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Sensitivity of microwave spectra of deuterated ammonia to the variation of the electron-to-proton mass ratio

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Abstract

We estimate sensitivity coefficients Q_μ to the variation of the electron-to-proton mass ratio μ for microwave transitions in partly deuterated ammonia NH_2D and ND_2H . Because of the mixing between rotational and inversion degrees of freedom the coefficients Q_μ strongly depend on the quantum numbers of the transition. This can be used for an astrophysical search for the possible variation of the constant μ .

1. Introduction

At present the discrete microwave spectra of molecules are used for astrophysical studies of the possible variation of the fine-structure constant $\alpha = e^2/(\hbar c)$, the electron-to-proton mass ratio $\mu = m_e/m_p$ and the nuclear g -factor g_n . Rapid progress in experiments with cold molecules can lead to new high-precision laboratory tests of the possible variation of μ (see, for example, review [1] and references therein). It was pointed out in [2] that inversion transitions in fully deuterated ammonia $^{15}\text{ND}_3$ have high sensitivity to μ -variation, $Q_\mu = 5.6$ (sensitivity coefficients Q are defined in section 2). The inversion transition in non-deuterated ammonia has a slightly smaller sensitivity to μ -variation, $Q_\mu = 4.5$ [3]. Molecular rotational lines have much smaller sensitivity, $Q_\mu = 1.0$. In astrophysics the observed frequency shifts are interpreted as Doppler shifts. Because of that, possible μ -variation would lead to apparent velocity offset between ammonia inversion line and rotational molecular lines, originated from the same gas clouds. This fact was used in [3–5] to establish very stringent limits on μ -variation over cosmological timescale $\sim 10^{10}$ years.

Recently, the ammonia method was applied to dense prestellar molecular clouds in the Milky Way [6–8]. These observations provide a safe bound of a maximum velocity offset between ammonia and other molecules at the level of $|\Delta V| \leq 28 \text{ m s}^{-1}$. This bound corresponds to $|\Delta\mu/\mu| \leq 3 \times$

10^{-8} , which is two orders of magnitude more sensitive than extragalactic constraints [3–5]. Taken at face value the measured ΔV shows positive shifts between the line centres of NH_3 and other molecules and suggests a real offset, which would imply a $\Delta\mu/\mu = (2.2 \pm 0.4_{\text{stat}} \pm 0.3_{\text{sys}}) \times 10^{-8}$ [8]. These results can be relevant to the theories, which predict the dependence of the fundamental constants on the local matter density [9].

One of the main possible sources of the systematic errors in such observations is the Doppler noise, i.e. stochastic velocity offsets between different species caused by different spatial distributions of molecules in the gas clouds [10, 11]. Because of that it is preferable to use lines with different sensitivity to the variation of fundamental constants of the same species. Recently, it was shown that sensitivity coefficients for Λ -doublet spectra of OH and CH molecules strongly depend on quantum numbers [12, 13]. In this context partly deuterated ammonia molecules NH_2D and ND_2H may also be interesting. Due to the broken symmetry, the rotational and inversion degrees of freedom for these molecules are strongly mixed. As we will show below, this leads to a significant variation of the sensitivity coefficients of different microwave transitions. Note that microwave spectra of NH_2D and ND_2H from the interstellar medium were recently detected [14–22].

2. Sensitivity coefficients

Let us define dimensionless sensitivity coefficients to the variation of fundamental constants so that

$$\frac{\delta\omega}{\omega} = Q_\alpha \frac{\delta\alpha}{\alpha} + Q_\mu \frac{\delta\mu}{\mu} + Q_g \frac{\delta g_n}{g_n}. \quad (1)$$

