Bundesministerium für Bildung und Forschung

SFB 1258 Neutrinos Dark Matter Messengers

Jagiellonian University in Krakow

A liquid-phase circulation-mode argon purification system

Christoph Vogl[1*], **Mario Schwarz**[1], **Patrick Krause**[1], **Grzegorz Zuzel**[2], and **Stefan Schönert**[1]

[1]TU-Munich, Physics Department, Garching, Germany
[2]Jagiellonian University, Institute of Physics, Cracow, Poland

[*]christoph.vogl@tum.de

High-purity noble liquids are difficult to procure, creating the need for on-site purification

Impurities in noble liquid detectors strongly deteriorate performance

Noble liquids often not available in required purity (or expensive, in limited amounts, etc.)

→ Experiments perform purification themselves

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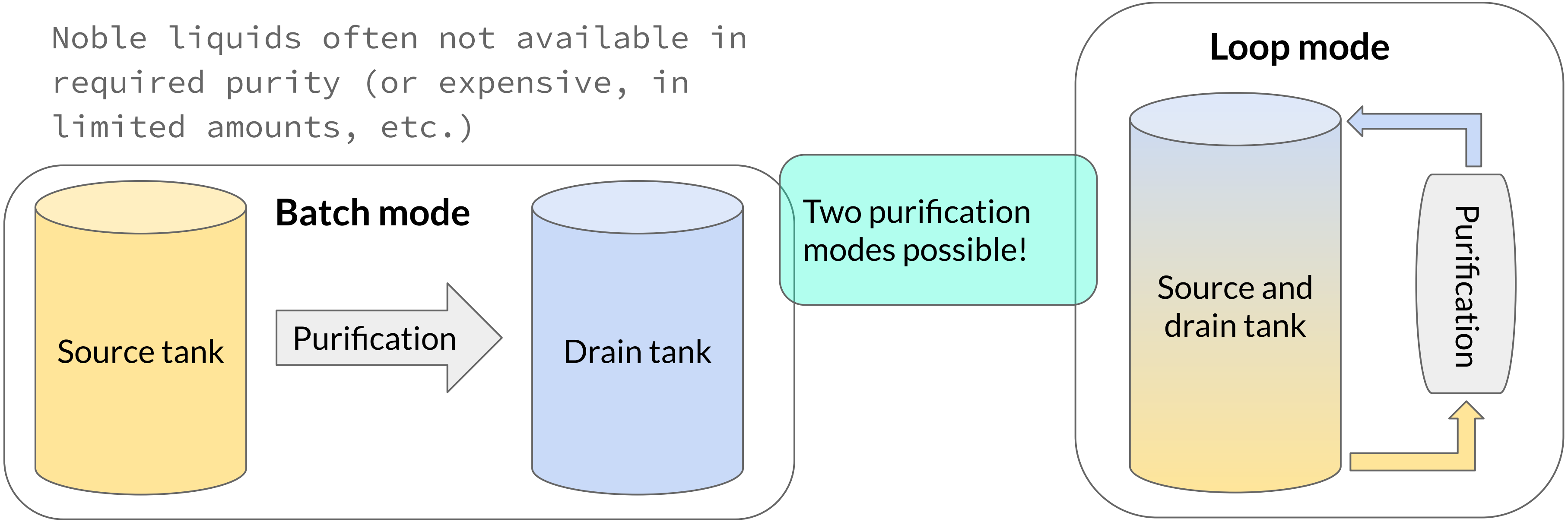
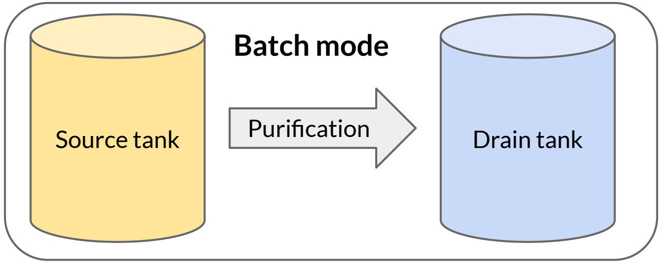


There are several modes in which noble liquids can be purified



Source tank ≠ drain tank

✅ Efficient

✅ Does not need a pump

✘ Needs two separate tanks

✘ only possible during detector filling, not possible during experimental runs

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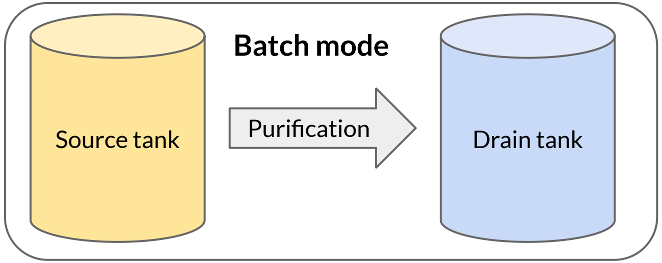

