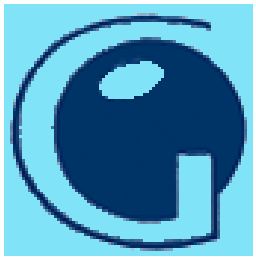
Using molecules to test variation of fundamental constants

Mikhail Kozlov



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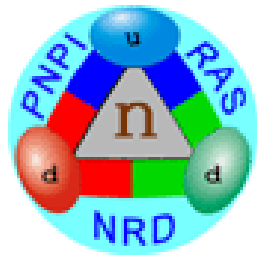

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Collaborators

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Fundamental constants in atomic physics

Fundamental constants, which influence atomic and molecular spectra:

- Fine structure constant $\alpha = e^2/(\hbar c)$ is a coupling constant in QED.
- Electron to proton mass ratio $\mu = m_e/m_p$. Because m_p is proportional to Λ_{QCD}, $\mu \sim m_e/\Lambda_{QCD}$.
- Nuclear gyromagnetic ratio g_n can be expressed in terms of Λ_{QCD} and quark masses, but for atomic physics g_n is independent constant. It always enters in combination $g_n\mu$. According to Flambaum & Tedesco (2006) the dependence of g_n on quark masses is weak.

Dimensionless sensitivity coefficients

If fundamental constants change, the frequency of any atomic transition also change:

$$\begin{aligned} \omega &= \omega_0 \left[1 + Q_\alpha \frac{\delta\alpha}{\alpha} + Q_\mu \frac{\delta\mu}{\mu} + Q_g \frac{\delta g_n}{g_n}\right], \\ \frac{\delta\omega}{\omega} &= \frac{\delta F}{F}, \qquad F = \alpha^{Q_\alpha} \mu^{Q_\mu} g_n^{Q_g}. \end{aligned}$$

In order to detect this variation we need to compare at least two transition frequencies:

$$\frac{\omega_i}{\omega_k} = \left(\frac{\omega_i}{\omega_k}\right)_0 \left[1 + \frac{\delta\Phi}{\Phi}\right], \qquad \Phi = \alpha^{\Delta Q_\alpha} \mu^{\Delta Q_\mu} g_n^{\Delta Q_g}.$$

Clearly, the effect is proportional to the differences of sensitivity coefficients ΔQ.

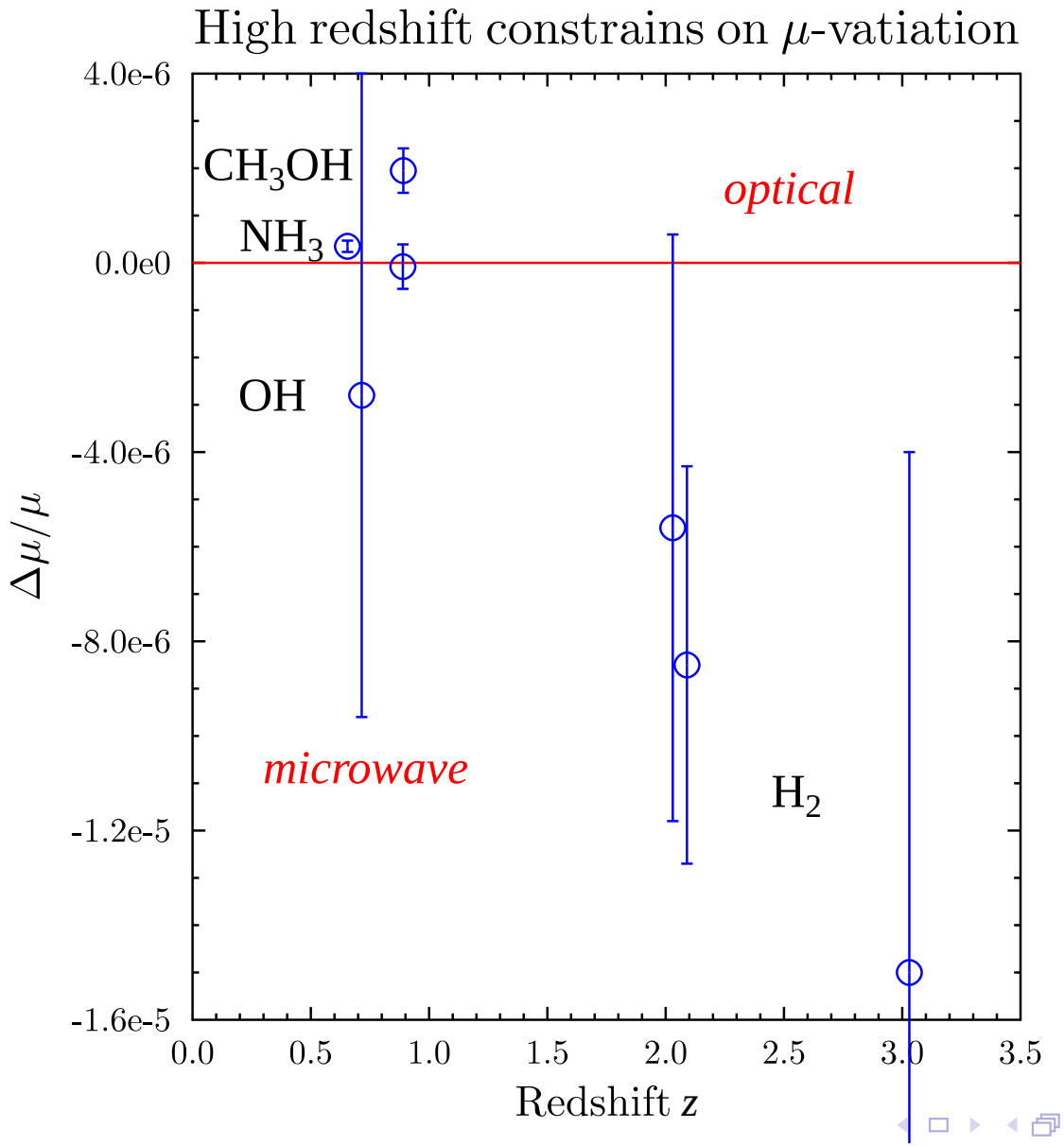
Sensitivity coefficients for different wavebands (in a.u.)

- For optical transitions in *light* ($Z \leq 30$) atoms and molecules $Q_\alpha, Q_\mu, Q_g \ll 1$.
- For optical transitions in *heavy* ($Z \gtrsim 80$) atoms and molecules $Q_\alpha \sim 1$.
- Fine structure (IR, FIR) $\sim \alpha^2 \quad \Rightarrow \quad Q_\alpha = 2$.
- Vibrational structure (IR) $\sim \mu^{1/2} \quad \Rightarrow \quad Q_\mu = \frac{1}{2}$.
- Rotational structure (FIR, microwave) $Q_\mu = 1$.
- Magnetic hyperfine structure (microwave) $Q_\alpha = 2;\ Q_\mu = 1;\ Q_g = 1$.
- Tunneling transitions in polyatomic molecules (FIR, microwave) $1 \lesssim Q_\mu \lesssim 10$.
- Microwave mixed tunneling-rotational lines $|Q_\mu| \gg 1$.
- Microwave Λ-doublet lines in diatomics $|Q_\alpha|, |Q_\mu| \gg 1$.

Early astrophysical limits on variation of constants

In **1956** Savedoff compared redshifts for optical transitions with that for 21 cm hyperfine line in hydrogen to get the limit on the variation $F = \alpha^2 \mu g_n$ on the order of 10^{-3} for the time 0.3 Gyr.

