Technical University of Munich

Bundesministerium für Bildung und Forschung

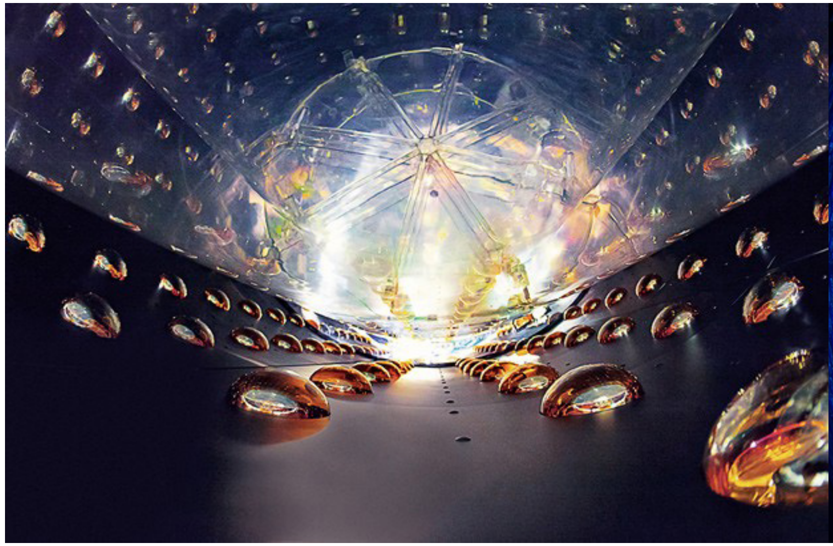
ORIGINS Excellence Cluster

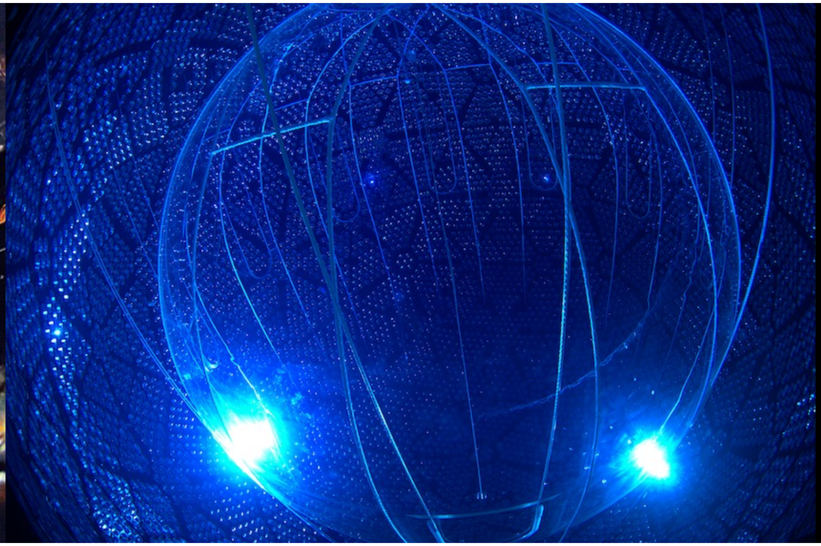
SFB 1258 Neutrinos Dark Matter Messengers

Novel Slow LAB-based Scintillators with C/S Separation with and without Metal Doping

Hans Th. J. Steiger[1, 2, 3] & Manuel Böhles[2,3]

[1]Technische Universität München, [2]Cluster of Excellence PRISMA+, [3]Johannes Gutenberg-Universität Mainz





JGU Johannes Gutenberg Universität Mainz

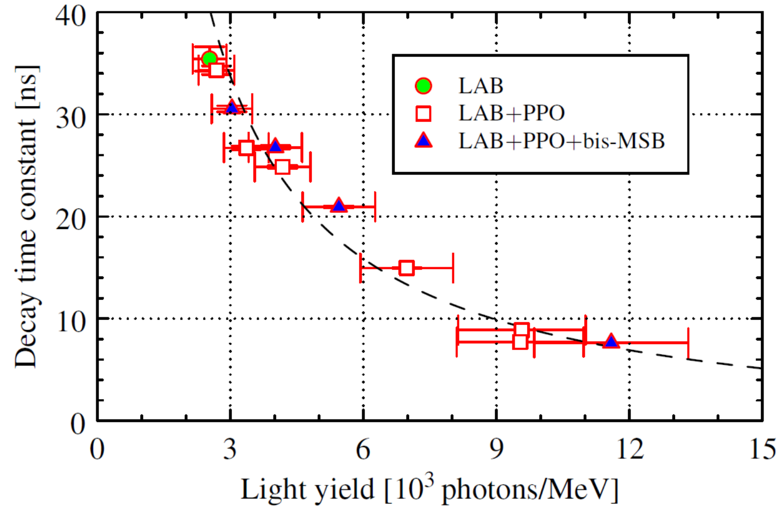
DFG

PRISMA+

Cherenkov and Scintillation Light Separation: How to get the LS slow?

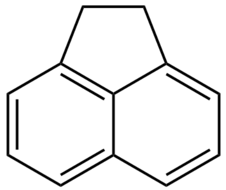
Three ways to get the scintillation emission slow:

- **Lower the fluor concentration**
  [Guo, Z. et al. – arXiv:1708.07781]
  - Low light yields
  - Limited PSD capabilities
  - Excellent transparency in case of LAB

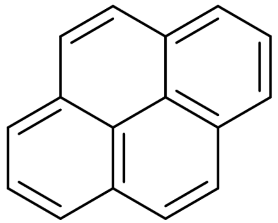


LAB/PPO-mixtures: Low PPO concentration leads to low LYs and no PSD!

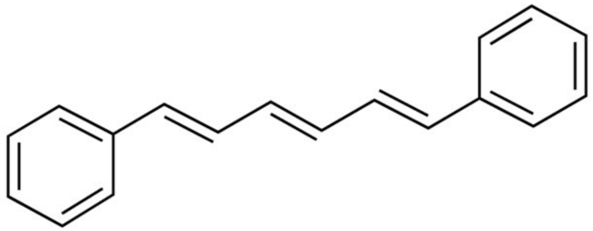
- **Slow fluors**
  [Biller, S. et al. – arXiv:2001.10825]
  - Expensive substances?
  - Toxic or cancerogenic compounds?
  - Slow scintillation comes often at the cost of losses in LY
  - Often emission wavelength maximum deep in the UV-region!
  - PSD not demonstated! --> test at INFN-LNL (Nov. 24)



Acenaphthene ($C_{12}H_{10}$)



Pyrene ($C_{16}H_{10}$)



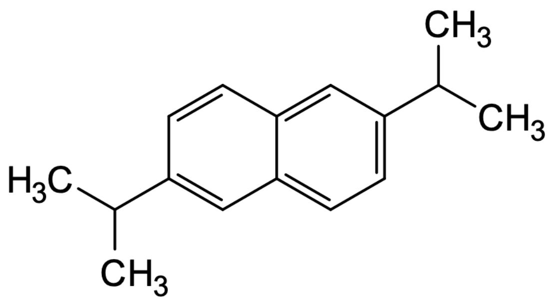
1,6-Diphenyl-1,3,5-hexatriene (DPH, $C_{18}H_{16}$)

- **Blended or multi-solvent cocktails**
  [Steiger, Hans Th. J. et al. – arXiv:2405.01100, 2024]
  - LY typically: 10^4 Ph./MeV, $\tau_1 = 12\text{–}45$ ns (adjustable)
  - LY and PSD can be enhanced with a carefully balanced selection of solvent and co-solvent
  - Cheap and easy to clean co-solvents

Prepared for submission to JINST

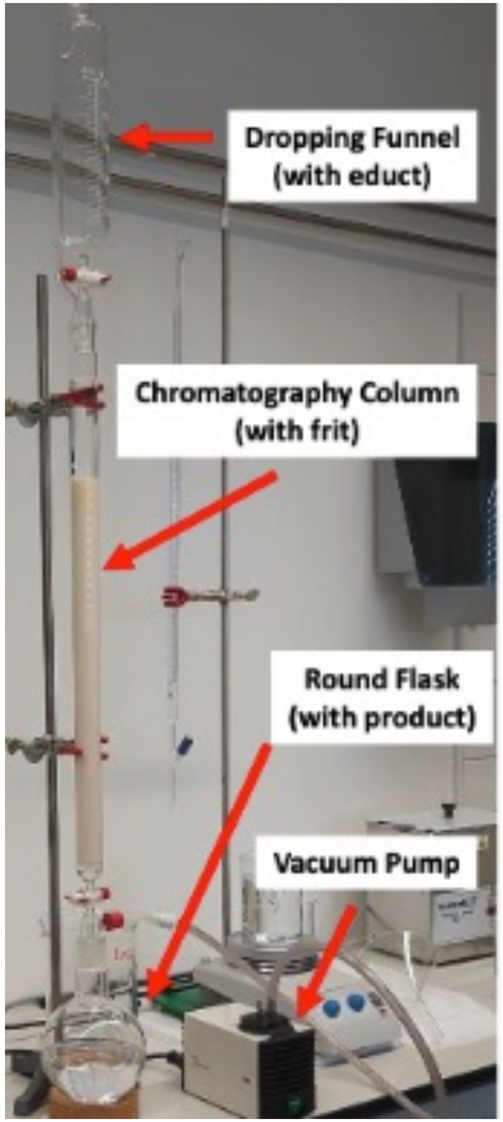
Development of a Bi-solvent Liquid Scintillator with Slow Light Emission

Hans Th. J. Steiger,[a,b,c,1] Matthias Raphael Stock,[c] Manuel Böhles,[a,b] Sarah Braun,[c] Edward J. Callaghan,[d,e] David Dörflinger,[c] Ulrike Fahrendholz,[c] Gabriel D. Orebi Gann,[d,e] T. Kaptanoglu,[d,e] Lennard Kayser,[c] Florian Kübelbäck,[c] Meishu Lu,[c] Lothar Oberauer,[c] Korbinian Stangler,[c] Michael Wurm,[a,b] Dorina Zundel[a,b]

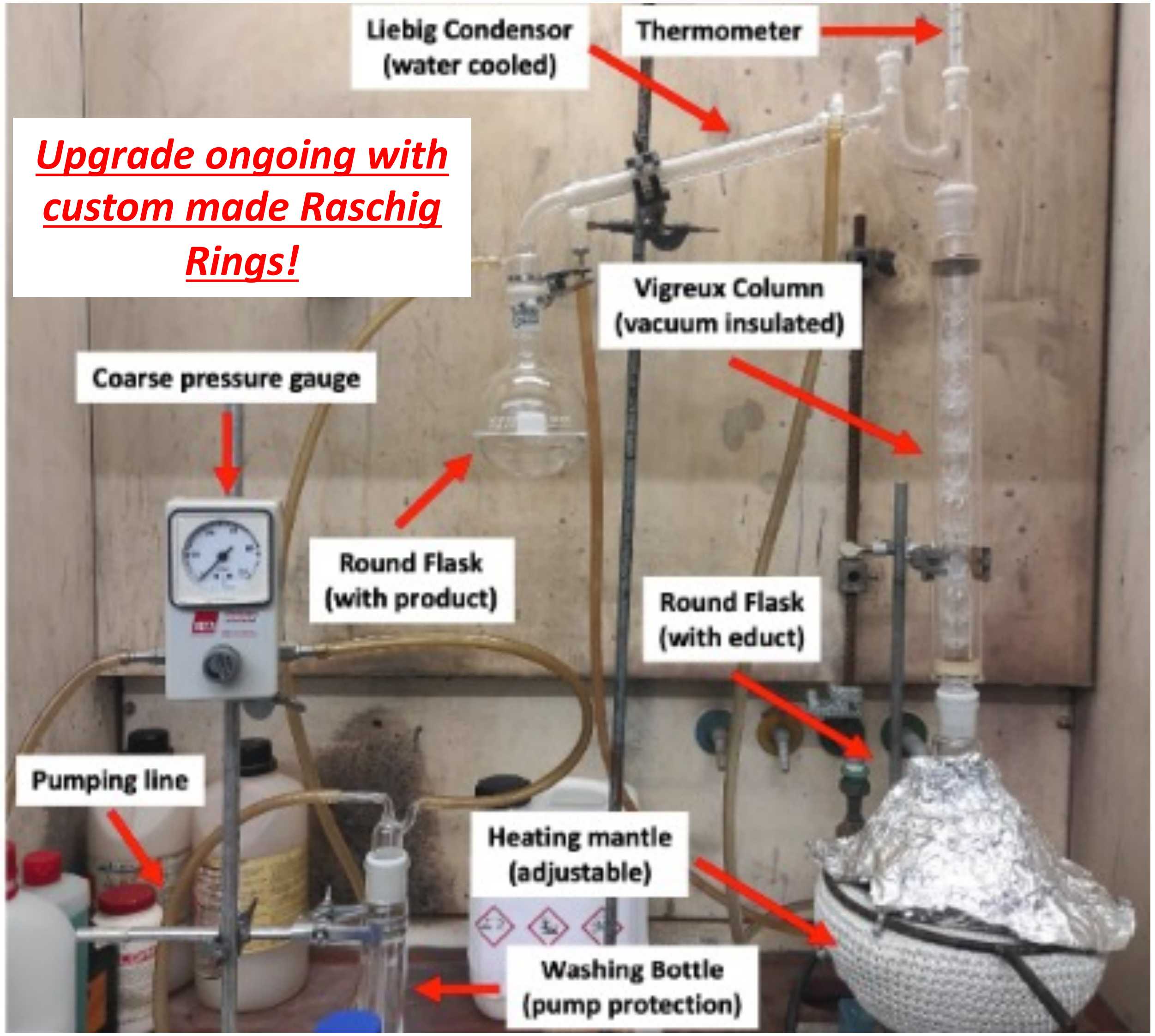


2,6-Diisopropylnaphthalene (DIPN, $C_{16}H_{20}$)

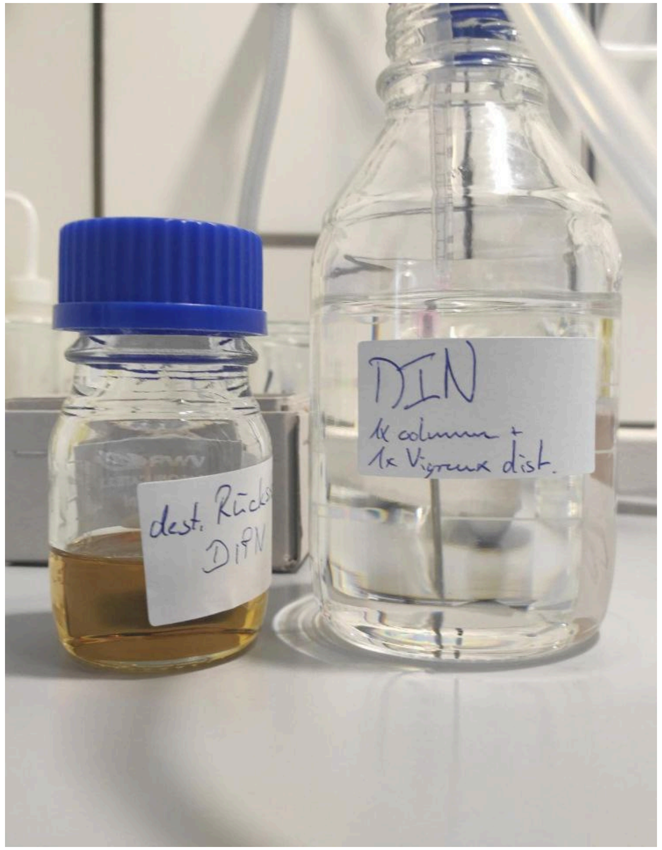
Purification of DIN (Ruetasolv Di-S)



Long and thin Al_2O_3 Colum
(basic alumina of activity Super-I



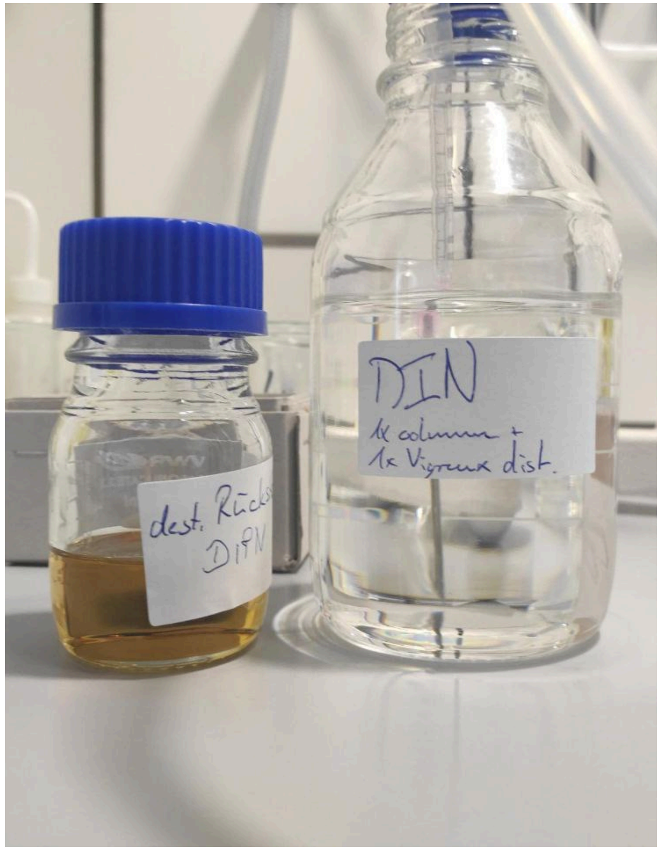
Low temperature vacuum Distillation + Vigreux Column
(product completely cooled down before exposed to air)



Residue and highly-pure DIN
after Distillation

Additional filtration:
200 nm + 50 nm + 20 nm
(Reduction of scattering!)

