

Detection of the Ammonium Ion in Space ¹

J. Cernicharo & B. Tercero

*Department of Astrophysics, CAB. INTA-CSIC. Crta Torrejón-Ajalvir Km 4. 28850
Torrejón de Ardoz, Madrid, Spain*

`jcernicharo@cab.inta-csic.es`

and

A. Fuente

Observatorio Astronómico Nacional, Apdo. 112, 28803, Alcalá de Henares, Spain

and

J.L. Domenech, M. Cueto, E. Carrasco, V.J. Herrero & I. Tanarro

Instituto de Estructura de la Materia, IEM-CSIC, Serrano 123, 28006 Madrid, Spain

and

N. Marcelino

NRAO, 520 Edgemont Road, Charlottesville, VA 22902, USA

and

E. Roueff

Luth, Observatoire de Paris, CNRS UMR8102, Place J. Janssen F-92190 Meudon, France

and

M. Gerin

*LERMA, Observatoire de Paris, CNRS UMR8112 and Ecole Normale Supérieure, 61
Avenue de l'Observatoire, F-75014 Paris, France*

and

J. Pearson

*Jet Propulsion Laboratory, 4800 Oak Grove Drive, MC 168-314, Pasadena, CA 91109,
USA*

ABSTRACT

We report on the detection of a narrow feature at 262816.73 MHz towards Orion and the cold prestellar core B1-bS, that we attribute to the 1_0-0_0 line of the deuterated Ammonium ion, NH_3D^+ . The observations were performed with the IRAM 30m radio telescope. The carrier has to be a light molecular species as it is the only feature detected over 3.6 GHz of bandwidth. The hyperfine structure is not resolved indicating a very low value for the electric quadrupolar coupling constant of Nitrogen which is expected for NH_3D^+ as the electric field over the N nucleus is practically zero. Moreover, the feature is right at the predicted frequency for the 1_0-0_0 transition of the Ammonium ion, 262817 ± 6 MHz (3σ), using rotational constants derived from new infrared data obtained in our laboratory in Madrid. The estimated column density is $(1.1 \pm 0.2) \times 10^{12} \text{ cm}^{-2}$. Assuming a deuterium enhancement similar to that of NH_2D , we derive $N(\text{NH}_4^+) \simeq 2.6 \times 10^{13} \text{ cm}^{-2}$, i.e., an abundance for Ammonium of a few 10^{-11} .

Subject headings: ISM: abundances — ISM: individual objects (B1-bS) — ISM: molecules — line: identification — molecular data

1. Introduction

Two of the major nitrogen bearing molecules in the ISM are N_2 and NH_3 , which are predicted to be present in many different media (see, e.g., Nejad et al. 1990; Aikawa et al. 2008; Harada et al. 2010). Whereas N_2 is expected to be largely in the gas phase, NH_3 should be mostly frozen onto the surface of dust grains for temperatures lower than 100 K. Diazenylium (N_2H^+) and Ammonium (NH_4^+), the protonated ions of N_2 and NH_3 , can also provide crucial information for the understanding of interstellar chemistry. The N_2H^+ ion is usually taken as a tracer for the apolar N_2 molecule, which is practically impossible to observe directly using conventional spectroscopic techniques. Detection of diazenylium has been reported from many different sources (Turner 1974; Green et al. 1974; Fuente et al. 1993; Daniel et al. 2007). The ion is assumed to be formed almost exclusively through the reaction of N_2 molecules with H_3^+ and to be destroyed mostly in collisions with electrons or with CO molecules. The existence of interstellar NH_4^+ is also predicted in various

¹This work was based on observations carried out with the IRAM 30-meter telescope. IRAM is supported by INSU/CNRS (France), MPG (Germany) and IGN (Spain)

astrochemical models. In particular, in hot cores (Aikawa et al. 2011) and in other high temperature environments like active galactic nuclei (Harada et al. 2010), grain evaporation leads to a large increase in the gas phase concentration of NH_3 . Ammonia has a high proton affinity and its protonated derivative, NH_4^+ , is predicted to become one of the dominant ions (Aikawa et al. 2008, 2011). Ammonium is also the dominant ion in many cold laboratory plasmas containing hydrogen and nitrogen (Carrasco et al. 2011, 2013). However, the symmetric character of NH_4^+ precludes its observation through radioastronomic methods, and NH_4^+ has never been detected in the ISM. Therefore, the deuterated variant NH_3D^+ , possessing a small permanent dipole moment, is more suitable for astronomical searches.