Present high redshift limits on μ-variation

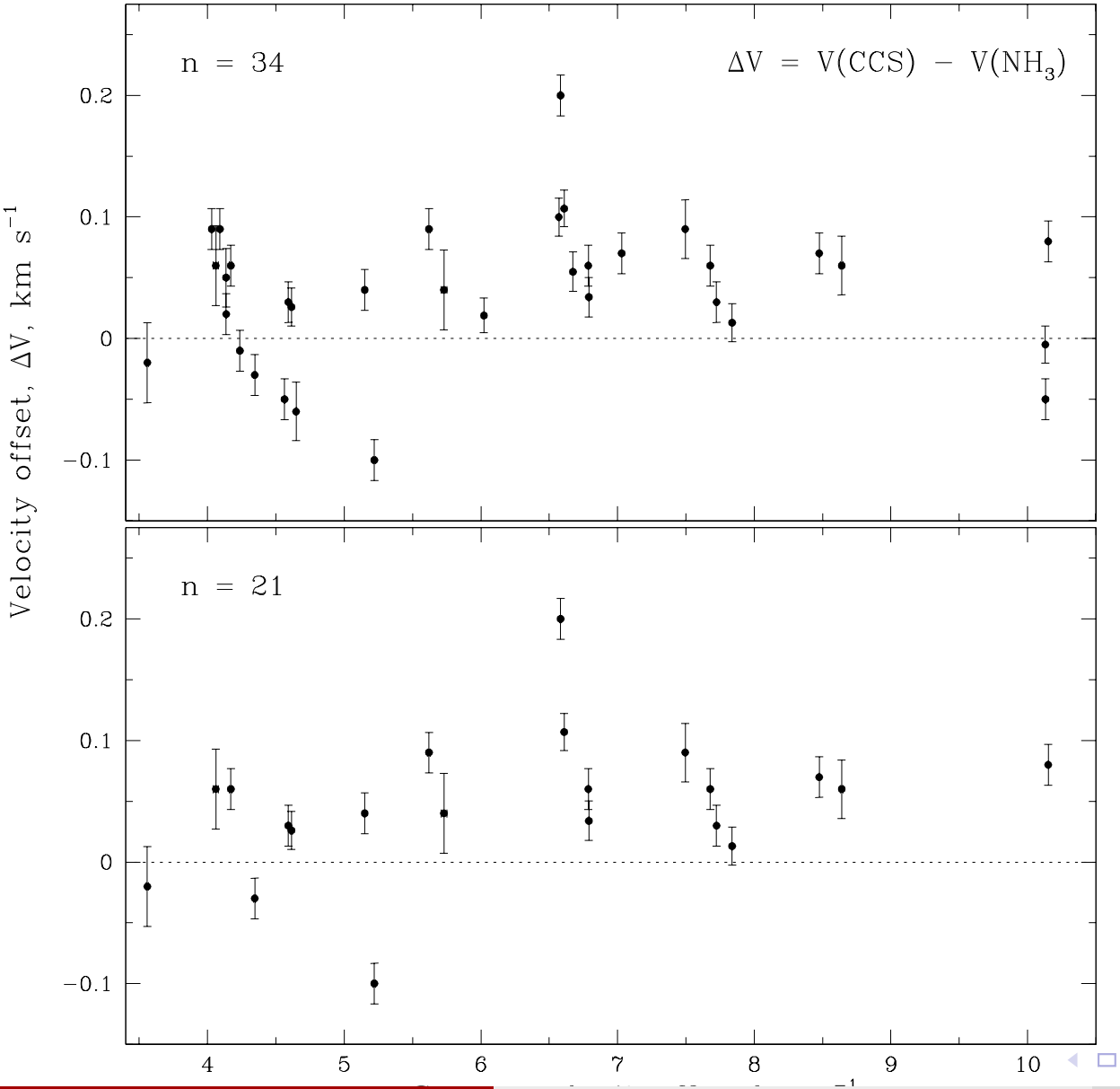


Cold molecular clouds in the Milky Way

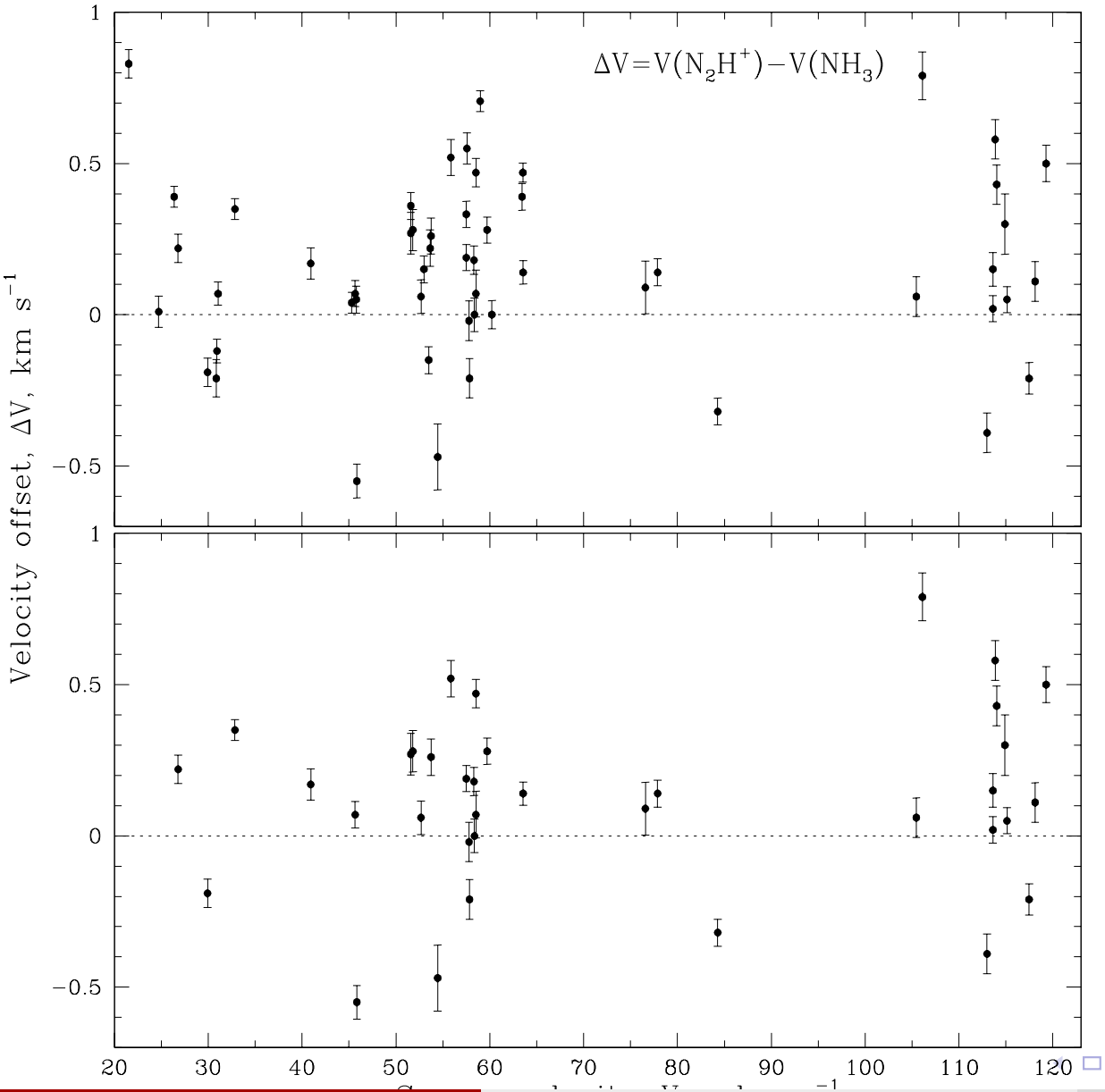
Emission lines of ammonia are often seen together with rotational lines of CCS, N_2H^+, and HC_3N from the cold molecular clouds in the Galaxy. These clouds include molecular cores, which have typical density $\lesssim 10^5$ cm^{-1} and temperature $T \lesssim 10$K. Molecular lines have typical width $\gtrsim 100$ m/s, which allows to measure frequency with relative accuracy up to 10^{-8}.

Good quality spectra are published for large number of cores (~ 200) in the Pipe Nebula (130 pc) [Rosolowsky et al. 2008], Perseus molecular cloud (260 pc) [Rathborne et al. 2008], and Infrared Dark Clouds ($>$ 2 kpc) [Sakai et al. 2008].

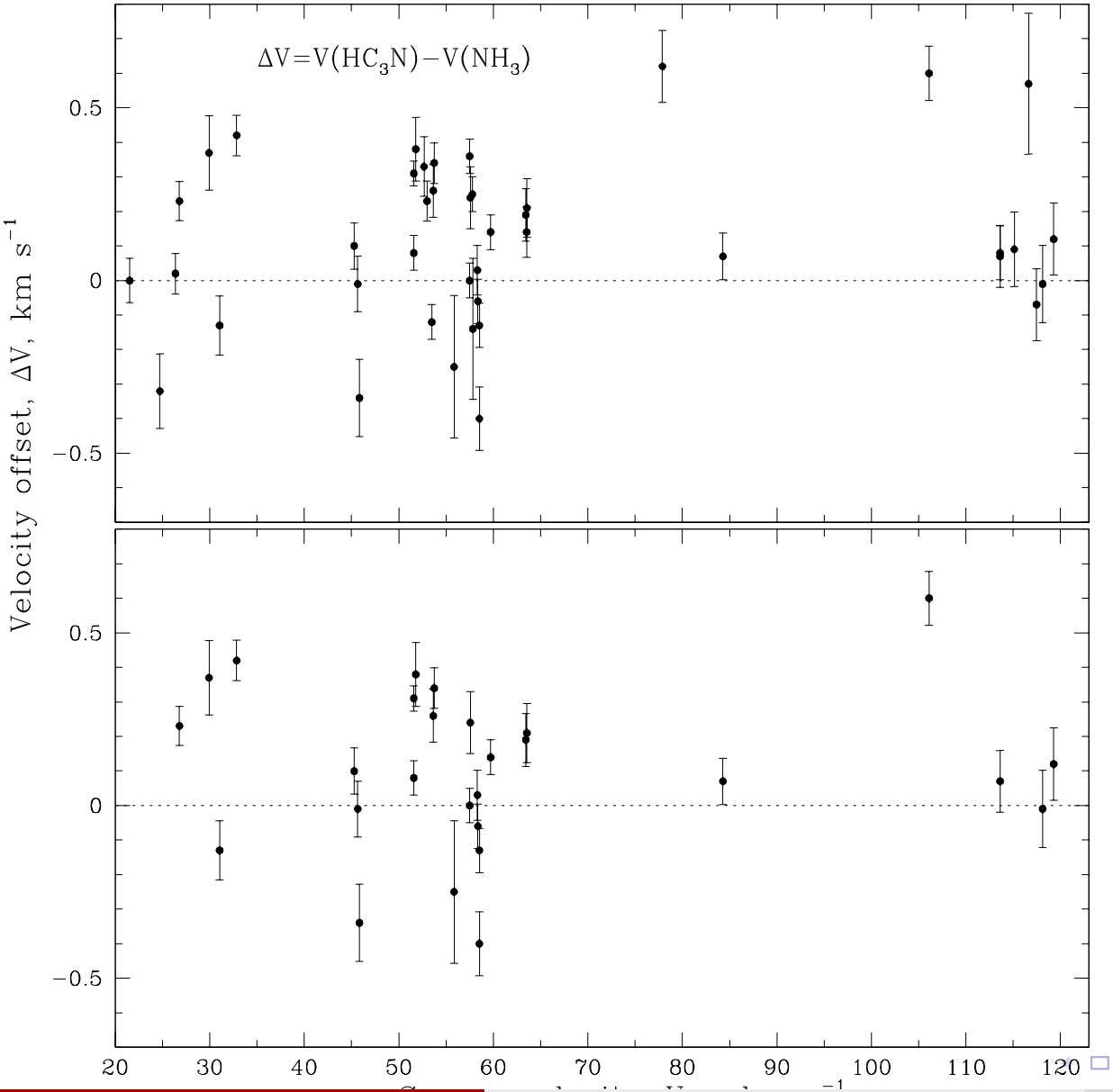
Pipe Nebula: ΔV(CCS-NH_3) (Rosolowsky et al. 2008)



IRDCs: $\Delta V(N_2H^+\text{-}NH_3)$ (Sakai et al. 2008)



IRDCs: $\Delta V(HC_3N\text{-}NH_3)$ (Sakai et al. 2008)



Comparison of ammonia tunneling transition with rotational transitions in CCS, N_2H^+, and HC_3N

Analysis of the published spectra from Pipe Nebula, Perseus molecular cloud, and Infrared Dark Clouds gave [Levshakov et al, arXiv:0808.0583, 2008]:

$$\Delta V = (52 \pm 7_{stat} \pm 14_{sys})\, m/s.$$

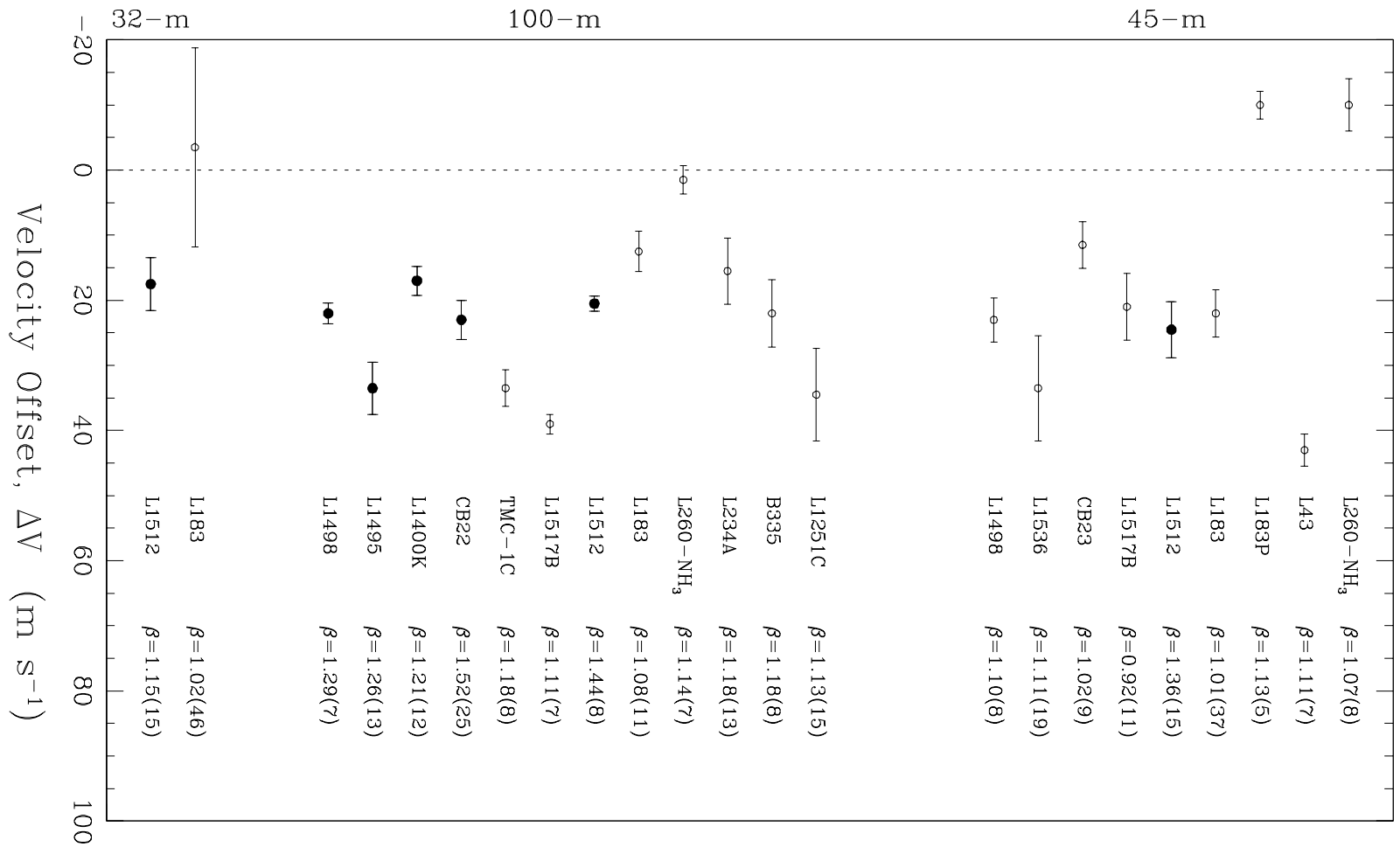
Being interpreted in terms of μ-variation this translates into:

$$\Delta\mu/\mu = (5.0 \pm 1.6) \times 10^{-8}.$$

Laboratory frequencies used in ammonia method

Molecule	Transition	$\nu_{\rm rest}$ GHz	$\lambda_{\rm rest}$ cm	ε_v m s^{-1}
CCS	$J_N = 2_1 - 1_0$	22.344033(1)	1.34	13
NH_3	$(J, K) = (1, 1)$	23.6944955(1)	1.27	1.3
HC_3N	$J = 5 - 4$	45.4903102(3)	0.66	2.8
N_2H^+	$J = 1 - 0$	93.173777(4)	0.32	14

