Liquid Scintillator Detectors

Minfang Yeh

NuFACT, Snowbird, Utah, 08/01-06/2022

Brookhaven National Laboratory
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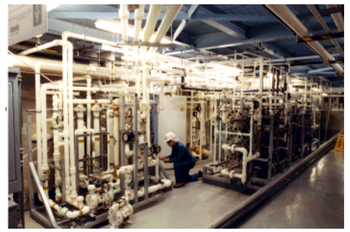
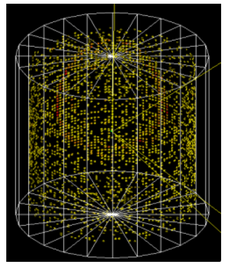
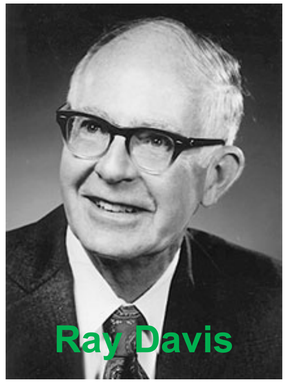
Detector Development
BNL Neutrino and Nuclear Chemistry Since 1960







HOMESTAKE — 615t $^{37}Cl + \nu_e \rightarrow {}^{37}Ar + e^-$

Gallex — 30-t $^{71}Ga + \nu_e \rightarrow {}^{71}Ge + e^-$

780t D_2O CC/NC — SNO

200-kt H_2O or 37-kt LAr — LBNF (DUNE)

