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

This article has been downloaded from IOPscience. Please scroll down to see the full text article.
(http://iopscience.iop.org/0953-4075/43/7/074003)

The Table of Contents and more related content is available

Download details:
IP Address: 134.197.22.10
The article was downloaded on 22/03/2010 at 02:34

Please note that terms and conditions apply.
Sensitivity of microwave spectra of deuterated ammonia to the variation of the electron-to-proton mass ratio

M G Kozlov1, A V Lapinov2 and S A Levshakov3

1 Petersburg Nuclear Physics Institute, Gatchina, 188300, Russia
2 Institute of Applied Physics, Ulyanova Str. 46, Nizhny Novgorod, 603950, Russia
3 Ioffe Physical-Technical Institute, Politekhnicheskaya Str. 26, 194021, St. Petersburg, Russia
E-mail: mgk@mf1309.spb.edu

Received 20 August 2009, in final form 7 October 2009
Published 19 March 2010
Online at stacks.iop.org/JPhysB/43/074003

Abstract
We estimate sensitivity coefficients $Q_\mu$ to the variation of the electron-to-proton mass ratio $\mu$ for microwave transitions in partly deuterated ammonia NH$_2$D and ND$_2$H. Because of the mixing between rotational and inversion degrees of freedom the coefficients $Q_\mu$ strongly depend on the quantum numbers of the transition. This can be used for an astrophysical search for the possible variation of the constant $\mu$.

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 $\mu$ (see, for example, review [1] and references therein). It was pointed out in [2] that inversion transitions in fully deuterated ammonia $^{15}$ND$_3$ have high sensitivity to $\mu$-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 $\mu$-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 $\mu$-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 $\mu$-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 $|\Delta V| \leq 28$ 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 $\Delta V$ shows positive shifts between the line centres of NH$_3$ and other molecules and suggests a real offset, which would imply a $\Delta \mu/\mu = (2.2 \pm 0.4_{stat} \pm 0.3_{sys}) \times 10^{-8}$ [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 $\Lambda$-doublet spectra of OH and CH molecules strongly depend on quantum numbers [12, 13]. In this context partly deuterated ammonia molecules NH$_2$D and ND$_2$H 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$_2$D and ND$_2$H 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

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

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 $\delta V$ 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

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

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

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

The typical values of $\delta V$ 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 $\Delta Q_i$.

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 $|Q_\mu| < 1$ and $|Q_g| < 1$. 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 $A$-doublet transitions, where $Q_\alpha$ and $Q_\mu$ 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$_2$D and ND$_2$H 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]

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

where $\omega_{\text{inv}}$ 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:

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

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

$$Q_\mu = \frac{1}{2} \left(1 + S + \frac{S}{2 \Delta U - \frac{1}{2} \omega_{\text{inv}}}ight). \quad (8)$$

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 $\omega_{\text{inv}}$ and $\omega_{\text{inv},\mu}$ for different isotopic variants of ammonia to find $S$ from (4) and estimate $Q_\mu$ using (8). Results are presented in table 1.

<table>
<thead>
<tr>
<th>Molecule</th>
<th>Action $S$</th>
<th>This work</th>
<th>[2]</th>
<th>[3]</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH$_3$</td>
<td>5.9</td>
<td>4.4</td>
<td>4.5</td>
<td></td>
</tr>
<tr>
<td>NH$_3$</td>
<td>6.0</td>
<td>4.4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>NH$_3$D</td>
<td>6.5</td>
<td>4.7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>ND$_2$H</td>
<td>7.3</td>
<td>5.1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>ND$_3$</td>
<td>8.4</td>
<td>5.7</td>
<td>5.6</td>
<td></td>
</tr>
<tr>
<td>ND$_3$</td>
<td>8.5</td>
<td>5.7</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

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 $\omega_{\text{rot}}$. For such a mixed transition

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

and sensitivity coefficient is equal to

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

where $Q_{\text{inv},\mu} = 1$ 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 $\omega_{\text{hf}}$. 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 \( Q_\mu \) for several lower \( \Delta K = 0 \) microwave transitions in NH\(_2\)D. The lower state energy \( E \) is given in the third column.

