuming all of the CH_4 is at the given CO temperature.
[d] Including the entire broad feature.
[e] Including only the narrow 7.68 μm feature.
[f] Assuming w_ν of $R(2) = 8 \times 10^{-3}$ (see text).

seen toward W33 A and the short wavelength absorption seen toward NGC 7538 IRS 1 are discussed below.

3. RESULTS

The column density and temperature of gaseous CH_4 can be calculated using the line strengths tabulated by Champion et al. (1989),[8] assuming the observed lines are optically thin. This assumption is probably valid for NGC 7538, which has relatively weak lines, but may well be in error for IRc2 and W33 A. The CH_4 results for W33 A and NGC 7538 IRS 1 and IRS 9 are compared with published CO measurements in Table 3.

Our observations of NGC 7538 IRS 1 are contradictory at the 2 σ level, as we observe an $R(0)/R(2)$ flux ratio less than 0.25, whereas the ratio cannot be less than 0.45, the high-temperature limit. Mitchell et al. (1990) observe two CO components toward IRS 1, with $N_{CO} = 1.4 \times 10^{19}$ cm^{-2} at 25 K and 1.3×10^{19} cm^{-2} at 176 K. Assuming that the CH_4 absorption comes from the hotter of these giving the smaller $R(0)/R(2)$ ratio and the weaker constraint on the CH_4 abundance, we derive $N_{CH_4} \leq 2 \times 10^{16}$ cm^{-2} from the $R(0)$ line, which is less affected by telluric interference than the $R(2)$ lines are. Toward NGC 7538 IRS 9 we detected both $R(0)$ and $R(2)$, with a flux ratio of 0.7 ± 0.2, giving a gas temperature of 86 K. Mitchell et al. find a CO temperature of 26 ± 5 K, substantially less than our CH_4 temperature. The most likely explanation of this discrepancy is that there is uncorrected telluric absorption at the wavelength of the $R(2)$ line pair, making the feature appear too strong in both IRS 1 and IRS 9. If we assume that the apparent $R(2)$ line in IRS 1 is entirely spurious and assume an equal amount of uncorrected telluric absorption in IRS 9, we derive an $R(2)$ equivalent width in IRS 9 of 8×10^{-3} cm^{-1}, and a temperature of 40 K, in much better agreement with the CO temperature. Since only one line was observed toward W33 A, we cannot derive a temperature of the CH_4 gas. Mitchell, Allen, & Maillard (1988) observe two components of CO gas toward W33 A, at 23 and 120 K. In Table 3 we give the column density of CH_4 required, assuming each temperature, but we emphasize that N_{CH_4} is very uncertain due to the uncertain temperature, telluric interference, and possible saturation of the observed line.

The interpretation of the solid CH_4 observations is complicated by the presence of an unexpectedly broad feature toward W33 A and either a broad feature or two narrow features toward NGC 7538 IRS 1. Three explanations seem reasonable for the broad feature. First, the CH_4 absorption can be broadened by thermal effects. CH_4 interacts very weakly with surrounding molecules, and so can rotate in an ice, resulting in a temperature-dependent line width. Allamandola (1990, private communication) has observed a line width of 20 cm^{-1} (0.12 μm) in pure solid CH_4 at 30 K. If the broadening is the same in interstellar ices, a temperature of ≈ 60 K could account for the line width observed in W33 A. Second, the CH_4 feature may broaden and shift in interstellar ices. Laboratory data indicate little broadening, but ions or radicals not found in the laboratory samples may have larger effects. Third, another unidentified molecule may be responsible.

The band strength of solid CH_4 is 6×10^{-18} cm^{-1}/cm^{-2} (d'Hendecourt & Allamandola 1986). The derived column densities of solid CH_4 are compared with those of gaseous CH_4 and of solid and gaseous CO in Table 3. In the solid phase, $CH_4/CO \approx 3$ toward NGC 7538 IRS 1 and ≈ 0.1 toward NGC 7538 IRS 9. If the broad 7.6 μm feature is due to solid CH_4, then CH_4 is significantly more abundant than CO on grains toward W33 A and NGC 7538 IRS 1. In the gas phase, the ratio of N_{CH_4}/N_{CO} is typically $\approx 10^{-3}$. CH_4 is predominantly solid, whereas CO is almost entirely gaseous. This result is perhaps not very surprising, as CH_4 is more condensible than CO in an ice matrix.

