

# Systematic effects in Laser Raman measurements for KATRIN



Forschungszentrum Karlsruhe  
in der Helmholtz-Gemeinschaft



Universität Karlsruhe (TH)  
Forschungsuniversität • gegründet 1825



# LARA

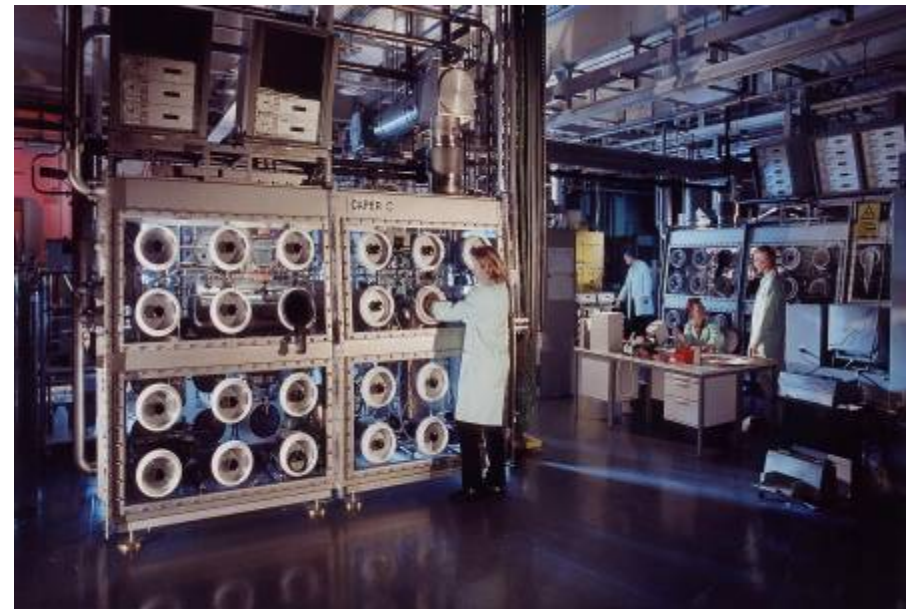
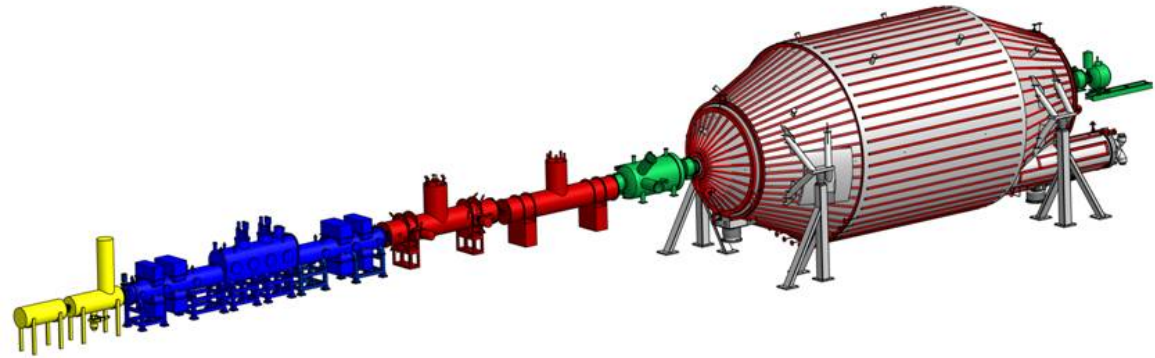


Teilprojekt A1

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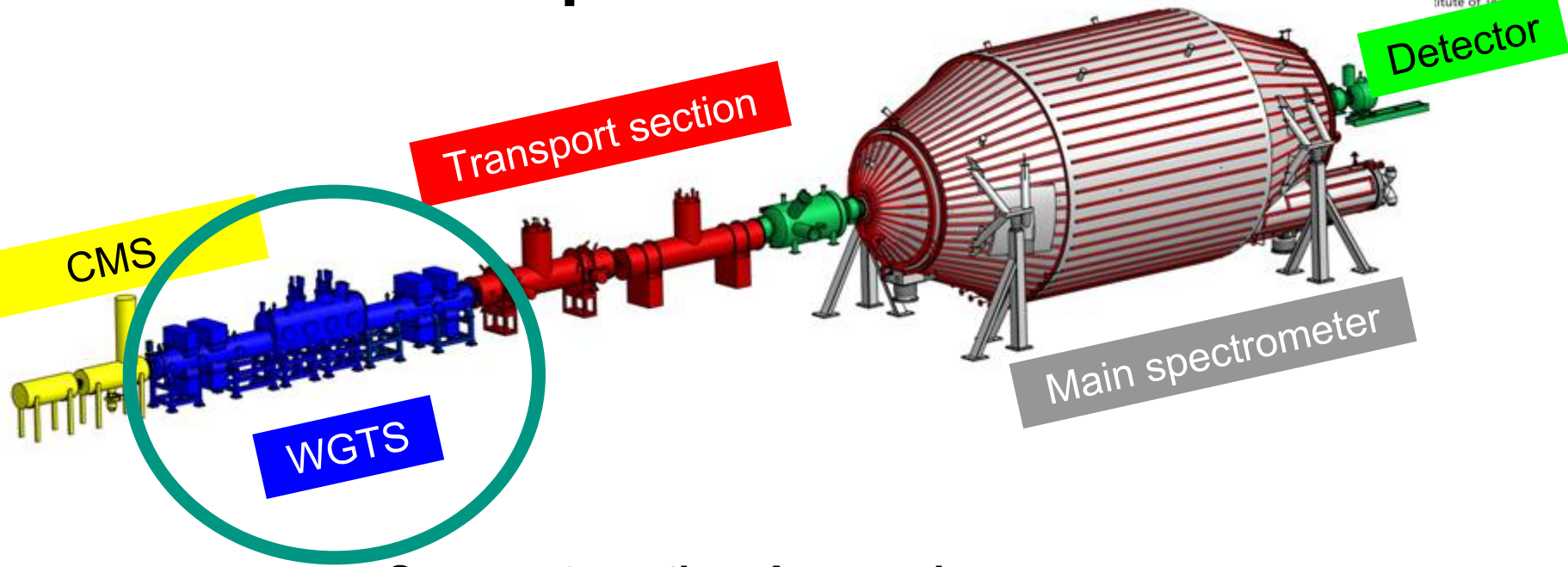
# Content

- LARA for KATRIN
- Fundamentals
- Setup of Laser-Raman system
- Systematic effect
  - Detection limit
  - Background features
  - Long term stability
- Quantitative analysis
- Summary / Outlook



Tritium Laboratory Karlsruhe

# The KATRIN-Experiment



$$\text{Source strength} = A_Q \cdot \rho d \cdot \epsilon_T$$

- column density
- tritium injection rate
- beam tube temperature
- tritium purity

$$\rho d = 5 \cdot 10^{17} \text{ cm}^{-2}$$

$$q_{in} = 1.8 \text{ mbar l s}^{-1}$$

$$T = 30 \text{ K}$$

$$\epsilon_T \geq 95\%$$

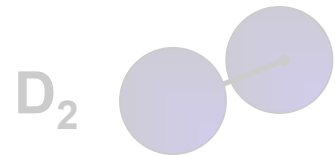
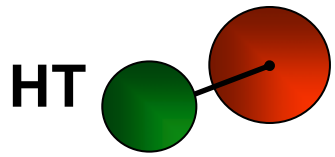
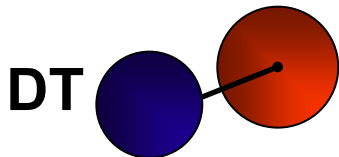
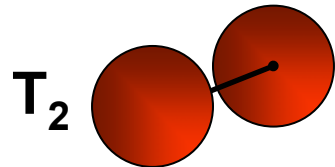
stability in the range of  $10^{-3}$

