Karlsruhe Institute of Technology

Tritium Laboratory Karlsruhe

Monitoring of the KATRIN source composition by Raman spectroscopy

Sebastian Fischer
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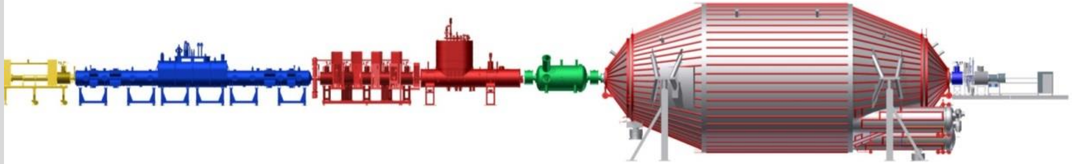
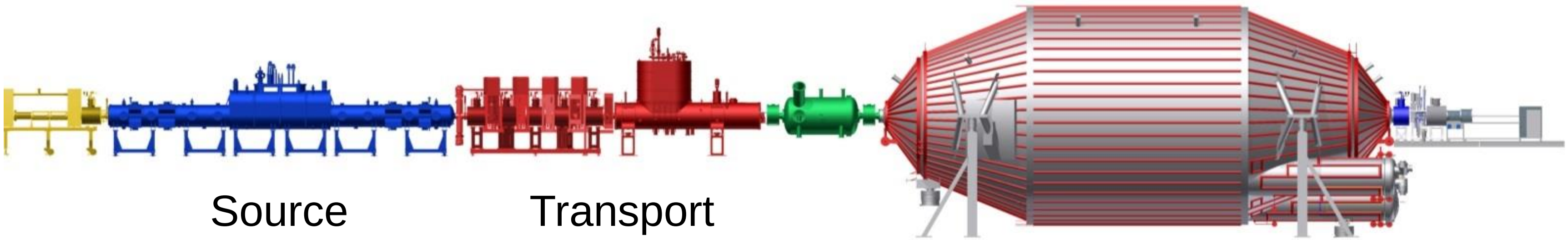
INSTITUTE OF TECHNICAL PHYSICS, TRITIUM LABORATORY KARLSRUHE



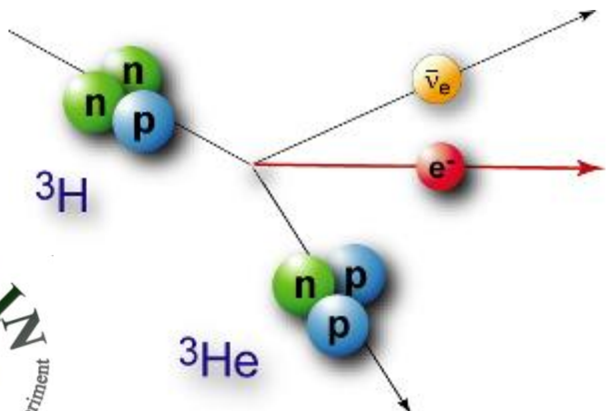
The KATRIN experiment



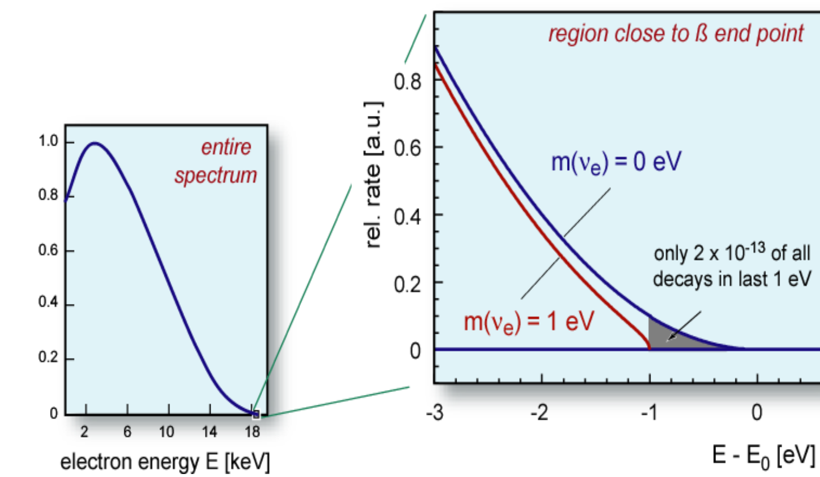
Source section

Transport section

Spectrometers and detector



$t_{1/2}$ = 12.3 years



$$\frac{\mathrm{d}N}{\mathrm{d}t} \propto p\cdot(E+m_e)\cdot(E_0-E)\cdot\sqrt{(E_0-E)^2-m_\nu{}^2}$$

Determination of neutrino mass with 200 meV/c² sensitivity (90 % C.L.)

$$m_\nu{}^2 = \sum_{i=1}^{3} |U_{ei}|^2 m_i{}^2$$

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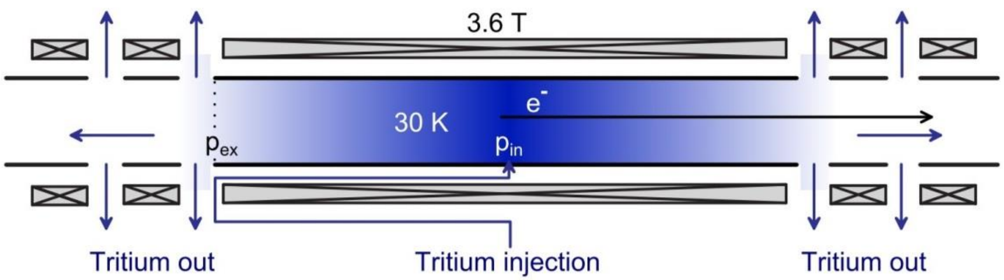
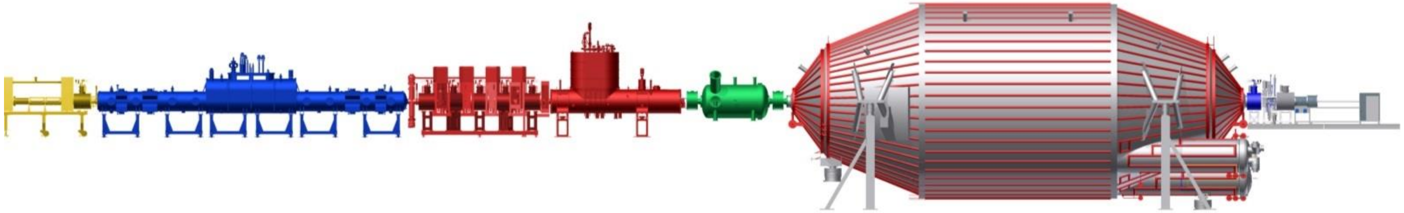
Windowless gaseous tritium source (WGTS)



- Continuous gas injection and removal
- Steady-state gas column inside source tube



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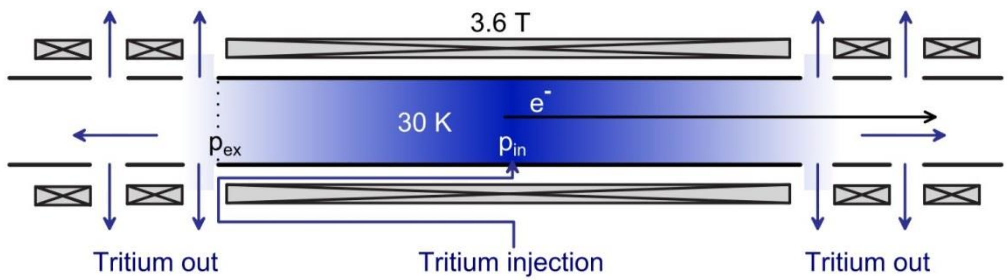

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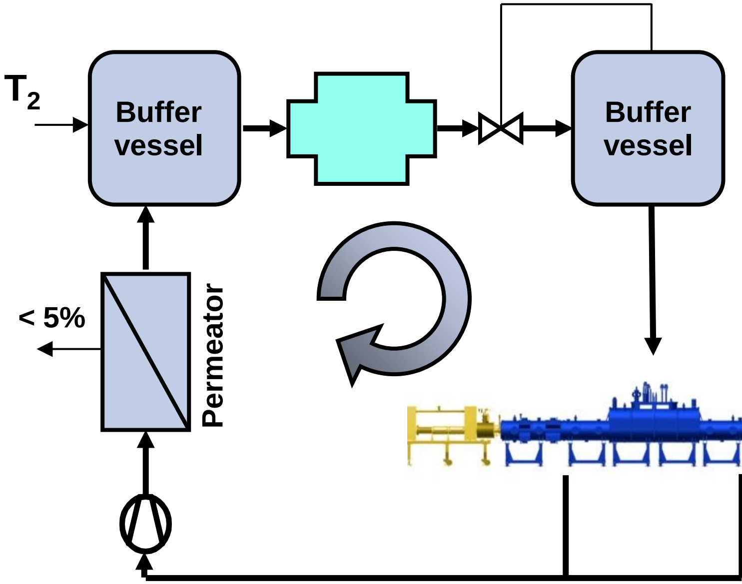
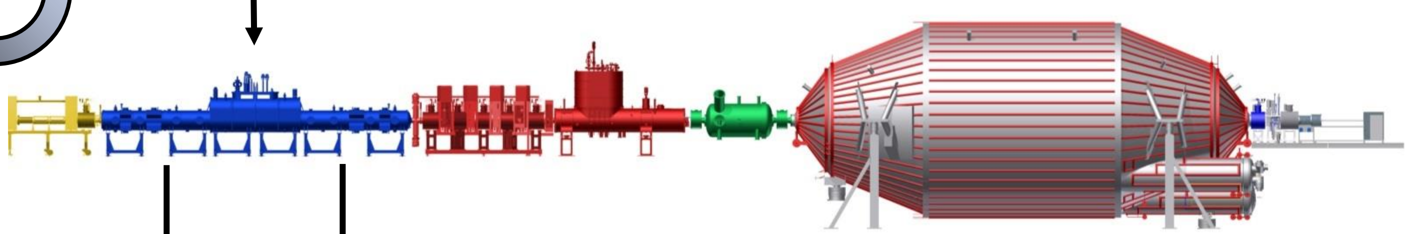


> 95% of tritium is kept inside "Inner loop"

Complete TLK infrastructure needed



Tritium throughput: 40 g / day

Control and monitoring of WGTS parameters

- Stability of WGTS is essential for m_ν measurement
- Essential source parameters are stabilized to 0.1% level
- Dedicated control and monitoring systems developed

  Monitoring of the operating parameters of the KATRIN Windowless Gaseous Tritium Source
  M. Babutzka et al., NJP 14 (2012) 103046

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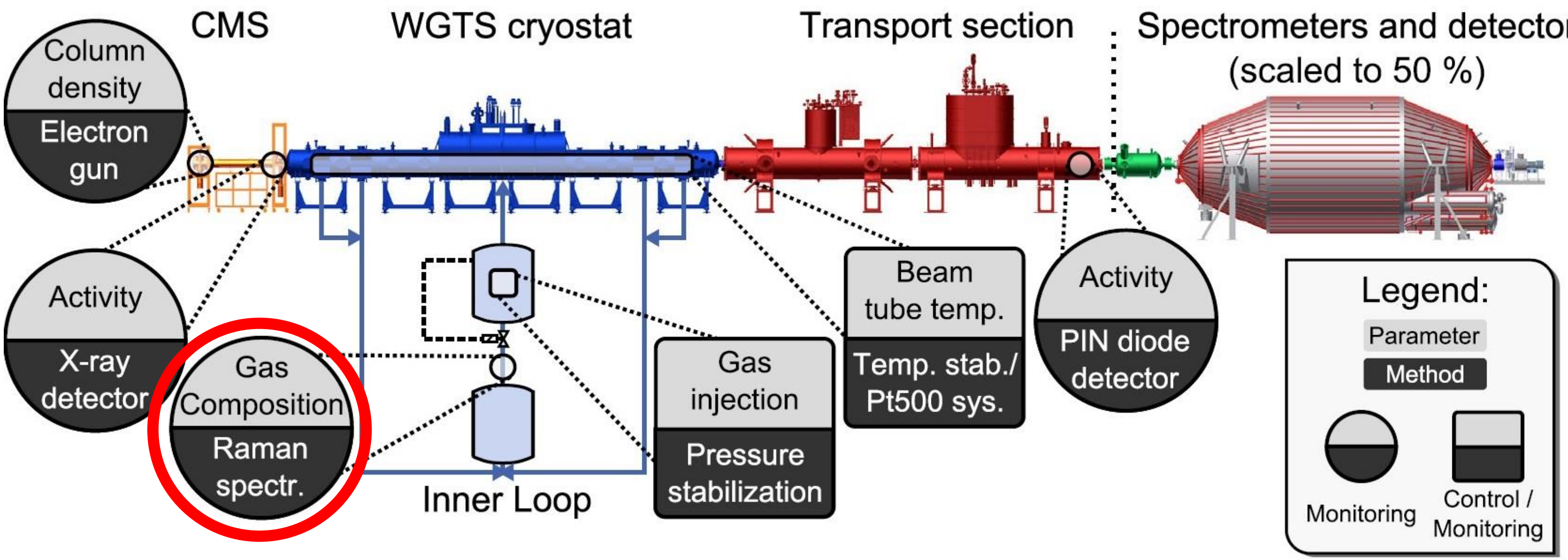