120-t 8% In-LS — LENS

200-t 0.1% Gd-LS — Daya Bay

780t 0.3% Nd/Te-LS — SNO+

^6Li, ^{10}B or Gd doped LS — PROSPECT/LZ

(Metal-doped) WbLS — 0νββ, dark-matter, medical, nonproliferation

Water-based LS

Scintillator

Radiochemical — Cherenkov

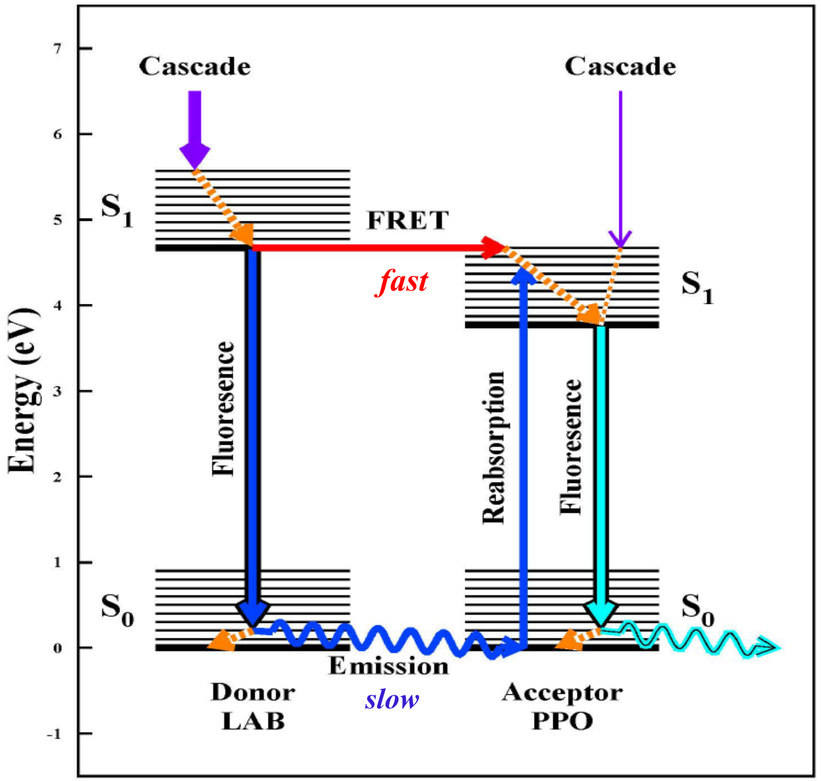
1960 1970 1980 1990 2000 2010 2020

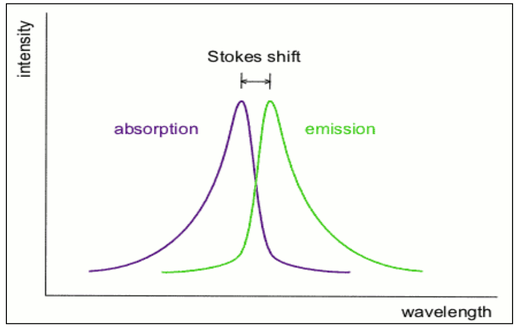
Outline

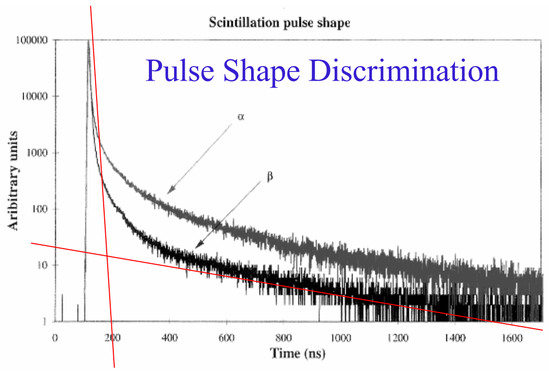
- Principle
- Current landscape
- Challenges and development for next generation liquid scintillator detectors
- Water-based Liquid Scintillator
- Summary

Liquid Scintillator Detectors

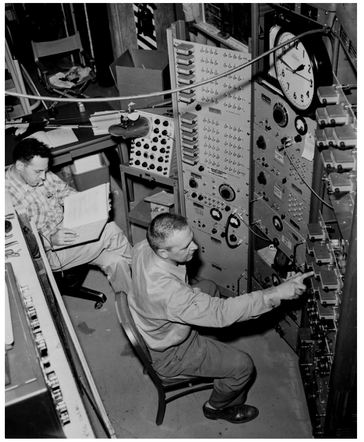
- $\bar{\nu}_e + p \rightarrow n + e^+;\ n + p \rightarrow d + \gamma$
- $\bar{\nu}_e + {}^{12}C \rightarrow e^+ + {}^{12}B \rightarrow {}^{12}C + e^- + \bar{\nu}_e$
- $\nu_e + {}^{12}C \rightarrow e^- + {}^{12}N \rightarrow {}^{12}C + e^+ + \nu_e$
- $\nu_x + {}^{12}C \rightarrow \nu_x + {}^{12}C^* \rightarrow {}^{12}C + \gamma$
- $\nu_x + e^- \rightarrow \nu_x + e^-$
- $\nu_x + p \rightarrow \nu_x + p$







S. Hans et. al., Light-yield quenching and remediation in liquid scintillator detectors, 2020 *JINST* 15 P12020



- Serve neutrino community since Reines and Cowan in 1950s
  - Stokes shift, photon-yield, timing structure, and C/H density determine responses

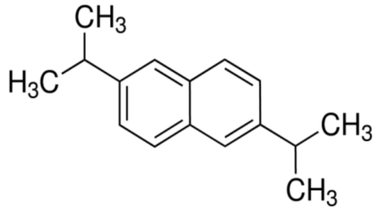
Liquid Scintillators

Table 1: Density, flash point and the wavelengths of the optical absorption/emission peaks (dissolved in cyclohexane) for several solvent candidates are shown.