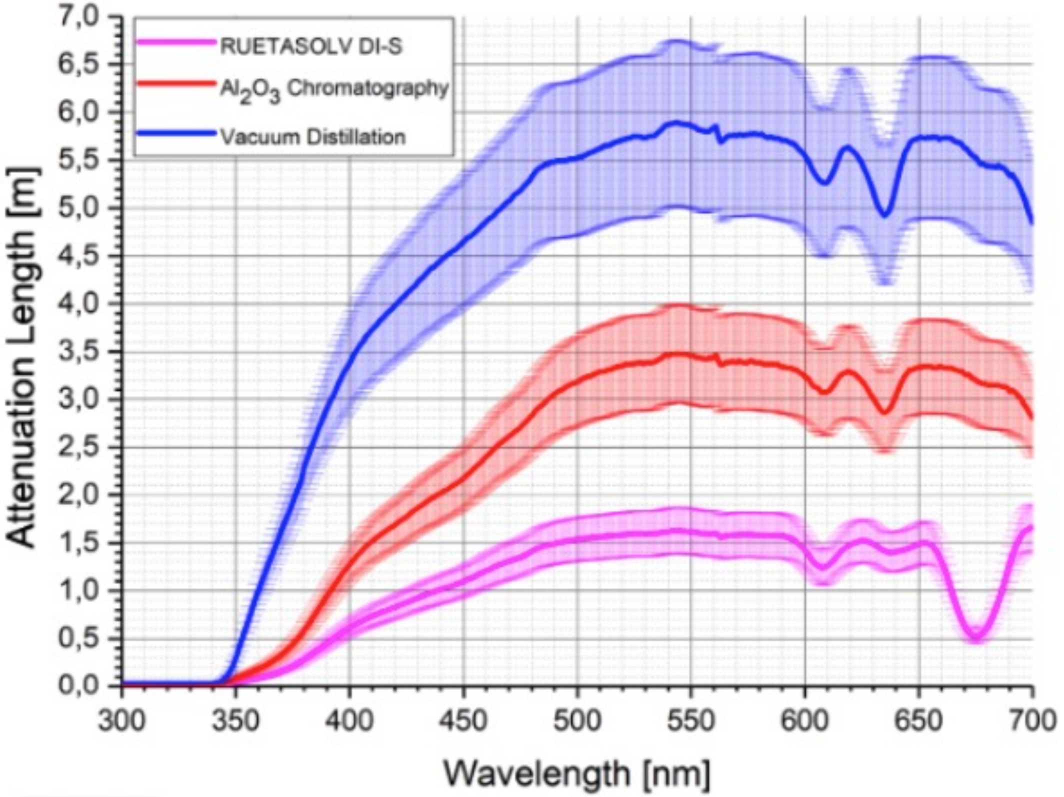
Purification of DIN (Ruetasolv Di-S): What we reached by now



Residue and highly-pure DIN
after Distillation

Attenuation Length with
UV/Vis-Spectrometer:

- Perkin Elmer Lambda 850+
- 10 cm cuvette
- large uncertainties, scattering, …



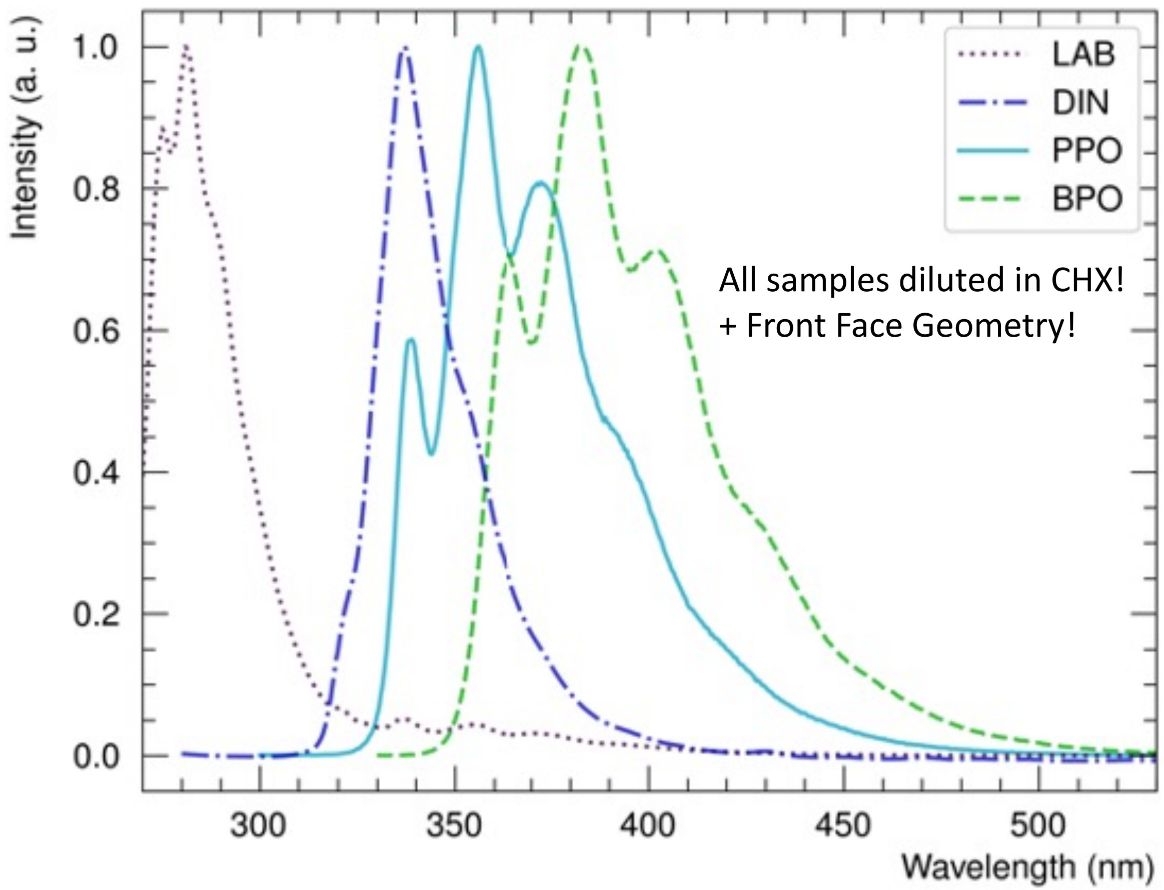
Attenuation Length after purification
(Improvement by distillation larger than by alumina column)

Calculation of the attenuation length of LAB/DIN mixtures
with JUNO-LAB (Daya Bay) with measured att. length:
Λ_{LAB} (430 nm) = (28.07 ± 2.94) m
(Measurement: S. Franke, PhD Thesis, PALM)

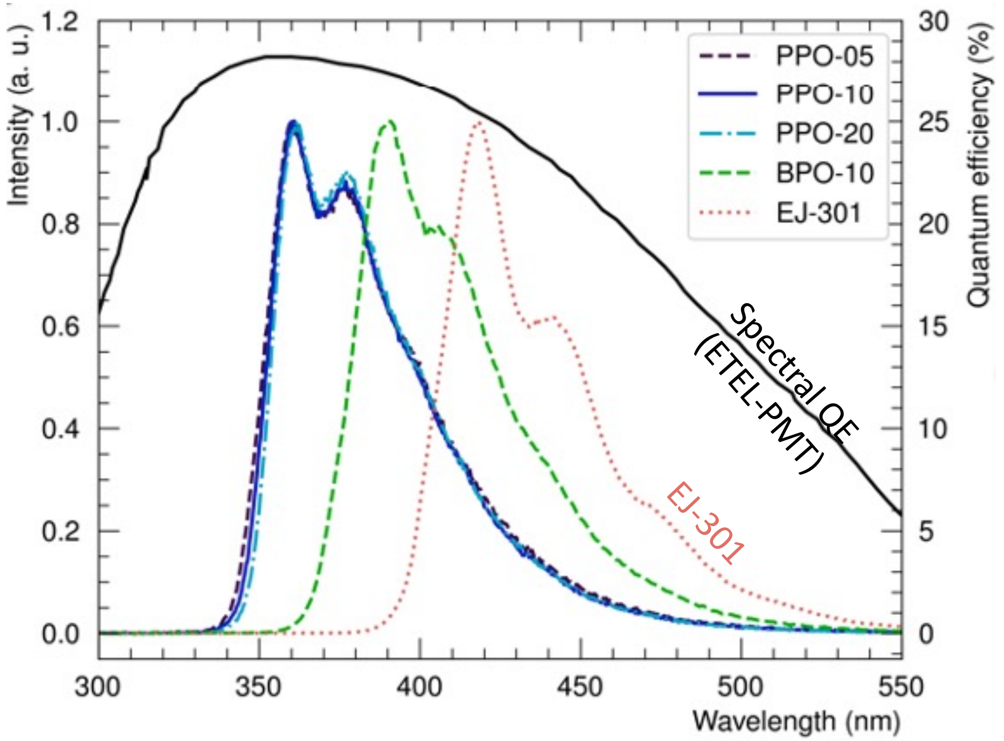
$$\frac{1}{\Lambda} = \sum_i \frac{1}{\Lambda_i} \qquad \begin{aligned} \Lambda_{90/10}(430\,\mathrm{nm}) &= (17.5 \pm 2.3)\,\mathrm{m} \\ \Lambda_{80/20}(430\,\mathrm{nm}) &= (13.9 \pm 1.6)\,\mathrm{m} \end{aligned}$$

Calculation in agreement
with UV/Vis direct
measurements: > 13.5 m.

Selection of Wavelength Shifters – Some Scintillator Samples under Investigation



Substance	Chemical Formula	Conc. in CHX	Exc. [nm]	Abs. Max. [nm]	Em. Max. [nm]
LAB	$(C_6H_5) - C_nH_{2n+1}$ with n = 10 – 13	10 ml/l	255 ± 1	260 [18]	281 ± 1
DIN	$C_{16}H_{20}$	10 ml/l	255 ± 1	279 [18]	337 ± 1
PPO	$C_{15}H_{11}NO$	40 mg/l	290 ± 1	303 [18]	356 ± 1
BPO	$C_{21}H_{15}NO$	100 mg/l	320 ± 1	320 [18]	383 ± 1

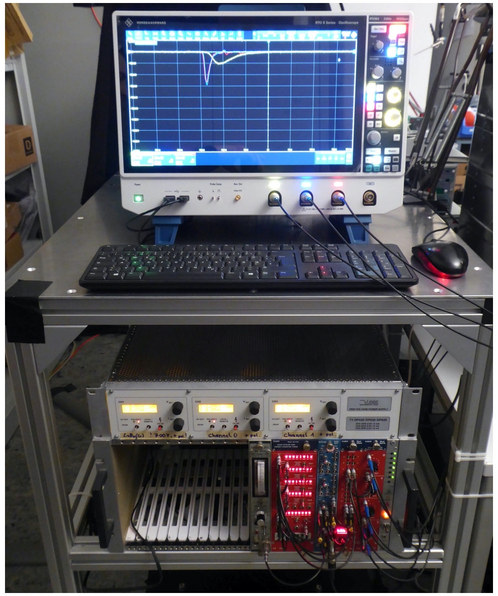


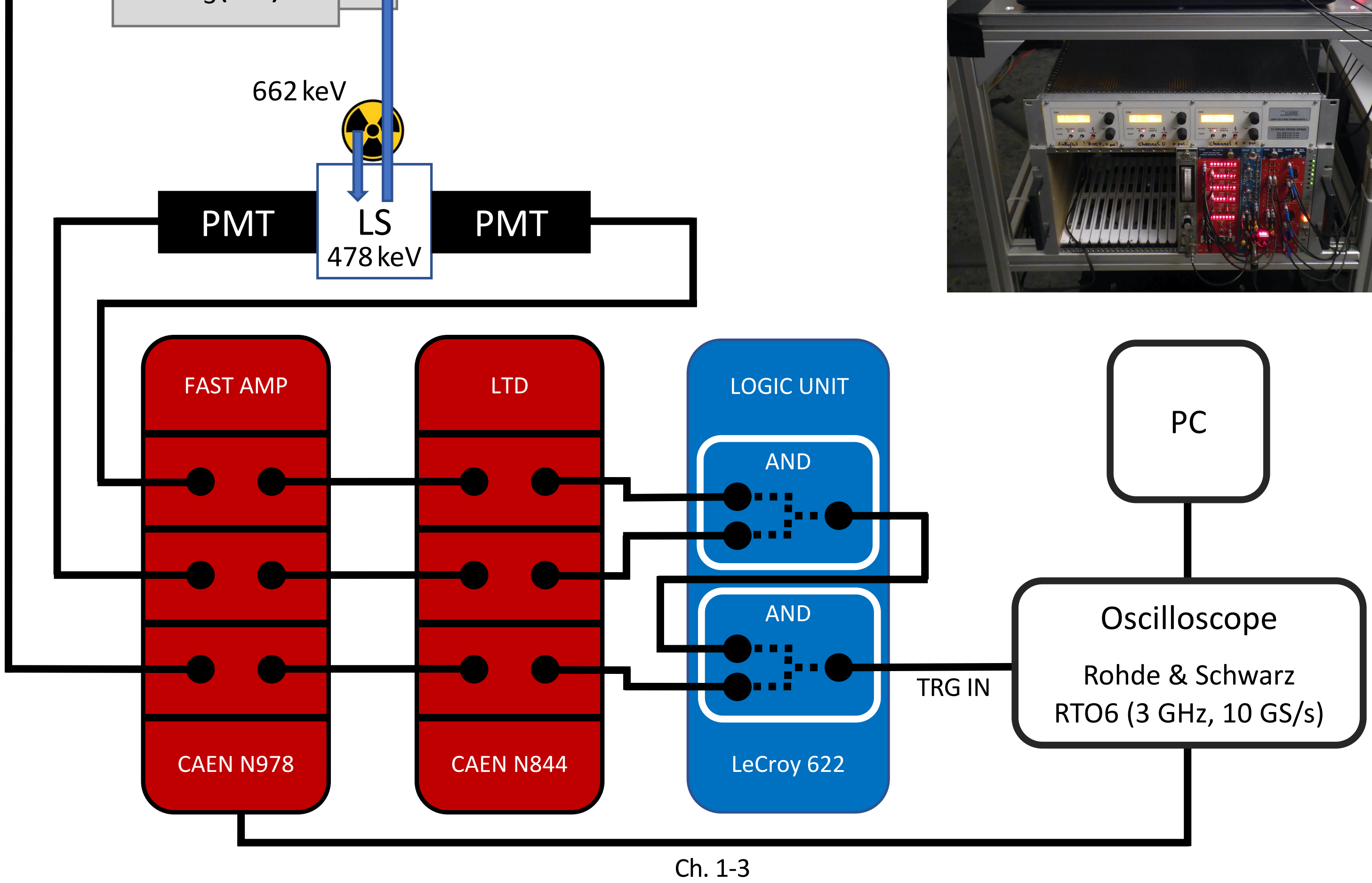
Sample Name	Solvent	Co-Solvent	Fluor	Fluor Concentration
PPO-05	90 % LAB	10 % DIN	PPO	0.5 g/l
PPO-10	90 % LAB	10 % DIN	PPO	1.0 g/l
PPO-20	90 % LAB	10 % DIN	PPO	2.0 g/l
BPO-05	90 % LAB	10 % DIN	BPO	0.5 g/l
BPO-10	90 % LAB	10 % DIN	BPO	1.0 g/l
BPO-20	90 % LAB	10 % DIN	BPO	2.0 g/l
PPO-05-20	80 % LAB	20 % DIN	PPO	0.5 g/l
PPO-10-20	80 % LAB	20 % DIN	PPO	1.0 g/l
PPO-20-20	80 % LAB	20 % DIN	PPO	2.0 g/l