These coefficients Q_i are most relevant in astrophysics, where lines are Doppler broadened and linewidth $\Gamma \approx \Gamma_D = \omega \times \delta V/c$, where δV is the velocity distribution width and c is the speed of light. The redshift of a given line is defined as $z_i = \omega_{\text{lab},i}/\omega_i - 1$. Frequency shift (1) leads to the change in the apparent redshifts of individual lines. The difference in the redshifts of two lines is given by

$$\frac{z_i - z_j}{1 + z} = -\Delta Q_\alpha \frac{\delta\alpha}{\alpha} - \Delta Q_\mu \frac{\delta\mu}{\mu} - \Delta Q_g \frac{\delta g_n}{g_n}, \quad (2)$$

where z is the average redshift of both lines and $\Delta Q_\alpha = Q_{\alpha,i} - Q_{\alpha,j}$, etc. We can rewrite (2) in terms of the variation of a single parameter \mathcal{F} :

$$\frac{z_i - z_j}{1 + z} = -\frac{\delta\mathcal{F}}{\mathcal{F}}, \quad \mathcal{F} \equiv \alpha^{\Delta Q_\alpha} \mu^{\Delta Q_\mu} g_n^{\Delta Q_g}. \quad (3)$$

The typical values of δV for extragalactic spectra are about few km s^{-1} . This determines the accuracy of the redshift measurements on the order of $\delta z = 10^{-5}$ – 10^{-6} , practically independent on the transition frequency. For gas clouds in the Milky Way the accuracy can be two orders of magnitude higher, $\delta z = 10^{-7}$ – 10^{-8} . In both cases *the sensitivity of astrophysical spectra to variations of fundamental constants directly depends on ΔQ_i* .

In the optical range the sensitivity coefficients are typically on the order of 10^{-2} – 10^{-3} , while in the microwave and far-infrared frequency regions $Q_i \sim 1$. However, equation (3) shows that we need lines with *different* sensitivities. It is well known that for rotational transitions $Q_\mu = 1.0$, whereas for vibrational transitions $Q_\mu = 0.5$. For both of them $|Q_\alpha| \ll 1$ and $|Q_g| \ll 1$. Inversion transition in NH_3 has $Q_\mu = 4.5$. In the microwave region one can also observe hyperfine transitions ($Q_\alpha = 2$, $Q_\mu = 1$, $Q_g = 1$) and Λ -doublet transitions, where Q_α and Q_μ strongly depend on quantum numbers and can be very large [13]. This makes observations in microwave and far-infrared wavelength regions potentially more sensitive to variations of fundamental constants, as compared to optical observations. Because of the lower sensitivity, systematic effects in the optical region are significantly larger [23].

2.1. Inversion transitions

Sensitivity coefficients for inversion transitions in NH_3 and ND_3 were obtained in [2, 3]. For non-symmetric molecules NH_2D and ND_2H selection rules are such that purely inversion transitions are not observable. Still, we will first estimate sensitivity coefficients for the inversion transition and then will proceed to the mixed inversion–rotation transitions.

In the WKB approximation the inversion transition frequency is given by the expression [24]

$$\omega_{\text{inv}} = \frac{\omega_v}{\pi} e^{-S}, \quad (4)$$

Table 1. Sensitivity coefficients Q_μ for the inversion transitions in different isotopologues of ammonia.

Molecule	Action S	Q_μ		
		This work	[2]	[3]
$^{14}\text{NH}_3$	5.9	4.4		4.5
$^{15}\text{NH}_3$	6.0	4.4		
$^{14}\text{NH}_2\text{D}$	6.5	4.7		
$^{14}\text{ND}_2\text{H}$	7.3	5.1		
$^{14}\text{ND}_3$	8.4	5.7		5.7
$^{15}\text{ND}_3$	8.5	5.7	5.6	

where ω_v is the vibrational frequency for the inversion mode and S is the action (we use atomic units $\hbar = m_e = e = 1$). Vibrational frequency is inversely proportional to the square root of the reduced mass M_1 and, hence, is proportional to $\mu^{1/2}$. The action

$$S = \int_{-a}^a \sqrt{2M_1[U(x) - E]} dx, \quad (5)$$

to a first approximation, is proportional to $\mu^{-1/2}$ (integration here goes between classical turning points, i.e. $U(\pm a) = E$). Therefore, we can rewrite (4) in the form [2]

$$\omega_{\text{inv}} = a_1 \mu^{1/2} \exp(-a_2 \mu^{-1/2}), \quad (6)$$

and find the respective sensitivity coefficient:

$$Q_\mu = \frac{1}{2}(1 + a_2 \mu^{-1/2}) = \frac{1}{2}(1 + S). \quad (7)$$

Deriving (7) we neglected that the energy E in (5) also depends on μ , $E = U_{\text{min}} + \frac{1}{2}\omega_v$. Taking this into account we get the corrected expression [3]

$$Q_\mu = \frac{1}{2} \left(1 + S + \frac{S}{2} \frac{\omega_v}{\Delta U - \frac{1}{2}\omega_v} \right). \quad (8)$$