Deuterated Ammonia NH_2D was detected in Orion by Rodriguez-Kuiper et al. (1978) and in SgrB2 by Turner et al. (1978). A detailed analysis of the $\text{NH}_2\text{D}/\text{NH}_3$ abundance ratio in Orion was first provided by Walmsley et al. (1987) who obtained a value of 0.003. Deuterated Ammonia is produced through the dissociative recombination of NH_3D^+ with electrons. NHD_2 , and ND_3 have been also detected towards a large variety of molecular clouds harbouring a large range of kinetic temperatures (Saito et al. 2000; Roueff et al. 2000; Lis et al. 2002; Roueff et al. 2005; Lis et al. 2006). Hence, the precursor molecule NH_3D^+ , and even more deuterated isotopologues of the Ammonium ion, could be present in hot and cold molecular clouds.

In this Letter we report on the first detection in space of the singly deuterated Ammonium ion towards the sources Orion-IRc2 and the cold prestellar core B1-bS, a source where the whole family of ^{15}N and deuterated isotopologues of Ammonia and NNH^+ have been detected (Lis et al. 2002, 2006; Saito et al. 2000; Daniel et al., in preparation).

2. Observations

The observations presented in this paper were motivated by the analysis of the Orion line survey carried out with the 30m IRAM telescope (Tercero et al. 2010, 2011). During the interpretation of the line survey we had to deal with more than 8000 unidentified features. Around 4400 of them have been successfully assigned to several isotopes of $\text{CH}_3\text{CH}_2\text{CN}$, CH_2CHCN , CH_3OCOH , their vibrational levels and those of all abundant molecules, and to the recently detected methyl acetate and the gauche conformer of ethyl formate in this source (Demyk et al. 2007; Carvajal et al. 2009; Margulès et al. 2009, 2010; Tercero et al. 2010, 2011, 2012, 2013; Motiyenko et al. 2012; Daly et al. 2013; López et al. in preparation). Among the strongest unidentified features we found one at 262816.7 MHz for $\nu_{\text{LSR}}=9 \text{ km s}^{-1}$, (see Figure 1) that was very close to the predicted frequency for the $J_K=1_0-0_0$ line of NH_3D^+ . This deuterated species of the Ammonium ion was observed in the laboratory

by Nakanaga & Amano (1986) through its ν_4 vibrational mode from which rotational constants for the ground state were derived. The molecule was incorporated some time ago to the MADEX code (Cernicharo 2012). The calculated frequency for the $J_K=1_0-0_0$ transition of NH_3D^+ from these rotational constants is 262807 MHz with a $\pm 3\sigma$ uncertainty of ± 9 MHz. The spectrum shown in Figure 1 shows a line 10 MHz away from the predicted one with a linewidth similar to those of the NH_2D lines observed in the same frequency survey. The observed feature, labelled U262816.7 in Figure 1, is practically free of blending from other species. None of the rotational transitions of the recently characterized isotopologues and vibrationally excited states of methyl/vinyl/ethyl cyanide, methyl formate, formamide, or from the abundant species found in Orion (Tercero et al. 2010) matches the frequency of U262816.7 except a possible very weak contribution from $^{13}\text{CH}_3\text{OCOH}$ in its first torsional state. Nevertheless, having still 3600 unidentified features in Orion, claiming a possible identification with NH_3D^+ will be extremely risky and speculative without a more precise frequency measurement in the laboratory (see Section 4).