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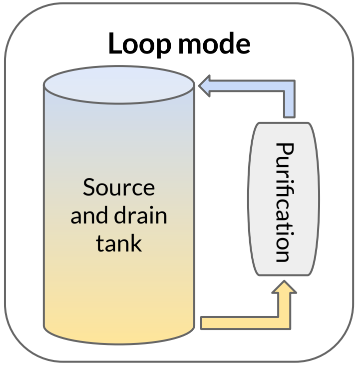
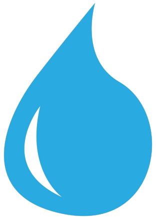


Source tank = drain tank

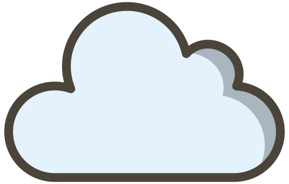
✘ Less efficient b/c impurities dilute
✘ Needs a pump
✅ Only needs one tank
✅ possible during experimental runs

In which phase to purify?



Liquid phase

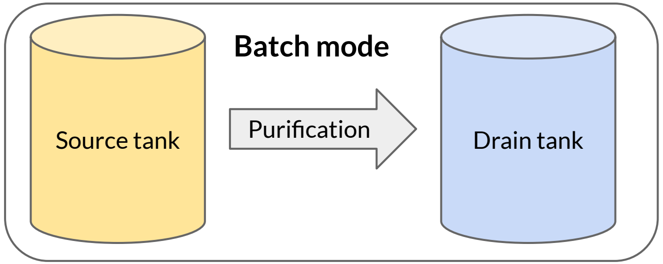
✅ No need to change phase (no boiling or condensing)
✅ High throughput due to high density
✅ Possible during detector filling in batch mode
✘ Less effective

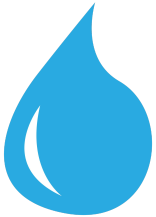


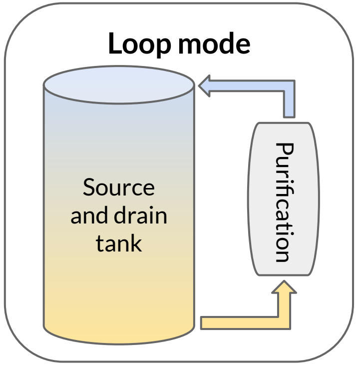
Gaseous phase

✅ Very effective due to high mobility
✘ Need to change phase at least once (recondensing and/or boiling)
✘ Less throughput

Here: Purification in liquid phase and either batch or loop mode





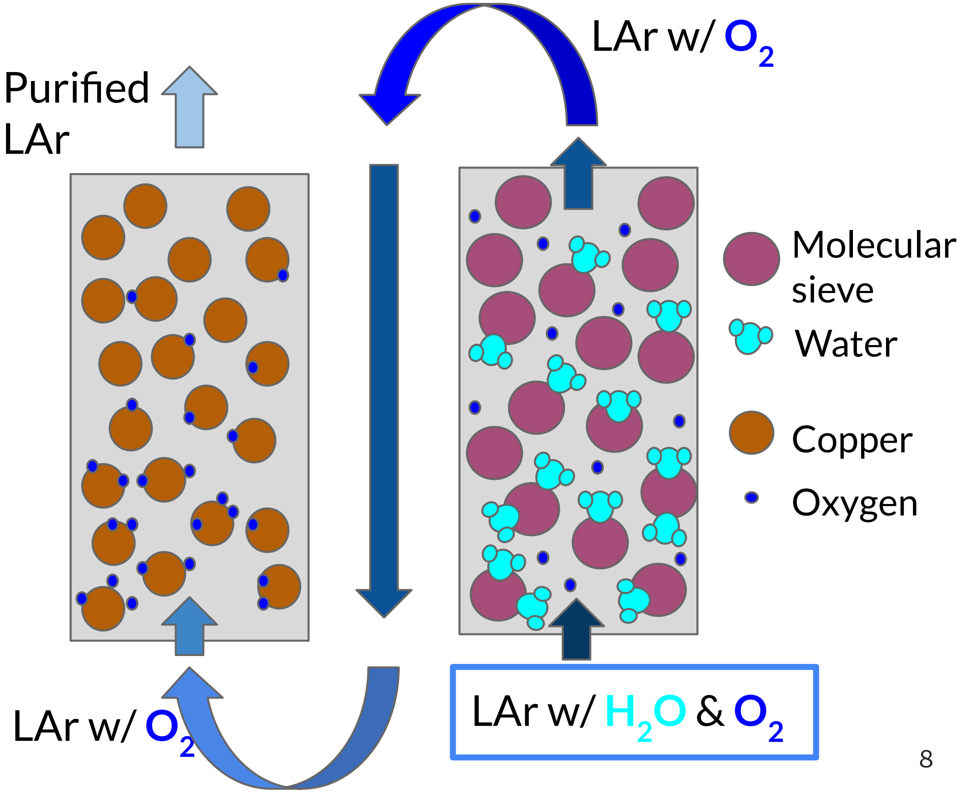


LAr can be purified with commercially available dispersed copper and molecular sieves