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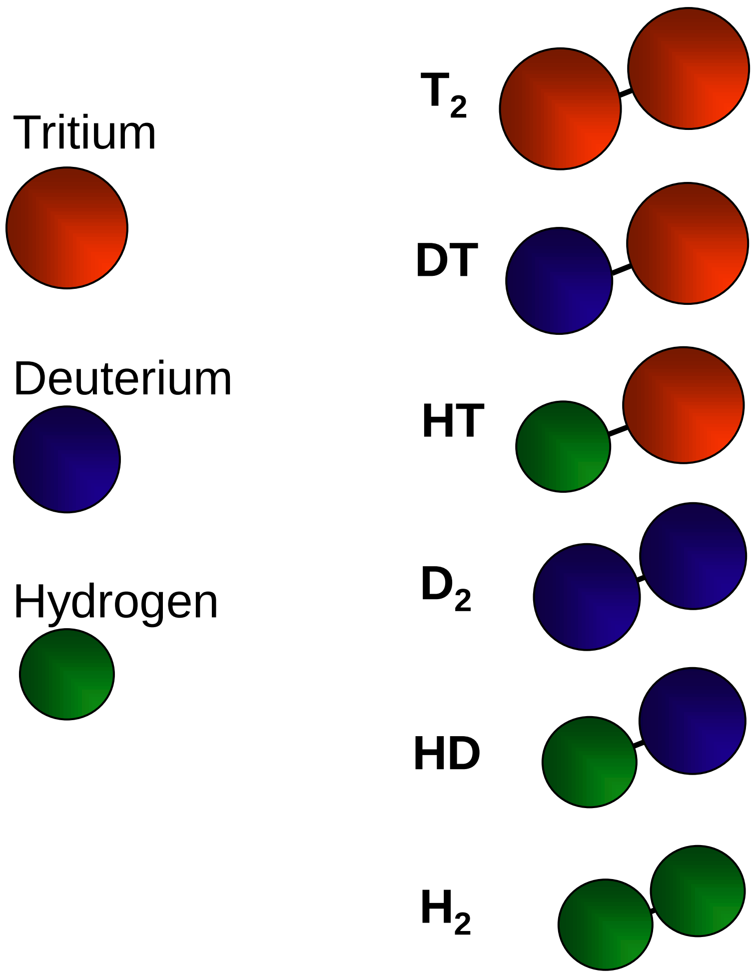
Gas composition inside WGTS

3 hydrogen isotopes

6 hydrogen isotopologues



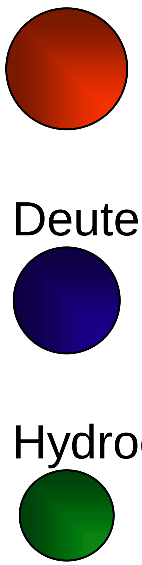
Gas composition inside WGTS

3 hydrogen isotopes

Tritium

Deuterium

Hydrogen



6 hydrogen isotopologues

T_2

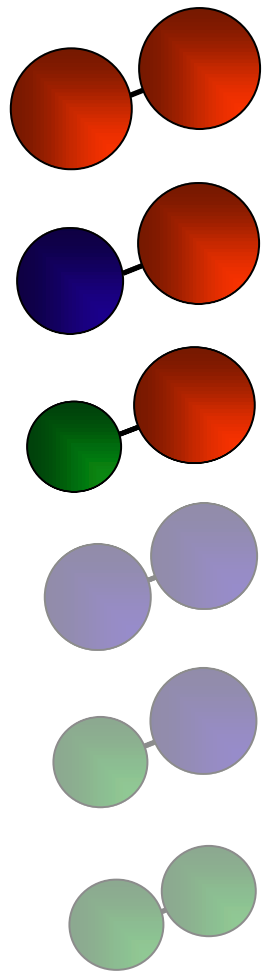
DT

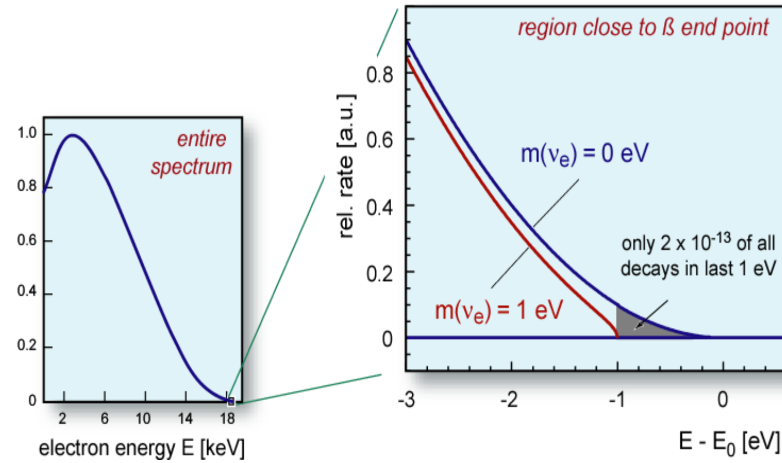
HT

D_2

HD

H_2





$> 91\ \%\ T_2$

$< 9\ \%\ DT$

$< 1\ \%\ D_2$

Tritium purity $\varepsilon_T > 95\ \%$

β-spectrum depends on gas composition

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Molecular effects on β spectrum

- Doppler broadening

$$\Delta E_{Doppler} \propto v_{therm} \propto \sqrt{\frac{R \cdot T}{m_{mol}}}$$

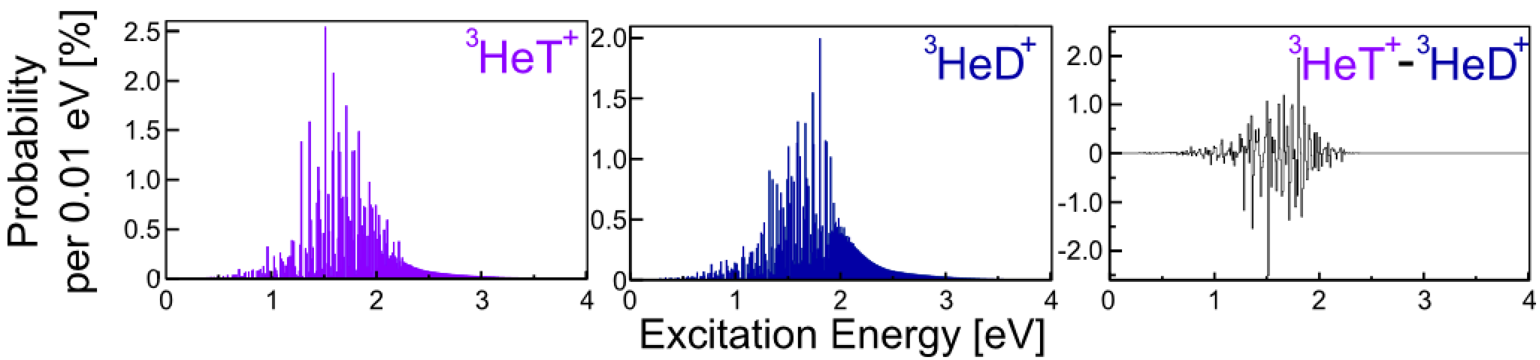
- Electron scattering with molecules

$$\Delta E_{scat,elast} \propto E_{e,kin} \cdot \frac{m_e}{m_{mol}}$$

- Nuclear recoil of daughter molecules (e.g. $^3HeT^+$)

$$E_{rec} \approx E_0 \cdot \frac{m_e}{m_{mol}}$$

- Final state distribution



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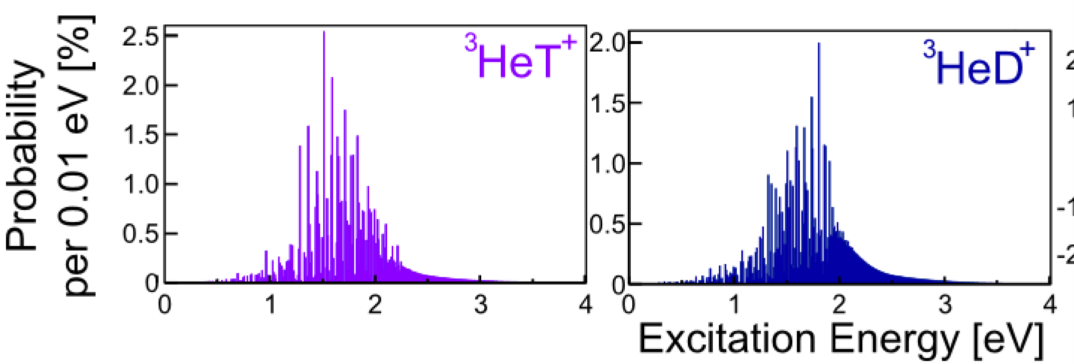


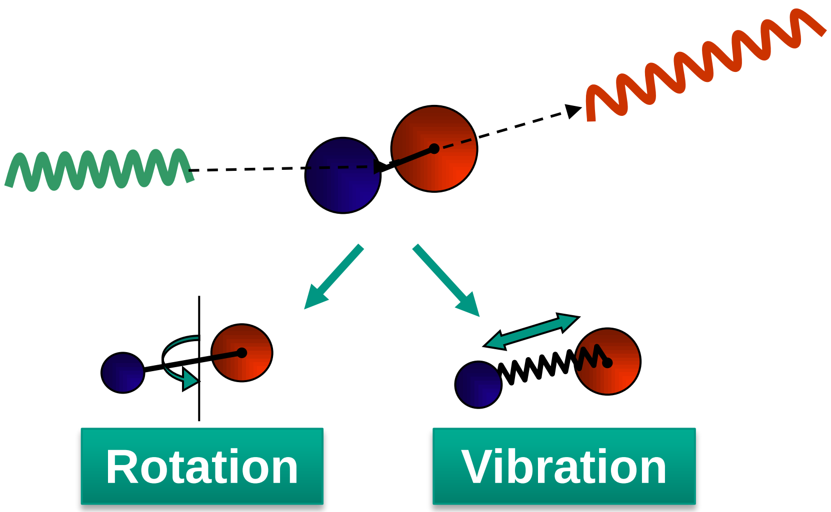
Continuous measurement of gas composition needed.

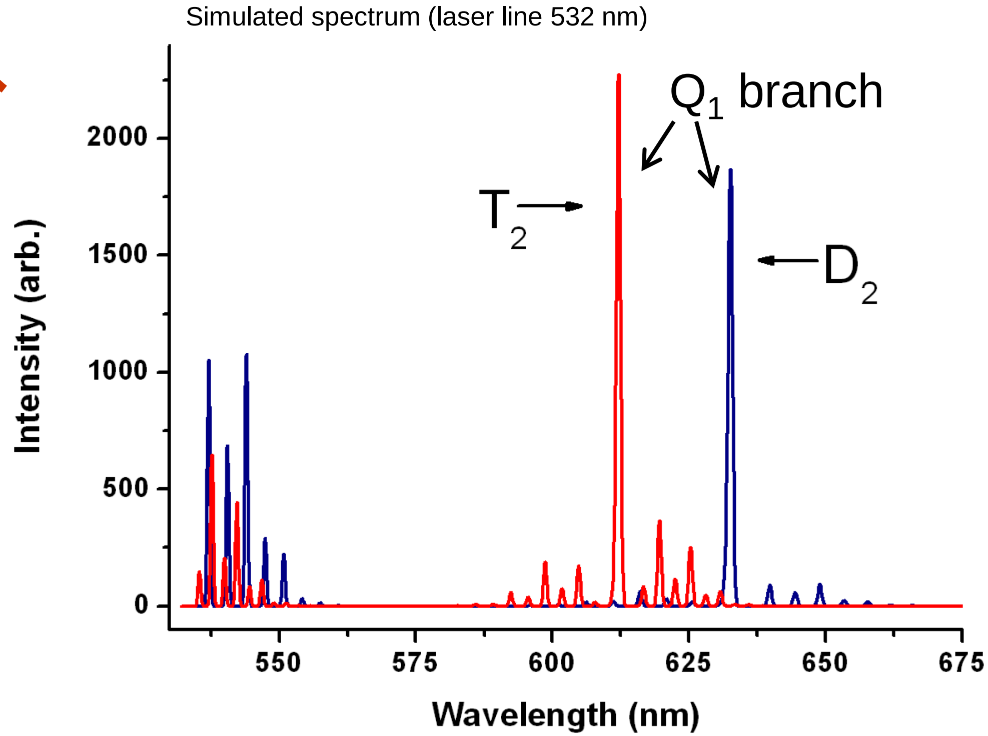
0.1% precision

< 10% accuracy

M. Schlösser et al., arXiv:1203.4099

The Raman Effect





Stokes Raman scattering

- Photon loses energy to molecule
  → Excitation of molecule
  → Change of wavelength