4. DISCUSSION

The observed gas-phase CH_4 abundances may be compared to calculations of interstellar chemistry. Most theoretical studies of the chemistry of molecular clouds concentrate on gas-phase ion-molecule reactions (e.g., Millar & Nejad 1985; Herbst & Leung 1986). These authors predict a peak CH_4/CO ratio $\approx 10^{-2}$ about 10^6 yr after formation of a molecular cloud from H I gas with a density of 10^4 cm^{-3}. The CH_4 abundance then falls to about 10^3 of CO, after about 10^7 yr. Given the substantial uncertainties in our CH_4 column densities, we

[8] The line strengths, $S_{ij}(cm^{-1}\ atm^{-1}\ cm^{-1})$ given by Champion et al. are for a gas at $T = 296$ K. The equivalent width w_ν for the column density of CH_4 of N_{CH_4} at a temperature T is given by:

$$w_\nu(cm^{-1}) = \left(\frac{N_{CH_4}}{n_0}\right) S_{ij}(T_0) \left(\frac{T_0}{T}\right)^{3/2} \exp\left[\frac{E_j}{k}\left(\frac{1}{T_0} - \frac{1}{T}\right)\right],$$

where $n_0 = 2.5 \times 10^{19}$ cm^{-3} is the atmospheric density at $T_0 = 296$ K, and we have used the high-temperature approximation for the rotational partition function.

cannot rule out a purely gas-phase origin of the observed gas-phase CH_4. In fact, either a young or a chemically evolved molecular cloud may be consistent with the observations. More definite conclusions will require observations of additional lines to resolve questions about saturation and temperature.

A more stringent test of the ability of purely gas-phase chemical models to explain the abundance of CH_4 is possible using the solid-phase CH_4 observations. If it is assumed that the CH_4 seen in the solid phase was made by gas-phase reactions and frozen onto grains, the total (gas plus solid) CH_4 abundance can never exceed its peak gas-phase value ($\approx 10^{-2}$ of CO). NGC 7538 IRS 1 and IRS 9 (Table 3) just reach this limit, and W33 A exceeds it if the broad 7.6 μm feature is due to solid CH_4. Although we cannot yet rule out CH_4 formation through purely gas-phase processes, efficient removal of CH_4 from the gas before it can be destroyed would be required, suggesting that some fraction of the CH_4 was formed on grains. d'Hendecourt et al. (1985) and Brown et al. (1988) model the chemistry of molecular clouds including gas-phase reactions, freeze-out of molecules onto grains, and a few grain-surface reactions. The grain-mantle CH_4 in these models is formed predominantly by solid-phase reactions. Both models predict a grain-mantle CH_4 abundance slightly larger than that of CO when cycling of molecules between the solid and gas phases is neglected, consistent with our results. d'Hendecourt et al. also consider the effect of continuous cycling of molecules between the gas and solid phases and find much lower CH_4 abundances due to destruction of CH_4 in the gas.

Although gas-phase reactions may be able to explain our observations of gaseous CH_4 and either gas-phase or grain-surface reactions may be able to explain our solid-phase CH_4 observations, a problem remains when the two sets of observations are considered together. One would expect that if the CH_4-rich grain mantles are heated sufficiently by a nearby star to evaporate the CH_4, a large gas-phase abundance of CH_4 ($\approx 10^{-2}$ of CO) should be observed. Brown et al. discuss this process, and Blake et al. (1987) suggest that a similar model can explain their observations of peculiar molecular abundances near OMC-1 IRc2. However, we observe low gas-phase abundances of CH_4 ($\approx 10^{-3}$ of CO) toward all of the sources we observe, including NGC 7538 IRS 1 and probably W33 A and OMC-1 IRc2, all of which have substantial amounts of warm gaseous CO, indicating that grains should have been heated enough to release both CO and CH_4 from their mantles. It may be that the CH_4 is destroyed by gas-phase reactions soon after being evaporated from the grains. Although we may have underestimated the abundance of gas-phase CH_4 toward W33 A and OMC-1 IRc2 if the $R(0)$ line is saturated toward these sources, it is hard to see how a significant $R(0)$ line could have escaped detection toward NGC 7538 IRS 1. We plan to observe additional lines of CH_4, and perhaps $^{13}CH_4$, to resolve this question.

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