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Letter to the Editor

Ground-based searches for interstellar H_2D^+

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Abstract. We present ground-based searches for the $1_{10} - 1_{11}$ line of interstellar H_2D^+ at 372 GHz which are more sensitive than those obtained from the Kuiper Airborne Observatory (KAO) by factors of 3–4 for extended sources and by more than two orders of magnitude for compact sources. The line was not detected in a variety of interstellar clouds, including NGC 2264 toward which a possible detection had been suggested previously. The inferred H_2D^+ abundance limits of $10^{-10} - 10^{-11}$ are still consistent with, but approach the abundances predicted by chemical models. Simultaneous observations of the DCO^+ 3–2 and N_2H^+ 4–3 lines have been used to place additional limits on the H_3^+ abundance, and suggest $10^{-11} < x(H_3^+) < 10^{-9}$. The N_2H^+ data also indicate that for NGC 2264, but perhaps not for the other sources, gas-phase N_2 contains a substantial fraction of the available nitrogen in the cloud.

Key words: Interstellar molecules: H_2D^+; Molecular Clouds

1. Introduction

The H_3^+ ion is thought to play a central role in the ion–molecule reaction scheme which dominates gas-phase interstellar chemistry (Herbst & Klemperer 1973; Watson 1976). The ubiquitous observations of protonated molecules such as HCO^+ and N_2H^+ testify to its presence. However, the H_3^+ ion has not yet been detected in interstellar clouds, despite sensitive searches for its infrared vibration–rotation lines in absorption against bright background sources (Geballe & Oka 1989; Black et al. 1990). In principal, the H_2D^+ isotopomer can more easily be observed by rotational spectroscopy owing to its permanent dipole moment. Moreover, at the low temperatures in interstellar clouds the reaction $H_3^+ + HD \rightleftharpoons H_2D^+ + H_2$ (1) leads to significant fractionation, so that the H_2D^+/H_3^+ ratio is much larger than the cosmic deuterium abundance of about 2×10^{-5}. Indirect evidence for the presence of H_2D^+ comes from the large observed abundances of deuterated molecules like DCO^+, DCN and N_2D^+ (e.g. Guélin, Langer & Wilson 1982; Wootten 1987).

H_2D^+ has ortho and para modifications, with the lowest 1_{11} ortho state lying 86 K above the true ground state. The ground-state $1_{01} - 0_{00}$ para-H_2D^+ line lies at 1370 GHz, and can only be observed from airborne platforms. The ground-state $1_{10} - 1_{11}$ ortho-H_2D^+ transition occurs at the lower frequency of 372.421 GHz (Bogey et al. 1984), but lies in an unfavorable part of the spectrum since it is close to the strong 380 GHz atmospheric water line. Observations of this transition have therefore previously been attempted only from the KAO (Phillips et al. 1985; Pagani et al. 1992a). However, under exceptional conditions, the line is also accessible from high ground sites. With the availability of the new large submillimeter telescopes and improved high-frequency receivers, the ground-based searches can be more sensitive than those from the KAO because of longer possible integration times, much smaller beams and fewer restrictions on the choice of sources.

Additional impetus for the H_2D^+ search stems from the uncertainty regarding the detection of the ion in interstellar clouds. Phillips et al. (1985) reported a possible line at the expected H_2D^+ frequency in the dense cloud NGC 2264, and found upper limits for the cold cloud TMC–1. Black et al. (1990) subsequently searched for the H_3^+ infrared lines in absorption against NGC 2264 IRS, but failed to detect the ion. More recently, Pagani et al. (1992a) obtained upper limits for the H_2D^+ 372 GHz line from the KAO in NGC 2264 and several other sources, which are comparable to the strength of the line reported by Phillips et al. We report here the results of ground-based searches for the 372 GHz line which are factors of 3–4 more sensitive than those obtained from the KAO for extended sources, and more than a factor 100 for small sources (<20″). We do not confirm the possible feature in NGC 2264, and obtain significant upper limits for other sources.

