

*Letter to the Editor***Detection of doubly deuterated ammonia in L134N**E. Roueff¹, S. Tiné², L.H. Coudert³, G. Pineau des Forêts⁴, E. Falgarone⁵, and M. Gerin⁶¹ Observatoire de Paris–Meudon and UMR 8631 du CNRS, 92195 Meudon, France (Evelyne.Roueff@obspm.fr)² Observatoire de Paris–Meudon and UMR 8631 du CNRS, 92195 Meudon, France (Stefano.Tine@obspm.fr)³ Laboratoire de Photophysique Moléculaire, 91405 Orsay, France (Laurent.coudert@ppm.u-psud.fr)⁴ Observatoire de Paris–Meudon and UMR 8631 du CNRS, 92195 Meudon, France (forets@obspm.fr)⁵ Ecole Normale Supérieure and UMR 8540 du CNRS, 75231 Paris, France (Edith.Falgarone@ira.ens.fr)⁶ Ecole Normale Supérieure and UMR 8540 du CNRS, 75231 Paris, France (Maryvonne.Gerin@ensapa.ens.fr)

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Abstract. ND₂H has been detected for the first time in the dense core of L134N, at the peak ammonia position. The abundance ratio [ND₂H]/[NH₂D] is 0.05, a slightly smaller value than the deuterium fractionation observed for ammonia [NH₂D]/[NH₃] \sim 0.1 (Tiné et al. 2000). This is the second doubly deuterated species detected so far in the interstellar medium, and the first one in a cold dense core. The chemical processes leading to the formation of a significant amount of doubly deuterated ammonia ND₂H need to be thoroughly investigated.

Key words: ISM: individual objects: L134N – ISM: molecules**1. Introduction**

Despite a low cosmic abundance of deuterium D/H \sim 1.4×10^{-5} in the solar neighborhood (Lemoine et al. 1999), singly deuterated interstellar molecules are generally present in cold molecular clouds (e.g. Guélin, Langer and Wilson 1982). These observations have been considered as a fairly successful test of the role of ion–molecule chemistry in building interstellar molecules, since chemical fractionation is a natural result of such reaction schemes, as emphasized by Watson (1974) already a long time ago. In such conditions, the deuteron exchange occurs in the forward direction only, the reverse reaction involving endothermicities of some hundreds of K being inhibited at low temperatures. Singly deuterated molecular species (HDO, HDCO, CH₃OD, DCN, DNC, DCO⁺ ...) and even one doubly deuterated molecule (D₂CO) (cf Turner 1990, Sutton et al. 1995, van Dishoeck et al. 1995, Ceccarelli et al. 1998) have also been observed in star forming regions (Orion–IRc2 and IRAS16293–2422). The presence of deuterated species in such places has generally been interpreted as the signature of an active surface chemistry. Desorption in the gas phase of the ad-

sorbed species is due to thermal evaporation in the rather warm environment (temperatures of the order of 100 K) (Rodgers and Millar 1996, Brown and Millar 1989a, 1989b). Time dependent effects may provide some additional enhancement of deuterated species even in absence of an effective desorption mechanism, due to the slower freeze out of deuterated species onto dust grains than for their H–bearing counterparts (Brown and Millar 1989a). However, while these effects will likely enhance the abundance of species such as OD (Brown and Millar 1989a), they are found to be less important for deuterated ammonia (cf the freezeout model of Willacy and Millar 1998). Moreover, doubly deuterated species have never been observed in cold regions up to now.

The L134N (also referred to as L183) dark cloud is an exceptional environment for deuterium fractionation. Tiné et al. (2000) present maps of DCO⁺, N₂D⁺, NH₂D in this dense core. The spatial distribution of N₂H⁺, N₂D⁺ and NH₂D shows a well defined peak, where both the abundances relative to H₂ and the deuterium fractionation are particularly large. This position coincides with an ammonia peak (Swade 1989, Ungerechts, Walmsley and Winnewisser 1980). The emission of NH₂D is so strong that saturation correction effects had to be applied in the analysis of the spectra. We considered thus the possibility of detecting the elusive doubly deuterated species in the same dense core. We report here the positive detection of ND₂H at 110 GHz in this cold dense molecular core.

