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AN INFRARED SPECTROSCOPIC SEARCH FOR THE MOLECULAR ION H_3^+

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ABSTRACT

A search for infrared vibration-rotation lines of H_3^+ at 4 μm toward five obscured infrared objects has produced upper limits corresponding to H_3^+ column densities of $4 \times 10^{14} \sim 10^{15}$ cm^{-2}.

Subject headings: interstellar: abundances — interstellar: molecules — line identifications

I. INTRODUCTION

The molecular ion H_3^+ is thought to play a fundamental role in the chemistry of the interstellar medium (Herbst and Klemperer 1973; Watson 1973, 1976; Dalgarno and Black 1976; Suzuki 1979). It is a well-bound molecular system with a large energy of formation ($H_2 + p \rightarrow H_3^+ + 4.35$ eV) and exists abundantly ($\sim 10^{11}$ cm^{-3}) in hydrogen discharges in the laboratory. In space, it is produced in large amounts in dense molecular clouds through cosmic-ray ionization of molecular hydrogen followed by the 1.7 eV exothermic ion-molecule reaction

$$H_2 + H_2^+ \rightarrow H_3^+ + H\,, \tag{1}$$

which has a large Langevin cross section of several hundred Å^2. The H_3^+ ion thus produced is expected to play the crucial role of protonator in the interstellar medium through the proton hopping reaction

$$X + H_3^+ \rightarrow HX^+ + H_2\,. \tag{2}$$

This reaction is exothermic and very efficient for most neutral atoms and molecules X of astrophysical interest with the notable exceptions, X = He, N, Ne, and O_2 (for which proton affinities are 1.9 eV, 3.4 eV, 2.1 eV, and 4.3 eV, respectively). The H_3^+ ion thus initiates a chain of reactions which lead to a variety of complex molecules which have been observed in interstellar space (Huntress 1977; Smith 1988).

Because of its structural symmetry (equilateral triangle), H_3^+ does not possess a permanent dipole moment and hence no rotational spectrum is expected except for the very weak forbidden rotational transitions recently discussed by Pan and Oka (1986). A possible detection of a submillimeter wave emission of its deuterated species H_2D^+ has been reported by Phillips *et al.* (1985). In order to detect H_3^+ directly in the interstellar medium, the most promising method is to use the ν_2 vibration-rotation fundamental band which appears at 4 μm (Oka 1980, 1981). In this paper we report on such an attempt in the last few years. Most of the molecular properties and numerical data of H_3^+ used in this paper may be found in Oka's review on H_3^+ (1983).

II. VARIOUS ESTIMATES

The ν_2-fundamental band of H_3^+ has a relatively large vibrational transition dipole moment of 0.157 debye theoretically calculated by Carney and Porter (1974, 1976); the observed intensities of spectral lines in the laboratory are consistent with this value. The standard intensity formula (Pugh and Rao 1976) gives the peak absorption as

$$\alpha \equiv \frac{\Delta I}{I} = 1.45 \times 10^{-15} N(H_3^+) f_{JK}(T) \frac{A_{JK}}{\Delta v}\,, \tag{3}$$

where $N(H_3^+)$ is the column density of H_3^+ in cm^{-2}, $f_{JK}(T)$ is the temperature-dependent fraction of molecules in the lower rotational level of the transition, and Δv is the half-width at half-maximum of the observed spectral line in km s^{-1}. A_{JK} is the Hönl-London factor for a perpendicular ($\Delta K = \pm 1$) transition which is one-fourth of the expression given in Herzberg's book (1945). The numerical factor 1.45×10^{-15} results from the expression $(8\pi^3 v \mu^2 / 3hc)(\ln 2/\pi)^{1/2}$.