Water removal: Adsorption on molecular sieve with pore size of 4 Angstrom

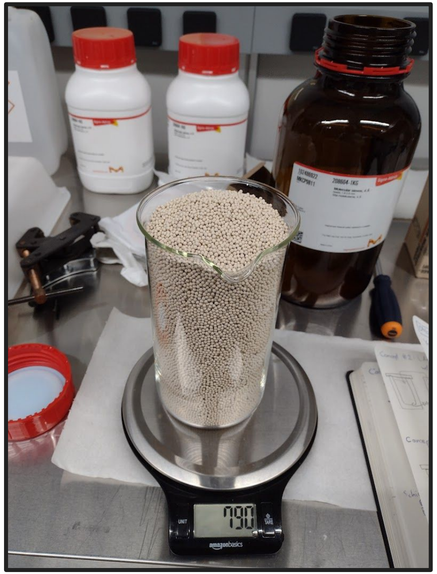
Oxygen removal: Chemical binding to copper via
$2Cu + O_2 \rightarrow 2CuO$

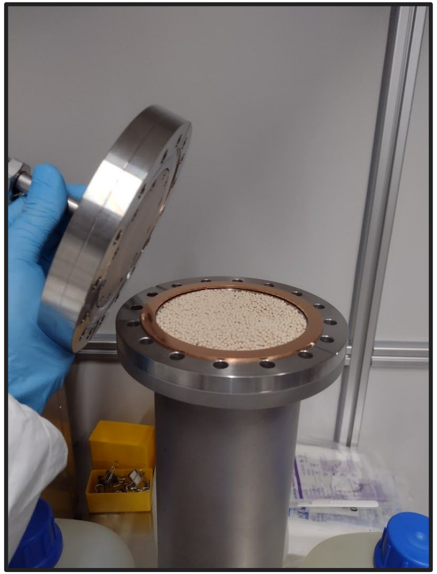
Some nitrogen is retained as well

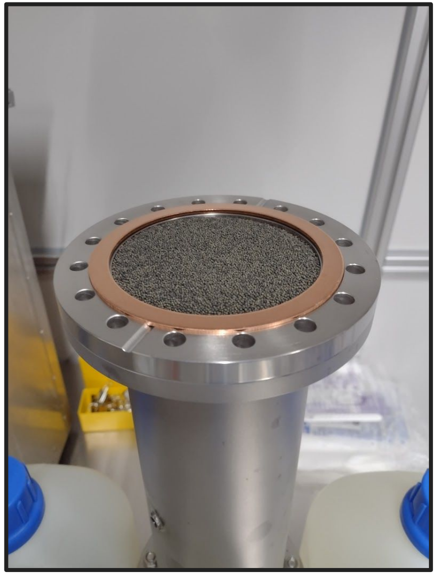


Filling of purification cartridges with molecular sieve and dispersed copper

Filled 2005 g of molecular sieve 4 A, and 2341 g of copper catalyst into stainless steel CF100 cylinders



