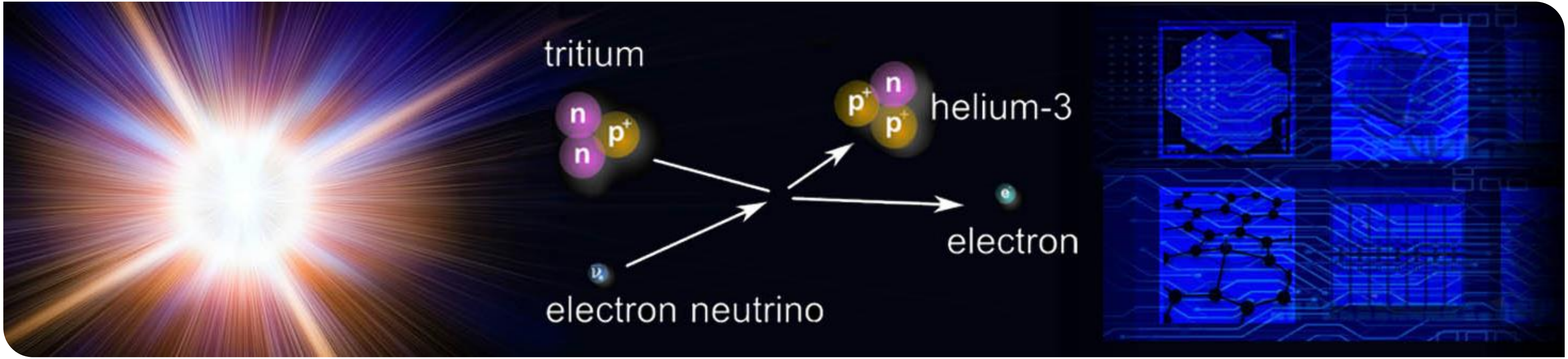
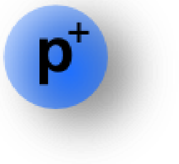
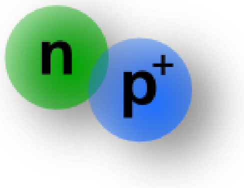
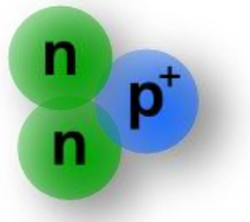
Fundamental physics with the heaviest isotope of hydrogen

Magnus Schlösser, Tritium Laboratory Karlsruhe



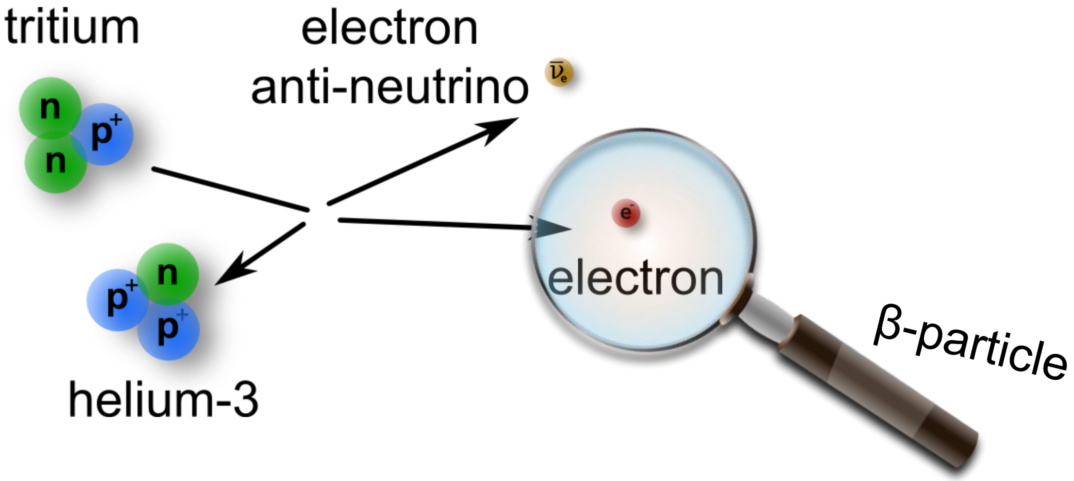
Three isotopes of hydrogen

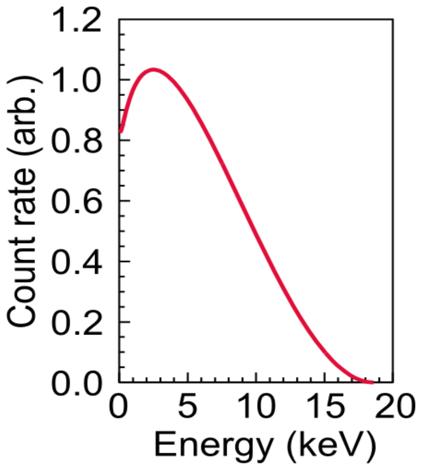
Hydrogen (H) (Protium)	Deuterium (D)	Tritium (T)
		
stable	**stable**	**unstable (radioactive)**
$m = 1$ $s = \frac{1}{2}$	$m = 2$ $s = 1$	$m = 3$ $s = \frac{1}{2}$

Chemistry similar,

But different mass

Tritium is radioactive



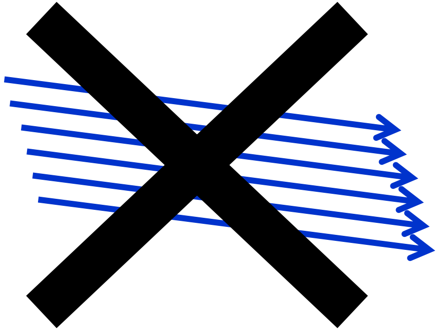


- Half live: 12.3 year
- Max. decay energy: 18.6 keV

1 g tritium:
≈ 3.6 × 10^{14} Bq ≈ 10 000 Ci

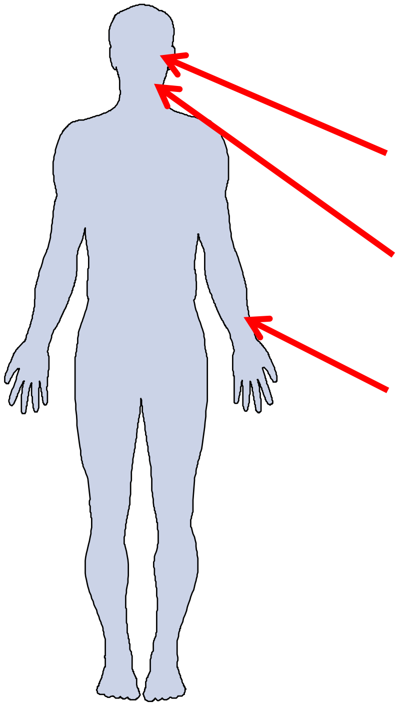
Danger of the radioactivity

External exposition



Max. decay energy: 18,6 keV

penetration in air ~ 6 mm
penetration in metal < 0.1 mm



Internal exposition

Inhalation

Ingestion

Incorporation via skin or wounds

$$H_2O + T_2 \rightarrow HTO + HT$$

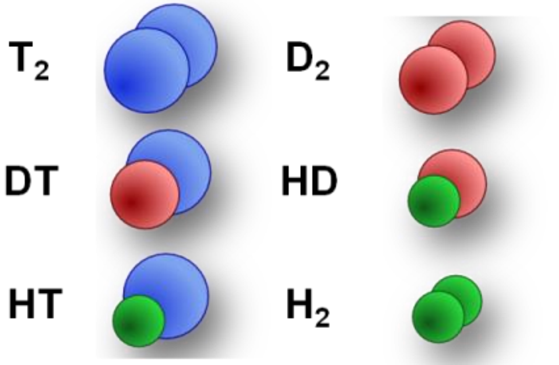
Lethal Dose:
T_2 ~ 20 l
T_2O ~ 2µl

Don‘t let tritium enter your body

Some tritium properties

3 main forms (Q means H, D, or T)

- Molecular (Q_2): gaseous, 6 isotopologues
- Oxidised (Q_2O): water, 6 isotopologues
- Bound in molecules (CQ_4, R-Q…)



High reactivity (like hydrogen)

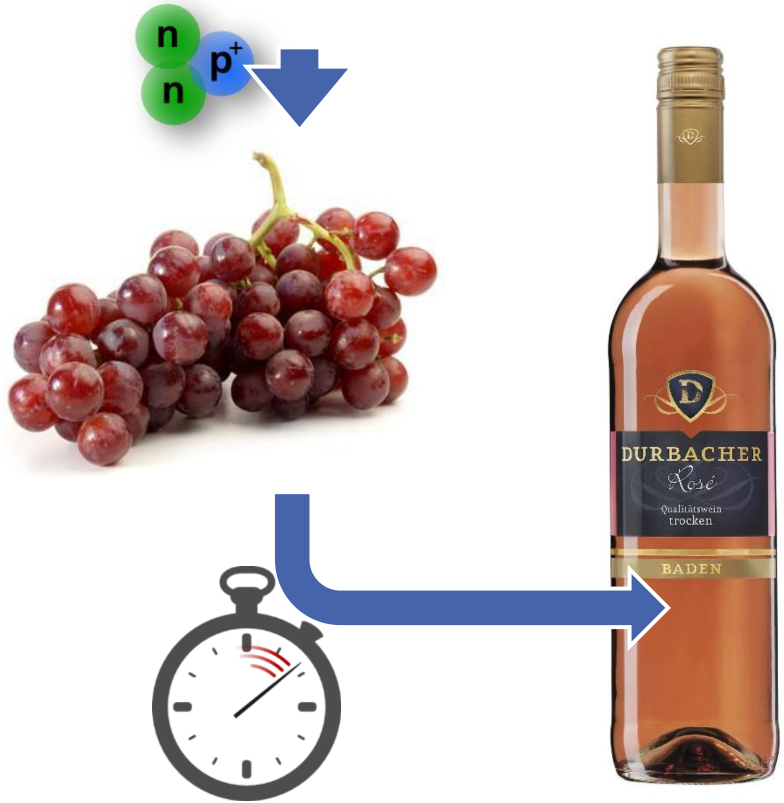
- Chemically very reactive (giving / taking e^-)
- Strong interaction with metals (solution, diffusion, permeation…)

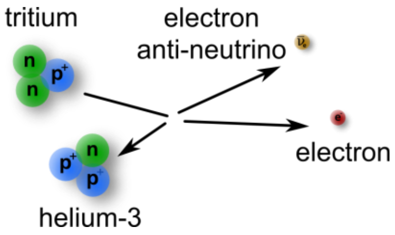
Thermodynamically equilibrated

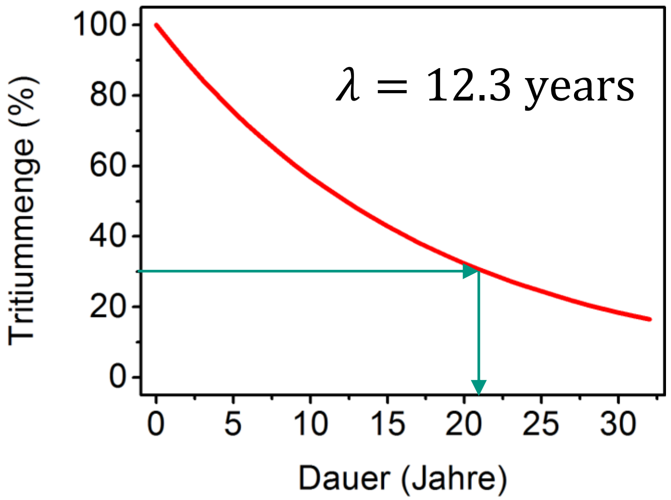
$H_2 + T_2 \Leftrightarrow 2\,HT$

$D_2 + T_2 \Leftrightarrow 2\,DT$

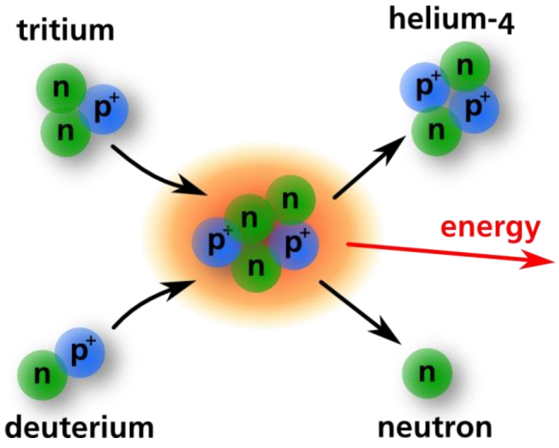
Radiometric dating of wine





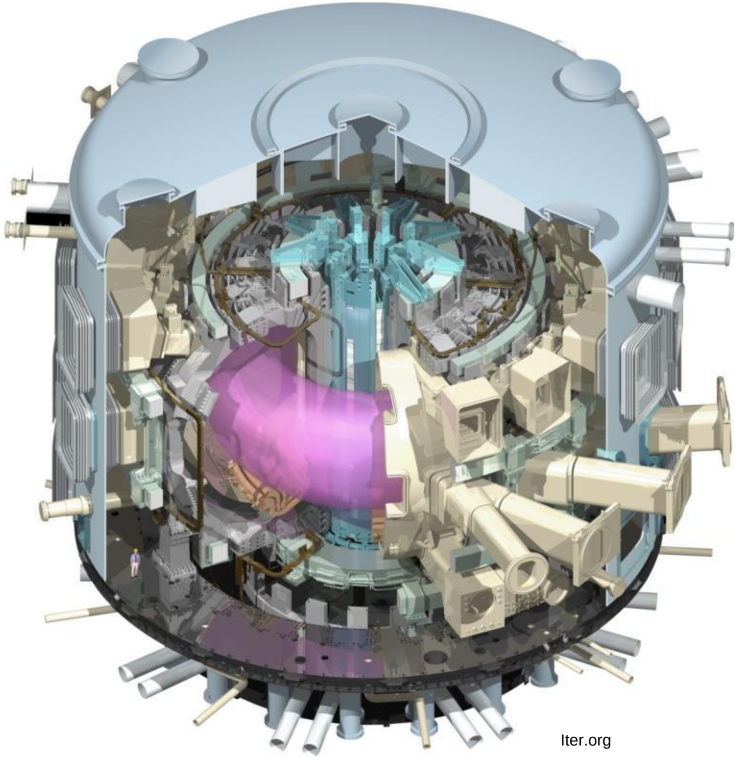


Fusion power using tritium



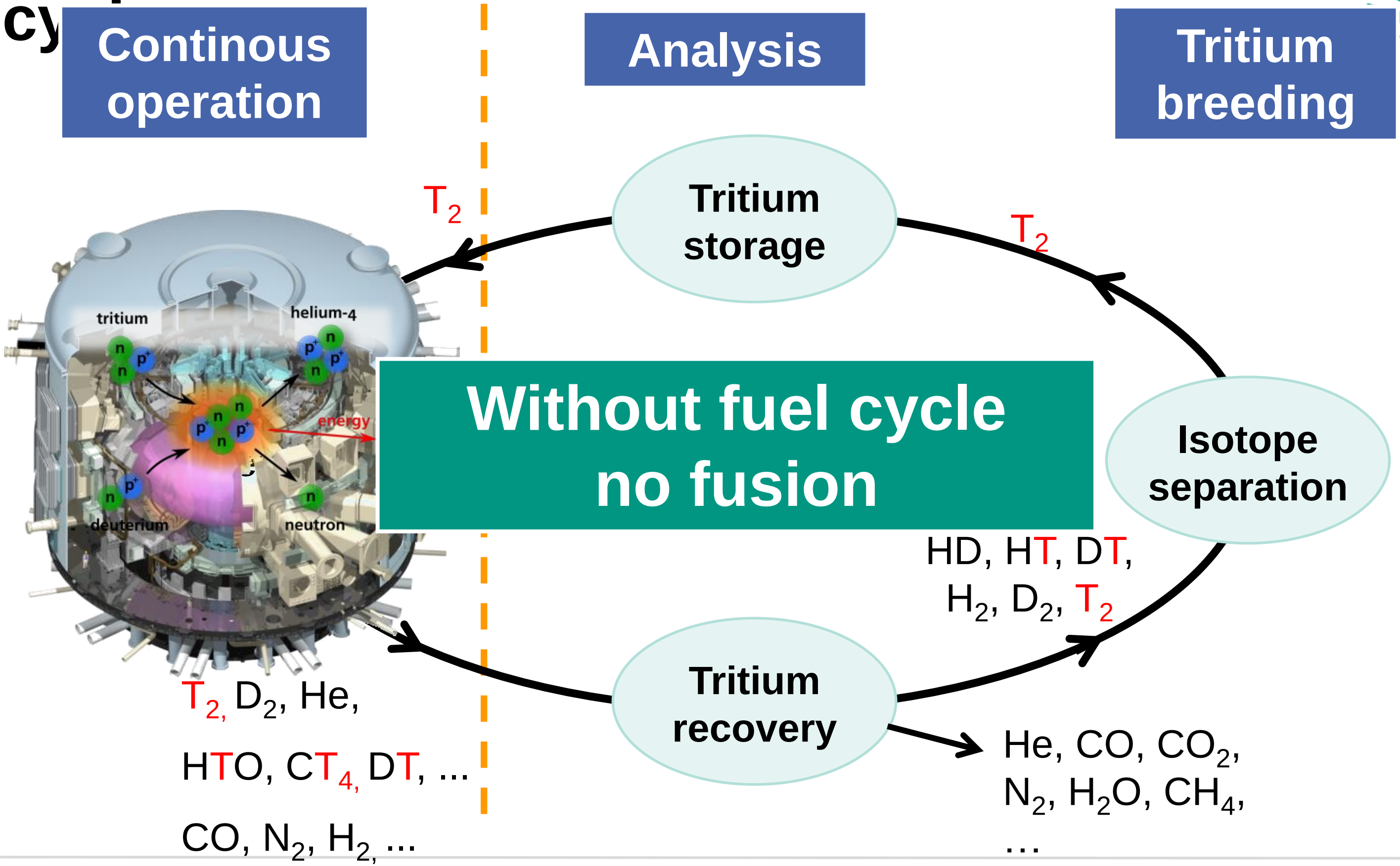
T ~ 100 Mio. °C

Fusion reactor ITER



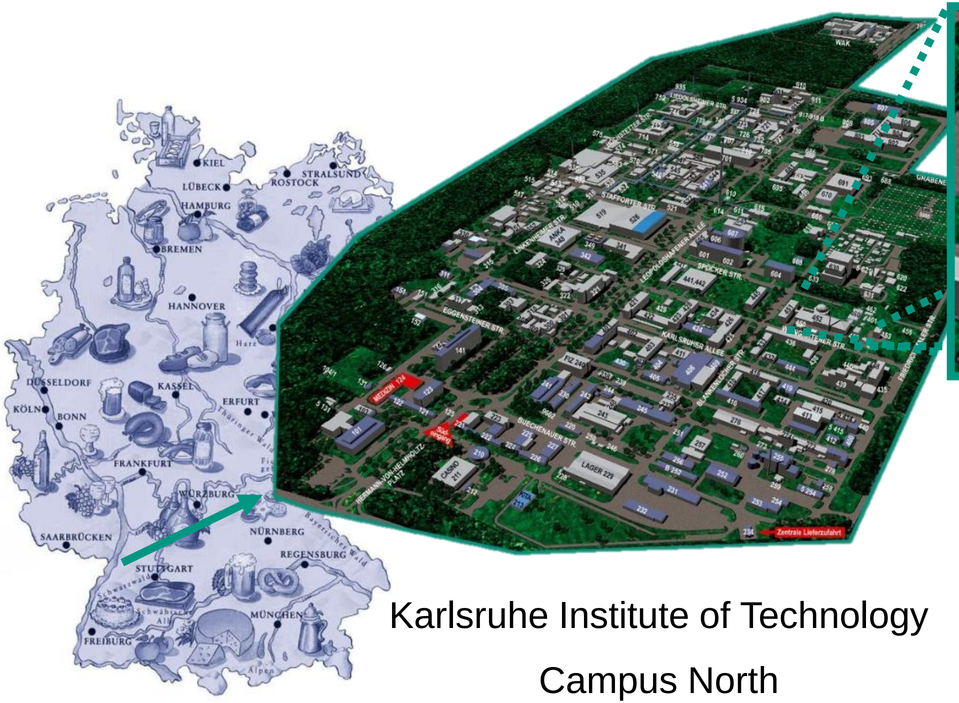
Iter.org

Fuel cy



Continous operation
Analysis
Tritium breeding
Without fuel cycle
no fusion
T_2
Tritium storage
T_2
Isotope separation
HD, HT, DT,
H_2, D_2, T_2
Tritium recovery
He, CO, CO_2,
N_2, H_2O, CH_4,
…
tritium
helium-4
energy
deuterium
neutron
T_2, D_2, He,
HTO, CT_4, DT, ...
CO, N_2, H_2, ...