Dedicated observations in 2009 of ammonia and HC_3N with 3 different radio telescopes



Results of dedicated observations in 2009

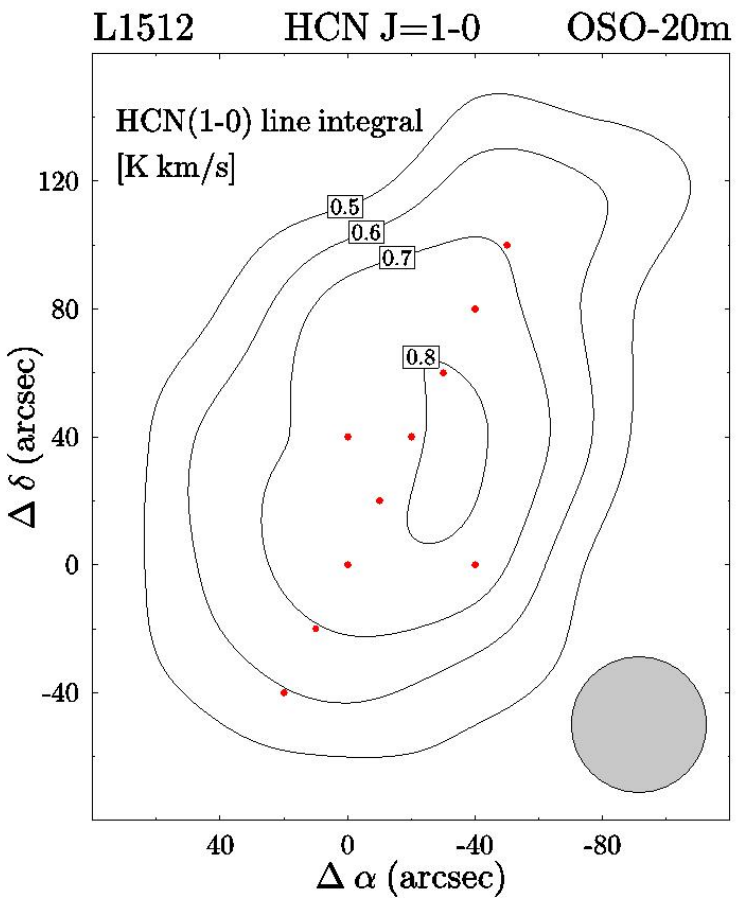
Observations were done in Medicina 32-m telescope, Effelsberg 100-m telescope, and Nobeyama 45-m telescope for rotational line in HC_3N and inversion line in NH_3. Comparison of these lines revealed systematic relative offset [Levshakov *et al.* A&A, **512**, A44 (2010)]:

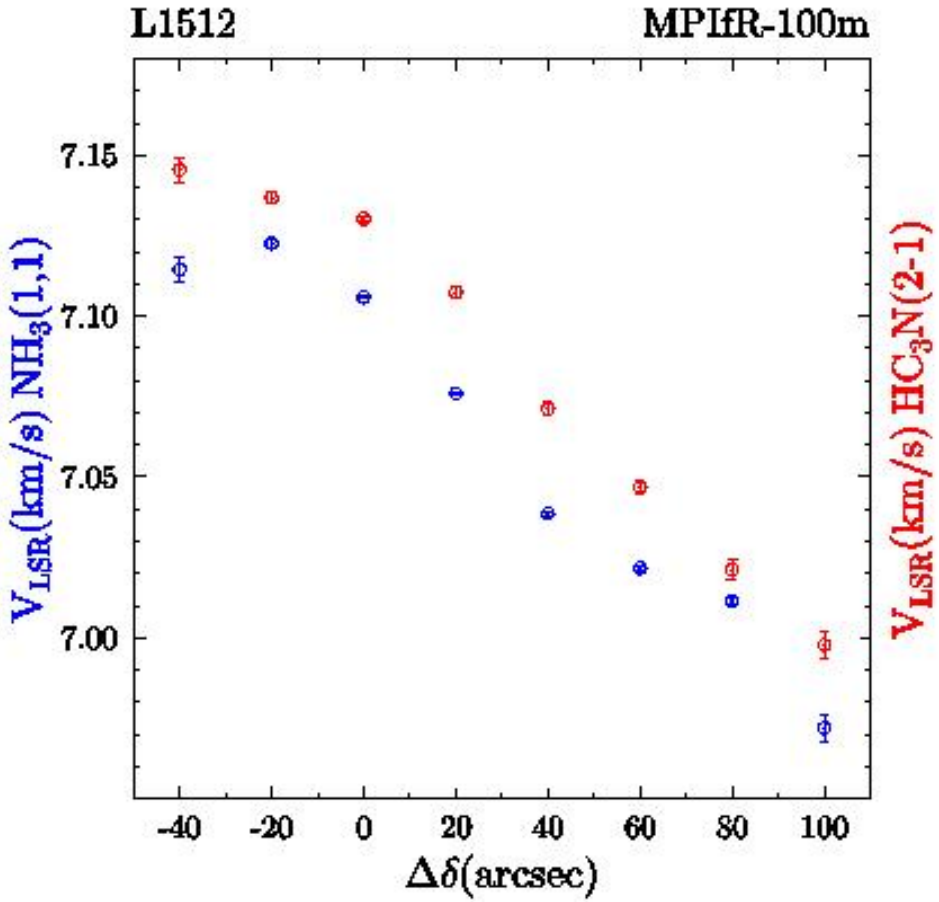
$$\Delta V = (27 \pm 4_{stat} \pm 3_{sys})\, m/s,$$

$$\Delta\mu/\mu = (2.6 \pm 0.4_{stat} \pm 0.3_{sys}) \times 10^{-8}.$$

2010 result from 100-m Effelsberg telescope

Mapping of molecular core L1512





Results of mapping in 2010

Four molecular cores were studied. For two of them (L1498 and L1512) the observed linewidths and velocity profiles are consistent with the assumption that two molecules trace the same gas. *For two other cores the molecules are not tracing the same gas.*
Result of the 2010 mapping of L1498 and L1512 cores give:

$$\Delta V = (26.9 \pm 1.2_{stat} \pm 3_{sys})\, m/s,$$

$$\Delta\mu/\mu = (2.6 \pm 0.1_{stat} \pm 0.3_{sys}) \times 10^{-8}.$$

This is in agreement with 2009 results.

Chameleon-like scalar field

Non-zero result from the Milky Way corresponds to the timescale of few hundred years, or the time-variation on the scale of $10^{-10}\,\mathrm{yr}^{-1}$. This is in sharp contradiction with both laboratory limit $[< 10^{-14}\,\mathrm{yr}^{-1}]$ and cosmological limit $[< 2 \times 10^{-16}\,\mathrm{yr}^{-1}]$.

Many theoretical models introduce additional scalar field to explain the cosmological Dark Energy. In Chameleon models such field is massless in the vacuum but becomes massive in the presence of matter. This leads to the dependence of fundamental constants on the local matter density.

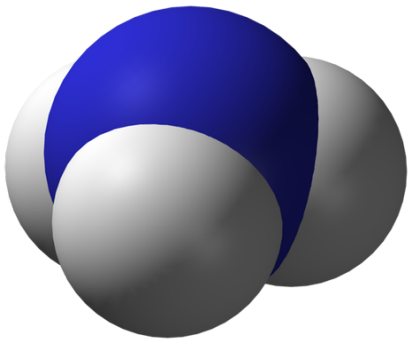
The matter density in the molecular clouds is $\sim 10^5\,\mathrm{cm}^{-3}$*, so observed non-zero variation agrees with prediction of Chameleon models*.

Current status

Studies of the possible systematics are going on on the 100-m Effelsberg telescope.

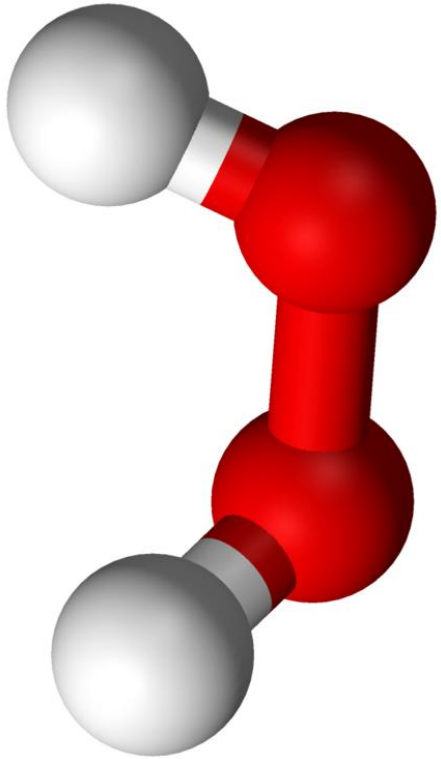
- Telescope software is tested
- Results from different detectors and in different polarizations are compared.
- Several effects on the level $\sim$ 1 m/s were found but no effects on the scale of 20 m/s.
- More mapping of molecular cores is planned.
- Plans for remeasuring laboratory frequencies.
- Plans to use other molecules with high sensitivity to μ-variation.

Examples of molecules with tunneling modes



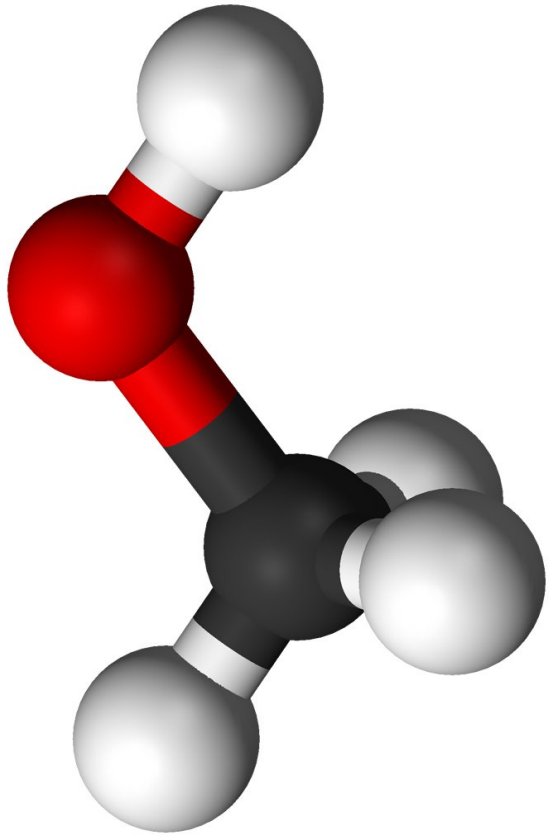
Hydronium – H_3O^+

(umbrella mode)



Peroxide – H_2O_2

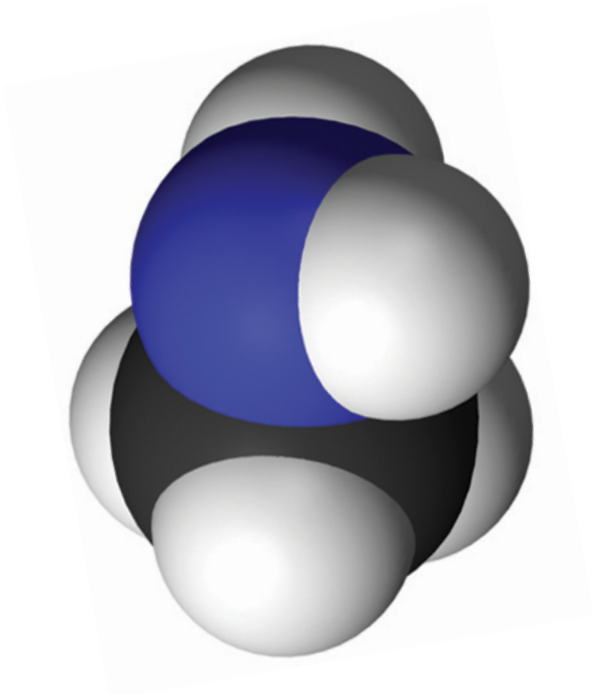
(inversion)



