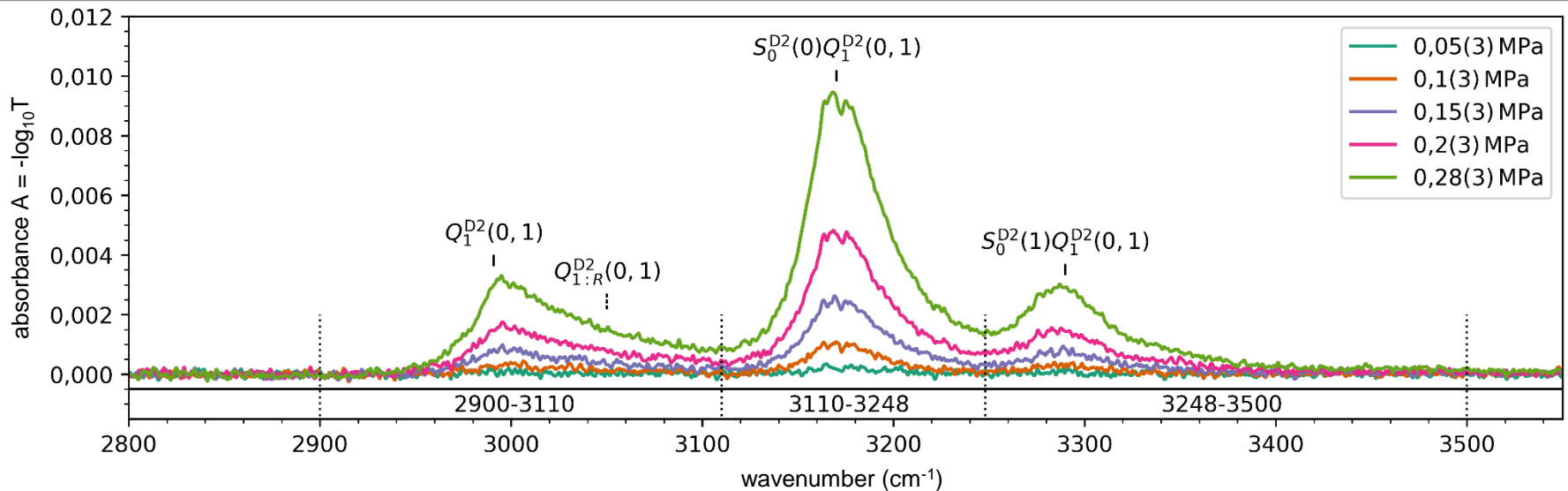
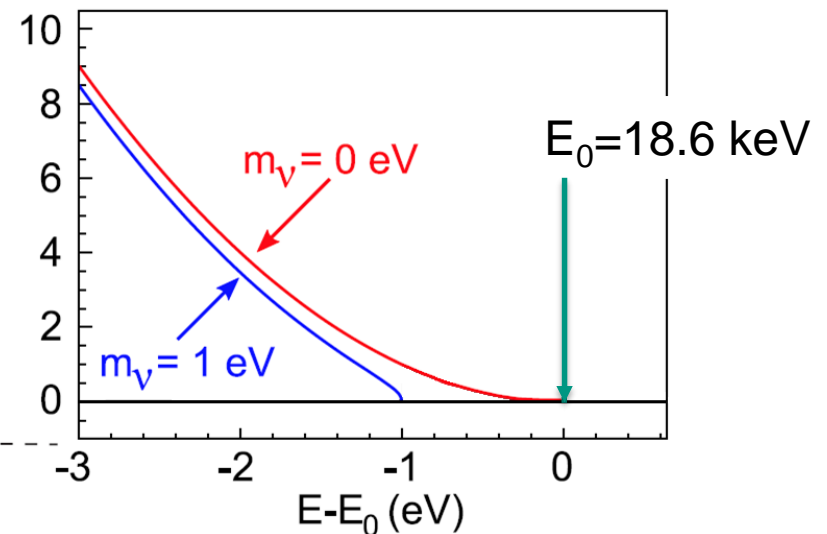
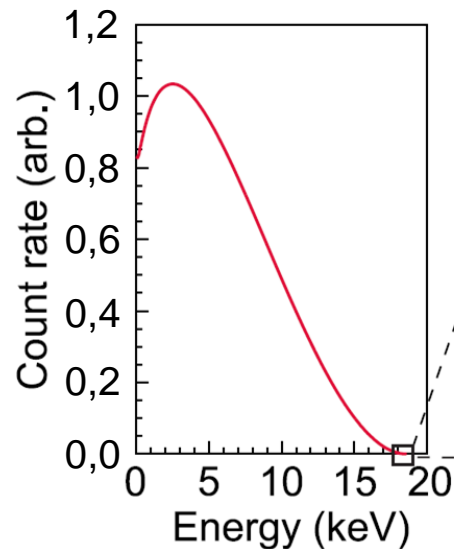
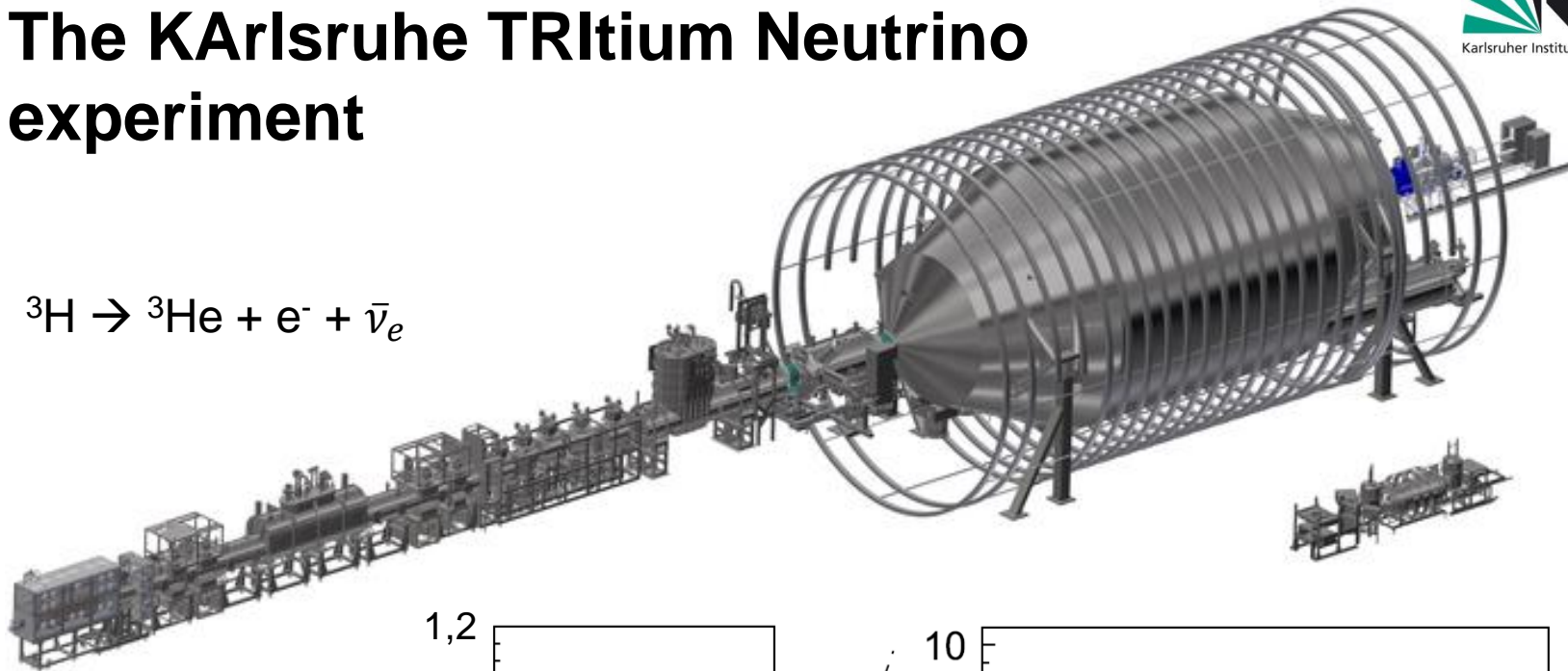
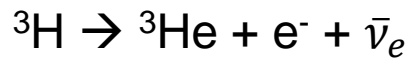


Van-der-Waals cluster formation as a systematic uncertainty for the KATRIN neutrino mass measurement

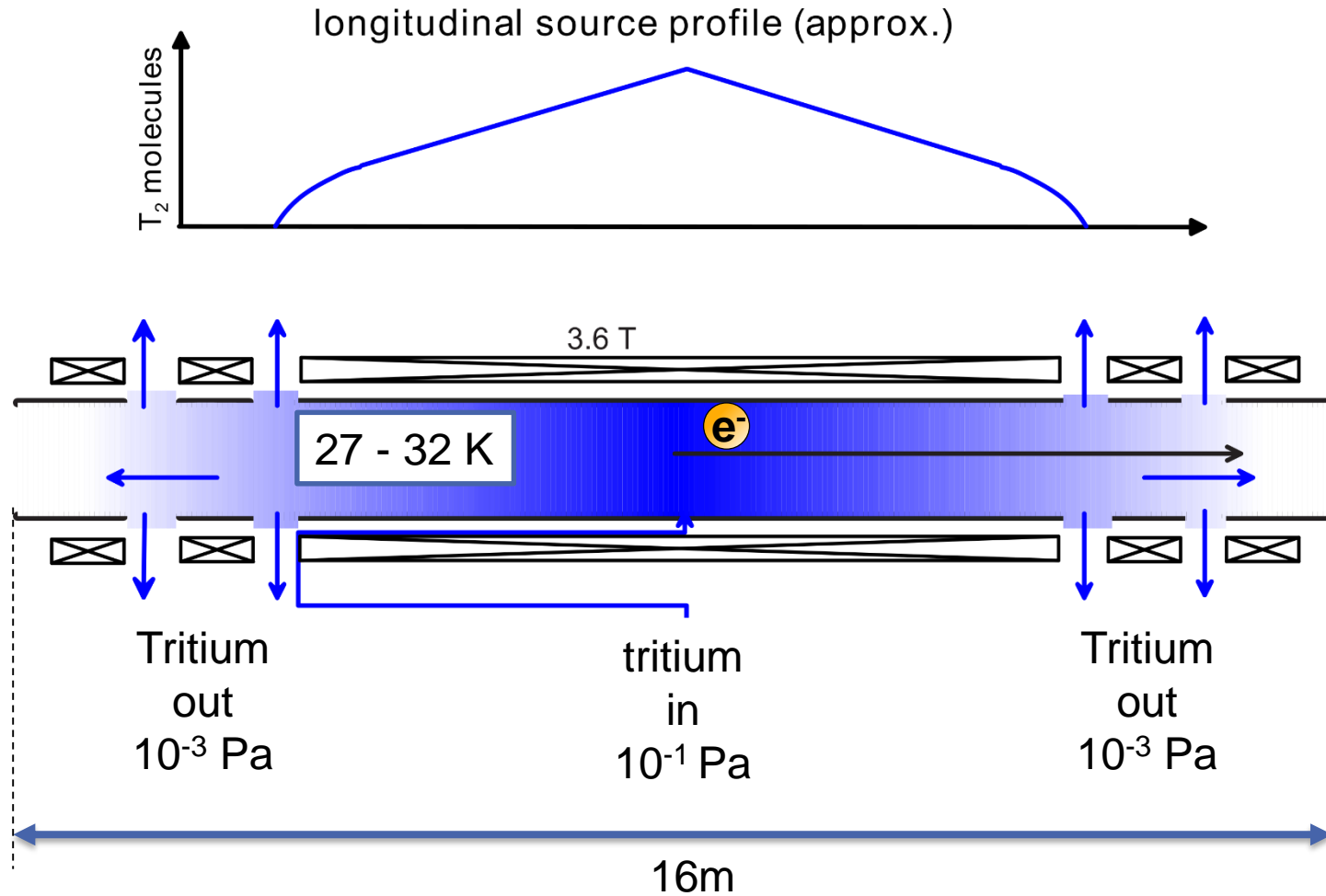
Sebastian Mirz – Tritium Laboratory Karlsruhe, Institute for Nuclear Physics, Karlsruhe Institute of Technology