monitoring

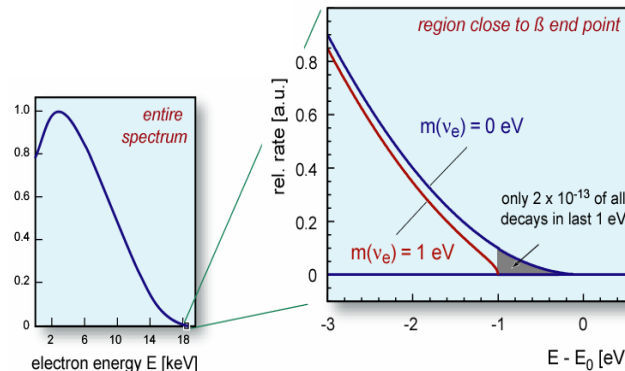
Laser-Raman Spectroscopy

# Isotopic purity ( $\epsilon_T$ )

Isotopologues  
of hydrogen



Direct contribution to  
count rate at endpoint



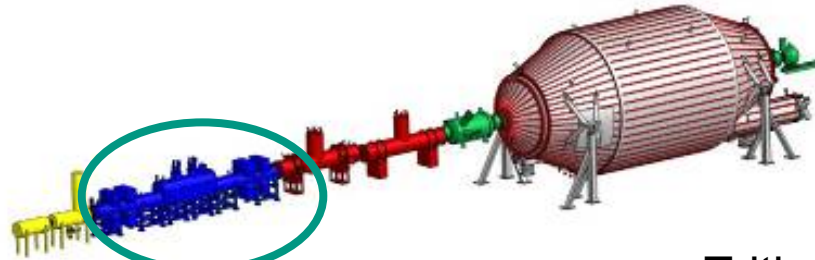
Different final states

## Requirements for LARA

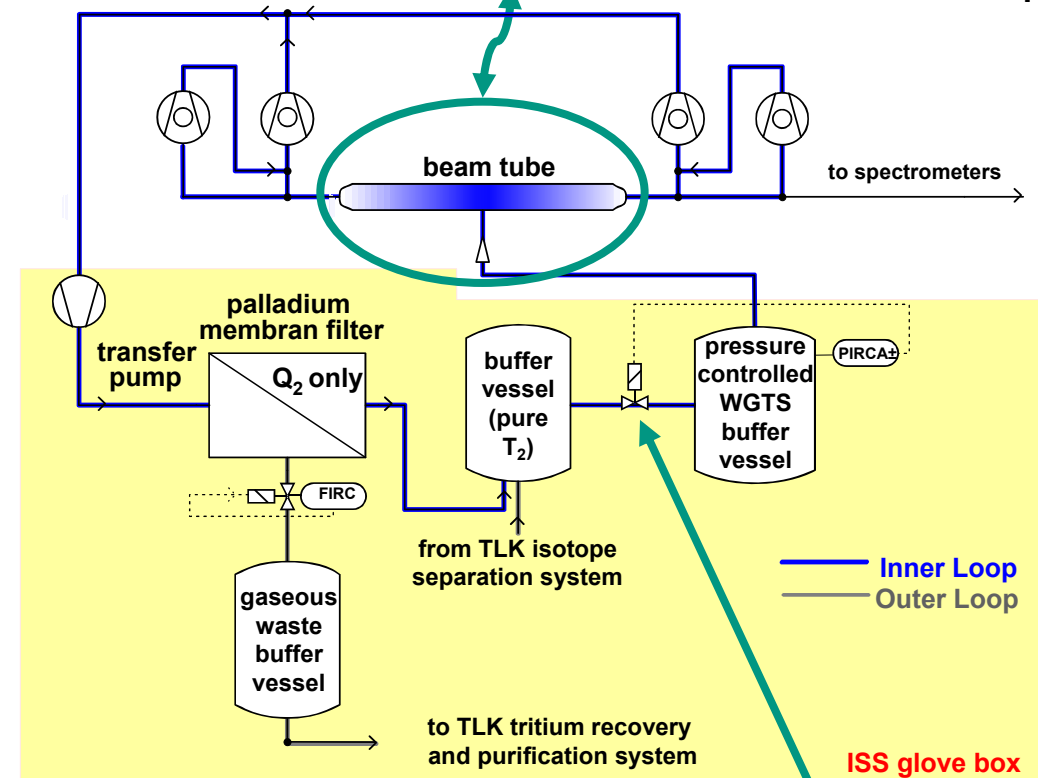
- Measurement of tritium purity before injection into WGTS with a precision of **0,1 %**
- Determination of isotopologue composition
- Acquisition time as short as possible (minutes)
- Entire T<sub>2</sub> pumped through LARA-cell (no Bypass)

# LARA for KATRIN

## Flow diagram of Inner Loops and Source



Tritium throughput  
**40 g/d =**  
 **$1,5 \cdot 10^{16}$  Bq/d**

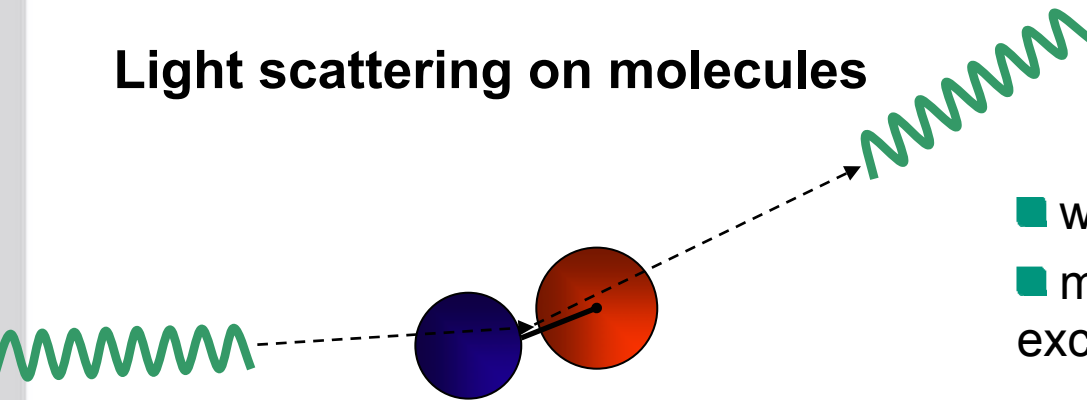


**LARA-Measurements @  $p \approx 100$  mbar**

Q = H, D, T

# Fundamentals

## Light scattering on molecules

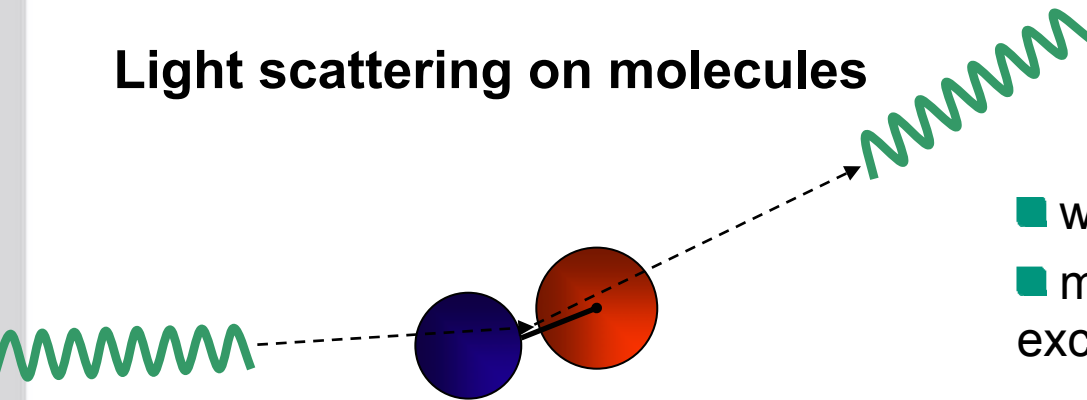


## elastic collisions Rayleigh-scattering

- wavelength of photons invariant
- molecule remains in state of excitement

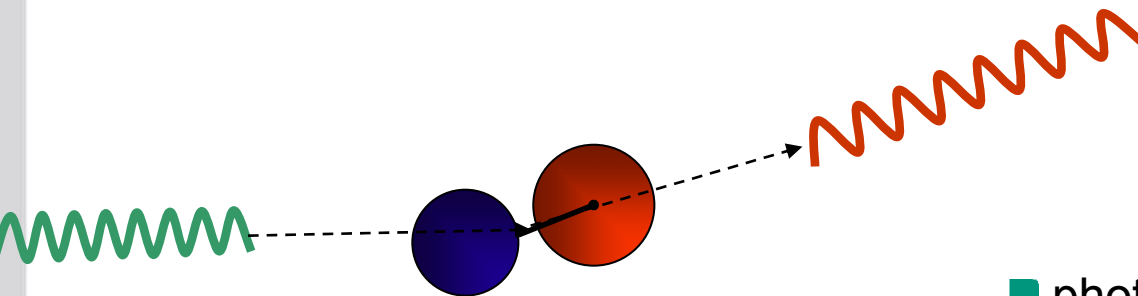
# Fundamentals

## Light scattering on molecules



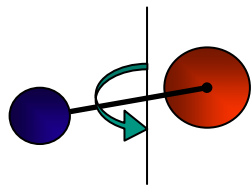
### elastic collisions Rayleigh-scattering

- wavelength of photons invariant
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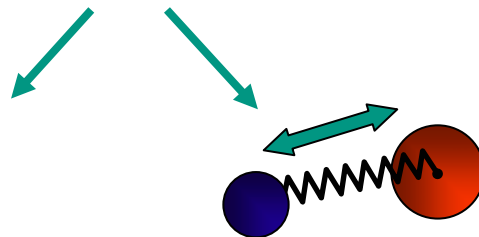


### inelastic collisions Raman-scattering

- photon loses a fraction of its energy  
→ wavelength increases
- energy is transferred to molecule
- **vice versa process also possible!**