2. Observations

The H_2D^+ observations at 372 GHz were performed with the Caltech Submillimeter Observatory (CSO) on December 9, 1991 during a night of exceptional submillimeter transparency. The optical depth at 225 GHz was 0.02–0.03, corresponding to a transmission at 372 GHz of 50–70%. The lead alloy SIS junction receiver built by Ellison et al. (1989) was used in conjunction with a 1024 channel, 500 MHz bandwidth acousto-optical spectrometer (AOS) as the backend. The H_2D^+ line was centered in the upper sideband, and typical system temperatures including atmospheric losses were 2000 K. The calibration was performed using the ambient chopper method. A correction

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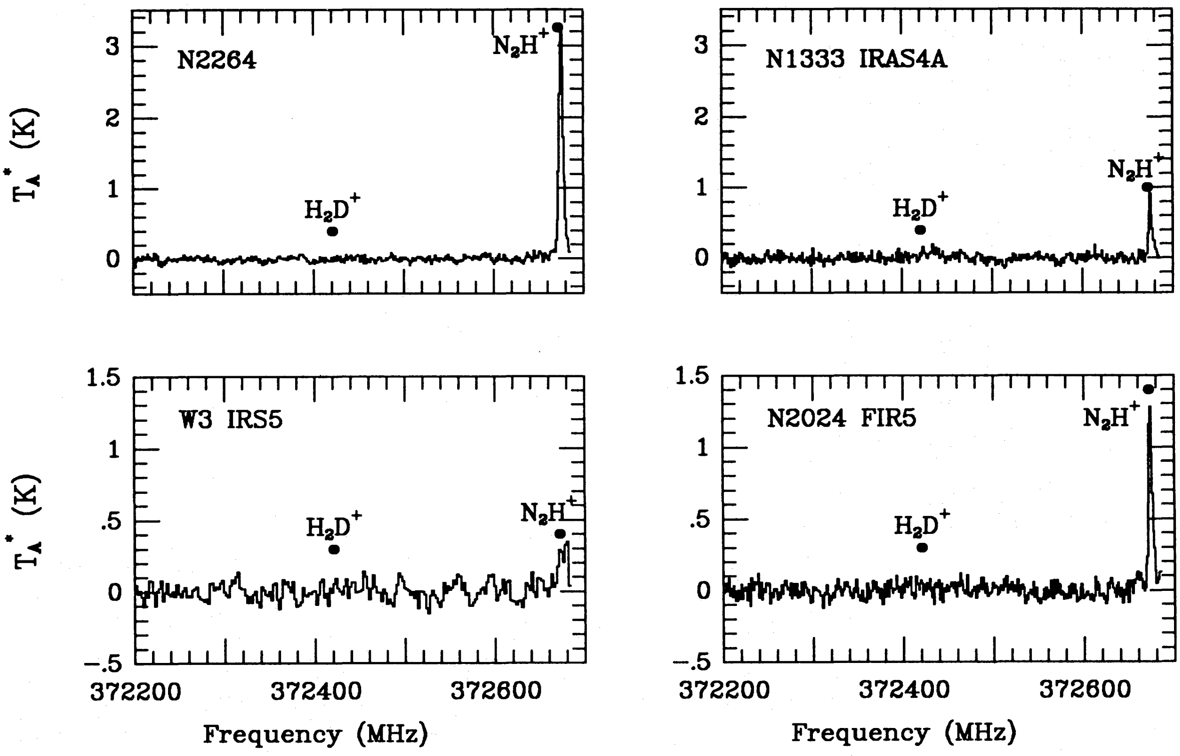

Fig. 1. Observed spectra at 372 GHz toward four of the five sources. The spectra toward NGC 2264, NGC 1333 IRAS 4A and NGC 2024 FIR5 have been smoothed to 1 MHz resolution, and that toward W3 IRS5 to 2 MHz resolution.

for unequal sideband atmospheric transmission of <10% was applied by means of an atmospheric model which has been adjusted over a two year period to best fit the Mauna Kea atmosphere. The CSO beam size at 372 GHz is about 18″, significantly smaller than the KAO beam of 3.′7. The main beam efficiency is about 55%, whereas the efficiency for extended sources is about 70%. The latter value has been used in the analysis. The overall uncertainty in the calibration is estimated to be of order 30%. The observed sources are summarized in Table 1. The position in NGC 1333 labelled WLS corresponds to the peak in the DCO^+ 1–0 map of Wootten et al. (1982).

The observed spectra at 372 GHz, presented in Figure 1, are summarized numerically in Table 1. It is clear that none of the sources shows a significant feature at the frequency of the H_2D^+ line. The 2σ limit for NGC 2264 obtained after 45 min on–source integration is about 90 mK when corrected for beam efficiency, which is substantially below the possible feature of $T_A^* \approx$230 mK by Phillips et al. (1985). Similar 2σ limits are found for the other sources; they are typically a factor 3 lower than the limits obtained by Pagani et al. (1992a). Since the KAO observations refer to a much larger beam over which the conditions necessary to excite the H_2D^+ line may not exist, the current ground–based limits are effectively even more sensitive by up to two orders of magnitude.