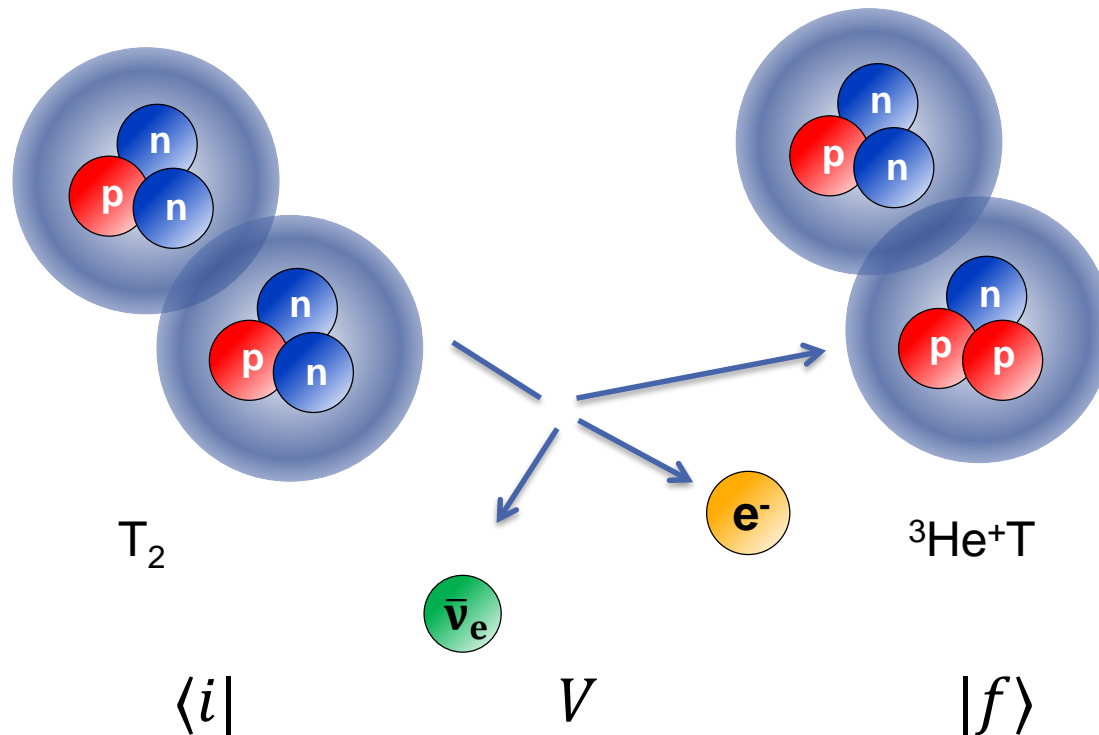
The Karlsruhe TRitium Neutrino experiment



Windowless Gaseous Tritium Source (WGTS)



Molecular β -decay

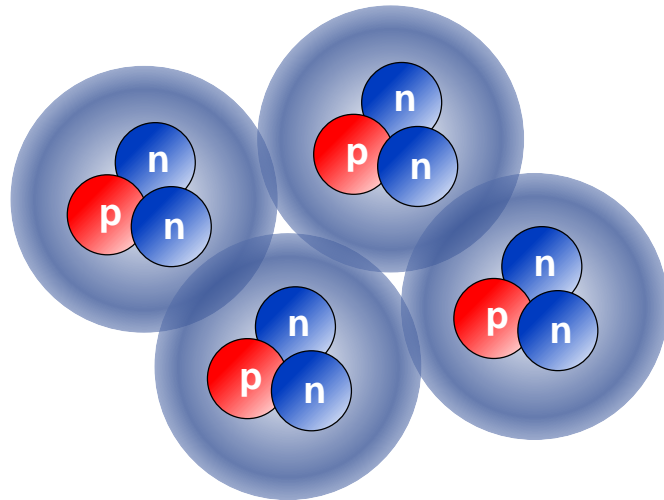


β -spectrum depends on initial and final state distribution

relevant for β -spectrum:

- excitation energy for an initial final state combination E_i
- probability $P(E_i)$

Influence of Van-der-Waals clusters



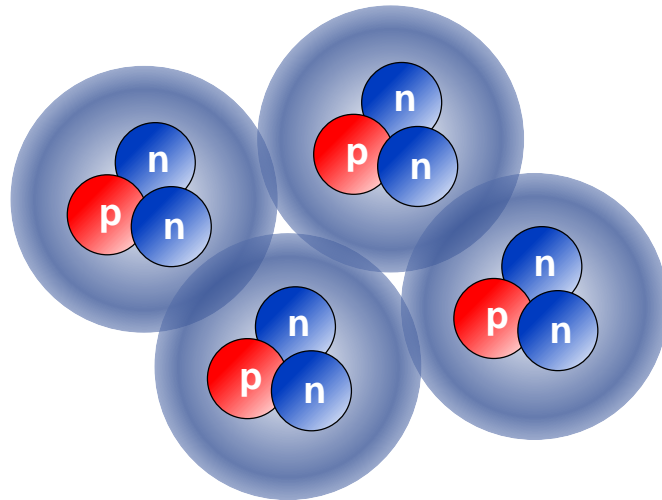
$(T_2)_2$ Van-der-Waals dimer

London dispersion force:

$$w(r) \propto \frac{\alpha_1 \alpha_2}{r^6}$$

α : polarizability

Influence of Van-der-Waals clusters



$(T_2)_2$ Van-der-Waals dimer

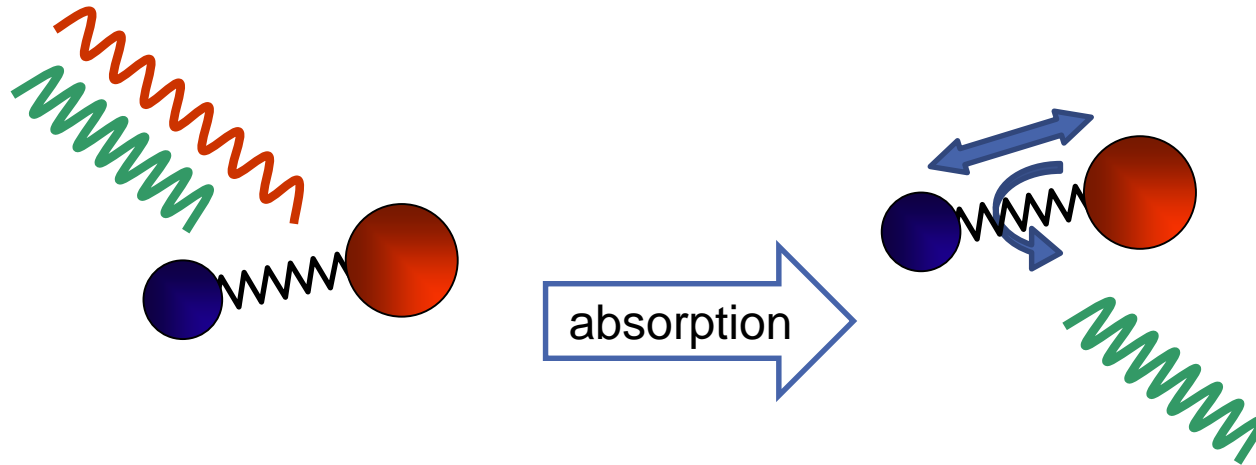
Influence by different...

- ... initial state distribution
- ... binding energy (e.g. 0.36 meV für $(H_2)_2$)
- ... recoil energy (1.7 eV for single T_2)
- ... final state distribution

Cluster formation to be included in uncertainty budget for FSD: 0.02 eV²

KATRIN needs the concentration and excitation energies of the clusters
Problem: No theoretically or experimentally determined values available for HT, DT and T_2

Experimental investigation: IR absorption spectroscopy

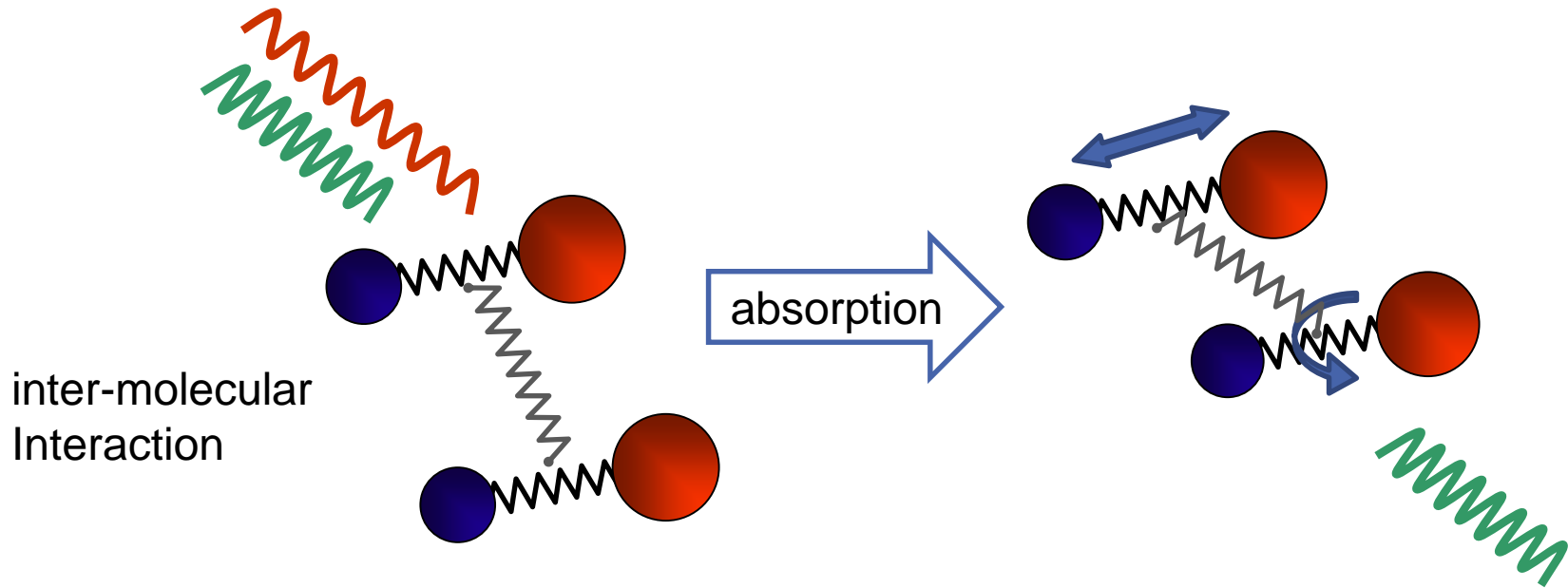


(Dipole-) absorption requires:

- Permanent electrical dipole moment
- or an electrical dipole moment, that changes during vibration

- H_2 , D_2 , T_2 : no electrical dipole moment
- HD , HT , DT : electr. dipole moment negligible ($3,3 \cdot 10^{-34}$ Cm for HD)

Experimental investigation: IR absorption spectroscopy



Inter-molecular interaction: Collision, Van-der-Waals binding
 → induced dipole moment → induced absorption

IR absorption spectroscopy is sensitive to Van-der-Waals clusters

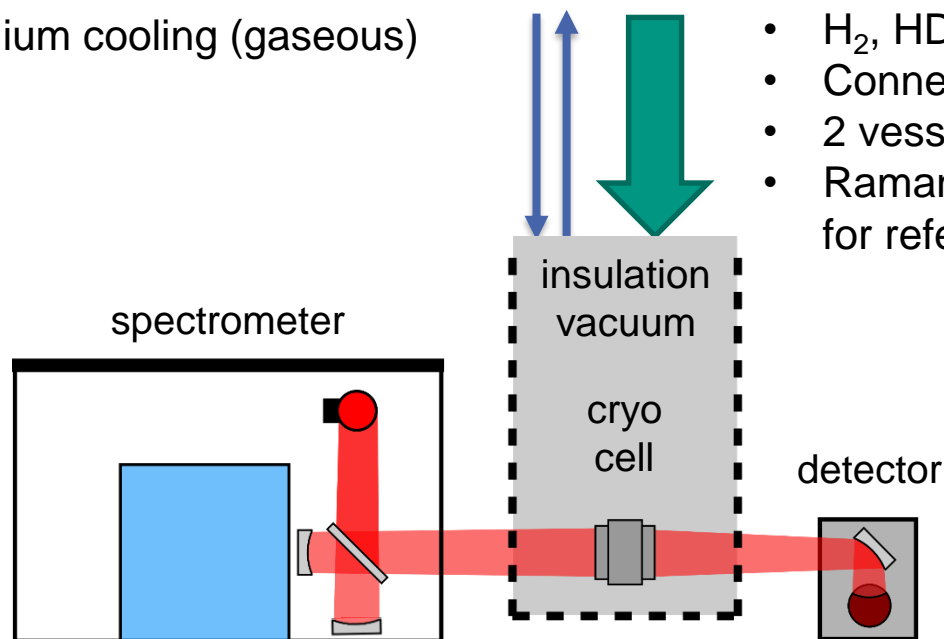
Experiment: TApIR at the Tritium Laboratory Karlsruhe

Cooling:

- helium cooling (gaseous)

Gas supply:

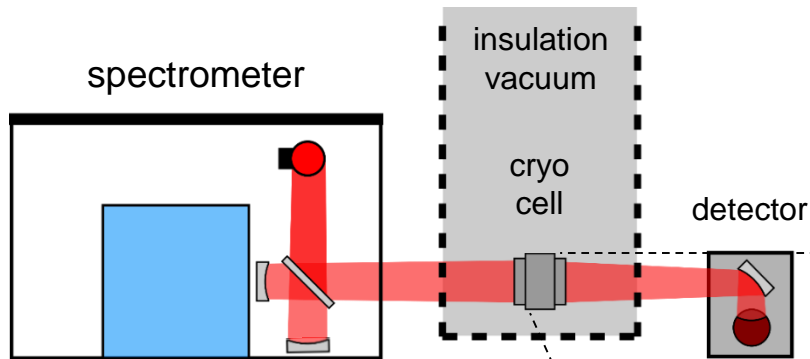
- H₂, HD, D₂
- Connection to cryo distillation
- 2 vessels: 100l + 1l
- Raman system for reference calibration



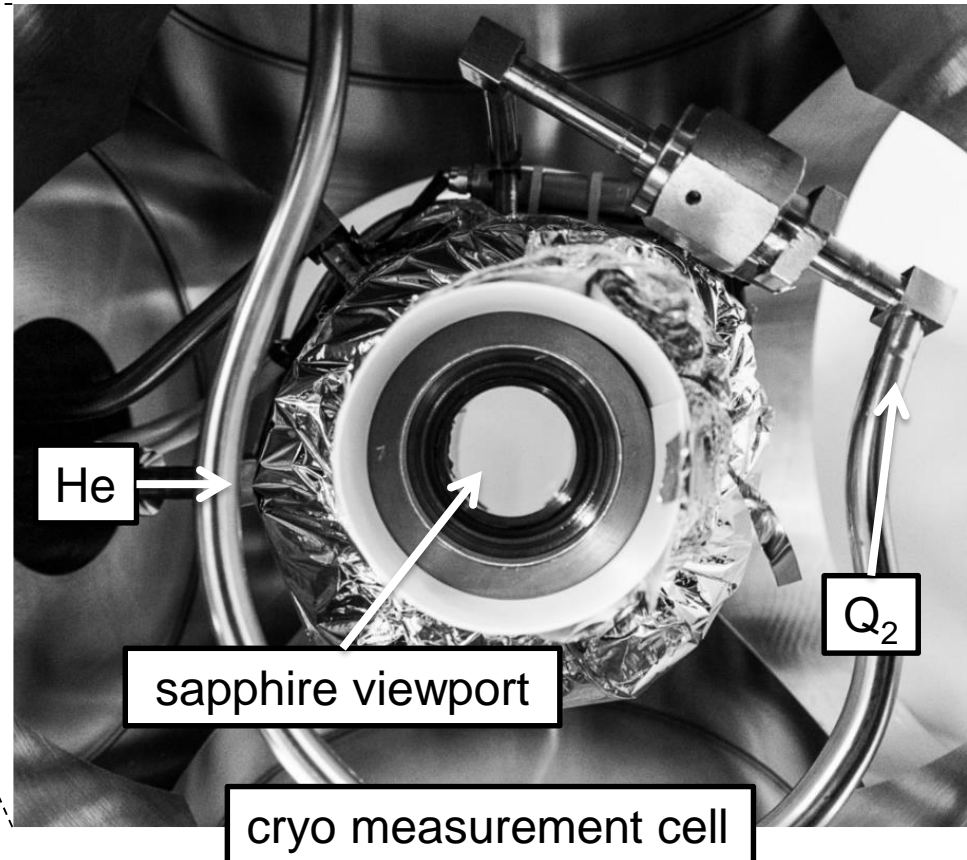
Spectrometer:

- Bruker TENSOR 27: Resolution $< 0.9 \text{ cm}^{-1}$ (0.2 nm @1500 nm)
- Bruker VERTEX 70: Resolution $< 0.4 \text{ cm}^{-1}$ (0.09 nm @1500 nm)

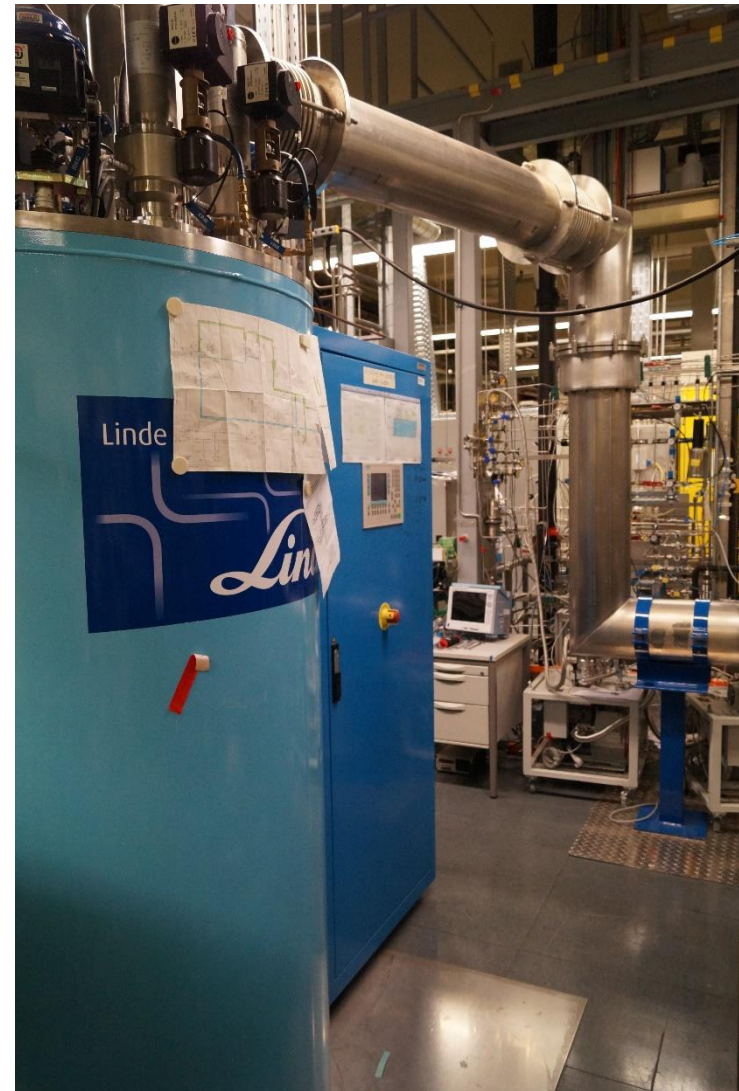
Measurement cell



- absorption length: 5 cm
- temperature: 18 K to 35 K
- pressure: 0.05 MPa to 0.28 MPa