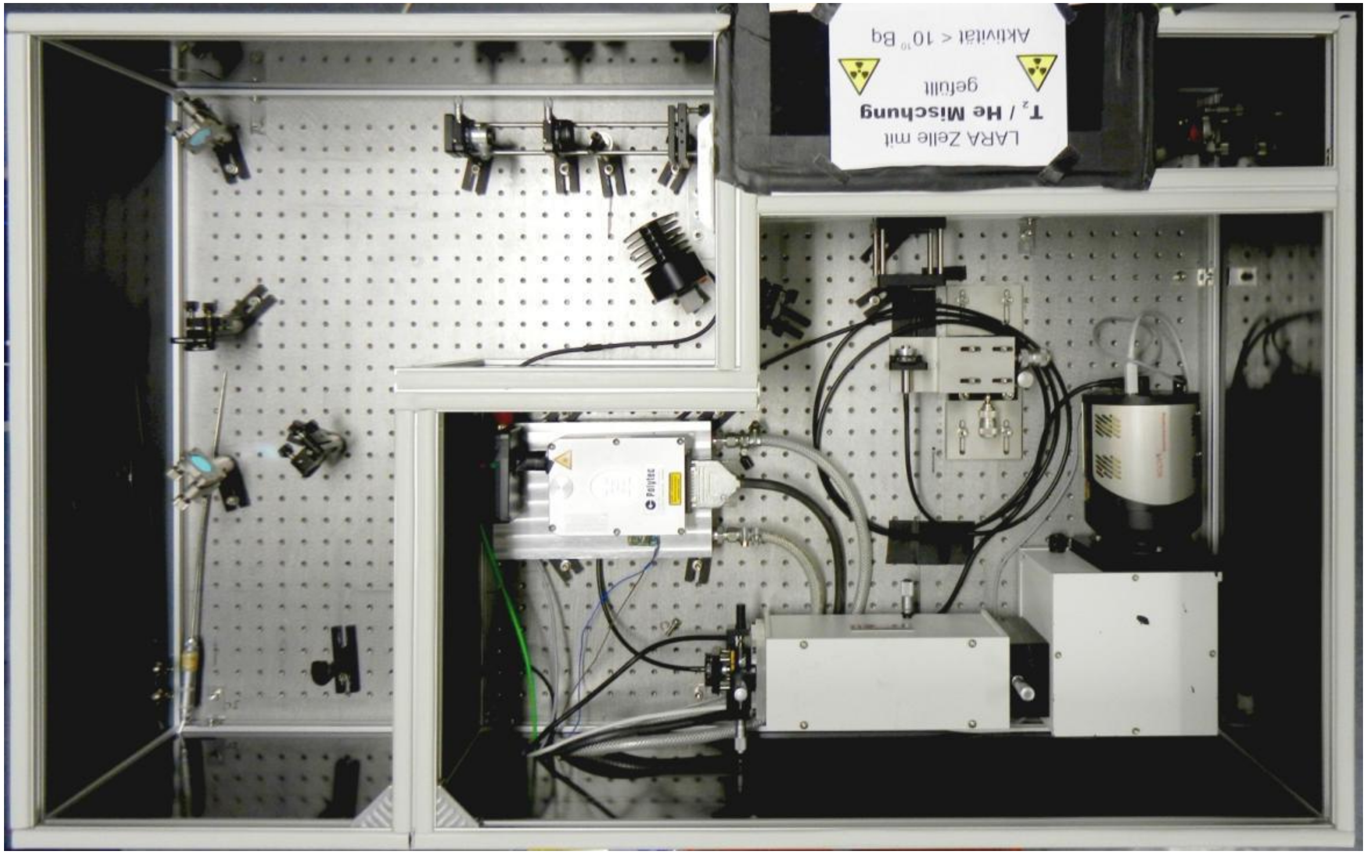
Analysis

- Line position → Qualitative analysis
- Line intensity → Quantitative analysis

Experimental setup

Swansea University
Prifysgol Abertawe

Karlsruhe Institute of Technology



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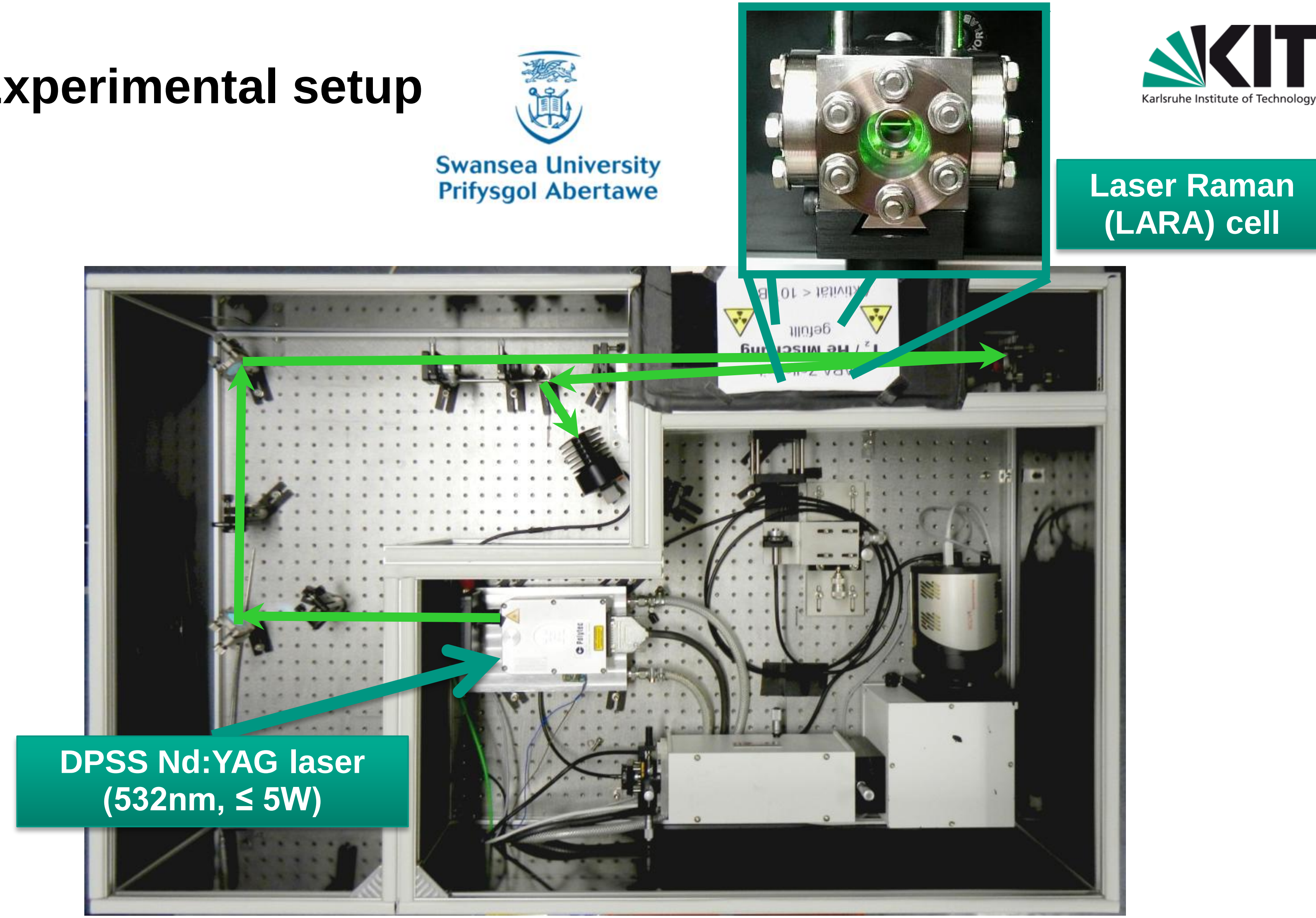

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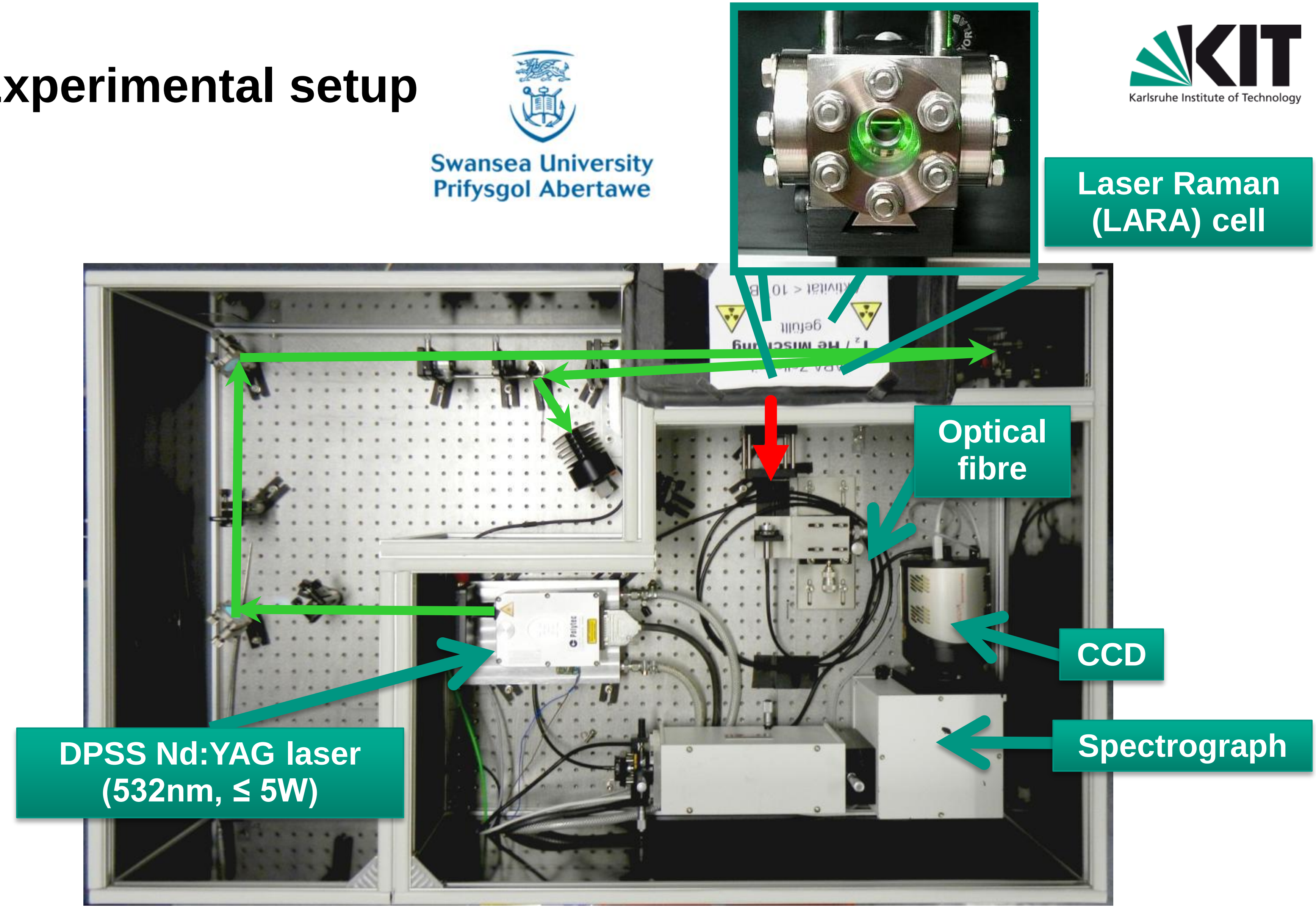
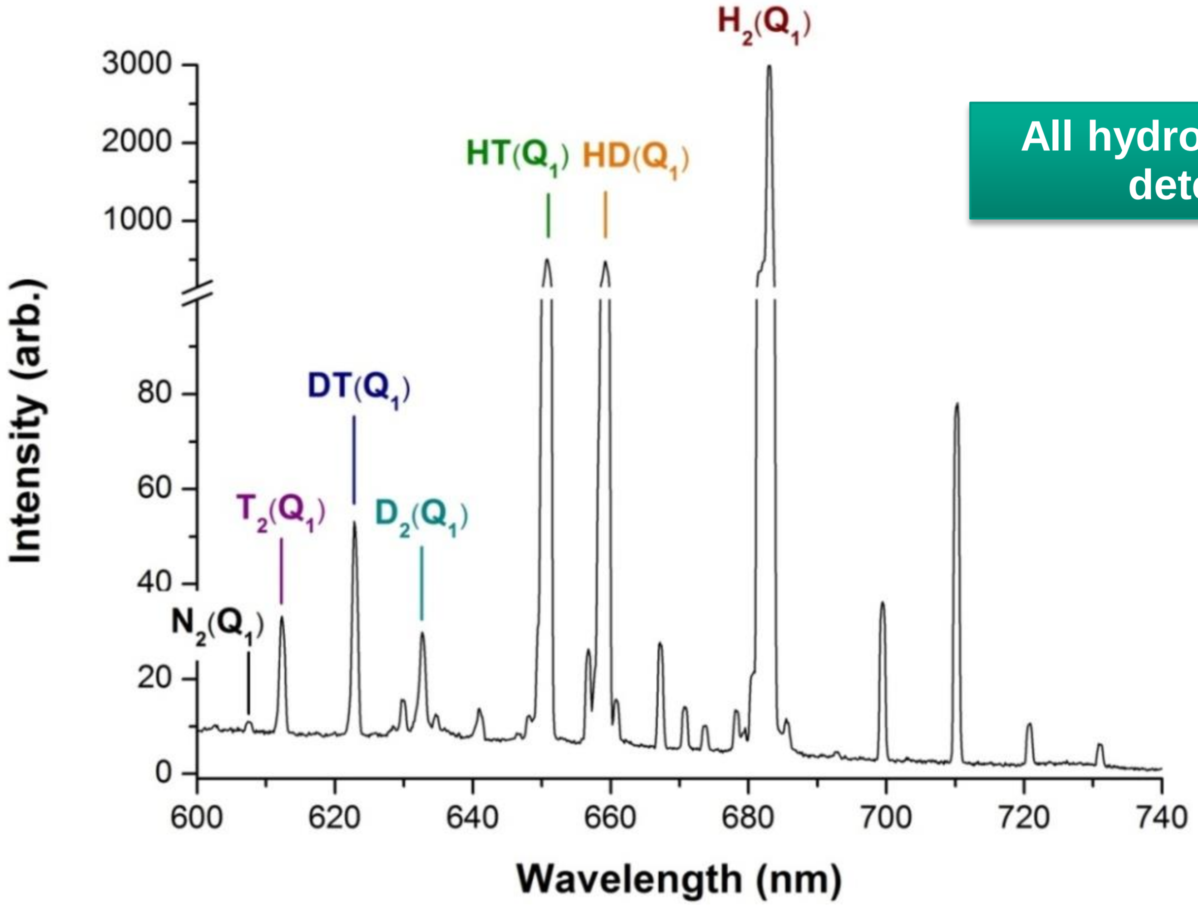


