

*Letter to the Editor***Detection of doubly deuterated formaldehyde towards the low-luminosity protostar IRAS 16293–2422**C. Ceccarelli¹, A. Castets¹, L. Loinard^{1,2}, E. Caux³, and A.G.G.M. Tielens⁴¹ Laboratoire d'Astrophysique, Observatoire de Grenoble – B.P. 53, F-38041 Grenoble cedex 09, France² Institut de Radio Astronomie Millimétrique, 300 rue de la Piscine, F-38406 St.-Martin-d'Hères, France³ CESR CNRS-UPS, B.P. 4346, F-31028 Toulouse cedex 04, France⁴ SRON, P.O. Box 800, 9700 AV Groningen, The Netherlands

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Abstract. We report the detection of the doubly deuterated form of formaldehyde towards IRAS 16293-2422, an extremely young protobinary system. This is the only detection of D₂CO in any astronomical source other than Orion (Turner 1990). The line searched was that corresponding to the 4_{2,2} – 3_{2,1} transition at $\nu_0 = 236.102$ GHz, and was detected at three positions, one centered on the source itself, and the other two 10'' West and 10'' East respectively. A survey of the literature revealed that two additional lines of D₂CO at 245.532 and 342.522 GHz were detected towards IRAS 16293-2422, but classified as “unidentified”. We failed to detect any signal towards the outflow (at offsets (–20'';0'') and (–30'';+10'')) where low-J CO emission lines are quite strong. The line profiles at the three positions show a dip at the zero rest velocity. We rule out the possibility that this dip may be due to a pure kinematic effect and show that it is actually due to *self absorption*. The gas responsible for the D₂CO emission is located at less than about 10'' ($\sim 2 \cdot 10^{16}$ cm) from the central source.

We used multi-transition H₂CO observations of van Dishoeck et al. (1995) to estimate the density ($n_{H_2} \sim 10^7$ cm^{–3}) and temperature ($T \sim 100$ K) of the emitting gas. From those values, we can derive a lower limit to the column density required for the D₂CO line we observed to be self-absorbed: $N(D_2CO) \sim 10^{14}$ cm^{–2}. Combining the previous and present observations of H₂CO, HDCO and D₂CO we obtain the following upper limits to the abundance ratios of the three species: $[HDCO] / [H_2CO] \leq 0.25$ and $[D_2CO]/[HDCO] \leq 0.5$. If the abundance of H₂CO in IRAS16293-2422 is comparable to that observed towards the Hot Cores in Orion ($\sim 10^{-8}$; Blake et al. 1987) and if the D₂CO 4_{2,2} – 3_{2,1} line is only moderately thick, D₂CO is only 1/10 less abundant than H₂CO. We finally speculate that such a large amount of warm D₂CO can only be produced by evaporation of this molecule from the grain mantles, where it was presumably copiously formed during the long pre-collapse period.

Key words: ISM: abundances – ISM: individual: IRAS 16293–2422 – ISM: molecules – stars: formation

1. Introduction

There is more and more evidence that grain surface chemistry plays a key role in the formation of several molecules in the ISM. For instance, the abundance of the doubly deuterated formaldehyde (D₂CO) towards the Hot Cores in Orion cannot be understood in terms of pure gas-phase chemistry (Turner 1990) but can be adequately explained if deuterated molecules form on grain surfaces and are subsequently evaporated into the gas phase (Tielens 1983).

The study of deuterated molecules around protostars provides information both on the *pre-collapse* and the *collapse* phases. Indeed, the D/H fractionation ratio depends on the D/H ratio at the moment of the mantle formation during the pre-collapse phase (we note that the gas phase composition retains this memory during the first 10⁵ years after mantle ejection into the gas phase – Charnley, Tielens & Millar 1992), while the release of the deuterated molecules into the gas phase occurs during the collapse phase, either through the heating of the grains by the central source, or through sputtering of icy grain mantles in a shock.