TApIR experiment



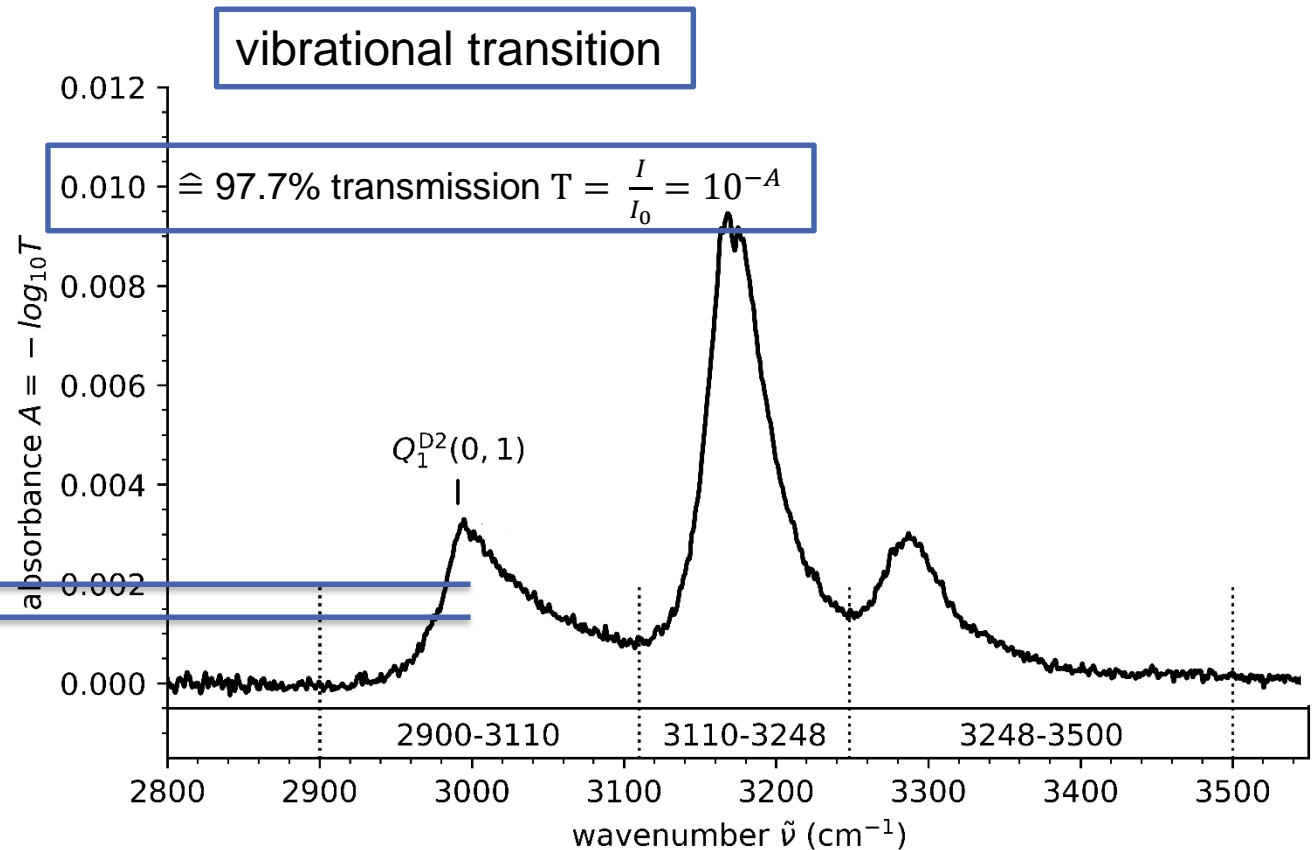
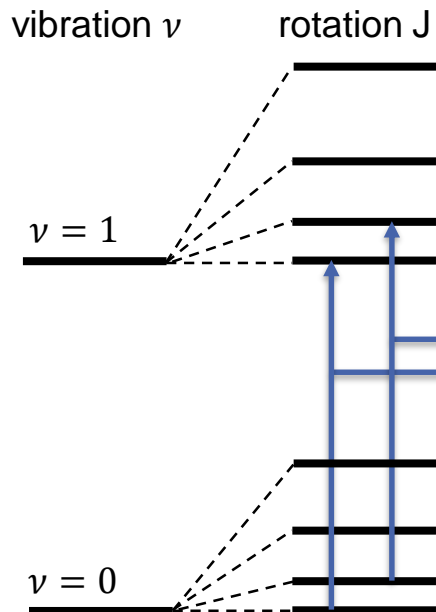
Structure of the IR spectra

Notation: $\Delta J_{\Delta \nu}(J_i)$

$\Delta J = 0 \rightarrow Q$

$\Delta J = 1 \rightarrow R$

$\Delta J = 2 \rightarrow S$



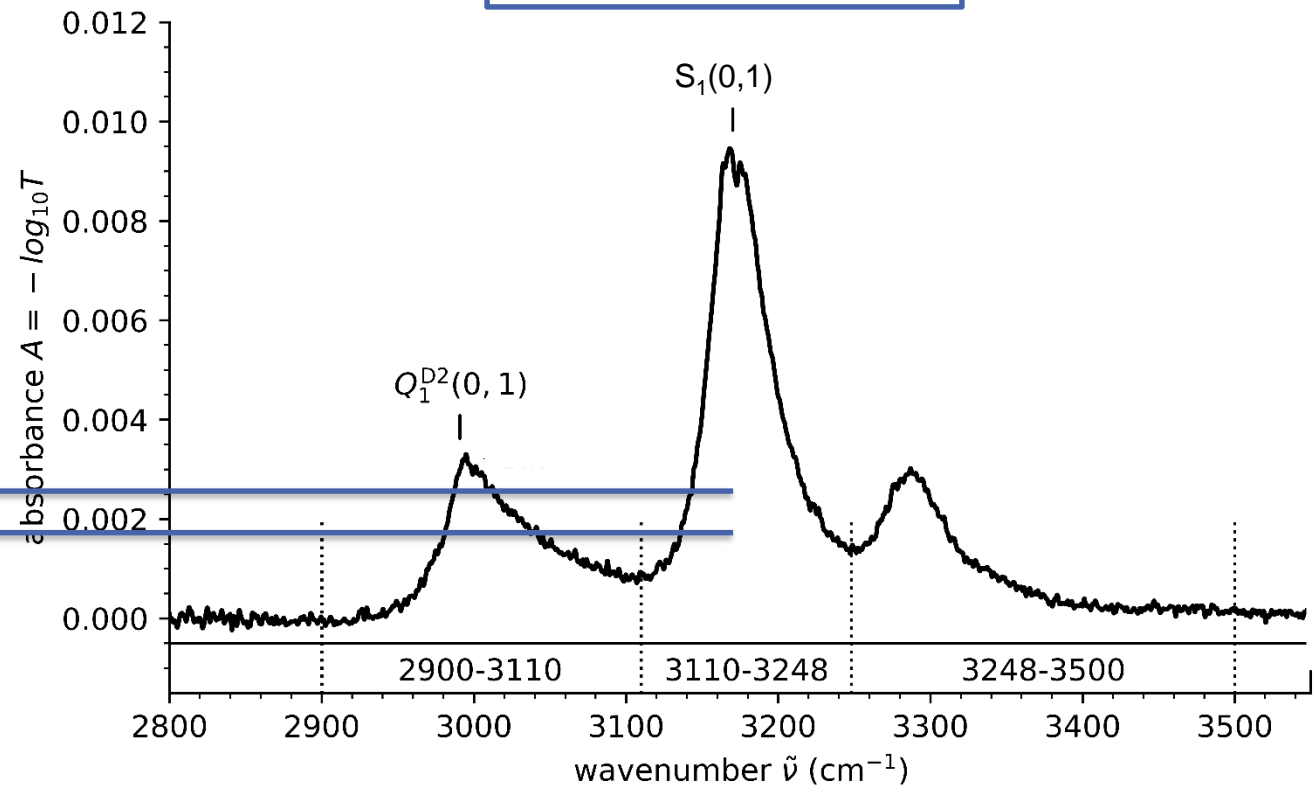
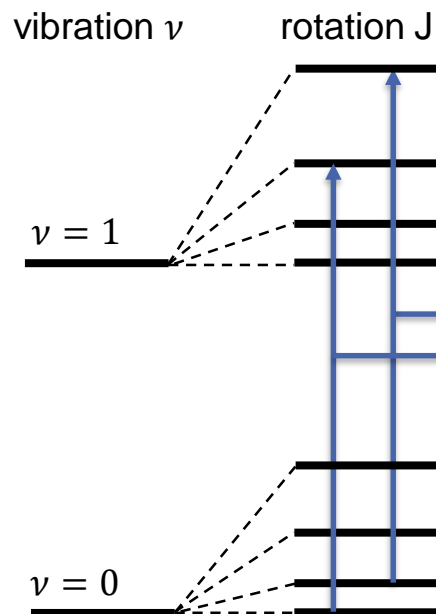
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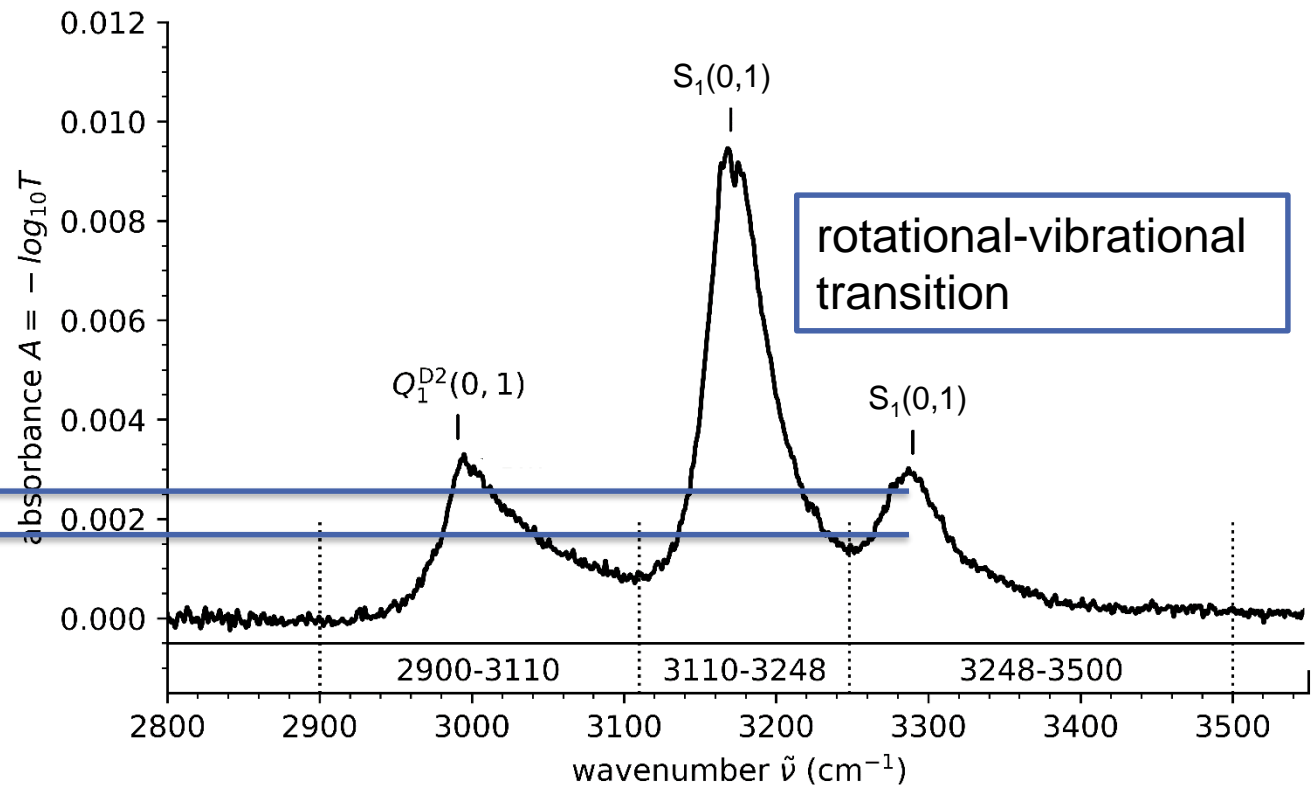
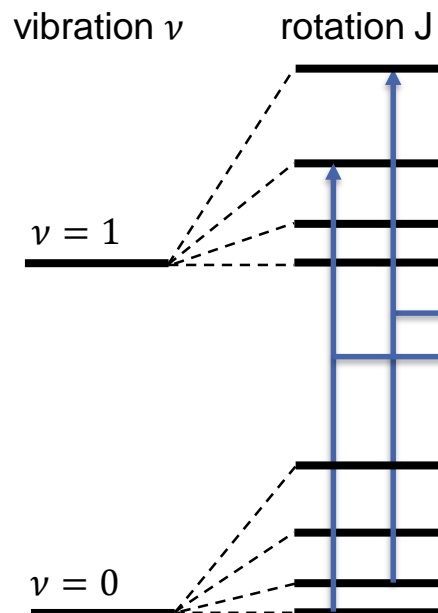
Structure of the IR spectra