Prompted by the observed line in Orion we started a search for a similar feature towards the cold prestellar cloud B1-bS. Complex organic molecules have been recently found in this cold core (Marcelino et al. 2009, 2010; Cernicharo et al. 2012). This source also shows an impressive enhancement in the deuteration of abundant molecules such as H_2CO , H_2CS , and NH_3 (Lis et al. 2002, 2006; Marcelino et al. 2005; Saito et al. 2000). With a kinetic temperature of $\simeq 12$ K the expected density of lines at 262.8 GHz is rather low. Previous observations at 3 and 2 millimeter wavelengths indicate that only some diatomic and triatomic species will have significant emission at these high frequencies (Marcelino et al. 2005, 2009, 2010; Cernicharo et al. 2012).

The observations were performed at the IRAM 30m telescope at Pico Veleta (Spain) using the 1 mm Eight MIXer Receivers (EMIR). The backend was a fast Fourier Transform Spectrometer (FTS) with a spectral resolution of 50 kHz and 37275 channels. Two bands, 1.82 GHz wide each, were centered at 262.816 and 265.886 GHz. These two bands were observed simultaneously in the two orthogonal polarizations of the EMIR 230 GHz receiver. All the observations were performed using the Wobbler Switching mode which produces remarkable flat baselines. Pointing errors were always within $3''$. The 30m beam size at the observing frequency is $11''$. The spectra were calibrated in antenna temperature corrected for atmospheric attenuation using the ATM package (Cernicharo 1985; Pardo et al. 2001).

Two different runs have been used for the present observations. The first one in July 2012 allowed us to detect a feature at the same frequency of the line observed in Orion (262816.73 MHz). During this run weather conditions were mostly average to moderate summer, with 3–10 mm of precipitable water vapor. System Receiver temperatures were

between 80 K and system temperatures always above 400 K. A second run in January 2013 provided much better weather conditions and sensitivities. The water vapour amount was 1-2 mm and system temperatures between 200-250 K. The final spectrum, with all data averaged, has a sensitivity of 4.9 mK with 48.9 KHz of spectral resolution (3.7 mK after a box smoothing of 2 channels). The total observing time was 50.8 hours with an averaged system temperature of 249 K.

3. Results

The resulting spectrum from the 30m telescope observations is shown in Figure 2. The upper panel shows the data at the original spectral resolution of 48.9 KHz. The lower panel shows the same data smoothed to 97.8 KHz. A feature is observed right at the expected frequency (see below) with a signal to noise ratio of 4.5 and 6 in the upper and lower panels, respectively. The line is narrow, having a width at half maximum of 0.65 km s^{-1} . It appears as having a double peak that could be due to the hyperfine structure but it is within the noise of the data. Due to the central position of the nitrogen atom in Ammonium the electric field on the nucleus is close to zero. Hence, the quadrupolar coupling constant of the nitrogen nucleus is probably negligible. The Deuterium atom has a spin of 1 and could introduce also some hyperfine splitting but the corresponding quadrupolar momentum is expected to be also very low. The identification relies on the close match between the frequency of the feature with that predicted from the rotational constants derived with our new laboratory measurements (see below). Moreover, this is the unique feature detected together with the lines of CCH $N=3-2$ over a bandwidth of 3.8 GHz. Taking into account the low kinetic temperature of the cloud no candidates can be found in MADEX, the CDMS, or JPL catalogues. Hence, the carrier has to be a light species and NH_3D^+ is the best candidate. Note that in this source NH_3 , $^{15}\text{NH}_3$, NH_2D , $^{15}\text{NH}_2\text{D}$, ND_2H , ND_3 have been detected plus NNH^+ , NND^+ , $^{15}\text{NNH}^+$, N^{15}NH^+ (see above), so the chemistry is well constrained and the abundance of NH_3 and its related species is high.