Proof of principle

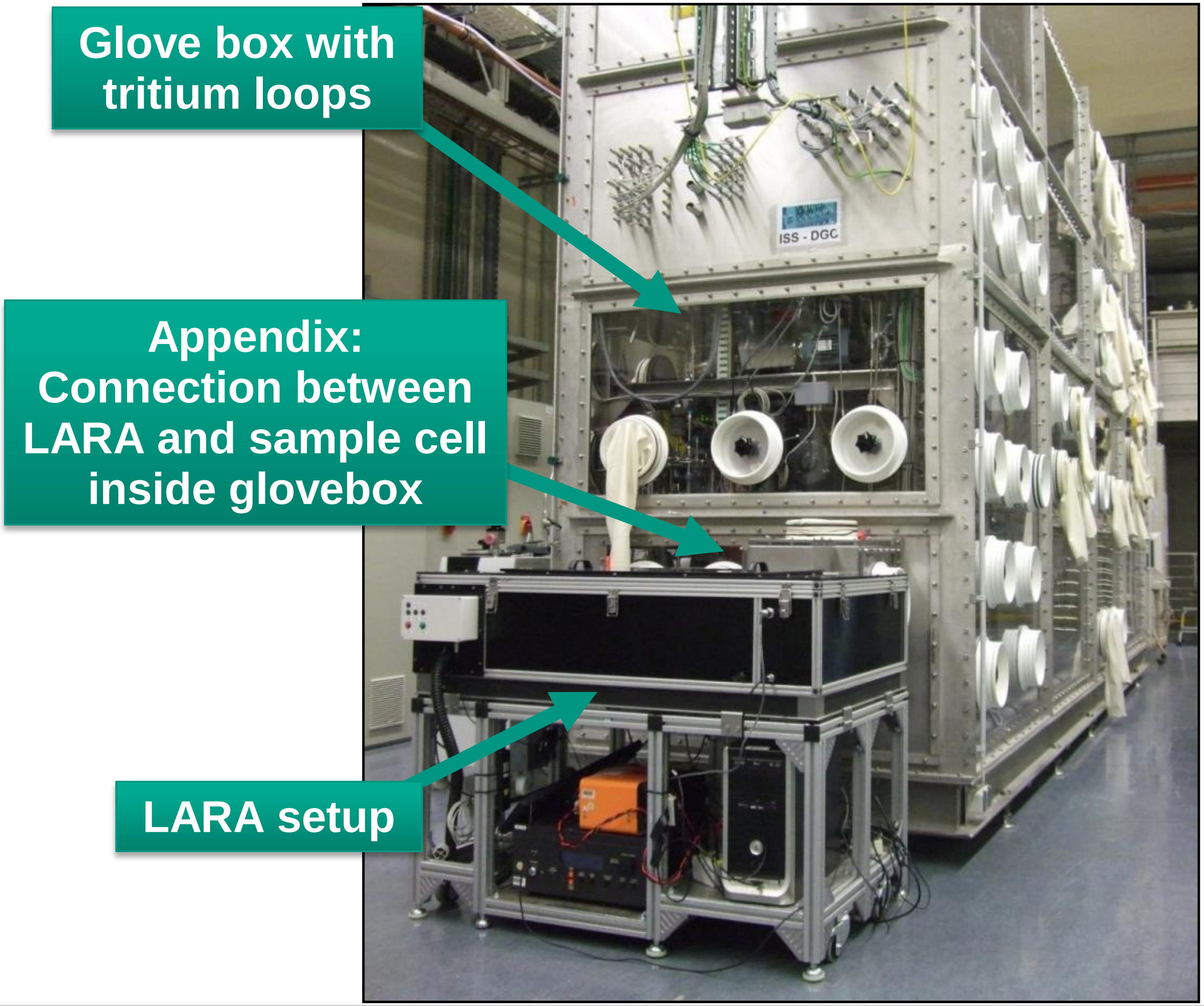
- Static samples with low tritium activity

M. Sturm et al., Laser Phys., 20, 2, 493 (2010)

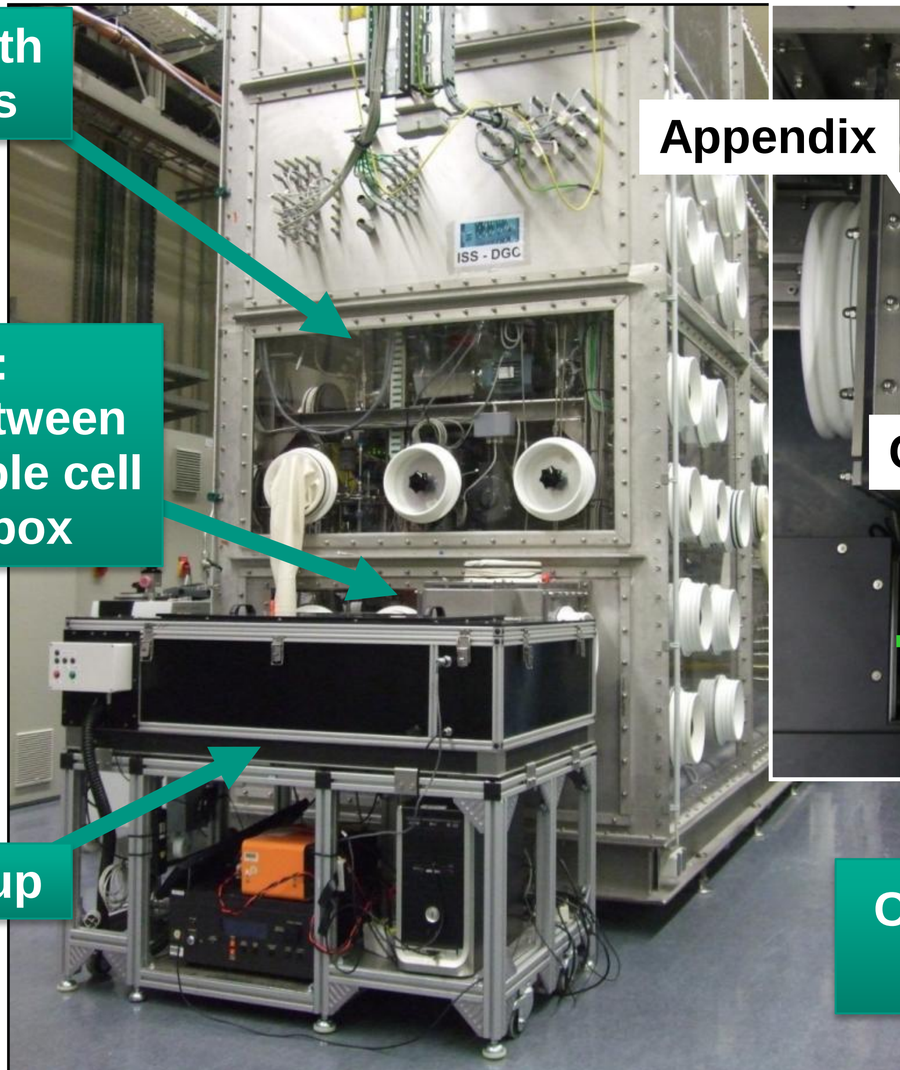
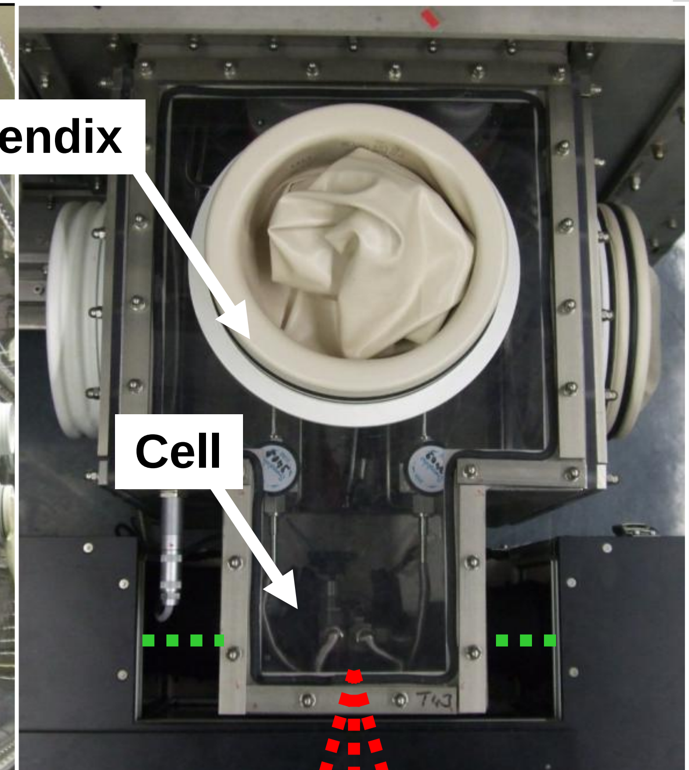


All hydrogen isotopologues can be detected simultaneously

LARA setup, tritium loops and the appendix



LARA setup, tritium loops and the appendix

Commissioned in 2009

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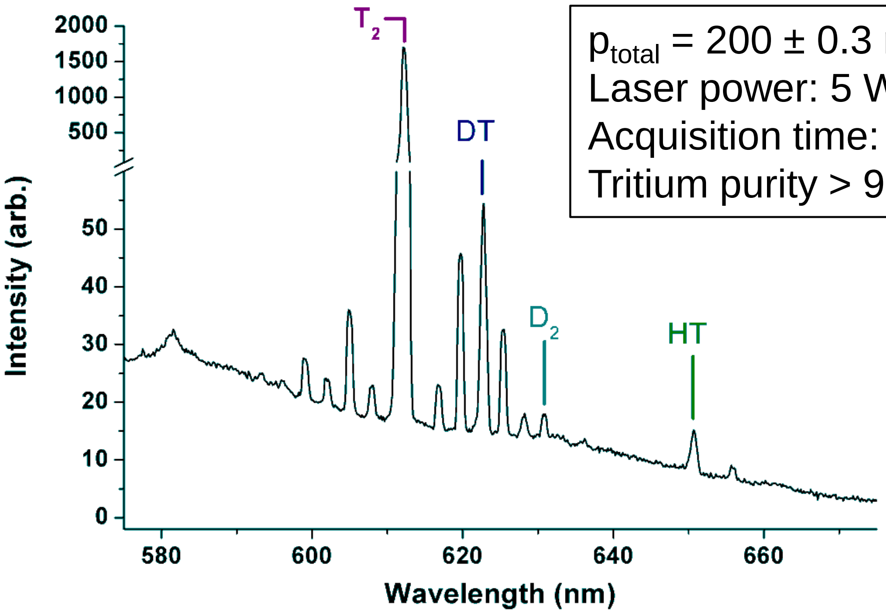
Long-term monitoring inside a test loop

- Non-stop monitoring for > 21 days



p_{total} = 200 ± 0.3 mbar
Laser power: 5 W
Acquisition time: 250 s
Tritium purity > 95 %