2. Observations

The observations have been performed with the 30m IRAM telescope in Pico Veleta (Spain) in July and August 1999. We had average summer weather conditions (T_{sys} = 170 K). The telescope is equipped with SIS receivers operated in single side band. The observations were performed in frequency switching mode with the frequency throw set to 7.7 MHz. As a backend we used the autocorrelator with a very high frequency resolution of 20 kHz. The corresponding velocity resolution is \simeq 0.05 km s⁻¹. The angular resolution at the 30m telescope is 22'' at 110 GHz. At

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Table 1. Spectroscopic parameters of the observed ND₂H lines. E_u is the energy of the upper level of the transition, S is the line strength and A the radiative emission probability. Values in parentheses refer to powers of ten.

Transition	Frequency (MHz)	E _u (K)	S debye ²	A s ⁻¹
(ortho)				
1 _{1,0} -1 _{0,1}	110812.85 ^a 110812.34 ^b 110812.59 ^c	18.7	0.129 ^c	6.8(-7) ^c
(para)				
1 _{1,0} -1 _{0,1}	110896.70 ^a 110896.15 ^b 110896.55 ^c	18.4	0.147 ^c	7.8(-7) ^c

^a De Lucia and Helminger (1975) (measured)

^b De Lucia and Helminger (1975) (calculated)

^c present work (calculated)

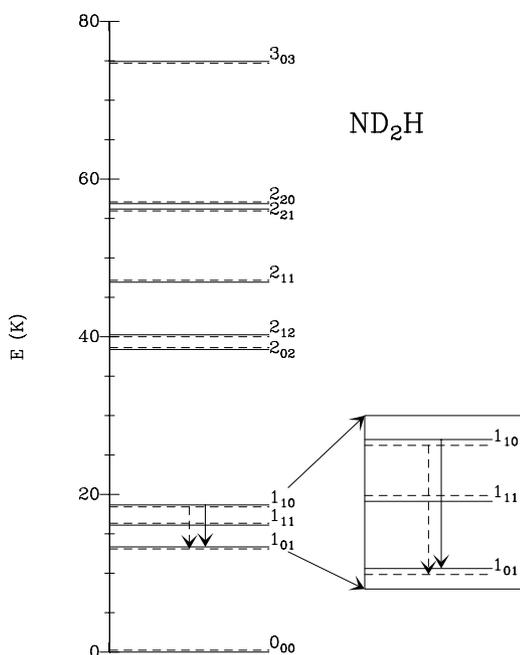


Fig. 1. Energy level diagram of ND₂H. The labels refer to J K_a K_c, the lower tunneling level is characterized by v = 0, the upper one by v = 1. Full lines correspond to ortho levels (K_a + K_c + v even) and dashed lines correspond to para levels (K_a + K_c + v odd). The arrows show the levels connected by the observed 110 GHz transitions.

this frequency the main beam efficiency is approximately 0.66. We observed the L134N dark cloud (RA(1950) = 15^h 51^m 30^s.0, DEC(1950) = -02°42'31'') at the offset (30''E, 18''N) where the NH₂D emission peaks (Tin e et al. 2000). We integrated a total of 9.5 hours to achieve a noise level of 7 mK.

3. Spectroscopic properties of ND₂H

The ground states of NH₂D and ND₂H have been studied through microwave, millimeter wave, sub-millimeter wave and