4. New Laboratory Experiments

The density of lines in B1-bS is low at these high frequencies and the agreement between the frequency of the observed feature with the predictions of Nakanaga & Amano (1986) is very good. However, a final assignment relies on the precise determination from laboratory data of the rotational constants of NH_3D^+ . We have measured again the ν_4 band of deuterated Ammonium with better absolute frequency calibration and more lines than those

reported by Nakanaga & Amano (1986) (2×10^{-4} vs. 1×10^{-3} cm $^{-1}$ and 76 vs. 61 lines respectively). A detailed description of these experiments and calculations are presented in the accompanying Letter (Doménech et al., 2013). The predicted frequency, calculated from the ground state rotational constants derived from a fit with the same parameters as those of Nakanaga & Amano (1986) is 262817 ± 6 MHz ($\pm 3\sigma$). The difference with the observed frequency is much less than one sigma deviation. Hence, we are fully confident that the detected feature in Orion and B1-bS corresponds to the 1_0-0_0 transition of NH_3D^+ .

5. Gas phase chemical modeling

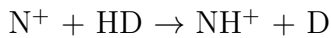
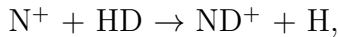
The Orion data will be interpreted in a forthcoming paper. In this work we will concentrate on the analysis of the B1-bS data. Due to the low dipole moment of NH_3D^+ , $\mu=0.26$ D, the Einstein coefficient for spontaneous emission is $A_{ij}=4.8 \times 10^{-6}$ s $^{-1}$ and the critical density to efficiently pump the 1_0 rotational level is just of a few 10^4 cm $^{-3}$. Hence, we could assume that the levels of NH_3D^+ are thermalized at $T_K=12$ K (Marcelino et al. 2005; Cernicharo 2012), we derive for the A species of NH_3D^+ a column density of $(5.5 \pm 1.0) \times 10^{11}$ cm $^{-2}$. The total column density of deuterated Ammonium is, hence, $N(\text{NH}_3\text{D}^+) = (1.1 \pm 0.2) \times 10^{12}$ cm $^{-2}$. The column density of molecular hydrogen derived by Hirano et al. (1999) is 1.3×10^{23} cm $^{-2}$. Therefore, the abundance of deuterated Ammonium is $\simeq 8 \times 10^{-12}$. NH_2D has been observed by Saito et al. (2000) who obtained a column density of $(3.2 \pm 1.2) \times 10^{14}$ cm $^{-2}$. Hence, the abundance ratio of deuterated Ammonia over the deuterated Ammonium ion is $\simeq 300$. Assuming a deuteration enhancement similar to that observed for NH_2D (Saito et al. 2000) the abundance of Ammonium is $\simeq 1.6 \times 10^{-10}$.

All deuterated isotopologues of Ammonia have been detected in B1-bS (Lis et al. 2002, 2006; Saito et al. 2000) and the corresponding gas phase chemical models have been presented by Roueff et al. (2005) who were able to obtain satisfactory agreement between observations and gas phase models in which depletion effects of C and O on grains have been simulated. In these models, Ammonia formation results directly from dissociative recombination of the deuterated Ammonium ion which gives preferentially the $\text{NH}_3 + \text{H}$ product (Öjekull et al. 2004). As a consequence, these models suggest that Ammonium is present at a significative level in this environment and its possible detection is a major witness of the occurrence of such a gas phase chemistry.

Nitrogen chemistry has received considerable recent interest (Dislaire et al. 2012) with the reevaluation of the starting step of gas phase chemical reactions leading to Ammonia via the Ammonium ion: $\text{N}^+ + \text{H}_2 \rightarrow \text{NH}^+ + \text{H}$. This reaction is known to have a slight endothermicity and its efficiency is very much dependent on the para/ortho state of H_2

i.e, whether rotational state of H_2 is 0/1, respectively. Indeed, based on low temperature experiments (Marquette et al., 1985), the endothermicity involved is 168.5 K with para- H_2 and 41.9 K when H_2 is in its “normal” form (1/4 para + 3/4 ortho). These values should be compared to the energy defect between the corresponding $J=0$ and $J=1$ levels of H_2 , which is 170.5 K. Dislaire et al. (2012) have reevaluated the reaction rate constant involved with the ortho form of H_2 with significant differences compared to the previous estimate of Le Boulot (1991). An additional complexity, which is not yet fully understood, is the occurrence of fine structure of N^+ , whose levels are also split by similar amounts. These effects are neglected up to now.