Because of the large rotational constants ($B = 43.56$ cm^{-1}, $C = 20.61$ cm^{-1}) of H_3^+, most of the H_3^+ molecules in dense molecular clouds populate the lowest ortho-level $J = 1$, $K = 0$ or the lowest para-level $J = 1$, $K = 1$ (note that the lowest $J = K = 0$ state is forbidden by the Pauli principle). The $J = 1$, $K = 1$ level (with a spin statistical weight of 1) is lower than the $J = 1$, $K = 0$ level (with a weight of 2) by 22.846 cm^{-1} ($\sim$33 K) (Watson *et al.* 1984) and has a higher population for $T < 48$ K. Unlike ortho- and para-H_2, the ortho- and para-H_3^+ equilibrate rapidly by proton hopping reactions (Oka 1981).

Out of the six vibration-rotation transitions starting from these two lowest levels, we chose the following two for our search:

$$^{r}Q(1, 0);\quad v = 1, J = 1, K = 1, l = 1 \leftarrow v = 0, J = 1, K = 0 \quad \text{at } 2529.724 \text{ cm}^{-1}$$

and

$$^{p}P(1, 1);\quad v = 1, J = 0, K = 0, l = -1 \leftarrow v = 0, J = 1, K = 1 \quad \text{at } 2457.290 \text{ cm}^{-1}\,.$$

The Hönl-London factor for these transitions are $\frac{1}{2}$ and $\frac{1}{6}$, respectively. The $^{r}Q(1, 0)$ transition is the strongest observed in the laboratory. The other strongest line $^{r}R(1, 0)$ at 2925.898 was not used because of interference by atmospheric absorption lines predicted by Traub and Stier (1976). The $^{r}Q(1, 0)$ transition is 0.2810 cm^{-1} below the weak $P(35)$ $2\nu_1 \leftarrow 0$ (telluric) transition of N_2O. The $^{p}P(1, 1)$ transition is the weakest of the four possible transitions starting from the $J = 1$,

$K = 1$ rotational level, although not by a large factor. It was chosen because it is free from atmospheric absorption, lying 0.499 cm^{-1} below the weak $P(5)\ \nu_1 + 2\nu_2{}^0 \leftarrow 0$ transition of N_2O.

If we assume LTE at 30 K and a spectral resolution Δv of 10 km s^{-1}, equation (3) gives for the two transitions

$$\alpha = 2.9 \times 10^{-17} N(H_3{}^+)$$

and

$$\alpha = 1.4 \times 10^{-17} N(H_3{}^+) ,$$

respectively. Since the minimum detectable absorption is on the order of 1%, we require for detection that the column density be on the order of 4×10^{14} cm^{-2} even for the most favorable cases. This is a rather large number in view of the fact that the observed column density of HCO^+, the most abundant observed molecular ion, in the Orion Molecular Cloud is $\sim 2 \times 10^{15}$ cm^{-2} (Rydbeck *et al.* 1981).

The abundance of $H_3{}^+$ in dense clouds has been estimated in several papers. In particular, discussions by Dalgarno and his colleagues are noteworthy (Dalgarno, Oppenheimer, and Berry 1973; de Jong, Dalgarno, and Boland 1980; Lepp, Dalgarno, and Sternberg 1987). Equating the production rate of $H_3{}^+$ by cosmic-ray ionization (followed by eq. [1]) $\zeta n(H_2)$, with the destruction rate due to the reaction of equation (2), $kn(H_3{}^+)n(X)$, we obtain the ratio of $n(H_3{}^+)/n(H_2)$

$$\frac{n(H_3{}^+)}{n(H_2)} = \frac{\zeta}{kn(X)} \sim \frac{10^{-8}}{n(X)} , \tag{4}$$

where $n(X)$ is the number density (in cm^{-3}) of species X, ζ is the cosmic ray ionization flux ($\sim 10^{-17}$ s^{-1}), and k is the Langevin rate constant ($\sim 10^{-9}$ cm^2) for the reaction of equation (2). Since CO is the most abundant neutral molecule to remove proton from $H_3{}^+$, the $n(H_3)/n(H_2)$ ratio is largest in objects in which gaseous CO is depleted. Thus de Jong, Dalgarno, and Boland (1980) and Lepp, Dalgarno, and Sternberg (1987) recommend search for $H_3{}^+$ in dense clouds with severe carbon depletion. The required $H_3{}^+$ column density of $N(H_3{}^+) \sim 10^{15}$ cm^{-2} will be realized if, for example, $n(CO) \sim 1$ cm^{-3} and $N(H_2) \gtrsim 10^{23}$ cm^{-2}. Recent controversy on the discrepancies between different reported values of the electron recombination rate of $H_3{}^+$ (Amano 1988) does not affect this value because of the small concentration of electrons compared to that of CO ($\sim 10^{-4}$). The $H_3{}^+$ ion may also be abundant in diffuse interstellar clouds as discussed by Black (1987).