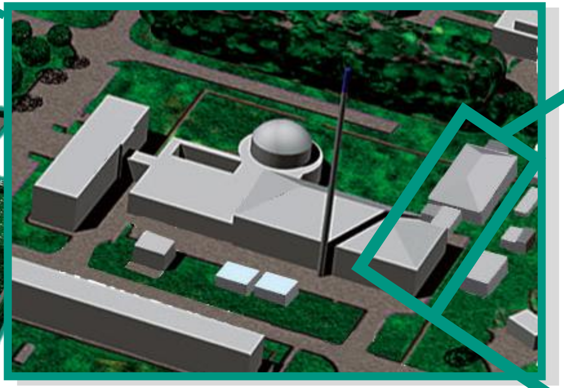
The Tritium Laboratory Karlsruhe

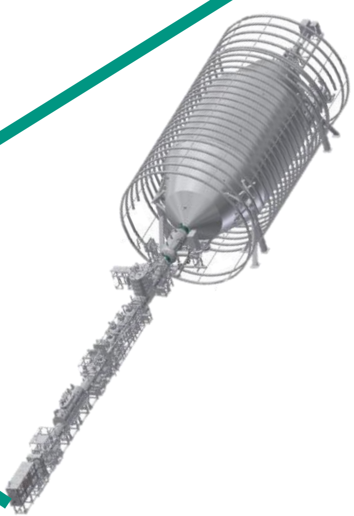


Karlsruhe Institute of Technology

Campus North

Tritium Laboratory Karlsruhe (TLK)

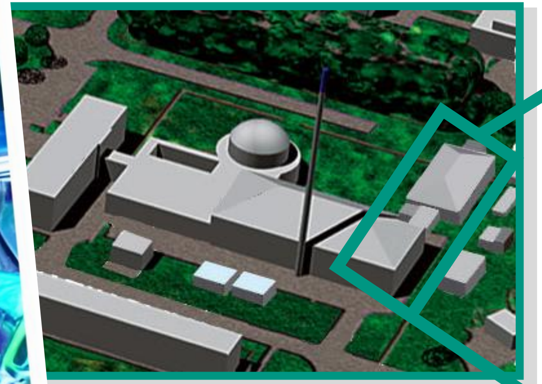




- Commissioning 1993
- Licensed for 40 g Tritium
- Two missions:
  - Fuel cycle for fusion reactors
  - KATRIN Experiment

The T ruhe

um Laboratory Karlsruhe (TLK)



Commissioning 1993

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Two missions:

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The Tritium Laboratory Karlsruhe

- Unique research facility
  - able to setup and operate a large variety of experiments with tritium (Astroparticle physics and fusion)
  - Closed tritium loop for recycling and purifying of tritium
  - License for 40 g of tritium ($\approx 1.5 \cdot 10^{16}$ Bq)

- 30 years of tritium experience
- Currently ~50 people “on board” including 6 doctoral researchers and many students
- 21 persons necessary for TLK baseline operation

Ideal facility for high-activity tritium experiments

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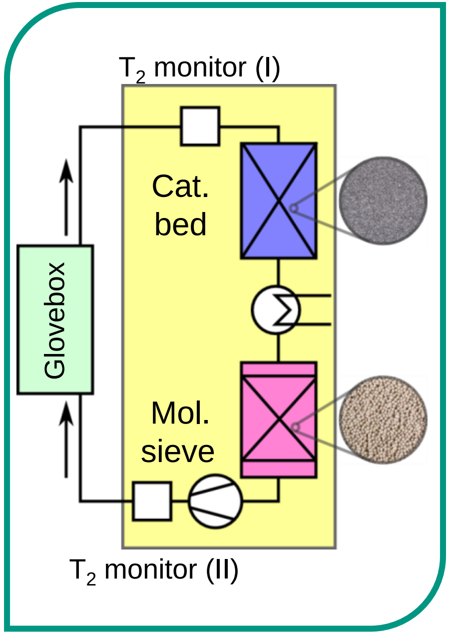
Ideal facility for high-activity tritium experiments

Safe tritium handling

Safety precautions to prevent harm to humans and environment

Enclosure concept

Fully metal components

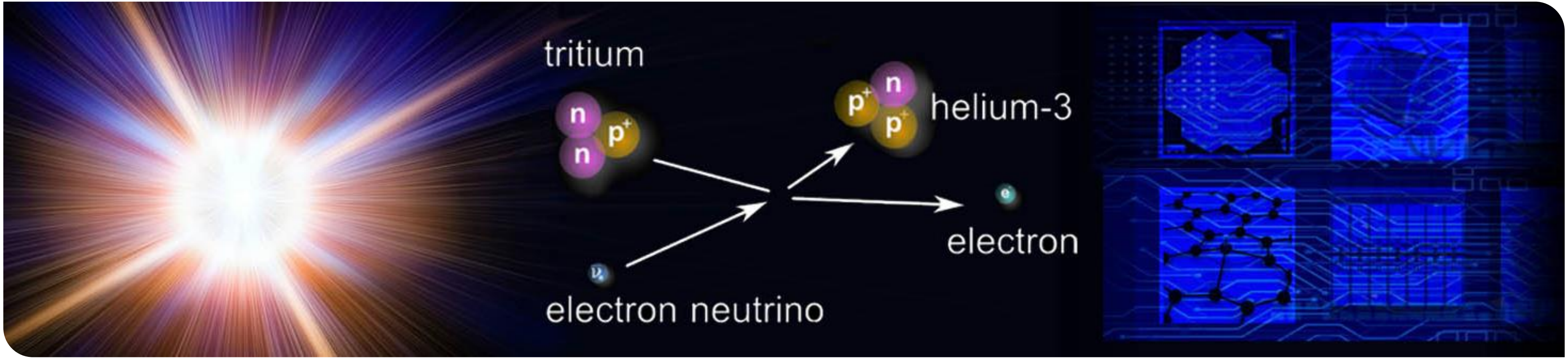


Tritium retention system

Fundamental physics with the heaviest isotope of hydrogen

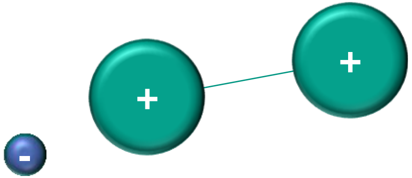
Part 1: Test of bound-state QED

Magnus Schlösser, Tritium Laboratory Karlsruhe



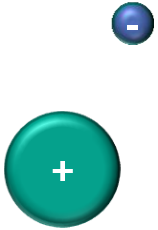
Simple systems of quantum electrodynamics

Free electron

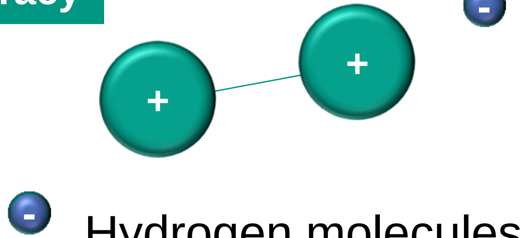


Hydrogen molecular cation
(H_2^+, HD^+, D_2^+, HT^+, DT^+, T_2^+)

Rather simple systems
Can be calculated to very high accuracy



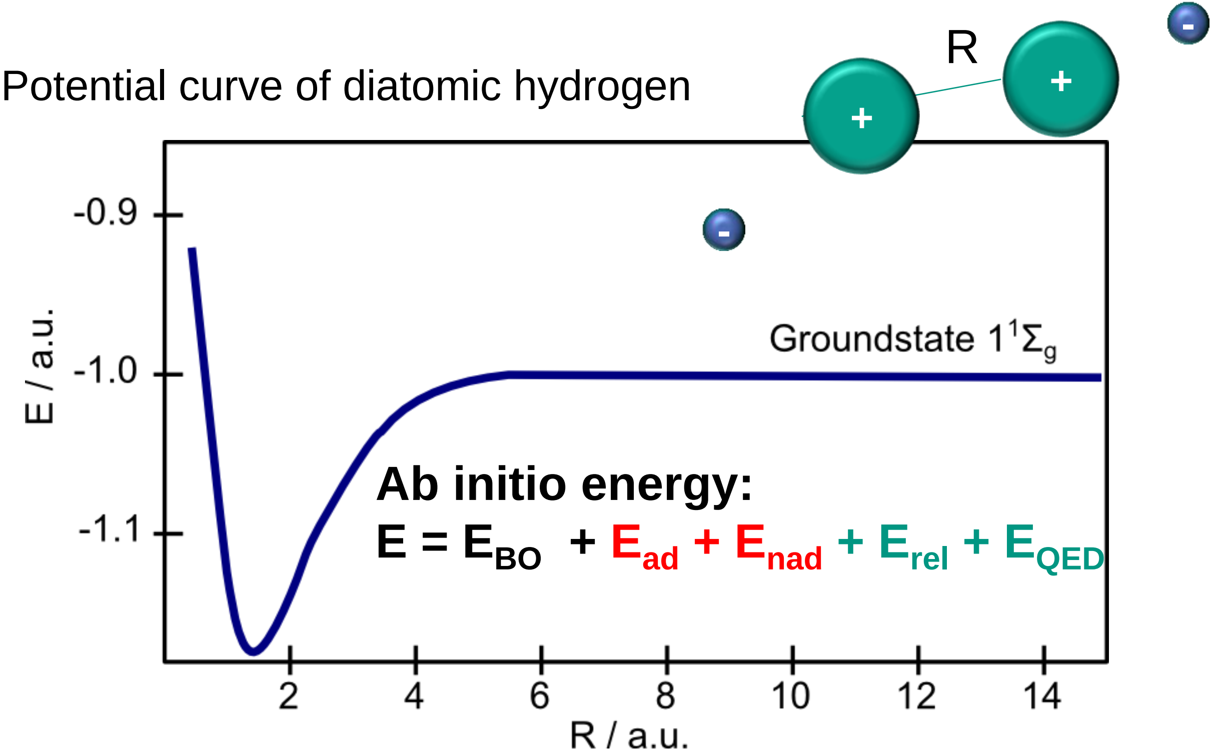
Hydrogen atom (H, D, T)



Hydrogen molecules
(H_2, HD, D_2, HT, DT, T_2)

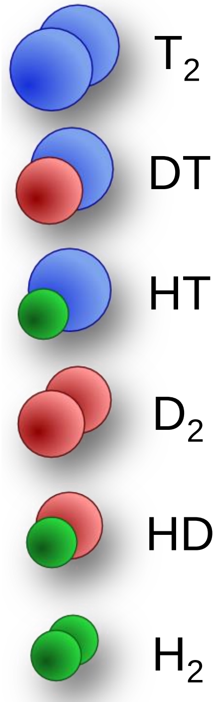
What does „calculation“ mean?

- Potential curve of diatomic hydrogen



Ab initio energy:

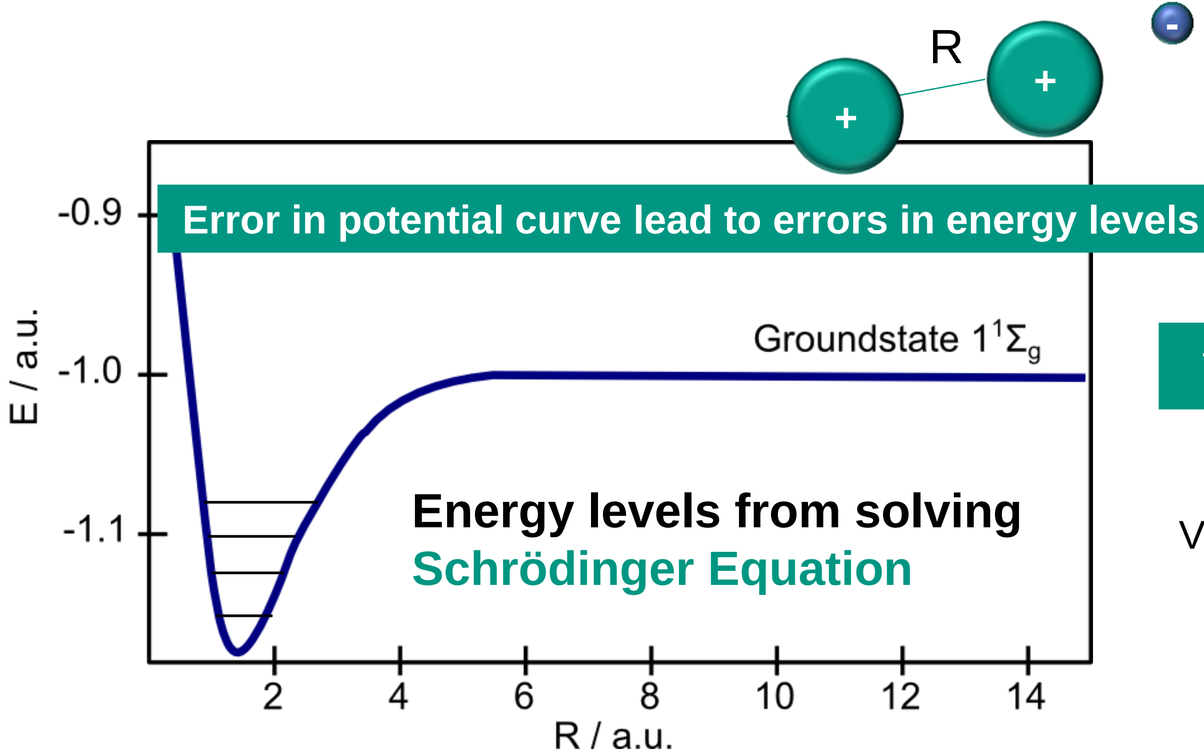
$$E = E_{BO} + E_{ad} + E_{nad} + E_{rel} + E_{QED}$$



Correction terms have different mass scaling

”Calculation” means determining the potential curve accurately

Energy levels



Error in potential curve lead to errors in energy levels

Energy levels from solving
Schrödinger Equation

Theory → ← Experiment

Validate fundamental theory

Effects by new physics

e.g. Hollik, Linster, Tabet
EPJ. C 80, 661 (2020)

Measuring energy levels → probing the potential curve

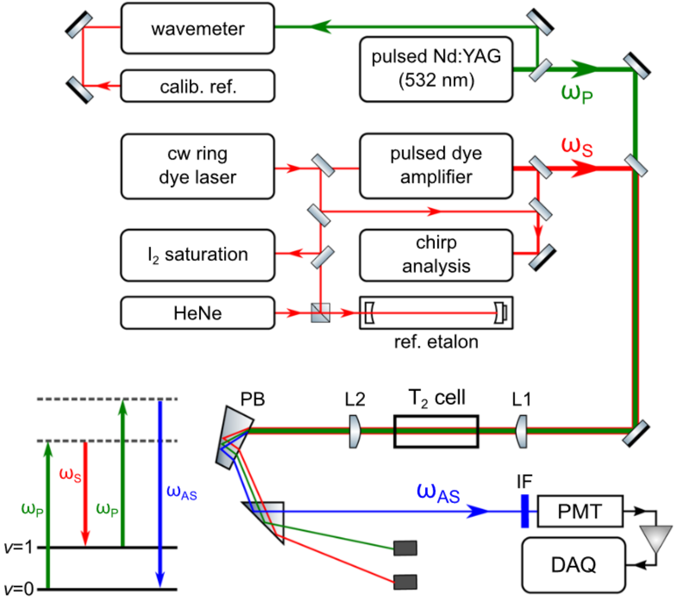
Experimental tests

Until recently: H_2, HD, D_2

Now additionally: T_2, DT, HT

Bring tritium cell with <1 GBq (exemption limit)

Example:
Coherent-Anti-Stokes **Raman**-Spectroscopy

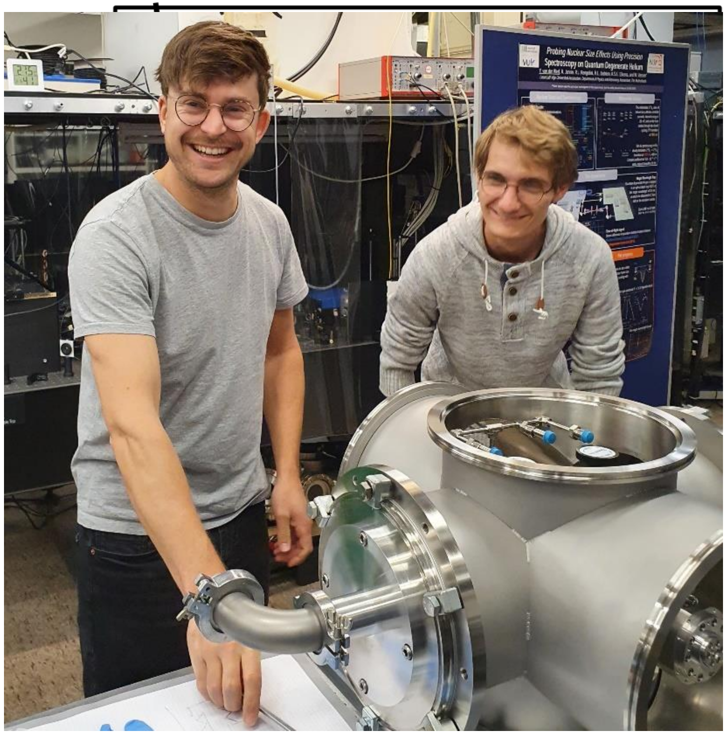


Trivikram, Schlösser, Ubachs, Salumbides, PRL 120 163002 (2018)

Signal if ΔE of two photons ≡ ΔE of two molecular levels

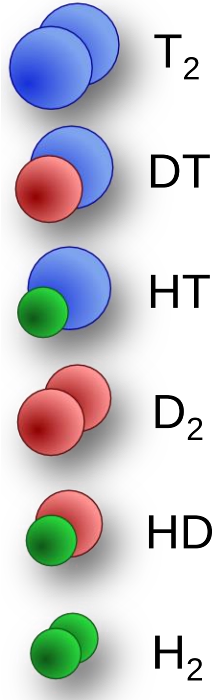
Benchmarking QED with hydrogen molecular spectroscopy

Karlsruhe Institute of Technology



2 4 6 8 10 12 14
R / a.u.