Interestingly enough, thanks to the lower zero-point vibrational energy of HD, the reactions



are found to have even smaller endothermicity. We derive a value of 16.3 K for the production of ND^+ and 100 K for the channel towards NH^+ . Once NH^+ or ND^+ are produced, they react with H_2 , HD, D_2 in a sequence of rapid exothermic reactions until formation of Ammonium. Walmsley and colleagues have been the first to include both para and ortho forms of H_2 , related ions (H_2^+ , H_3^+), and their multiply deuterated isotopologues for completely depleted regions (Walmsley et al. 2004; Flower et al. 2004, 2006a,b), following an earlier suggestion of Pagani et al. (1992). We have extended this approach to a full chemical network including oxygen, carbon, sulfur and nitrogen (Pagani et al. 2011). For the purpose of the present paper, we have updated this chemical network with the recent findings on nitrogen chemistry reported above on the one hand and included also some recently determined neutral-neutral reaction rate constants (Daranlot et al. 2012). As a result, the present model contains 220 species (including ortho/para forms) of singly to fully deuterated substitutes and more than four thousand reactions. We display in Figure 3 the corresponding time evolution of the fractional abundances relative to molecular hydrogen of NH_4^+ and NH_3D^+ . The physical conditions correspond to those supposed to hold in B1-bS: the proton density is taken as $2 \times 10^5 \text{ cm}^{-3}$, a temperature $T=12 \text{ K}$ is assumed, and the elemental abundances correspond to model 2 of Roueff et al. (2005). We see that the fractional abundances of NH_4^+ and NH_3D^+ peak are $\simeq 10^{-10}$ (0.7 Myr) and $\simeq 10^{-11}$ (1 Myr), respectively which is consistent with the observational derivations. The abundance ratio $X(NH_4^+)/X(NH_3D^+)$ is $\simeq 10$. This value is similar to that found for NH_3/NH_2D by Saito et al. (2000). For $t=1 \text{ Myr}$ this ratio is $\simeq 2$.

Our chemical models predict that the abundance of $NH_2D_2^+$ could be a factor of two lower than that of NH_3D^+ . Consequently, we could expect to detect more deuterated species

of the Ammonium ion in cold dark clouds. However, more accurate spectroscopic data are needed for NH_2D_2^+ and NHD_3^+ in order to search for these species.

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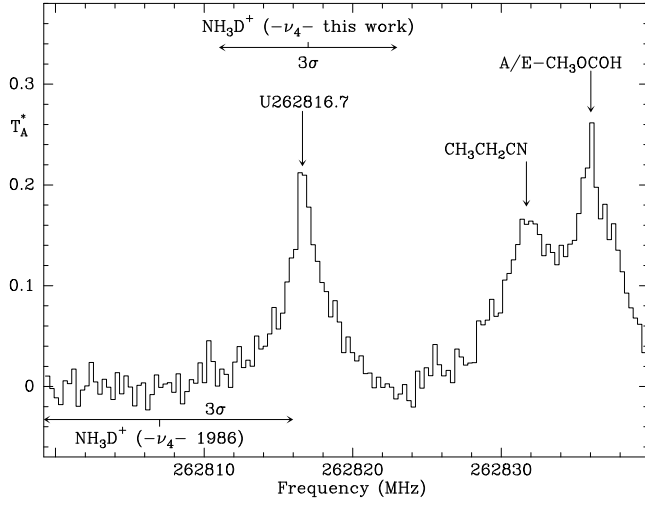


Fig. 1.— Observed spectrum towards Orion-IRc2 around the expected frequency of the $J_K=1_0-0_0$ line of the deuterated ammonium ion. Identification of all other lines in this frequency range is indicated. The predicted frequency and its 3σ error bar is indicated at the bottom for the Nakanaga & Amano (1986) measurements and at the top for our new laboratory data (Domenech et al., 2013; see accompanying Letter).

