

Axion detection by atomic/molecular transitions

Dark matter axions may cause transitions between atomic states

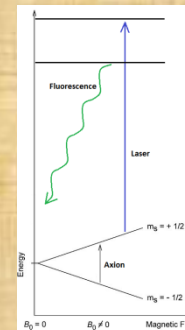
P. Sikivie PRL, 113, 201301, (2014)

Due to very low interaction cross section, it necessary to look for axion induced transition on a mole of atoms/molecules to find tens of events.

The sample has to be cooled at low temperature T in order to populate only the ground state. In particular, on an Avogadro number of particles, the population of the "eventually axion excited state" must be zero

$$N_A \text{Exp}\left(-\frac{E_{ex} - E_{gr}}{kT}\right) \ll 1$$

Detection



The axion mass is unknown and the frequency of transition is proportional to axion mass

- Buffer gas cooling
- Atom/molecule matrix isolation spectroscopy
- Rare earth and transition metal doped crystal
- Atom/molecule in parahydrogen matrix

Buffer Gas cooling

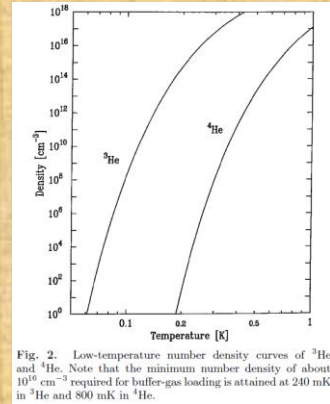
Gas Sample

Laser cooling, Stark or Zeeman selection
can not cool such amount of particles



He3 BGC

R. deCarvalho, Eur. Phys. J. D 7, 289-309 (1999)



The characteristics of the candidate molecule are:

- considerable vapour pressure for capillary filling
- large rotational relaxation cross section
- the Zeeman splitting has to be in axion mass range of interest

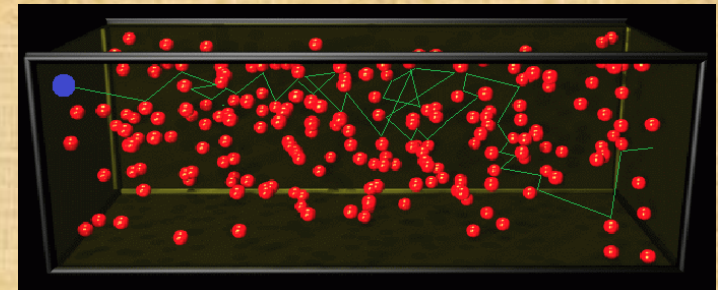
It is necessary to calculate if all particles are thermalized for the available helium density at 280 mK. In other word it is necessary to calculate the minimum oxygen path length in a buffer gas environment necessary to thermalize an Avogadro number of particles.

$$\sigma_{el} \approx 10^{-18} m^{-2}$$

^3He density at 280 mK is about $3 \cdot 10^{16} \text{ cm}^{-3}$

$$\sigma_{rot} \approx 10^{-20} m^{-2}$$

Random walk (velocity dependent) of an oxygen molecule in a Helium 3 environment



Molecular Oxygen

Only odd N in ground state

$$\Psi_t = \Psi_e \Psi_v \Psi_r \Psi_n$$

Ψ_t symmetric under nuclei exchange

Ψ_n symmetric (^{16}O spin = 0)

Ψ_v symmetric (it depends only on R)

\sum_g^3 is the e.g.s. (antisymmetric)

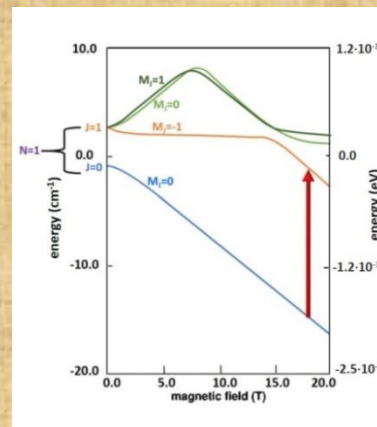


$$\Psi_r$$

antisymmetric \Rightarrow N odd

The good quantum number are:

N, J, M



The transition frequency is tuned by magnetic field (1.2 - 1.9 meV)

The molecule promoted to (1,1,-1) state is detected by exciting it to B5 state (B5 P1Q1 transition) and monitoring the fluorescence. ($S = 1.2 \cdot 10^{(-20)}$ cm, $A = 228$ Hz, 52556 cm $^{-1}$) (several transition)

Being a gaseous molecular system the interpretation of result is simple
The linewidth is about 25 KHz

The acquisition time required for a single event is several days

Matrix isolation Spectroscopy

Originally developed to trap or produce chemical species and preserving them in solidified inert gases at cryogenic temperatures, MIS has grown to become a methodology for obtaining information on atomic/molecular structures and chemical reactions

G. C. Pimentel, J. Am. Chem. Soc., **80**, 62-64 (1958)

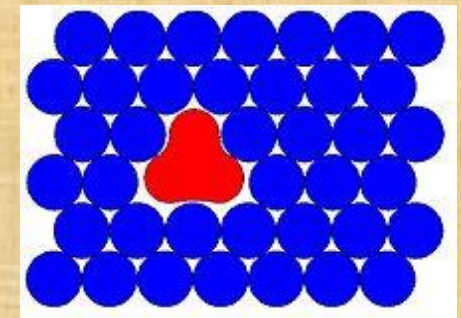
D. C. Silverman and M. E. Fajardo, J. Chem. Phys., **106**, 8964

Y. Bouteiller et al. Chem. Phys. **386** (2011) 29-40

- provides lower temperatures
- larger sample densities

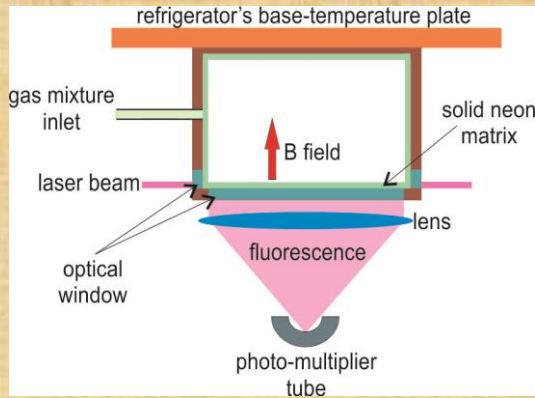
- additional broadening in the spectral linewidth of the matrix-isolated species (in comparison to the gas phase), caused by the interaction with the environment

Rare gas matrices have been widely used in recent studies because of their chemically inert property, excellent optical properties, including isotropy and transparency from the far IR to the vacuum UV



Axion detection based on atomic transition does not require high resolution \sim tens of GHz but requires large sample density

Experimental setup



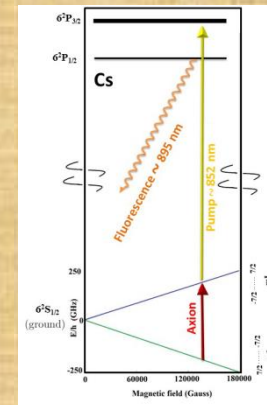
Because of lower obtainable temperature, the axion mass investigation range can be extended

Temperature is lower than BGC \Rightarrow $v_a \approx 1 \text{ cm}^{-1}$
 $m_a \approx 0.1 \text{ meV}$



MIS suitable technique for axion detection

Detection scheme



Considering neon as host matrix and a volume of order of a typical MIS experiment, 10 hours integration time are sufficient.
 It is on order of magnitude less than BGC oxygen but the interpretation is more difficult.