The evolutionary time scales of low-mass protostars are believed to be much longer than that of high-mass protostars such as the Hot Cores in Orion, and the D/H fractionation ratio might consequently be quite different in those two kinds of sources. Hence, we searched for D₂CO in IRAS 16293-2422 (hereafter IRAS16293), a very young, low luminosity ($\sim 30L_{\odot}$) protostar, probably in a very early stage of its evolution (e.g. Bontemps et al. 1996 and Saraceno et al. 1996). IRAS16293 is located at a distance of ~ 160 pc in an isolated, cold molecular cloud core within the ρ Ophiuchus complex (Wootten 1989). Since its discovery, IRAS16293 has been the target of numerous observational studies which revealed the presence of several components (see the review by van Dishoeck et al. 1995; hereafter

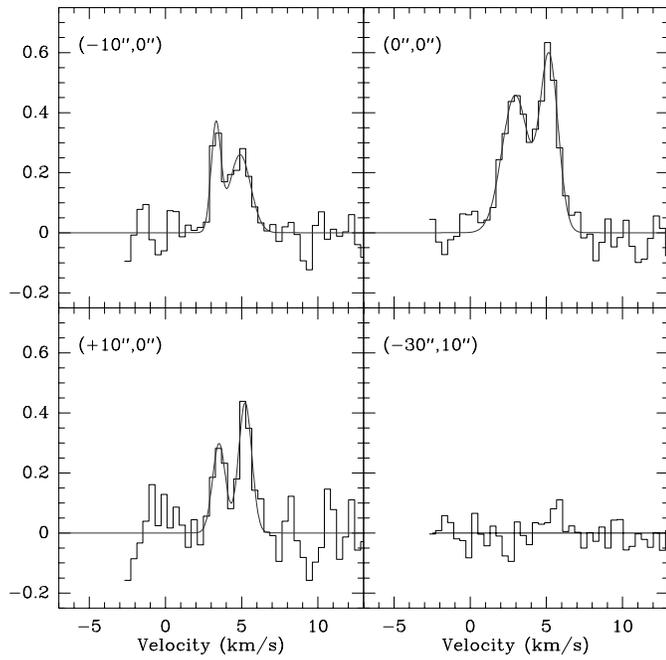


Fig. 1. $4_{2,2} - 3_{2,1}$ D_2CO spectra obtained towards IRAS16293 at offsets $(-10''; 0'')$, $(0''; 0'')$ and $(+10''; 0'')$ from the nominal center at $\alpha(2000) = 16^h 32^m 22^s.6$, $\delta(2000) = -24^\circ 28' 33''$. The spectra have been smoothed to a velocity resolution of 0.4 km s^{-1} to improve the signal-to-noise ratio. The intensities are expressed in units of main beam brightness temperature T_{mb} ; the velocities are relative to LSR. The best two-component gaussian fit is shown in each case.

vDBJG95); an ambient molecular core, a rotating and infalling envelope, an outer cold disk, two warm circumstellar disks and finally a multiple outflow. More recently, several tracers of energetic shocks such as [OI]63 μm fine structure line, high J CO transitions, and H_2O , OH and CO^+ rotational lines were detected in the far infrared (Ceccarelli et al. 1997, 1998a, 1998b).

From a chemical point of view IRAS16293 is a benchmark. It seems to be one of the most active regions ever observed. Blake et al. (1994) and vDBJG95 carried out spectral surveys in the 239–250 GHz and 338–347 GHz bands, detecting around 300 lines from 25 different chemical species. One remarkable result of those surveys was the discovery of fairly high degrees of deuteration of the molecules, a fact that lead us to choose this source for our search of D_2CO .

2. Observations and results

The observations reported here were collected in February 1998 with the IRAM 30-m telescope located on top of Pico Veleta near Granada (Spain). The line observed was that corresponding to the $4_{2,2} - 3_{2,1}$ transition of D_2CO at 236.102 GHz. Five positions were observed at offsets $(+10''; 0'')$, $(0''; 0'')$, $(-10''; 0'')$, $(-20''; 0'')$ and $(-30''; +10'')$ from the nominal position of IRAS16293 at $\alpha(2000) = 16^h 32^m 22^s.6$, $\delta(2000) = -24^\circ 28' 33''$. The receiver was connected to an autocorrelator, yielding a spectral resolution of $\sim 78 \text{ kHz}$ for a total bandwidth of 700 MHz. At the frequency considered, this corresponds to a velocity res-

Table 1. Parameters of the three lines of D_2CO detected towards IRAS16293. The line at $\nu = 236.102 \text{ GHz}$ corresponds to the spectra on Fig. 1, and the parameters for the two other lines are taken from Blake et al. (1994).