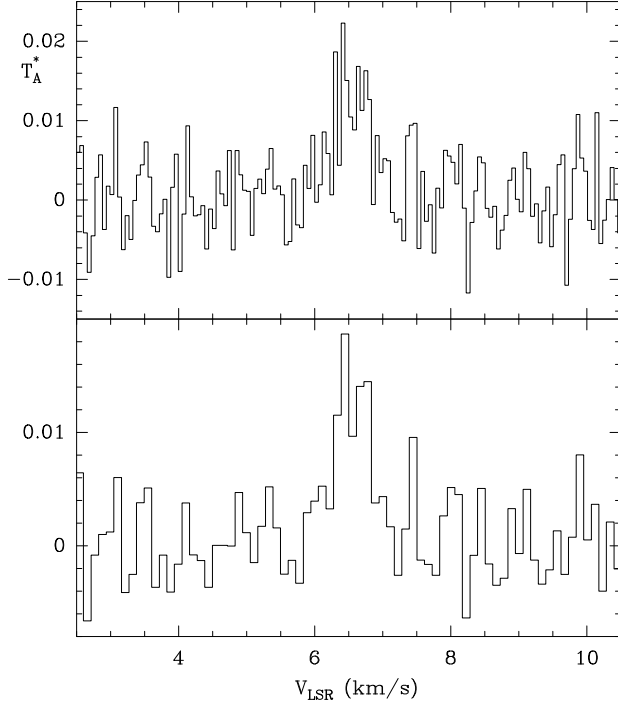


Fig. 2.— Observed spectrum towards B1-bS ($\alpha_{2000}=03^h 33^m 21.34^s$, $\delta_{2000}=31^\circ 07' 26.7''$) at the expected frequency of NH_3D^+ $J_K=1_0-0_0$ transition. The upper panel contains the raw data resulting from a total integration time of 51 hours with a spectral resolution of 48.8 KHz (0.054 km s^{-1}). The bottom panel shows the same data smoothed to 98 KHz. For a LSR velocity of the source of 6.5 km s^{-1} the derived line frequency for the observed feature is $262816.73 \pm 0.1 \text{ MHz}$.

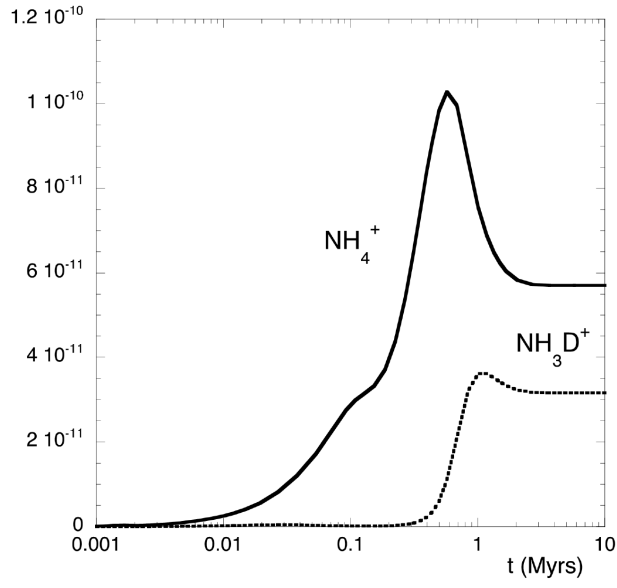


Fig. 3.— Abundances of NH_4^+ and NH_3D^+ as a function of time for the chemical model described in the text.