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$\Delta J = 2 \rightarrow S$



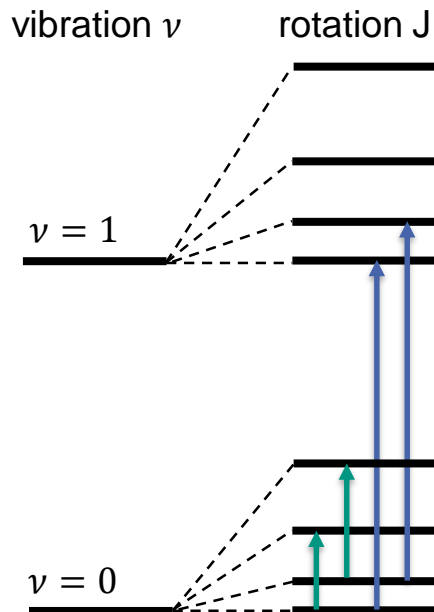
Structure of the IR spectra

Notation: $\Delta J_{\Delta \nu}(J_i)$

$\Delta J = 0 \rightarrow Q$

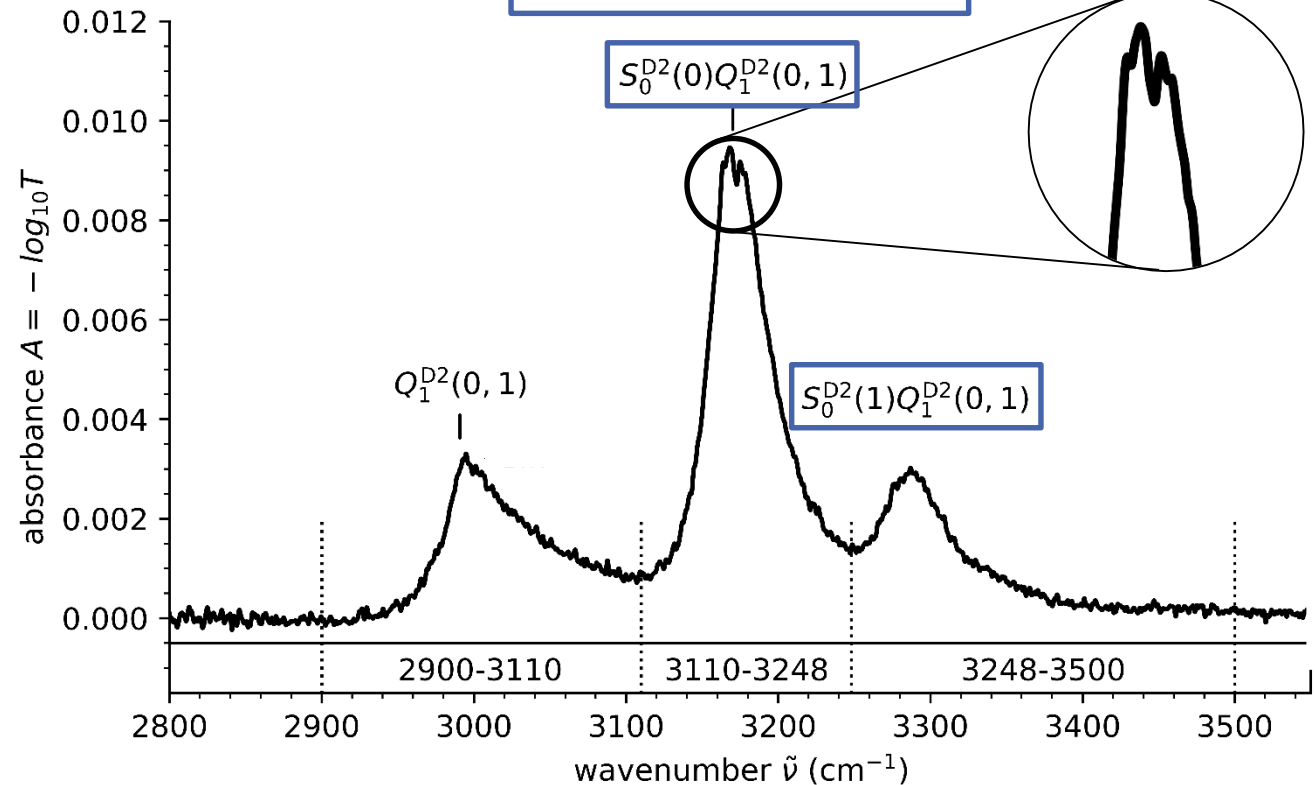
$\Delta J = 1 \rightarrow R$

$\Delta J = 2 \rightarrow S$



splitting:

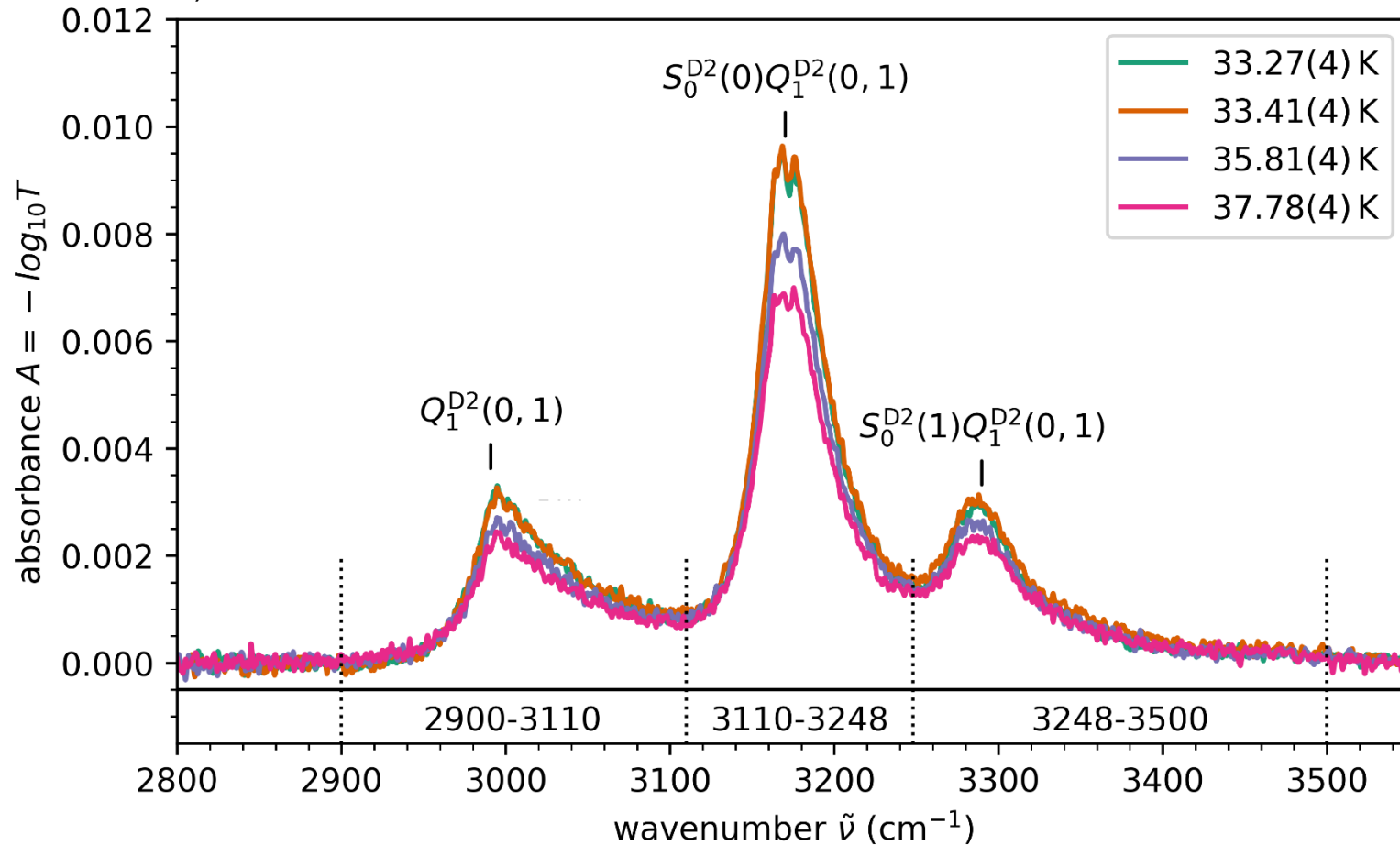
- double transition
- **dimer rotation**