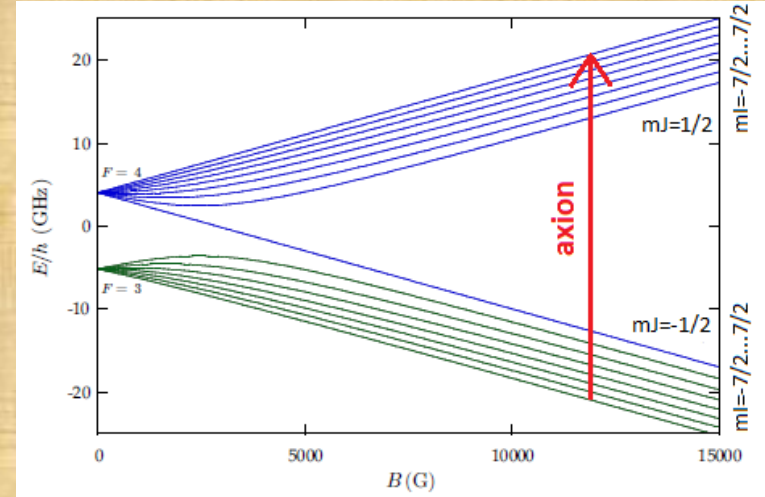
Caesium

Alkali atom $I=7/2$

Ground state $6^2S_{1/2}$

D1 line 894.593 (nm) $6^2S_{1/2} \rightarrow 6^2P_{1/2}$

D2 line 852.347 (nm) $6^2S_{1/2} \rightarrow 6^2P_{3/2}$



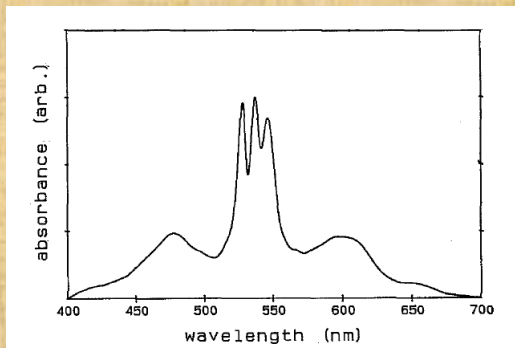
Alkali atom spin exchange

collision cross section $\sigma_{SE} \sim 10^{-28} m^{-2}$ D.F. Kimball, PRA, 82, 062714 (2010)

The thermal relaxation would require a km path in the buffer gas cell

BGC is impossible !

The alkali atoms in noble gas solid matrix (Xe, Kr, Ar) have been already studied and the linewidth is too large for our purposes.



S.Tam J. Chem. Phys. **99**, 854 (1993)

Jacquet 2011 J. Chem. Phys. **135**, 174503 (2011)

Ryan 2010 J. Phys. Chem. A, **114**, 3011–3024, (2010)

Neon is missing

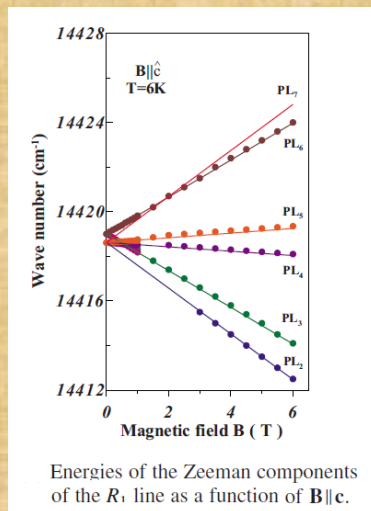
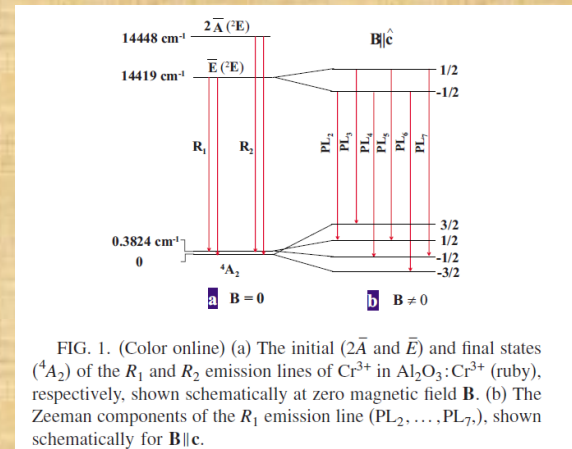
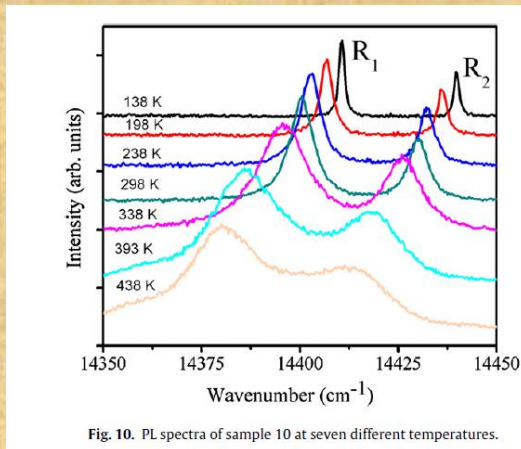
He and Ne polarizability is an order of magnitude lower than other noble gases (He is too hard to solidify)