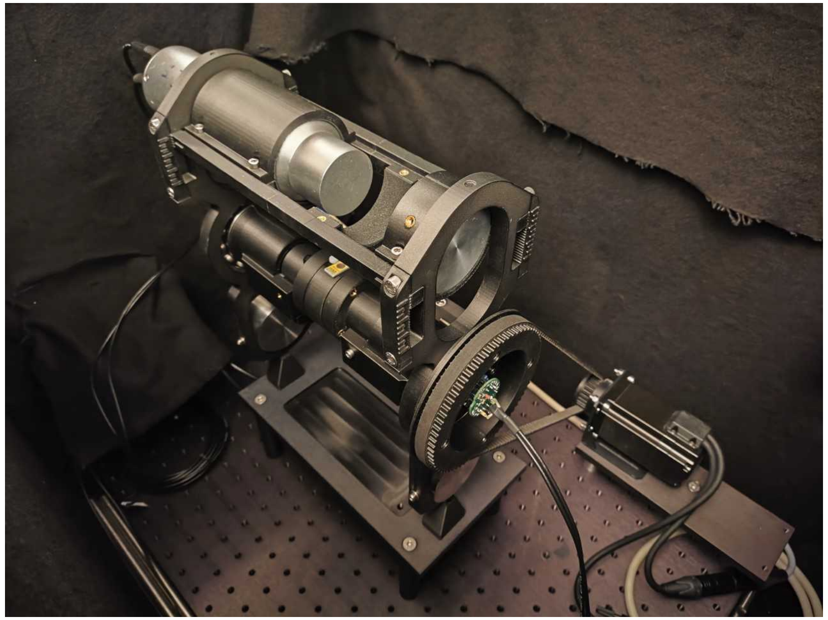
Light Yield and Nonlinearity Study in the Mainz Scintillation LY Setup

Manuel Böhles
(PhD Student, JGU Mainz)

- Solvent Purification
- Loading
- Light Yields
- UV/Vis and Fluorimetry
- …





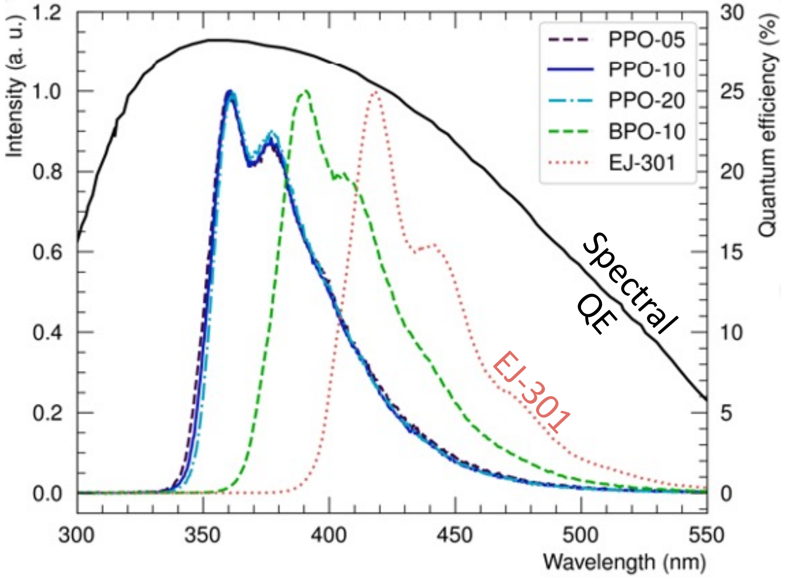


The Mainz LY Setup: Glowrats
(Geared Light yield Observation With Rotating Apperatus for Testing Scintillators)

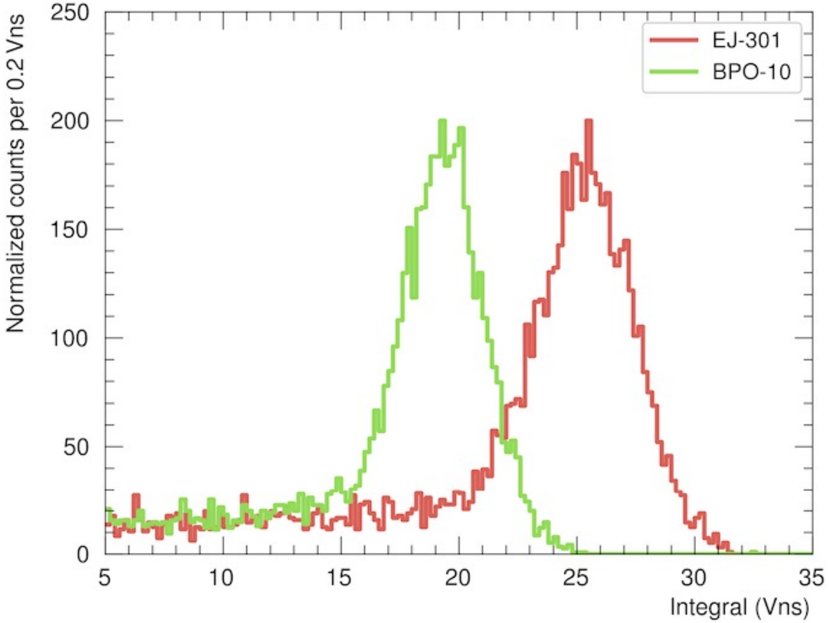
PRiSMA+ JG|U

Light Yield Study in the Mainz Scintillation LY Setup

EJ-301 as standard
(Xylene+Naphtalene + …)



Spectral Emission and spectral QE of the PMT have to be considered!

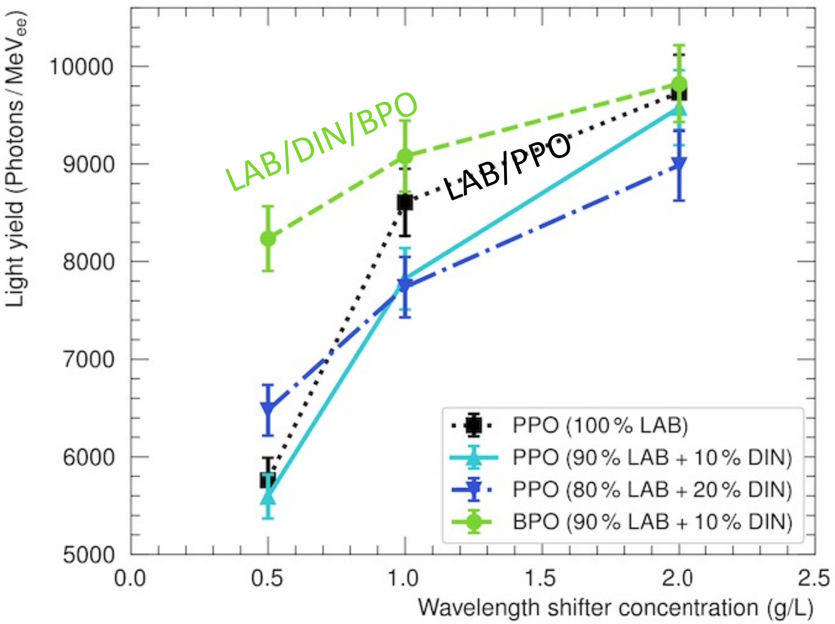


Backscatter Spectra of BPO-10 Sample and EJ-301
(uncorr. LY of EJ-301 larger than BPO-10 sample)

Admixture of DIN:

- With PPO no or nearly no increase in LY compared to pure LAB/PPO!
- **With BPO strong increase in LY especially at low concentrations!**
  - efficient energy transfer
    → low critical concentration of WLS

Note:
All LSs were measured with a **250 ns integration window!**
→ **slow LS would benefit from longer acquisition.**

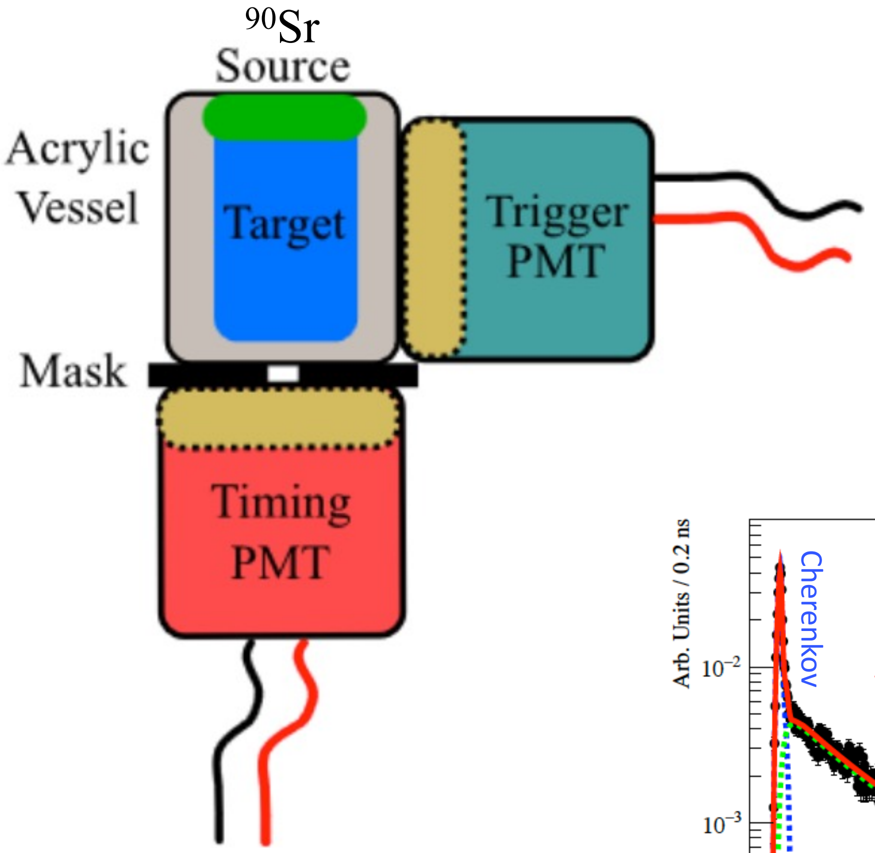


LY with spectral correction of the samples

Sample Name	Rel. LY in %	LY [Ph./MeV]	LY Spectr. Corr. [Ph./MeV]
Anthracene	100	17400	-
EJ-301	78	13572	13572
LAB + 0.5 g/l PPO	39.5 ± 1.6	6877 ± 275	5756 ± 230
LAB + 1.0 g/l PPO	59.1 ± 2.4	10281 ± 412	8605 ± 345
LAB + 2.0 g/l PPO	66.8 ± 2.7	11622 ± 465	9728 ± 390
PPO-05	38.4 ± 1.6	6679 ± 268	5590 ± 225
PPO-10	53.7 ± 2.2	9345 ± 374	7822 ± 314
PPO-20	65.7 ± 2.7	11440 ± 458	9575 ± 384
BPO-05	53.8 ± 2.2	9367 ± 375	8234 ± 330
BPO-10	59.4 ± 2.4	10329 ± 414	9079 ± 364
BPO-20	64.2 ± 2.6	11173 ± 447	9821 ± 393
PPO-05-20	44.5 ± 1.8	7737 ± 310	6476 ± 260
PPO-10-20	53.1 ± 2.2	9244 ± 370	7737 ± 310
PPO-20-20	61.7 ± 2.5	10735 ± 430	8985 ± 360

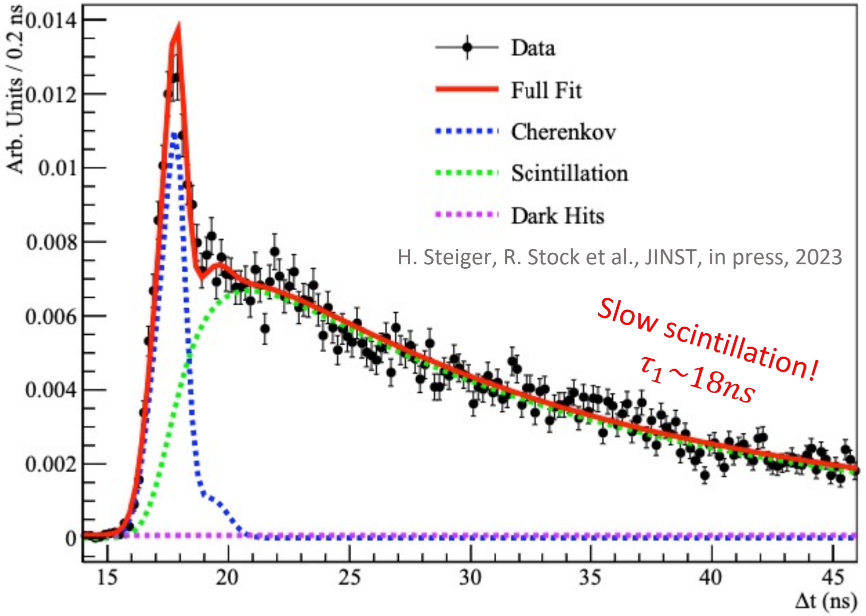
C/S-Separation

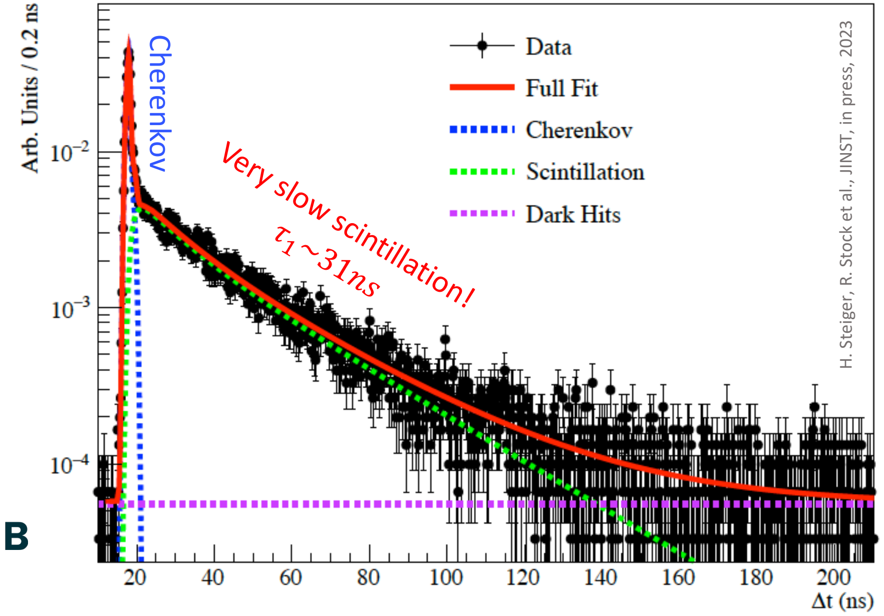
C/S-Separation Setup at UC-Berkeley (G. Orebi-Gann)



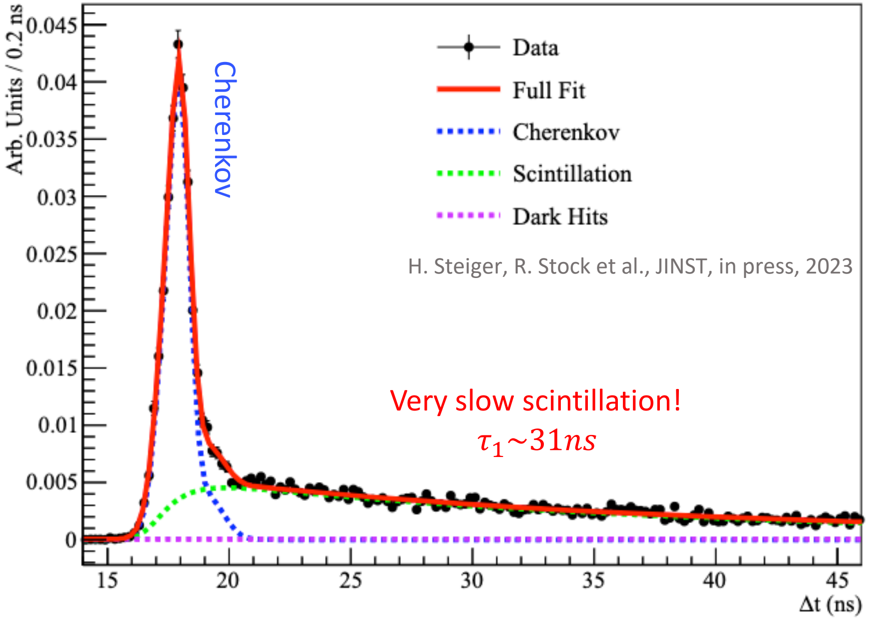
C/S Separation Setup

90% LAB + 10% DIN + 1.0 g/l PPO





90% LAB + 10% DIN + 0.5 g/l PPO



Berkeley UNIVERSITY OF CALIFORNIA
BERKELEY LAB

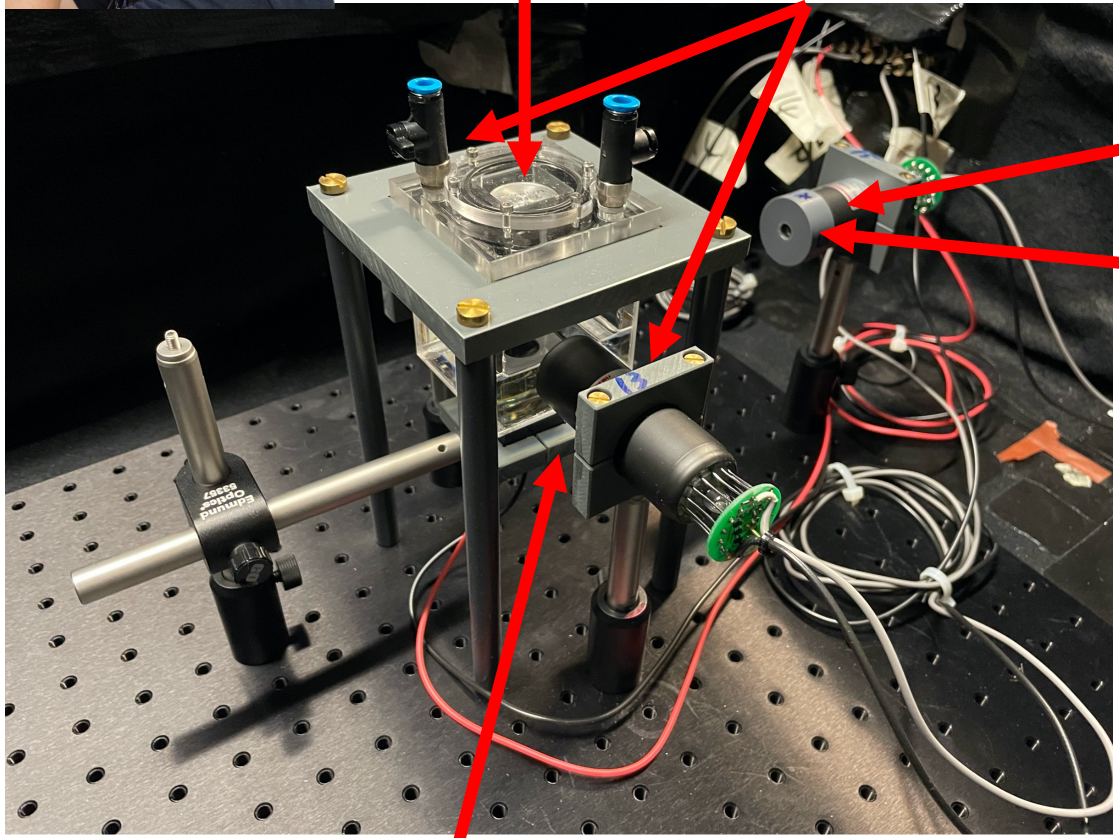
The Munich Scintillation Cherenkov Separator (MSCS) – A new Tool @ TUM

MSc Thesis:
Meishu Lu
(now PhD Thesis in JUNO)

- All PMTs show a TTS well below 70ps.
- ADC sampling rates: 2GS/s – 10 GS/sS by timing only!
- No chromatic filtering or ND filters are necessary!