Hydrogen isotopologues



T_2: 3000 MHz (Veirs 1987)

10 MHz TLK/VU PRL 120, 16002 (2018)

DT: 3000 MHz (Veirs 1987)

10 MHz TLK/VU PRR 1, 033124 (2019)

HT: 300 MHz (Zare 1987)

10 MHz TLK/VU PCCP 22, 8973 (2020)

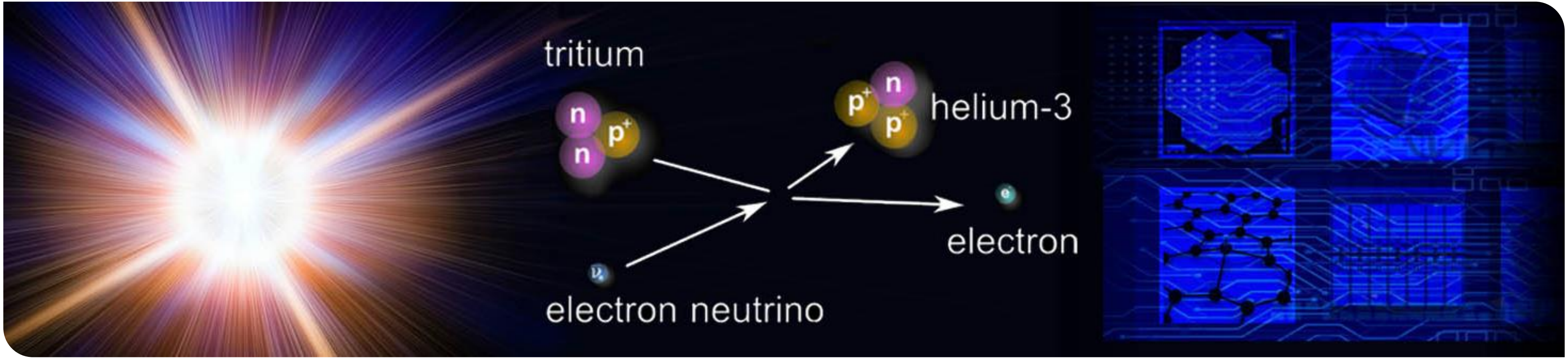
HD: 20 kHz: NICE-OHMS technique
TLK/VU 2020 PRA 105, 6 (2022)

April 23: 10 kHz for HT with NICE-OHMS technique

Fundamental physics with the heaviest isotope of hydrogen

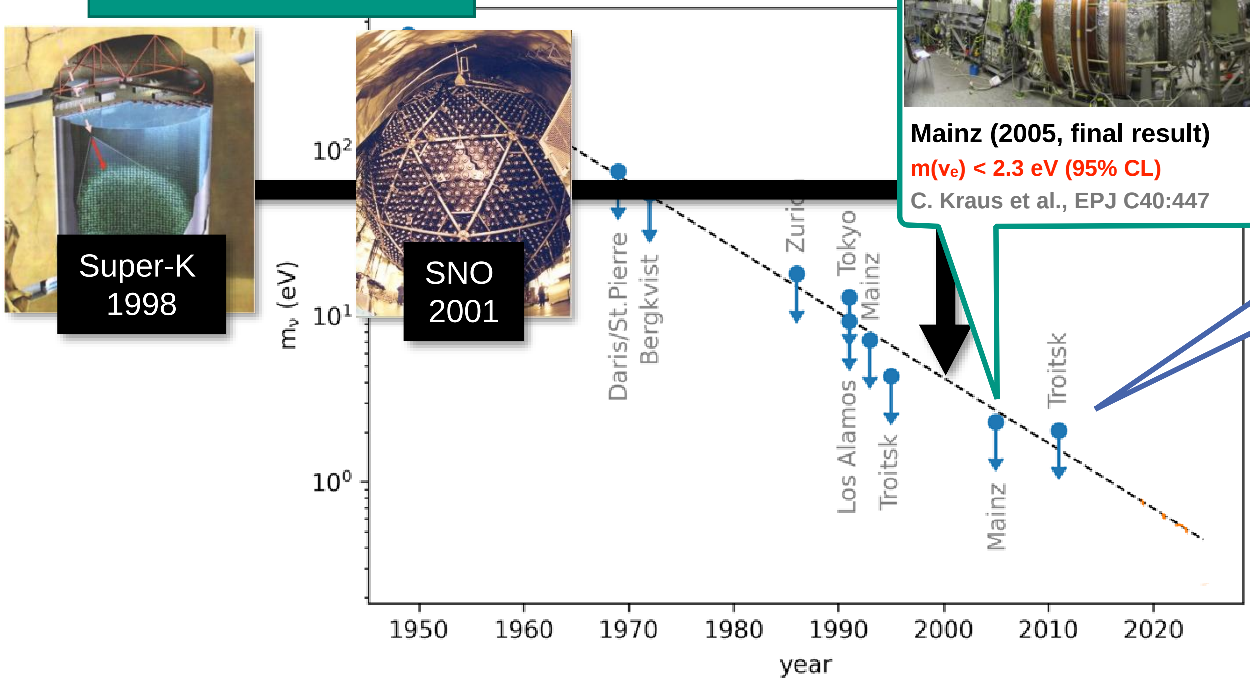
Magnus Schlösser, Tritium Laboratory Karlsruhe

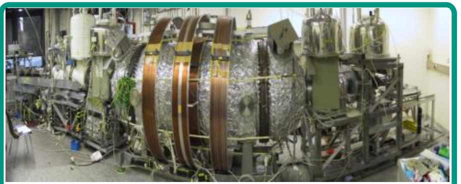
Part 2: Measurement of neutrino mass



Moore's Law of direct neutrino mass searches

Neutrino oscillation
→ neutrinos have mass!





Mainz (2005, final result)
$m(\nu_e)$ < 2.3 eV (95% CL)
C. Kraus et al., EPJ C40:447

Troitsk (2011, re-analysis)
$m(\nu_e)$ < 2.05 eV (95% CL)
V. N. Aseev et al., PRD 84:112003

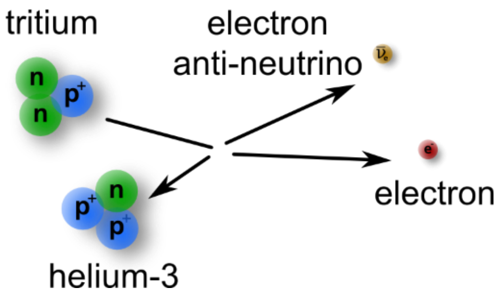
previous limits (2 eV)
ν ruled out as DM

upcoming (KATRIN: 200 meV)
degeneracy scale

future approaches (40 meV)
hierarchy scale

Tritium beta decay experiments

- Direct, model-independent access to neutrino mass

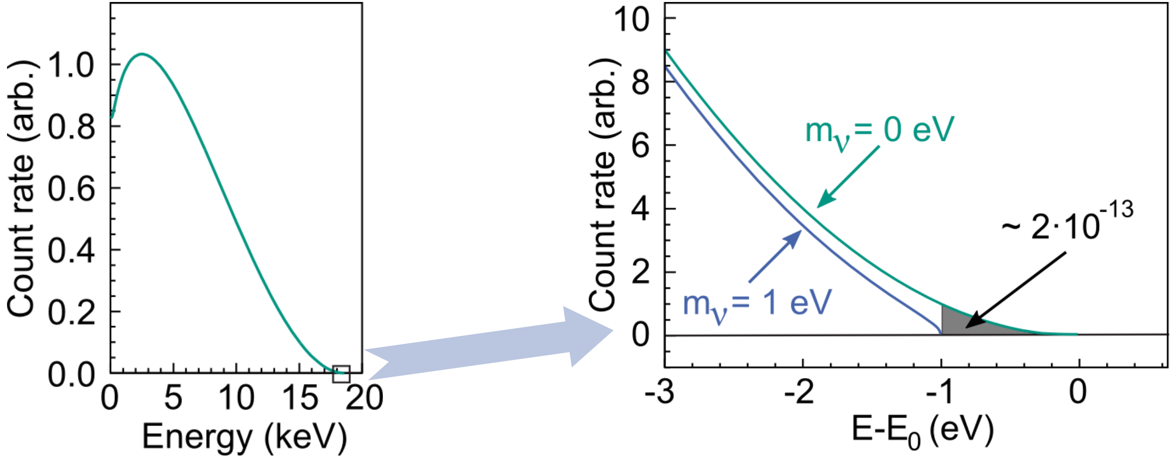


$$m_\beta^2 = \sum_i |U_{ei}|^2 m_i^2$$

$$\frac{dN}{dE} \propto \sqrt{(E_0 - E)^2 - m_{\bar{\nu}_i}^2 c^4}$$

E_0 = 18.6 keV

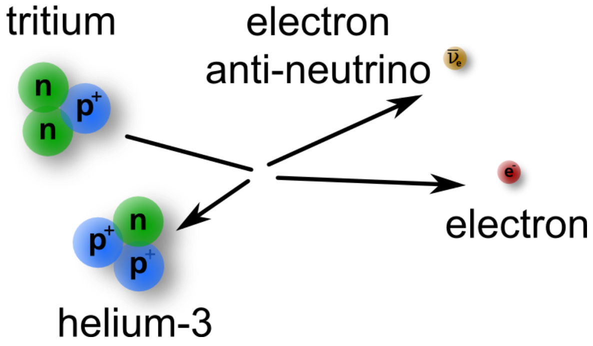
$T_{1/2}$ = 12.3 y



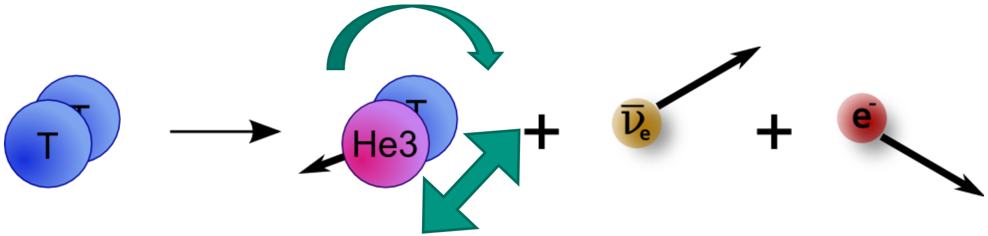
KATRIN's aim: Measurement of m_ν with a sensitivity of 0.2 eV/c²

Molecular decay

Atomic decay

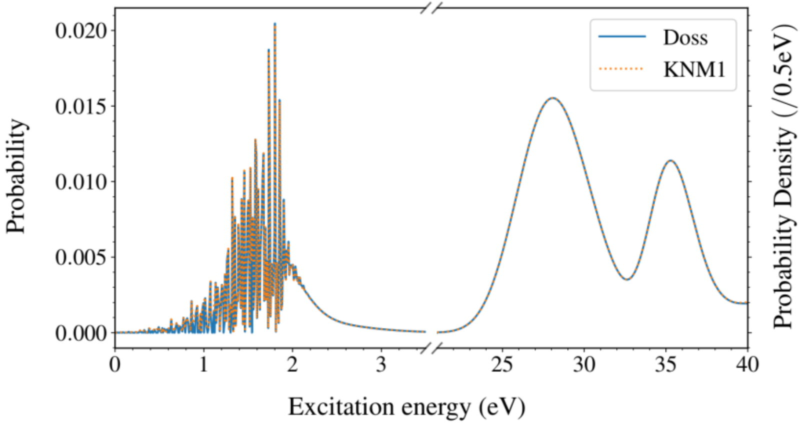


Decay from a molecule



+ further inner excitations (rotation / vibration)

Final-state distribution

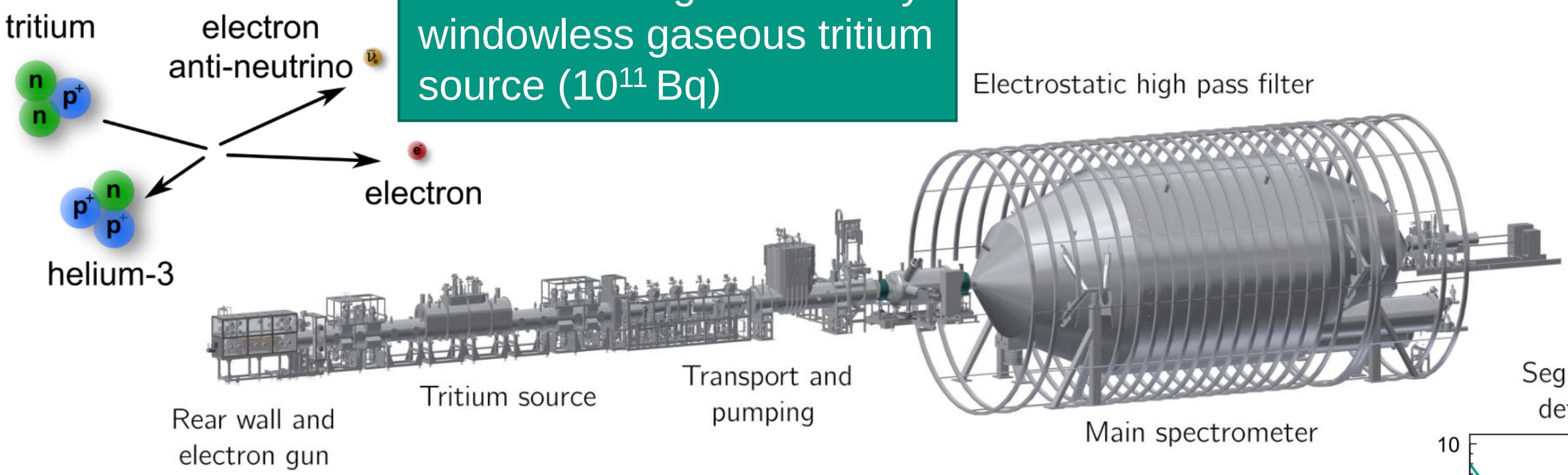


Molecular effects need to be taken into account in neutrino mass analysis

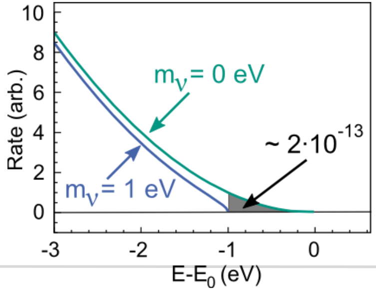
„model-dependence“

Karlsruhe Tritium Neutrino Experiment (KATRIN)

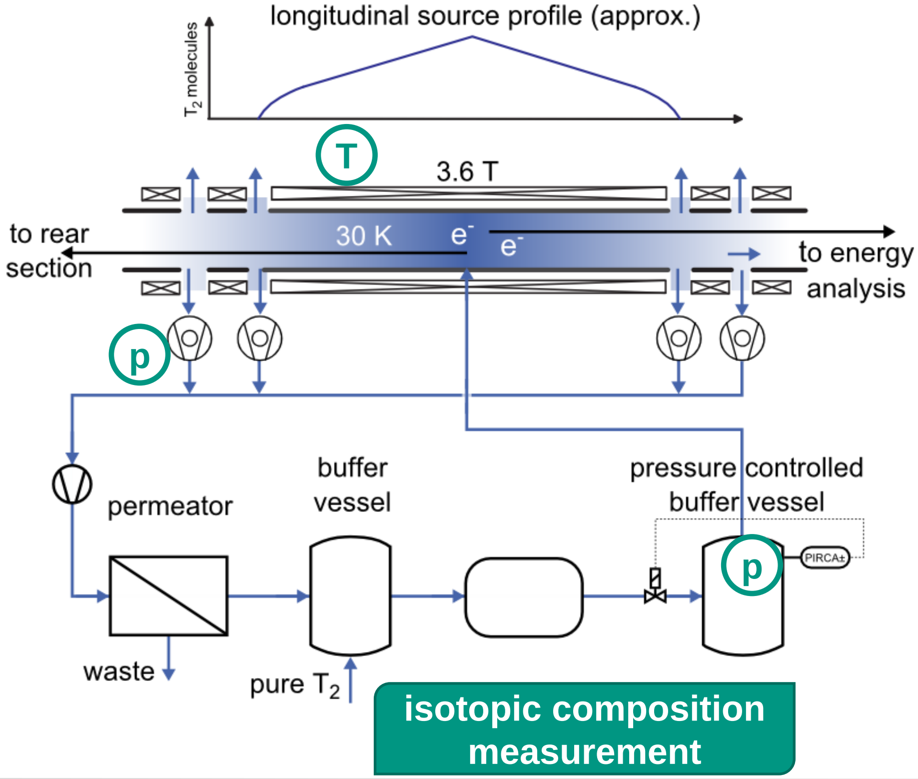
ultra-stable high-luminosity windowless gaseous tritium source (10^{11} Bq)



high-resolution MAC-E filter with < 1 eV energy resolution

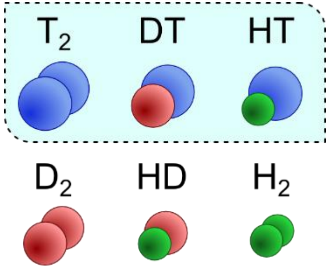


The stable tritium source



- T_2 purity > 95%
- Source activity 10^{11} Bq
- Source profile stable to 10^{-3} level

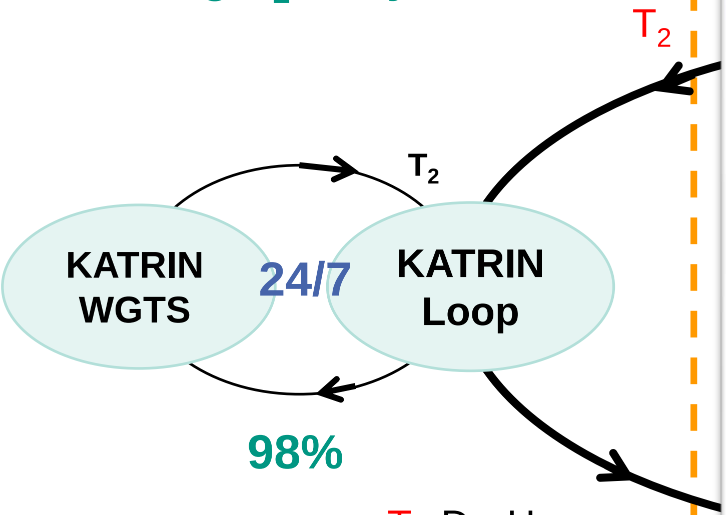
Hydrogen isotopologues



- T_2 throughput ~ 40 g/day
- Operation 24/7, 60 days/run
- Necessary inventory >15 g

Tritium supply

40 g T_2 / day



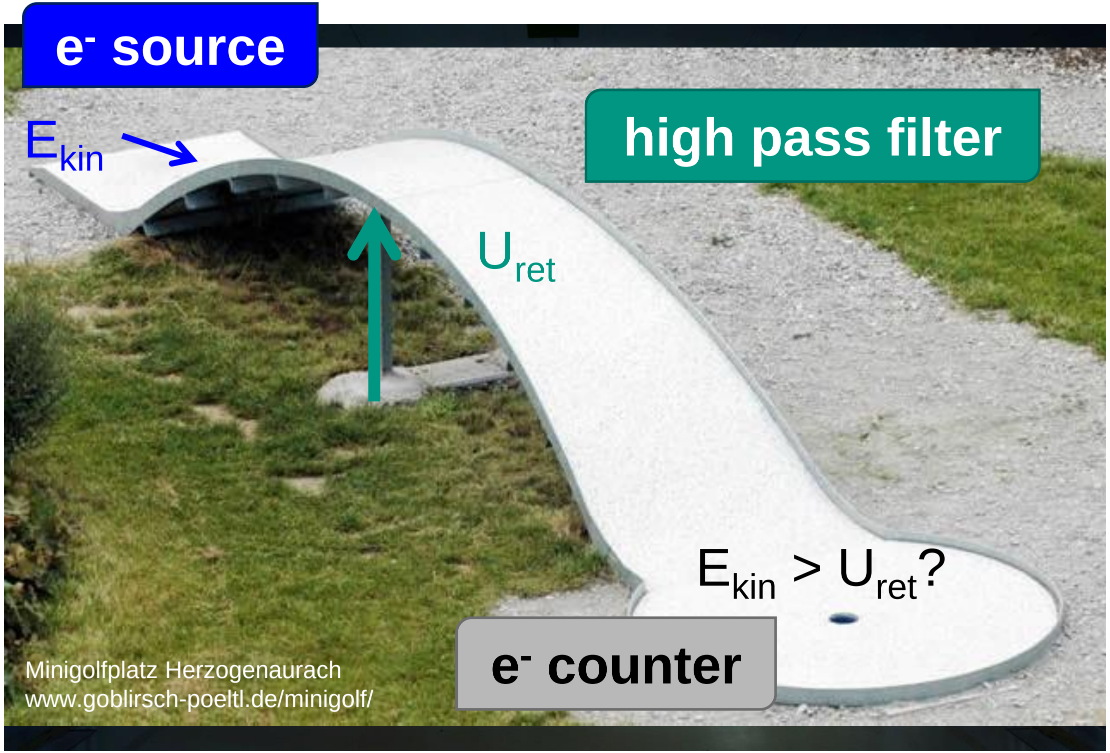
T_2, D_2, He,

H**T**O, CT_4, D**T**, ...