The detection can be carried out using fluorescence or REMPI with extraction of electron from neon crystal (also molecular Oxygen is a good candidate)

A. W. Wiederkehr Mol Phys. 110, 1807 (2012)

**Rare earth and transition
metal doped crystal**

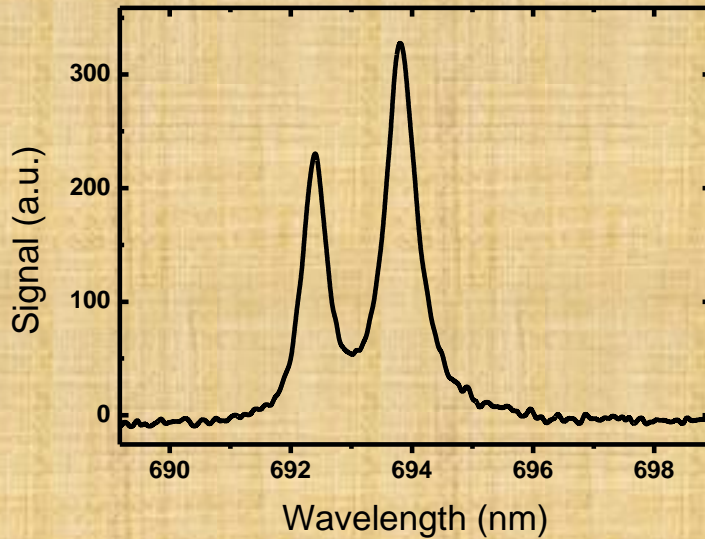
Ruby($Al_2O_3:Cr^{3+}$)



Narrow linewidth,
R1 and R2 are very close (1,5nm)