Following [25] we can estimate $\Delta U \equiv U_{\text{max}} - U_{\text{min}}$ for ammonia to be approximately 2020 cm^{-1} . Now we can use experimental frequencies ω_v and ω_{inv} for different isotopic variants of ammonia to find S from (4) and estimate Q_μ using (8). Results are presented in table 1.

2.2. Mixed transitions

For partly deuterated ammonia inversion levels have different ortho–para symmetry. As a result, inversion transitions can be observed only in combination with rotational transitions ω_r . For such a mixed transition

$$\omega = \omega_r \pm \omega_{\text{inv}}, \quad (9)$$

and sensitivity coefficient is equal to

$$Q_\mu = \frac{\omega_r}{\omega} Q_{r,\mu} \pm \frac{\omega_{\text{inv}}}{\omega} Q_{\text{inv},\mu}, \quad (10)$$

where $Q_{r,\mu} = 1$ and $Q_{\text{inv},\mu}$ is given in table 1. Equation (10) shows that the sensitivity coefficient of the mixed transition is simply a weighted average of those of constituents. For ammonia all observed transitions have a complex hyperfine structure. That means in addition to two terms in (9) there is a third hyperfine term ω_{hf} . However, the hyperfine contribution is typically very small, $|\omega_{\text{hf}}/\omega| \ll 1$. Therefore, we can neglect hyperfine corrections to the sensitivity coefficients.

Table 2. Sensitivity coefficients Q_μ for several lower $\Delta K_c = 0$ microwave transitions in NH_2D . The lower state energy E is given in the third column.

Transition		E (cm $^{-1}$)	ν (GHz)	Q_μ
$1_{1,0}(a) \rightarrow 0_{0,0}(s)$	<i>para</i>	0.00	494.5	1.09
$1_{1,0}(s) \rightarrow 0_{0,0}(a)$	<i>ortho</i>	0.41	470.3	0.90
$1_{1,1}(a) \rightarrow 1_{0,1}(s)$	<i>para</i>	11.10	110.2	1.41
$1_{1,1}(s) \rightarrow 1_{0,1}(a)$	<i>ortho</i>	11.51	85.9	0.48
$2_{1,2}(a) \rightarrow 2_{0,2}(s)$	<i>para</i>	32.78	74.2	1.61
$2_{1,2}(s) \rightarrow 2_{0,2}(a)$	<i>ortho</i>	33.19	50.0	0.10
$2_{2,1}(a) \rightarrow 2_{1,1}(s)$	<i>ortho</i>	40.01	305.7	1.15
$2_{2,1}(s) \rightarrow 2_{1,1}(a)$	<i>para</i>	40.41	282.1	0.84

Exceptions to this rule take place when there are accidental degeneracies between different levels and respective transition frequencies become comparable to the hyperfine structure. This happens, for example, for some Λ -doublet transitions in NO and LiO molecules [13].

3. Molecules NH_2D and ND_2H

Partly deuterated ammonia NH_2D and ND_2H are asymmetric tops. Their rotational levels are classified by the rotational quantum number J and two projections K_a and K_c , which correspond to two limiting cases of prolate and oblate symmetric tops. Thus, rotational levels are labelled as J_{K_a, K_c} . Symmetric and asymmetric levels, which correspond to the inversion tunnelling mode, are labelled with indexes s and a , respectively.