^{90}Sr or ^{241}Am Source

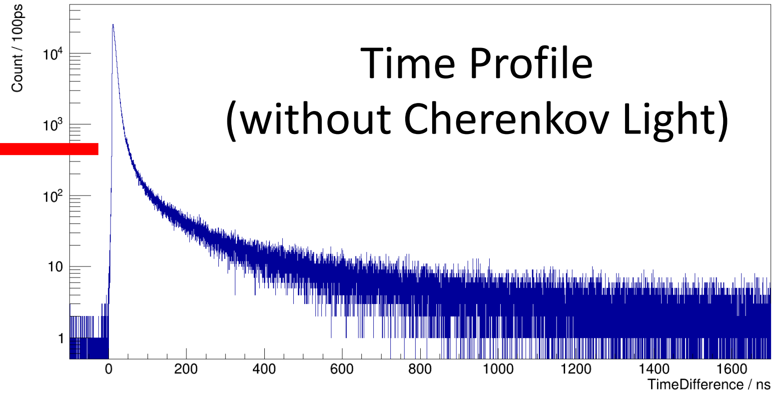
Trigger PMTs
(strongly coupled)

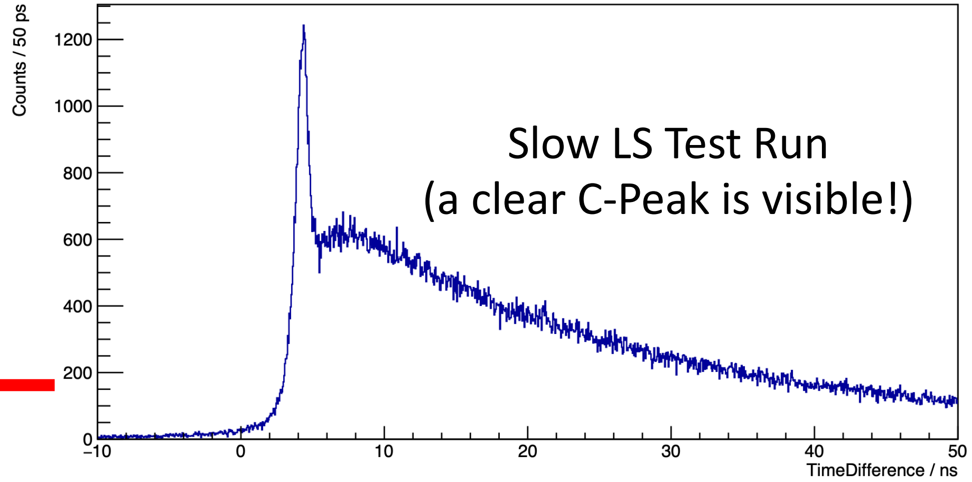


Time Profile PMT

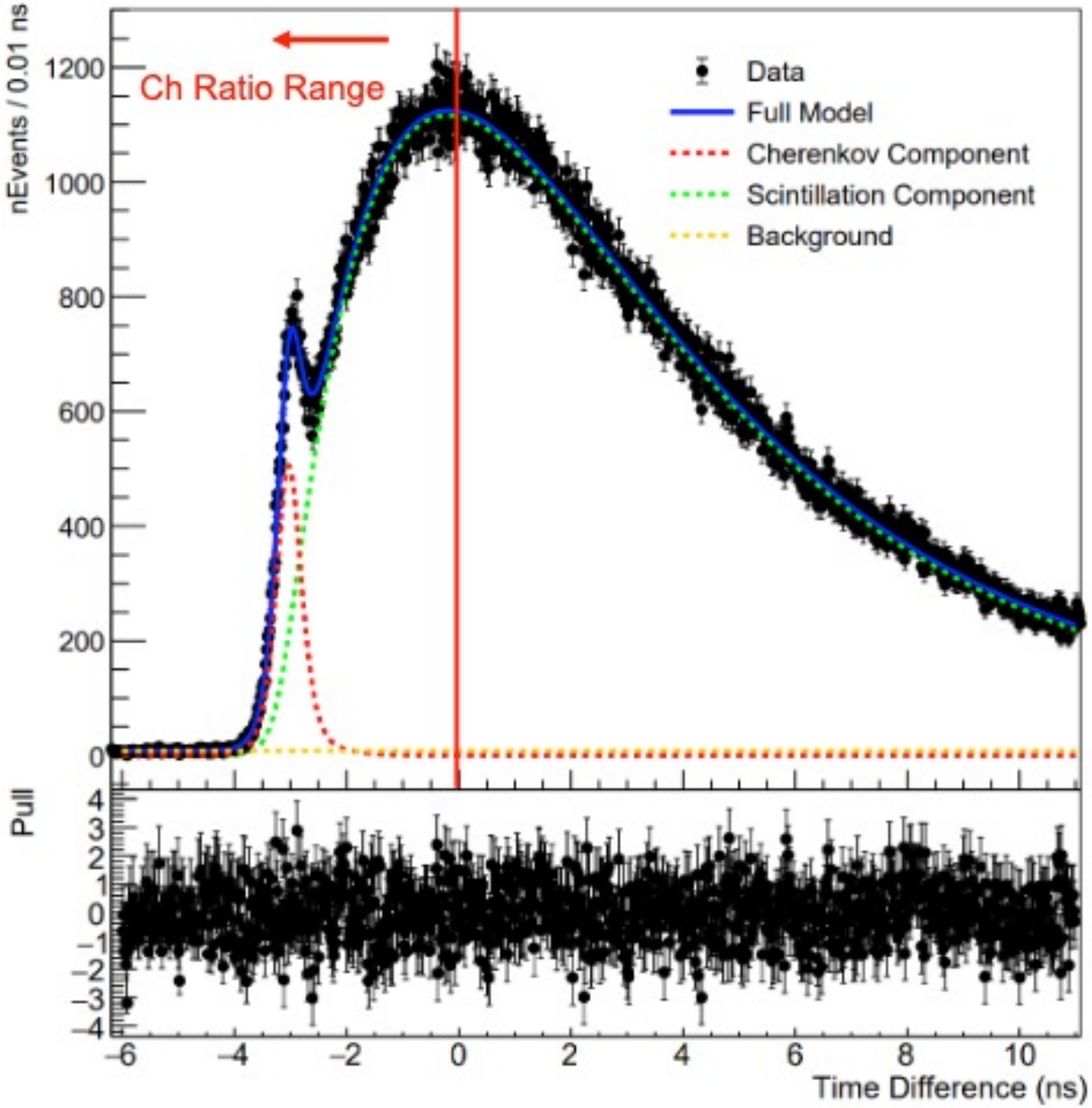
Aperture

C/S - PMT





Separation Study of the JUNO Scintillator

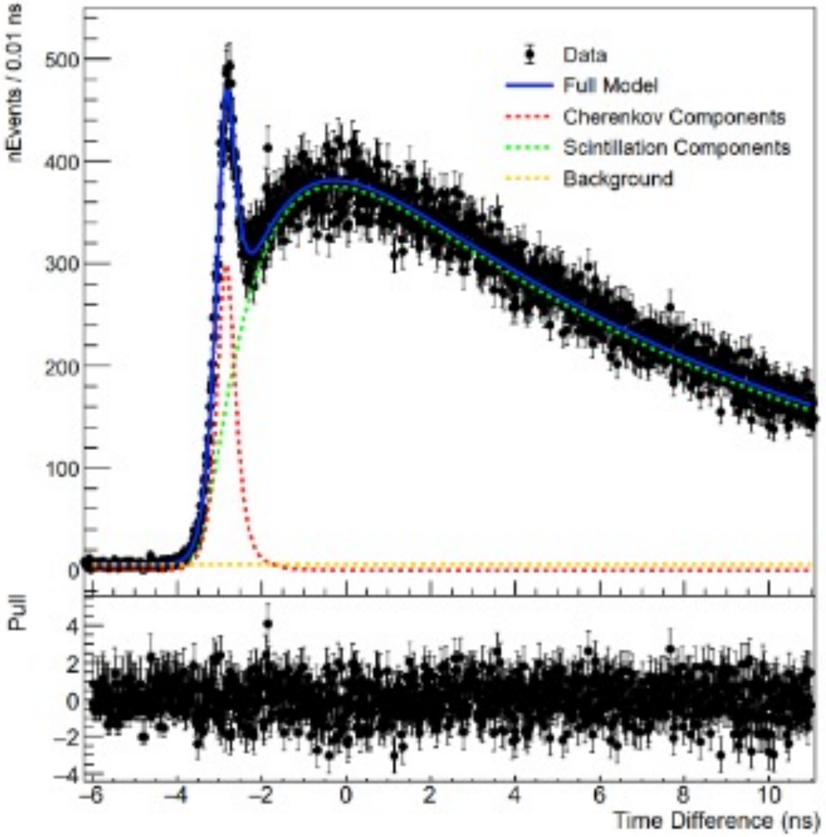


Calculating the Cherenkov Ratio until the Scintillation Peak

Ch Ratio = 10.5 %

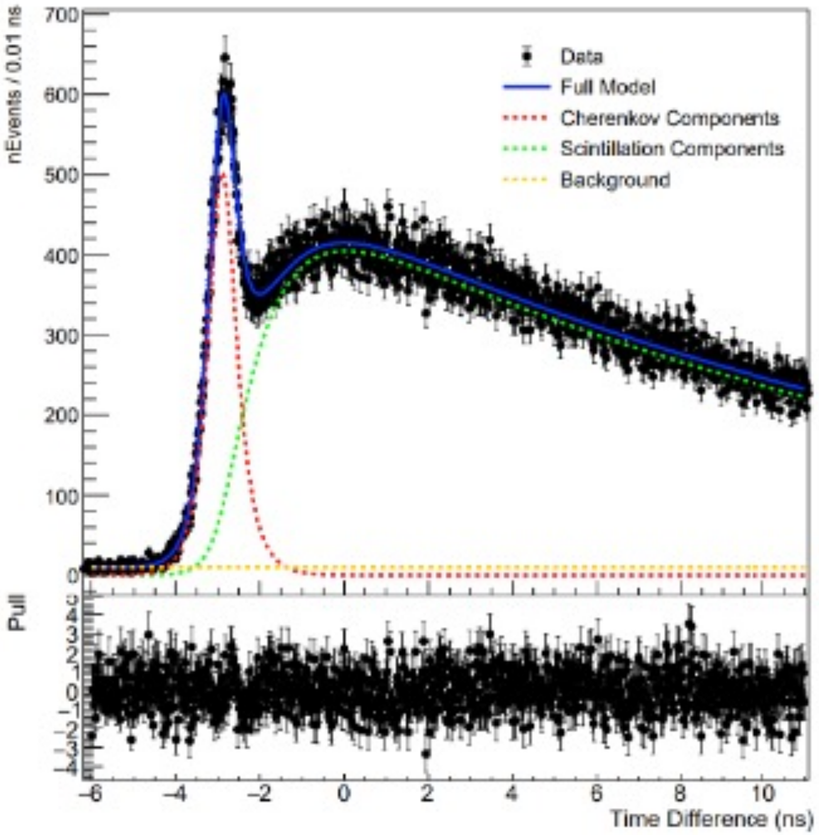
Scintillation decay components in good consistence with the previous study by Raphael Stock

Separation Study of the Bi-solvent Slow Scintillator



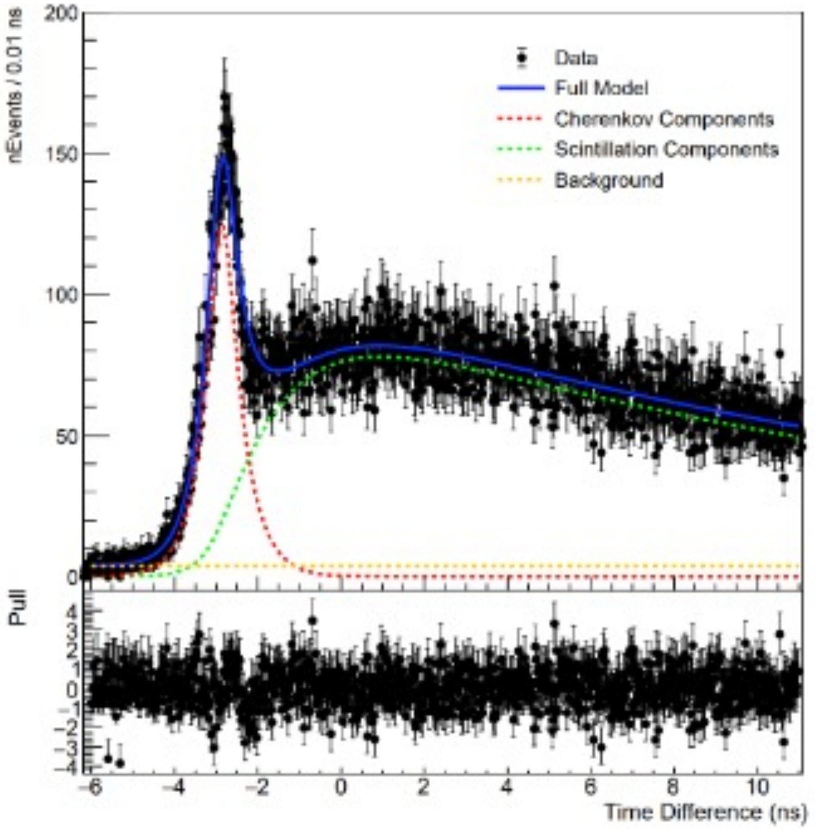
PPO-20

15.3 %



PPO-10

28.3 %



PPO-05

29.1 %

PSD Studies: Time Correlated Single Photon Counting with Pulsed Neutron Beams

Time Profile Experiment

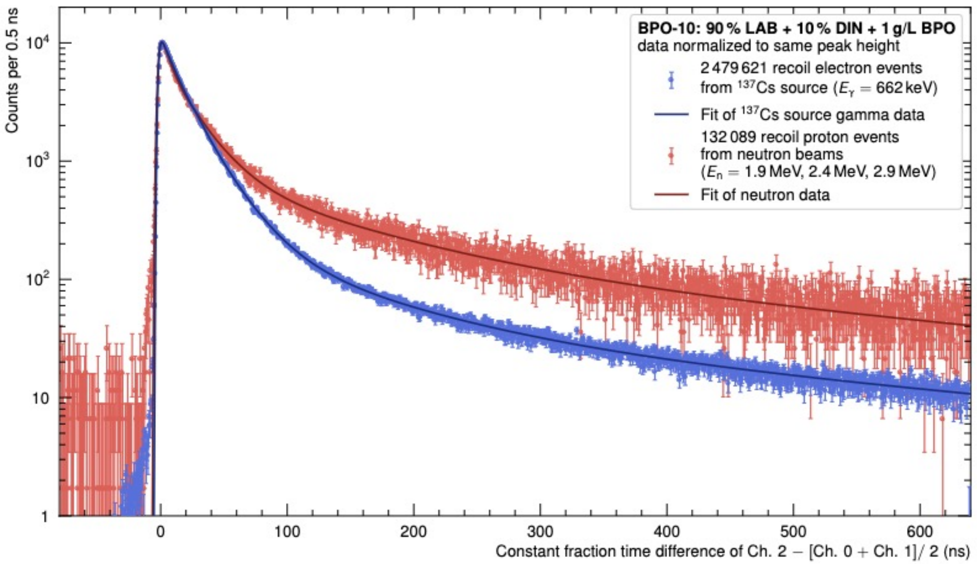
M. R. Stock
(PhD Thesis)