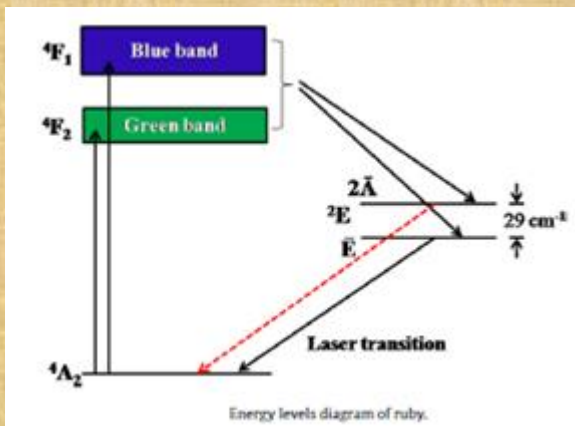
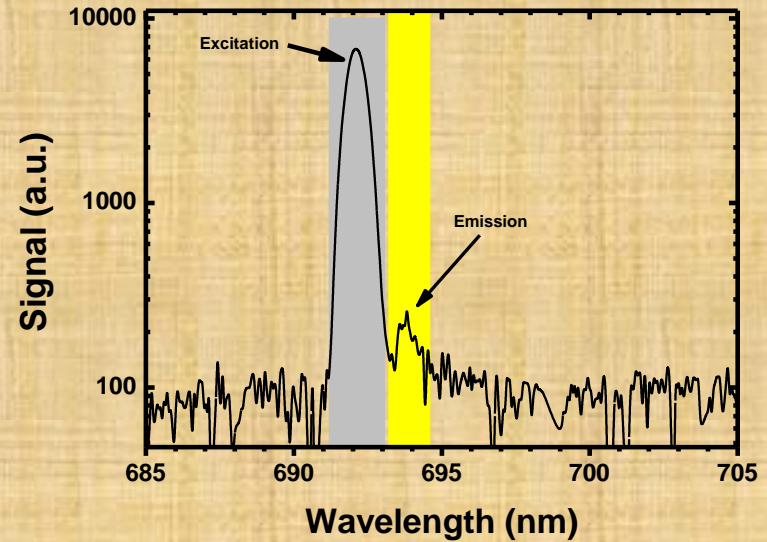
Emission

excitation wavelength 540 nm. Grating 1200 grooves/mm



PLE

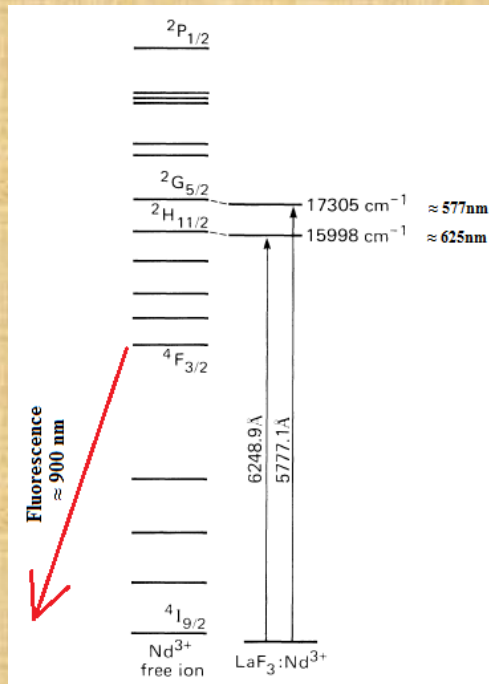
Acquisition mode: 100 um slits, spile-removal multi-mode, acquisition 5 sec)



The peak are resolved also in excitation but the efficiency is low

It is necessary a transmittance spectrum to estimate crystal heating

$LaF_3:Nd^{3+}$



625nm transition
2.4 GHz linewidth

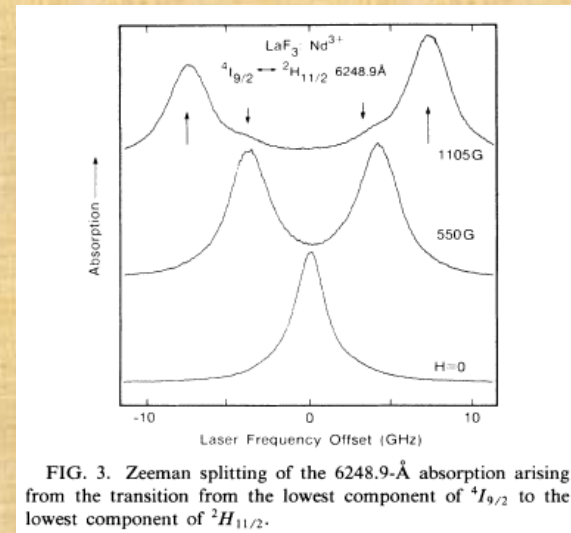
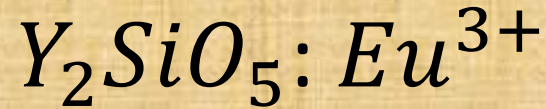


FIG. 3. Zeeman splitting of the 6248.9 Å absorption arising from the transition from the lowest component of $4I_{9/2}$ to the lowest component of $2H_{11/2}$.