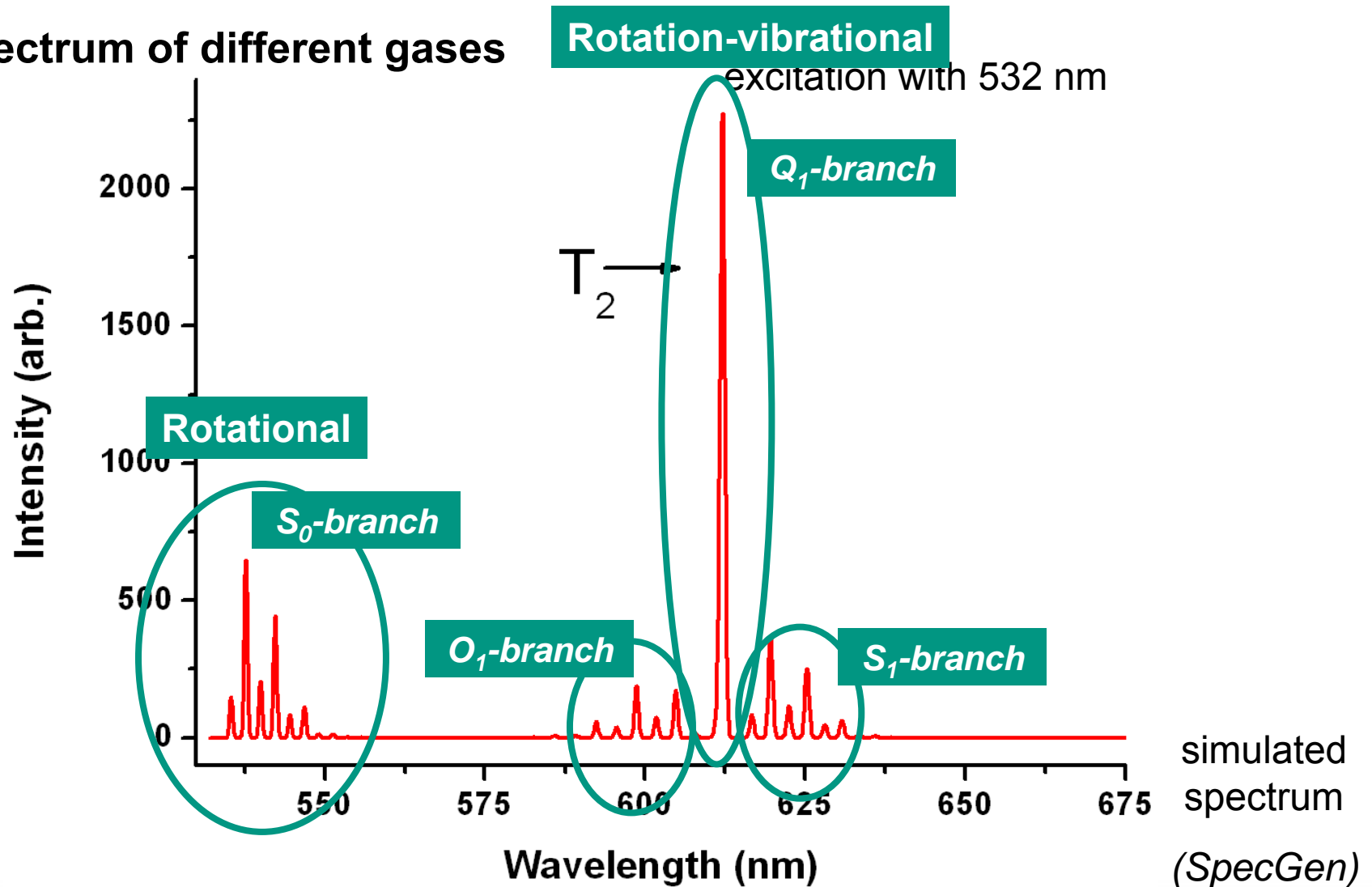


Rotation



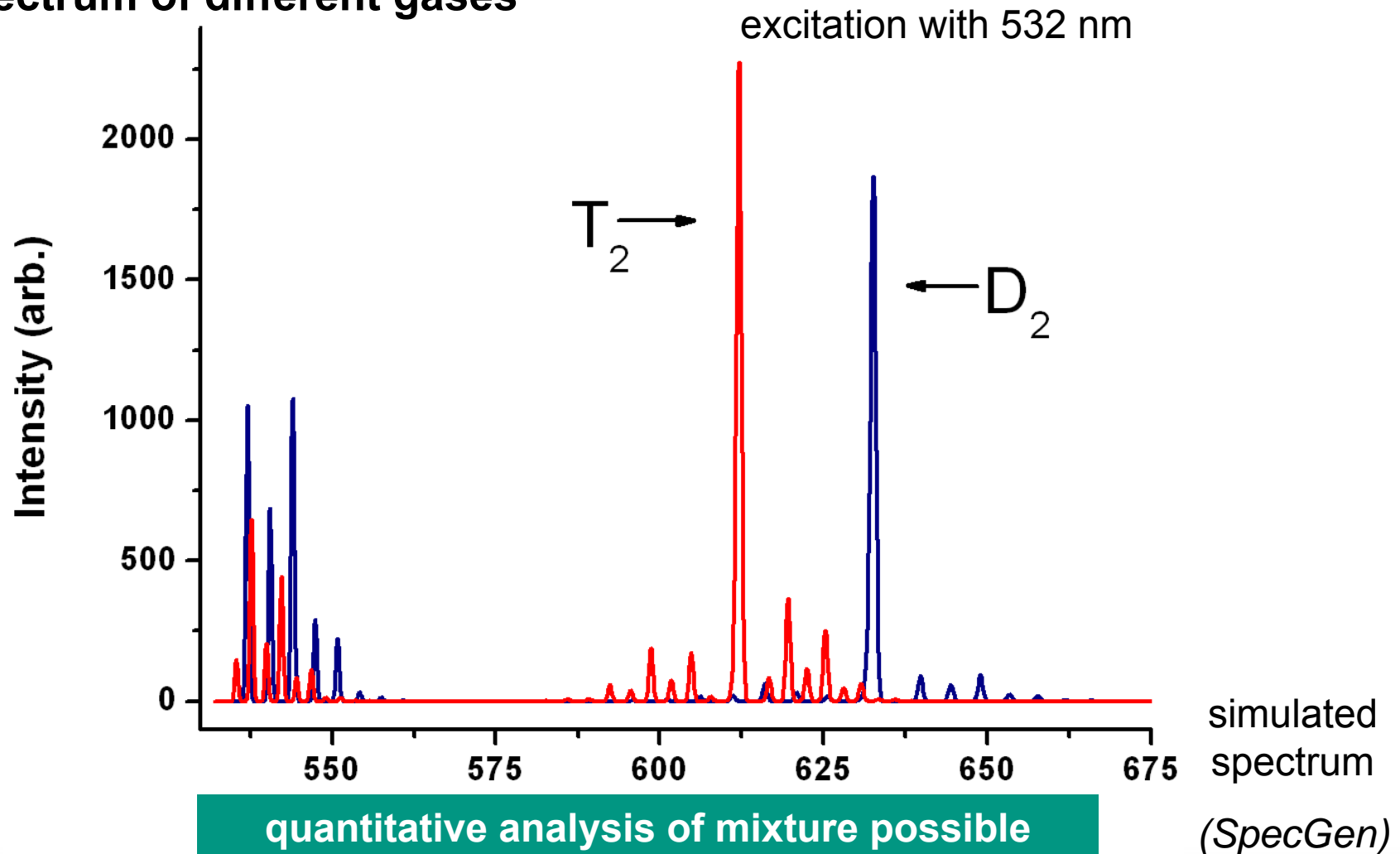
Vibration

## Spectrum of different gases

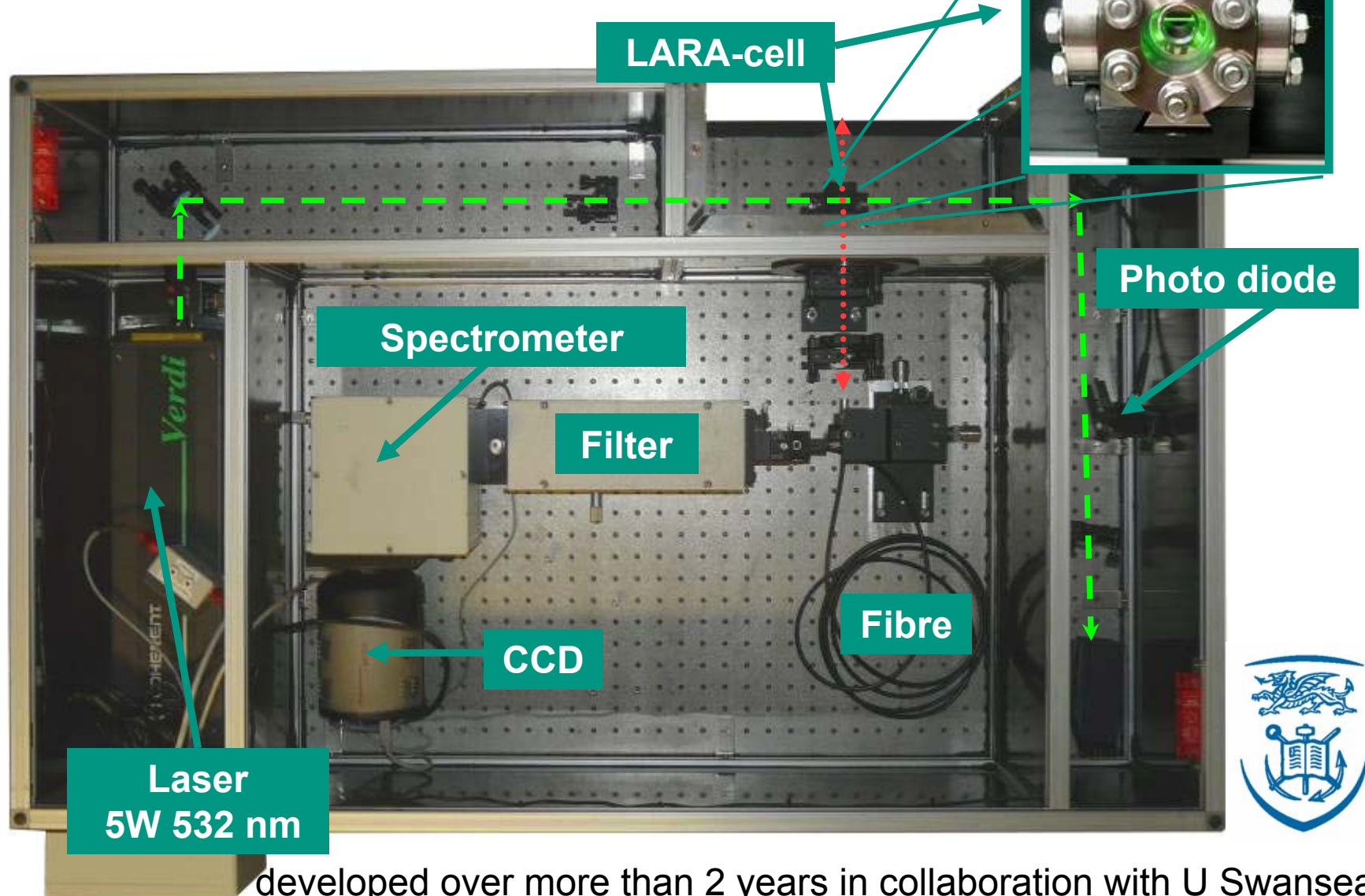




## Spectrum of different gases



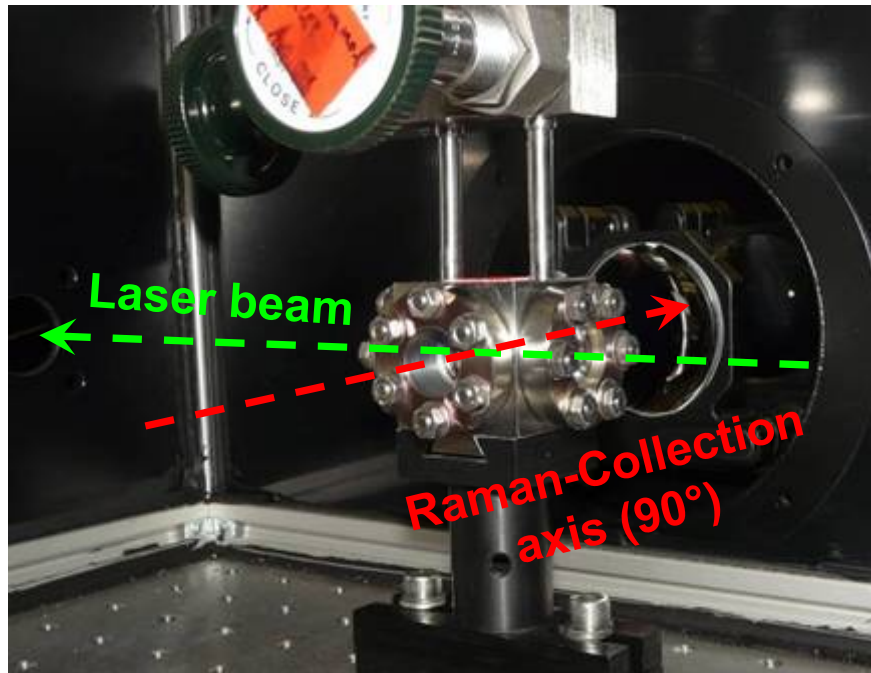
# Setup of LARA-system



developed over more than 2 years in collaboration with U Swansea

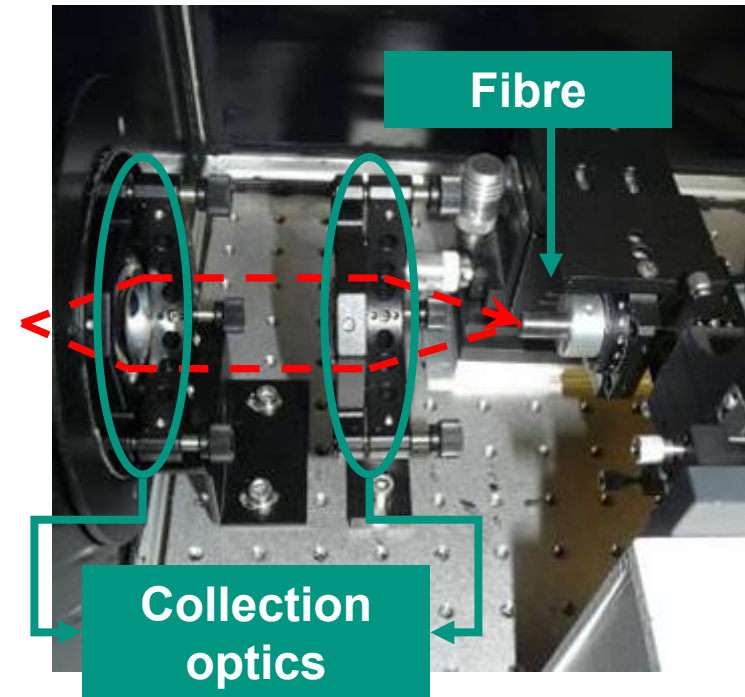
# Setup of LARA-system

## 1:1 image of scattered light on a fibre bundle



### Laser-Raman-Cell

- Volume: 7,1 cm<sup>3</sup>