Molecular clouds often contain massive young stars that are bright infrared sources and thus may be used to probe the cloud. From the above estimates it is apparent that the strongest $H_3{}^+$ lines produced in such columns are at the edge of detectability with current astronomical infrared spectrometers operating at 4 μm.

III. OBSERVATIONS

High-resolution spectral searches for the vibration-rotation $H_3{}^+$ lines have been made toward five obscured infrared sources. All of the spectra were obtained at the United Kingdom Infrared Telescope on Mauna Kea. The instruments used were scanning Fabry-Perot interferometers in series with a liquid and solid nitrogen–cooled grating spectrometer. The Fabry-Perot interferometers, which were used at ambient temperature, have resolutions of 9 and 18 km s^{-1} (determined from measurements of low pressure lines of CO and N_2O). Blocking filters and gratings (the latter with rulings of 383 and 303 grooves mm^{-1}) were used to reject unwanted Fabry-Perot orders. Standard chopping and nodding practices were employed. The detector viewed a circular field of 5″ diameter. An observing log is given in Table 1.

Four of the infrared sources, GL 2591, LkHα 101, BN, and NGC 2024/IRS 2 were observed at 9 km s^{-1} resolution, in steps of 3.2 km s^{-1}, on 1985 October 12, in 90 km s^{-1} wide intervals centered on the ${}^rQ(1, 0)$ line at 2529.724 cm^{-1} (3.953 μm). The lower level of this transition $(J, K) = (1, 0)$ will be well populated at temperatures of many tens of degrees which are thought to be the case in the molecular clouds surrounding the above sources. One of the above sources, NGC 2024/IRS 2, shows an absorption at 4.675 μm due to solid CO (Geballe 1986), indicating at least some carbon depletion in the interstellar gas. A fifth source, W33 IR, was observed in a 90 km s^{-1} wide interval centered on the ${}^pP(1, 1)$ line at 2457.290 cm^{-1} (4.070 μm). A substantial fraction of the cloud surrounding this object is believed to be at temperatures in the range 20–40 K (Goldsmith and Mao 1983). For such a low temperature, the $J = 1$, $K = 1$ level is more populated than the $J = 1$, $K = 0$ as discussed earlier. Infrared spectra of W33 IR show very strong absorption of solid CO and other probable carbon-bearing molecules suggesting high depletion of gaseous CO (Geballe *et al.* 1985). A Fabry-Perot interferometer of resolution 18 km s^{-1} was used to search for the $H_3{}^+$ line in W33 IR, because the source was too faint to detect easily at higher resolution. The spectrum was sampled in 5 km s^{-1} steps.

Wavelength calibrations were made by observing a reference gas cell containing N_2O, which has several vibration-rotation bands in the 4 μm region. The atmospheric transmission, which is flat apart from N_2O lines and the broad tail of the 4.3 m CO_2 band, was determined from measurements of a standard star or the Moon.

IV. RESULTS

The final spectra, in which the telluric absorption lines are removed, are shown in Figures 1 and 2. These spectra have been smoothed by a three-point triangle function, resulting in displayed resolutions of 10 and 19 km s^{-1}. The spectra are

TABLE 1

OBSERVING LOG

Source Name	Date (UT)	$H_3{}^+$ Line	Resolution (km s^{-1})	Integration Time (s $point^{-1}$)	Calibration Object
GL 2591	1985 Oct 12	${}^rQ(1, 0)$	9	120	BS 2479
LkHa$^\alpha$ 101	1985 Oct 12	${}^rQ(1, 0)$	9	56	BS 2479
BN	1985 Oct 12	${}^rQ(1, 0)$	9	104	BS 2479
NGC 2024/IRS 2	1985 Oct 12	${}^rQ(1, 0)$	9	160	BS 2479
W33 IR	1987 Jul 6	${}^pP(1, 1)$	18	1332	Moon