Total inversion symmetry of the rotation-inversion levels is $(-1)^{K_c}$ for s levels and $(-1)^{K_c+1}$ for a levels. Inversion accompanied by rotation over π around one of the principle axes corresponds to the transposition of the two identical nuclei (H for NH_2D , or D for ND_2H). The allowed levels are determined by the total spin of the identical nuclei. Because of that a purely inversion transition is strongly suppressed as an *ortho* \leftrightarrow *para* one.

These selection rules lead to two types of allowed transitions. For transitions with $\Delta K_c = 0$ the allowed combination is $s \leftrightarrow a$ and for transitions with $\Delta K_c = \pm 1$ the allowed combinations are $s \leftrightarrow s$ and $a \leftrightarrow a$. In other words, transitions with $\Delta K_c \neq 0$ are pure rotational, while transitions with $\Delta K_c = 0$ are mixed. Consequently, all transitions with $\Delta K_c \neq 0$ have $Q_\mu = 1$, while transitions with $\Delta K_c = 0$ according to (10) have sensitivities

$$Q_\mu(\omega) = 1 \pm \frac{\omega_{\text{inv}}}{\omega} (Q_{\text{inv}, \mu} - 1). \quad (11)$$

For NH_2D molecule the inversion frequency is 12.2 GHz and $Q_{\text{inv}, \mu} = 4.7$, so

$$Q_\mu(\omega) = 1 \pm \frac{45 \text{ GHz}}{\omega}. \quad (12)$$

For ND_2H molecule the inversion frequency is $\omega_{\text{inv}} = 5.11$ GHz and $Q_{\text{inv}, \mu} = 5.1$. Thus,

$$Q_\mu(\omega) = 1 \pm \frac{21 \text{ GHz}}{\omega}. \quad (13)$$

Results for several strong transitions between lowest rotational levels of these molecules are given in tables 2 and 3.

Table 3. Sensitivity coefficients Q_μ for $\Delta K_c = 0$ transitions in ND_2H .

Transition		E (cm $^{-1}$)	ν (GHz)	Q_μ
$1_{1,0}(a) \rightarrow 0_{0,0}(s)$	<i>ortho</i>	0.00	388.7	1.05
$1_{1,0}(s) \rightarrow 0_{0,0}(a)$	<i>para</i>	0.17	378.5	0.94
$1_{1,1}(a) \rightarrow 1_{0,1}(s)$	<i>para</i>	9.10	67.8	1.31
$1_{1,1}(s) \rightarrow 1_{0,1}(a)$	<i>ortho</i>	9.27	57.7	0.64
$2_{1,2}(a) \rightarrow 2_{0,2}(s)$	<i>ortho</i>	26.66	38.7	1.54
$2_{1,2}(s) \rightarrow 2_{0,2}(a)$	<i>para</i>	26.83	28.6	0.27
$2_{2,1}(a) \rightarrow 2_{1,1}(s)$	<i>ortho</i>	32.59	193.0	1.11
$2_{2,1}(s) \rightarrow 2_{1,1}(a)$	<i>para</i>	32.76	183.2	0.89

We see that differences between coefficients Q_μ for ND_2H are somewhat smaller, than for NH_2D . In combination with significantly smaller abundance, this makes ND_2H a less attractive candidate for the search of μ -variation. Table 2 shows that coefficients Q_μ for $\Delta K_c = 0$ transitions in NH_2D molecule with $J \leq 2$ vary from 0.1 to 1.6. Thus, the largest Q_μ for deuterated ammonia is almost three times smaller than Q_μ for the inversion line in NH_3 . However, in NH_3 there are no close lines with different sensitivities and we are forced to use rotational lines of other molecules as a reference. For deuterated ammonia there are four pairs of lines for *ortho* and *para* molecules which have close frequencies and excitation temperatures, but significantly different sensitivities. For two of these pairs $\Delta Q_\mu \sim 1$. In addition, there are several transitions with $\Delta K_c = \pm 1$, which all have $Q_\mu = 1$. This gives a very characteristic pattern of frequency variation caused by the variation of μ and can be used to suppress various systematic effects. Consequently, reliability of the results can be significantly improved. We conclude that microwave spectra of deuterated ammonia can be used as an important supplement to the ammonia method used previously in [3–8].

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