At the edge of the spectra, a strong line with $T_A^* \approx$1–3 K is seen for most of the sources. It can be identified with N_2H^+ 4–3, previously seen only by Pagani et al. (1992a) in NGC 6334. Because the effective resolution of the AOS was only 3 km s^{-1} at the time of observations, not all lines are resolved and only integrated antenna temperatures are given in Table 1. The presence of the N_2H^+ line in the H_2D^+ search band is fortunate, since it provides important indirect constraints on the H_3^+ abundance in the clouds, as well as the excitation conditions. For completeness, the DCO^+ 3–2 line at 217 GHz was observed using the sensitive new SIS receiver built by Kooi et al. (1992). The telescope beam size at this frequency is about 34″ and the efficiency for extended sources about 77%.

3. Analysis

3.1. Excitation model

The derivation of column densities from observed line strengths requires information about the excitation conditions. The upper levels of the H_2D^+ $1_{10} - 1_{11}$, N_2H^+ 4–3 and DCO^+ 3–2 transitions lie at 104, 45 and 20 K, whereas the critical densities are about 2×10^5, 10^7 and 5×10^6 cm^{-3}, respectively. Thus, high densities ($n > 10^5$ cm^{-3}) and not–too–low temperatures are required to excite the lines, even though the abundance of the H_2D^+ ion is greatly enhanced at T <20 K. The physical conditions in W3 IRS5 and NGC 1333 IRAS 4A have been constrained from observations of high–excitation lines of other species such as H_2CO, HCO^+ and CS in a similar beam (Phillips et al. 1992; Blake et al. 1992). The temperature in the protostellar NGC 1333 IRAS 4A region is somewhat uncertain, but is unlikely to be much different from the dust temperature of 37 K throughout the source (Sandell et al. 1991). Because of the lack of lines at the NGC 1333 WLS position, it will not be considered any further. The conditions in NGC 2264 and NGC 2024 are based on literature data (Black et al. 1990;

Table 1. Observations

Source	V_{LSR} (km s^{-1})	T_A^* (K)	$\int T_A^* dV$ (K km s^{-1})	$\int T_A^* dV$
		H_2D^+ $1_{10}-1_{11}$	N_2H^+ 4–3	DCO^+ 3–2
W3 IRS5	−39.0	≤0.09[a]	1.8	≤0.3
NGC 1333 IRAS 4A .	+6.0	≤0.11[b]	3.1	1.7
NGC 1333 WLS	+7.5	≤0.15[b]	1.0	≤0.1
NGC 2024 FIR5 15″S	+10.2	≤0.07[c]	5.1	≤0.2
NGC 2264 IRS	+6.5	≤0.06[d]	15.0	0.9

Note: All limits are 2σ for the indicated line width.
[a] ΔV=5; [b] ΔV=1.5; [c] ΔV=2.5; [d] ΔV=3.5 km s^{-1}.
Positions (1950): W3 IRS5: α=02 21 53.3, δ=61 52 21; NGC 1333 IRAS4A: 03 26 04.8, 31 03 14; WLS: 03 25 53.0, 31 10 30; NGC 2024: 05 39 12.8, –01 57 19; NGC 2264: 06 38 25.0, 09 32 29.

Mauersberger et al. 1992), together with unpublished H_2CO spectra.

The adopted H_2D^+ model has been described by Black et al. (1990). In brief, the excitation *within* each of the ortho– and para–ladders is controlled by radiative transitions and inelastic collisions with H_2, whereas transitions *between* the two ladders can only occur through reactive collisions with H_2. The latter collisions are rapid enough that the ortho ladder cannot be considered separately, but has the 0_{00} para level as its effective ground state. A constant de–excitation rate coefficient of 5×10^{-10} cm^3 s^{-1} was used for all inter–ladder transitions. For sufficiently high densities, this model approaches the "thermal" model of Phillips et al. and the LTE model of Pagani et al. with $T_{ex} = T$. At the high densities considered here, excitation by far–infrared radiation does not influence the results for the 372 GHz line, although it affects the 1370 GHz line.

3.2. Column densities and abundances

The inferred column densities are summarized in Table 2. The most sensitive H_2D^+ limits are about $5 \times 10^{12} - 1 \times 10^{13}$ cm^{-2}. Determination of the H_2D^+ abundance requires knowledge of the H_2 column densities along the lines of sight, which are highly uncertain. We adopt the values listed in Table 2. Except for NGC 1333, these have been derived from $C^{18}O$ 2–1 measurements and refer to a similar beam of 20–30″ (Phillips et al. 1992). The H_2 column density toward NGC 2264 is constrained to better than 50% (Black et al. 1990). On the other hand, that toward NGC 1333 IRAS 4A is based on submillimeter continuum measurements of the dust (Sandell et al. 1991), and is uncertain by a factor of two due to uncertainties in the temperature and the emissivity of the grains. Using this $N(H_2)$, the $C^{18}O$ data give a ^{12}CO abundance of only a few times 10^{-6}, suggesting that CO (but not H_3^+ or H_2D^+) may be significantly depleted onto the grains in the bulk of the cloud (Blake et al. 1992). A similar situation may apply to NGC 2024 FIR5 (Mezger et al. 1992).