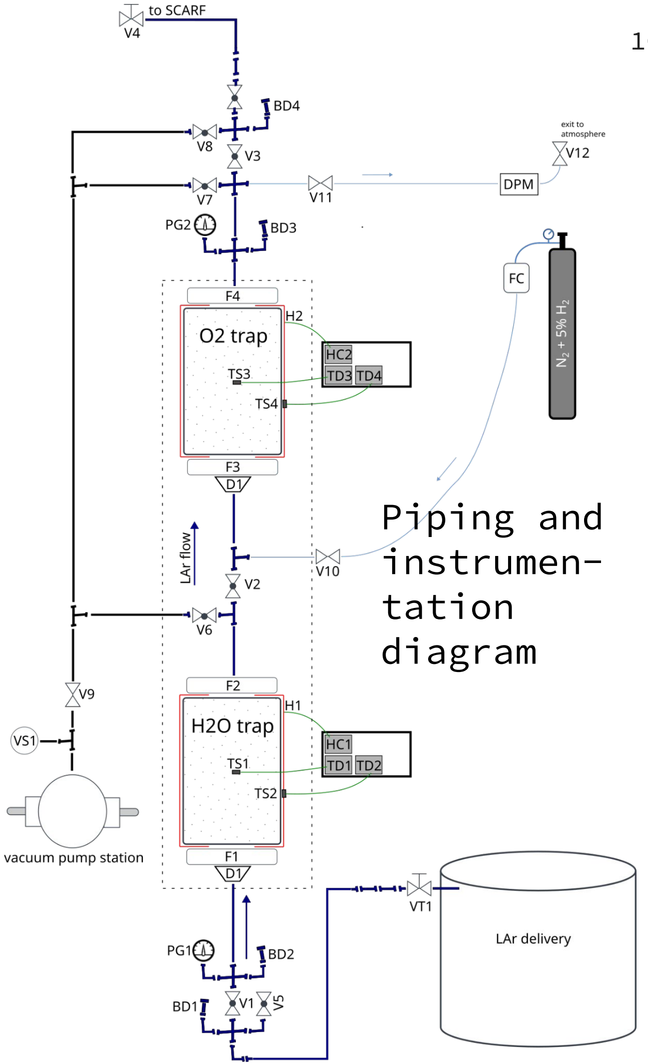




Piping and instrumentation

Purification setup w/o thermal insulation

CAD drawing of the purifier



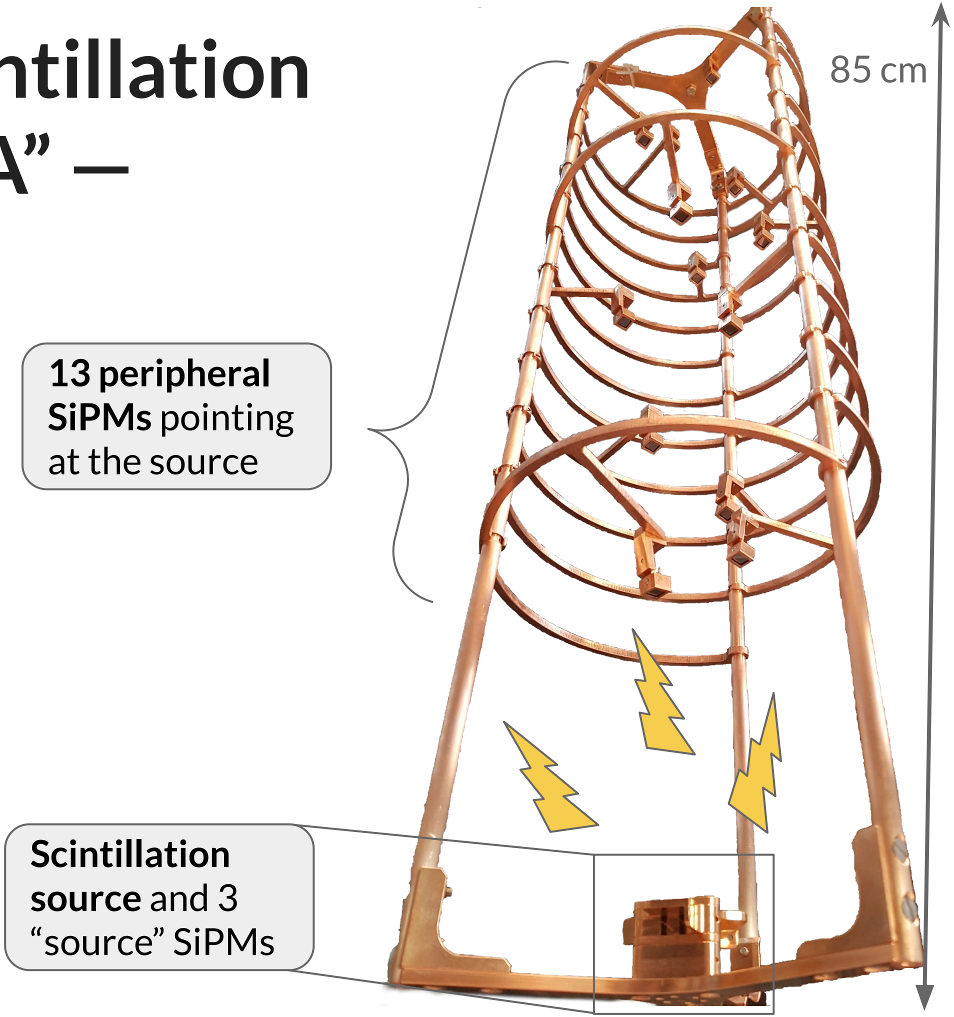
Piping and instrumentation diagram

Determination of the scintillation parameters with “LLAMA” — a triggered SiPM array

The LEGEND Liquid Argon Monitoring Apparatus (LLAMA) operates **completely submerged in LAr**, and features[3]

- ^{241}Am scintillation light source
- 3 “source” SiPMs providing the **trigger** and **p.e. yield**
- 13 peripheral SiPMs providing **time structure** and **propagation information**

[3] [M. Schwarz et al., *ANIMMA 2021* (July 2021)]



LLAMA is operating in SCARF, a 1 ton LAr test stand

Located in shallow underground laboratory (depth of 10 m w.e.)

Muon flux reduced by factor of 4

Able to house LLAMA for various LAr scintillation studies, e.g. XeDLAr at LIDINE 2021: C. Vogl et al 2022 JINST 17 C01031

Alternative setup: Up to two germanium detectors + fibers w/ SiPMs

Application #1: Filling of “SCARF” cryostat

First performance test when filling 1 ton of LAr into empty cryostat “SCARF”

Initial impurities in delivered 600 L LAr tanks (vendor certificates):

1) $[O_2]$ = 0.5 μL/L
   $[N_2]$ = 0.5 μL/L
   $[H_2O]$ < 3 μL/L
2) $[O_2]$ = 0.2 μL/L
   $[N_2]$ = 0.3 μL/L
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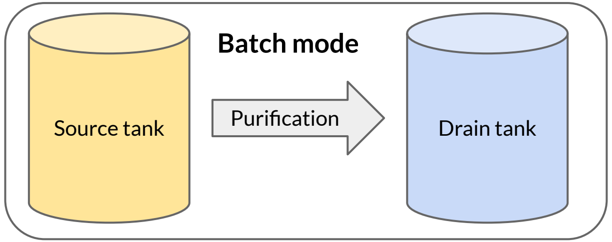