Optical transition

R. M. Macfarlane, PRB, vol. 36, n. 7 (1987)

The shape of 577nm transition is not showed but the reported inhomogeneous linewidth is about 3 GHz (50 ppm, 1.6 K)



Pumped at 578nm and detected at 585nm

Unfortunately Eu^{3+} has not linear Zeeman splitting but Y_2SiO_5 crystals exhibit very narrow inhomogeneously broadened linewidths, are commercial crystals and can be doped also with Er^{3+} (very narrow) and Nd^{3+}

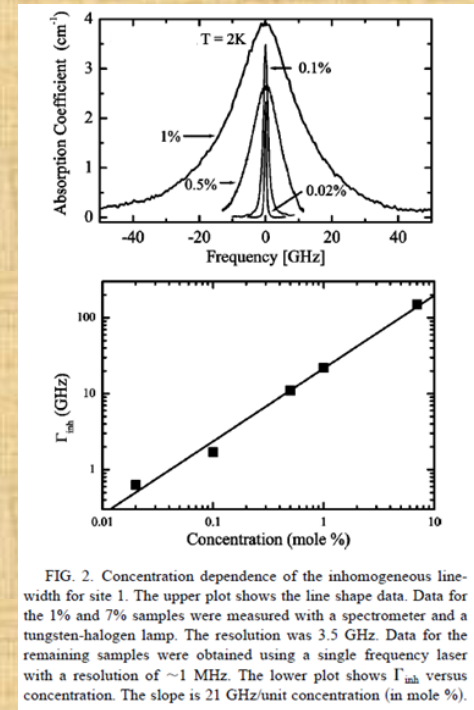
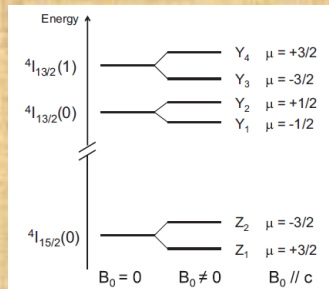


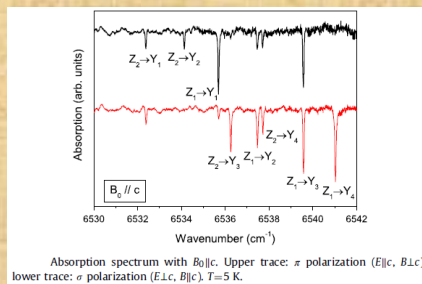
FIG. 2. Concentration dependence of the inhomogeneous linewidth for site 1. The upper plot shows the line shape data. Data for the 1% and 7% samples were measured with a spectrometer and a tungsten-halogen lamp. The resolution was 3.5 GHz. Data for the remaining samples were obtained using a single frequency laser with a resolution of ~ 1 MHz. The lower plot shows Γ_{inh} versus concentration. The slope is 21 GHz/unit concentration (in mole %).

Energy level structure and optical dephasing under magnetic field in $\text{Er}^{3+}:\text{LiYF}_4$ at $1.5 \mu\text{m}$

R. Maribo, Journal of Luminescence **169** (2016) 478–482

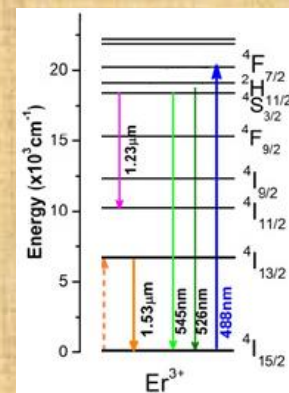


The Zeeman field splitting is in our range of interest (about 100GHz at 2T)



Inhomogeneous linewidth is smaller than the typical hyperfine splitting at zero field