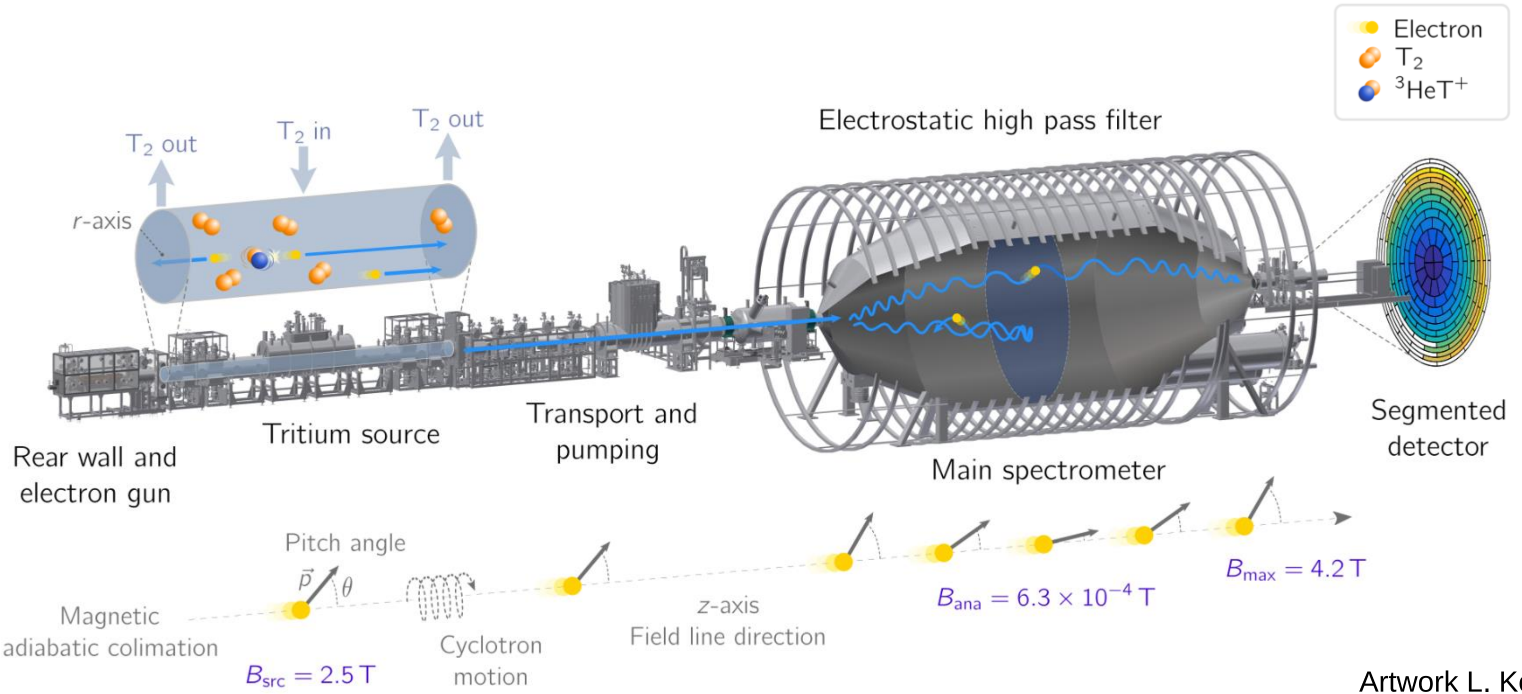
CO, N_2, H_2, ...

...

Measurement principle analogy



Working principle of KATRIN

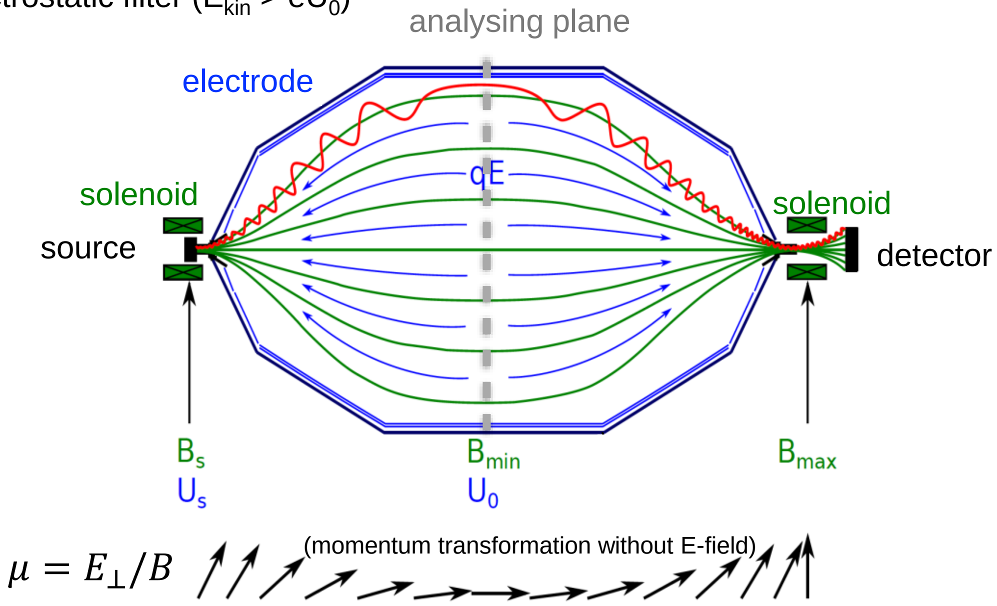


Artwork L. Köllenberger

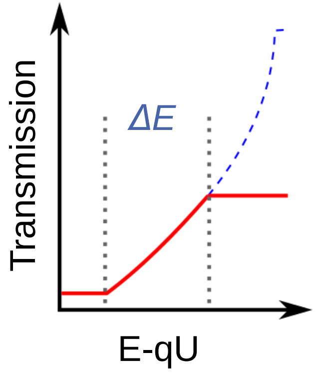
High-resolution spectrometer: MAC-E filter

Magnetic **A**diabatic **C**ollimation & **E**lectrostatic **Filter**

Integrating electrostatic filter ($E_{kin} > eU_0$)



Sharp high pass filter:

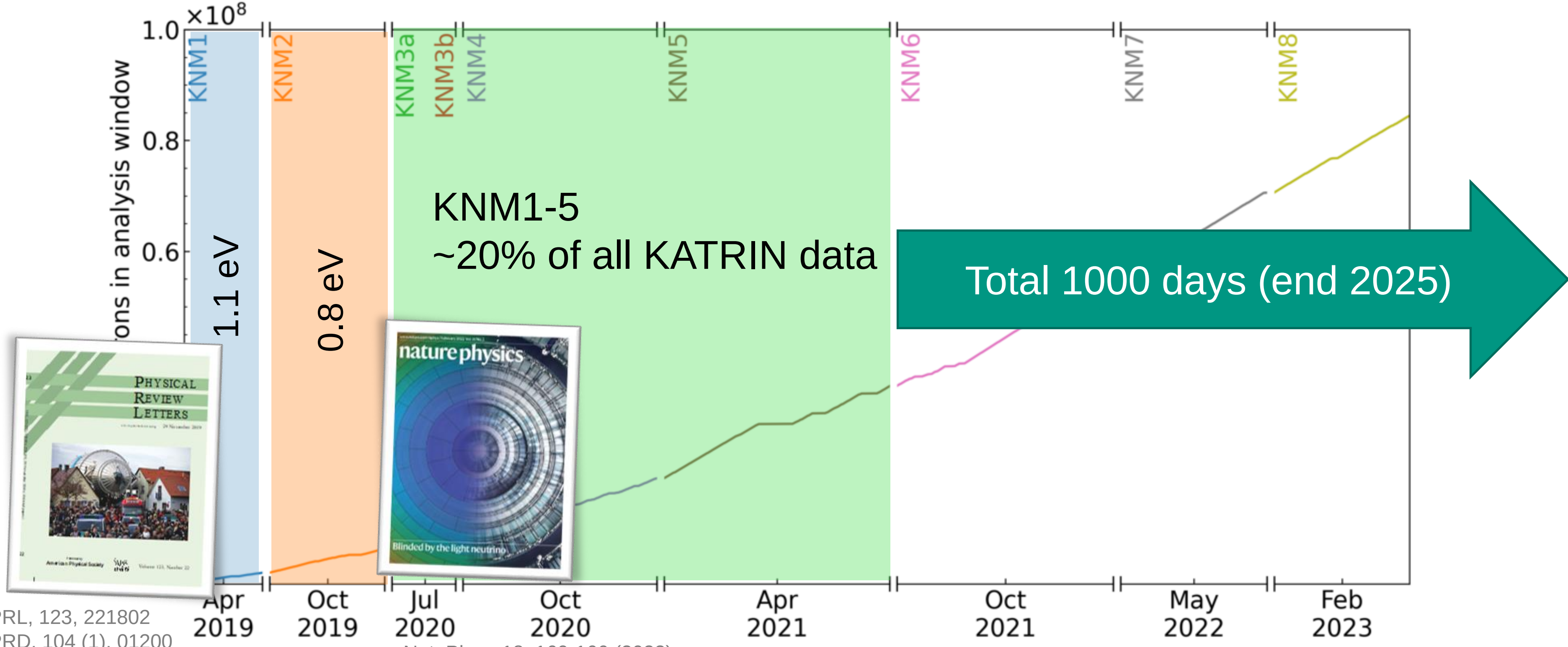


$$\frac{\Delta E}{E} = \frac{B_{min}}{B_{max}}$$

→ **ΔE < 1 eV** at 18.6 keV

e.g. Kleesiek et al., EPJ. C 79, 204 (2019)

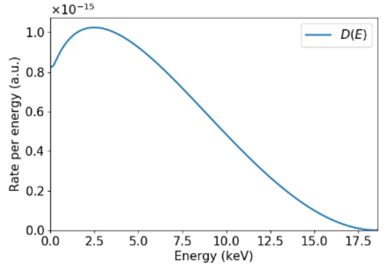
KATRIN experimental overview (data release)



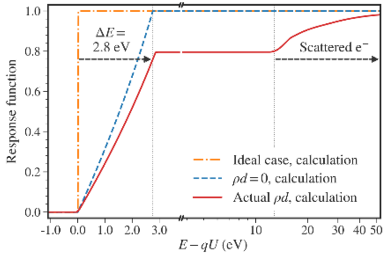
PRL, 123, 221802
PRD, 104 (1), 01200

Nat. Phys. 18, 160-166 (2022)

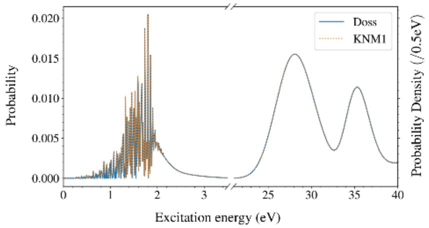
Ingredients for integral spectrum



Differential spectrum



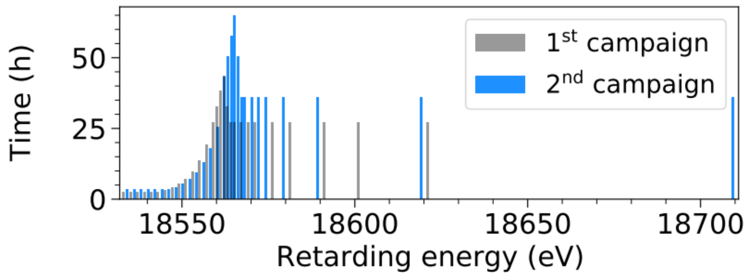
Experimental response
(scattering, transmission, ..)

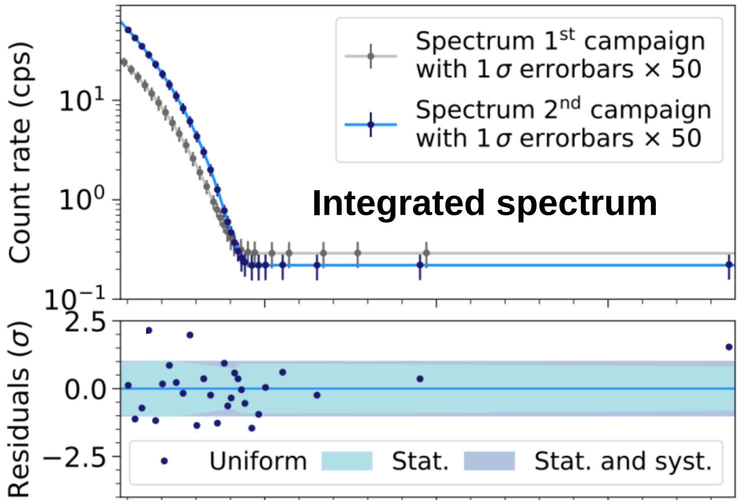


Molecular final state distribution

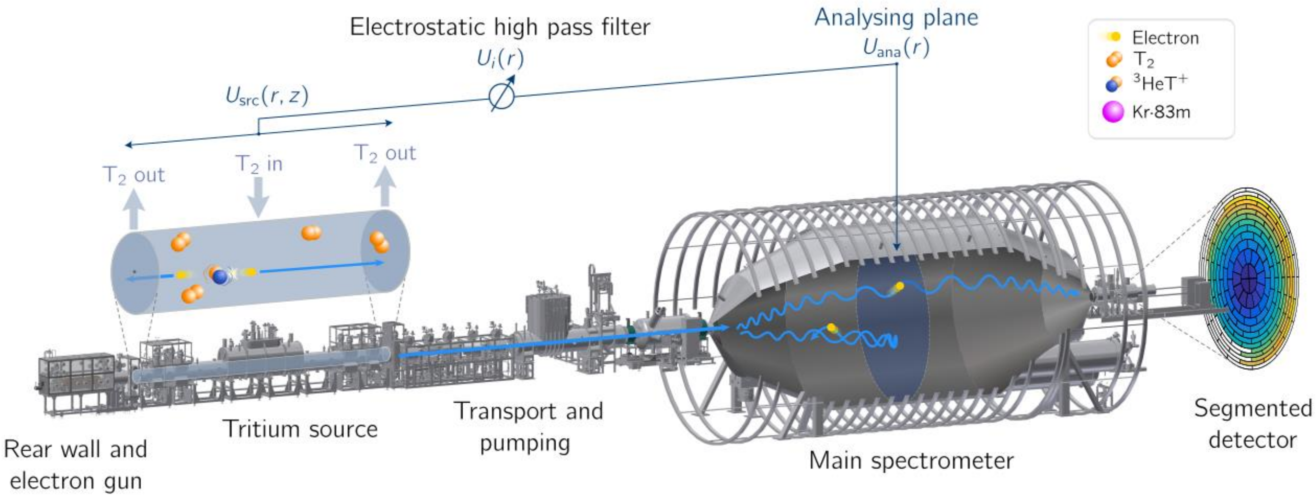
(+ artificial unknown smearing until data unblinding)

Measurement time distribution



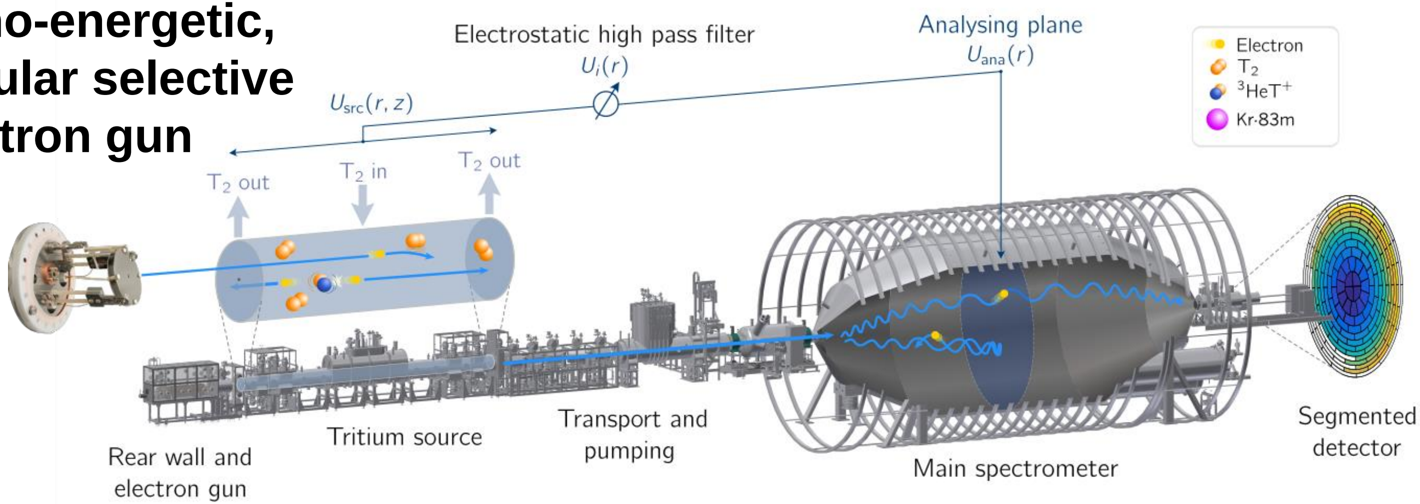


Measurement of key systematics



Measurement of key systematics

Mono-energetic, angular selective electron gun

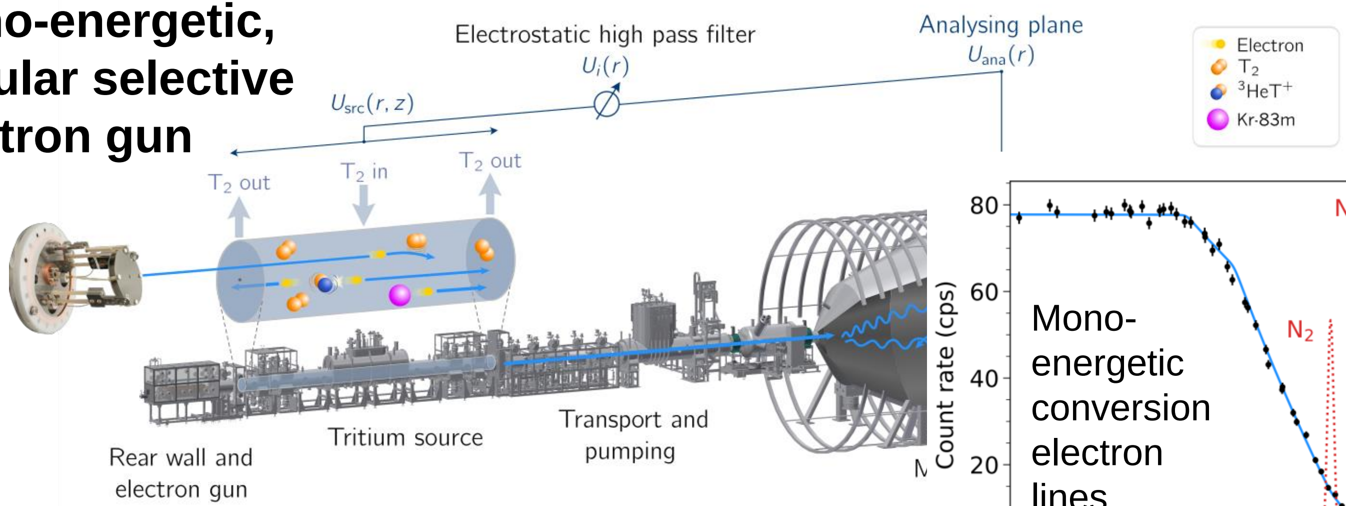


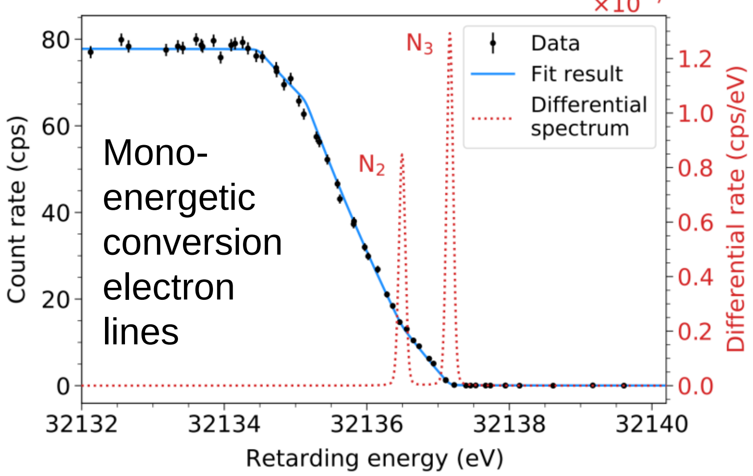
Determination of gas density and energy losses

Measurement of key systematics

Co-circulation of meta-stable Kr-83m

Mono-energetic, angular selective electron gun





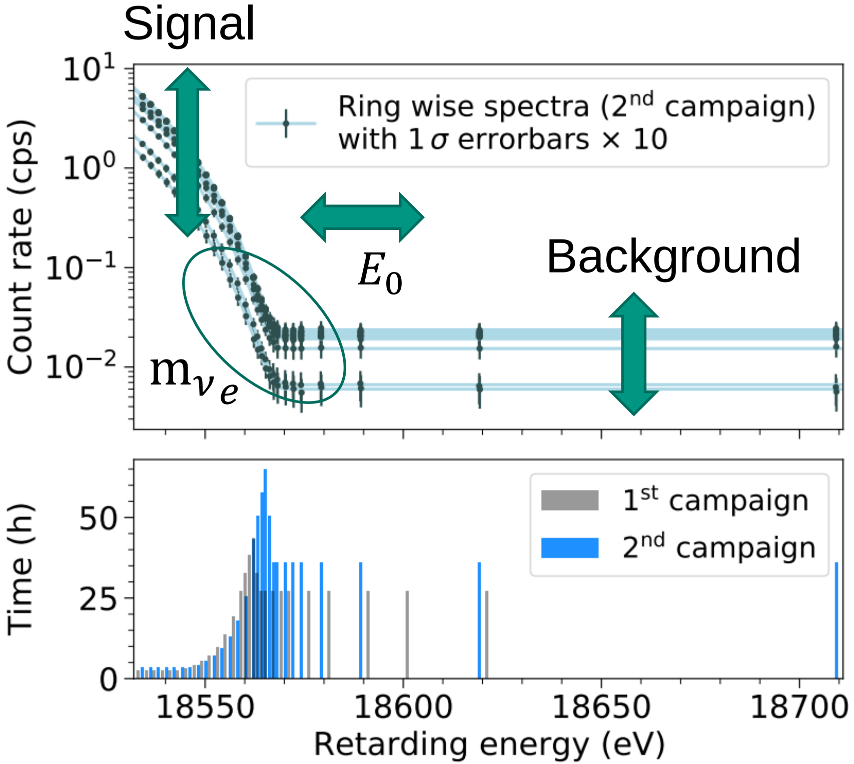
Determination of gas density and energy losses