Long-term monitoring inside a test loop

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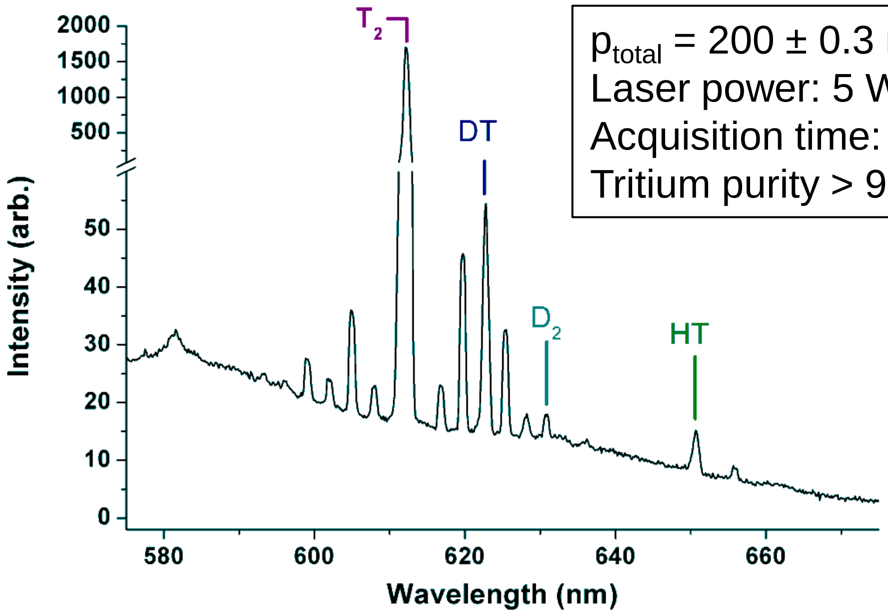

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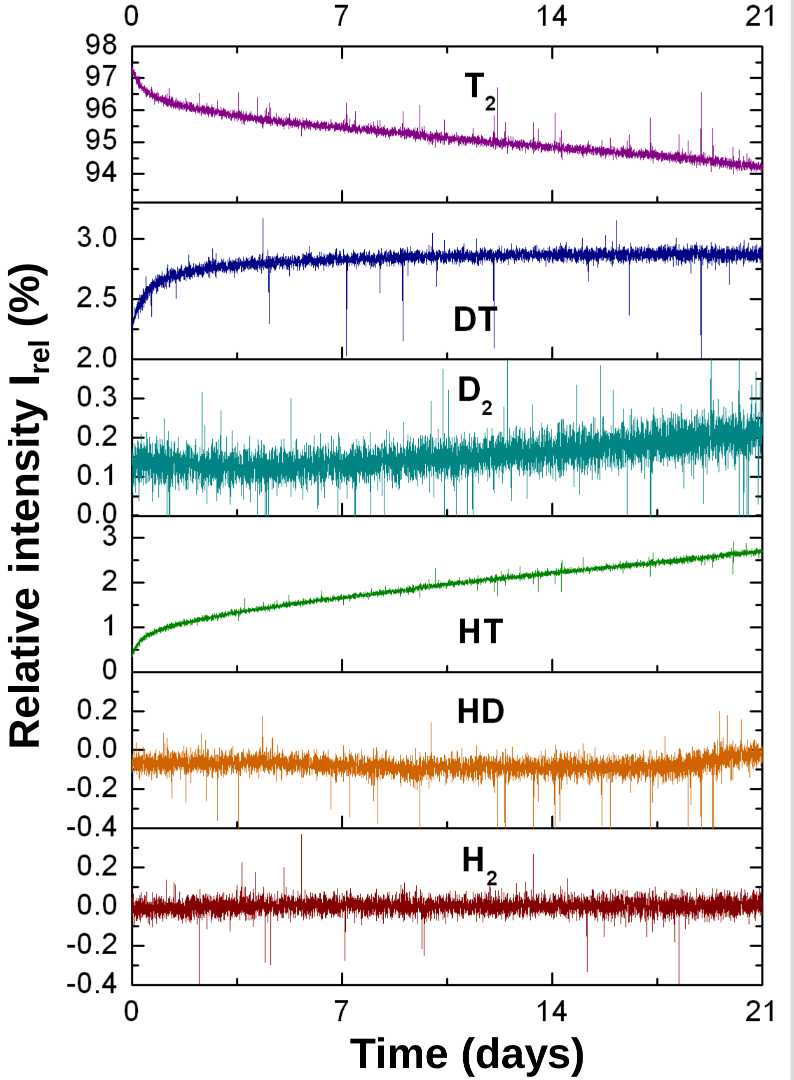

S. F. et al, Fusion Sci Technol. 60 3, 925-930 (2011)

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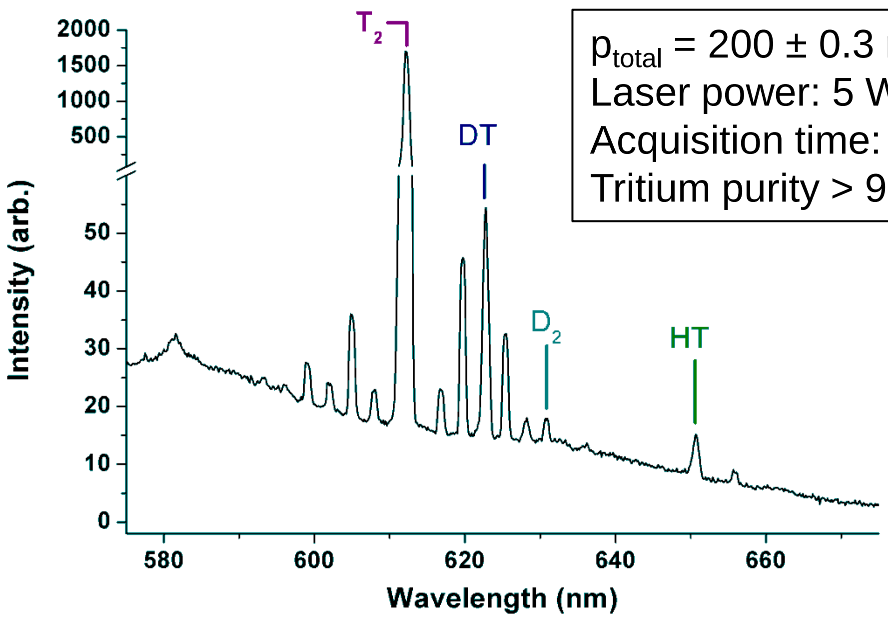

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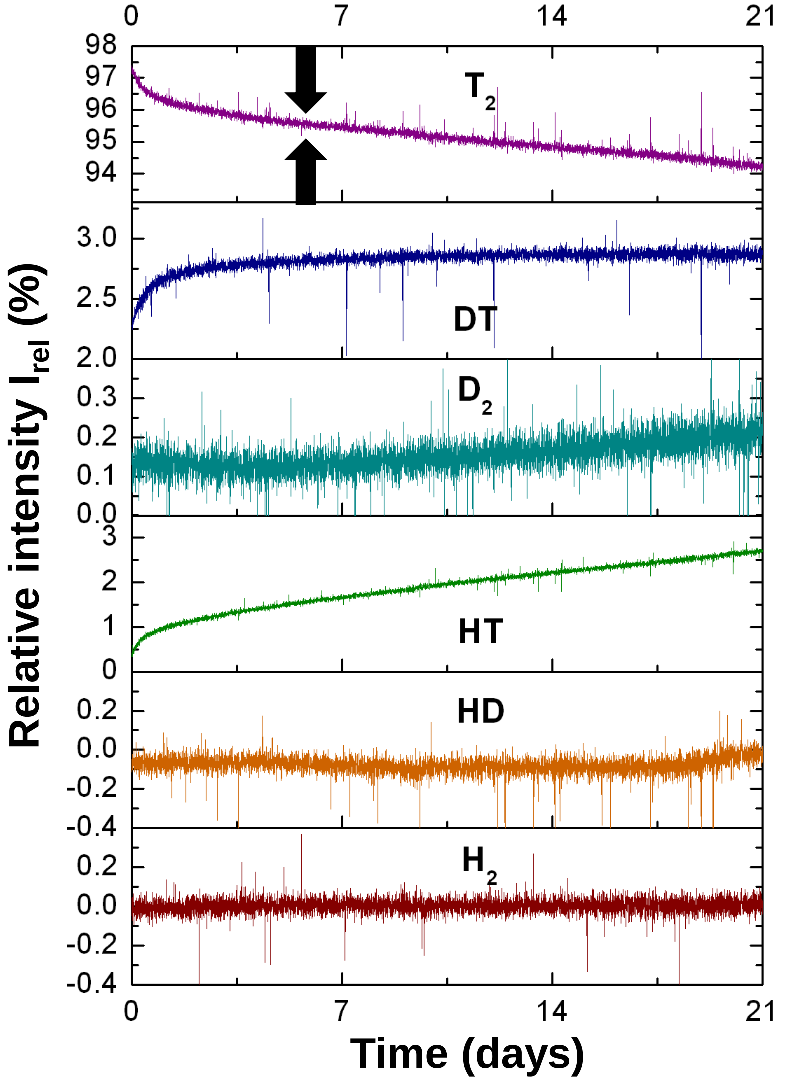


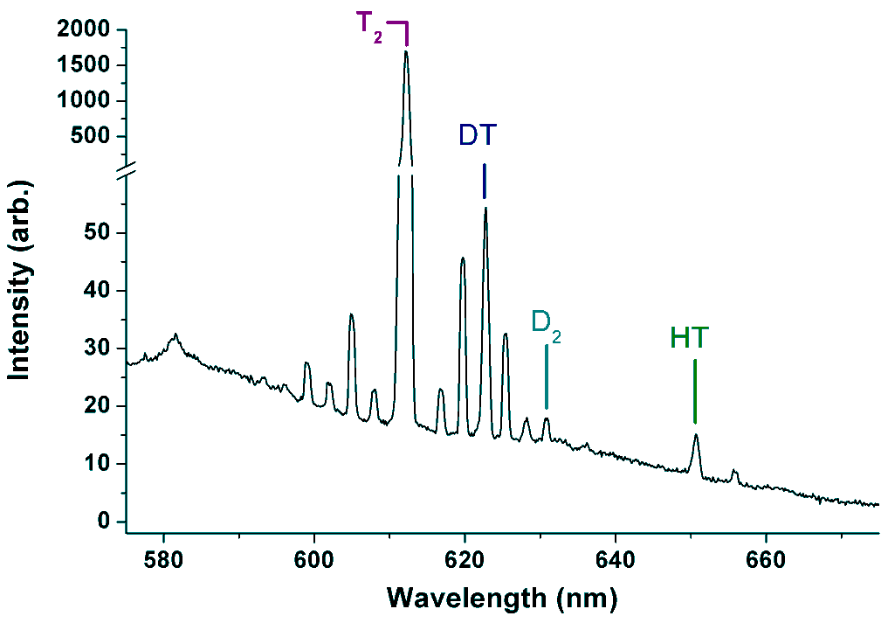
Changes on the 0.1% level can be monitored

S. F. et al, Fusion Sci Technol. 60 3, 925-930 (2011)

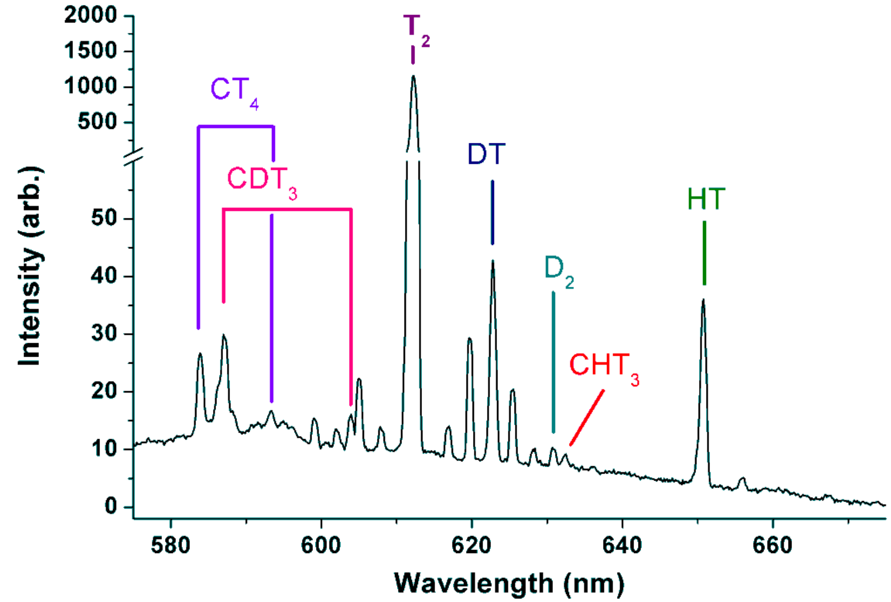
Long-term monitoring inside a test loop

- Generation of impurities

First spectrum



Last spectrum (after > 21 days)

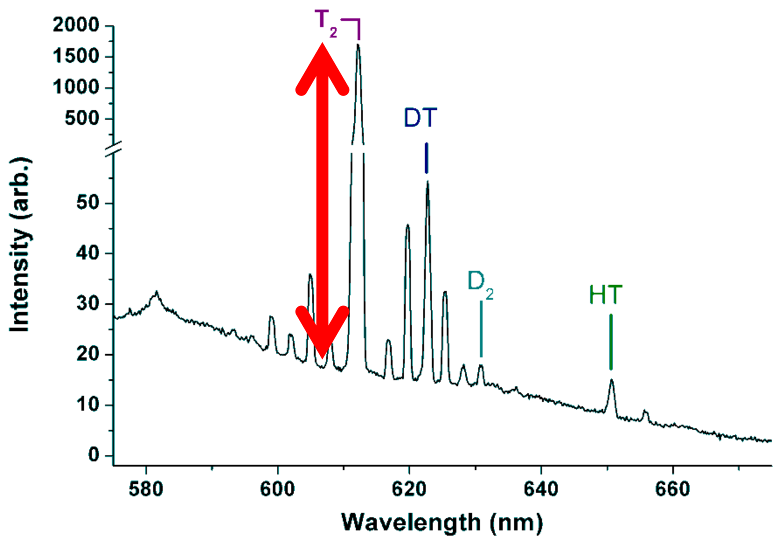


Formation of tritiated methane species (from carbon in stainless steel)