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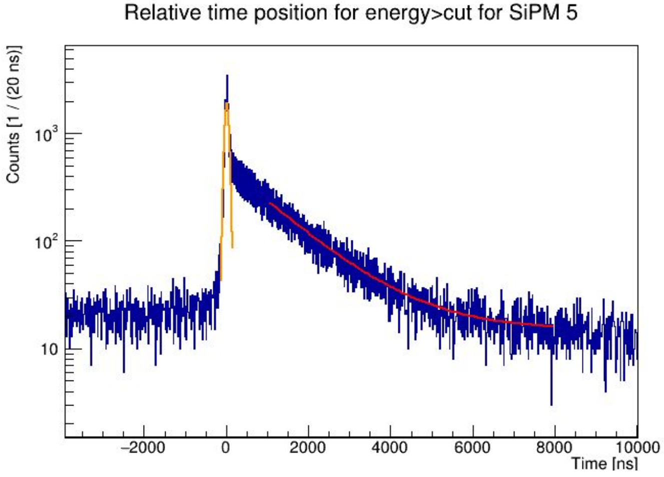
Measured triplet lifetime after purification:
1.31 ± 0.02 µs

Corresponds to[1, 2]
< 0.01 µL/L O_2-equiv.
and < 0.8 µL/L N_2



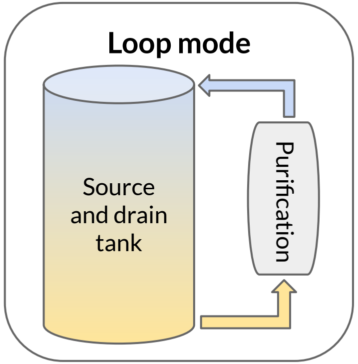
Successful purification of 1 ton of LAr!

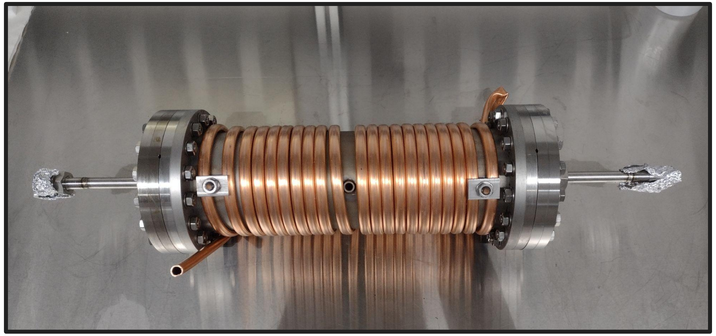
[1] [R Acciarri et al. 2010 JINST 5 P05003]
[2] [R Acciarri et al. 2010 JINST 5 P06003]

Application #2: Loop-mode purification of contaminated LAr

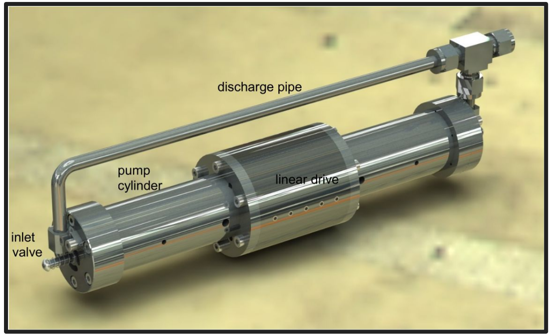
Triplet lifetime in SCARF reduced to 1.00 ± 0.02 µs due to contamination with air

Upgrade of setup to accommodate loop-mode purification: Install **active LN_2 cooling** and use **submerged cryogenic liquid pump**