Mapping of E/B fields

Determination of source potential (absolute, broadening, shifts)

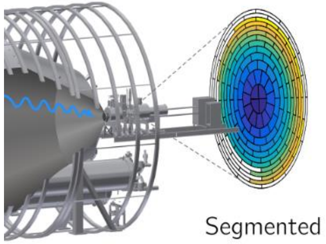
Nat. Phys. 18, 160-166 (2022)

Inference of parameters



- **Ring wise fitting**
  (allowing for radial effects
  in source and spectrometer)

12 x endpoint
12 x signal normalization
12 x background rate
1x neutrino mass
→ **37 parameters**



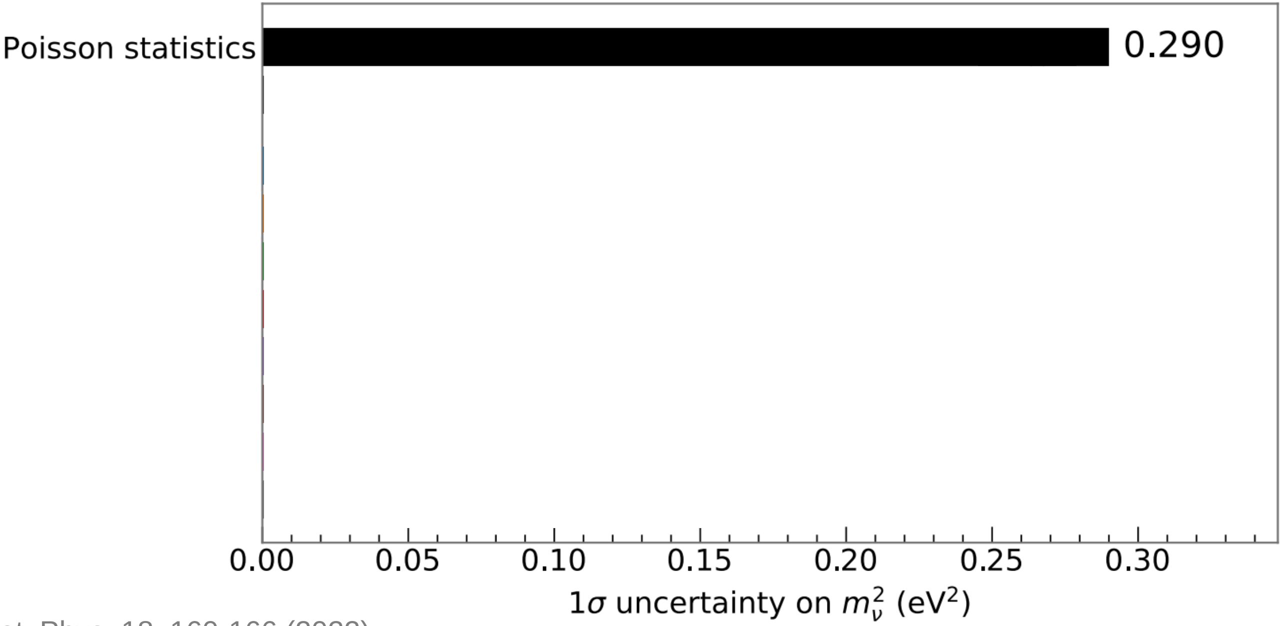
- **Excellent agreement of data to fit-model** $\chi^2/\mathrm{ndof} = 277/299$

- **Consideration of systematic effects**

Nat. Phys. 18, 160-166 (2022)

Breakdown of systematic uncertainty

- Breakdown after 1st of 5 years of KATRIN operation



Nat. Phys. 18, 160-166 (2022)

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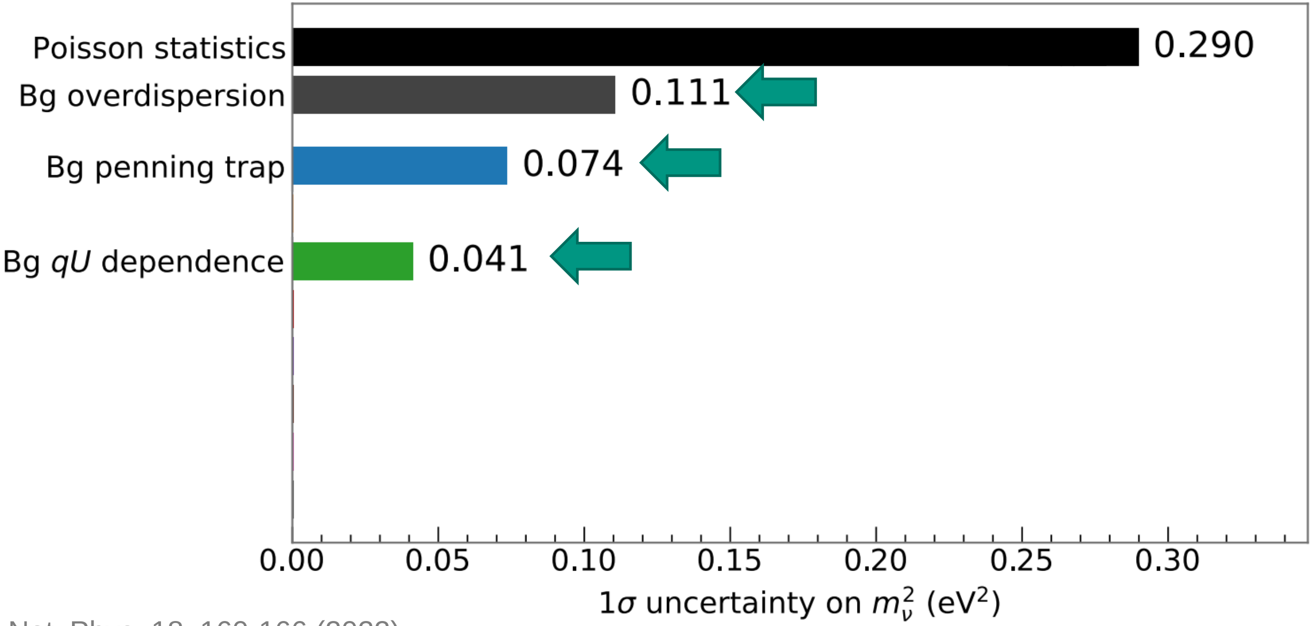

- Background systematics dominate

Nat. Phys. 18, 160-166 (2022)

Breakdown of systematic uncertainty

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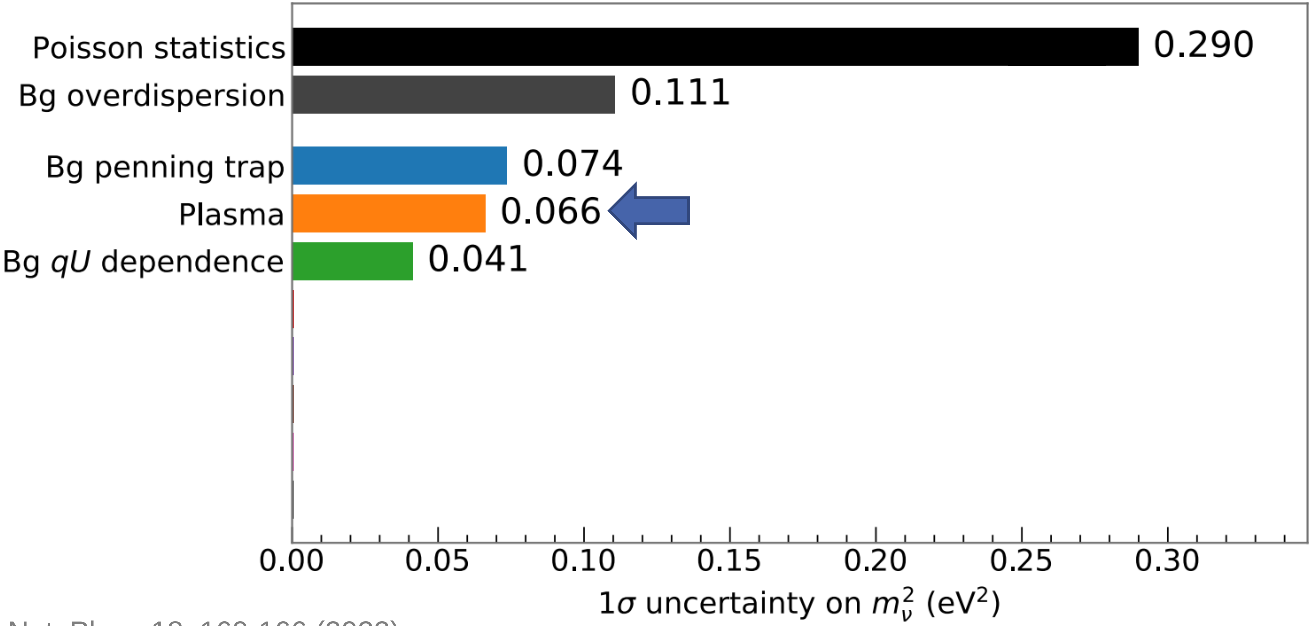

- Background systematics dominate
- Plasma characterisation performed with strong monoenergetic ^{83m}Kr source

Nat. Phys. 18, 160-166 (2022)

Breakdown of systematic uncertainty

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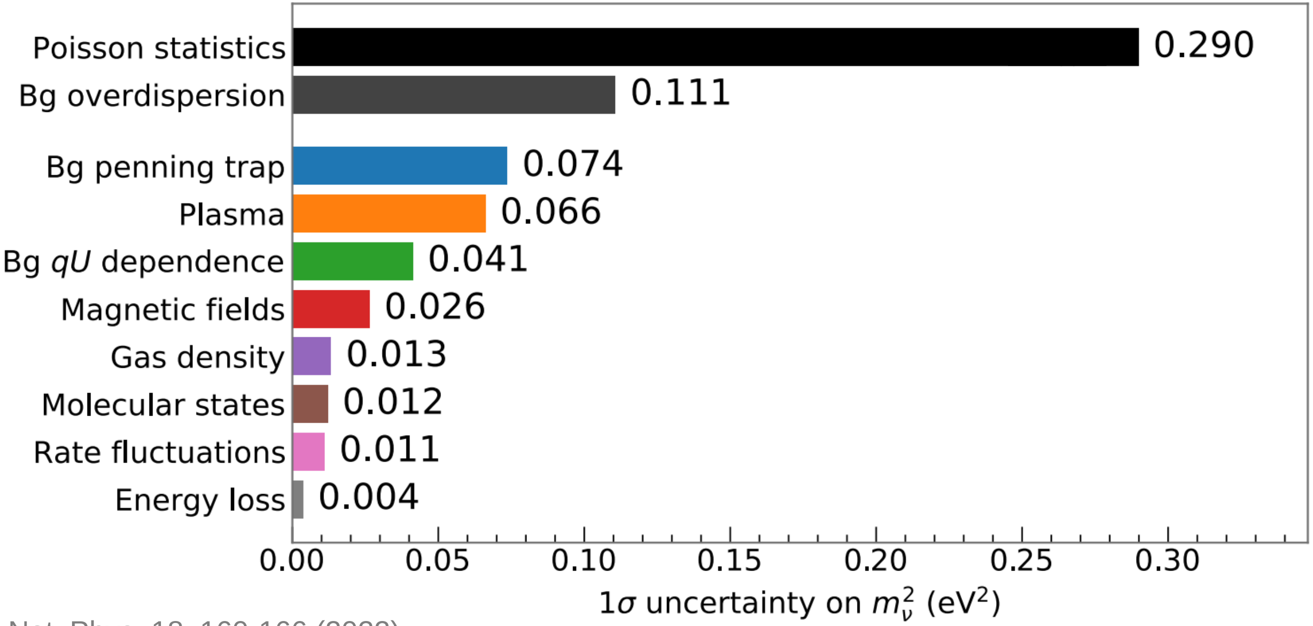
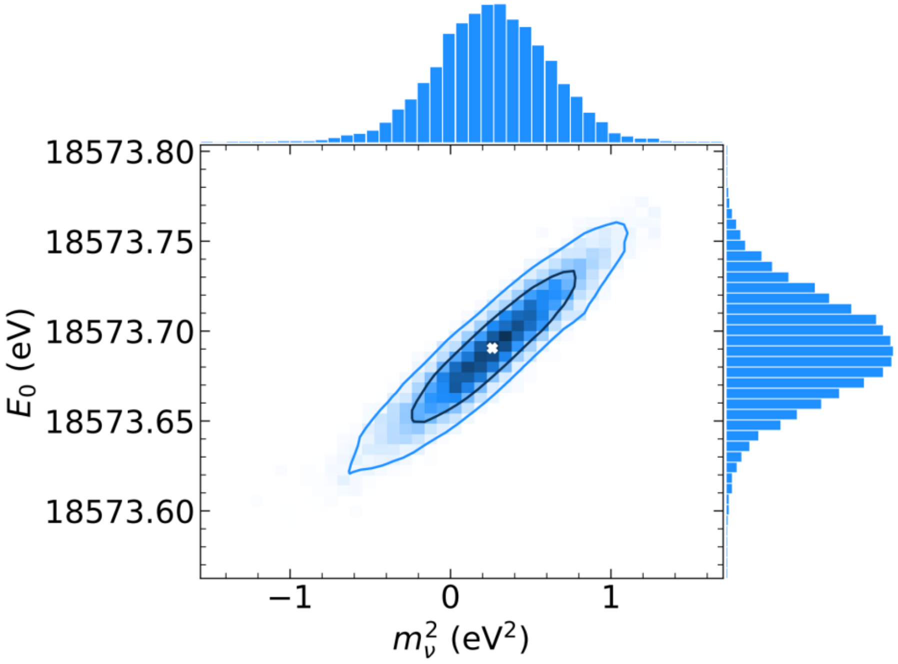


Nat. Phys. 18, 160-166 (2022)

- Background systematics dominate
- Plasma characterisation performed with strong monoenergetic ^{83m}Kr source
- „Other systematics“

Best fit value for m_ν^2



- **Value from 37-free parameter best-fit**

$m_\nu^2 = 0.26 \pm 0.34\ \mathrm{eV}^2$ (90% CL)

First campaign

$m_\nu^2 = (-1.0^{+0.9}_{-1.1})\ \mathrm{eV}^2$ (90% CL)

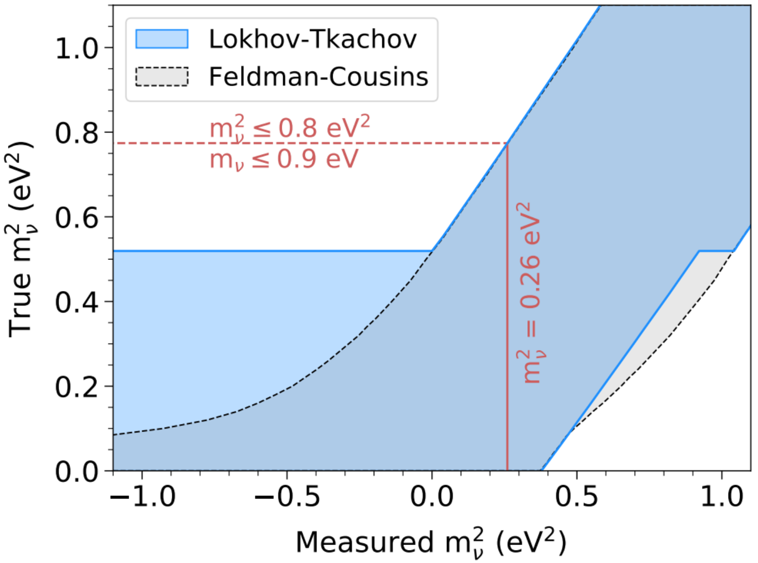
$E_0 = 18573.69 \pm 0.03\ \mathrm{eV}$

In agreement with Q-value from $m(^3\mathrm{He}):m(\mathrm{T})$ mass measurements

Nat. Phys. 18, 160-166 (2022)

New upper limit

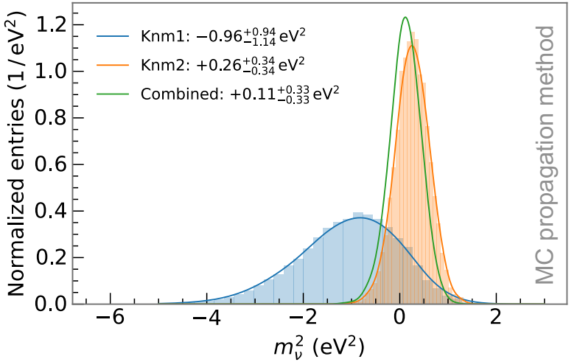
- Frequentist limit construction
  (Lokhov-Tkachov and Feldman-Cousins)



Experimental sensitivity $m_\nu < 0.7$ eV (90% CL)

New limit from 2nd campaign
$m_\nu < 0.9$ eV (90% CL)

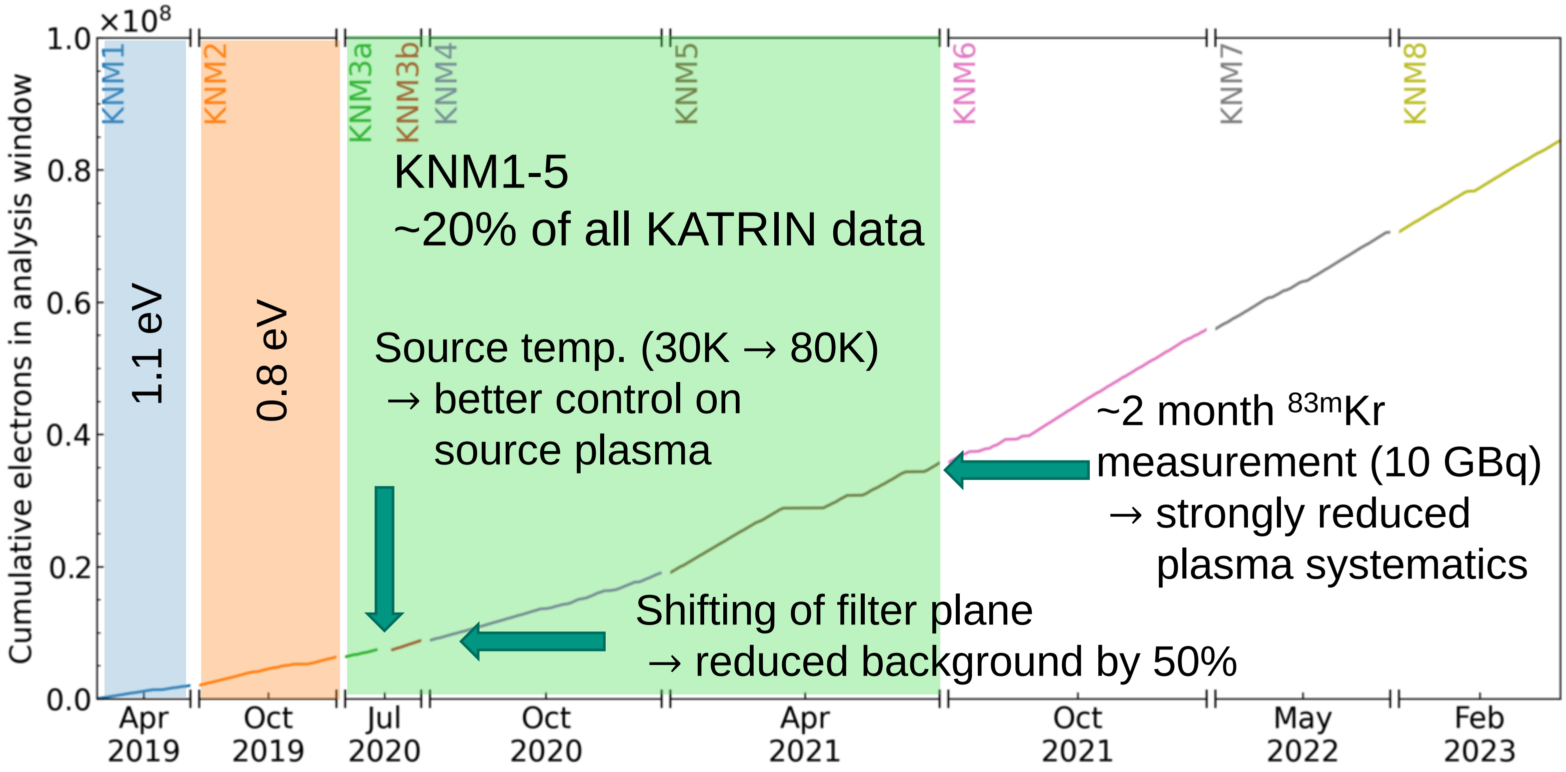
First campaign $m_\nu < 1.1$ eV (90% CL)