Methanol – CH_3OH

(hindered rotation)

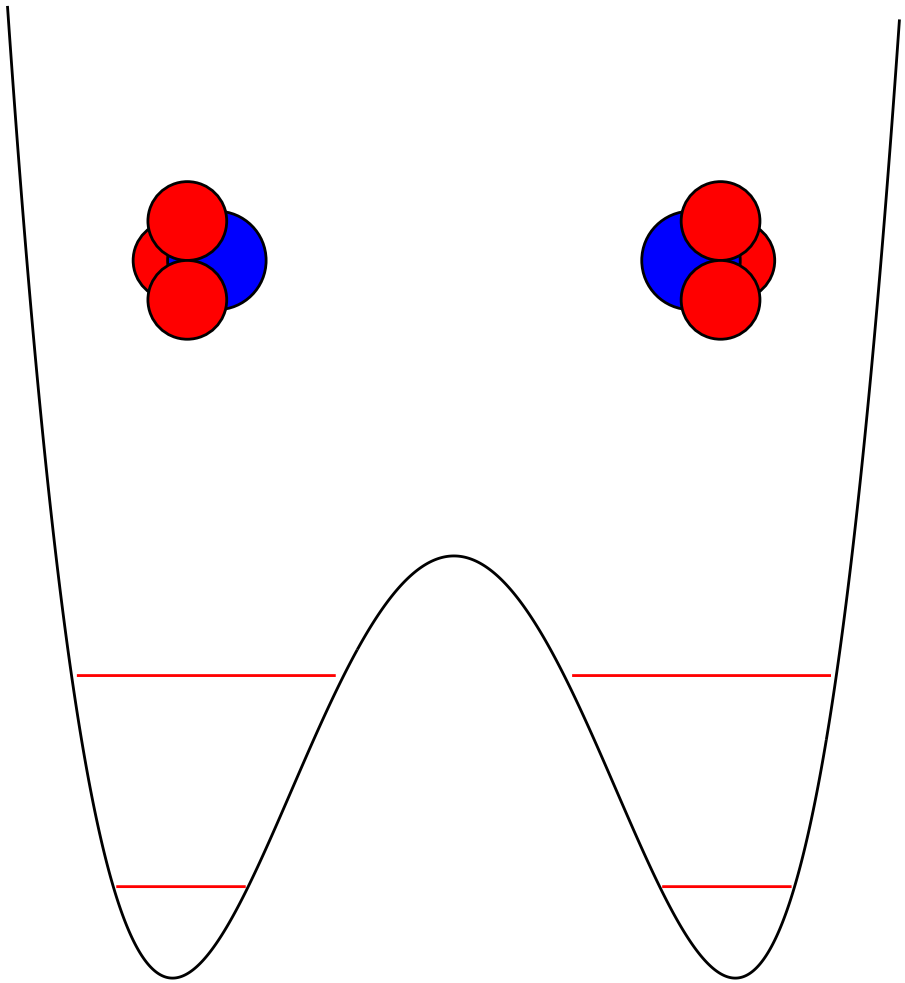
Molecules with more than one tunneling mode



methylamine – CH_3NH_2
(one wagging & one torsion modes)

hydrazine – N_2H_4
(two wagging & one inversion modes)

Inversion line in NH_3



Analytical solution [Landau & Lifshitz]

WKB approximation for tunneling frequency in the ground state E_0 reads:

$$\omega_{\rm tun} = \frac{2E_0}{\pi}\exp\left(-S\right)$$
$$= \frac{2E_0}{\pi}\exp\left(-\frac{1}{\hbar}\int_{-a}^{a}\sqrt{2M_1(U(x)-E_0)}\mathrm{d}x\right),$$
$$\frac{\delta\omega_{\rm tun}}{\omega_{\rm tun}} \approx \left(\frac{1}{2}+\frac{S}{2}+\frac{S}{2}\frac{E_0}{U_{\rm max}-E_0}\right)\frac{\delta\mu}{\mu} = Q_\mu\frac{\delta\mu}{\mu}.$$

Sensitivity coefficients Q_μ for inversion transition in different isotopologues of ammonia.

Molecule	Action S	Q_μ
$^{14}NH_3$	5.9	4.4
$^{15}NH_3$	6.0	4.4
$^{14}NH_2D$	6.5	4.7
$^{14}ND_2H$	7.3	5.1
$^{14}ND_3$	8.4	5.7
$^{15}ND_3$	8.5	5.7
$^{15}ND_3$ *		5.6

*) van Veldhoven *et al.* [Eur. Phys. J. D,**31**, 337 (2004)].

Mixed tunneling-rotational transitions

In some molecules tunneling mode is strongly mixed with rotational degrees of freedom. For example, in partly deuterated ammonia inversion lines have different ortho-para symmetry. Because of that inversion transition goes only in combination with rotational transitions. For such mixed transitions

$$\omega = \omega_{\rm r} \pm \omega_{\rm tun} \,,$$

and sensitivity coefficients are equal to

$$Q_\mu = \frac{\omega_{\rm r}}{\omega} Q_{{\rm r},\mu} \pm \frac{\omega_{\rm tun}}{\omega} Q_{{\rm tun},\mu} \,,$$

where $Q_{{\rm r},\mu} = 1$ and $Q_{{\rm tun},\mu} \gtrsim 2$. If $\omega \ll \omega_{\rm r,tun}$, then $|Q_\mu| \gg 1$.

Effective tunneling-rotational Hamiltonian

More accurate theory for mixed tunneling-rotational transitions can be based on effective tunneling-rotational Hamiltonians. However, we need to know how effective parameters scale with μ.

Consider example of the rotational constant B_0 for diatomic molecule in its ground vibrational state:

$$B_0 = \frac{1}{M}\left\langle v=0 \left| \frac{1}{R^2} \right| v=0 \right\rangle = B_e - \alpha_e + \ldots$$

$B_e = 1/(MR_0^2) \quad \Longrightarrow \quad B_e \sim \mu$

α_e – depends on vibrational wavefunction $\quad \Longrightarrow \quad \alpha_e \sim \mu^{3/2}$.

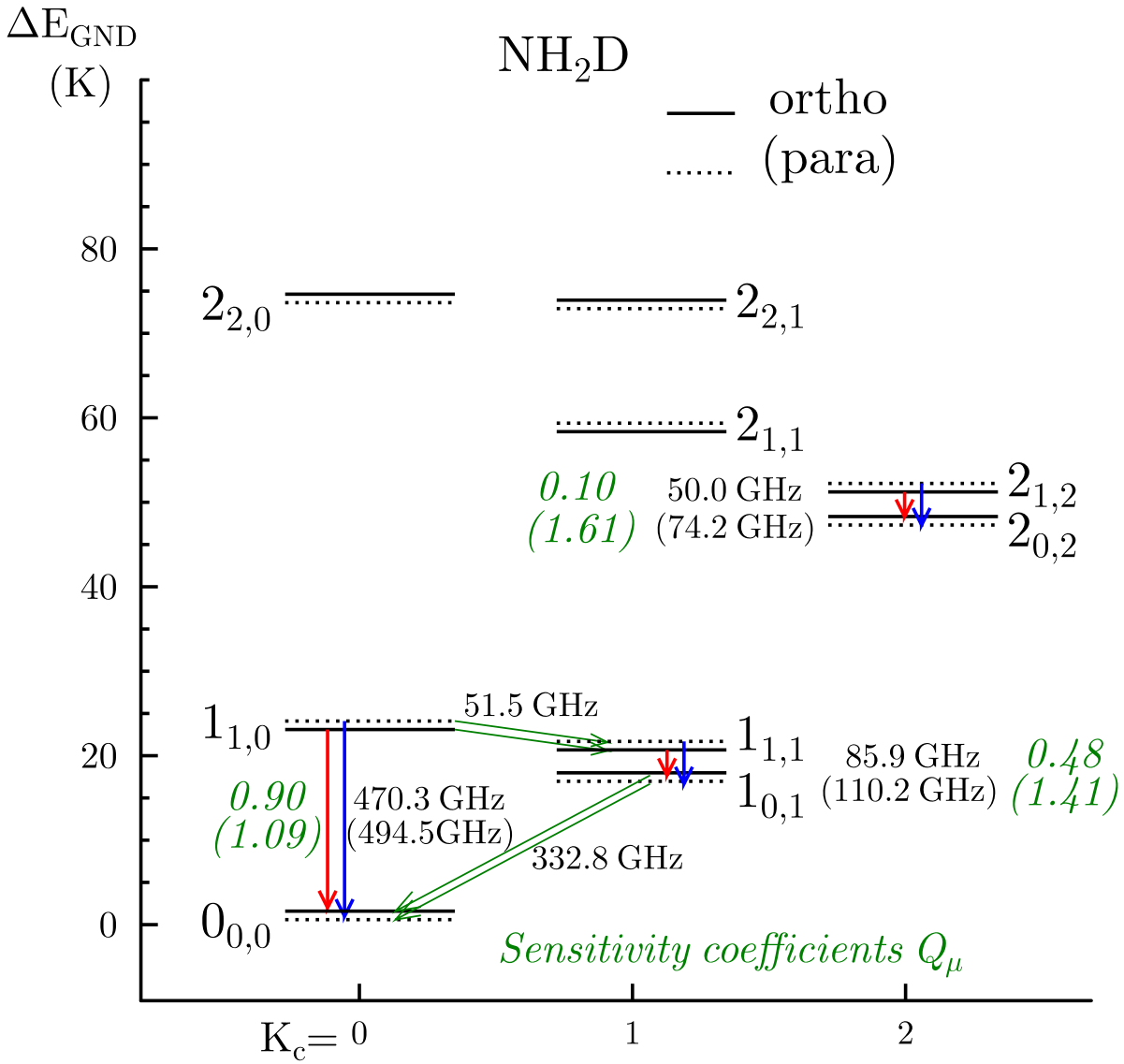
Result: $\boxed{\delta B_0/B_0 = Q_\mu^r \, \delta\mu/\mu; \quad Q_\mu^r \neq 1}$.

- ◇ Typically, $\alpha_e/B_0 \sim 10^{-2}$;
- ◇ NO molecule: $Q_\mu^r = 0.995$;
- ◇ HF molecule: $Q_\mu^r = 0.981$.

μ-scaling of parameters in effective Hamiltonians

- There is no one-to-one correspondence between terms of the effective Hamiltonian and the terms of the Born-Oppenheimer perturbation theory, therefore parameters only approximately scale as integer powers of μ.
- Accuracy for sensitivity coefficient is limited by approximate scaling of parameters.
- No need to use complex Hamiltonians, which provide spectroscopic accuracy for transition frequencies, but have many parameters ($\sim 10^2$).
- Simple models provide 10 – 20% accuracy even for large sensitivity coefficients.