broad background by collisions + sharp dimer excitations

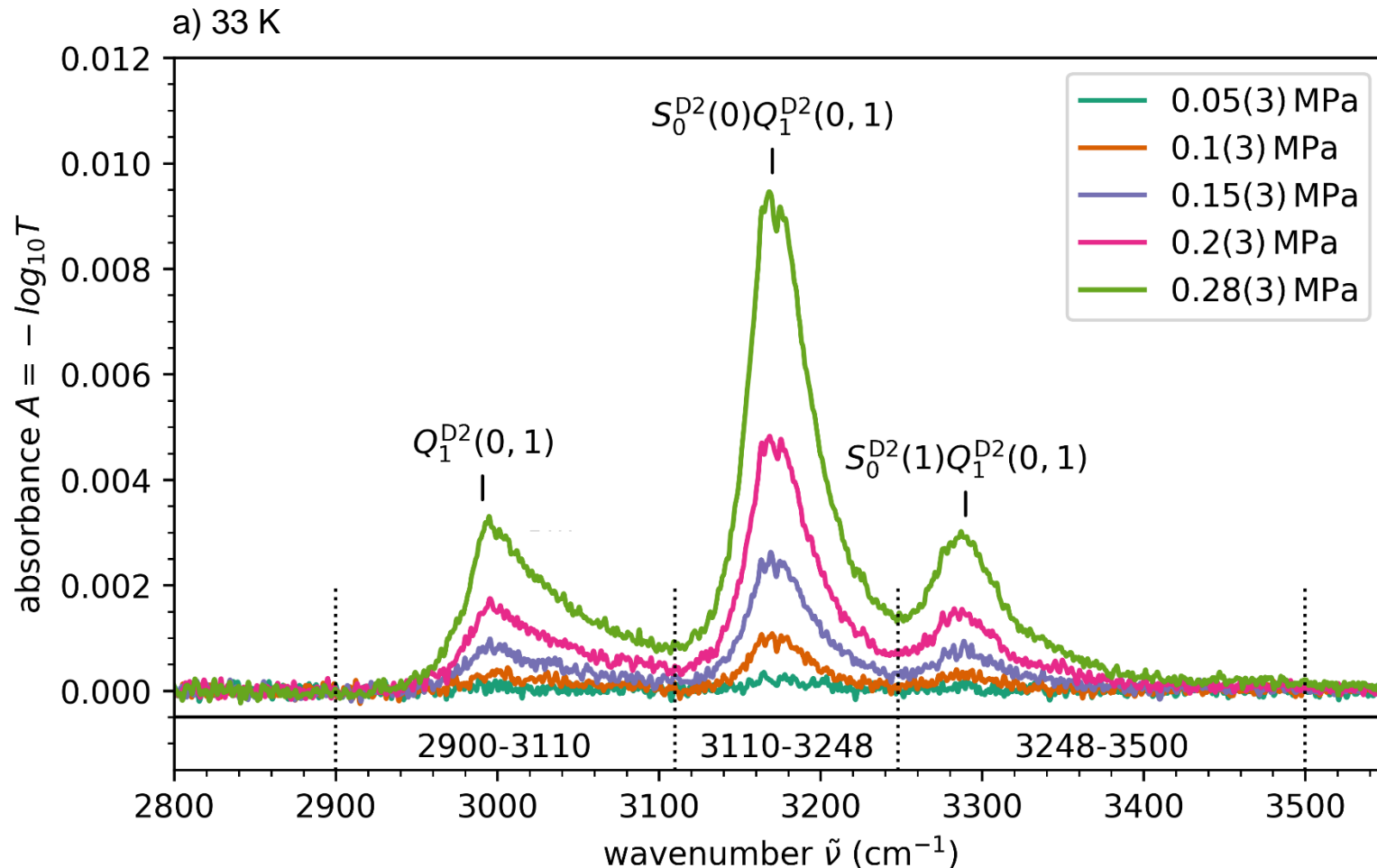
Line selection: Gaseous D₂

a) 0.28 MPa



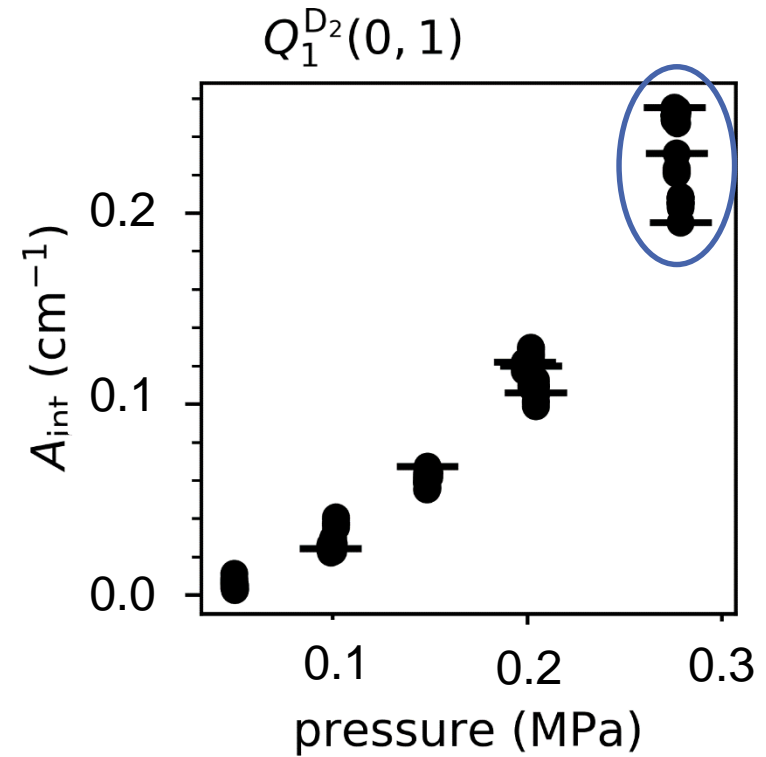
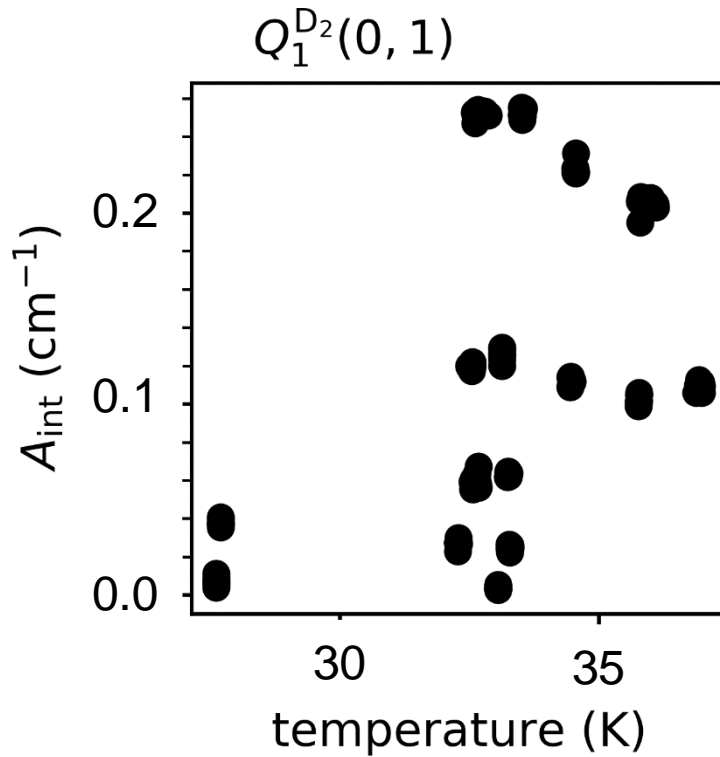
higher temperature → less absorbance

Line selection: Gaseous D₂



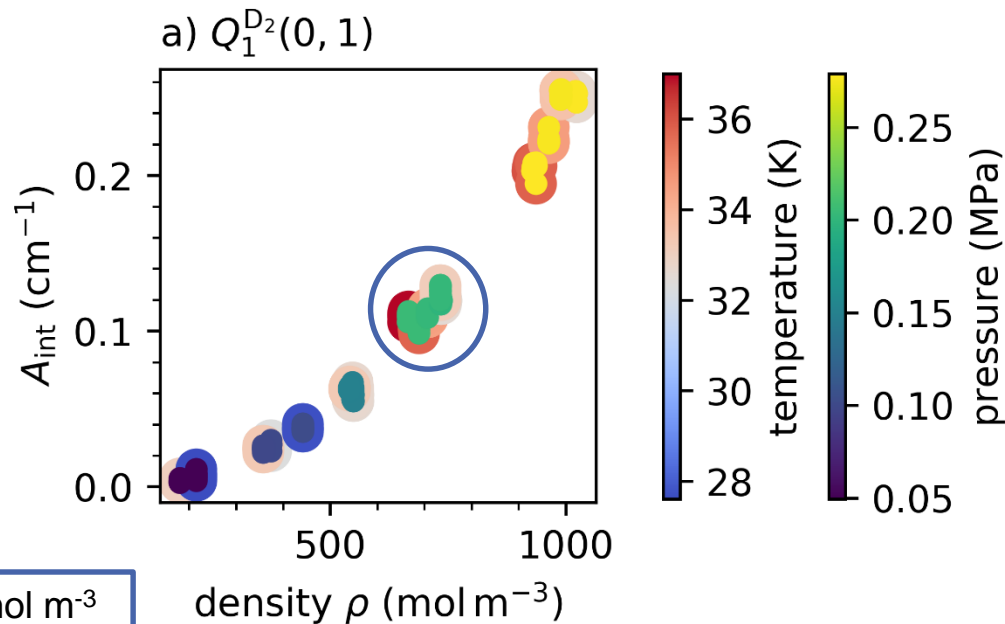
higher pressure → more absorbance

Pressure and temperature dependency



no clear correlations visible

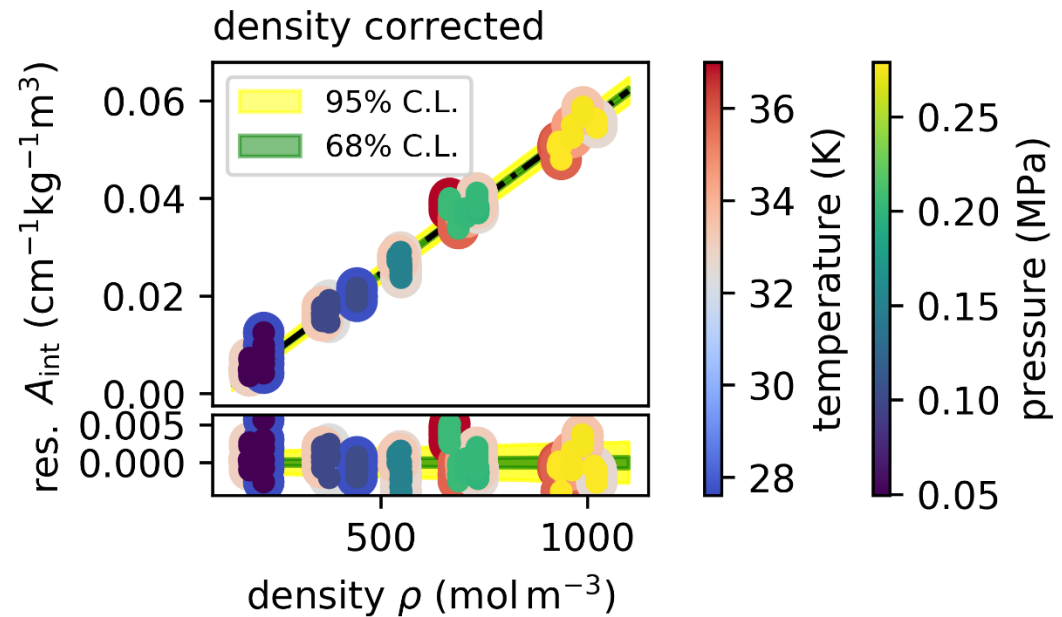
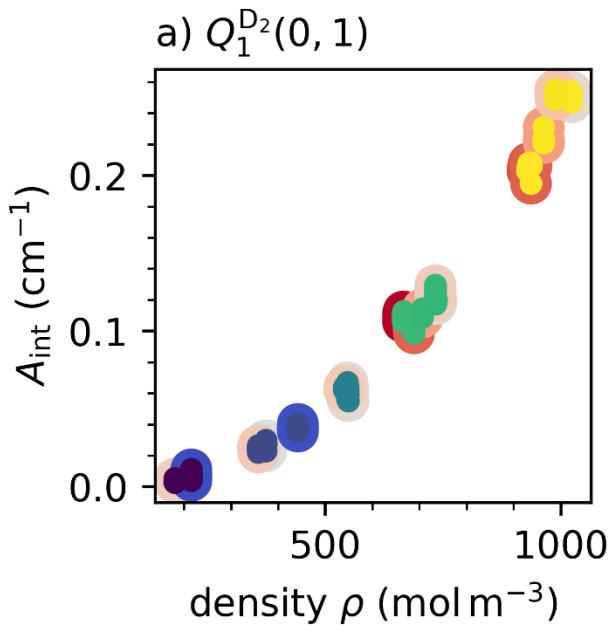
Solution: Density dependency