New limit from KATRIN
$m_\nu < 0.8$ eV (90% CL)

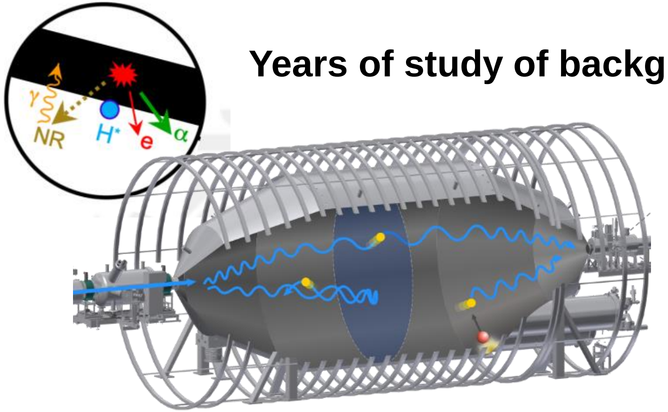
Nat. Phys. 18, 160-166 (2022)

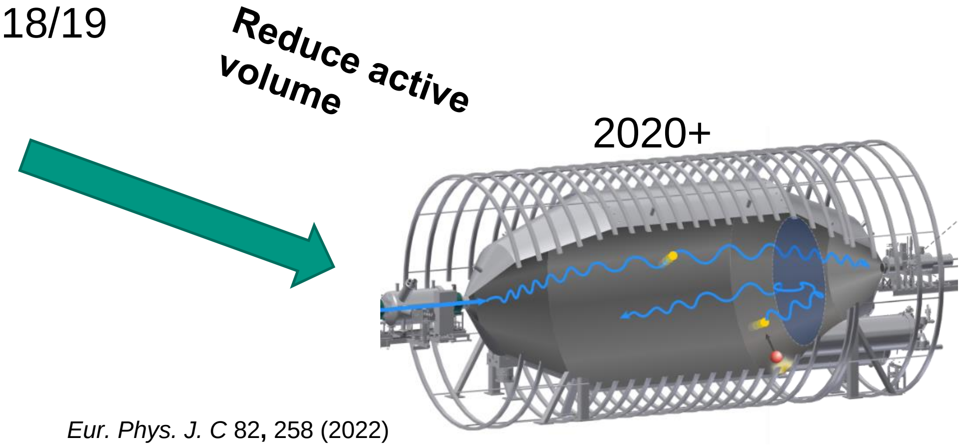
KATRIN experimental overview (data release)



The background as main obstacle for sensitivity

Years of study of background





- Initial **radioactive** decay
- Excited **neutral** atoms (Rydberg states)
- **Ionization** in spectrometer volume
- Background electron **at filter energy**

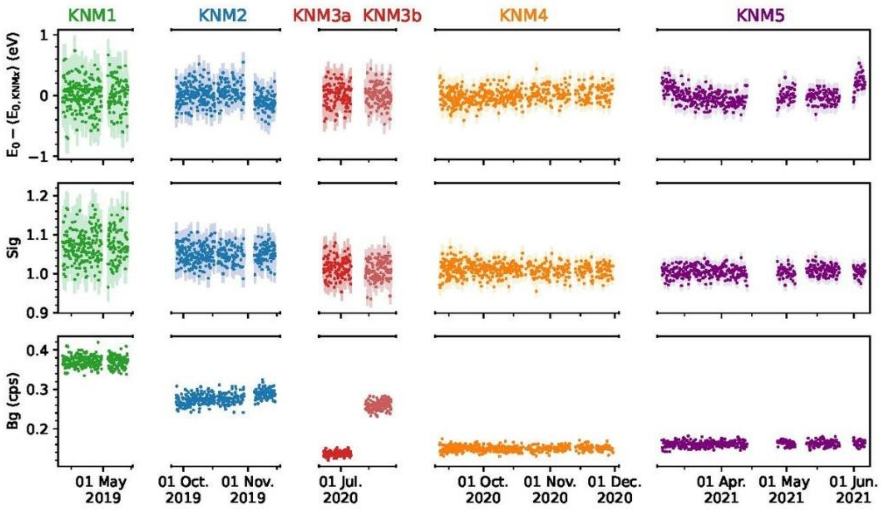
Eur. Phys. J. C 82, 258 (2022)

Background rate: 0.22 counts s^{-1} → 0.13 counts s^{-1}

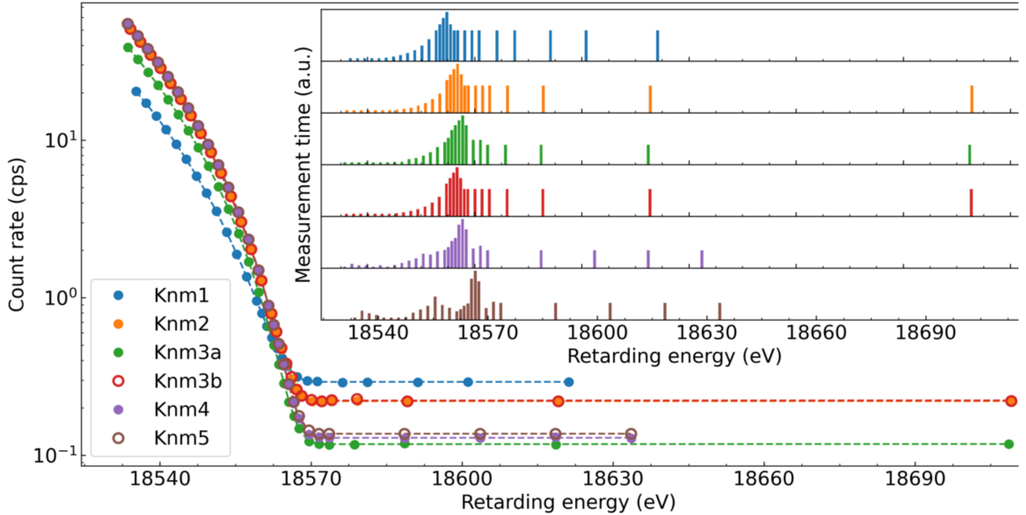
Analysis „plane“ distorted → 14 radially resolved spectra

Peak into latest data

- Fit parameters from 1757 spectral scans



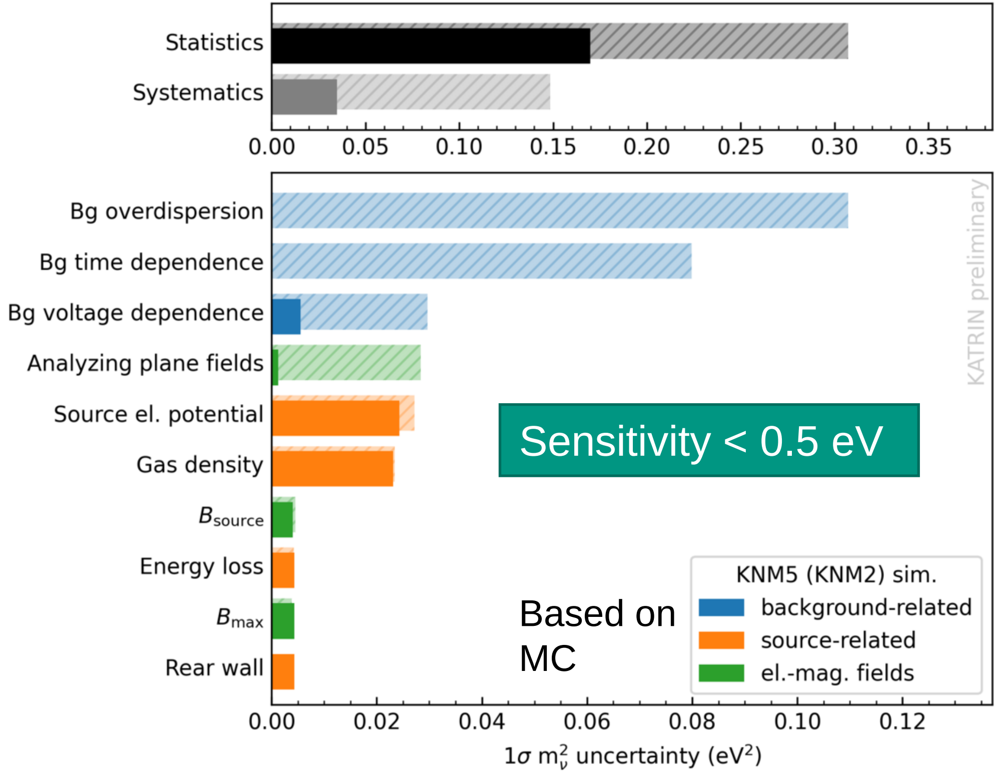
- „Uniform“ spectra



→ **1259 data points** (runs with shifted analysis plane have 14 individual patches) and **136 free parameters**

Sensitivity of KNM1-5 data set

Karlsruhe Institute of Technology



Achievements

- Reduced backgrounds by improved field settings

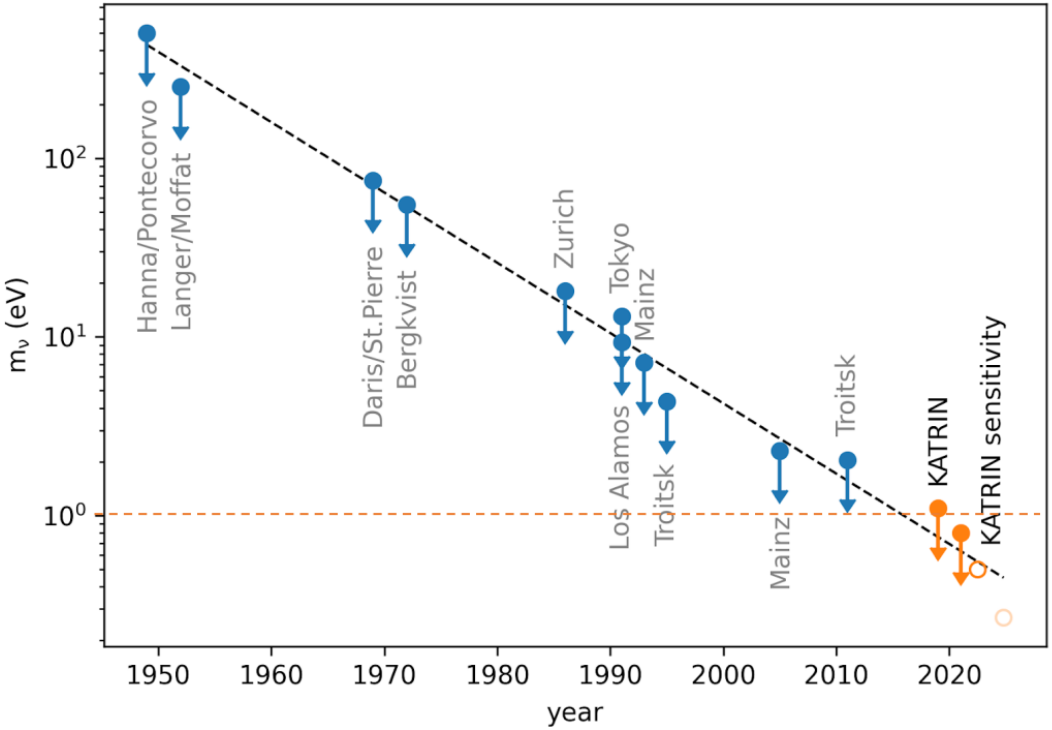
Planned improvements

- Refine plasma characteristics
- Improved measurement of gas density

… both with **new egun**

*KATRIN sensitivity is still **limited by statistics***

Evolution of neutrino mass measurements

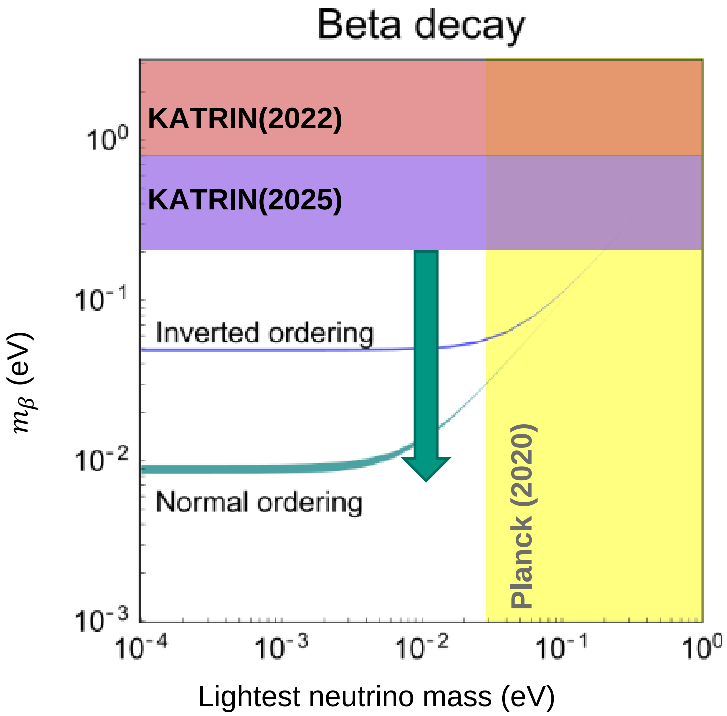


eV limit

KNM1-5 sensitivity

KATRIN goal

How to go beyond the KATRIN aim?



More statistics

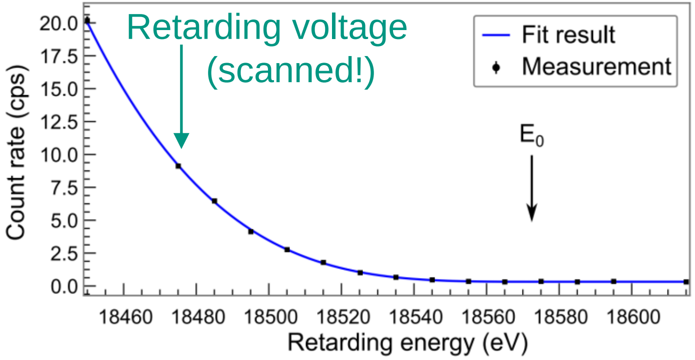
More signal	Less background
More tritium	Avoiding of BG-sources
More efficient detection principle	Mitigate BG source
	Better discrimination

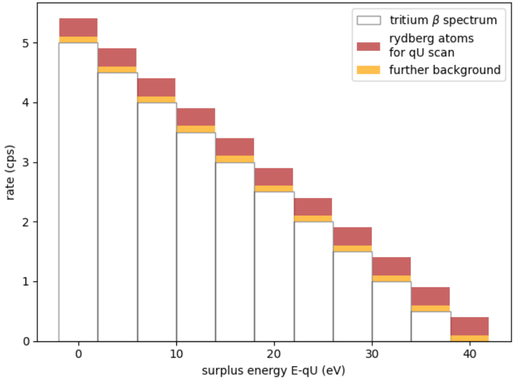
Better understanding of systematics

- Avoiding of systematics
- Migitation / measurement of systematics

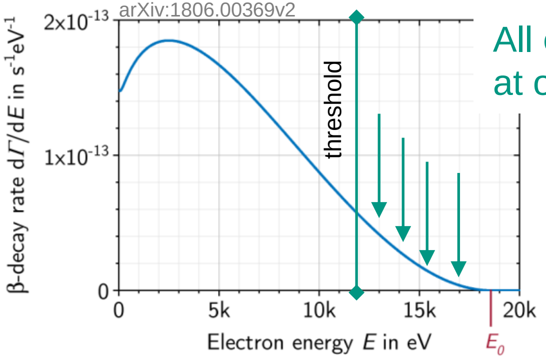
Improved measurement principle

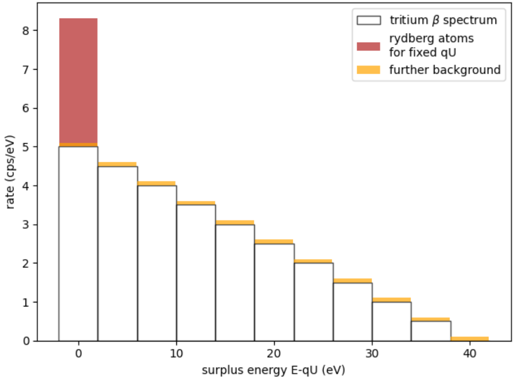
Integral measurement (high pass filter)
- Energy resolution determined by filter
- Detector „only“ counts
- Reduced statistics





Differential measurement
- Energy resolution determined by detector or time of flight

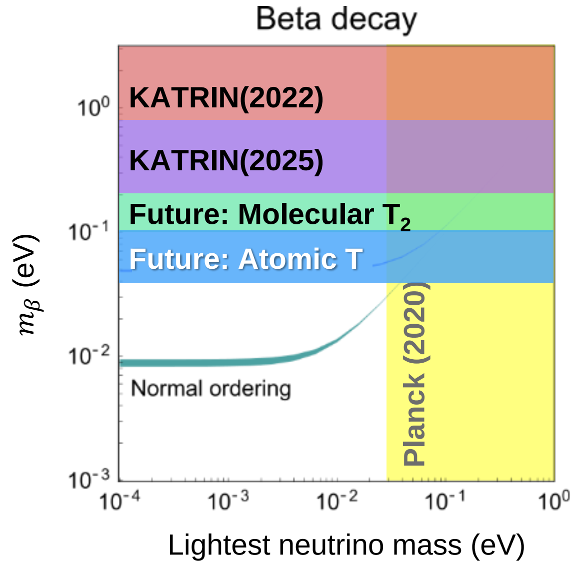


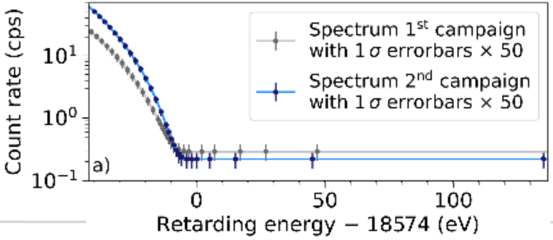


More signal

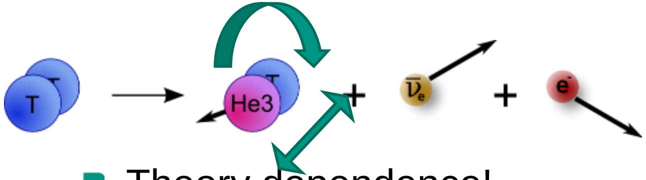
Less background

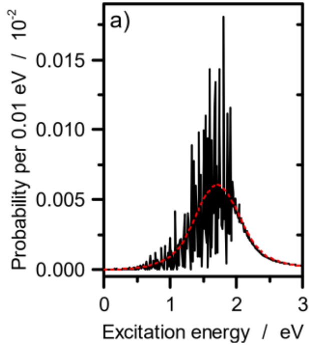
Conquering new frontiers with new technology





- **Molecular tritium decay**





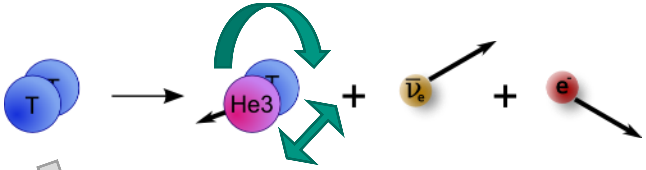
  - Theory dependence! (calculation accuracy)
  - Effective smearing of energy resolution ($\sigma \sim 400$ meV)