Spectrum of NH_2D



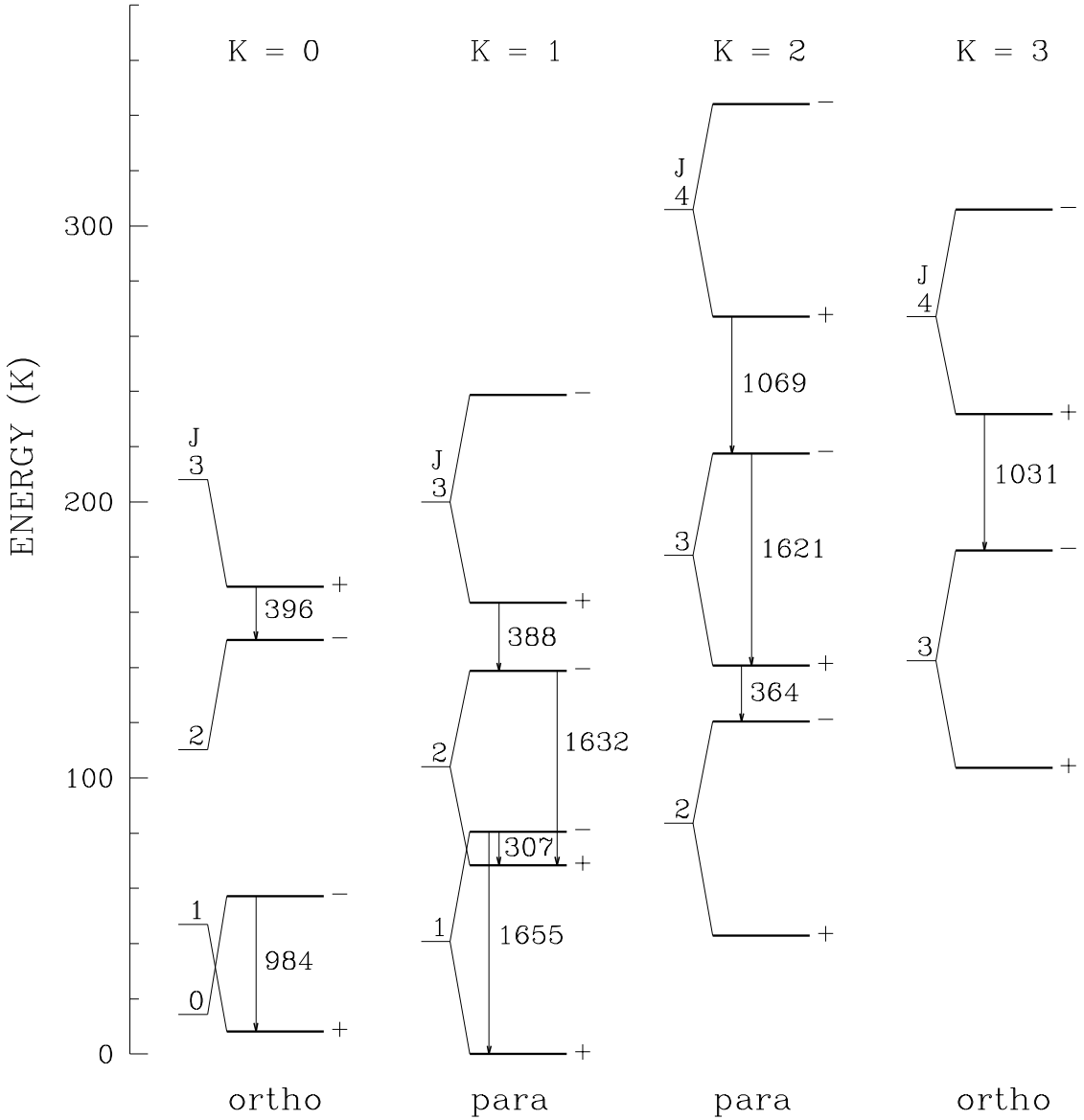
Mixed transitions in H_3O^+, H_2O_2, and CH_3OH

In hydronium ion H_3O^+ and in peroxide the inversion frequencies (55 and 11 cm^{-1}) are much higher, than in ammonia and are comparable to rotational frequencies. Because of that, there are “low frequency” mixed transitions with high sensitivities of different signs. Some of these transitions were observed from the interstellar medium.

In methanol molecule CH_3OH hindered rotation of the OH group is strongly mixed with rotation of the molecule as a whole. Again, there are several low frequency mixed transitions with enhanced sensitivity [Jansen *et al.* 2011]. Very strong methanol lines are often seen from warm molecular clouds with $n > 10^6\,\mathrm{cm}^{-3}$.

Very promising candidates for the μ-variation search!

Low frequency mixed transitions in H_3O^+



Sensitivities of mixed transitions of hydronium ion

Transition						Frequency	Q_μ	ISM
J	K	s	J'	K'	s'	(MHz)		
1	1	-1	2	1	$+1$	307192.41	$+6.4$	✓
3	2	$+1$	2	2	-1	364797.43	-3.5	✓
3	1	$+1$	2	1	-1	388458.64	-3.1	✓
3	0	$+1$	2	0	-1	396272.41	-3.0	
0	0	-1	1	0	$+1$	984711.91	$+2.7$	
4	3	-1	3	3	$+1$	1031293.74	-0.6	
4	2	-1	3	2	$+1$	1069826.63	-0.5	
3	2	-1	3	2	$+1$	1621738.99	$+2.0$	
2	1	-1	2	1	$+1$	1632090.98	$+2.0$	
1	1	-1	1	1	$+1$	1655833.91	$+2.0$	

Sensitivities of mixed transitions of peroxide molecule

$J_{K_A,K_C}(\tau)$ upper	lower	$E_{\rm up}$ (K)	ω (MHz) theory	exper.	Q_μ	ISM
$0_{0,0}(3)$	$1_{1,0}(1)$	17	14818.8	14829.1	$+37(3)$	
$2_{1,1}(1)$	$1_{0,1}(3)$	21	37537.0	37518.3	$-13(1)$	
$1_{0,1}(3)$	$1_{1,1}(1)$	19	67234.5	67245.7	$+8.8(6)$	
$2_{0,2}(3)$	$2_{1,2}(1)$	24	68365.3	68385.0	$+8.7(6)$	
$3_{0,3}(3)$	$3_{1,3}(1)$	31	70057.4	70090.2	$+8.5(6)$	
$3_{0,3}(3)$	$2_{1,1}(1)$	31	219163.2	219166.9	$+3.4(2)$	✓
$6_{1,5}(1)$	$5_{0,5}(3)$	66	252063.6	251914.7	$-1.1(2)$	✓
$4_{0,4}(3)$	$3_{1,2}(1)$	41	268963.7	268961.2	$+3.0(2)$	✓
$5_{0,5}(3)$	$4_{1,3}(1)$	53	318237.7	318222.5	$+2.7(1)$	✓

✓Bergman *et al.* (2011)

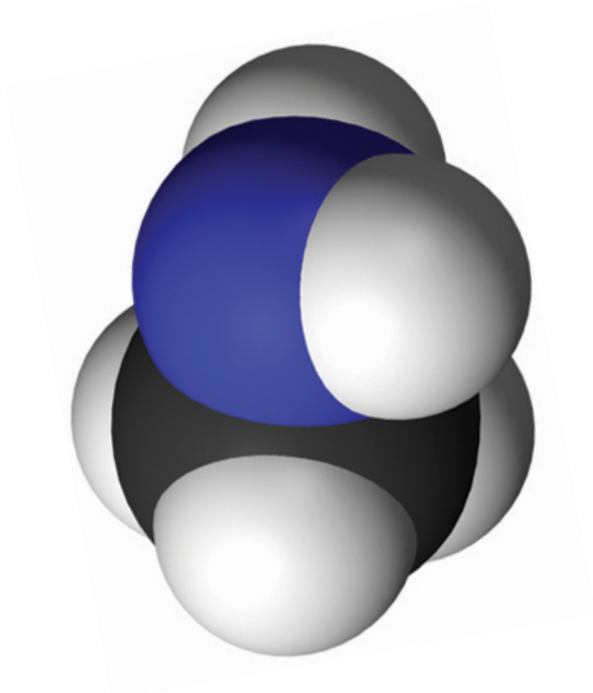
Sensitivities of mixed transitions of methanol molecule

J_1	K_1	J_2	K_2	σ	ν(MHz)	T(K)	Q_μ	ISM
6	0	5	1	A^+	6669	48	+42.9	✓
3	−1	2	0	E	12179	19	+32.4	✓
3	1	3	2	E	24929	35	−16.6	✓
4	1	4	2	E	24933	43	−16.6	✓
2	1	2	2	E	24934	27	−16.6	✓
3	0	4	−1	E	36169	26	−9.6	✓
6	1	7	0	A^+	44069	63	−5.3	✓
2	−1	1	0	E	60531	13	+7.3	✓✓
4	0	3	1	A^+	107014	23	+3.6	✓
5	−1	5	0	E	157179	39	+3.4	✓
4	−1	4	0	E	157246	27	+3.4	✓
2	0	2	1	A^+	304208	7	+1.9	✓

✓Milky Way; ✓✓ redshift $z = 0.89$ (Muller *et al.* 2011)

Methylamine

- Tunneling modes have following frequencies: 7480.542 MHz (torsion mode) and 3098.372 MHz (wagging mode). Thus, we can expect enhanced sensitivity for mixed transition in a few GHz range.
- Recently methylamine was detected for the first time in a high redshift object ($z = 0.89$) [Muller *et al.* 2011].



Sensitivity coefficients for methylamine are calculated by Vadim Ilyushin *et al.*

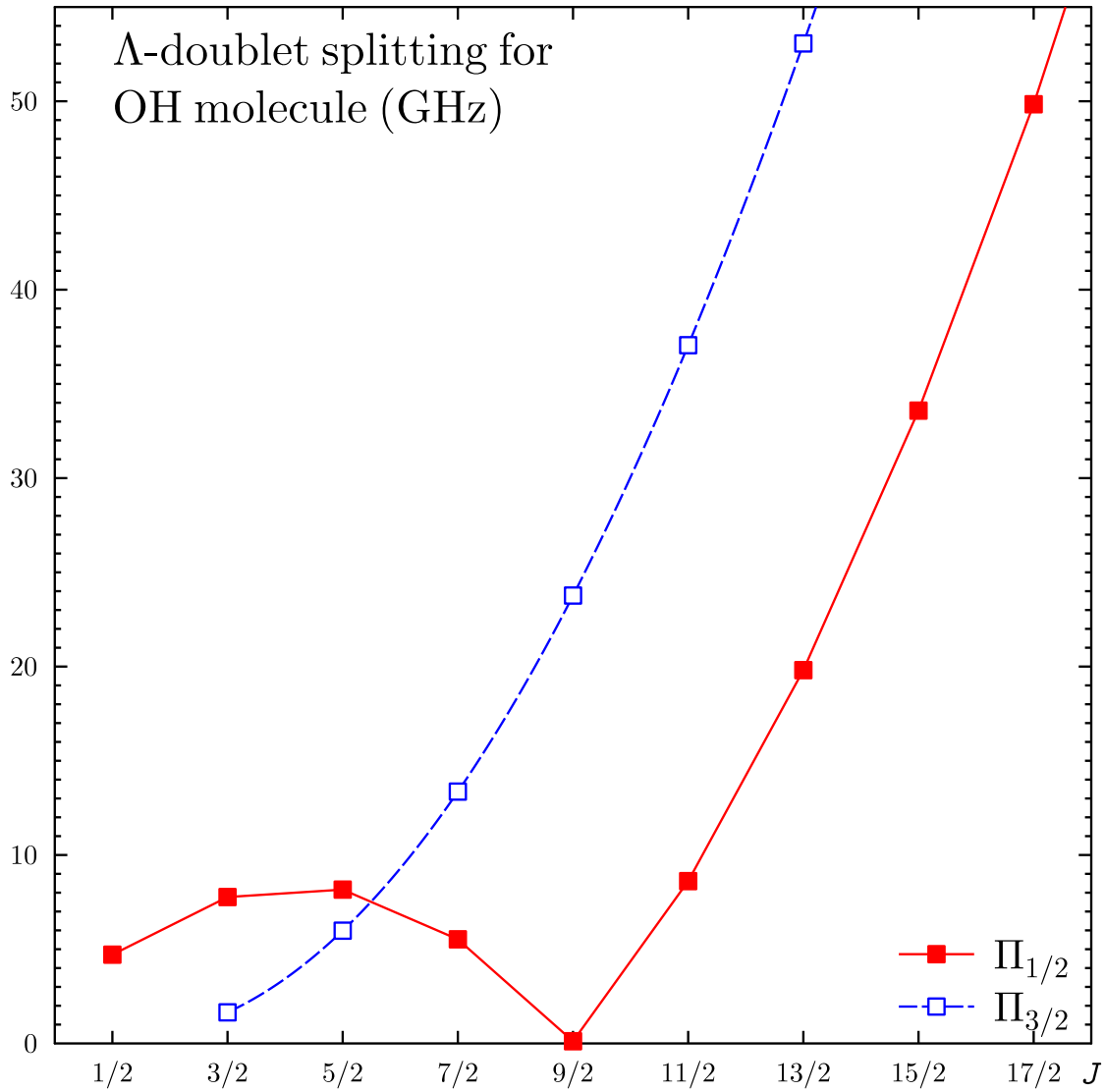
Λ-doublet transitions in OH, CH, NO, and NH^+

In molecules OH and CH electronic spin $\boldsymbol{S}$ is weakly coupled to the molecular axis for low J and decoupled from the axis for higher J. This leads to gradual transformation of Ω-doubling into Λ-doubling.