- Operation: „static“ or „in flow“



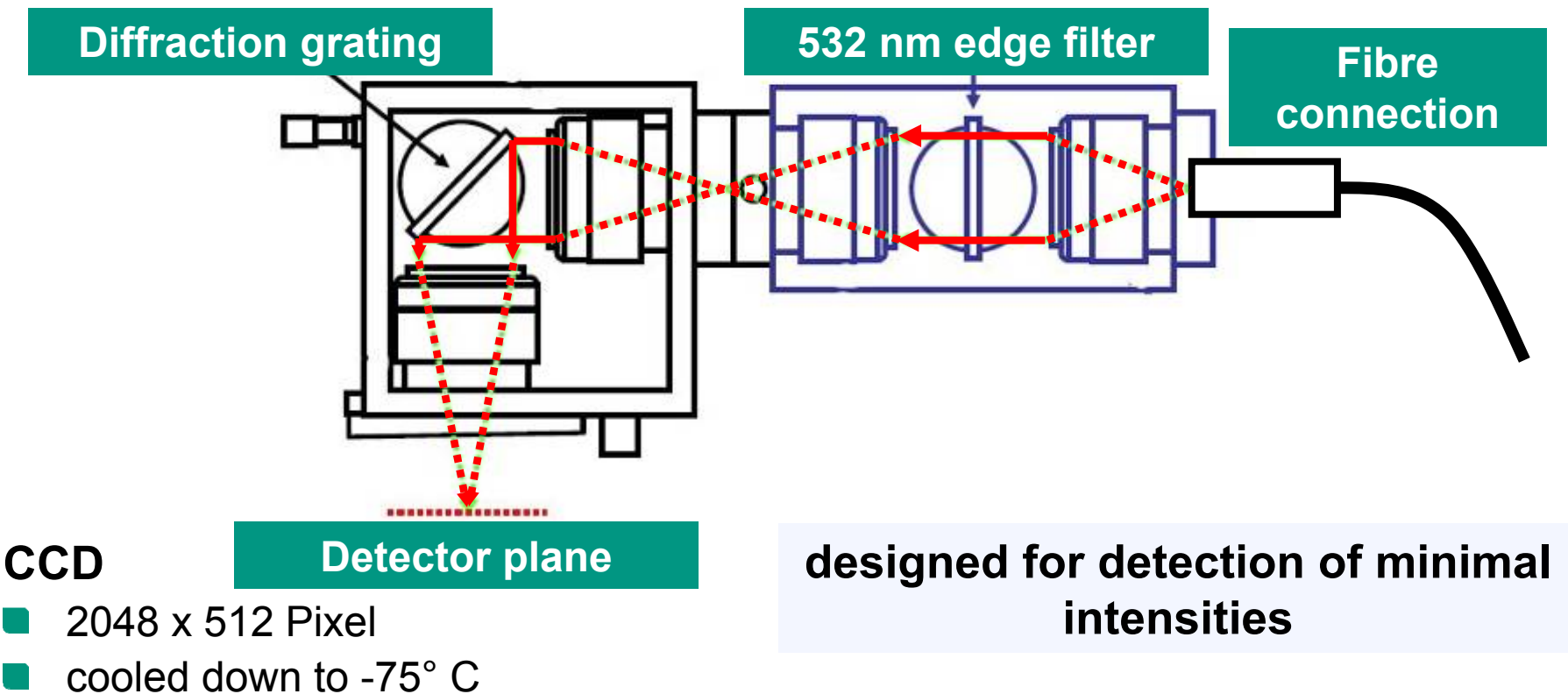
### Fibre

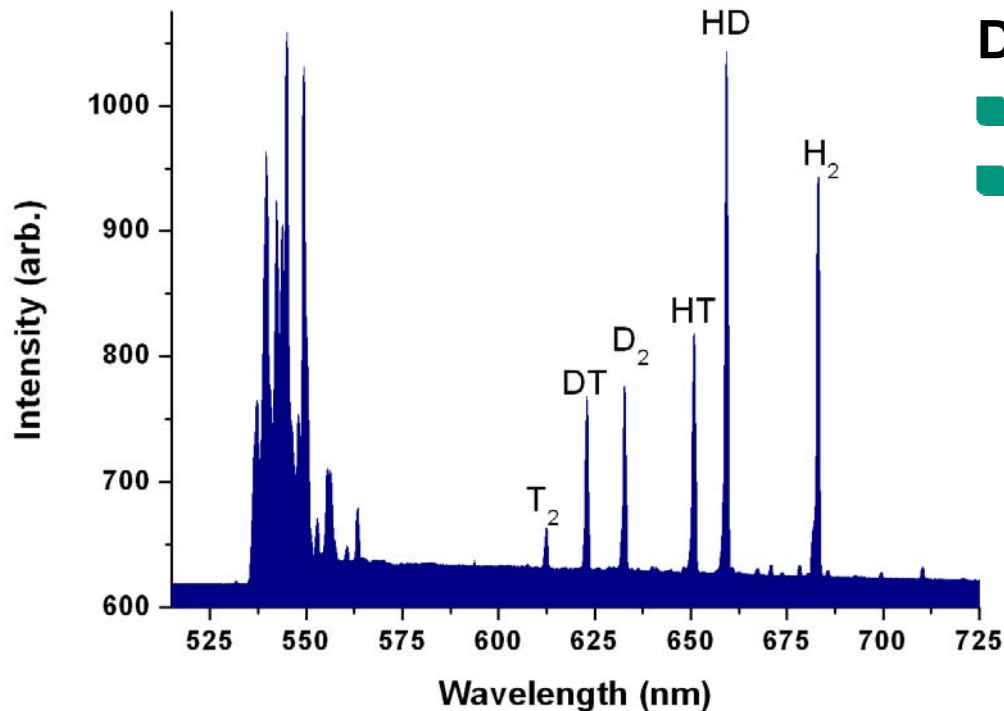
- 48 single fibres adjoining
- simple adjustment
- robust construction

# Setup of LARA-system

## Transmission Spectrometer

- high light throughput  $\rightarrow$  high intensity
- moderate resolution  $600\text{mm}^{-1} \rightarrow$  covers region of interest





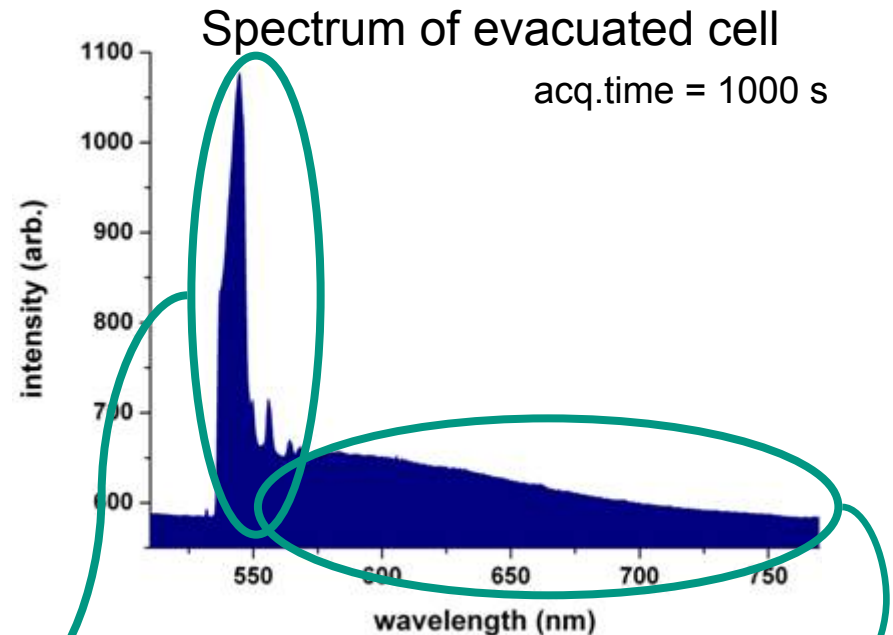