Less prominent formation in inner loop expected (due to permeator)

S. F. et al, Fusion Sci Technol. 60 3, 925-930 (2011)

Everything done? Not yet

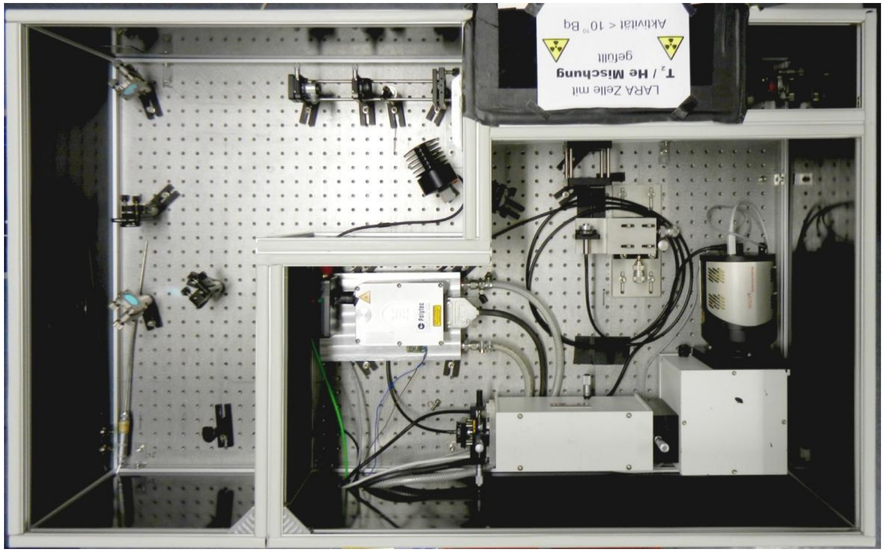


Extraction of peak intensities

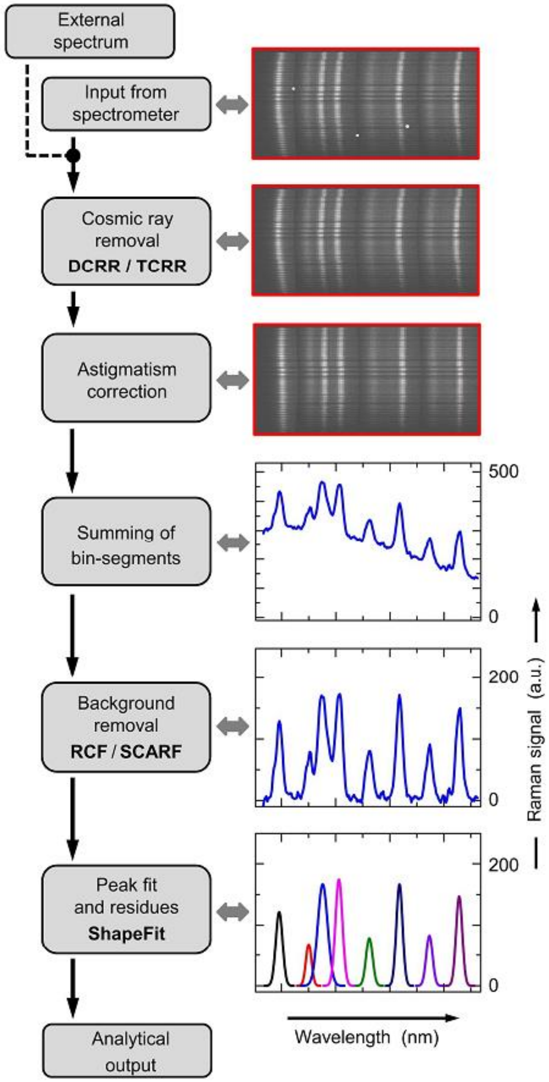
- Accurate, automated data analysis
- Conversion of peak intensities into concentration → Calibration

Hardware

- Simplification of beam path
- Monitoring of system performance
- Tritium resistant optical coatings



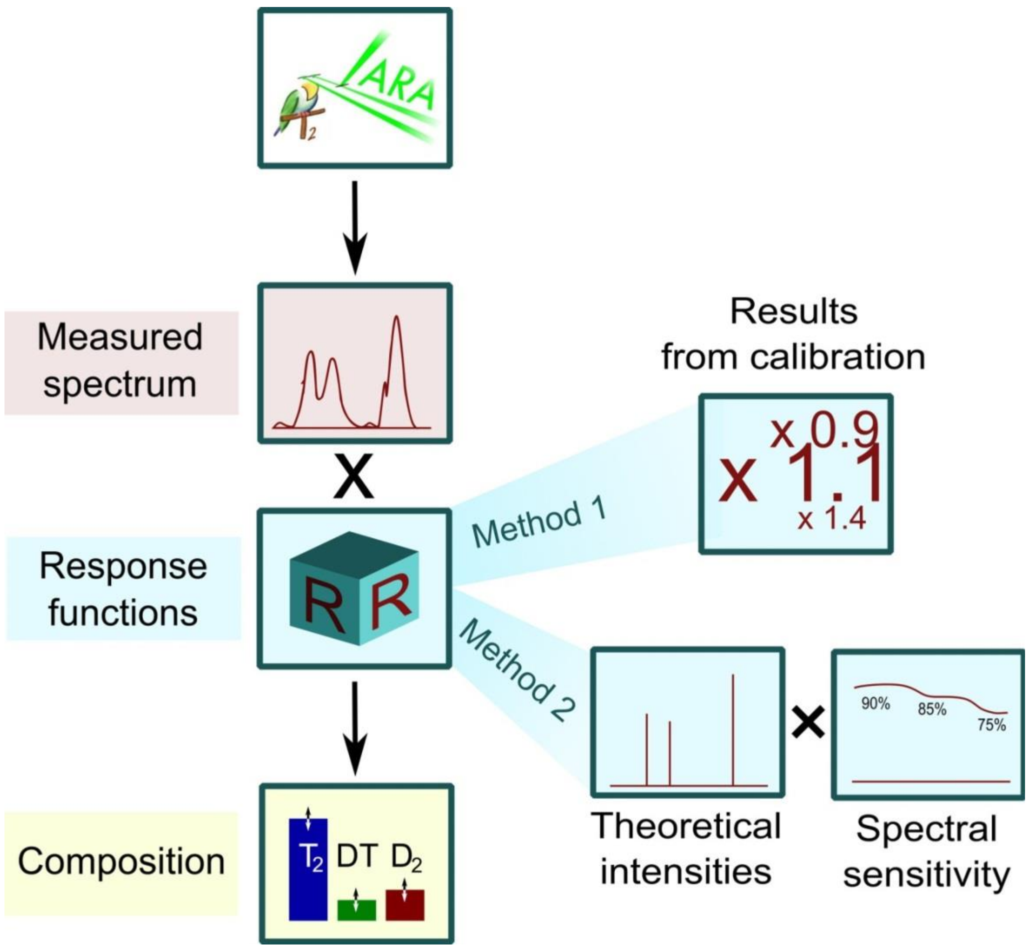
Data analysis: Accurate, robust and automated



- Development of analysis chain
  - Fully documented and tested

    T. M. James et al., Applied Spectroscopy 67 (8) 949 (2013)
  - LabVIEW code available on http://spectools.sourceforge.net
- Real time analysis implemented into data acquisition
- Validation
  - Analysis of ambient air. Extraction of natural abundance of ^{17}O, ^{18}O, ^{15}N
  - Application in calibration of LARA system

Calibration



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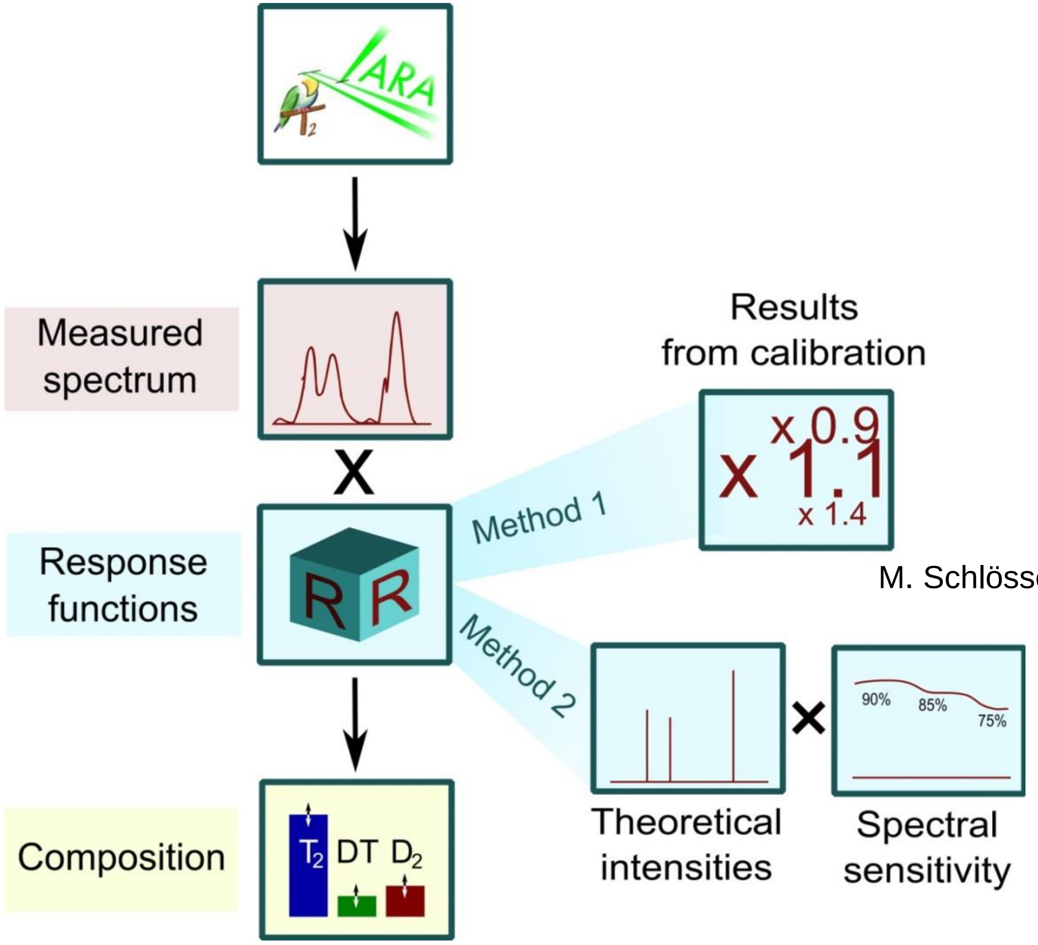
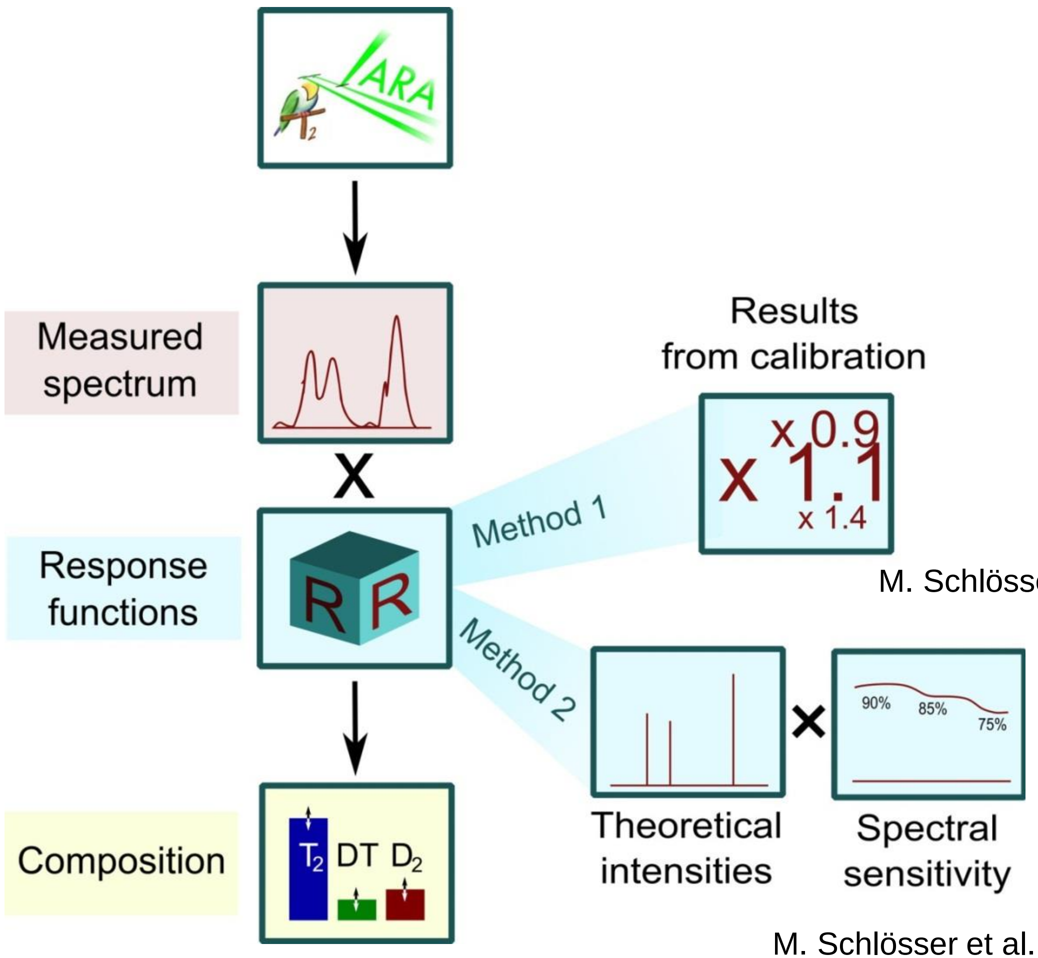