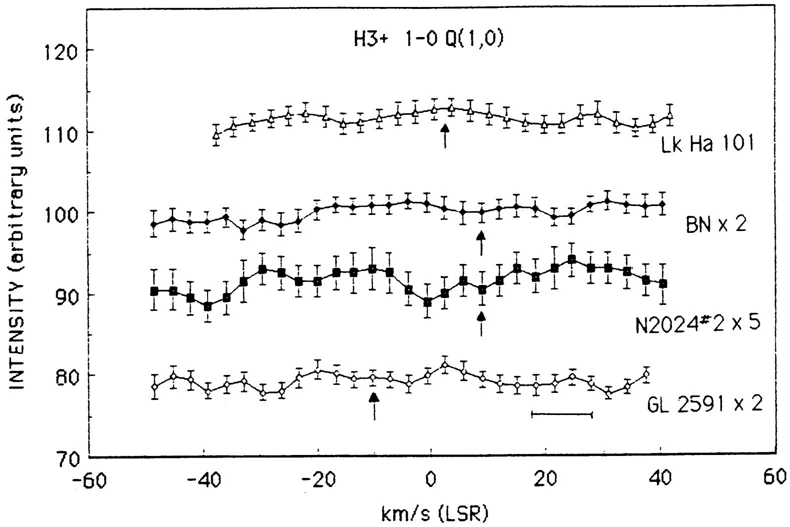

FIG. 1.—Spectra of four embedded infrared sources near the $^rQ(1, 0)$ line of H_3^+ at 2529.724 cm^{-1}. Arrows indicate the expected positions of the line. The velocity resolution (FWHM) is denoted by a horizontal bar. Error bars are ± 1 σ.

essentially flat to within the error bars. In these figures arrows denote the velocities at which the peak of H_3^+ lines are expected; the velocity for LkHα 101 is from Dewdney and Roger (1982), that of GL 2591 is from Bally and Lada (1983) and Geballe and Wade (1985), and that for W33 IR from Goldsmith and Mao (1983).

None of our observational results represents a detection of H_3^+. The 2 σ upper limits are listed in Table 2 first as equivalent widths of unresolved lines observed at the instrumental resolution, and second as column densities. Equations and constants from § II have been used to derive the column densities. As the derived column densities are dependent on the cloud temperature, the upper limits are given for various assumed temperatures of 3 K, 10 K, 30 K, 100 K, and 300 K. The upper limits for the $^rQ(1, 0)$ line have minima at intermediate temperature reflecting the fact that at a very low temperature most of the molecules populate the (1, 1) level while at a high temperature higher J levels.

V. DISCUSSION

The H_3^+ molecular ion is perhaps the most important molecular species yet to be detected in interstellar space in order to confirm and further develop the currently accepted ion-molecule reaction scheme of interstellar chemistry. The upper limits listed in Table 2 can be viewed in various ways in relation to other astronomical observations and conjectures.

a) OMC-1 BN Source

The present work provides the first reliable upper limit for the column density of H_3^+ in front of the BN source. The earlier value (Oka 1981) was not accurate because of the poor weather conditions and the incorrect Hönl-London factor (a factor of 4 mentioned earlier in this paper). The present upper limit of $3 \sim 4 \times 10^{14}$ is much less than the column densities of abundant neutral molecules such as H_2, CO, H_2O, CH_3OH, NH_3, SO, SO_2, HCN, OH, H_2CO, OCS, SiO, $(CH_3)_2O$, $HCOOCH_3$, CN, etc., which range from 2×10^{23} cm^{-2} to