far-infrared spectroscopy (De Lucia & Helminger 1975, Cohen and Pickett 1982, Fusina et al. 1988) as well as with the help of infrared spectroscopy (Coudert, Valentin and Henry 1986) which yielded values for their rotational and inversion spectroscopic constants. These molecules have a complicated pattern of energy levels characteristic of light asymmetric tops. A further complication arises from vibration-rotation interactions which connect levels having opposite symmetry with respect to inversion. The two transitions of ND₂H we have searched for around 110 GHz correspond to pure rotational transitions of the ortho and para forms with nuclear statistical weight of 6 (=I(I+1)(2I+1)) and 3 (=I(2I+1)) respectively where I (=1) is the nuclear spin of D¹. We have computed the energy pattern using the data reported by De Lucia and Helminger (1975) and by Fusina et al. (1988). Line intensities were calculated assuming that the total dipole moment is the same as for NH₂D ($\mu = 1.47$ debye). This has been shown to be a valid approximation in the case of the vibrational ν_2 band (Kartha et al. 1988). In the energy pattern as well as in the line intensities calculations the xyz axis system was attached to the molecule (Cohen and Pickett 1982) so that the inertia tensor is non diagonal and displays a non vanishing I_{xz} component. This choice leads to the following values for the two nonzero components of the dipole moment μ_x and μ_z : -0.18 and 1.463 debye, respectively.

Particular care has been devoted to the phase conventions and we have checked our program by recalculating the Stark coefficients C₂ of NH₂D and comparing them with the observed values given by Cohen and Pickett (1982). Fig. 1 displays the lowest rotational-tunneling energy levels expressed in K. The vibrational quantum number v relative to inversion sublevels is equal to 0 for the lower sublevel and to 1 for the upper one.

Table 1 gives the molecular properties corresponding to the two observed transitions. The hyperfine structure was not resolved in De Lucia and Helminger (1975) measurements. The comparison between measured and calculated values of the transition frequencies is also emphasized in Table 1.

4. Results and discussion

Fig. 2 displays the resulting spectra. The top panel is for the ortho form of ND₂H and the bottom one for the para form, in the rotational transitions observed (see Table 1).

The results of Gaussian fits to these lines are displayed in Table 2 in the T_A^{*} scale. Because the source is not likely to be very extended, we choose to use the main beam temperature scale to derive the column density. The values listed in Table 2 have been multiplied by the ratio of forward and main beam efficiencies of the 30m IRAM telescope (~ 1.37) to obtain the best estimate of the radiation temperature T_R.

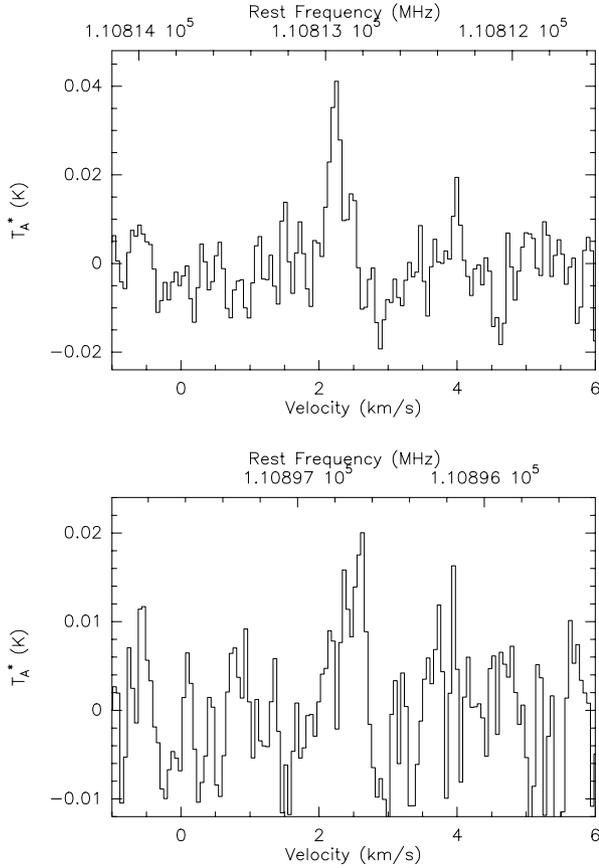
From the solution of the equation of radiative transfer one has

$$T_R = [J_\nu(T_{ex}) - J_\nu(T_{bg})](1 - e^{-\tau}) \quad (1)$$

¹ Turner (1990) used erroneously an ortho to para ratio of 3 for D₂CO and ND₂H.

Table 2. Results of observations. Values in parentheses refer to powers of ten.