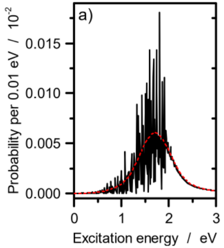
Solution:
Use atomic tritium

- **New detection methods**
  - Differential methods *(MAC-ToF, Calorimetric, CRES)* (Higher resolution, reduced background)
  - More statistics, better control of systematics

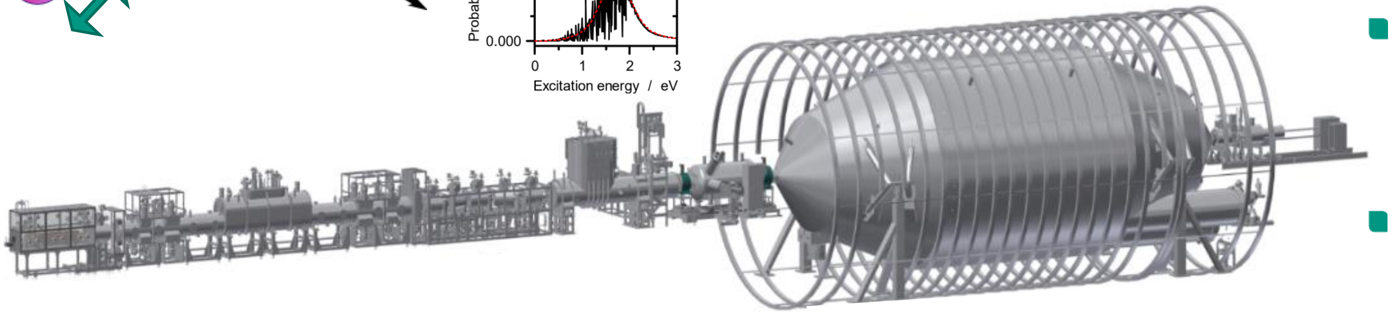
KATRIN and TLK as ideal R&D facilities

- Molecular effects → spectral broadening

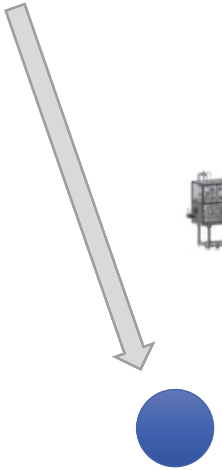




Quantum detector technology

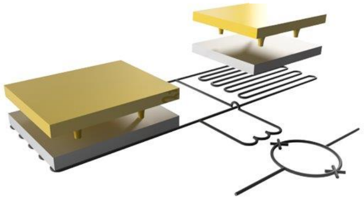


- eV resolution for **differential** detection
- **immune to** Rydberg-like **backgrounds**



Atomic source technology

- µm-size calorimeters



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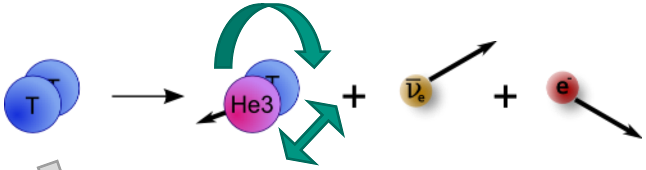

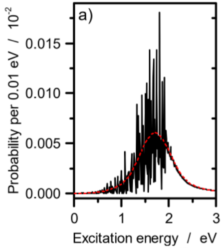

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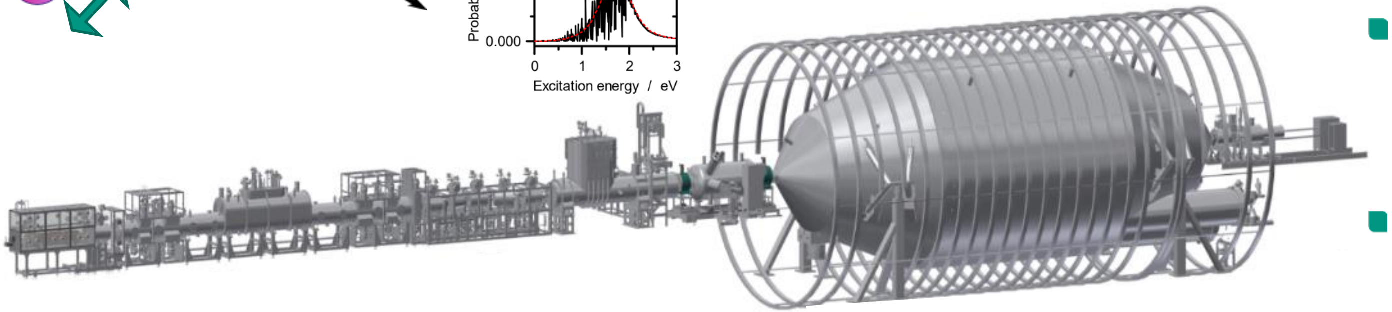
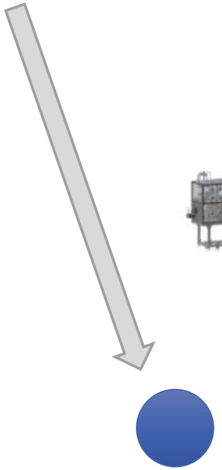
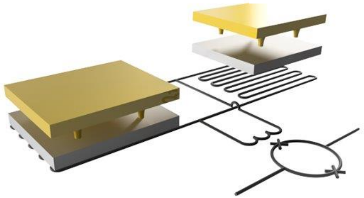


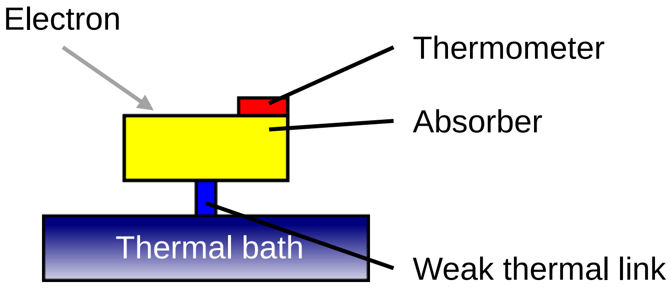


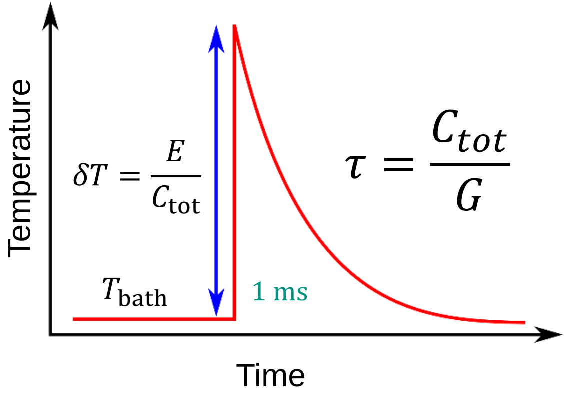
Atomic source technology

- µm-size calorimeters



Quantum sensors as high resolution differential detectors





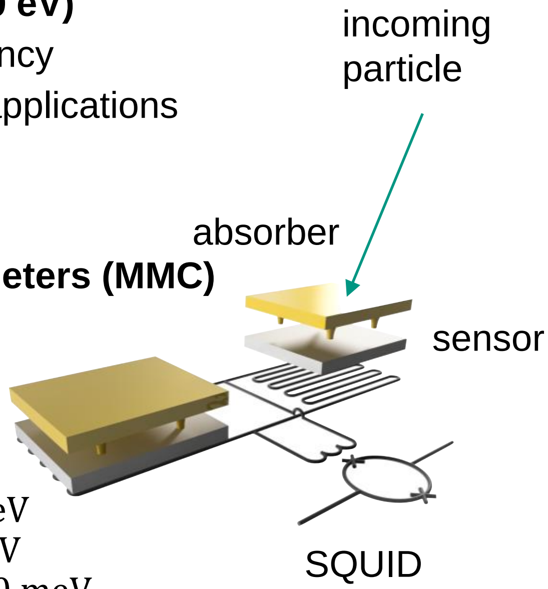
$$\tau = \frac{C_{tot}}{G}$$

Advantages

- Energy resolution ***O*(eV)** compared to conventional detectors ***O*(100 eV)**
- Nearly 100% quantum efficiency
- Broad spectrum of possible applications

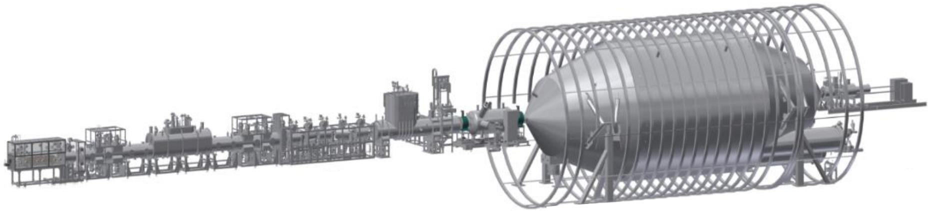
e.g. Metallic Magnetic Calorimeters (MMC)

- Temperature-dependence in sensor magnetization
- Read-out by SQUID
- Energy resolution:
  - Current: $\Delta E \lesssim 2$ eV
  - Midterm: $\Delta E \lesssim 1$ eV
  - Future: $\Delta E \sim 100$ meV



Not yet tested with external electrons

Differential detector – Challenges



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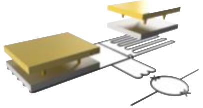
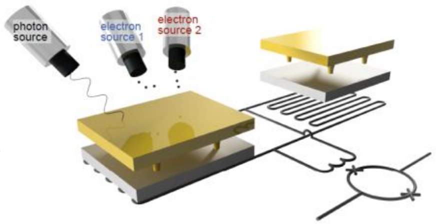


- Concept of coupling **quantum sensor detector array** to a **KATRIN-like infrastructure**
  - **Type** of quantum sensor
  - Operation in **magnetic field**
  - Coupling of **mK cryo-platform** with room temp spectrometer
  - Large area detector and **multiplexing**
  - **Limits to energy resolution**

ELECTRON Project

KIT-Future field stage 2

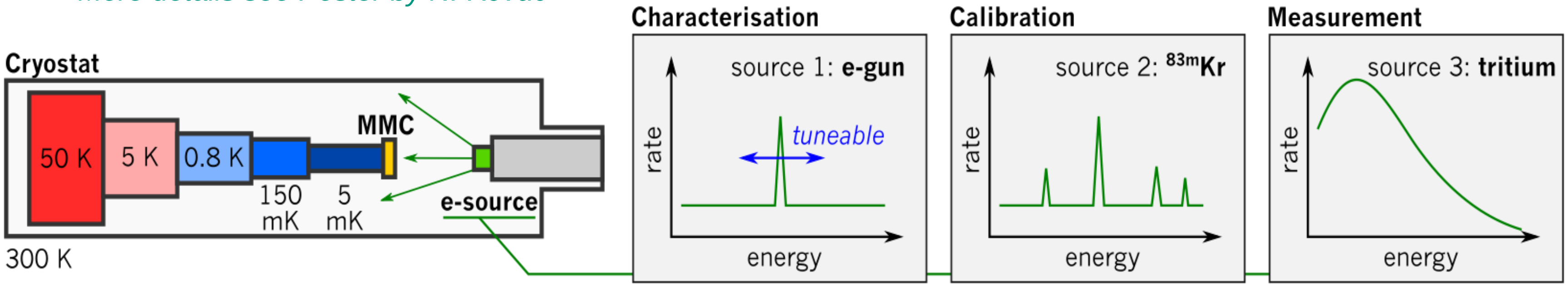
Measure external electrons with MMC detectors
Cooperation of IAP, IAP-TLK with
world-leading expert Prof. Kempf (KIT-IMS)



- Electron-gun → Characterization of the detector-electron interplay
- ^{83m}Kr source → Calibration of the detector response
- Tritium → Differential spectrum

Started in summer 2022

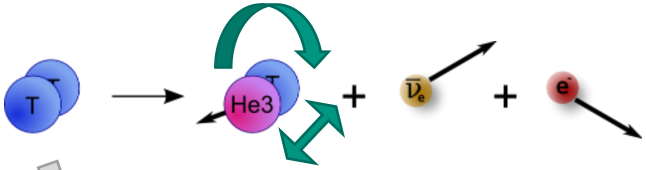
More details see Poster by N. Kovač

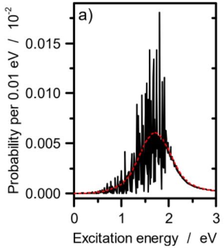


First krypton measurements with MMCs in mid of April 2023.

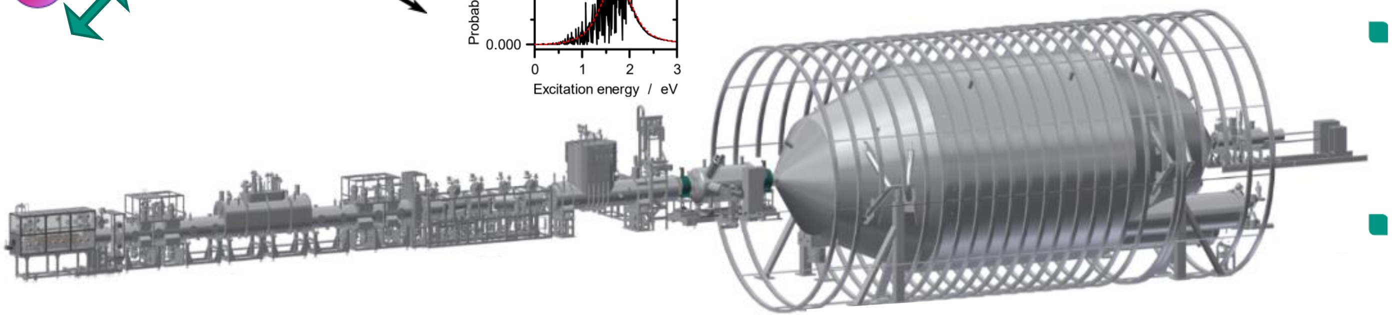
KATRIN and TLK as ideal R&D facilities

- Molecular effects → spectral broadening





Quantum detector technology



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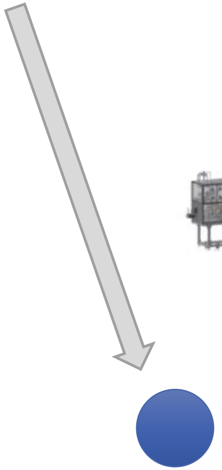
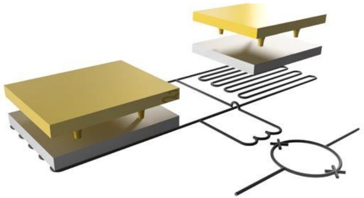
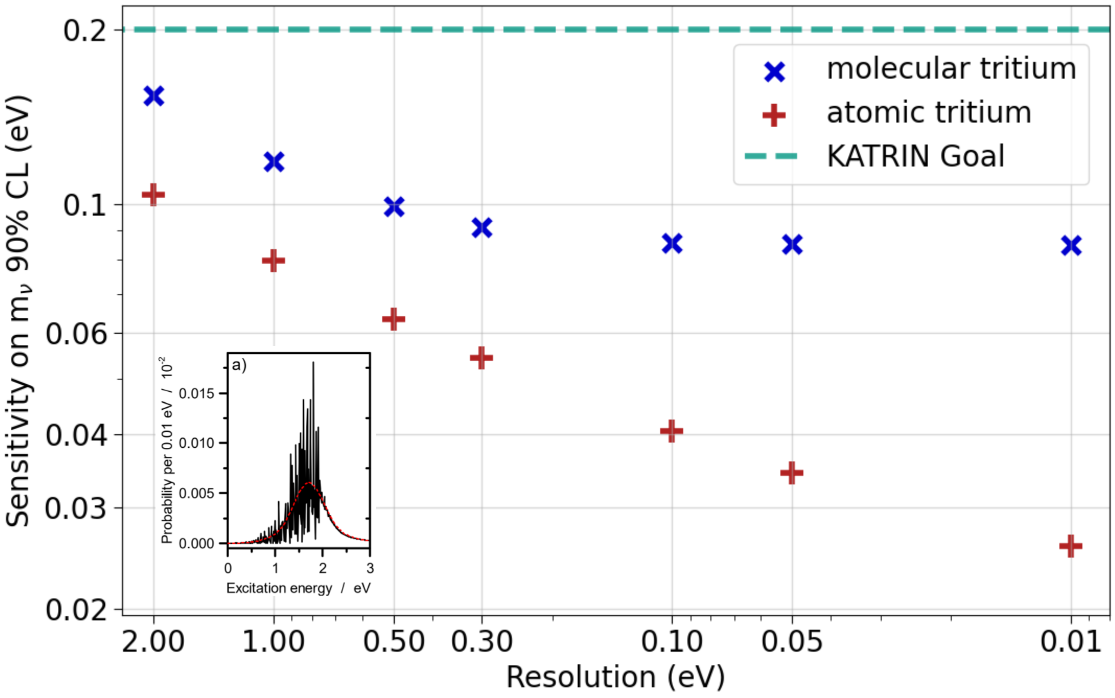


Atomic source technology

- µm-size calorimeters



Sensitivity with atomic source



By Svenja Heyns

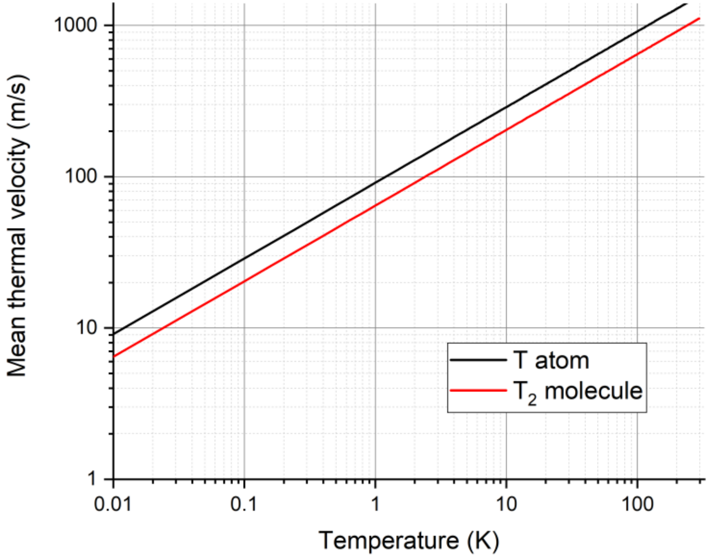
$qU = 18520$ eV
Fitrange: $E_0 - 30$ eV
CD @ 75%
$m_\nu = 0.00$ eV
stat. bg = 0.00 cps/eV
'statistics only'
Scattering included

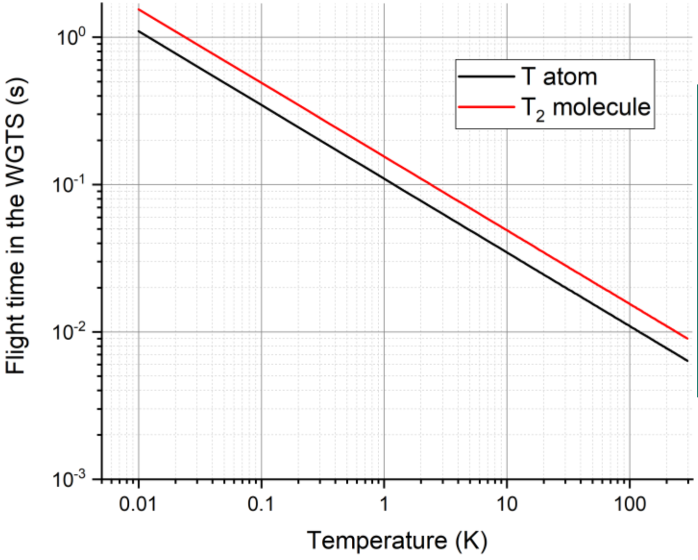
Sensitivity limited by molecular broadening

Activity from an tritium source

$$\text{Activity} \propto \text{Tritium injection} \times \text{flight path} \times \text{velocity}^{-1}$$