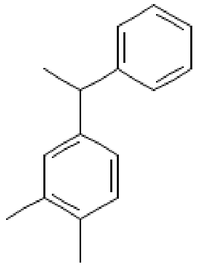
C. Buck and M. Yeh, J. Phys. G: Nucl. Part. Phys. 43 093001 (2016)

Molecule	chemical formula	density [kg/l]	flash point	abs. max.	em. max.
PC	C_9H_{12}	0.88	48°C	267 nm	290 nm
toluene	C_7H_8	0.87	4°C	262 nm	290 nm
anisole	C_7H_8O	0.99	43°C	271 nm	293 nm
LAB	–	0.87	∼ 140°C	260 nm	284 nm
DIN	$C_{16}H_{20}$	0.96	> 140°C	279 nm	338 nm
o-PXE	$C_{16}H_{18}$	0.99	167°C	269 nm	290 nm
n-dodecane	$C_{12}H_{26}$	0.75	71°C	–	–
mineral oil	–	0.82 – 0.88	> 130°C	–	–

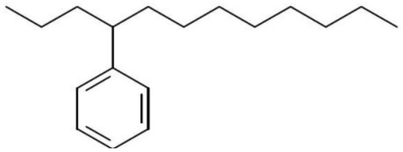
New scintillator materials provide better compatibility and less chemical hazard



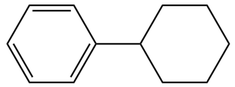
Di-isopropylnaphthalene (DIN)



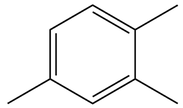
1-phenyl-1-xylyl-ethane (PXE)



Linear alkylbenzene (LAB)



Cyclohexylbenzene (PCH)



1,2,4-trimethylbenzene (PC)

Primary Fluors and Secondary Shifter

C. Buck and M. Yeh, J. Phys. G: Nucl. Part. Phys. 43 093001 (2016)

Table 2: The wavelengths of the optical absorption and emission peak maxima for primary and secondary fluor molecules diluted in cyclohexane are listed.

Molecule	chemical formula	abs. max.	em. max.
PPO	$C_{15}H_{11}NO$	303 nm	358 nm
PBD	$C_{20}H_{14}N_2O$	302 nm	358 nm
butyl-PBD	$C_{24}H_{22}N_2O$	302 nm	361 nm
BPO	$C_{21}H_{15}NO$	320 nm	384 nm
p-TP	$C_{18}H_{14}$	276 nm	338 nm
TBP	$C_{28}H_{22}$	347 nm	455 nm
bis-MSB	$C_{24}H_{22}$	345 nm	418 nm
POPOP	$C_{24}H_{16}N_2O_2$	360 nm	411 nm
PMP	$C_{18}H_{20}N_2$	295 nm	425 nm