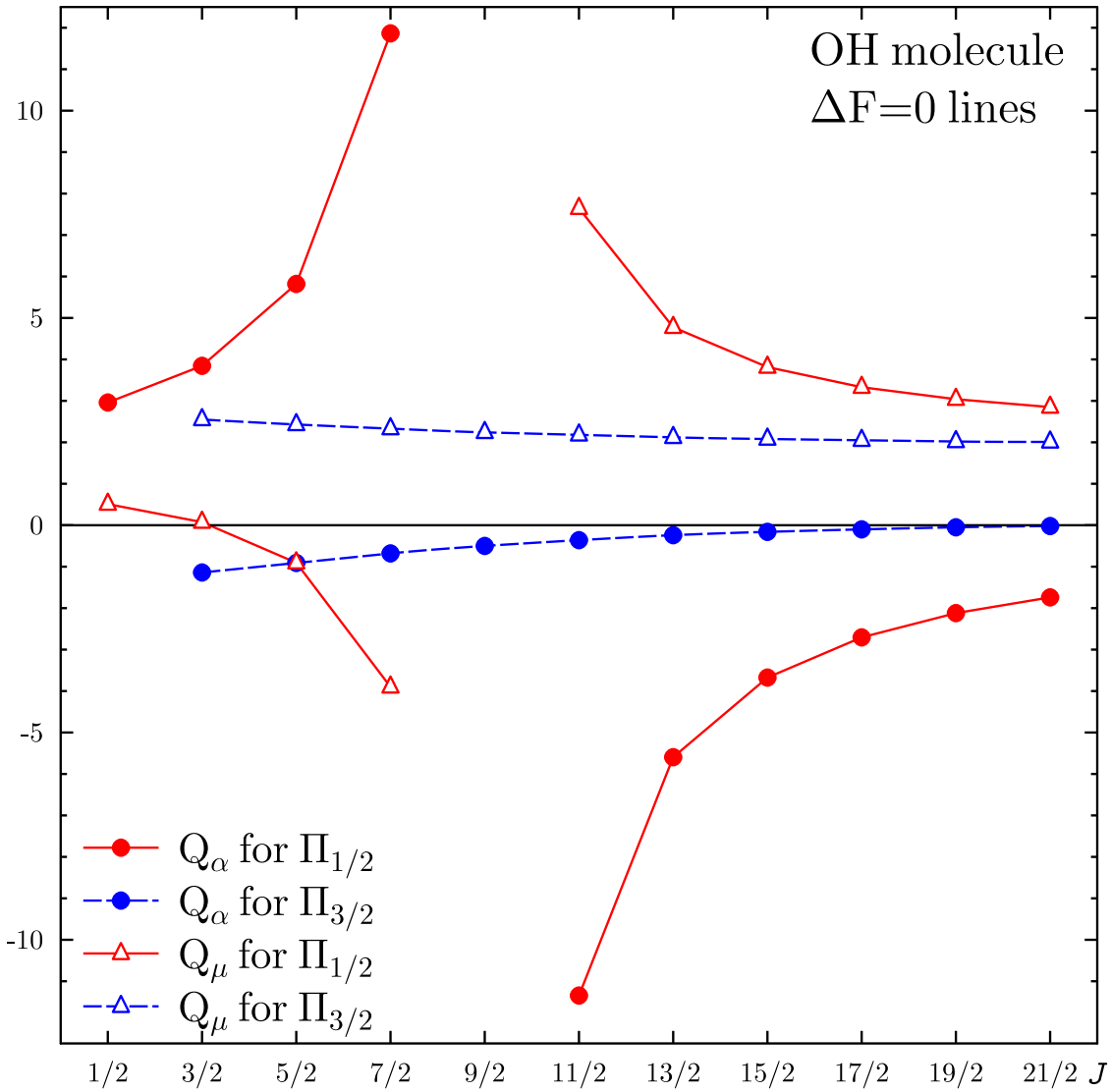
For electronic state $\Pi_{1/2}$ of OH molecule and $\Pi_{3/2}$ state of CH molecule the decoupling of $\boldsymbol{S}$ causes line crossing and huge enhancement of the sensitivity coefficients Q_α and Q_μ. Sensitivity coefficients for two other fine structure levels smoothly depend on the quantum number J.

In NH^+ ion additional enhancement is caused by proximity of the level $^4\Sigma^-$. Separation between $^2\Pi$ and $^4\Sigma^-$ levels is only about 300 cm^{-1}.

Frequencies of Λ-transitions in OH



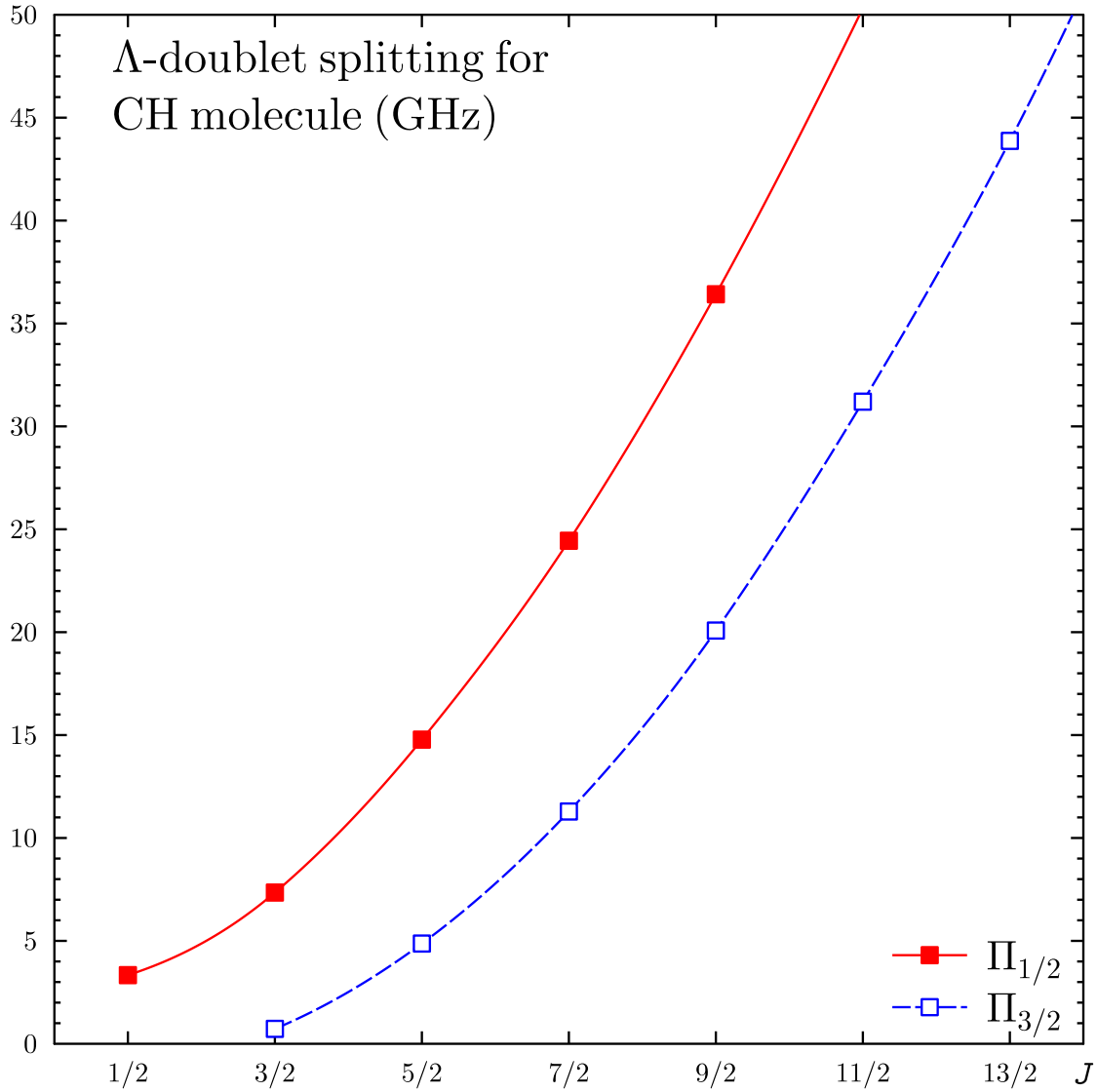
Sensitivities for Λ-transitions in OH



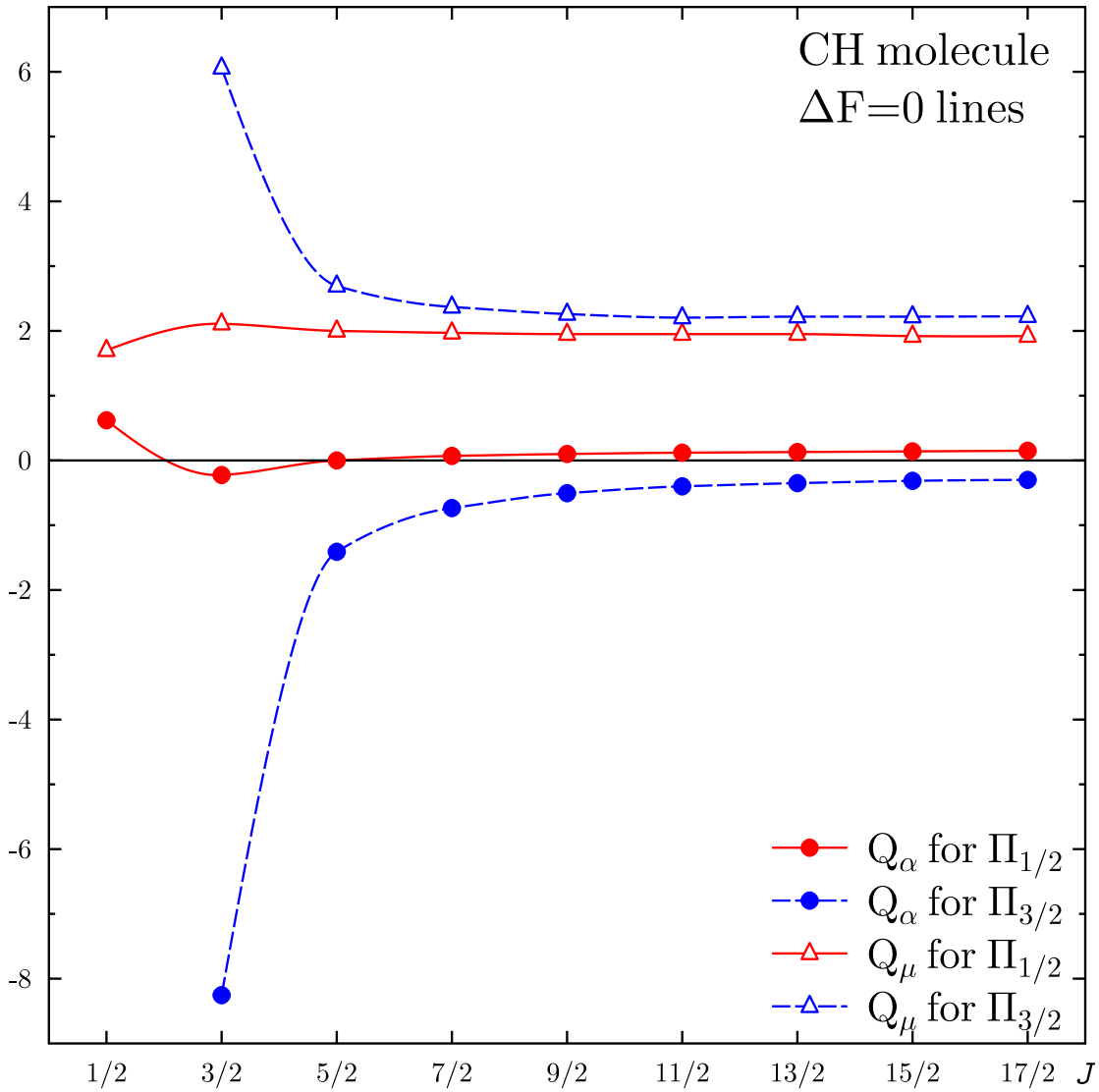
Frequencies and sensitivities of hyperfine structure for $J = \frac{9}{2}$ transition in $^2\Pi_{1/2}$ state of OH.