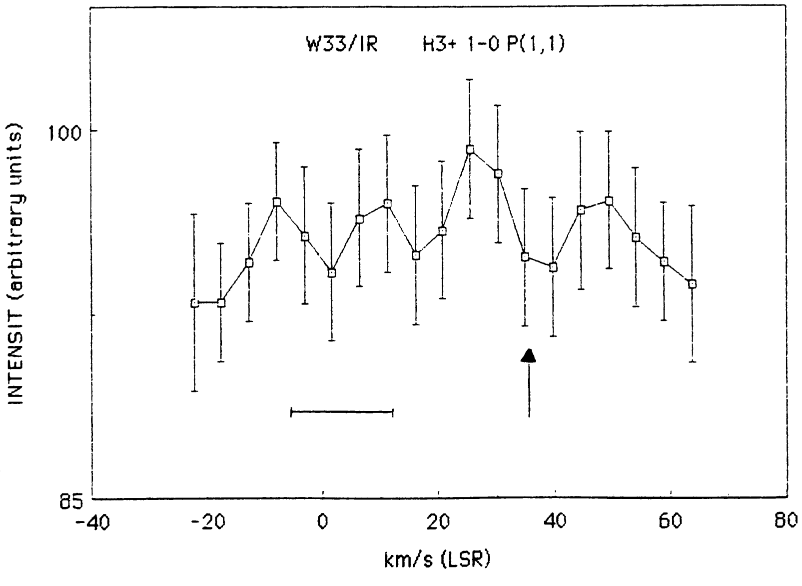

FIG. 2.—Spectrum of W33 IR near the $^rP(1, 1)$ line of H_3^+ at 2457.290 cm^{-1}

TABLE 2

UPPER LIMITS TO H_3^+

SOURCE NAME	H_3^+ LINE	W(ν) (cm^{-1})	2 σ UPPER LIMITS 3 K	10 K	$N(H_3^+)(10^{14}$ cm$^{-2})$ 30 K	100 K	300 K
GL 2591	$^rQ(1, 0)$	0.0020	...	22	3.8	3.3	14
LkHα 101	$^rQ(1, 0)$	0.0015	...	17	2.9	2.5	10
BN	$^rQ(1, 0)$	0.0020	...	22	3.8	3.3	14
NGC 2024/IRS 2	^{r}Q(1, 0)	0.0030	...	33	5.8	4.9	21
W33 IR	$^pP(1, 1)$	0.0070	14	15	23	43	240

2×10^{15} cm^{-2} (Rydbeck and Hjalmarson 1985; Guélin 1985). Most of these molecules are produced by chemical reactions initiated by H_3^+ (Huntress 1977; Dalgarno 1985; Smith 1988) but H_3^+ itself does not have a high steady-state concentration because of its high reactivity with neutral molecules through equation (2).

The observed upper limit of H_3^+ is smaller, but comparable to the observed column density of the most abundant molecular ion HCO^+ which has been reported to be 2×10^{15} cm^{-2} (plateau) and 3×10^{14} cm^{-2} (ridge) by Rydbeck *et al.* (1981) and listed as 1×10^{15} cm^{-2} by Guélin (1985). The predicted ratio of H_3^+ to HCO^+ ranges from $\sim 7 \times 10^{-2}$ (Herbst and Klemperer 1973) to ~ 1 (de Jong, Dalgarno, and Boland 1980) in depleted dense clouds. The millimeter observations of HCO^+ cover a wide region of, and sample the entire line of sight through, Orion A, while the infrared observations cover only the narrow line of sight to BN. Were the densities of ionic species considerably higher than average in the vicinity of BN, H_3^+ might have been detected; however, the observations demonstrate that this is not sufficiently true.

The column density of CO in front of BN has been determined by Scoville *et al.* (1983) to be 1.3×10^{19} cm^{-2}. Assuming that $C/H = 1 \times 10^{-4}$ in the gas, this corresponds to $N(H_2) = 7 \times 10^{22}$ cm^{-2}. Then the upper limits of the ratios H_3^+/CO and H_3^+/H_2 are $\sim 3 \times 10^{-5}$ and 5×10^{-9}, respectively. This upper limit to H_3^+/H_2 is compared with some recent calculations in Table 3. The upper limit is similar to some of the predictions and therefore may constrain some chemical models of the Orion molecular cloud. We note that the H_3^+/H_2 ratio estimated from the possible observation of H_2D^+ emission in NGC 2264 by Phillips *et al.* (1985) based on a thermal model is comparable to the present upper limit, although their estimate is critically dependent on the assumed temperature. In view of the fast exchange reaction between H_2D^+ and H_2 the ortho-para conversion of H_3^+ must be rapid in a molecular cloud.