Purification cartridge wrapped in copper coil for active LN_2 cooling

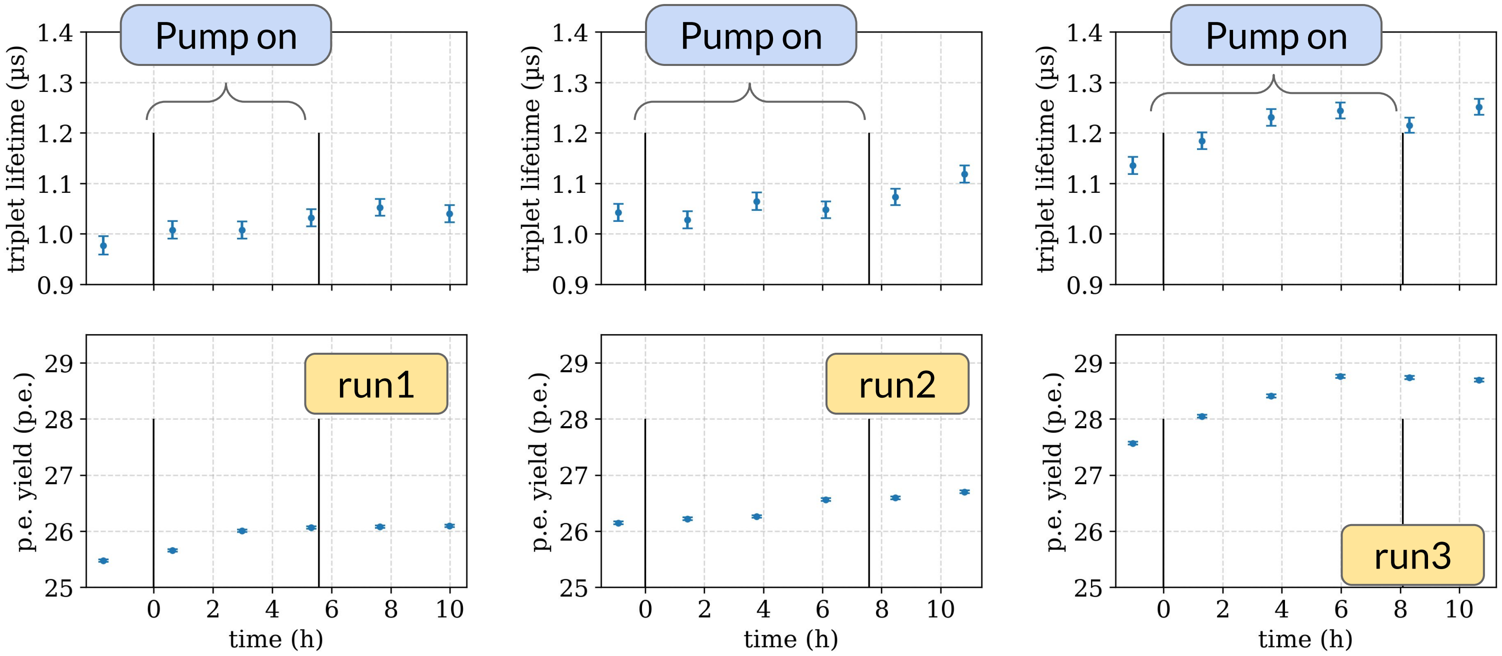


Cryogenic submerged LAr pump

Single-day purification runs in loop mode

3 purification runs at different pumping speeds between 225 L/h to 450 L/h

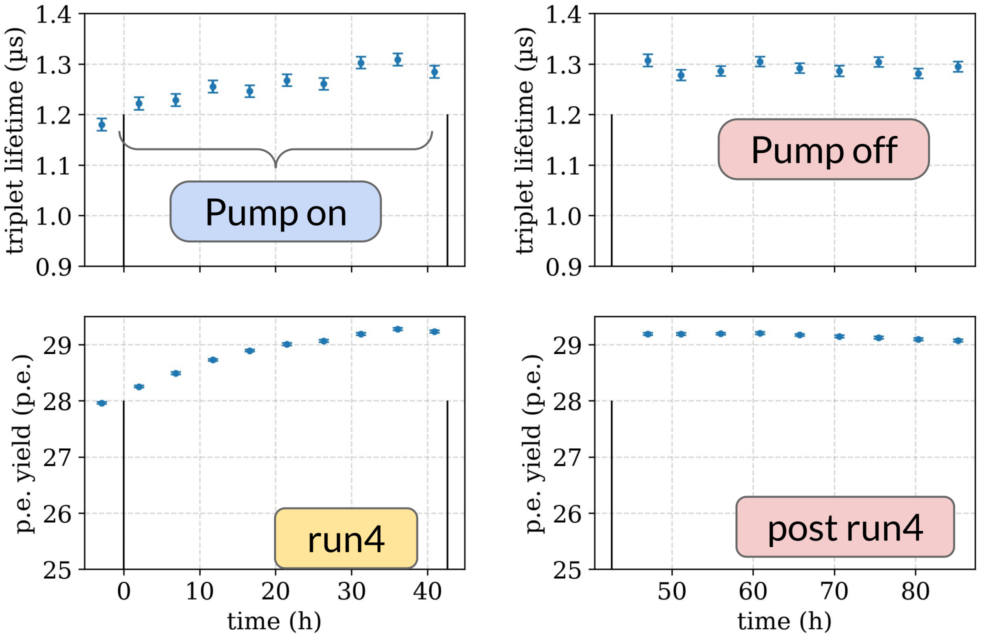
Increase from 0.98 ± 0.02 μs to 1.25 ± 0.02 μs after ~ 9 volume exchanges



42 h purification run and aftermath

Long ~ 42 hour continuous purification run (10.4 volumes exchanged)

Increase back to ~ 1.3 μs! Purification complete!