- The vessel containing ~180 cm^3 LS is placed between two PMTs
  - provide the start signal of the time measurement
- third PMT is placed in a certain distance to ensure the detection of only a single photon from each event!
  - provides the stop signal (TCSPC)

Neutron production at the CN Accelerator:

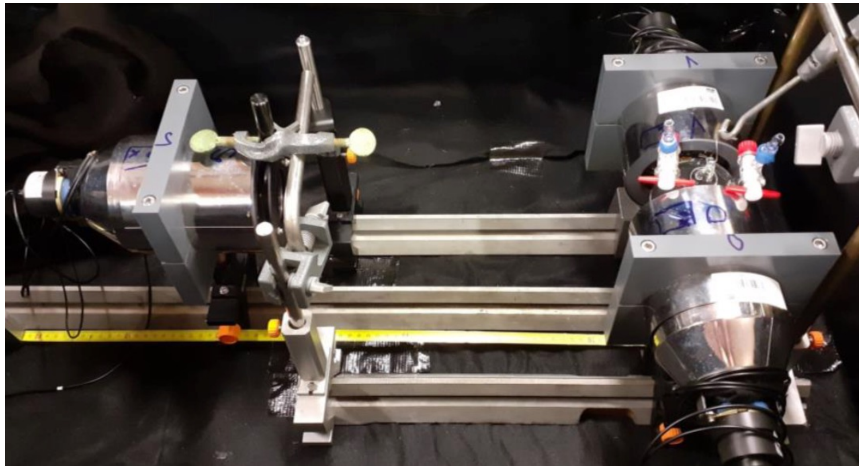
$$^7Li + p \rightarrow\ ^7Be + n$$

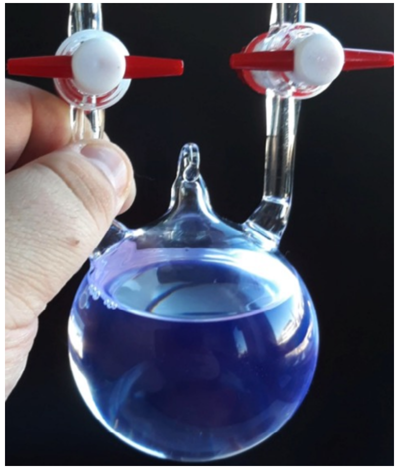
Nuclear reaction for quasi-monoenergetic neutron production
(Reaction Threshold: 1877 keV)



PSD exceending the JUNO-LS PSD by far! Similar to Borexino LS!
(even with lower fluor concentrations)

Distinguish neutron interactions from beam correlated gammas by time-of-flight (ToF) measurements!
→ very clean neutron sample





The second and therefore slower component is the dominant one!

The LS can be tuned to be very slow without loss in LY!

Effective Liftime: approx. 60 ns!

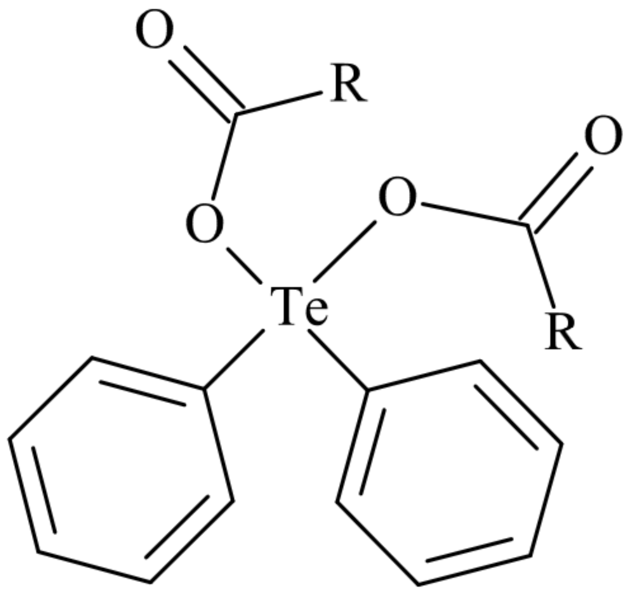
Sample	PPO-05	
Source	e$^-$	p
A_1 (%)	$18.66 \pm 0.20_{-8.06}$	$14.20 \pm 1.46^{+2.21}$
A_2 (%)	$68.03 \pm 0.26^{+5.06}$	48.40 ± 1.44
A_3 (%)	$7.64 \pm 0.09^{+2.94}$	21.46 ± 1.61
A_4 (%)	$5.67 \pm 0.06_{-0.38}$	15.94 ± 0.87
τ_r (ns)	$1.94 \pm 0.02^{+0.42}_{-0.42}$	$1.37 \pm 0.02^{+0.43}_{-0.43}$
τ_1 (ns)	$17.79 \pm 0.18^{+0.08}_{-4.64}$	$11.53 \pm 0.63^{+1.03}_{-0.16}$
τ_2 (ns)	$32.40 \pm 0.33_{-2.15}$	32.24 ± 1.21
τ_3 (ns)	$104.68 \pm 1.05^{+5.82}_{-17.88}$	135.00 ± 19.71
τ_4 (ns)	$436.13 \pm 7.52^{+55.57}_{-17.70}$	732.53 ± 361.00
τ_{life}	$60.03 \pm 0.56^{+4.74}_{-3.27}$	$164.37 \pm 58.11^{+0.52}_{-0.43}$

Tellurium Doping of Slow LS Cocktails

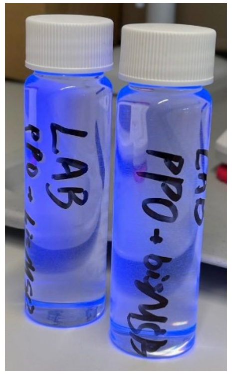
Ideal Dopant:

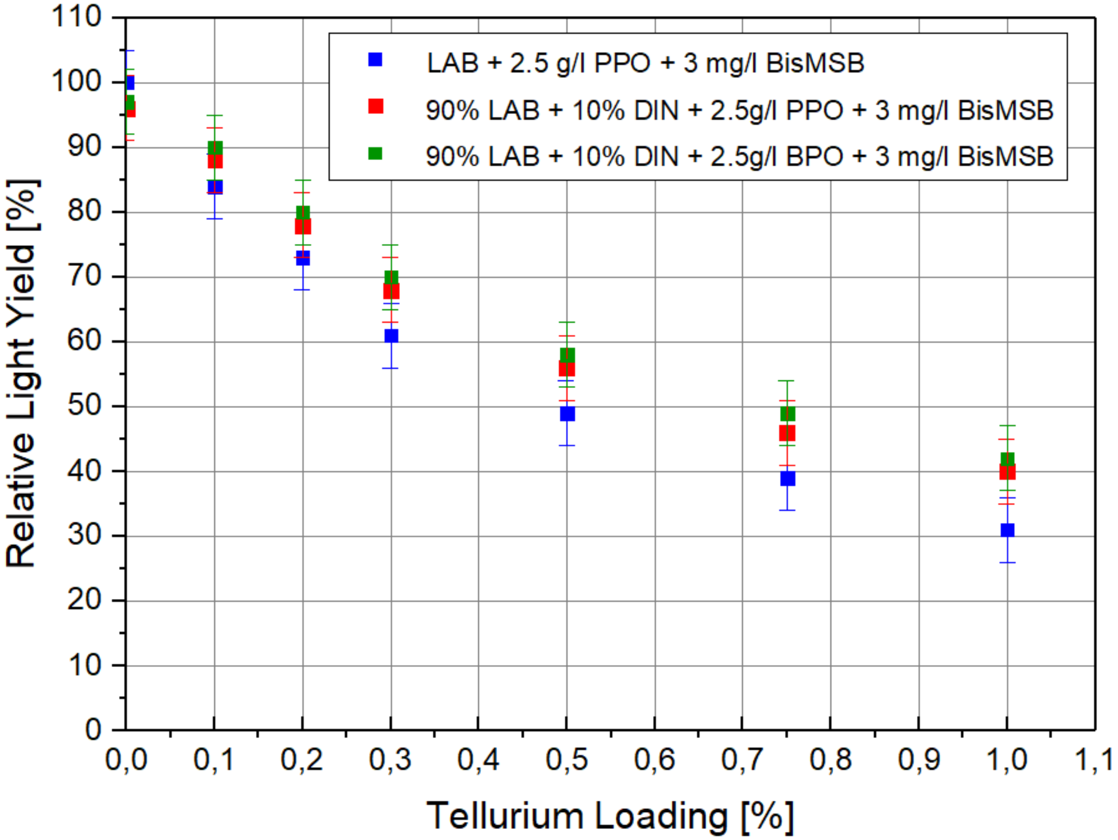
- **high solubility in LAB**
- individual compound
- solid material (powder)
- **no absorption bands in the emission region of the LS**
- **chemical stability** against humidity, oxygen + material compatibility
- low toxicity
- high flashpoint

Loading via the method of **I.A. Suslov et al., NIM-A, 1040 (2022) 167131**



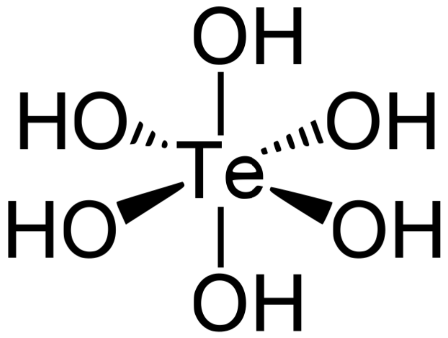
Diphenyltellurium dicarboxylates:
(a) dipivalate
(b) diisovalerate
(c) di-2-ethylhexanoate



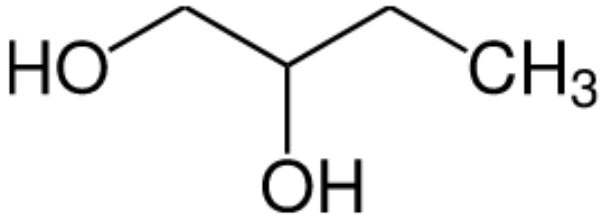


- Loading up to 0.2% by mass Tellurium do not harm the LY too much
- Nearly no visible influence on LS transparency (in UV/Vis measurement)
- Admixtures of DIN: reduce light-loss by Te-dopant a lot!
- BPO instead of PPO: better LY (better energy transfer from DIN)
- Excellent stability over 1 year (see also Suslov et al., 2023)

Tellurium Doping of Slow LS Cocktails using Te-diols

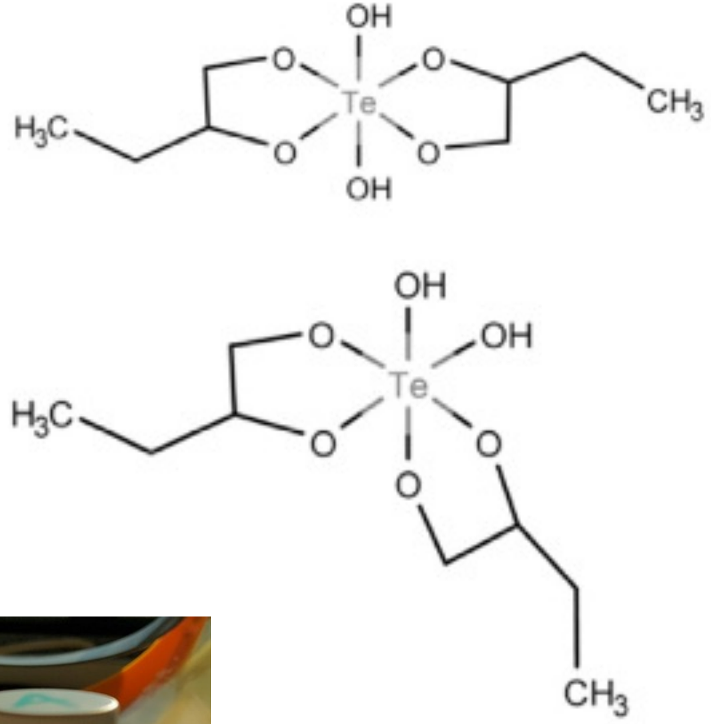


Telluric Acid (TeA)



1,2-Butanediol

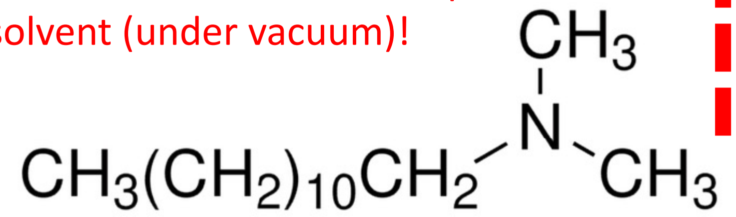
Azeotropic Distillation
(Toluene, Benzene etc.)



Neutralization of TeA

N$_2$ bubbling and mixing with toluene/benzene as an azeotropic solvent (under vacuum)!

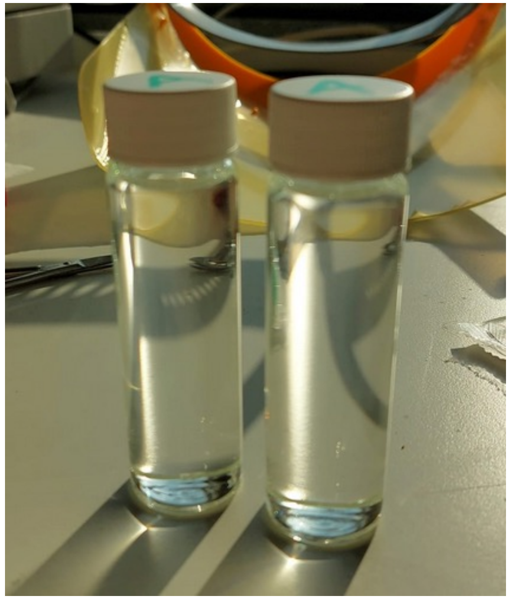
Weak acid – but solid at 20°C



N,N-Dimethyldodecylamin

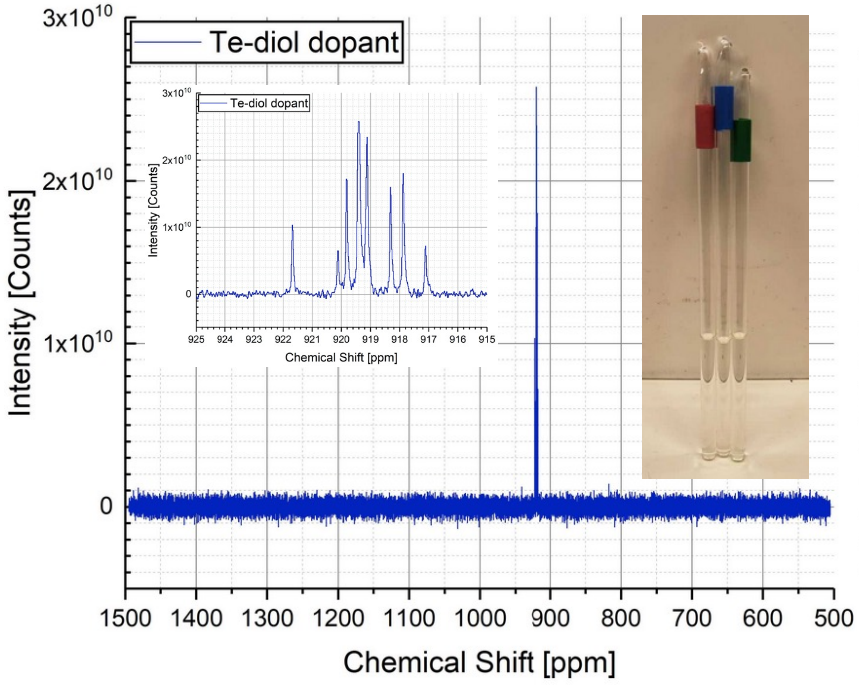
Two ways:

- Hot synthesis in aqeous solution
  → Oligomerization + DDA
- Cold way via neutralization with amine (no water!)