TABLE 3

COMPARISON OF OBSERVED UPPER LIMIT FOR $[H_3^+]/[H_2]$ ($< 5 \times 10^{-9}$) IN BN WITH PREDICTED VALUES

Model	Value
Langer and Graedel 1989	$1 \times 10^{-7} \sim 3 \times 10^{-9}$
Herbst and Leung 1986	1.6×10^{-9}, 1.8×10^{-9}
Brown and Rice 1986	6.3×10^{-9}
Watt 1985	5×10^{-11}, 1.3×10^{-10}
Leung, Herbst, and Huebner 1985	3.0×10^{-9}, 1.3×10^{-9}
Millar and Freeman 1984*a*, *b*	5.3×10^{-10}
Graedel, Langer, and Frerking 1982	5.2×10^{-9}, 4.7×10^{-10}
Prasad and Huntress 1980	1.1×10^{-9}, 9×10^{-11}
Phillip *et al.* 1985	2.3×10^{-9}

Since H_3^+ is a crucial agent for formation of many molecules, its abundance can be gauged by the abundance of some product neutral species. Thus Lepp, Dalgarno, and Sternberg (1987) recently predicted that $[H_3^+] = 0.1[OH]$ for a cold dark cloud. Using the column density of OH in Orion A of 1×10^{16} cm^{-2} (Guélin 1985) the calculated $[H_3^+]$ is 1×10^{15} cm^{-2} which is negated by the present upper limit of $3 \sim 4 \times 10^{14}$ cm^{-2} albeit by a small margin. Such a comparison, however, may not be very meaningful because the microwave emission signal may well be contributed by molecules behind the infrared star.

b) Other Sources

In addition to BN, line-of-sight measurements of CO column densities have been made in three of the objects we searched for H_3^+. Black and Willner (1984) found a CO column density of $\sim 1 \times 10^{19}$ cm^{-2} toward NGC 2024/IRS 2. Upper limits to H_3^+/CO and H_3^+/H_2 in front of this object are probably a factor of 2 higher (and less significant) than toward BN. The column density of CO in front of GL 2591 is $\sim 2 \times 10^{19}$ (G. F. Mitchell, private communication); however, because much of this CO is warm (G. F. Mitchell, private communication; Geballe and Wade 1985), the upper limit to H_3^+ (and hence H_3^+/H_2) is also probably less significant than toward Orion. Recently, Mitchell, Allen, and Maillard (1988) have measured the column density of cold (23 K) CO to be $\sim 1.3 \times 10^{19}$ cm^{-2} toward W33 IR and the total CO column density to be $\sim 2.7 \times 10^{19}$ cm^{-2}. The significance of our (rather large) H_3^+ upper limit toward this object is difficult to judge, because of the possibility that C is severely depleted in the cold gas in front of this object. The visual extinction toward LkHα 101 is only ~ 10 mag (McGregor, Persson, and Cohen 1984); hence, the upper limit to $N(H_3^+)$ here is less significant than toward BN.

VI. CONCLUSION

Although the sensitivities of current high-resolution infrared spectrometers apparently are insufficient to detect H_3^+ directly in interstellar space, present-day instruments probably are very close to detection. An increase in sensitivity by an order of magnitude, which should occur within the next few years, is likely to lead to the discovery of interstellar H_3^+.

Note added in manuscript (1989 March 3).—The overtone band $2\nu_2 \rightarrow 0$ emission spectrum of H_3^+ has now been identified in Jupiter's south polar hot spot (Maillard and Drossart 1989; J. K. G. Watson, private communication).

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