Only possible for H_2, HD, D_2
Calibration uncertainty < 2%

M. Schlösser et al., Anal. Chem., 85 (5) 2739–2745 (2013)

Calibration



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Applicable to all isotopologues

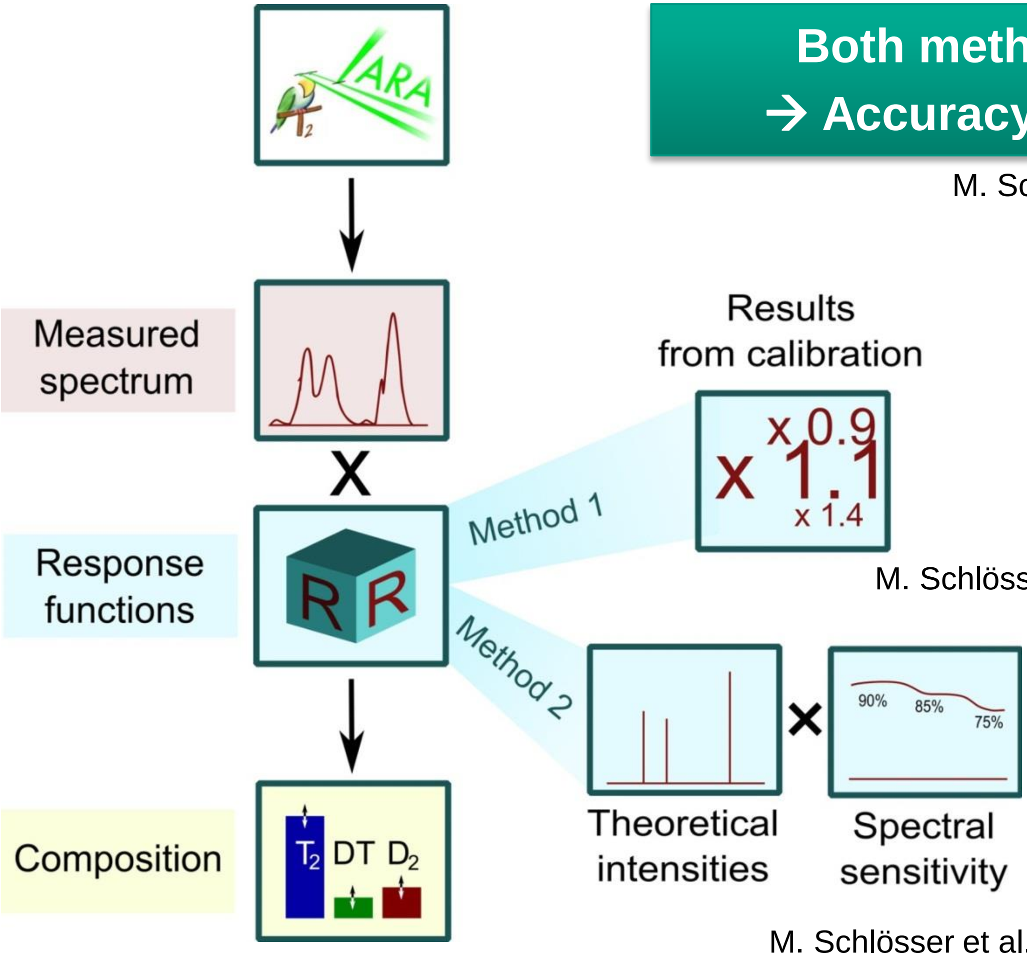
Validation of molecular constants within 3%

M. Schlösser et al., J. Raman Spectrosc., 44 (6) 857-865 (2013)
M. Schlösser et al., J. Raman Spectrosc., 44 (3) 453-462 (2013)

Calibration

Both methods agree within exp. errors.
→ Accuracy of Raman measurement < 6%

M. Schlösser et al., J. Mol. Struct., 1044 61-66 (2013)



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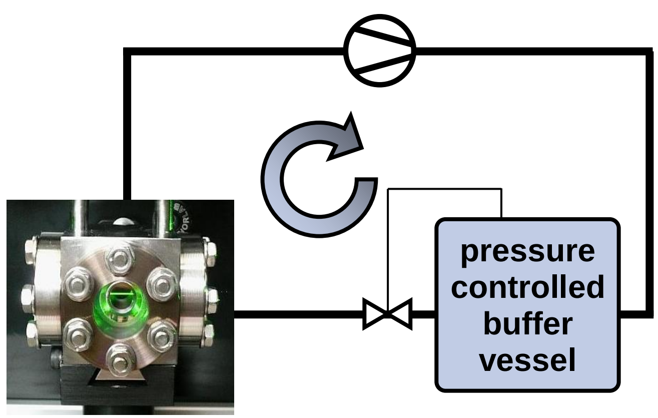
Coating degradation

- Anti-reflection coated windows (electron beam deposition)
- Degradation of coating on inner window surface after 3 months exposed to nominal tritium atmosphere (p = 200 mbar)



Damage not acceptable for long-term operation

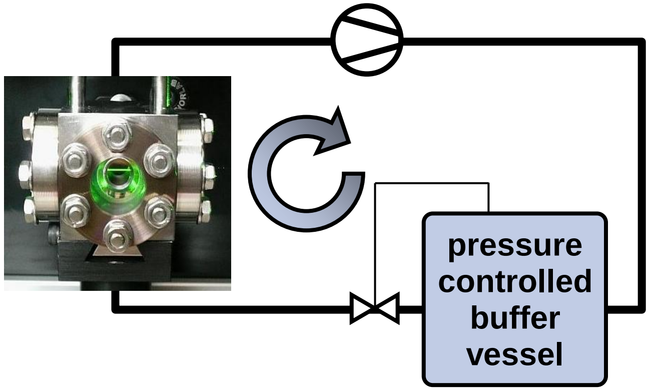
(Potential) reasons for coating damage

- Radiation damage

Other cells were successfully operated with pure tritium → Radiation damage is unlikely

- Formation of hydrofluoric acid (TF)

  Clark, Shanahan WSRC-STI-2006-00049 (2006)



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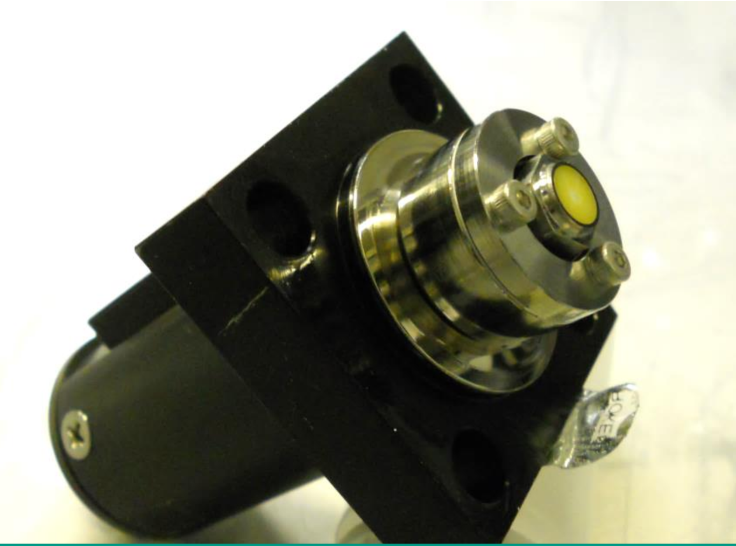
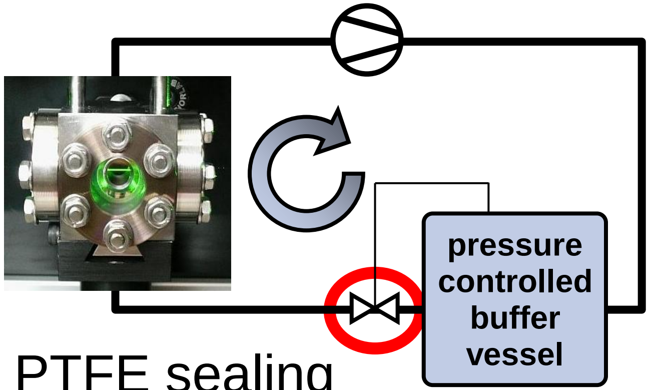
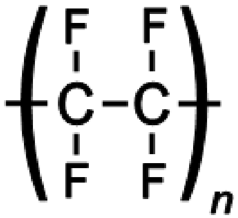


Discoloration of PTFE sealing → Indication for TF formation



Valve with PTFE sealing

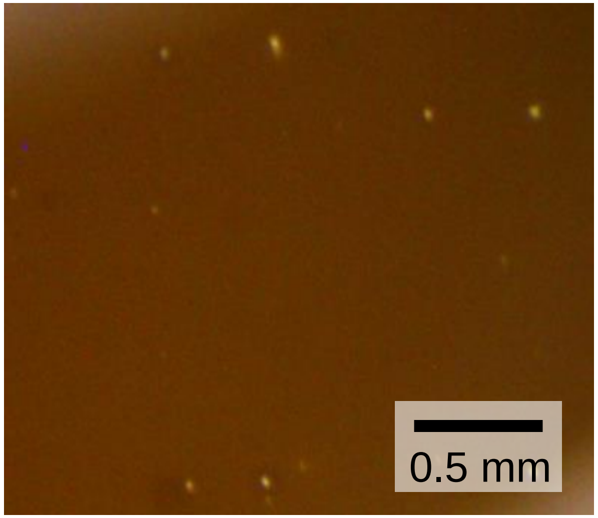


Approaching the problem

- Long-term test without PTFE sealing valve

Severe damage not reproduced

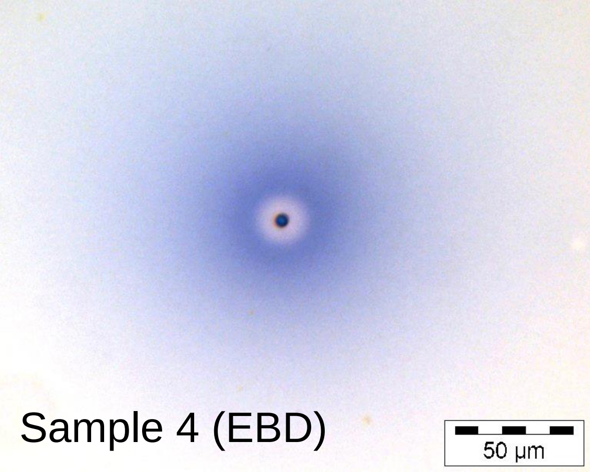
Small spots appeared on inner surface
→ No issue yet, but in future?



- Test of commercially available coatings
  - Sputtering vs. Electron beam deposition
  - 4 manufacturing methods tested
  - Sputtered coatings are likely more resistant than electron beam deposited ones

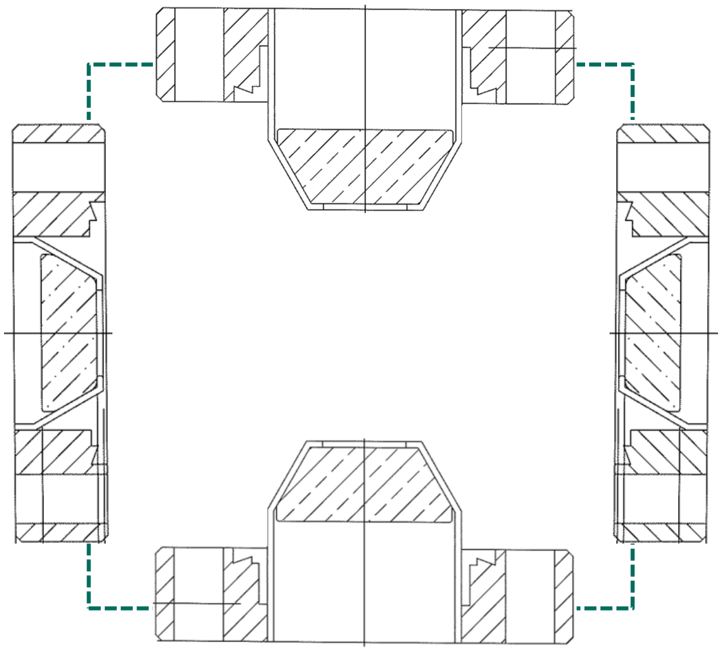
Spots on EBD coating
Intrinsic weakness of EBD coating?