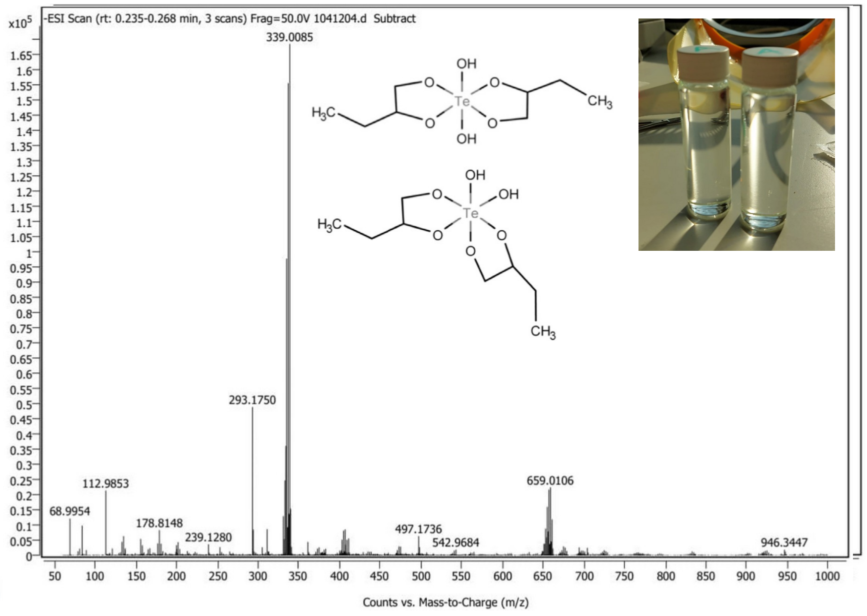


First samples: 1%$_m$ and 20%$_m$ Te-loading in LAB!
→ sent to NMR in Mainz

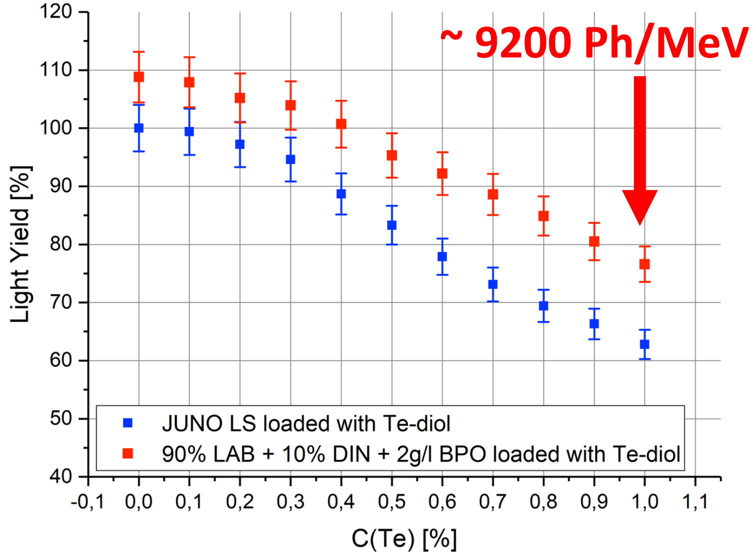
Tellurium Doping of Slow LS Cocktails with Te-diols



NMR Spectrum of the Te-complexes



ESI-MS Spectrum



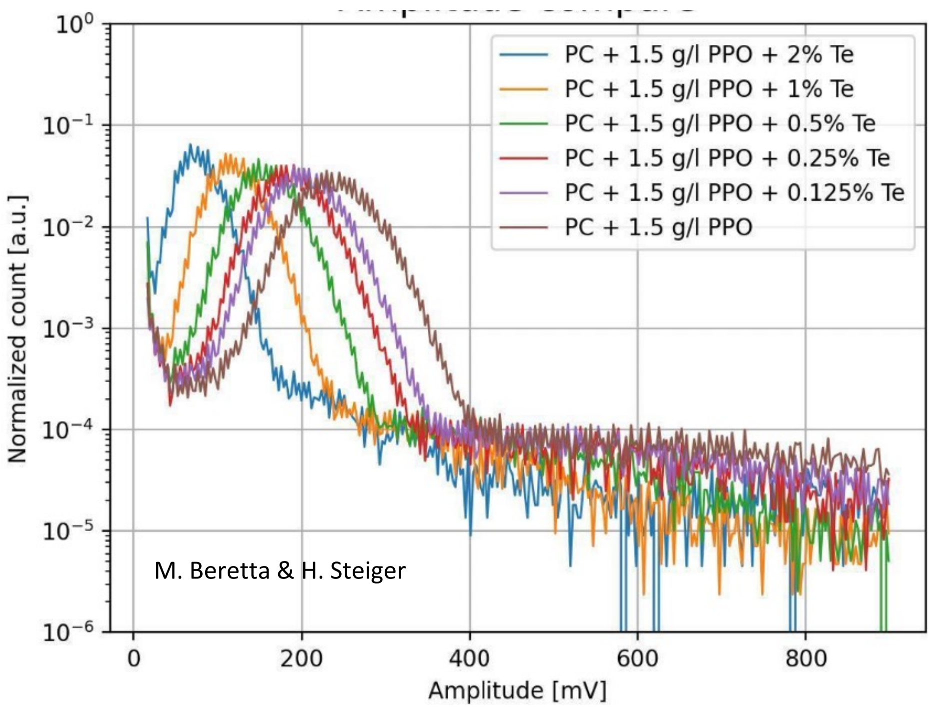
Slow LS exceeds LY of conventional LS with loading by far!

Prof. Dr. Mihail Mondshki:
Chemist at JGU Mainz
→ Performs all the chemical analysis for us!
→ Mass Spectrometry, NMR Spectroscopy

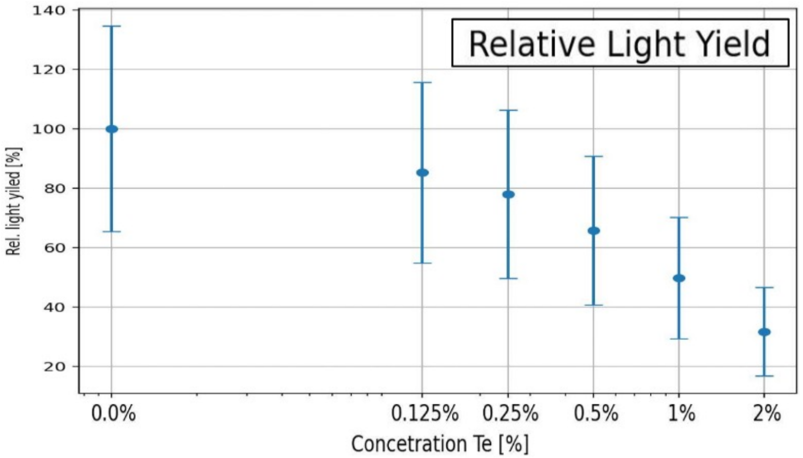
First samples (cold):
$1\%_m$ and $20\%_m$ Te-loading in LAB, PC, Xylene, DIN
→ Loading confirmed with NMR and ESI-MS

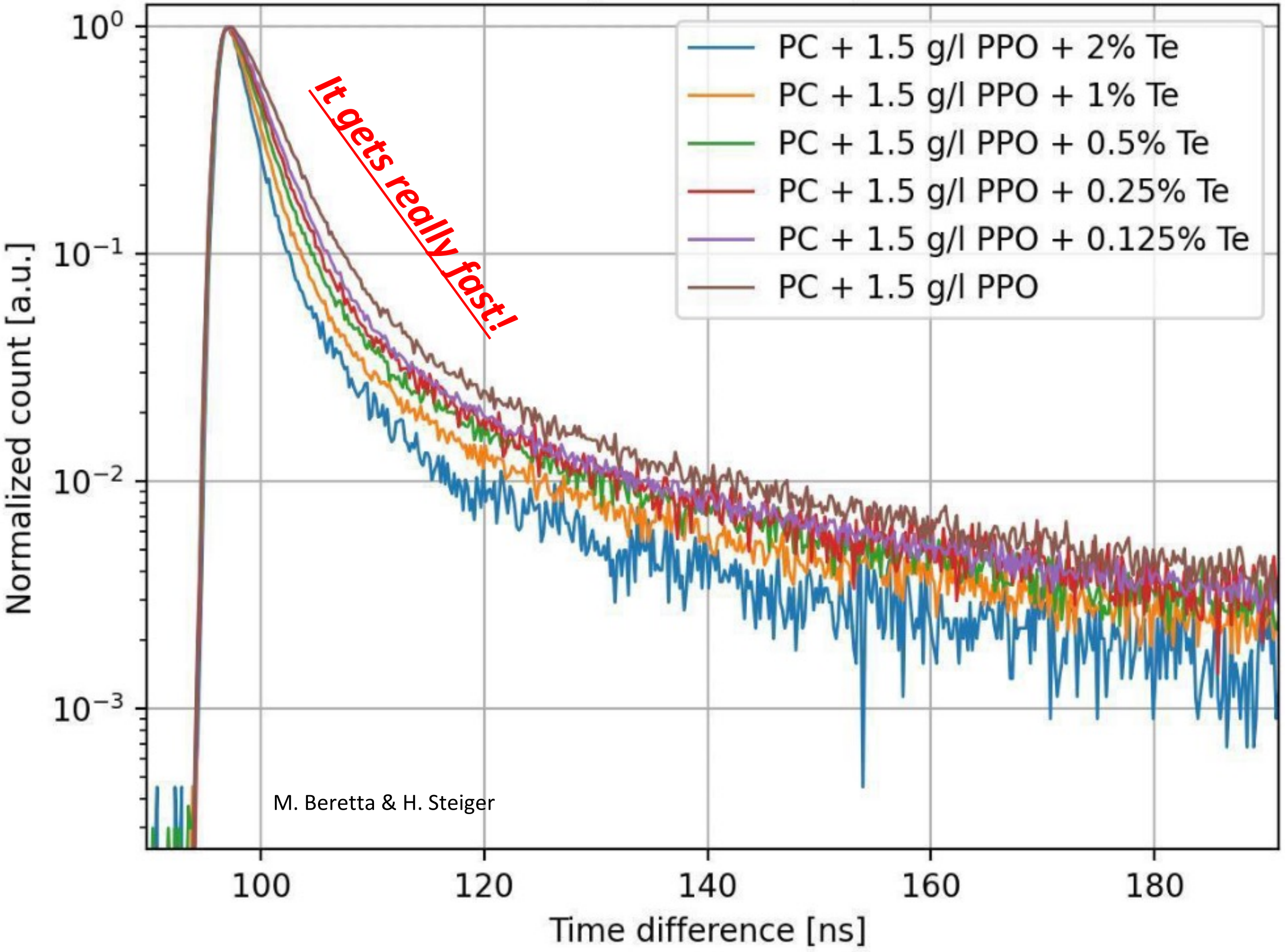
- No aging observed so far (1 year).
- No degradation in LY or transparency.
- High loadings can be nicely diluted → master solution
- Currently other diols and amines are tested.
- Constant quality checks by M. Böhles and M. Mondeshki!

Tellurium Doping of the Borexino LS (with Marco Beretta @ INFN Milano)



Light Yield extracted from the time profile setup (beta-source)





Precision Time Profile Study (INFN Milano) of Te-doped Borexino LS produced at TUM

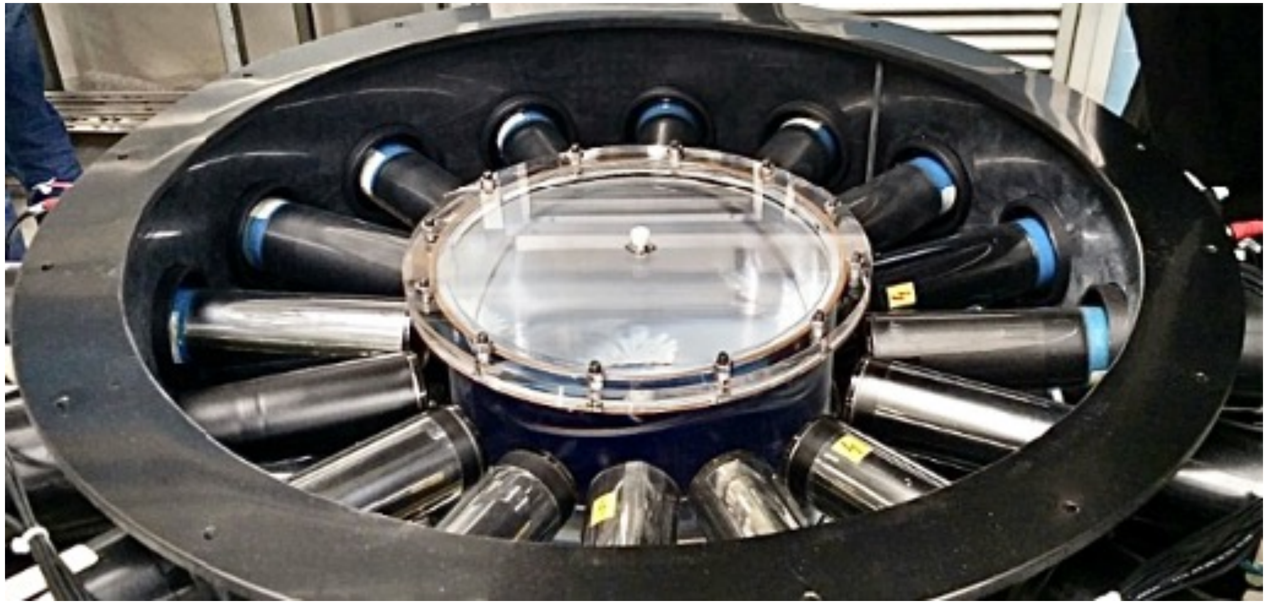
Higher loading → faster decay times!

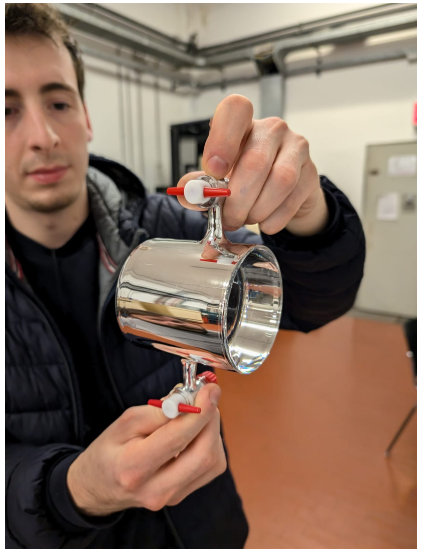
Study will be published soon in JINST in a joint paper.

Testbeam at the INFN-LNL for Slow Scintillators with and without Te-doping (Nov. 2024)

TUM: H.S., Meishu Lu, Shijiao Gao, Ronja Huber, Armin Siebert
JGU Mainz: Manuel Böhles
Uni Padova: Alberto Garfagnini, Andrea Serafini, Benedetta Rasera, Luca Silvestrin

Approx. 50 Tbyte of new data → to be analyzed soon!





Filling of Lavatrice: 90% LAB + 10% DIN + 1g/l PPO (very large amounts of data)

Samples studied for time profile and quenching:

- Borexino LS + 0.3% Te
- JUNO LS + 1% Te
- 95% LAB + 5% DIN + 1g/l PPO
- 95% LAB + 5% DIN + 1g/l BPO
- 90% LAB + 10% DIN + 1g/l BPO + 0.1% Te
- 90% LAB + 10% DIN + 1g/l BPO + 0.3% Te
- 90% LAB + 10% DIN + 1g/l BPO + 0.9% Te
- LAB + 4g/l Acenaphtene + 40mg/l BPO


AG Wurm
Heating °C
Stirrer rpm
MH 15
ROTH
PRISMA
1
2
3
04/217

Backup Slides

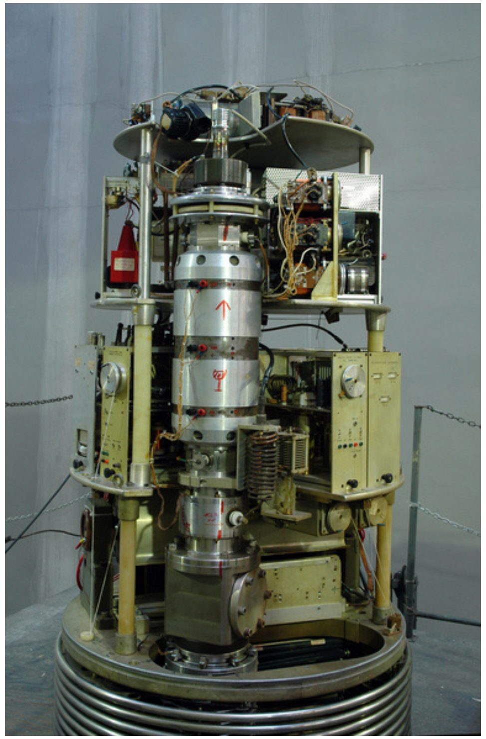
The CN Van de Graaff Particle Accelerator of the INFN-LNL as source of quasi-monoenergetic neutrons

INFN Istituto Nazionale di Fisica Nucleare — Laboratori Nazionali di Legnaro

Aerial view of the LNL with the tower of the CN accelerator


The CN HV Column


CN in operation
(closed pressure vessel)


Ion source and buncher

Proton beam with energies from 3.5 - 5.5 MeV.
(0.8-3 MV requires shorting parts of the accelerating column)