Line	T_A^* (K)	$\int T_A^* dv$ (K km s ⁻¹)	σ (K)	v_{LSR} (km s ⁻¹)	Δv (km s ⁻¹)	τ	N_{tot} (cm ⁻²)
110 GHz (ortho)	0.037	0.0092	0.0073	2.24	0.23	1.8(-2)	1.0(13)
110 GHz (para)	0.016	0.0056	0.0069	2.50	0.33	7.7(-3)	1.1(13)

**Fig. 2.** Spectra of ortho (top) and para (bottom) lines of ND₂H towards L134N (30'', 18'')

where $J_\nu(T)$ is the radiation temperature of a black body at temperature T , $T_{bg} = 2.7$ K (the temperature of the cosmic background radiation) and τ is the optical depth in the line. We adopted a simple LTE approximation, for lack of data on the collisional cross sections of this molecule. Hence, the excitation temperature T_{ex} for the lines is an adopted quantity, assumed here to be equal to 6K. This value was inferred from high S/N results of the single deuterium isotope NH₂D towards this same position (Tin e et al. 2000). The previous equations can then be solved to obtain τ and one can eventually derive the total column density N_{tot} of ND₂H as follows

$$N_{tot} = \frac{8\pi\nu^3}{c^3} \frac{Q(T_{ex})}{g_u A} \Delta v \frac{e^{-\frac{E_u}{kT_{ex}}}}{e^{\frac{h\nu}{kT_{ex}}} - 1} \tau \quad (2)$$

where g_u is the degeneracy of the upper state including the nuclear statistical weight and $Q(T)$ is the partition function for the

Table 3. Observed molecular fractions

line of sight	$\frac{NH_2D}{NH_3}$	$\frac{ND_2H}{NH_2D}$	$\frac{HD_2CO}{H_2CO}$	$\frac{D_2CO}{HDCO}$
L134N	0.1	0.05		
Orion (compact ridge)	0.06		0.01–0.14	0.02
IRAS 16293–2422	~ 0.1		0.14	≤ 0.5

molecule, obtained by summing over all energy levels of importance. Both quantities have been calculated by using 2 (1) for the nuclear statistical weight of ortho (para) states respectively. The results for τ and N_{tot} are shown in the last two columns of Table 2. N_{tot} , inferred from either the ortho or the para lines, is the total column density of ND₂H.

Thus, the intensity ratio of the lines is consistent with thermodynamic equilibrium between ortho and para forms. The detection of the para form is at the 3 σ level. The small difference between the central velocities of the ortho and para lines is consistent with the accuracy on the rest frequencies of the observed lines which is about 100 kHz.

Table 3 gives the observed deuterium fractionation of NH₂D and ND₂H towards the core of L134N as well as in the compact ridge in Orion, and towards IRAS 16293–2422, where another doubly deuterated molecule, D₂CO, has been detected by Turner (1990), Sutton et al. (1995) and Ceccarelli et al. (1998).

The gas in these latter lines of sight is fairly warm (temperature of about 80 K) and very dense ($n_H \sim 5 \times 10^6$ cm⁻³) as determined by Sutton et al. (1995) in their study of the distribution of molecules in the core of OMC–1 and by van Dishoeck et al. (1995) for IRAS 16293–2422. These physical conditions are appropriate for ice mantles evaporation combined with a complex gas phase chemistry. Indeed, Brown and Millar (1989b) and Charnley, Tielens and Rodgers (1997) evaluate the relative abundances of doubly deuterated species for hot core regions arising from grain surface formation and obtain the reported order of magnitude for the molecular fractions. The situation is clearly different for L134N where the temperature is very low for thermal evaporation to be efficient. The abundances of molecular ions in L134N such as HCO⁺, DCO⁺ do not support a large value for the cosmic ionization rate, hence cosmic rays desorption is not expected to be efficient either.

It is usually assumed that pure gas phase chemistry can not produce significant abundances of doubly deuterated species in interstellar conditions. However, this question has never been considered really quantitatively. The present detection of ND₂H in a dense core raises thus very stimulating questions about the chemical processes at work. Other searches for this molecule should be made in similar environments to appreciate

the exceptional properties of the L134N line of sight. It would also be interesting to observe other doubly deuterated species in the same location.

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