No effects on sputtered coatings



(Probably) solving the problem

- Current interpretation
  - Severe damage was caused by HF formation
  - Spots observed in tests due to weakness of EBD coatings



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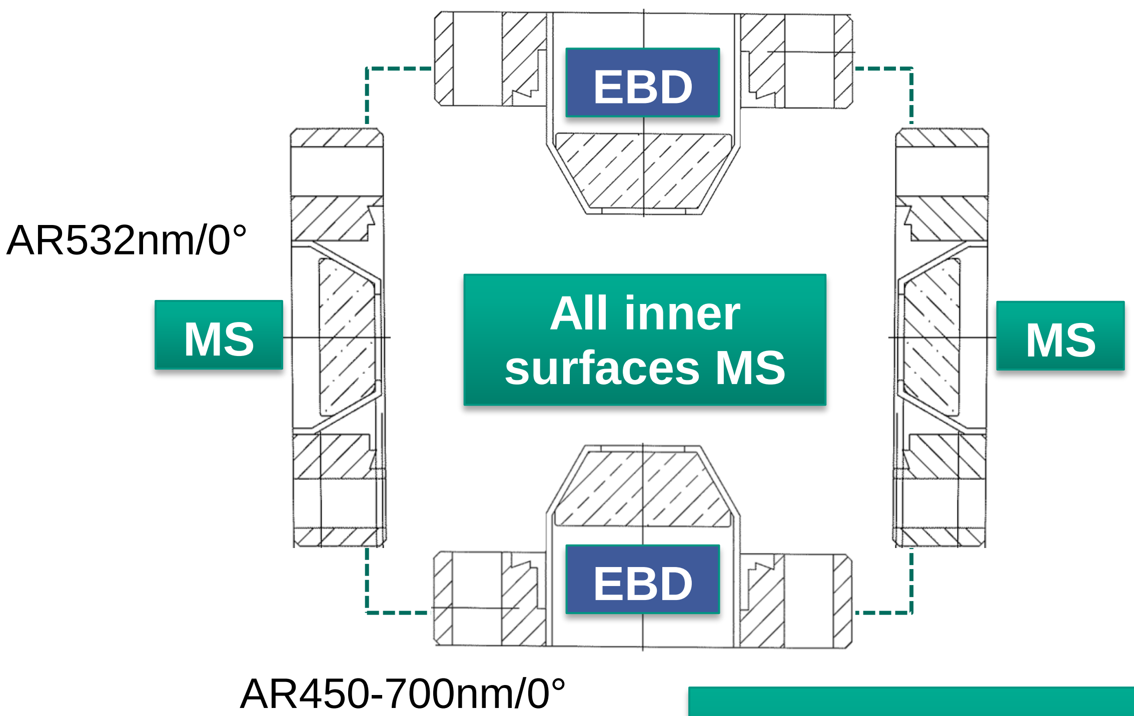


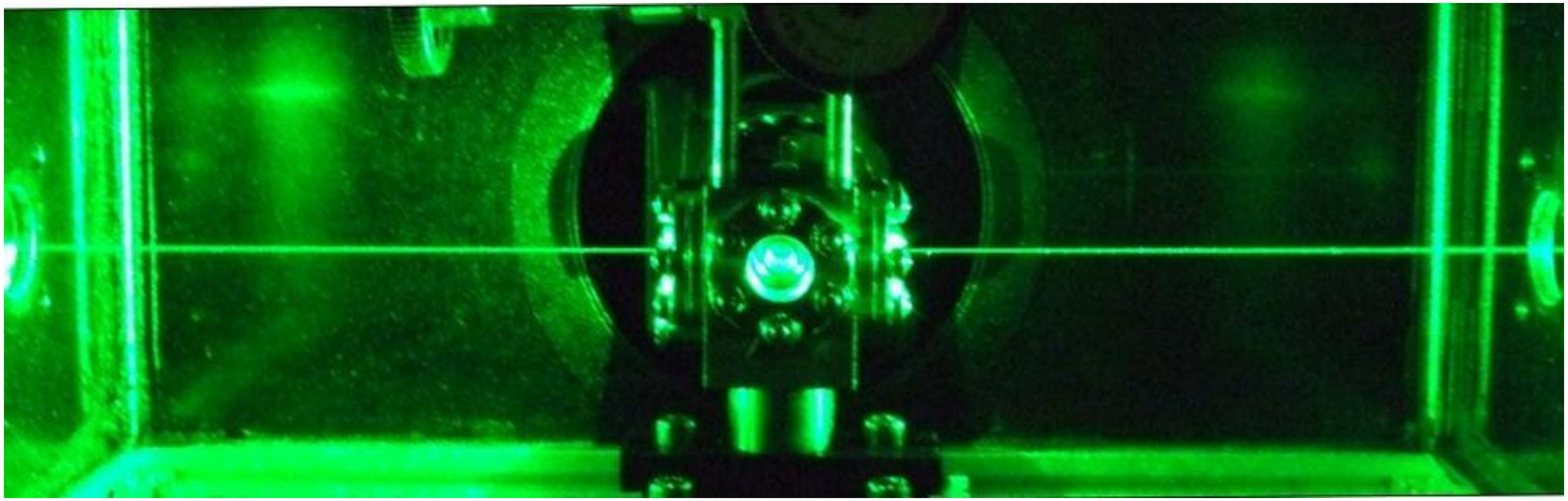
Magnetron sputtering = MS

Electron beam deposition = EBD

First test of coatings in December 2013

Conclusion

- Control and monitoring of WGTS parameters on 0.1% scale is essential and well on track
- Monitoring of gas composition by Raman spectroscopy (LARA)
- LARA performance demonstrated (0.1% precision and < 6% accuracy, robust data analysis)
- Coating issue understood, solution on the way



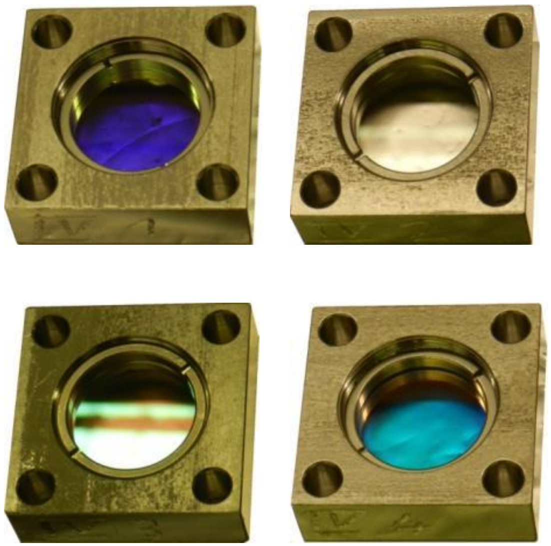
The LARA group

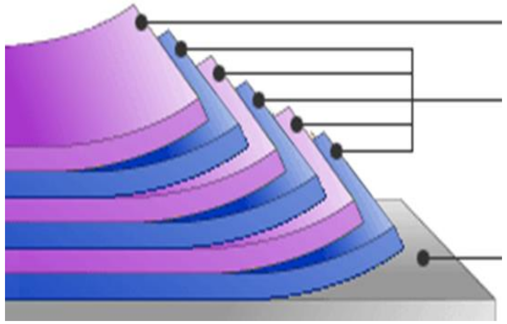
Karlsruhe Institute of Technology

Coating manufacturing methods

- Electron beam depositing (EBM)
- Ion assisted beam depositing (IAM)
- Magnetron sputtering (MS)
- Ion beam sputtering (IBS)

Decreasing porosity

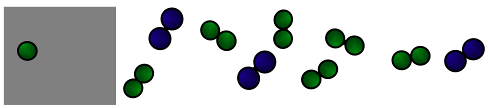




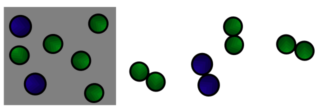
Intensity variations

- Isotope exchange reactions in gas
- Gas - wall interactions

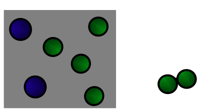
Stainless steel vessel wall with (H_2, D_2)



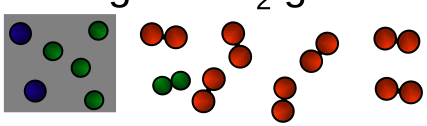
Permeation into stainless steel



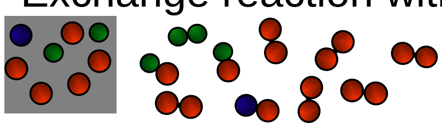
Evacuation of vessel

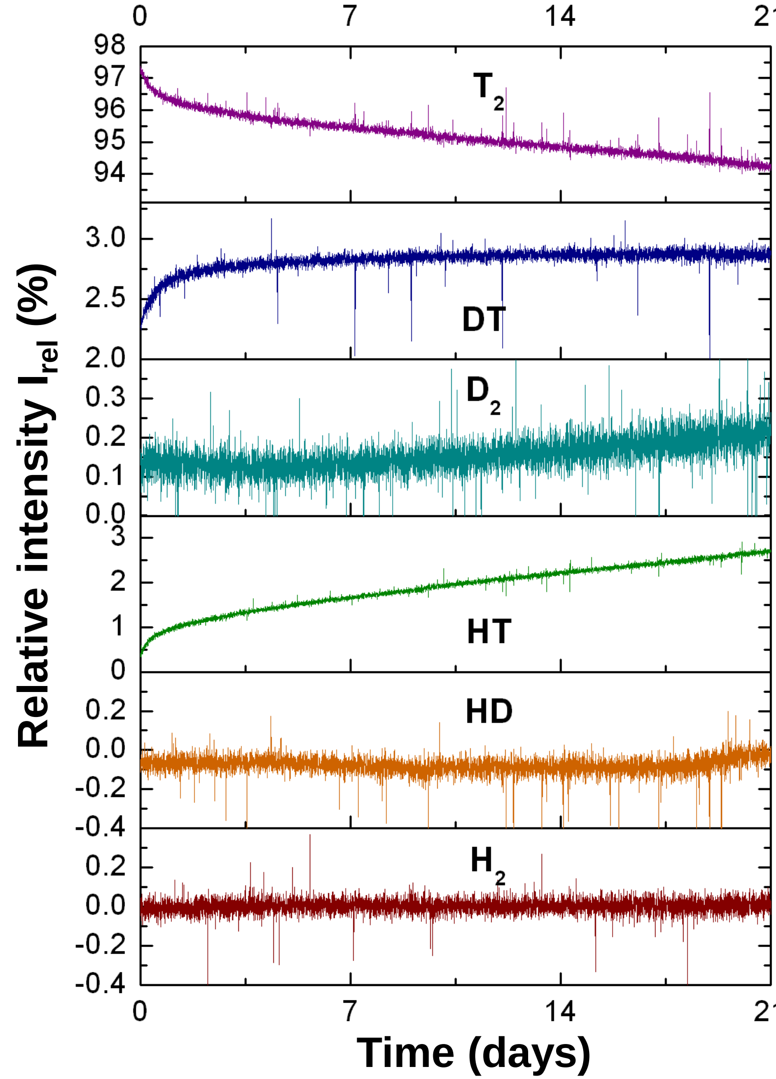


Filling with T_2 gas



Exchange reaction with T_2 of next filling

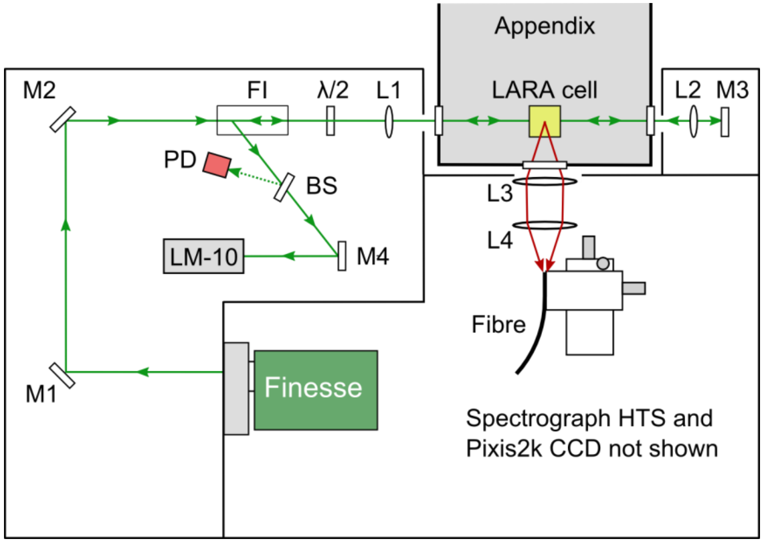




Beam path simplification, system monitoring

- Installation of faraday isolator
  → Simplification of beam alignment
- Training of personnel for maintenance and repair

ongoing



- Definition of control procedures and hardware status parameters