ν (GHz)	Transition	E_u (cm^{-1})	$\int T_{mb} dv$ (K km s^{-1})	T_{mb} (K)	Offset
236.102	o- $4_{22} - 3_{21}$	34.6	1.86	0.53	$(0'', 0'')$
			0.73	0.33	$(-10'', 0'')$
			0.79	0.38	$(+10'', 0'')$
245.532	p- $4_{13} - 3_{12}$	24.2	0.58	0.27	$(0'', 0'')$
342.522	o- $6_{06} - 5_{05}$	40.4	1.52	0.37	$(0'', 0'')$

olution of 0.1 km s^{-1} , and a total coverage of 90 km s^{-1} . The rejection level of the receiver was always better than 15 dB and checked against standard calibration sources. The observations were made by switching alternatively between the source and two OFF positions $3'$ East and West from the source ($\Delta\alpha = \pm 3'$, $\Delta\delta = 0$). A H_2CO map of the region (Loinard et al., in prep) shows that those positions are free of emission. The system temperature was about 800K during the observing; the total (ON+OFF) integration time of 45 minutes spent on each position yielded an r.m.s. noise level of 80 mK (T_{mb}) per channel. Pointing and focus were monitored regularly; pointing corrections were always less than $3''$. At the frequency considered, the angular resolution of the telescope is $11''$.

While the spectra at offset $(-20''; 0'')$ and $(-30''; +10'')$ yielded only upper limits of $\sim 0.1 \text{ K}$ (3σ), the spectra at offsets $(-10''; 0'')$, $(0''; 0'')$ and $(+10''; 0'')$ show very good signal-to-noise ratios, double-peaked profiles (Fig. 1). Although this is the first reported detection of D_2CO in any astronomical source other than Orion, two other lines of D_2CO were detected towards IRAS16293 during the spectral survey carried out by Blake et al. (1994) in the 239–250 and 338–347 bands, but classified as unidentified. Those were the $6_{0,6} - 5_{0,5}$ transition at 342.522 GHz. The sideband of one other unidentified line was not established but we surmise that it concerns the $4_{1,3} - 3_{1,2}$ transition at 245.532 GHz. Incidentally, those are the lines with the highest spontaneous emission coefficients in the intervals covered¹. The parameters of the three lines deduced from our spectra and taken from Blake et al. (1994) are listed in Table 1.

3. Discussion

3.1. Origin of the $4_{22} - 3_{21}$ line emission

The non-detection at offset $(-30''; +10'')$, where low-J CO emission lines due to the outflow have a strong peak (e.g. Walker et al. 1993), indicates that the line does not originate in the outflow. That the two peaks detected in our profiles originate in two distinct components along the line of sight can be ruled out, since it was not observed in any previous observation of IRAS16293 (see e.g. vDBJG95). The double peak could *a priori* result from

¹ Here, and in all what follows, the molecular data are taken from the JPL catalog (Pointer & Pickett 1992).

purely kinematical effects in a rotating disk, with the blue peak coming from the “approaching” side and the red peak coming from the “receding” side. However, the fact that the intensity of the two peaks (the red and the blue peak respectively) at the two lateral positions is practically the same, while only a factor two lower than that at the central position, is incompatible with a geometry of a pure rotating disk. The relatively constant signal of the two peaks at the lateral positions would imply a *very small radius*, much smaller than the $11''$ beamsize, of such an hypothetical disk (which would only be picked up by the edge of the beam at the $10''$ East and West observations). This is in contrast with the factor of two between the emission at the central and lateral positions which implies a *rather large radius*, comparable to the beamsize, of the disk. We therefore conclude that the origin of the double-peaked structure is not purely kinematical, but is due to *self absorption*. Finally the relative signal in the three positions indicates that, if the emission is uniformly distributed, it originates in a region of $\sim 20''$ in diameter, i.e. from a region closer to the central source than $\sim 2 \cdot 10^{16}$ cm.

3.2. D_2CO column density

Several transitions from H_2CO were previously observed towards IRAS16293 by Mangum & Wootten (1993) and vDBJG95. By considering the higher lying lines (whose upper level energy and line width are comparable with the D_2CO $4_{2,2} - 3_{2,1}$ line), vDBJG95 derive a density as high as 10^7 cm^{-3} and a temperature of ~ 100 K. At this density the H_2CO lines are likely to be thermalized (Mangum & Wootten 1993). Assuming that the $4_{2,2} - 3_{2,1}$ line originates approximatively in the same region and that LTE distribution is also valid for D_2CO , we can estimate the D_2CO column density required for the line to be self-absorbed in its central 0.5 km s^{-1} , via the relation:

$$N_x \geq \frac{8\pi \delta\nu}{A\lambda^3} [\exp[(E_u - E_l)/kT] - 1]^{-1} \times \left[\frac{g_u \exp[-E_u/kT]}{Q_T} \right]^{-1} \quad (1)$$

where $\delta\nu$ is the dip width, λ the wavelength, A the spontaneous emission coefficient, E_l and E_u the lower and higher energy level, g_u the statistical weight of the upper level of the transition $4_{2,2} - 3_{2,1}$ and Q_T the partition function. Fig. 2 shows the value of the column density required to have the line opacity equal to unity as a function of the temperature. Although the bulk of the $4_{2,2} - 3_{2,1}$ line emission originates in a gas at about 100 K, the absorption could be due to a more extended and colder layer of material. Hence, a lower limit to the D_2CO column density of 10^{14} cm^{-2} seems appropriate.