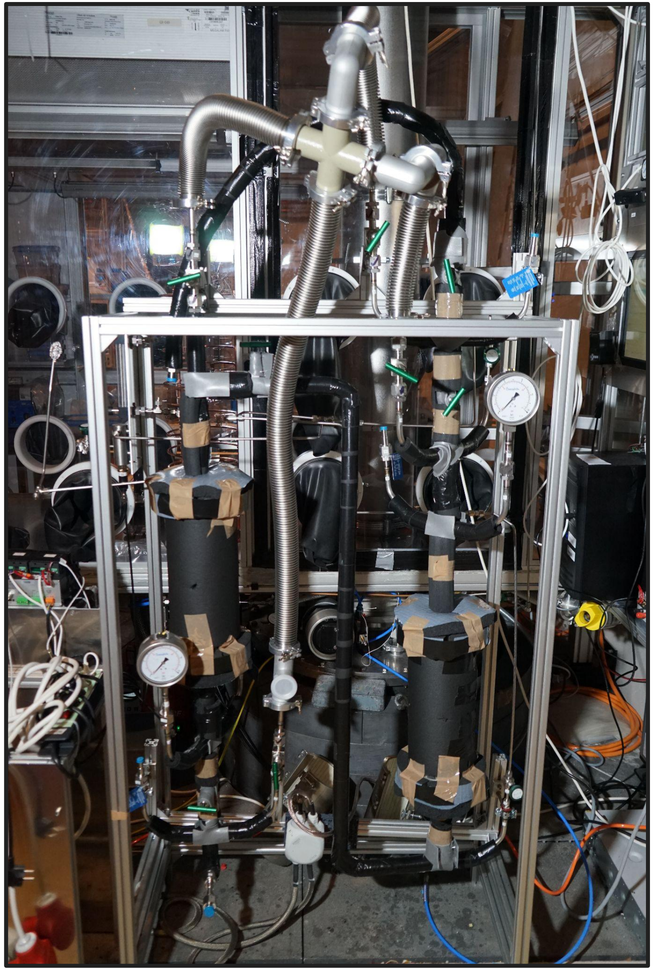
However, **photo-electron yield not completely recovered!**
29.21 ± 0.02 p.e. after purification,
34.56 ± 0.01 p.e. before contamination.

Origin unclear (as of yet)

[1] [R Acciarri et al. 2010 JINST 5 P05003]
[2] [R Acciarri et al. 2010 JINST 5 P06003]

Conclusions

- High chemical purity is paramount for noble liquid detectors
  → on-site purification necessary
- We constructed a **LAr purification system** using dispersed **copper** and 4 A **molecular sieve**
- It purifies in **batch and loop mode** in liquid phase **to sub-ppm levels** (triplet lifetime ~ 1.3 μs)
- Outlook: Build model (improve understanding) of purification columns and provide vacuum insulation to reduce boil-off.



Backup

Regeneration procedure

Molecular sieve saturated with water:
Heat to ~ 250 °C and apply vacuum for a few hours.

Oxidized copper:
Heat to 200–250 °C and flush with hydrogen in low concentrations (≤5 %) in an inert carrier gas (e.g. nitrogen or argon) until some 100 cartridge volumes have been exchanged. Finally, apply vacuum.

Radon emanation from adsorbers

- O_2 adsorber (CU-0226 S 14 X 28): (0.43 ± 0.03) Bq/kg
- H_2O adsorber (MS 5A): (1.32 ± 0.04) Bq/kg

Loop mode purification details

Run #	Frequency	Volume flow	Duration	Total volume exchanged	# Cryostat volumes exchanged
1	3 Hz	450 L/h	5.6 h	2510 L	3.5
2	2 Hz	300 L/h	7.6 h	2280 L	3.2
3	1.5 Hz	225 L/h	8.1 h	1820 L	2.5
4a	1	150 L/h	28.5 h	4275 L	6.0
4b	1.5 Hz	225 L/h	14.1 h	3173 L	4.4
Sum	—	—	63.9	14058 L	19.6