<table>
<thead>
<tr>
<th>Transition</th>
<th>( E ) (cm(^{-1}))</th>
<th>( \nu ) (GHz)</th>
<th>( Q_\mu )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1,0((a) → 0,0((s)) para</td>
<td>0.00</td>
<td>494.5</td>
<td>1.09</td>
</tr>
<tr>
<td>1,0((s) → 0,0((s)) ortho</td>
<td>0.41</td>
<td>470.3</td>
<td>0.90</td>
</tr>
<tr>
<td>1,1((a) → 1,0((s)) para</td>
<td>11.10</td>
<td>110.2</td>
<td>1.41</td>
</tr>
<tr>
<td>1,1((s) → 1,0((a)) ortho</td>
<td>11.51</td>
<td>85.9</td>
<td>0.48</td>
</tr>
<tr>
<td>2,2((a) → 2,2((s)) para</td>
<td>32.78</td>
<td>74.2</td>
<td>1.61</td>
</tr>
<tr>
<td>2,2((s) → 2,2((a)) ortho</td>
<td>33.19</td>
<td>50.0</td>
<td>0.10</td>
</tr>
<tr>
<td>2,1((a) → 2,1((s)) ortho</td>
<td>40.41</td>
<td>305.7</td>
<td>1.15</td>
</tr>
<tr>
<td>2,1((s) → 2,1((a)) para</td>
<td>40.41</td>
<td>282.1</td>
<td>0.84</td>
</tr>
</tbody>
</table>

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 \( \Lambda \)-doublet transitions in NO and LiO molecules [13].

3. Molecules NH\(_2\)D and ND\(_2\)H

Partly deuterated ammonia NH\(_2\)D and ND\(_2\)H 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 \( a \) and \( s \), respectively.

Total inversion symmetry of the rotation-inversion levels is \((-1)^{K_c}\) for \( s \) levels and \((-1)^{K_a+1}\) for \( a \) levels. Inversion accompanied by rotation over \( \pi \) around one of the principle axes corresponds to the transposition of the two identical nuclei (H for NH\(_2\)D, or D for ND\(_2\)H). 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 ↔ para one.

These selection rules lead to two types of allowed transitions. For transitions with \( \Delta K = 0 \) the allowed combination is \( s \leftrightarrow a \) and for transitions with \( \Delta K = \pm 1 \) 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 \( \Delta K \neq 0 \) have \( Q_\mu = 1 \), while transitions with \( \Delta K_c = 0 \) according to (10) have sensitivities

\[
Q_\mu(\omega) = 1 \pm \frac{\omega}{\omega_{inv}} (Q_{inv,\mu} - 1).
\]

For NH\(_2\)D molecule the inversion frequency is 12.2 GHz and \( Q_{inv,\mu} = 4.7 \), so

\[
Q_\mu(\omega) = 1 \pm \frac{45 \text{ GHz}}{\omega}.
\]

For ND\(_2\)H molecule the inversion frequency is \( \omega_{inv} = 5.11 \) GHz and \( Q_{inv,\mu} = 5.1 \). Thus,

\[
Q_\mu(\omega) = 1 \pm \frac{21 \text{ GHz}}{\omega}.
\]

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

We see that differences between coefficients \( Q_\mu \) for ND\(_2\)H are somewhat smaller, than for NH\(_2\)D. In combination with significantly smaller abundance, this makes ND\(_2\)H a less attractive candidate for the search of \( \mu \)-variation. Table 2 shows that coefficients \( Q_\mu \) for \( \Delta K = 0 \) transitions in ND\(_2\)H molecule with \( J < 2 \) vary from 0.1 to 1.6. Thus, the largest \( Q_\mu \) for deuterated ammonia is almost three times smaller than \( Q_\mu \) 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 \( \mu \) 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].

Acknowledgments

This research is partly supported by RFBR grants 08-02-00460, 09-02-00352 and 09-02-12223.

References