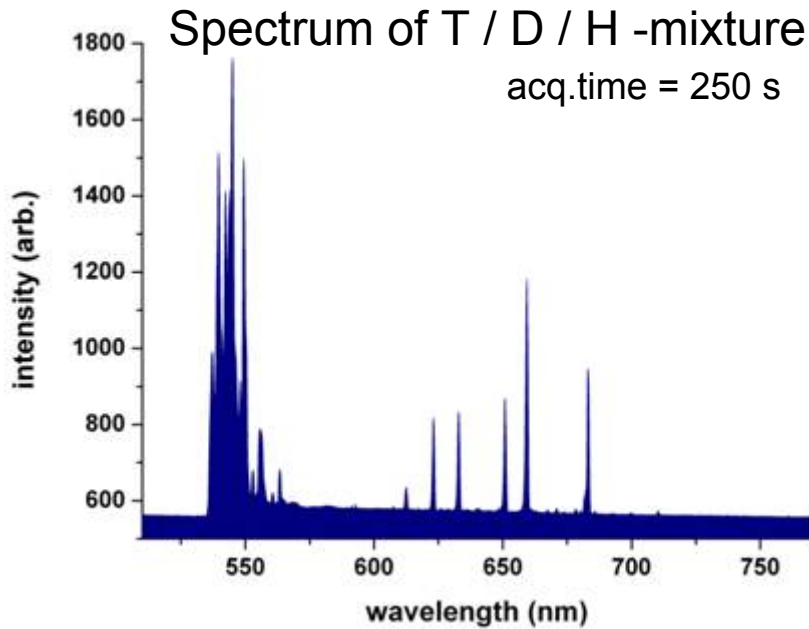
## Detection limit

- Peak height  $\approx 2 \cdot$  noise-level
- This sample:
  - Total pressure:  
 $p_{\text{tot}} \approx 110$  mbar
  - noise-level = 0.32
  - Sum of all 6 peak intensities:  
 $\Sigma_{\text{tot}} \text{signal} = 1274$
  - minimal detectable partial pressure:

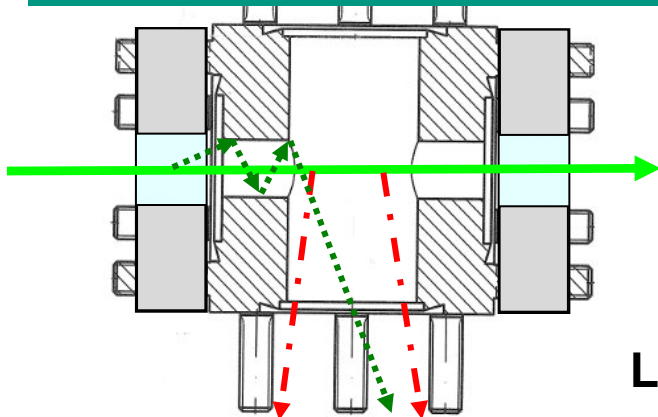
$$p_{\text{min}} = \frac{2 \cdot \text{noise-level}}{\Sigma_{\text{tot}} \text{signal}} \cdot p_{\text{tot}}$$