The derived upper limits on the H_2D^+ abundance are typically $10^{-11} - 10^{-10}$, with factors of a few uncertainty. Such limits are still just consistent with current chemical models, which predict $x(H_2D^+) \approx 10^{-10}$ for $T \approx 30$ K, decreasing to 10^{-11} for $T \approx 50$ K (Watson 1976; Millar et al. 1989; Pagani et al. 1992b). The limits on the H_3^+ abundance are typically $10^{-9} - 10^{-8}$ if the fractionation scheme of Pagani et al. is used with their standard parameters. However, the models usually refer to a cloud with a density of only 10^4 cm^{-3} in which the CO abundance is normal, about 10^{-4}. Since the H_3^+ abundance is inversely proportional to density, the model predictions for H_2D^+ could be as low as 10^{-12} at $T \approx 30$ K for densities of 10^6 cm^{-3} or larger. On the other hand, H_3^+ is significantly enhanced if CO is depleted, since reaction with this species is the principal removal mechanism. The enhancement for H_2D^+ may be even stronger if other heavy species such as O_2, N_2, and O are depleted as well, because reaction (1) then becomes the main destruction path of H_3^+. For regions such as NGC 1333 IRAS 4A and NGC 2024 FIR5, the effects of high density and low CO abundance may therefore cancel, and the H_2D^+ abundance may still be of order 10^{-11} or more.

A second method to infer $x(H_3^+)$ is to combine the H_2D^+ limits with the DCO^+ data. The derived DCO^+ column densities toward NGC 2264 and NGC 1333 IRAS 4A, together with unpublished $H^{13}CO^+$ data, imply $DCO^+/HCO^+ \approx 0.01$–0.02 and $DCO^+/H_2D^+ > 0.1$ and 0.2. In the original deuteration scheme of Watson, a simple relation $DCO^+/HCO^+ \approx \frac{1}{3} H_2D^+/H_3^+$, or equivalently $DCO^+/H_2D^+ \approx \frac{1}{3} HCO^+/H_3^+$, is found. Although the realization that atomic deuterium may be abundant inside dense clouds alters this argument somewhat (Dalgarno & Lepp 1984), the models of Millar et al. (1989) still indicate steady state ratios close to $\frac{1}{3}$. In that case, the observed DCO^+/H_2D^+ limits imply $HCO^+/H_3^+ > 0.25$ and 0.55, to be compared with model predictions of $HCO^+/H_3^+ \approx 10$ at high densities. The HCO^+ abundances derived from $H^{13}CO^+$ assuming an isotopic ratio of 50 are approximately 2×10^{-9} in NGC 2264 and 2×10^{-10} in NGC 1333, respectively, resulting in upper limits on the H_3^+ abundance of about 7×10^{-9} and 3×10^{-10}. For comparison, the limit inferred by Black et al. (1990) toward NGC 2264 is $< 1.4 \times 10^{-8}$. The DCO^+ limits toward W3 IRS5 and NGC 2024 FIR5 are significantly lower than those for H_2D^+, and indicate that these regions are not good places to search for H_2D^+.

Finally, information on the H_3^+ abundance can be derived from the N_2H^+ lines. The inferred N_2H^+ column densities range from $10^{12} - 10^{14}$ cm^{-2} and the abundances from $10^{-11} - 10^{-9}$. Thus, significant variations in the N_2H^+ abundance occur from cloud to cloud. N_2H^+ is formed mostly by the reaction of N_2 with H_3^+ and is destroyed by reactions with CO, O and electrons. If we assume that at least 50% of the gas–phase nitrogen is in the form of N_2 with $x(N_2) \approx 5 \times 10^{-5}$ and $x(CO) \approx 10^{-4}$, then a simple chemical scheme suggests $x(H_3^+) \approx x(N_2H^+)$. For lower N_2 abundances, the H_3^+ abundances will be higher. For NGC 2264, this leads to lower limits on the H_3^+ abundance of $4 \times 10^{-10} - 3 \times 10^{-9}$, which are barely consistent with the upper limits derived above.