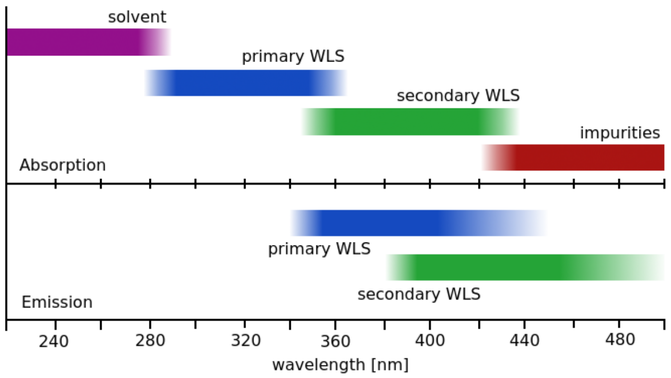
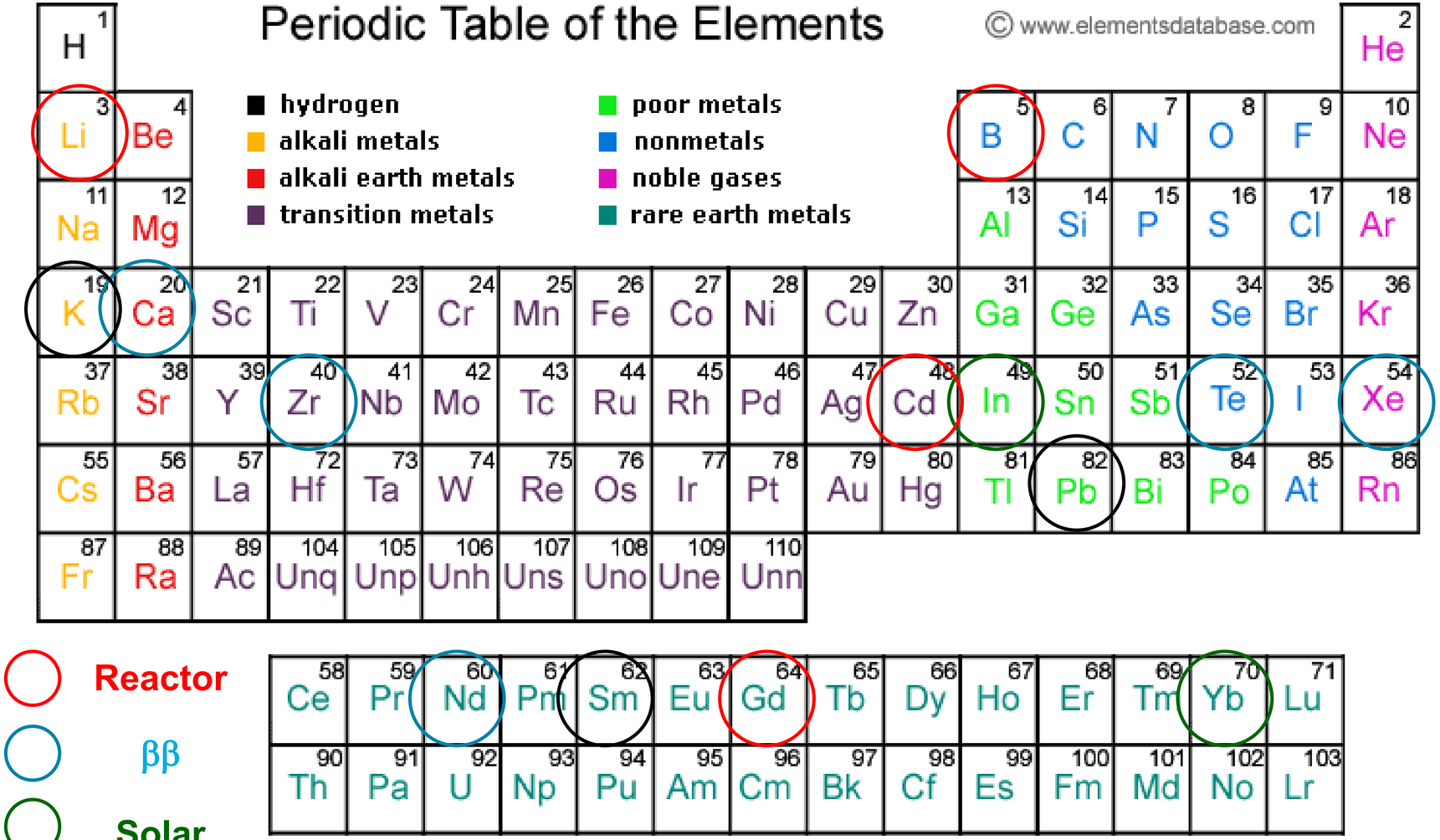


Figure 2: The plot shows for each scintillator component the wavelength region for which its own absorbance is dominating in the liquid. In addition the typical wavelength ranges for scintillator emission is shown for the case of just a primary or including a secondary wavelength shifter.