**Detection limit for hydrogen isotopologues < 0,06 mbar partial pressure (250s, 5W)**

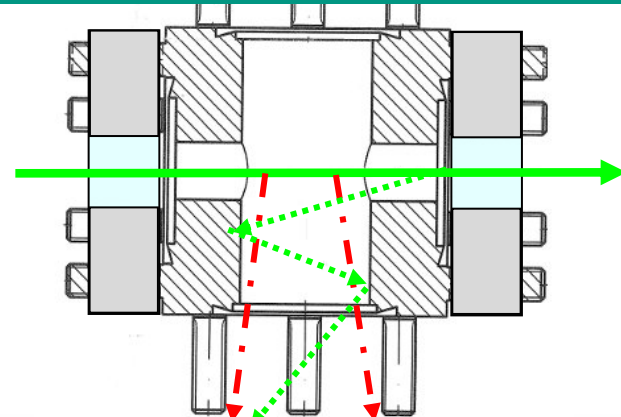
# Background features



Raman-scattering in  $\text{SiO}_2$



Reflections on windows



LARA - Cell

# Precision

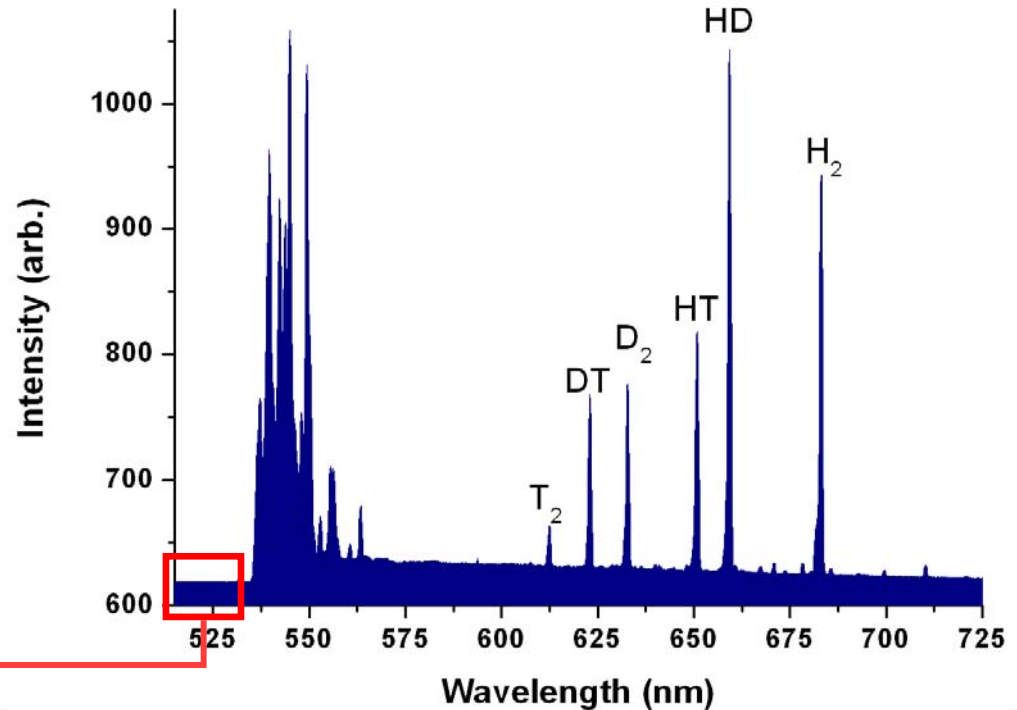
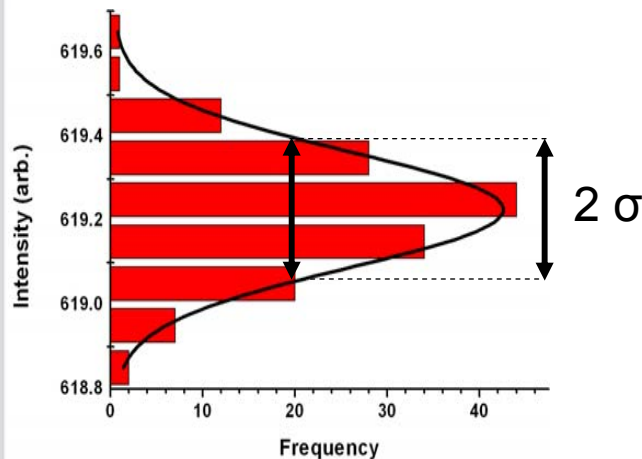
## Noise

Definition: Noise-amplitude  $N = 2 \sigma$

## Precision

$$\Delta I = \frac{\text{Noise}}{\text{Signal}}$$

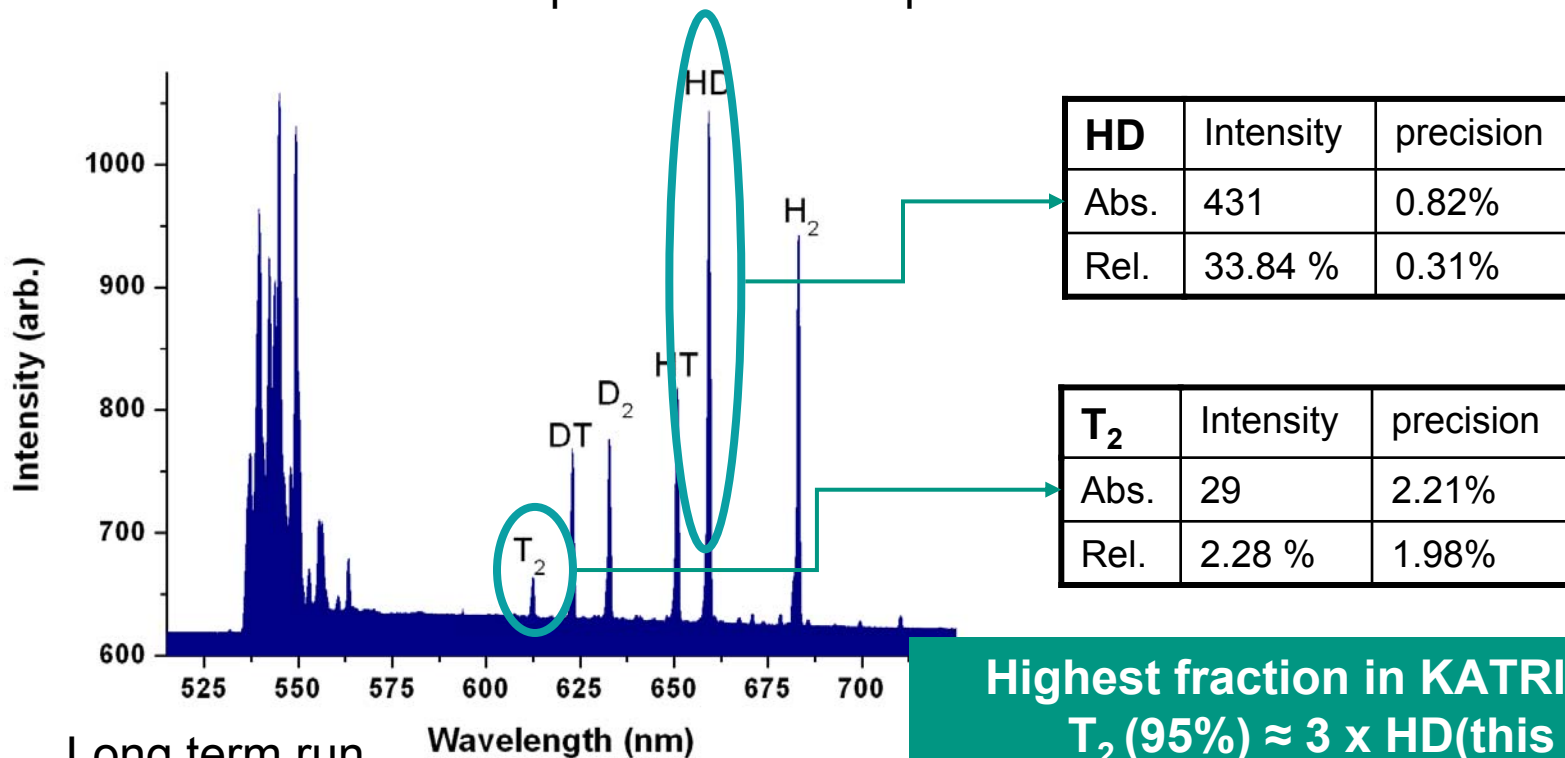
| Isotop.        | Intensity | S/N  | Precision = N/S |
|----------------|-----------|------|-----------------|
| T <sub>2</sub> | 33        | 108  | 0.92 %          |
| DT             | 139       | 462  | 0.22 %          |
| HT             | 190       | 636  | 0.15 %          |
| D <sub>2</sub> | 148       | 494  | 0.20 %          |
| HD             | 417       | 1392 | 0.07 %          |
| H <sub>2</sub> | 318       | 1060 | 0.09 %          |



# Stability of long term runs

## Test of stability

- Repeated measurements of static mixture
- All fluctuations are related to acquisition and analysis
- Calculate  $\sigma$  for each peak from all acquisitions



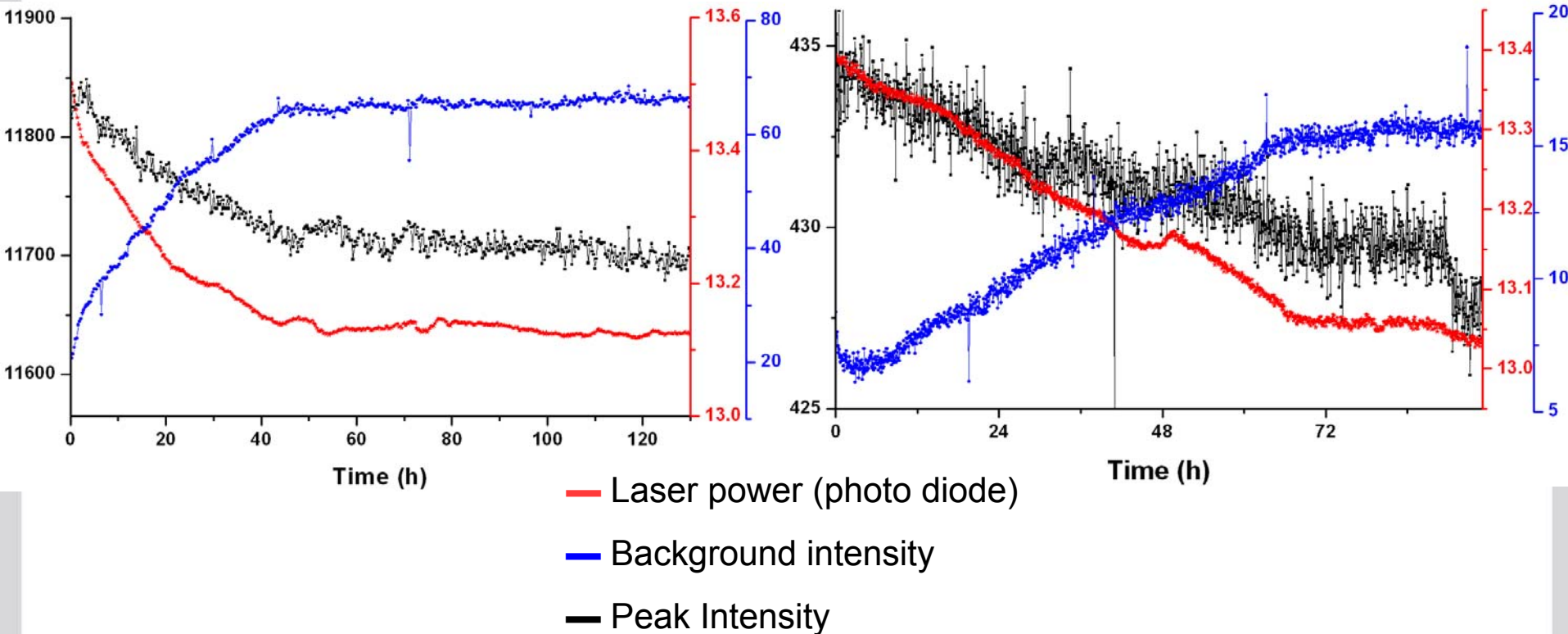
Long term run  
( $p \approx 100$  mbar) 1329 x 250 s = 95 h