temperature and absorbance not clearly correlated

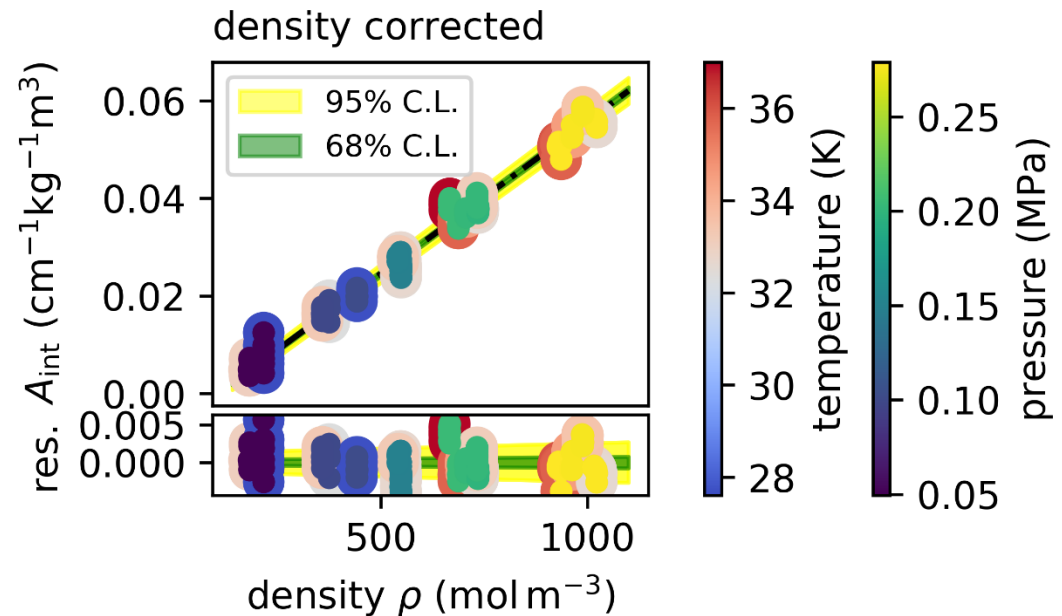
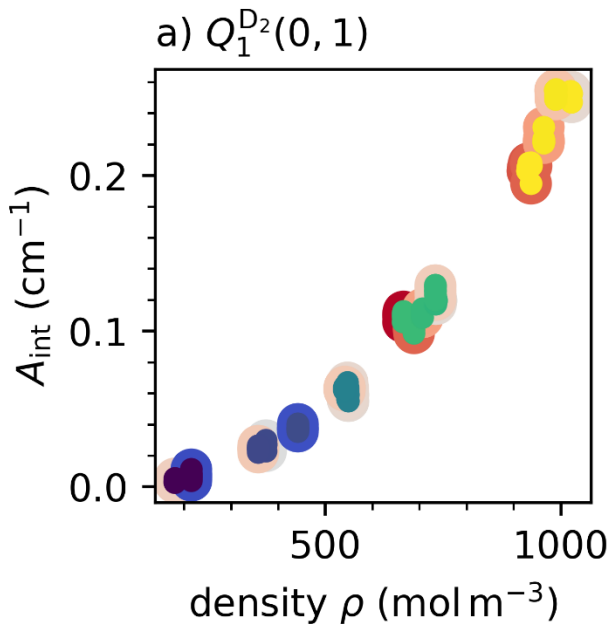
temperature dependency is actually a density dependency

Density dependency



quadratic density dependency: $A_{\text{int}} \propto \rho^2$

Density dependency



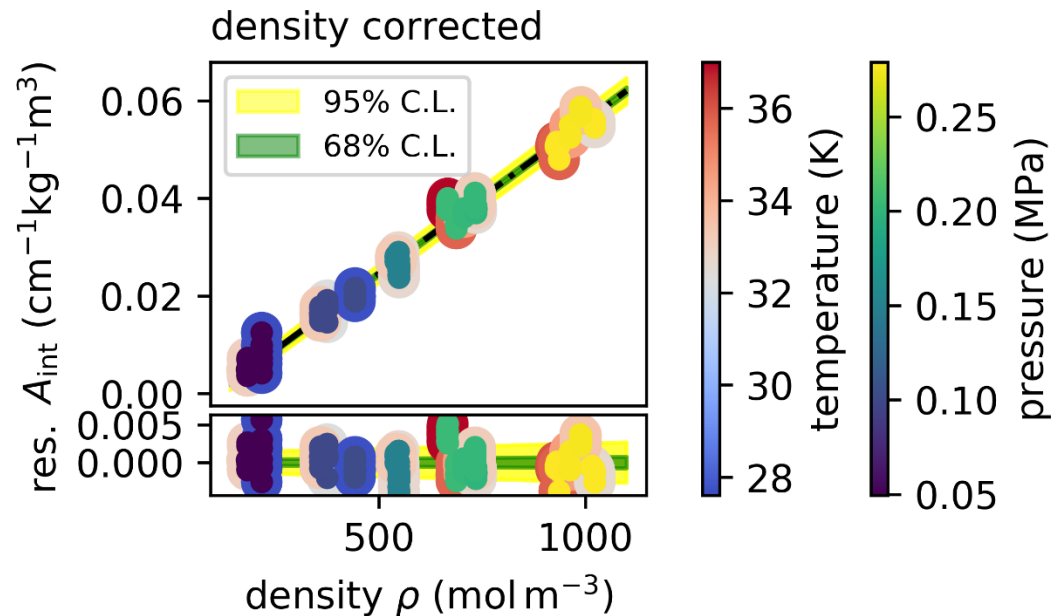
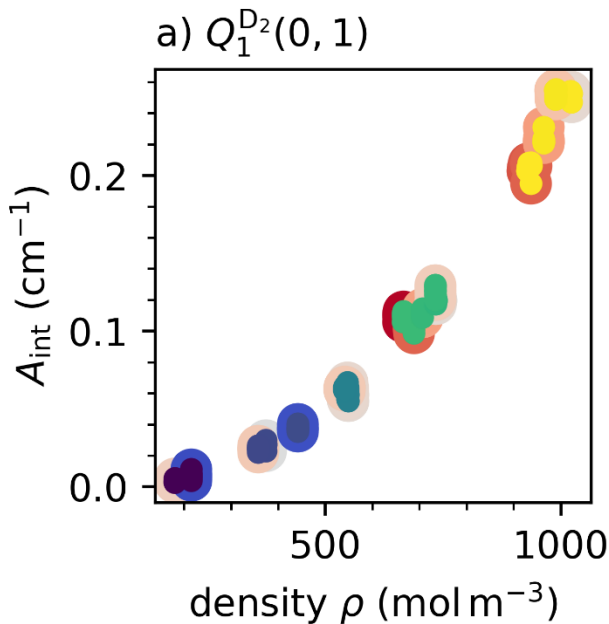
integrated absorbance: $A \propto A_1\rho + A_2\rho^2 + A_3\rho^3$

↑
intrinsic

↑
2 molecule interaction

↑
3 molecule interaction

Density dependency



integrated absorbance: $A \propto A_1\rho + A_2\rho^2 + A_3\rho^3$

intrinsic

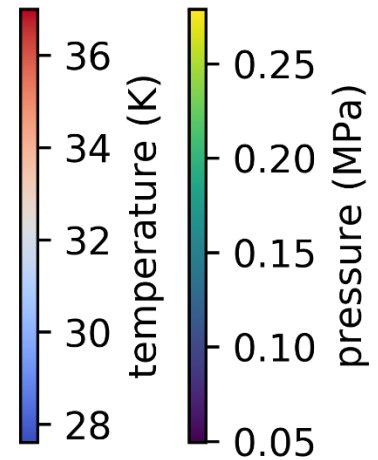
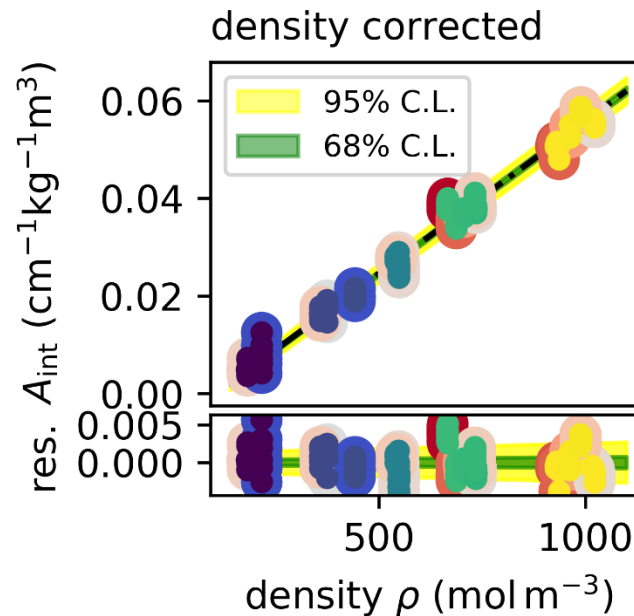
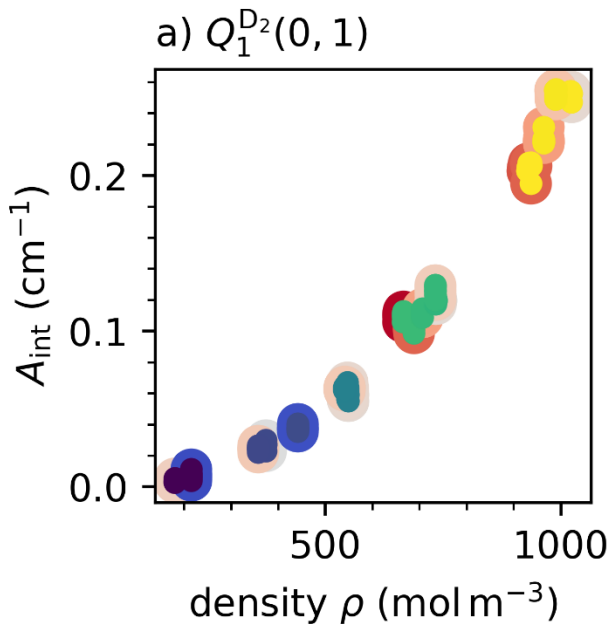
2 molecule interaction

3 molecule interaction

- free-free-scattering at the Van-der-Waals potential
- dimer formation $\sim \rho^2$

quadratic density dependency

Density dependency : Dimers?