Energy stability: 2-3 keV

Currents: continous up to 3 uA, pulsed: 1 uA at 3 MHz

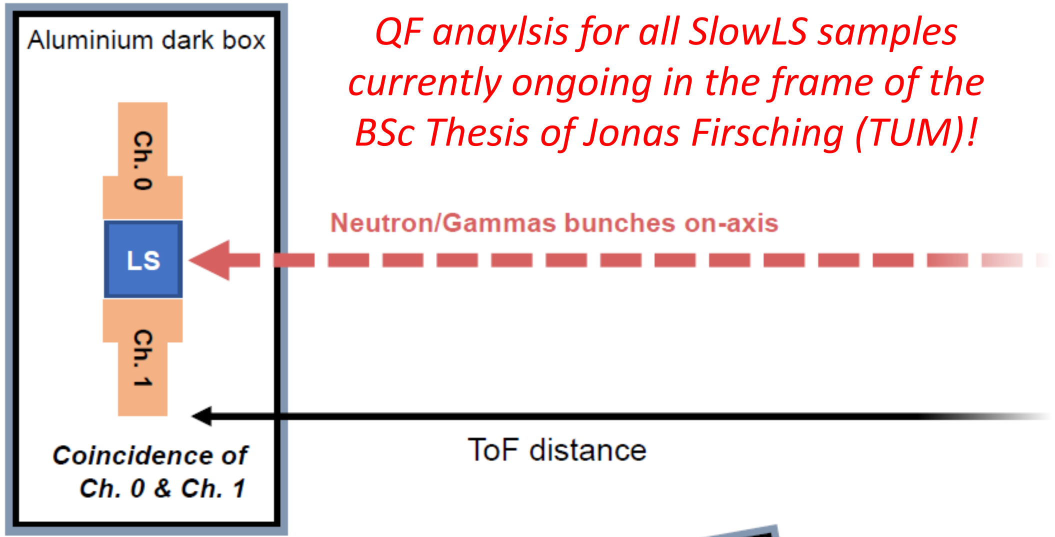
Pulse width: < 1ns

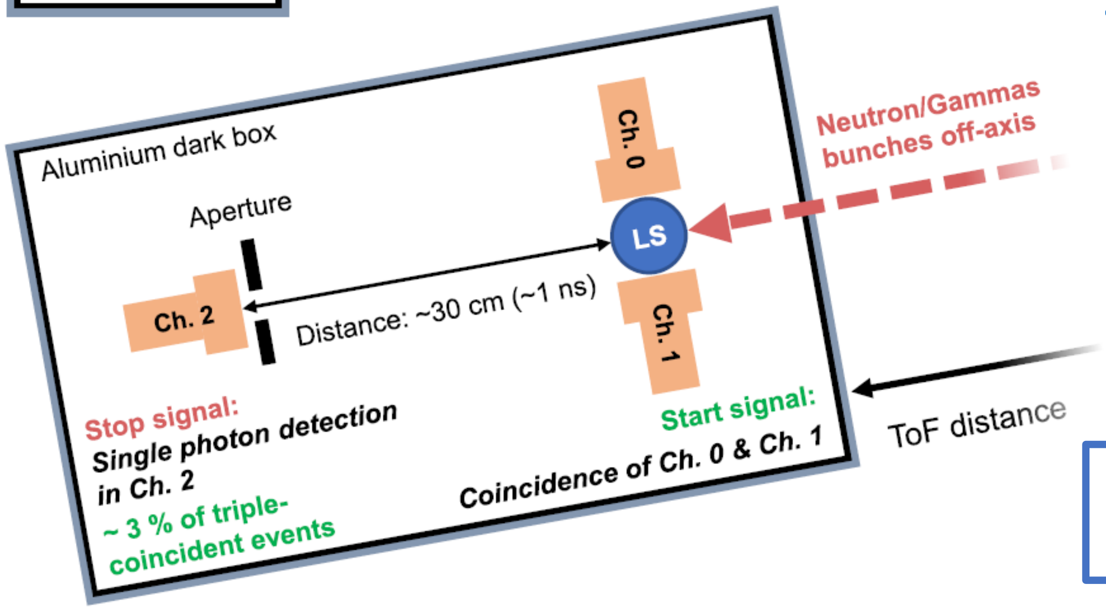
$$^{7}Li + p \rightarrow {}^{7}Be + n$$

Nuclear reaction for quasi-monoenergetic neutron production
(Reaction Threshold: 1877 keV)

Pulse Shape Discrimination and p-QF Study

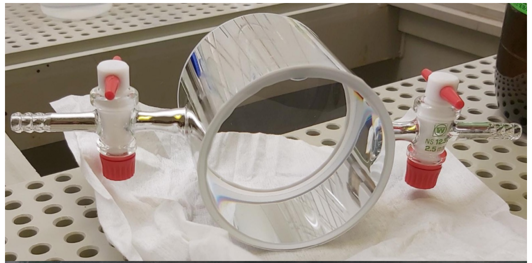
QF anaylsis for all SlowLS samples currently ongoing in the frame of the BSc Thesis of Jonas Firsching (TUM)!

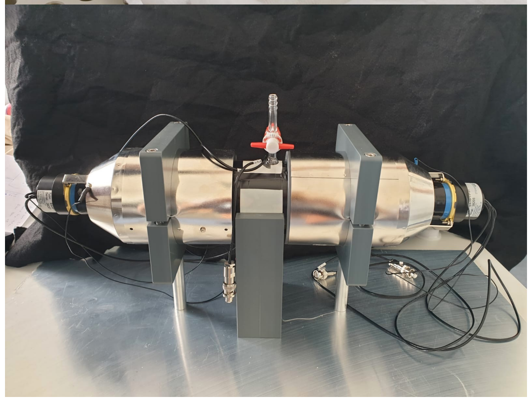




We simultaneously operate two experiments.

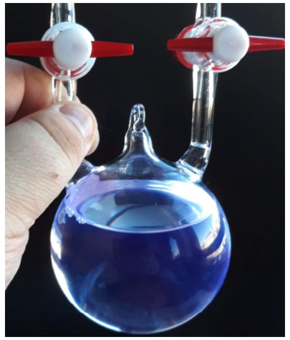
Quenching Factor (QF) experiment





- positioned directly on the beam axis
- detector placed in its own dark box
- target vessel contains ~400 cm^3 of LS
- optimized for low energy threshold with an efficient noise suppression:
  - coincidence of 2 PMTs with the beam trigger
  - vessel walls with highly reflective aluminum mirrors (BX-CTF)

Time Profile Experiment

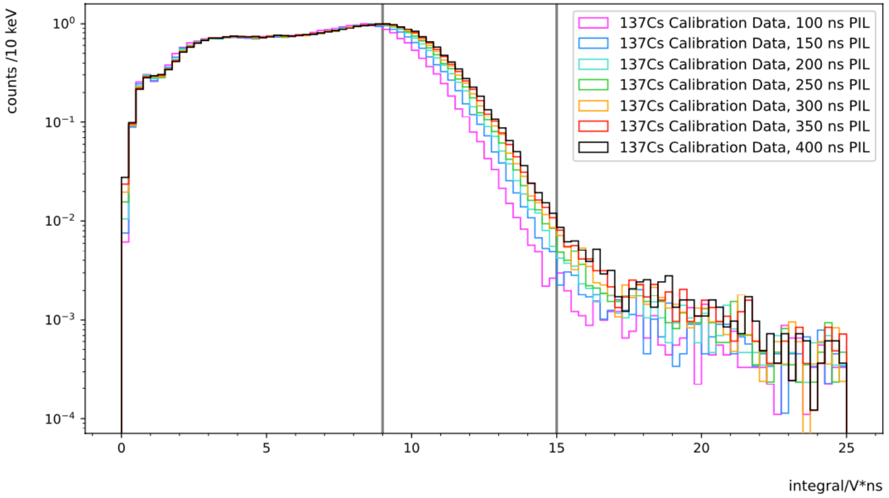


- Setup is placed in its own dark box.
- The vessel containing ~180 cm^3 LS is placed between two photomultiplier tubes (PMTs)
  - provide the start signal of the time measurement.
- third PMT is placed in a certain distance to ensure the detection of only a single photon from each event!
  - provides the stop signal.

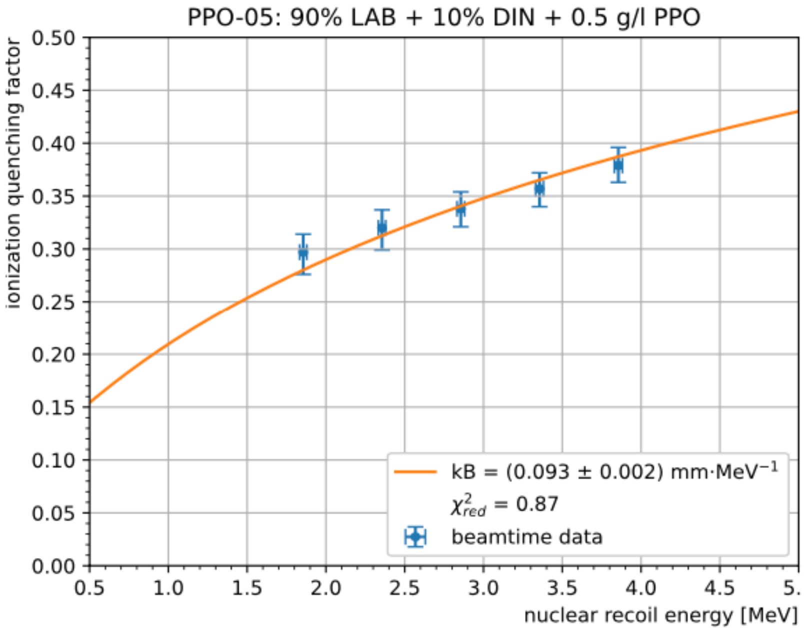
In both experiments we distinguish neutron interactions from beam correlated gammas by time-of-flight (ToF) measurements!

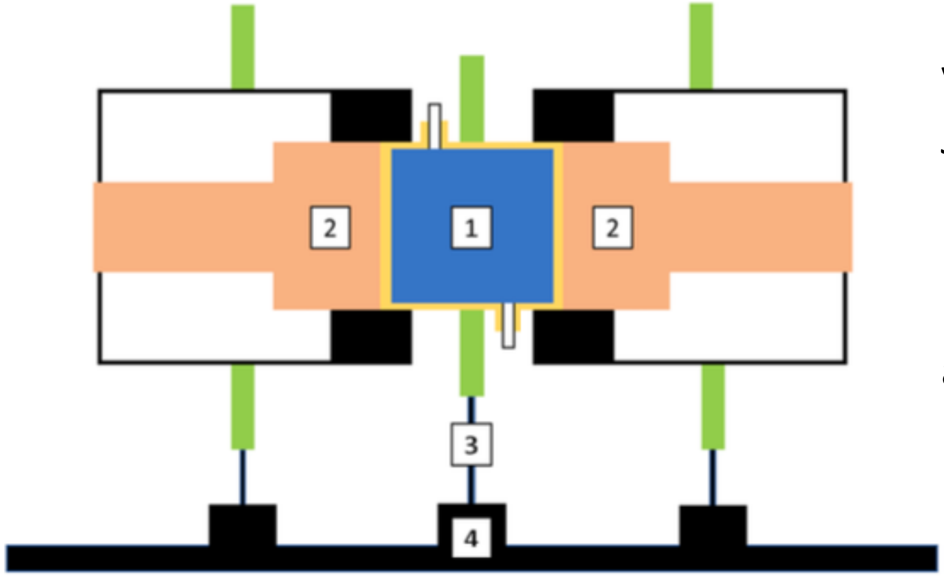
Proton Recoil Quenching Factors and Nonlinearity

Jonas Firsching
(BSc Thesis)



Slow scintillation requires long integration windows!

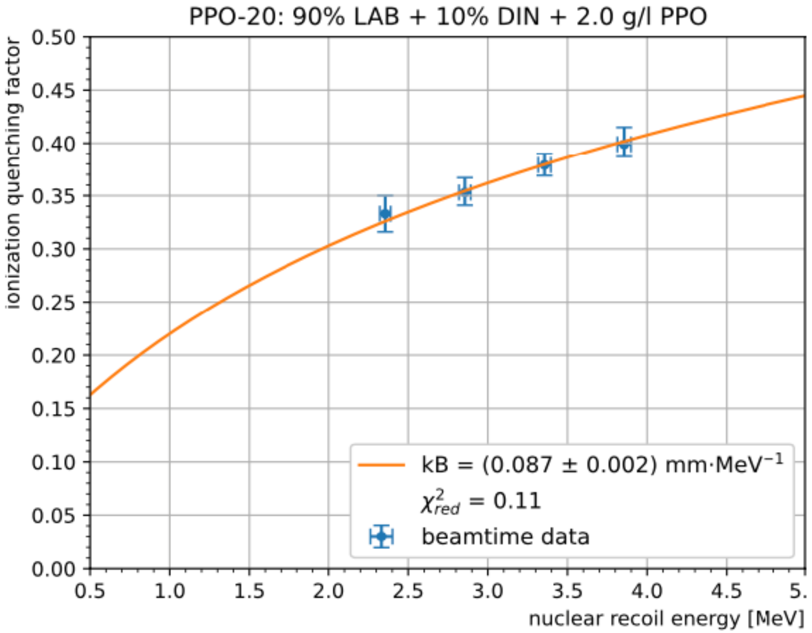




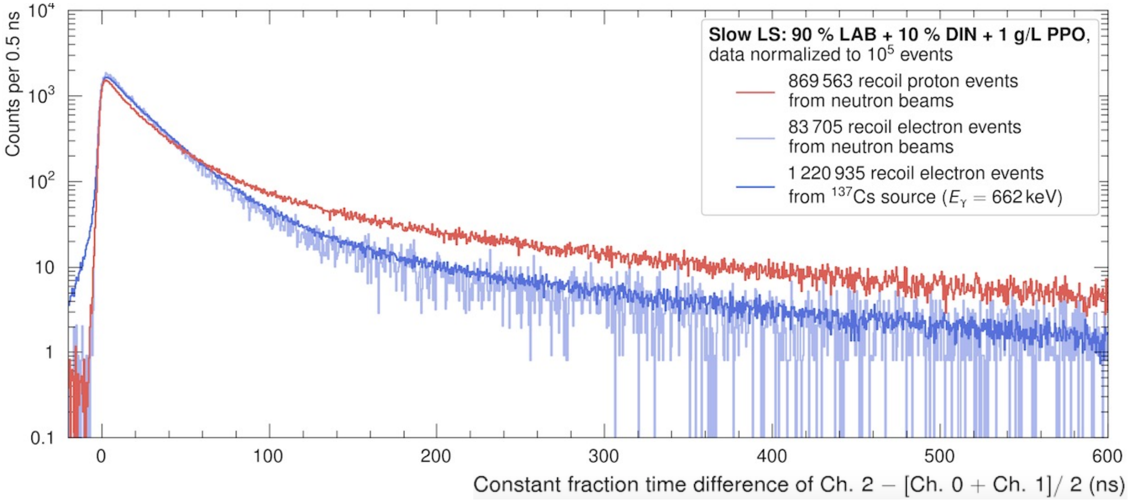
Experimental setup irradiated by a pulsed neutron beam

Very similar results to a JUNO-like LAB mixture or PC mixtures.

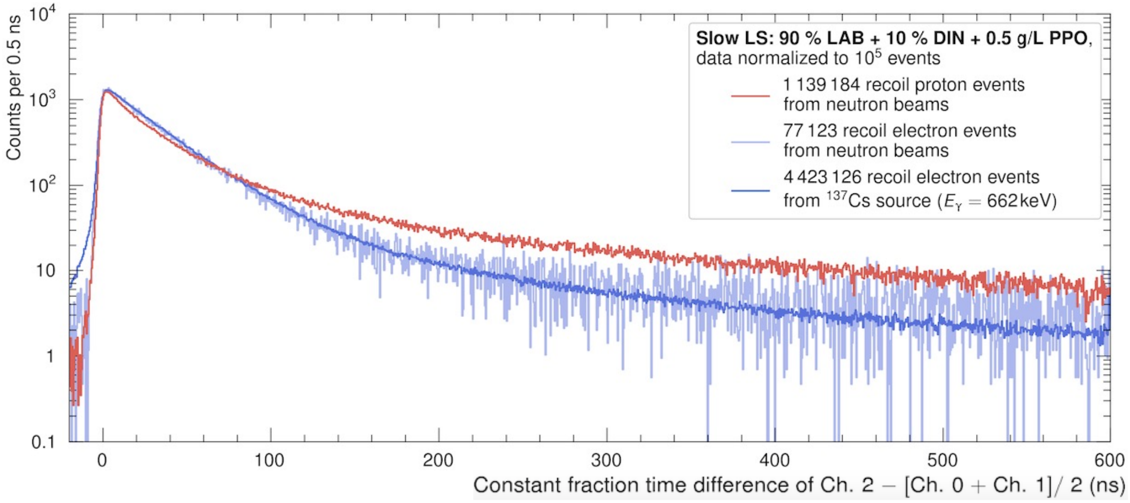
No trouble by multi-solvent approach!