Erbium ion shows more energetic transition for fluorescence detection



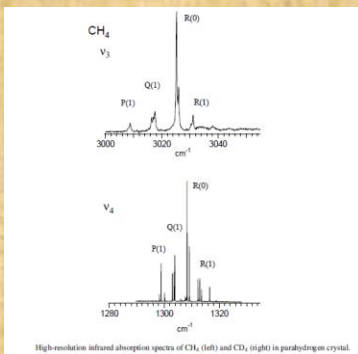
**Atom/molecule trapped in
solid parahydrogen (or methane)**

High resolution spectroscopy is not applicable to system in condensed phase because of spectral broadening due to strong intermolecular/atomic interaction

An exception is represented by PC because of weak intermolecular interaction in solid parahydrogen.

- at cryogenic temperature $J=0 \rightarrow$ spherically symmetric \rightarrow no averaged multipole exists and only the weak force assembles the molecules into quantum solid
- since the binding energy of solid parahydrogen is extremely weak, the neighboring intermolecular separation 0.3783
- the amplitude of the zpv amounts to almost 20% of the intermolecular distance.
- small mass of the constituent

solid parahydrogen provides an ample and "soft" space for guest molecules. The softness allows guest molecules to undergo vibrational and rotational motions with little hindrance.



T. Momose, Vibrational Spectroscopy 34 (2004) 95–108

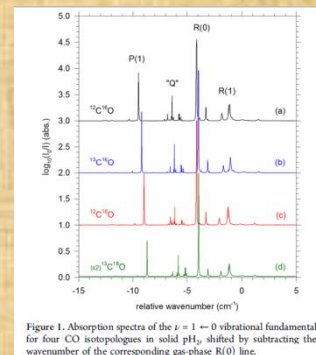
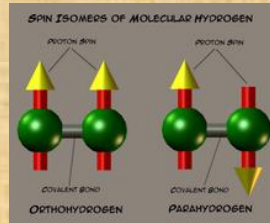


Figure 1. Absorption spectra of the $\nu = 1 \rightarrow 0$ vibrational fundamental for four CO isotopologues in solid pH_2 , shifted by subtracting the wavenumber of the corresponding gas-phase R(0) line.

M.E. Farjardo, J. Phys. Chem. A 2013, 117, 13504–13512

The electronic transition are narrow too?
In IR region are fluorescence measurements possible in MIS?

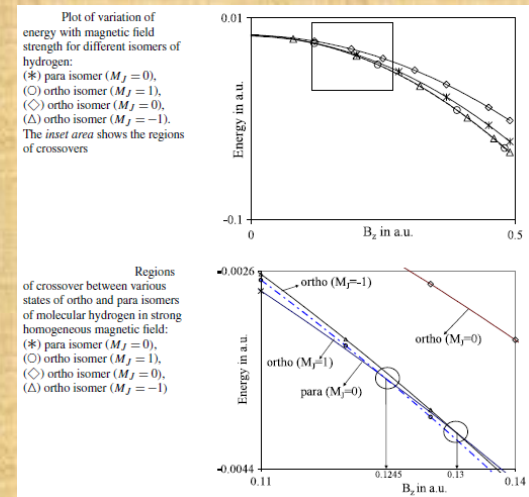
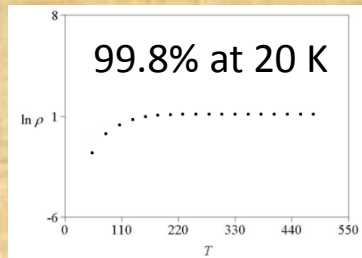
The orto/para fraction



Orto $I=1 \rightarrow J$ odd
 Para $I=0 \rightarrow$ even

At room temperature the ratio is 3:1

$$\rho = \frac{\sum_{J=1,3,\dots} \sum_{M_J} \sum_{M_I} \exp[-E_{ortho}(J, M_J, M_I)/k_B T]}{\sum_{J=0,2,\dots} \sum_{M_J} \exp[-E_{para}(J, M_J)/k_B T]}$$



The crossover take place at very large field

Thank you !