The three methods used to derive the H_3^+ abundances are summarized in the last few columns of Table 2. Each has its advantages and disadvantages. The first method assumes that the H_2D^+ fractionation is well understood, and depends mostly on the adopted temperature. The second method requires additional independent data, and assumes that HCO^+ and H_3^+ are similarly distributed, which may not be valid if CO is heterogeneously depleted. The last method involves the most assumptions, and is very sensitive to the density adopted in the N_2H^+ analysis. In all methods, a major uncertainty is the H_2

Table 2. Derived Column Densities and Abundances.

Source	T	n	N (cm^{-2})				$x = N(X)/N(H_2)$						
	(K)	(cm^{-3})	H_2D^+	DCO^+	N_2H^+	H_2	H_2D^+	DCO^+	N_2H^+	H_3^{+a}	H_3^{+b}	H_3^{+c}	$N_2{}^d$
W3 IRS5	50	5(6)	<7(12)	<2(11)	2(12)	2(23)	<4(-11)	<1(-12)	1(-11)	<2(-7)	...	>1(-11)	1(-5)
	100	5(6)	<5(12)	<2(11)	1(12)		<3(-11)	<1(-12)	5(-12)	<3(-7)	...	>5(-12)	5(-6)
N 1333 4A	35	1(6)	<4(12)	1(12)	1(13)	3(23)	<1(-11)	3(-12)	3(-11)	<3(-9)	<3(-10)	>3(-11)	2(-5)
	30	1(7)	<5(12)	1(12)	4(12)		<1(-11)	3(-12)	1(-11)	<3(-9)	<2(-10)	>1(-11)	6(-5)
N 2024 FIR5	50	5(5)	<3(12)	<3(11)	3(13)	2(23)	<2(-11)	<2(-12)	2(-10)	<1(-7)	...	>2(-10)	3(-5)
	30	5(6)	<6(12)	<1(11)	7(12)		<3(-11)	<5(-13)	4(-11)	<4(-9)	...	>4(-11)	4(-5)
N 2264 IRS	30	5(5)	<1(13)	2(12)	2(14)	8(22)	<1(-10)	3(-11)	3(-9)	<1(-8)	<3(-9)	>3(-9)	4(-4)
	30	5(6)	<9(12)	8(11)	3(13)		<1(-10)	1(-11)	4(-10)	<1(-8)	<7(-9)	>4(-10)	6(-4)

[a] From H_2D^+ assuming the theoretical H_3^+/H_2D^+ ratio computed by Pagani et al. (1992*b*) at the appropriate T for their standard model.
[b] From the $DCO^+/H^{13}CO^+$ analysis (see text).
[c] From the N_2H^+ analysis, assuming most gas-phase nitrogen is in the form of N_2.
[d] From N_2H^+ using model N_2/N_2H^+ ratios with $\zeta_o = 5 \times 10^{-17}$ s^{-1} at the appropriate density.

column density, especially for NGC 1333 and NGC 2024. For the best case, NGC 2264, it is seen that the three methods are only consistent if the density is higher than 5×10^5 cm^{-3}. For the other sources, a large range of values is still possible.

Alternatively, the N_2H^+ abundance can be used to infer the gas-phase N_2 abundance, if the CO and H_3^+ abundances are assumed to be standard. Chemical models give $N_2/N_2H^+ \approx 4 \times 10^4 - 10^5$ (Herbst et al. 1977; Millar et al. 1991), independent of time but linearly dependent on density and inversely proportional to the cosmic ray ionization rate ζ_o. The resulting N_2 abundances are included in Table 2 and range from 10^{-5} in W3 IRS5 to $> 10^{-4}$ in NGC 2264. Although they are not more accurate than an order of magnitude, it appears indeed that most of the available nitrogen is locked up in gas-phase N_2 in NGC 2264. N_2 may, however, contain only 10% or less of the nitrogen in W3 IRS5. The nitrogen could be either in the form of gas-phase atomic N in this cloud, or be depleted onto grains in some solid form, presumably solid N_2.

4. Conclusions

The observations presented in this letter show that sensitive ground-based searches for the H_2D^+ ion are possible under exceptional weather conditions. The derived limits are up to two orders of magnitude more sensitive than those obtained previously from the KAO. The inferred H_2D^+ and H_3^+ abundances are still consistent with chemical models; however, several indirect lines of argument based on DCO^+ and N_2H^+ observations suggest that at least for NGC 2264, only a factor of a few improvement in S/N is required to detect the ion. Dense clouds with temperatures in the range 20–30 K remain the best sources to search for the H_2D^+ 372 GHz line, both in terms of abundance and excitation.

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