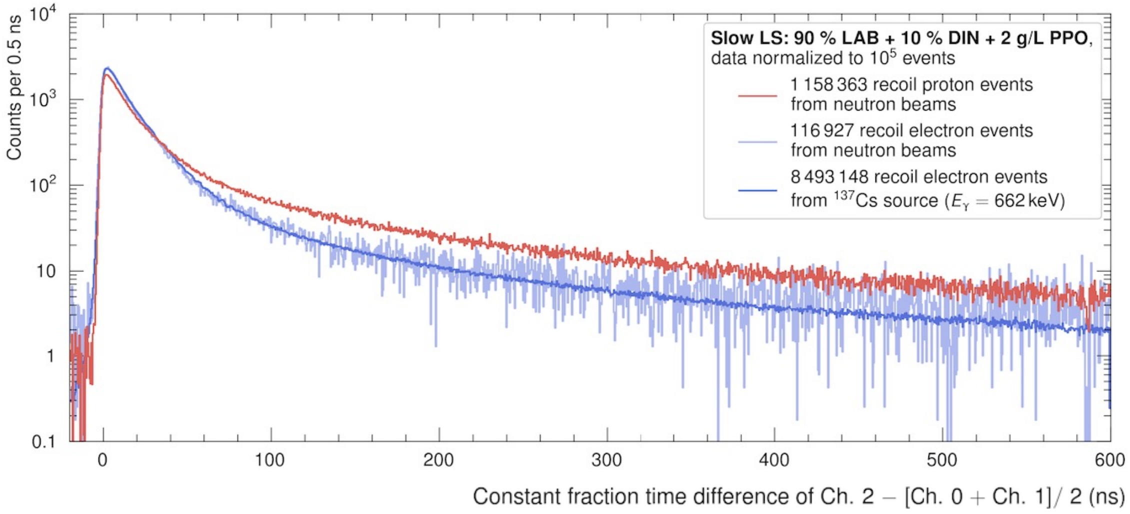
Pulse Shape Discrimination – Strong even with low Fluor Concentration



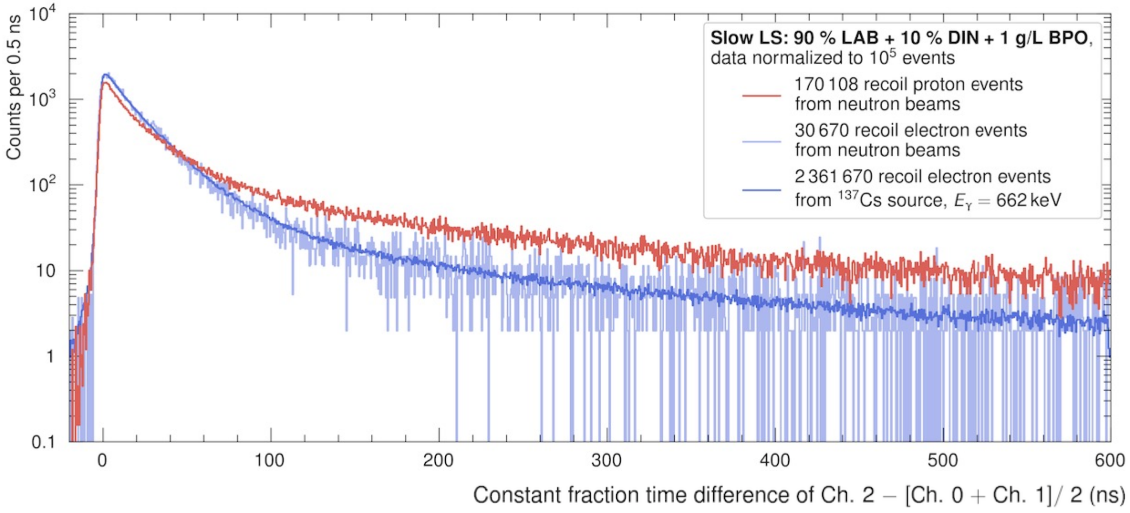
90% LAB + 10% DIN + 1g/l PPO ($\tau_1 \sim 18ns$)



90% LAB + 10% DIN + 0.5 g/l PPO ($\tau_1 \sim 31ns$)



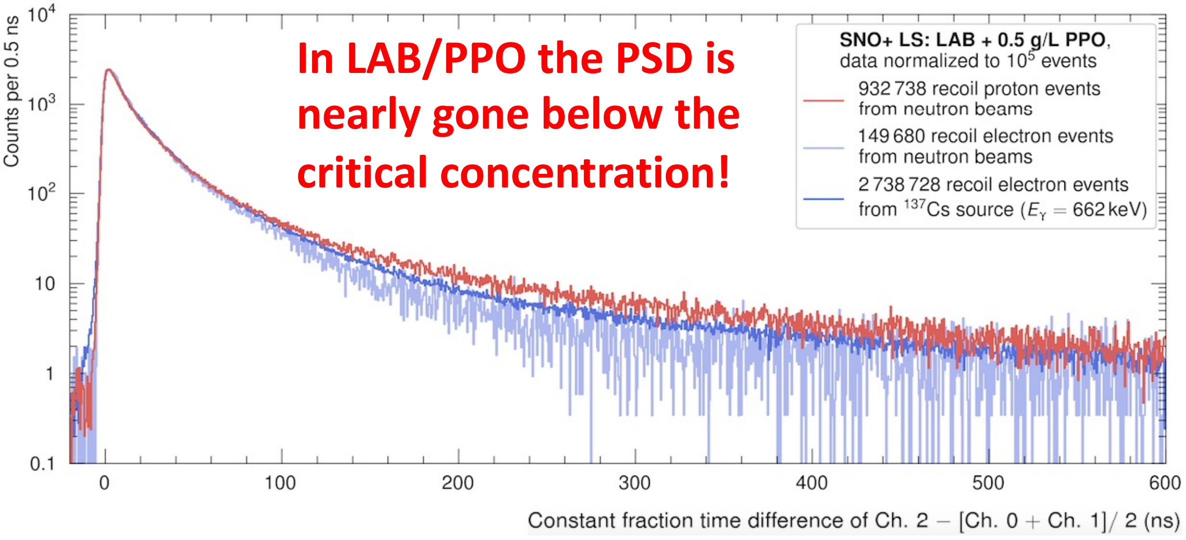
90% LAB + 10% DIN + 2g/l PPO ($\tau_1 \sim 12ns$)



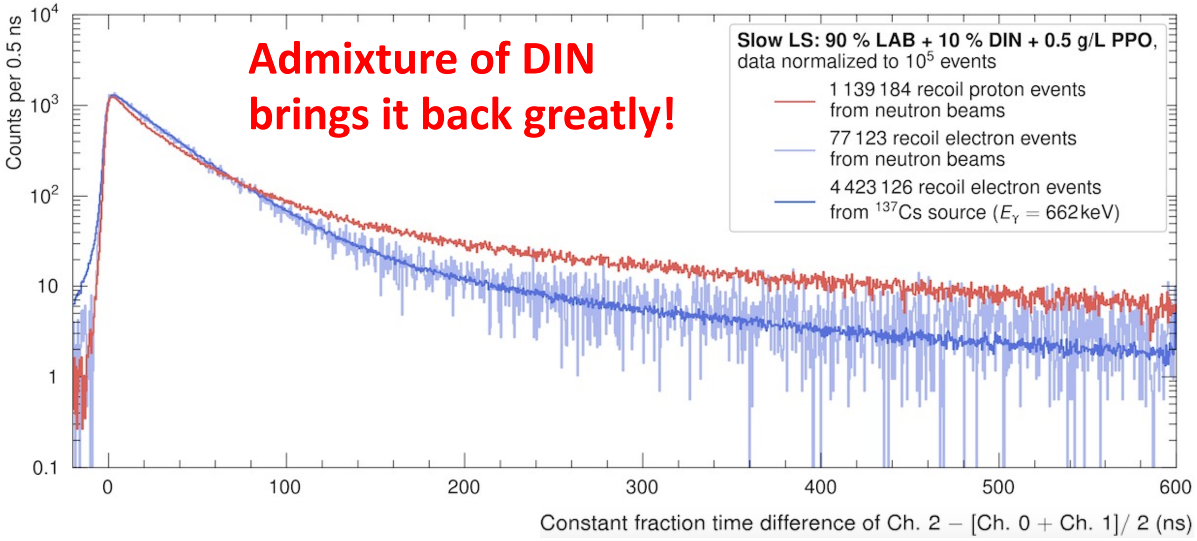
90% LAB + 10% DIN + 1 g/l BPO ($\tau_1 \sim 17ns$)

All plots from R. Stock (TUM)

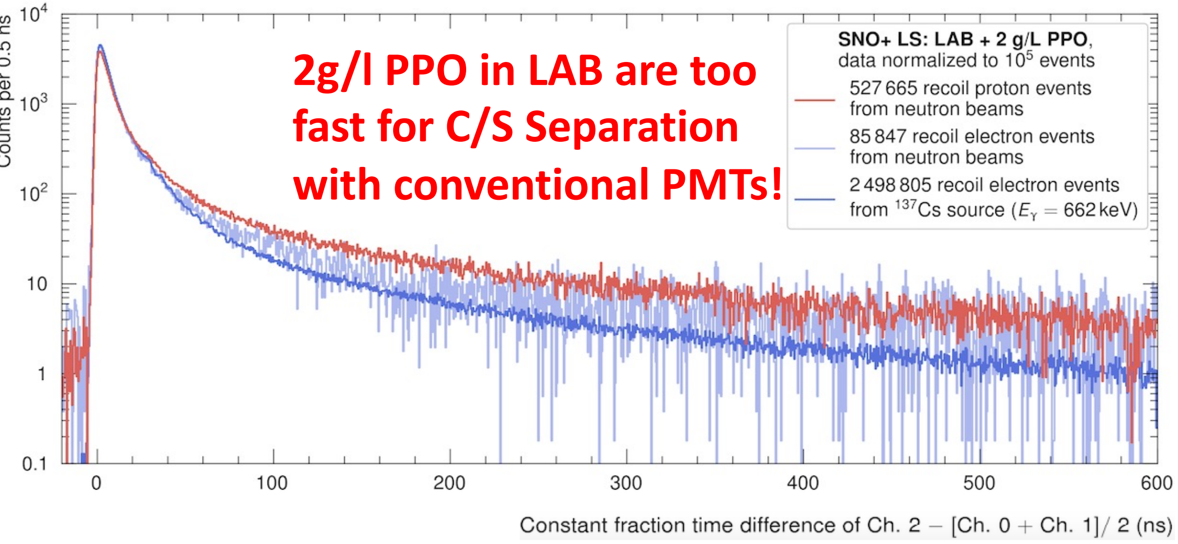
Pulse Shape Discrimination – A Comparison



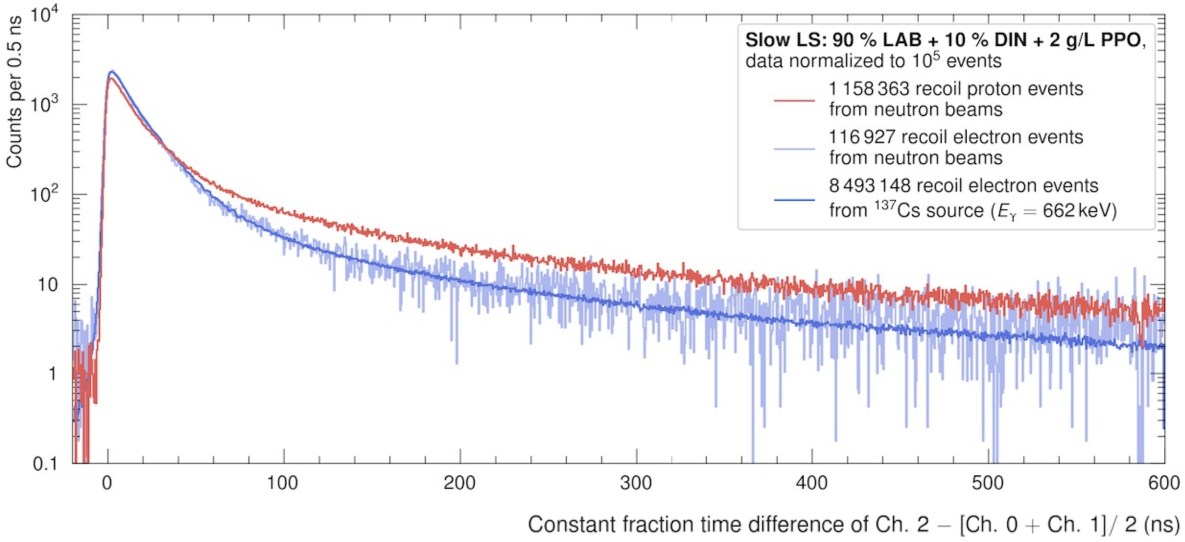
LAB + 0.5 g/l PPO (still quite fast scintillation!)



90% LAB + 10% DIN + 0.5 g/l PPO ($\tau_1 \sim 31ns$)



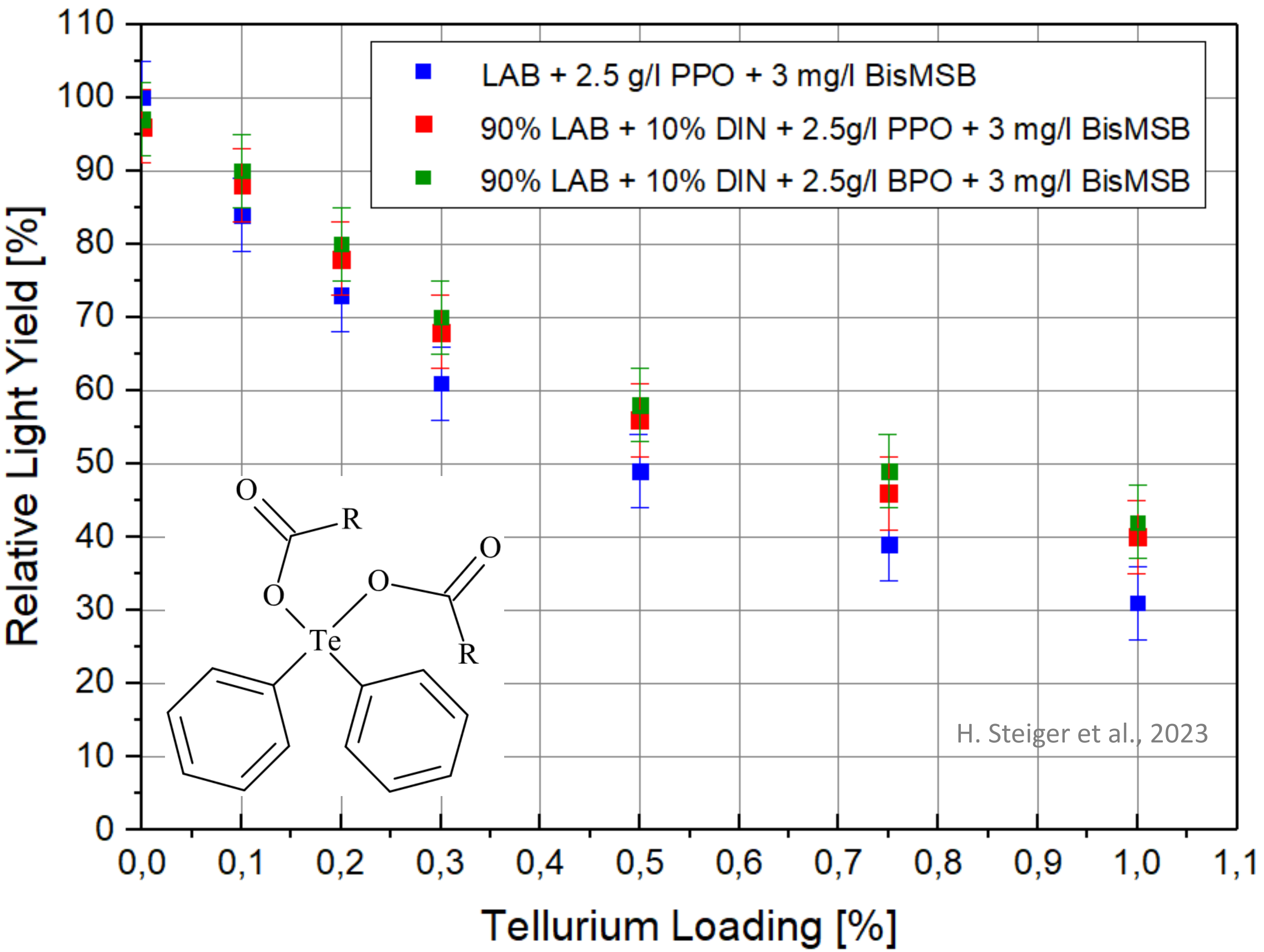
LAB + 2g/l PPO



90% LAB + 10% DIN + 2g/l PPO ($\tau_1 \sim 12ns$)

All plots from R. Stock (TUM)

Tellurium Doping of Slow LS Cocktails – Influence on the Light Yield



- Loading up to 0.2% by mass Tellurium do not harm the LY too much. (40t Te in JUNO-II?)
- No visible influence on LS transparency (in UV/Vis measurement)
- Admixtures of DIN: reduce light-loss by Te-dopant a lot!
- BPO instead of PPO: better LY (better energy transfer from DIN)
- Excellent stability over 1 year (see also Suslov et al., 2023)
- Questions:
    - 1. Is the radiopure synthesis of the Te-dopant possible?
    - 2. Can we do better with Xe-loading?