3.3. Relative abundances of D_2CO , $HDCO$ and H_2CO

In our analysis of the emission, we assume LTE conditions and *low optical depths*. The relative abundances of the various isotopes of formaldehyde can then be determined from multi-transition observations, using rotational diagrams. The column density N_x of a certain species can be expressed as:

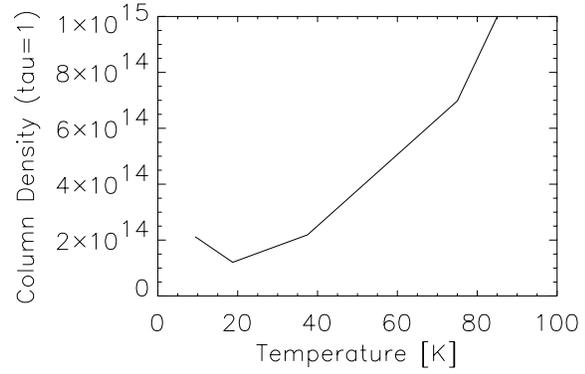


Fig. 2. Column density of D_2CO required to have the optical depth of the $4_{22} - 3_{21}$ line equal to unity, assuming LTE conditions and an absorption over the central 0.5 km s^{-1} interval.

$$\ln Y = \ln \left[\frac{I_\nu 3c^3}{64\pi^4 \nu^4 \mu^2 S_j} \right] = \ln \left[\frac{N_x}{Q_T} \right] - \frac{E_u}{kT} \quad (2)$$

where I_ν is the line intensity, while ν , μ and S_j are the transition frequency, dipole moment and line strength respectively. The abundance ratios are then derived from the ratios of the ordinates on the rotational diagrams of the three species respectively, divided by the rotational partition functions.

However, we *did show* in §3.1 that the D_2CO $4_{2,2} - 3_{2,1}$ line is *self absorbed* due to the colder foreground gas (cf. vDBJG95); the profile of the $HDCO$ $5_{1,5} - 4_{1,4}$ line, whose higher level energy is comparable to the D_2CO $4_{2,2} - 3_{2,1}$ line and which has been observed by vDBJG95 (their Fig. 18), also shows self absorption. We infer that the H_2CO high lying lines are also self absorbed, but that the relatively low spectral resolution ($\sim 1.5 \text{ km s}^{-1}$) of the observations obtained by vDBJG95 precluded its recognition. Because of self absorption, the rotational diagram method will only yield *upper limits* to the $[HDCO]/[H_2CO]$ and $[D_2CO]/[HDCO]$ ratios (we implicitly assumed $[H_2CO] \geq [HDCO] \geq [D_2CO]$, and took the ortho-to-para ratio equal to 3:1 for both H_2CO and D_2CO). Fig. 3 shows the rotational diagrams of H_2CO , $HDCO$ and D_2CO respectively. The $HDCO$ and H_2CO diagrams were built using the data from vDBJG95, while the D_2CO diagram includes our detection as well as the data from Blake et al. (1994). Our analysis leads to the following values for the relative abundances: $[HDCO]/[H_2CO] \leq 0.25$ and $[D_2CO]/[HDCO] \leq 0.5$.

3.4. Implications

Combining our lower limit to the D_2CO column density and upper limit to the $[D_2CO]/[H_2CO]$ ratio (≤ 0.1), we find that the lower limit to the H_2CO column density is 10^{15} cm^{-2} . This estimate is about five times larger than that quoted by vDBJG95 (obtained *assuming* that the H_2CO lines were *not* self absorbed), but remains compatible with their upper limits to the $H_2^{13}CO$ high lying lines. If the H_2CO emission originates in a region whose extent is similar to that of D_2CO ($\sim 10^{16}$ cm), and whose density is $\sim 10^7 \text{ cm}^{-3}$, then the abundance of H_2CO is larger