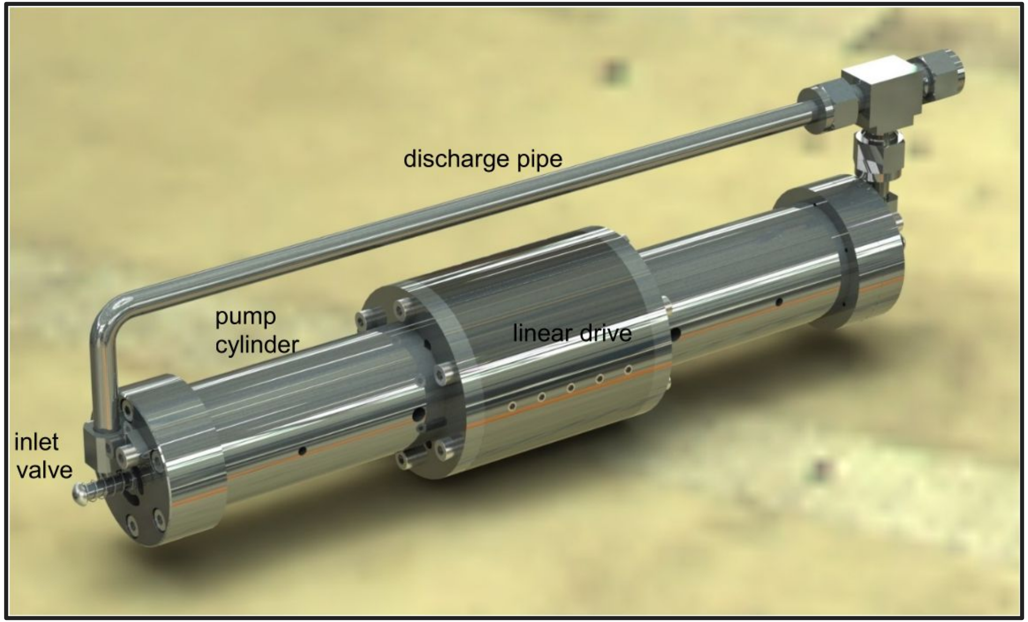
The LAr pump

Procured from the institute of air handling and refrigeration (IKL) Dresden

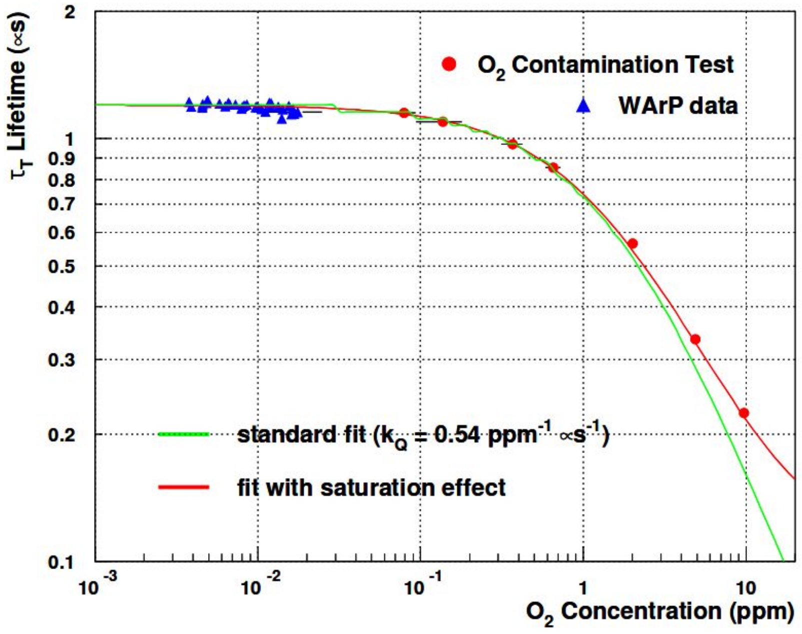
Linear drive piston pump.

Geometric flow rate of 150 L/h @ 1 Hz operating frequency

Working frequency range: 0.5 Hz to 3.3 Hz, best performance 1 to 2 Hz



Contaminant removal in concentration (oxygen)



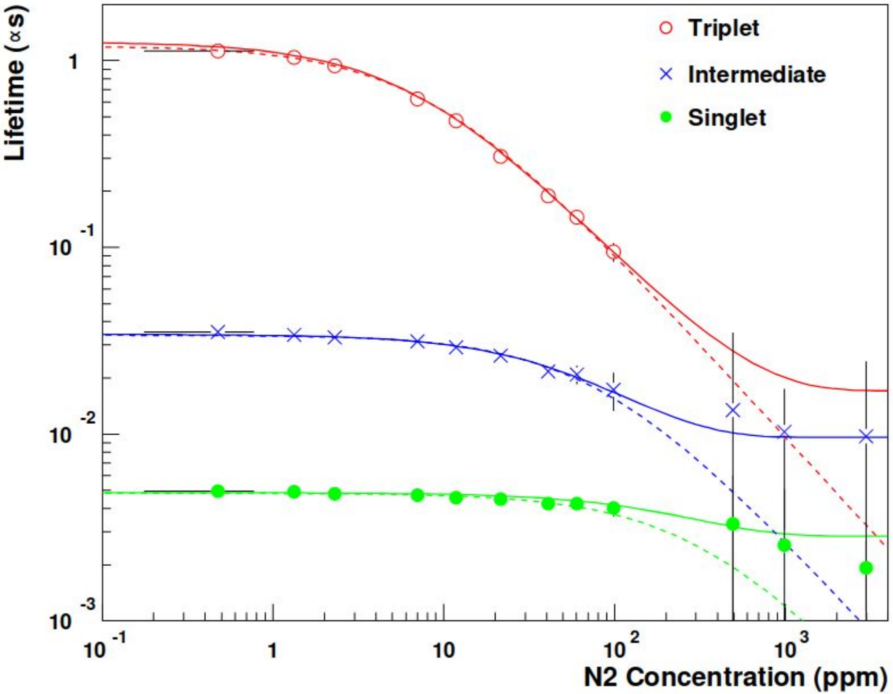
[R Acciarri et al 2010 JINST 5 P05003, fig. 6]

Assume that the contamination only stems from O_2 (not true, but provides a nice estimate)

Then we went from ~ 0.3 ppm O_2 equivalent before purification to < 0.01 ppm afterwards!

In loop mode!

Triplet lifetime and nitrogen concentration



[R Acciarri et al 2010
JINST 5 P06003, fig. 10]