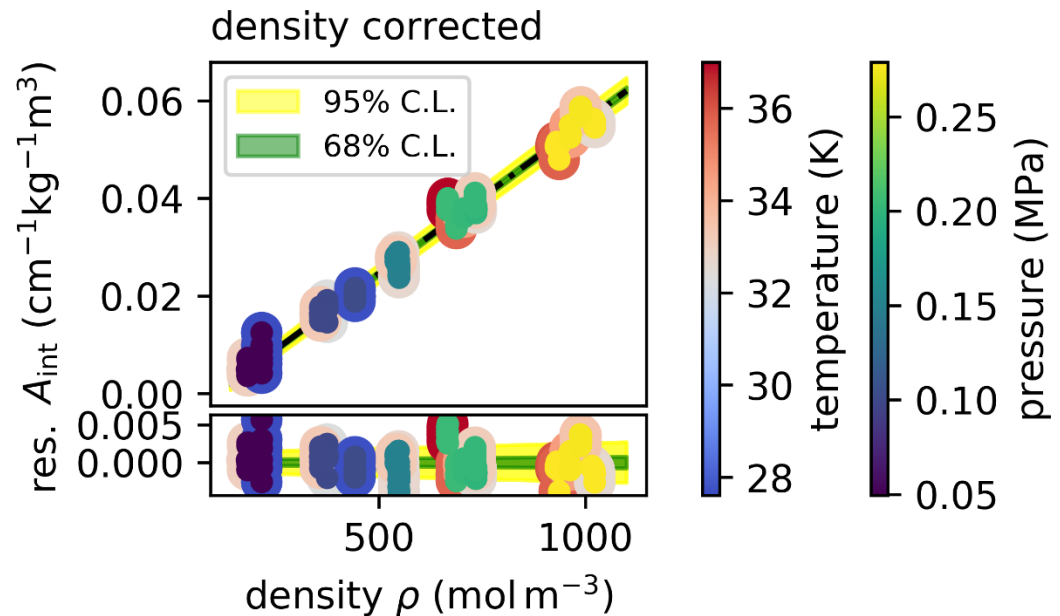
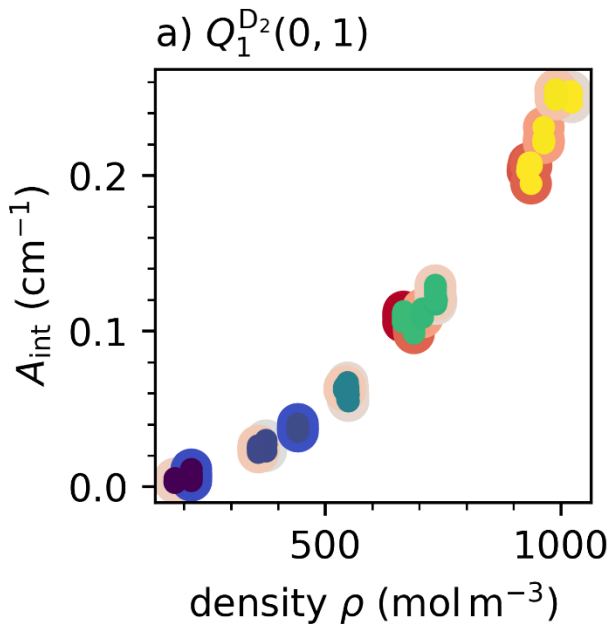
integrated absorbance: $A \propto A_1\rho + A_2\rho^2 + A_3\rho^3$

dimer intrinsic

2 dimer interaction

3 dimer interaction

Density dependency : Dimers?



integrated absorbance: $A \propto A_1\rho + A_2\rho^2 + A_3\rho^3$

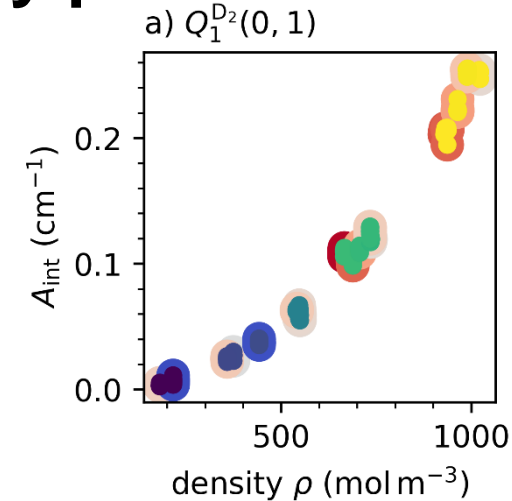
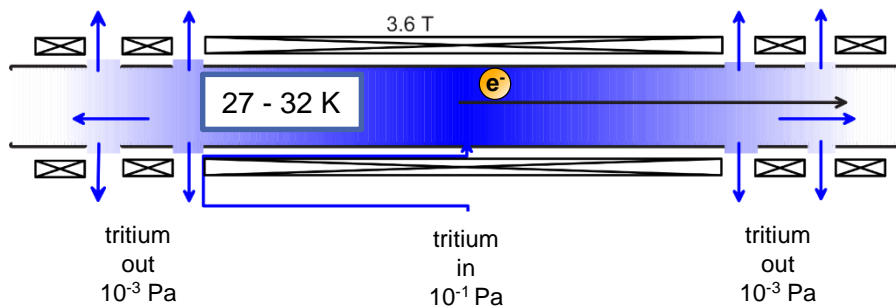
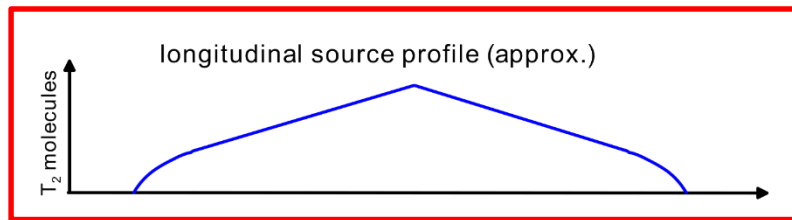
dimer intrinsic

2 dimer interaction

3 dimer interaction

dimer-dimer-interaction
→ larger clusters?

Impact on KATRIN: WGTS density profile



absorption \propto density²

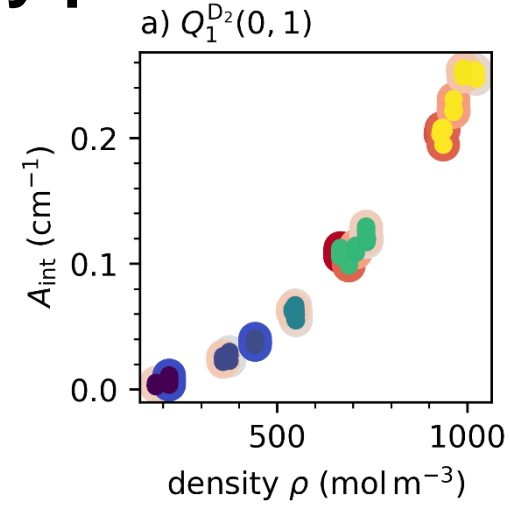
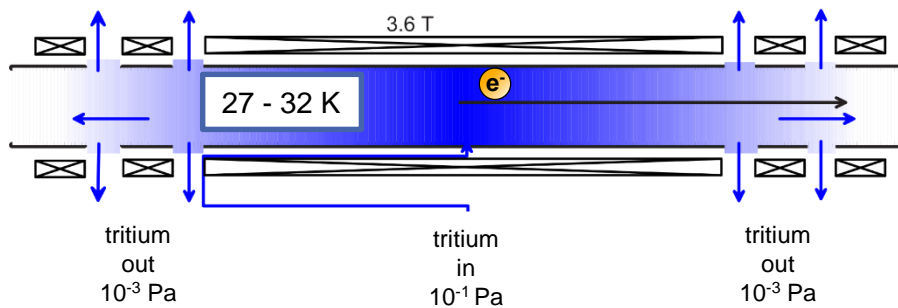
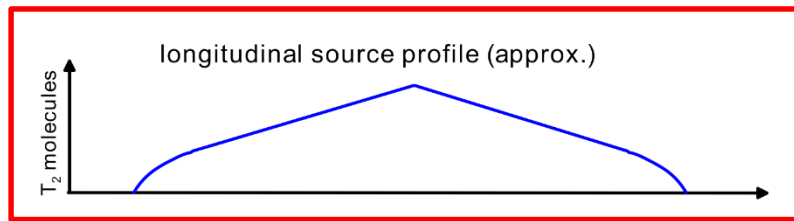
Extrapolation:

Assumption: 100% dimers at 0.05 MPa
 \rightarrow 10^{-6} % dimers at the WGTS inlet

Comparison with literature values:

- 10^{-4} % $(H_2)_2$ dimers under WGTS conditions
 (own calculations according to Frommhold 1993)
- 0.1 % $(H_2)_2$ dimers at 20 K, no pressure given (Watanabe et al. 1964)
- a few % $(H_2)_2$ dimers at 20 K, no pressure given (Watanabe et al. 1965)

Impact on KATRIN: WGTS density profile

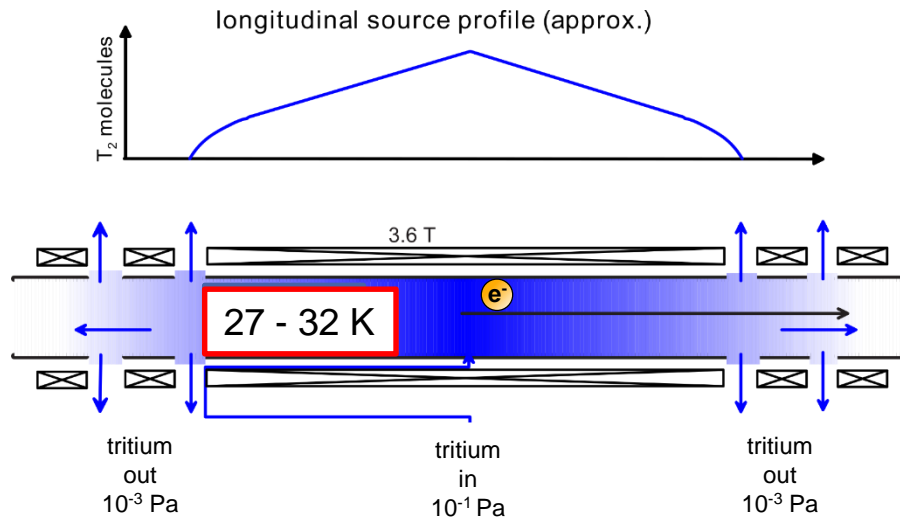


absorption \propto density²

Possible variation of the Van-der-Waals cluster density along the WGTS density profile

Future measurements with lower densities necessary to clarify this influence
 → T₂ApIR experiment: Commissioning + first data in 2020

Impact on KATRIN: WGTS temperature



~~“To use much lower temperatures than 30K is not possible, because tritium clusters may be created out of the T_2 molecules.”~~

KATRIN Design Report 2004

An increase of the WGTS temperature cannot significantly reduce the cluster concentration
 → tritium data needed to quantify or neglect the influence
 → T_2 ApIR experiment



THANK YOU FOR YOUR ATTENTION!