		ω (MHz)				
F	F'	Recom.	Uncert.	Q_α	Q_μ	Q_g
5	4	88.9504	0.0011	−921.58	459.86	−0.56
5	5	117.1495	0.0011	−699.65	349.59	−0.19
4	4	164.7960	0.0011	−496.67	248.77	0.16
4	5	192.9957	0.0011	−424.05	212.68	0.28

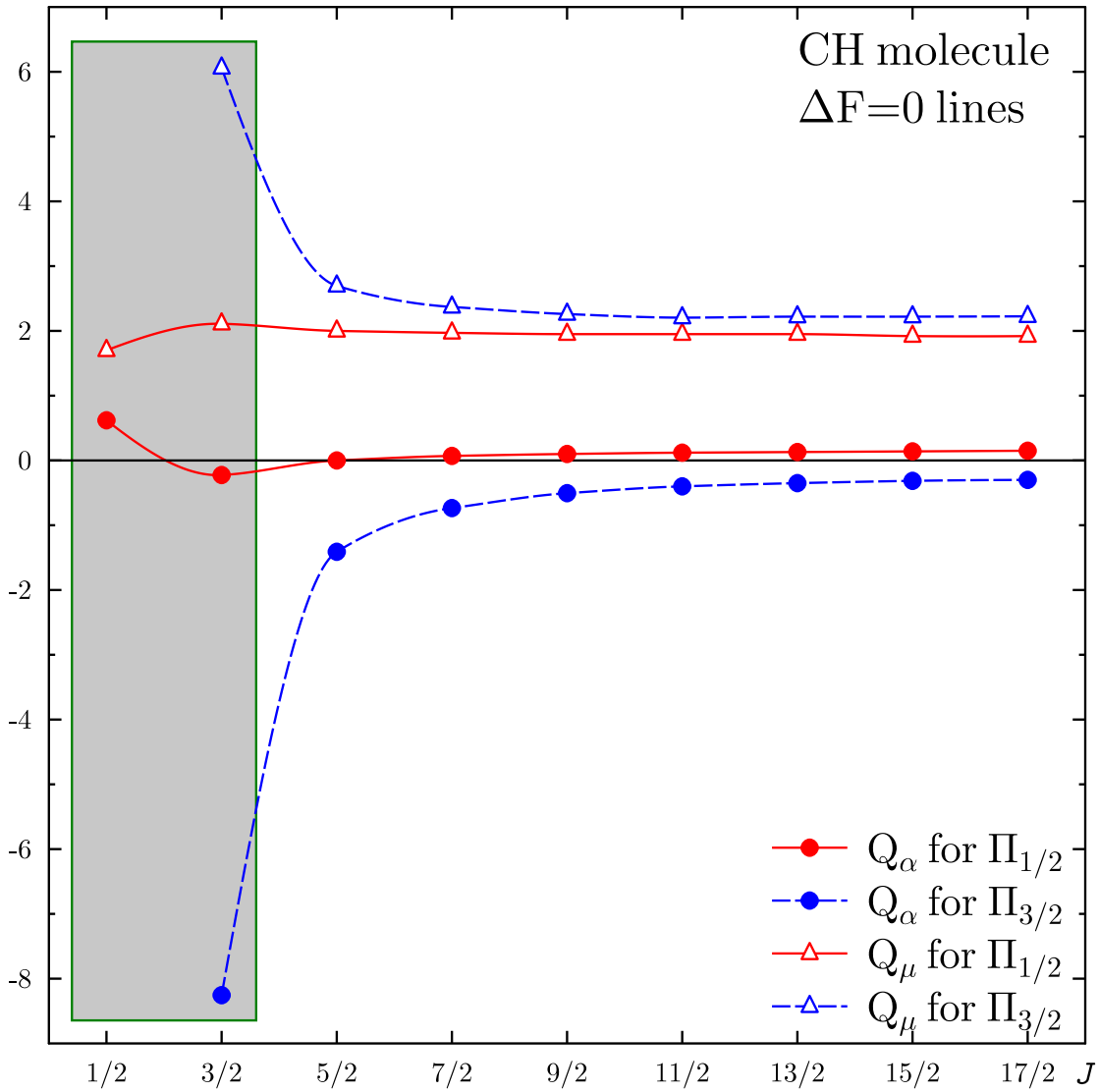
Frequencies of Λ-transitions in CH



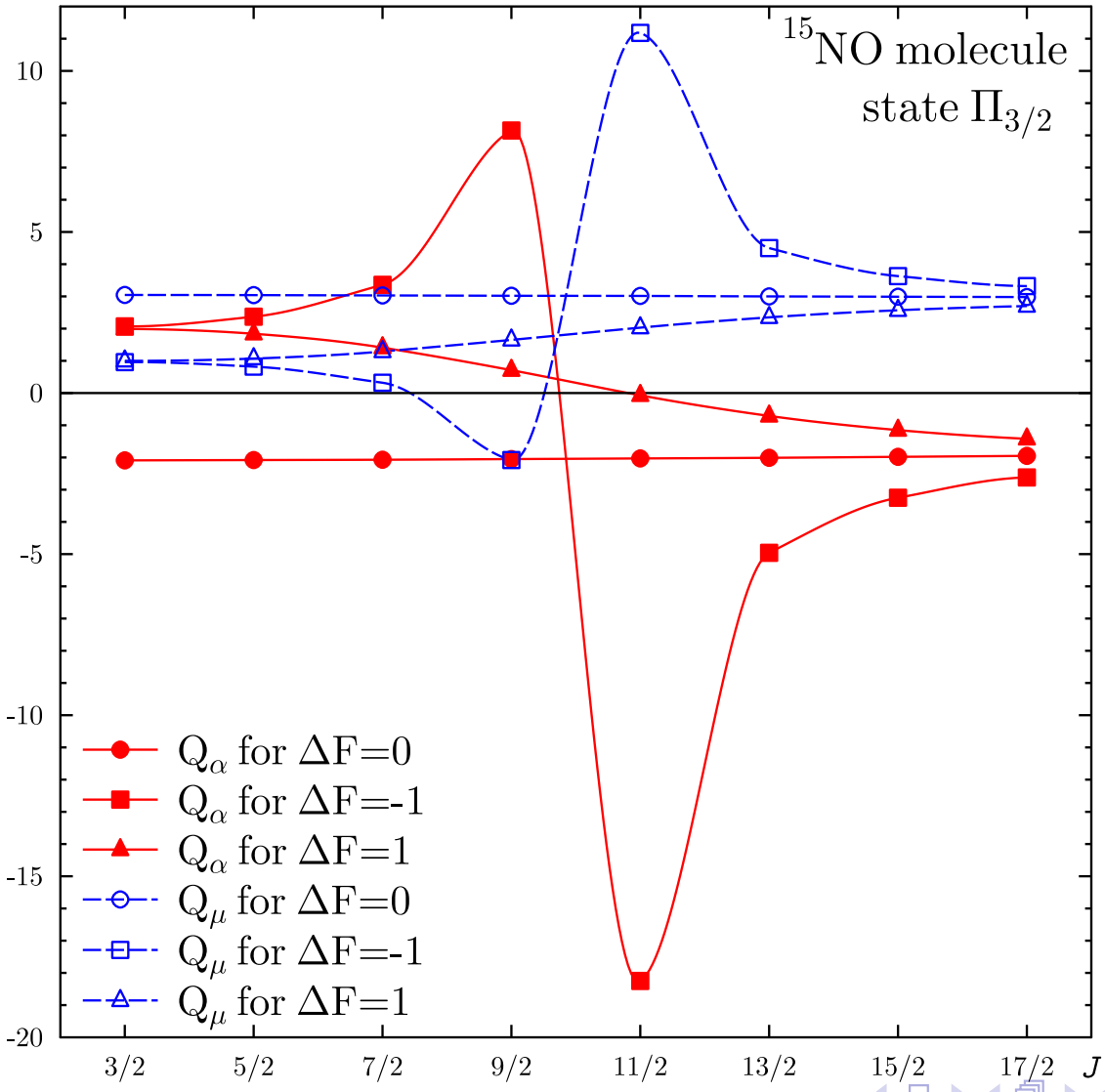
Sensitivities for Λ-transitions in CH



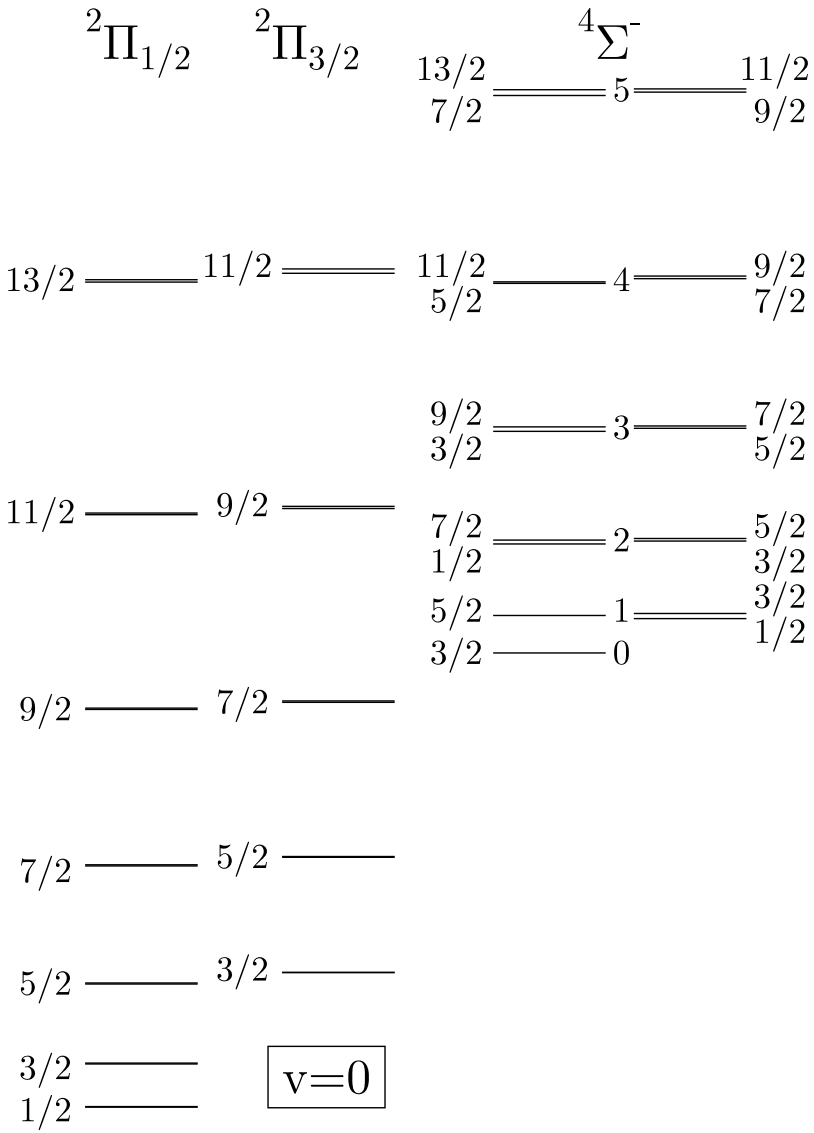
Sensitivities for Λ-transitions in CH

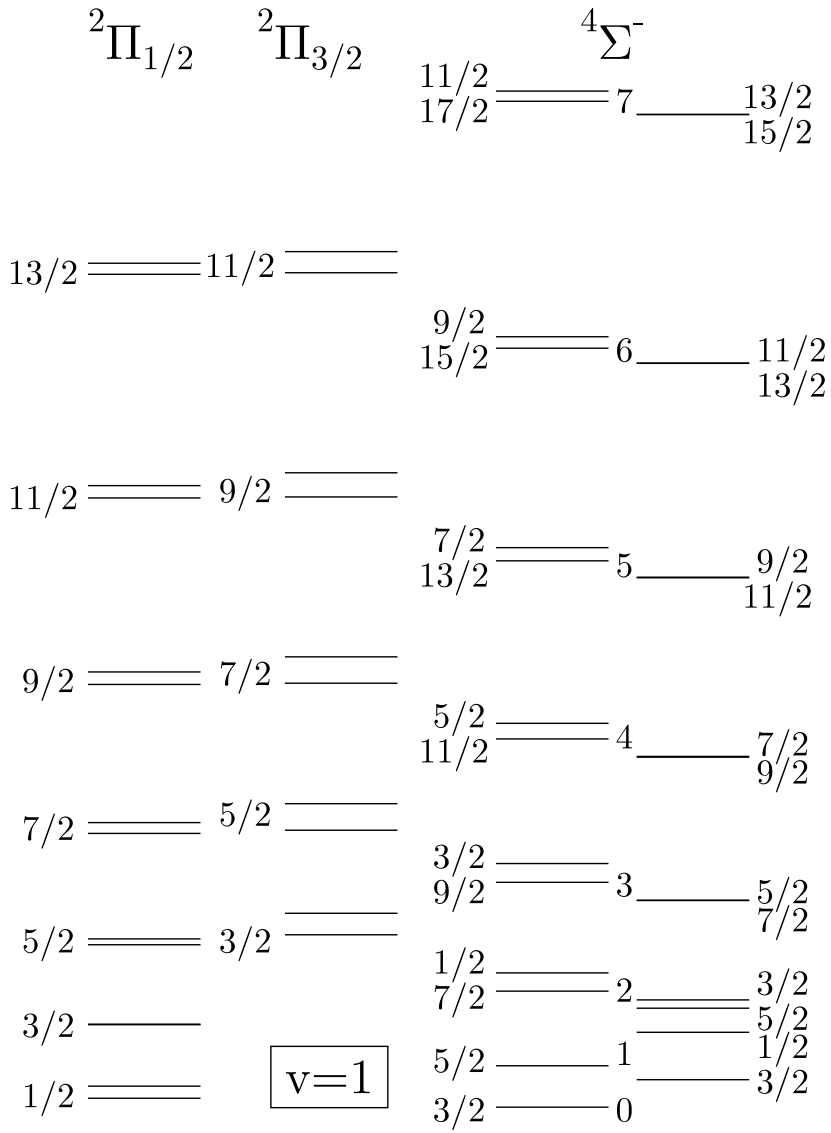


Sensitivities of hyperfine components of Λ-transitions in $^2\Pi_{3/2}$ state of ^{15}NO

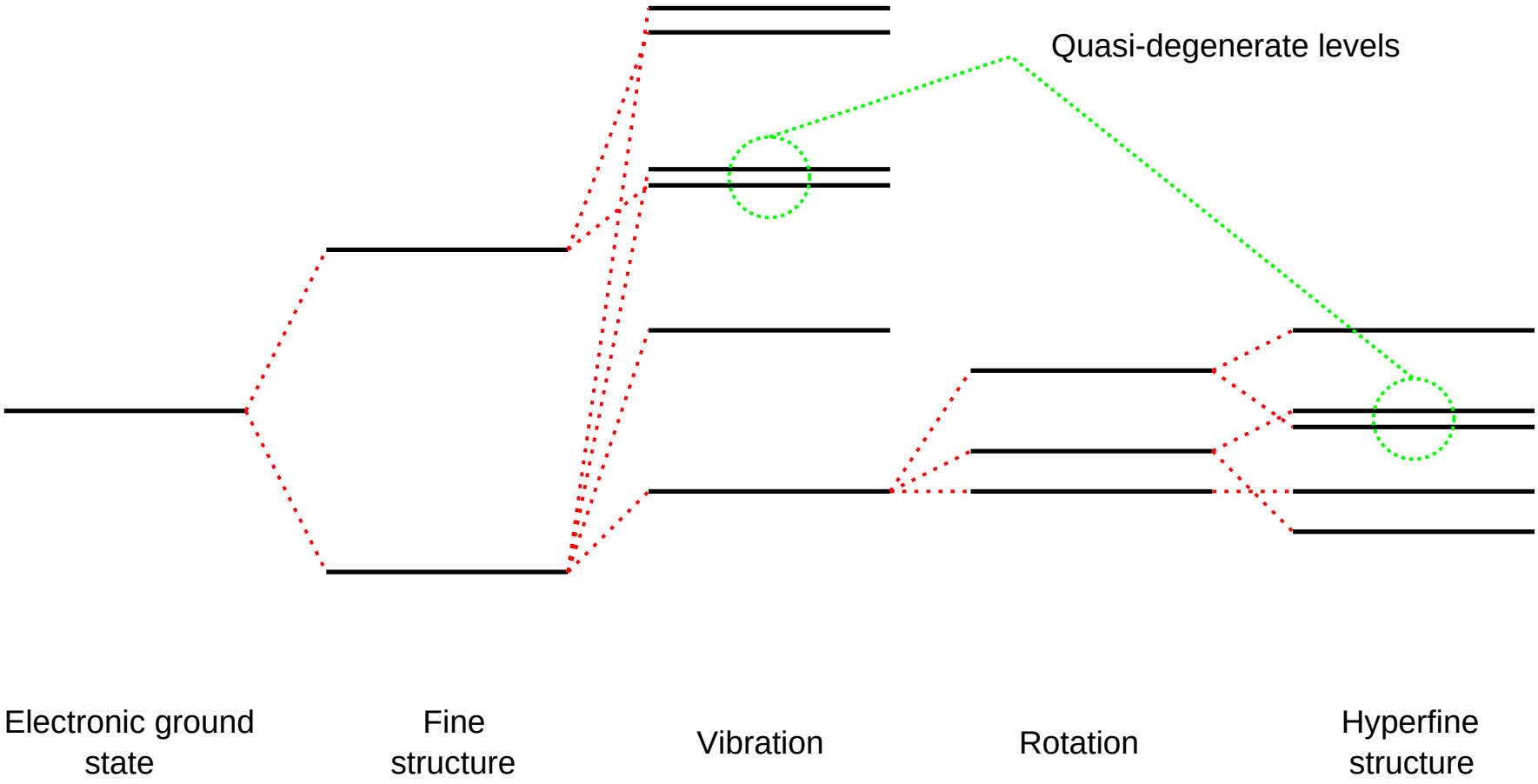


Level structure of NH^+ ion

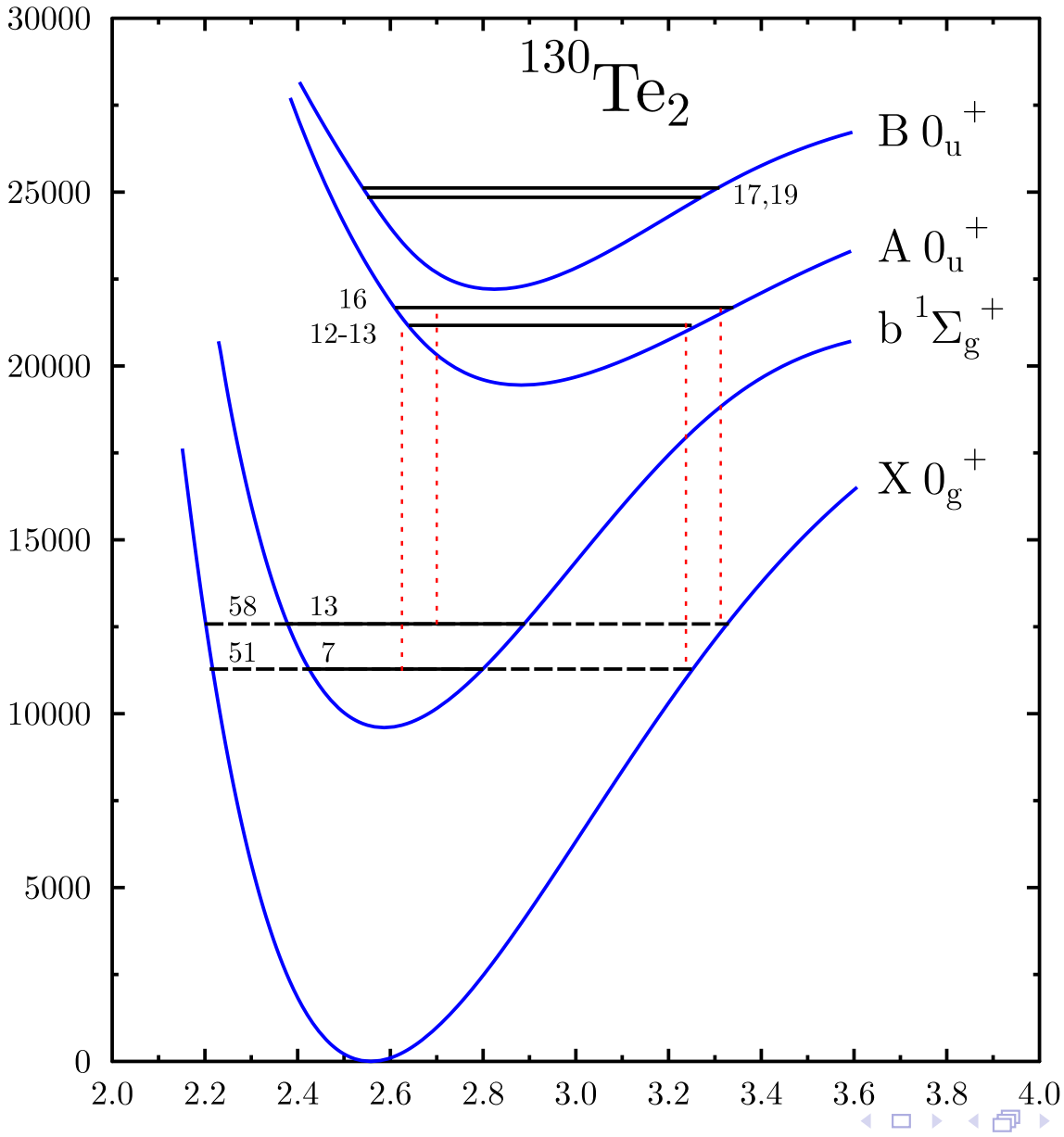




Accidental degeneracies in diatomics



Te_2 [DeMille *et al.* 2011]



Laboratory reference frequencies for astrophysics

Astrophysicists need laboratory frequency measurements with relative accuracy 10^{-8} for following transitions:

- Mixed inversion-rotational lines for NH_2D, H_3O^+, CH_3OH, H_2O_2, and CH_3NH_2;
- 3.3 GHz, 7.3 GHz, and 720 MHz Λ-doublet lines in CH;
- 4.8 GHz and 6.0 GHz Λ-doublet lines in OH;
- Rotational lines $2_1 - 1_0$ for CCS and $1 - 0$ for N_2H^+.

Publications

- V F Flambaum & M G Kozlov, Phys.Rev.Lett. **98**, 240801 (2007).
- S A Levshakov, P Molaro, and M G Kozlov, arXiv: 0808.0583.
- M G Kozlov,Phys.Rev.A, **80**, 022118 (2009).
- P Molaro, S A Levshakov, and M G Kozlov, Nuc. Phys. B Proceedings Supplements, **194**, 287 (2009).
- M G Kozlov, A V Lapinov, and S A Levshakov, J. Phys. B, **43**, 074003 (2010).
- S A Levshakov *et al.* Astron. Astrophys., **524**, A32 (2010).
- M G Kozlov & S A Levshakov, Astrophys. J., **726**, 65 (2011).
- M G Kozlov, S G Porsev and D Reimers, Phys.Rev.A, **83**, 052123 (2011).
- K Beloy *et al.* Phys. Rev. A, **83**, 062514 (2011).