Monitoring of the KATRIN source composition by Raman spectroscopy

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The KATRIN experiment

Source section

Transport section

Spectrometers and detector

$t_{1/2} = 12.3 \text{ years}$

Determination of neutrino mass with 200 meV/c$^2$ sensitivity (90 % C.L.)
Windowless gaseous tritium source (WGTS)

- Continuous gas injection and removal
- Steady-state gas column inside source tube
Windowless gaseous tritium source (WGTS)

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

> 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 $m_\nu$ 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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Gas composition inside WGTS

3 hydrogen isotopes
- Tritium
- Deuterium
- Hydrogen

6 hydrogen isotopologues
- $T_2$
- DT
- HT
- $D_2$
- HD
- $H_2$
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$\beta$-spectrum depends on gas composition

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> 91 % $T_2$
< 9 % DT
< 1 % $D_2$

Tritium purity $\varepsilon_T > 95 %$
Molecular effects on $\beta$ spectrum

- Doppler broadening

- Electron scattering with molecules

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

- Final state distribution

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

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

\[ E_{\text{rec}} \approx E_0 \cdot \frac{m_e}{m_{\text{mol}}} \]
Molecular effects on $\beta$ spectrum

- Doppler broadening
- Electron scattering with molecules
- Nuclear recoil of daughter molecules (e.g. $^3$HeT$^+$)
- Final state distribution

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
Experimental setup

Laser Raman (LARA) cell

DPSS Nd:YAG laser (532nm, ≤ 5W)
Experimental setup

Laser Raman (LARA) cell

Optical fibre

DPSS Nd:YAG laser (532nm, ≤ 5W)

CCD

Spectrograph
Proof of principle

- Static samples with low tritium activity

All hydrogen isotopologues can be detected simultaneously
LARA setup, tritium loops and the appendix

Glove box with tritium loops

Appendix: Connection between LARA and sample cell inside glovebox

LARA setup
LARA setup, tritium loops and the appendix

Glove box with tritium loops

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LARA setup

Commissioned in 2009

Appendix

Cell
Long-term monitoring inside a test loop

- Non-stop monitoring for > 21 days

\[ p_{\text{total}} = 200 \pm 0.3 \text{ mbar} \]
Laser power: 5 W
Acquisition time: 250 s
Tritium purity > 95 %
Long-term monitoring inside a test loop

- Non-stop monitoring for > 21 days

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Laser power: 5 W
Acquisition time: 250 s
Tritium purity > 95 %

S. F. et al, Fusion Sci Technol. 60 3, 925-930 (2011)
Non-stop monitoring for > 21 days

\[ \rho_{\text{total}} = 200 \pm 0.3 \text{ mbar} \]
Laser power: 5 W
Acquisition time: 250 s
Tritium purity > 95%

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 $^{17}\text{O}$, $^{18}\text{O}$, $^{15}\text{N}$
  - Application in calibration of LARA system
Calibration

- Measured spectrum
  - Response functions
    - Method 1
    - Method 2
  - Composition: $T_2$, DT, D$_2$

Results from calibration:

- $0.9 \times 1.1 \times 1.4$

- Theoretical intensities
- Spectral sensitivity

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Calibration

- Measured spectrum
- Response functions
- Composition

Results from calibration:

\[ x \times 0.9 \times 1.1 \]

Only possible for H\(_2\), HD, D\(_2\)

Calibration uncertainty < 2%

Calibration

Only possible for H₂, HD, D₂
Calibration uncertainty < 2%


Applicable to all isotopologues
Validation of molecular constants within 3%

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

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

Only possible for H₂, HD, D₂
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Applicable to all isotopologues
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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)
(Potential) reasons for coating damage

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  Discoloration of PTFE sealing → Indication for TF formation
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

AR532nm/0°

AR450-700nm/0°
(Probably) solving the problem

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

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
Coating manufacturing methods

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

Decreasing porosity

Top Coating (SiO$_2$)
Alternating metal oxides and SiO$_2$
Cell window
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

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Time (days)

Relative intensity \(I_{rel}\) (%)

- \(T_2\)
- \(DT\)
- \(D_2\)
- \(HT\)
- \(HD\)
- \(H_2\)

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Beam path simplification, system monitoring

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

Definition of control procedures and hardware status parameters

ongoing