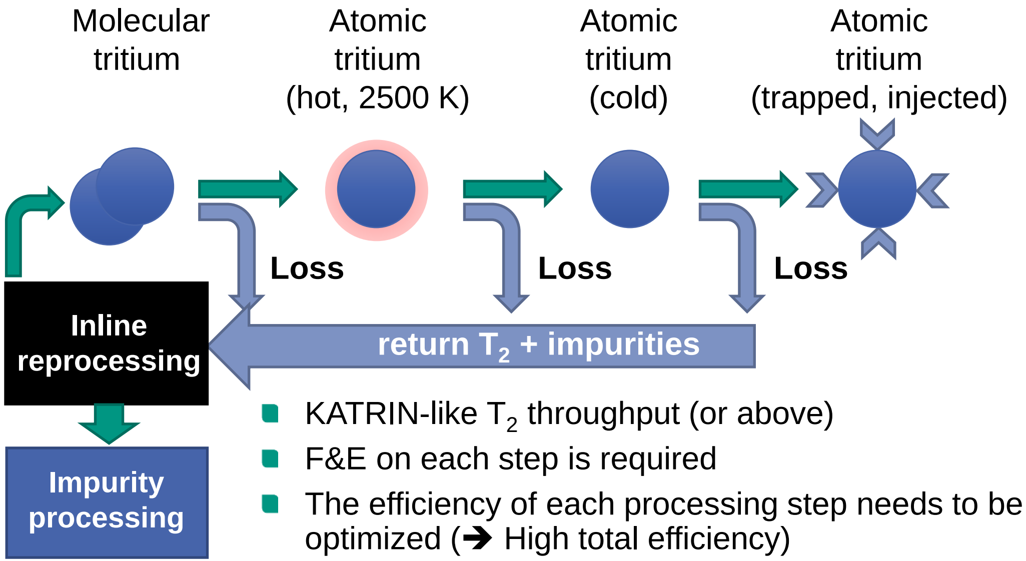
Example length KATRIN source ~ 10 m





Needed:
highly efficient atomic tritium generation, cooling and transportation

Atomic Tritium at Tritium Laboratory Karlsruhe



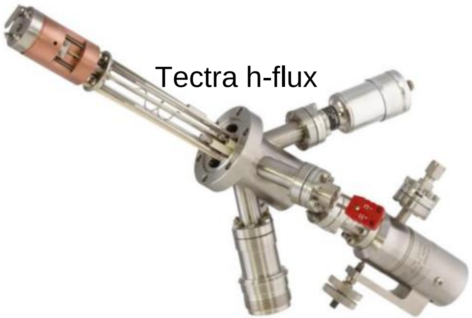
- KATRIN-like T_2 throughput (or above)
- F&E on each step is required
- The efficiency of each processing step needs to be optimized (➔ High total efficiency)

TLK is unique infrastructure for atomic source development **with partners**

Total glove box volume	**190 m³**
Experimental & Infrastructure area	**1600 m²**
Gloveboxes	**20**

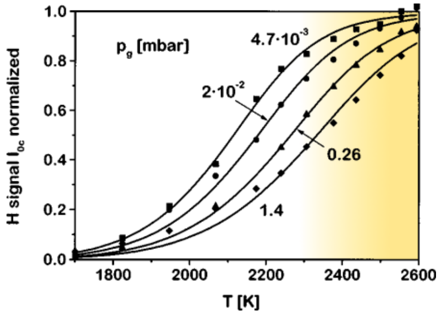
First stage: Tritium dissociation

$$T_2 + \text{energy} \rightarrow T + T$$

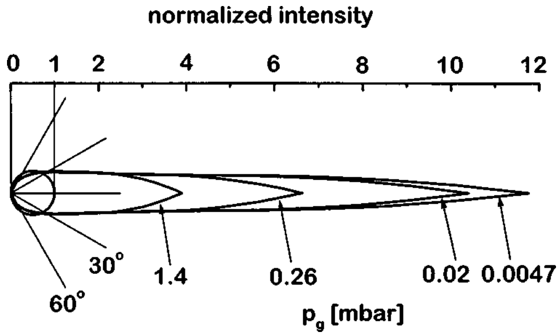


- Different technologies exists and are employed in literature
  - Photo-dissociation
  - RF-discharge
  - **Thermal cracking (current method of choice; following P8 approaches)**

Degree of dissociation of H_2



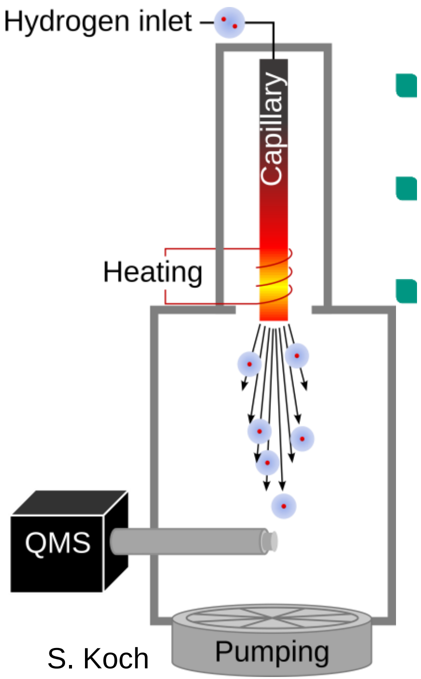
Angular distribution of H



Tschersich. J. Appl. Phys. 87, 2565–2573 (2000). Figures modified.

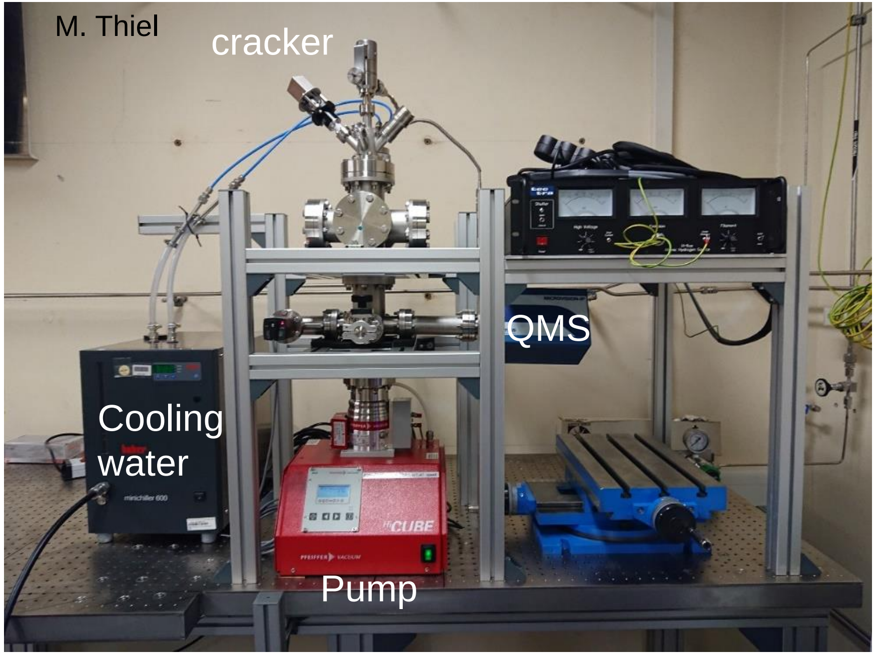
Atomic hydrogen source Test Mk 1 (AHS1)

- **Concept**

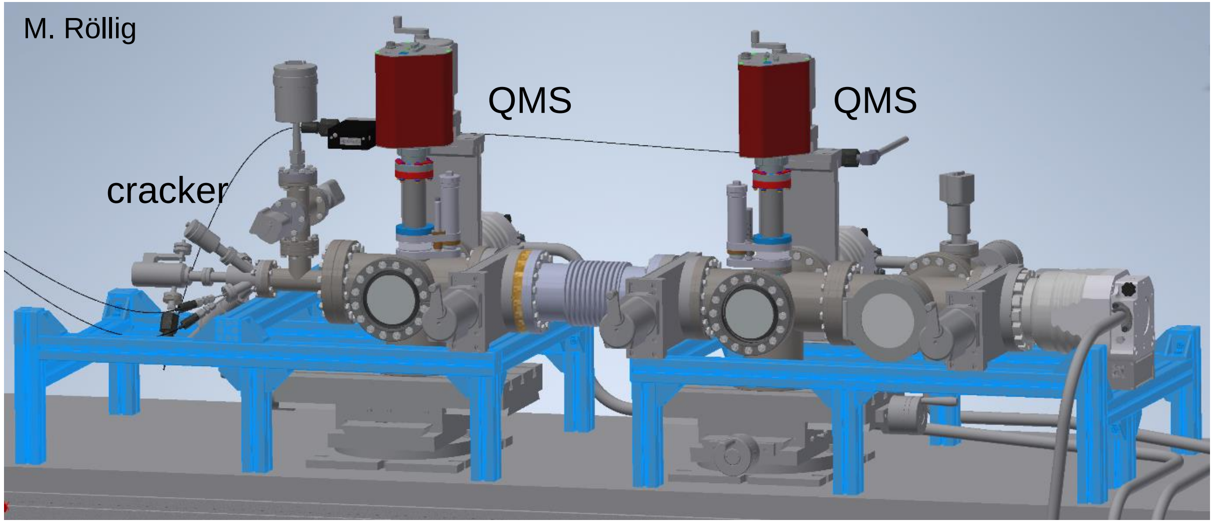


- Commercial Tectra h-flux source
- Single chamber design
- „standard“ quadrupole mass spectrometer (QMS)

- **Test setup in TLK „cold lab“**



Atomic hydrogen source Test Mk 2 (AHS2)



- Avoid short-comings of AHS1.0 → multi-chamber, multi-skimmer, 2+ TMPs, …
- Very modular (standard components, manipulators, ..)
- **ToF** option (short-term), **nozzle cooling** (mid-term), **velocity selection** (long-term)

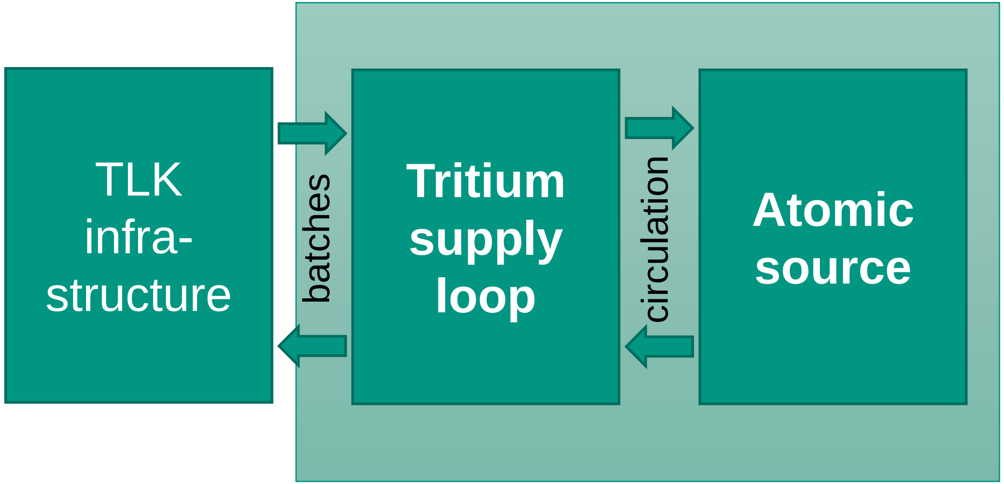
Towards first atomic tritium source at TLK

„AHS 1.5“ Emptied ALTEX box

Preparation of glovebox integration for **first atomic tritium** experiment ongoing

See Poster by Leonard Hasselmann

Configuration with O(5g) tritium inventories

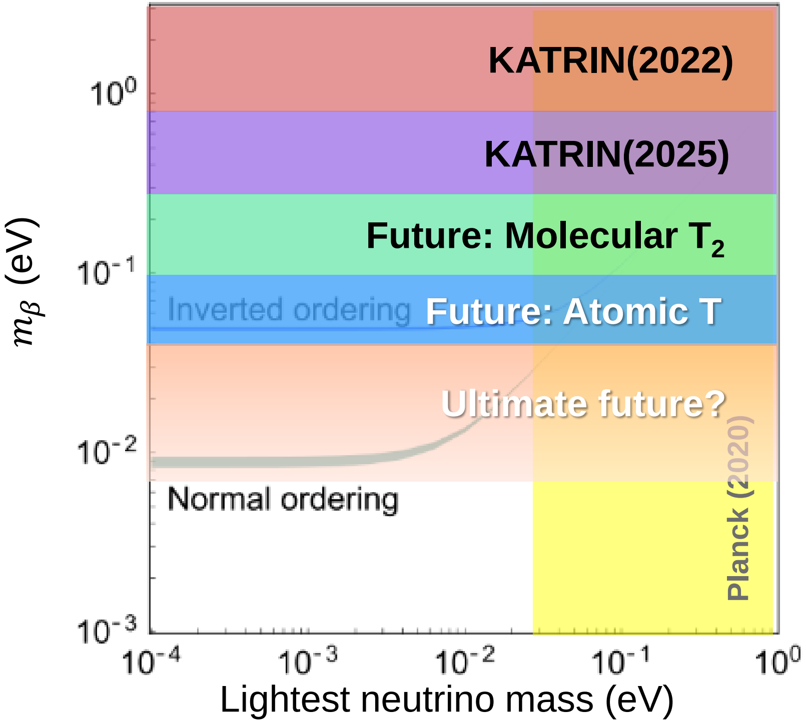


- Universal loop for provision of 20 sccm flow (T_2) ~ 20% of KATRIN
- Simple cracker-based test experiment

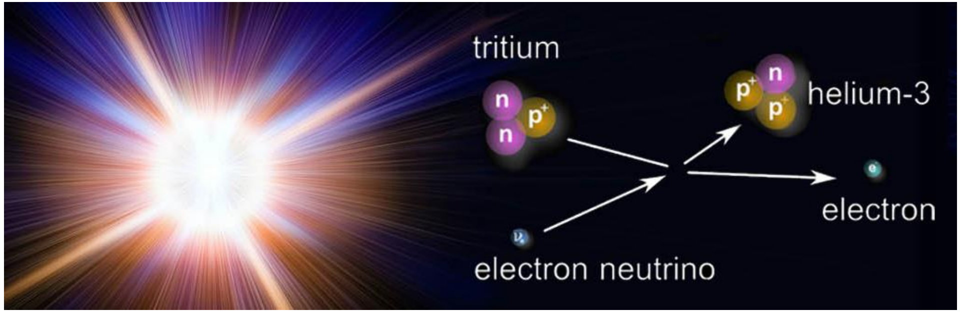
First experiments planned in 2024

Conquering new frontiers

- Model-independent **measurement of m_ν**



- „Holy grail“ quest
  Direct detection of relic neutrinos



New technology needed!

High-resolution differential detectors

Atomic tritium sources

Active within KCETA / KSETA

30 years of TLK

- We invite KCETA / KSETA to join our festivities
- More information on: https://www.iap.kit.edu/tlk/english/410.php

iap.kit.edu/tlk

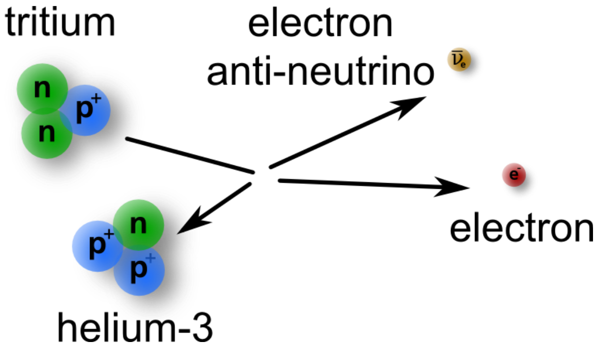
twitter.com/tritiumlab

instagram.com/tritiumlab

sciencemastodon.com/@tritiumlab

$$t_{\text{universe}} = 13.6 \times 10^{9}\ a$$

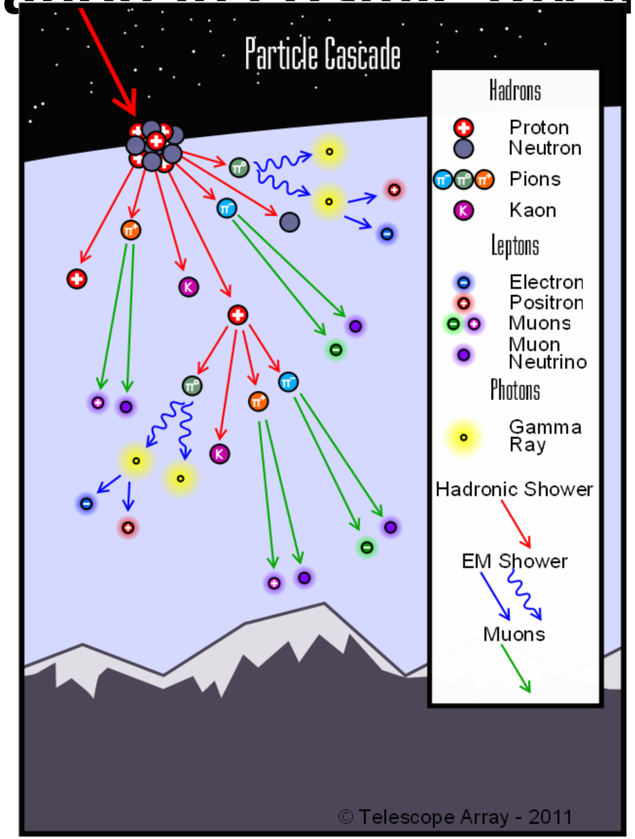
$$t_{solar} = 4.6 \times 10^{9}\ a$$



$$\tau_{1/2} = 12.3\ a$$

WHERE DOES THE TRITIUM COME FROM?

Generation in cosmic ray interactions



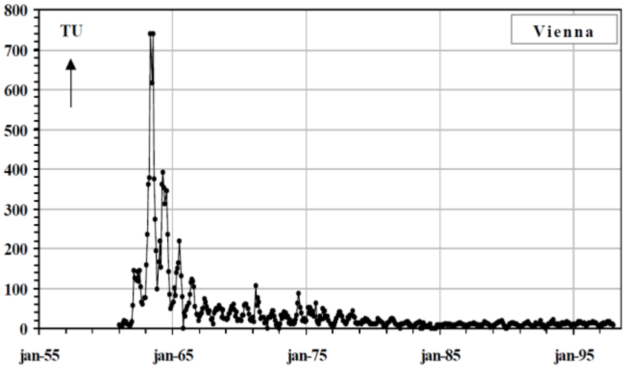
- Production of T mainly in upper troposphere and lower stratosphere

$$^{14}_{7}\mathrm{N} + ^{1}_{0}n \rightarrow ^{12}_{6}\mathrm{C} + ^{3}_{1}\mathrm{H}$$

- Yearly production ~ 200 g

Natural tritium content
3.5 kg

Atmospheric nuclear bomb tests

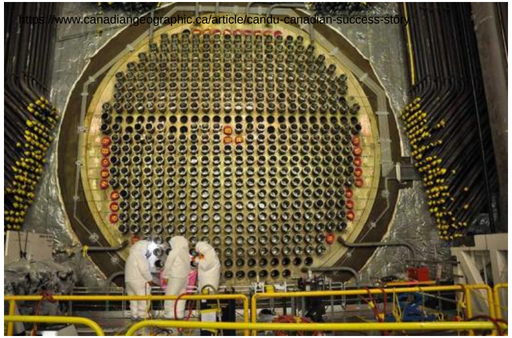


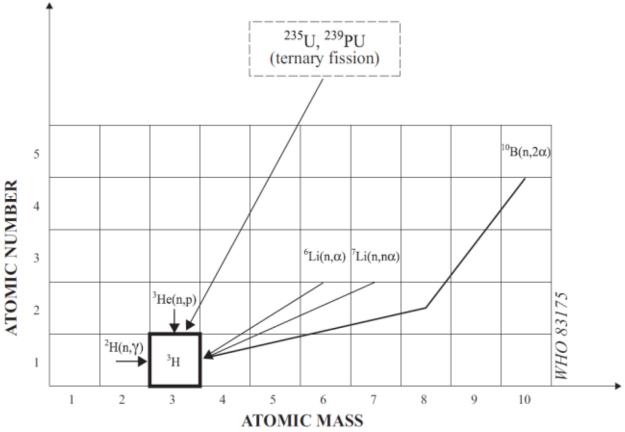
from IAEA (http://www-naweb.iaea.org/napc/ih/documents/global_cycle/vol%20II/cht_ii_05.pdf)

- **Atmospheric nuclear test brought about 1.5 kg per mton**
- **Test were stopped after 1963**
- **In 2007, there remained around 40 kg**

Nuclear bomb tritium today (2019)
~ 20 kg

Tritium (side) production in nuclear reactors





- **Tritium is produced in ternary nuclear reaction in the fission fuel**
- **Heavy water reactor (D_2O moderated)**

$$ {}^{2}_{1}\mathrm{H} + {}^{1}_{0}n \rightarrow {}^{3}_{1}\mathrm{H} + \gamma $$

- **Deuterium activation**

~ 100 g of tritium per year per heavy water reactor (600 MW class)