Highest fraction in KATRIN will be  
T<sub>2</sub> (95%)  $\approx 3 \times$  HD (this Run)  
→ 0.1% seems to be feasible in 250 s



# Stability of long term runs

## Long term measurement - Laser power

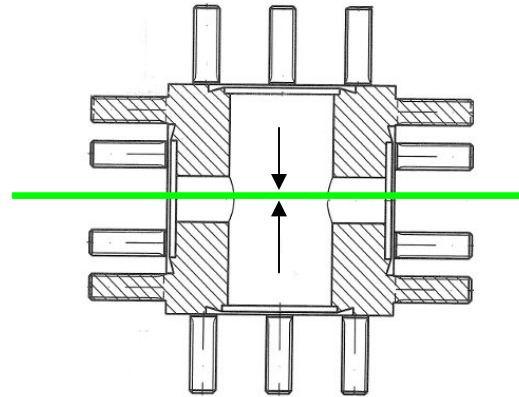


**Diametric trend of laser power / background visible!**

**Wear out of coating? Thermal misalignment? Pointing stability?**

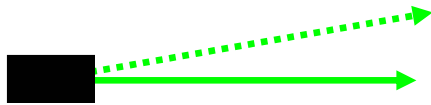
# Stability of long term runs

## Influence on laser power and background



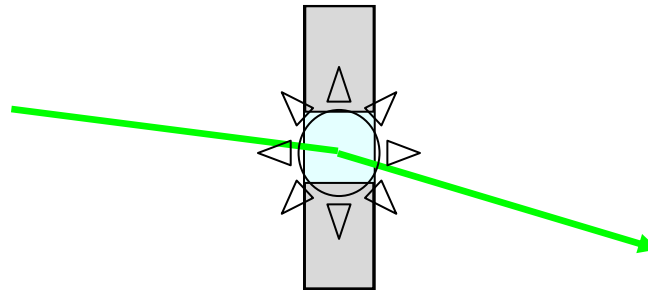
scattering region  
beam width  $\approx 20\mu\text{m}$

## Pointing stability



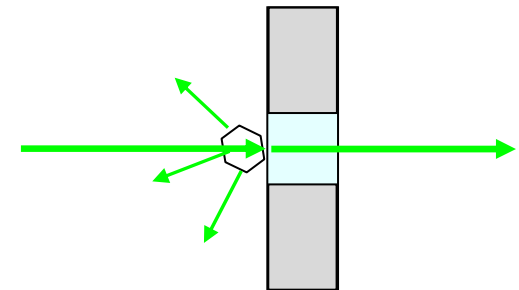
- Trace laser beam with modified webcam
- Laser service

## Thermal lensing



- Sequential shutter up/down measurements

## Dust on optics or laser beam

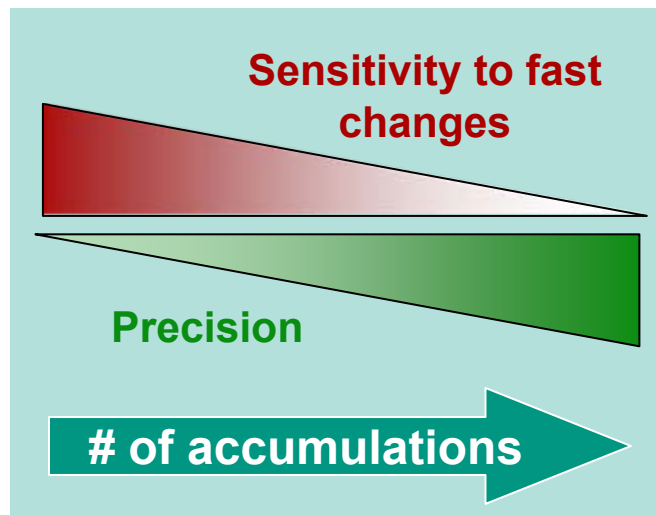
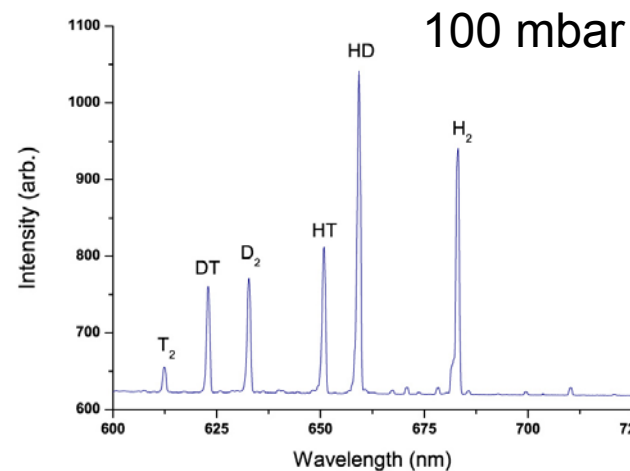
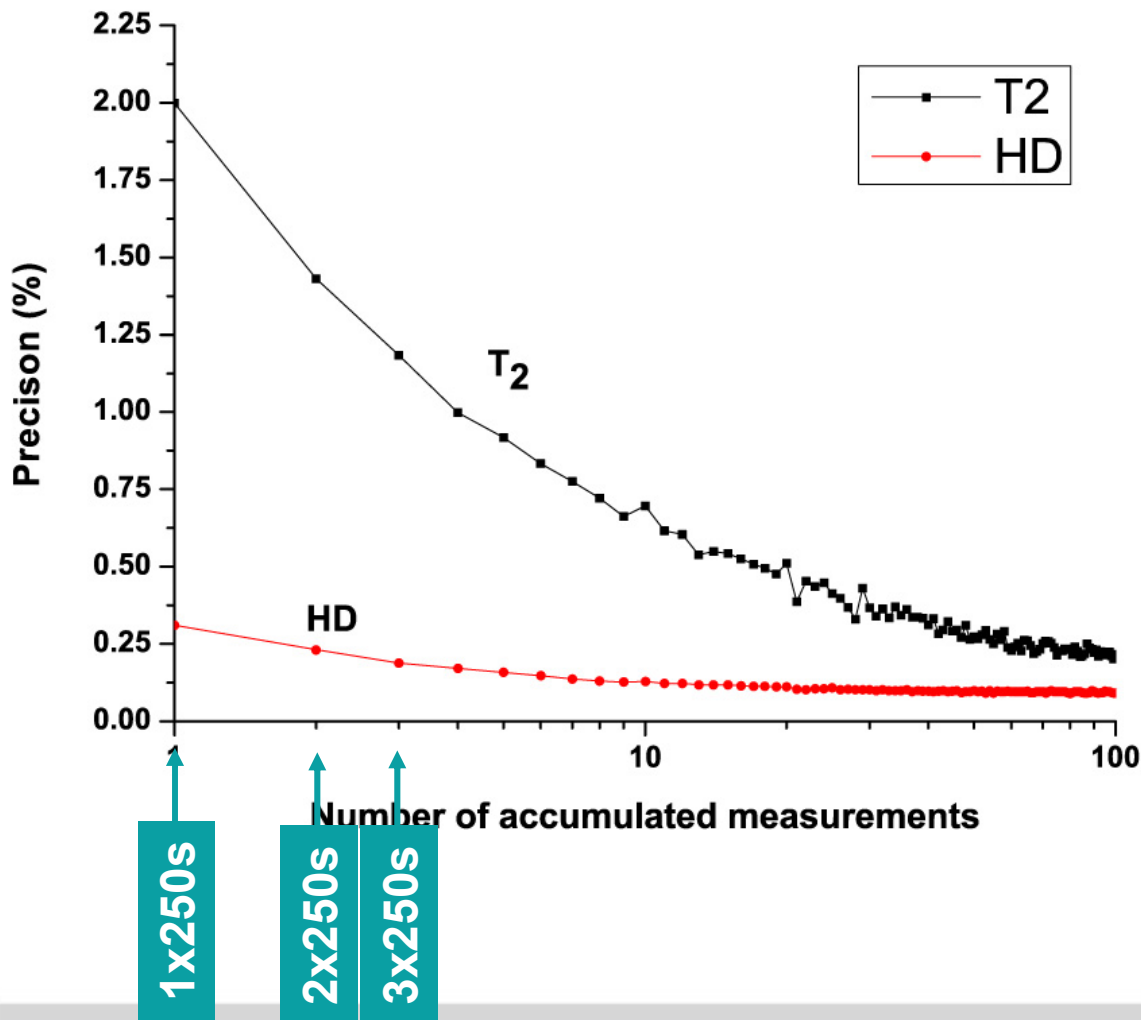