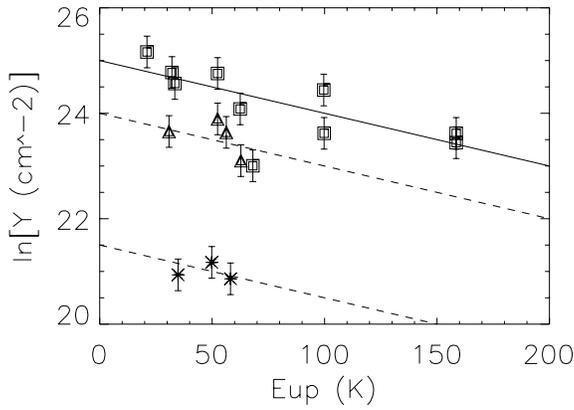


Fig. 3. Rotational diagrams for H_2CO , HDCO and D_2CO . The squares represent the H_2CO transitions, the triangles the HDCO and the crosses the D_2CO transitions. The error bars represent 30% of the signal of each line. The solid line corresponds to a rotational temperature of 100K. The dashed lines are the best fits of the HDCO and D_2CO lines with the same rotational temperature.

than 10^{-8} , a value comparable to what observed towards the Hot Cores in Orion (Blake et al. 1987).

This simple analysis implies that the fraction of D_2CO compared to H_2CO is fairly large. We note that lowering this ratio would require to increase the H_2CO abundance by the same factor. Either way, we face unusually high values of the two quantities, higher than those observed towards Orion, where the $[\text{D}_2\text{CO}]/[\text{H}_2\text{CO}]$ ratio is only 0.003 (Turner 1990). Turner (1990) claimed that the relatively high $[\text{D}_2\text{CO}]/[\text{HDCO}]$ ratio (~ 0.02) observed towards Orion was a sign of “active grain surface chemistry”, where these species are formed through reactions of H and D atoms with CO on grain surfaces. Our findings of $[\text{D}_2\text{CO}]/[\text{HDCO}]$ ratios up to 25 times higher than their value in Orion seems to indicate not only deuterium fractionation caused by grain chemistry, but also rather extreme conditions. The deuteration on a grain surface is set by the atomic D over atomic H ratio in the gas accreting onto the mantles. To get an $[\text{D}_2\text{CO}]/[\text{H}_2\text{CO}]$ ratio around 0.1 requires a very high $[\text{D}]/[\text{H}]$ ratio in the gas phase. If we consider that atomic H has an approximately constant density of $\sim 1 \text{ cm}^{-3}$, while a substantial fraction of Deuterium is locked up in atomic D (Tielens 1983), then a density in excess of 10^5 cm^{-3} is required to get such a $[\text{D}]/[\text{H}]$ ratio. In contrast, the formaldehyde released from grain mantles in Orion probably formed at somewhat lower densities ($\sim 10^4 \text{ cm}^{-3}$). The emerging picture is that D_2CO was formed on the grain surfaces during the very first and cold phases of the collapse, when the parental clump started to contract becoming denser, and successively D_2CO was desorbed into the gas phase either because of heating of the dust by the central accreting source or because of an intervening shock. Furthermore, both

the high $[\text{D}_2\text{CO}]/[\text{H}_2\text{CO}]$ ratio and high H_2CO abundance testify for the youth of the source. Modeling of the time needed to destroy both D_2CO and H_2CO suggests a time $\leq 10^5$ years since the desorption occurred (Charnley et al. 1992; Rodgers & Millar 1996).

4. Conclusions

We observed the $4_{2,2} - 3_{2,1}$ (236.102 GHz) transition of the doubly deuterated formaldehyde towards IRAS16293, a low-mass protostar. We detected emission from the source itself, and from lateral positions $10''$ East and West respectively, while we failed to detect any D_2CO emission at two positions along the outflow. The line is self-absorbed at all three positions which implies a lower limit to the D_2CO column density of 10^{14} cm^{-2} .

Combining the present observations with previous observations of H_2CO and HDCO by vDBJG95, we obtain upper limits to the abundance ratios for these species of $[\text{HDCO}]/[\text{H}_2\text{CO}] \leq 0.25$ and $[\text{D}_2\text{CO}]/[\text{HDCO}] \leq 0.5$. This implies $[\text{H}_2\text{CO}] \geq 10^{-8}$ and $[\text{D}_2\text{CO}]/[\text{H}_2\text{CO}] \leq 0.1$. Such high values are clear signs of grain surface chemistry, where these species are formed during a previous, cold dark core phase. Upon heating by the newly formed star of the ice mantles in its surroundings, these species evaporate into the gas phase in a Hot Core environment.

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