- Keep dust out (Tubes)

# Stability of the Laser Raman System

## Long term measurement

### Peak stability for accumulated intensity values



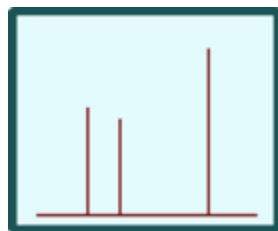
# Quantitative analysis

## Until now:

- Investigation of systematic effect via **peak area** measurements
- relative spectral intensities not proportional to relative composition

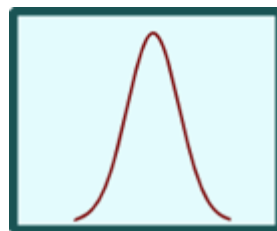
## Indirect Hard Modelling

- Uses quantum mechanical models for **quantitative** analysis
- Further advantage: Use of theoretical known peak shapes **reduces systematic uncertainties** (e.g. base line detection)



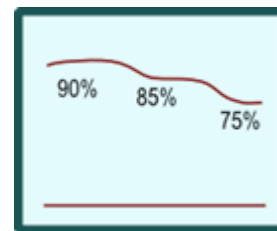
Intensities  
from QM

+

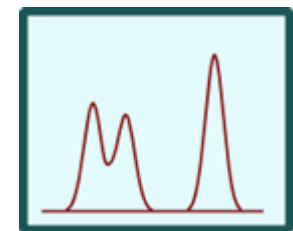


Spectrometer  
resolution

+



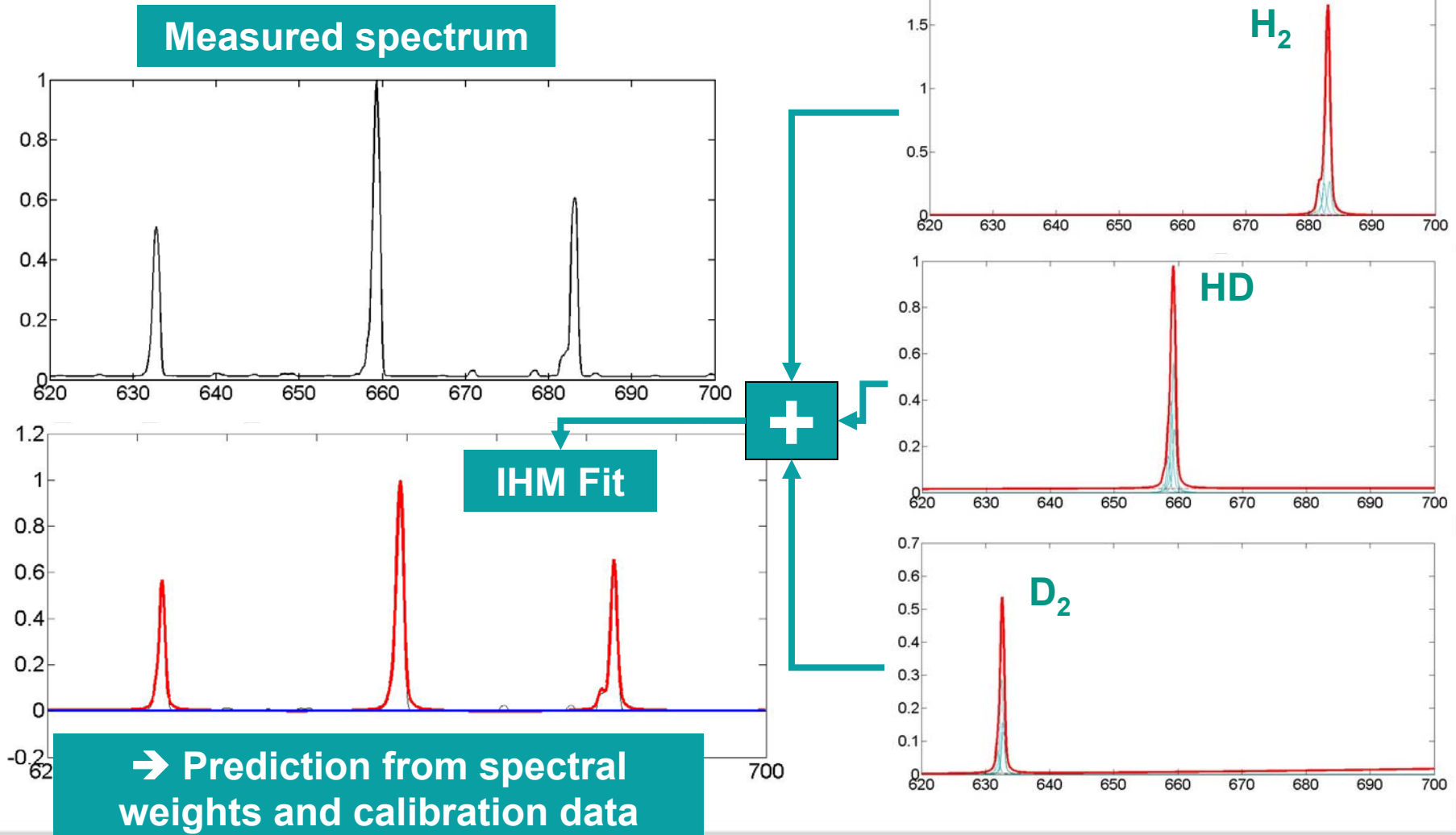
Spectral  
efficiency



Theoretical  
model

# Indirect Hard Modelling

## Principle of indirect hard modelling



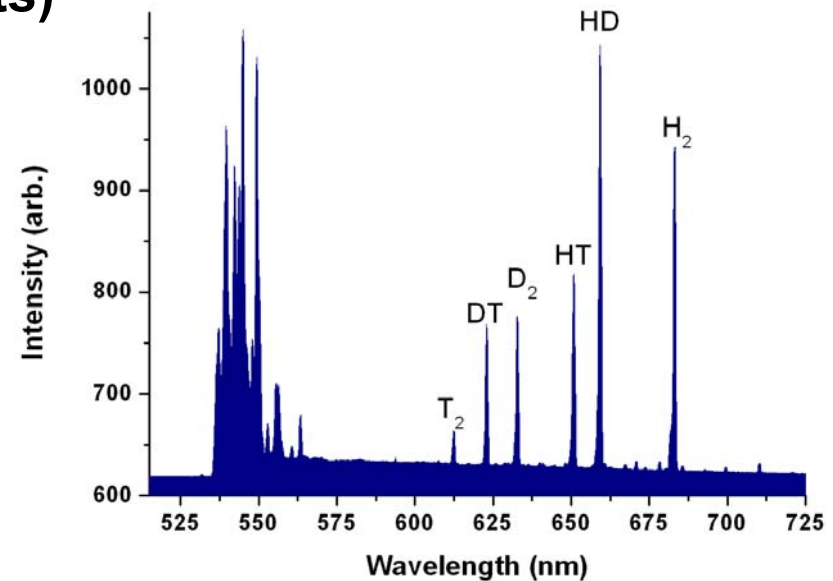
# Indirect Hard Modelling

## Comparison of precision (First results)

- First tests (demonstration)
  - semi-quantum mechanical model used
  - spectral corrections missing
  - no calibration
- Stability of relative measurements

| Precision    | T <sub>2</sub> | HD    |
|--------------|----------------|-------|
| “Peak areas” | 1.50%          | 0.28% |
| IHM          | 1.02%          | 0.20% |

*each time analysis of 322 spectra*



Most KATRIN-a-like  
measurement  
(p≈100mbar, 250s)

**IHM increases precision by about 30%**



# Summary and Outlook

- Laser Raman monitors the isotopic purity for KATRIN
- System has been investigated for more than 8 months on systematic effects (e.g. background, long term stability,...)

## Status

- KATRIN requirements (0.1% precision) reachable
- At the moment: Laser stability is not satisfying
- IHM method can improve analysis precision and allows quantitative analysis
- Actual detection limit < 0.06 mbar in 250 s



## Next steps

- Investigations and improvements of laser stability
- Theoretical modelling and spectra corrections for IHM
- Determination of spectroscopic data for tritium

**Michael Sturm, Sebastian Fischer, Helmut Telle, Richard Lewis,  
Magnus Schlösser, Beate Bornschein**





# Data-Processing