Redshift scintillation light to match sensible region of photodetectors

Metal-doped (Water-based) LS

for neutrino physics and other frontiers



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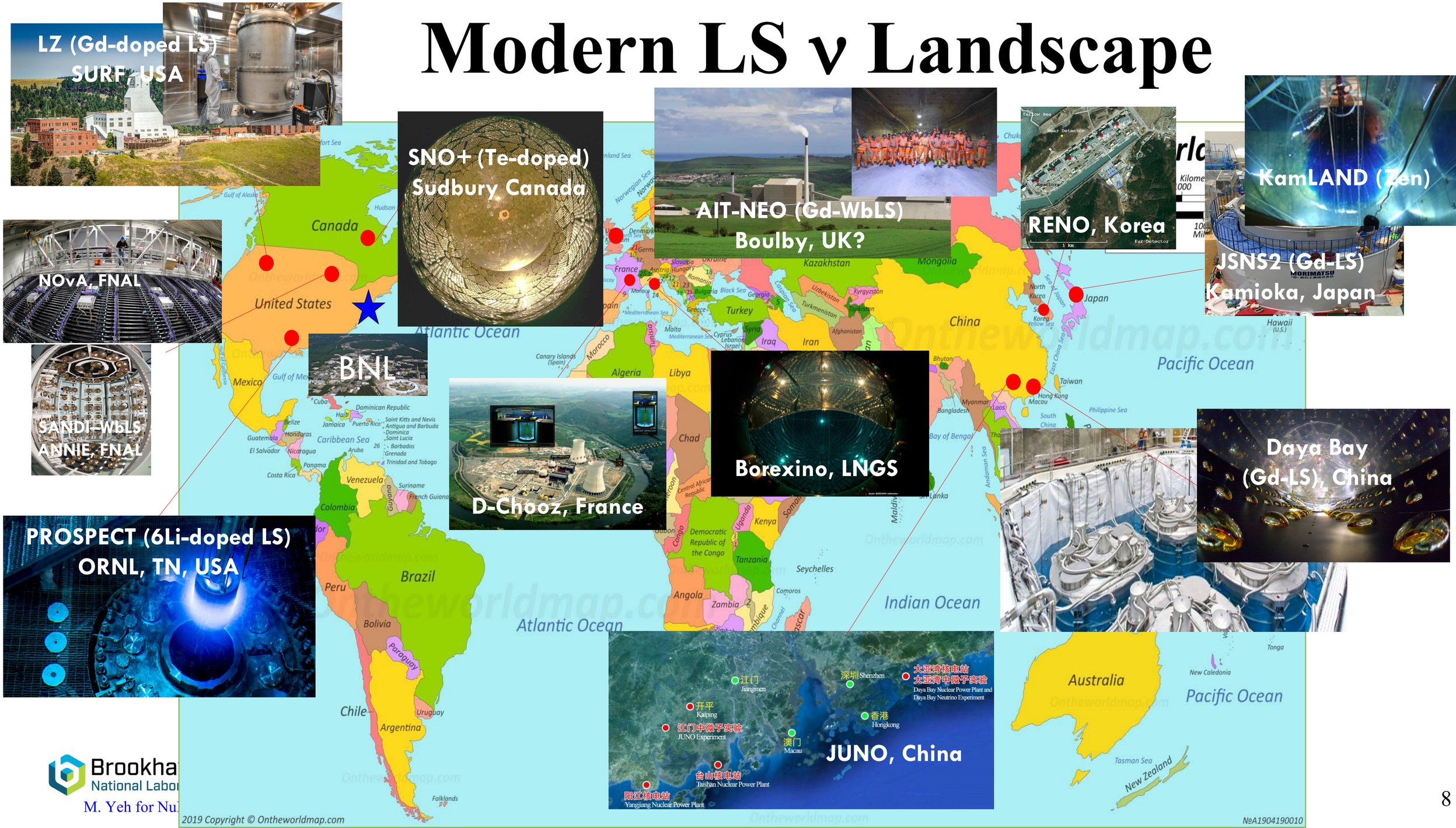
Modern LS ν Landscape



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Challenges and Developments of Next-generation Liquid Scintillator Detectors

- *Metal-doped techniques*
- *Scintillator directionality*

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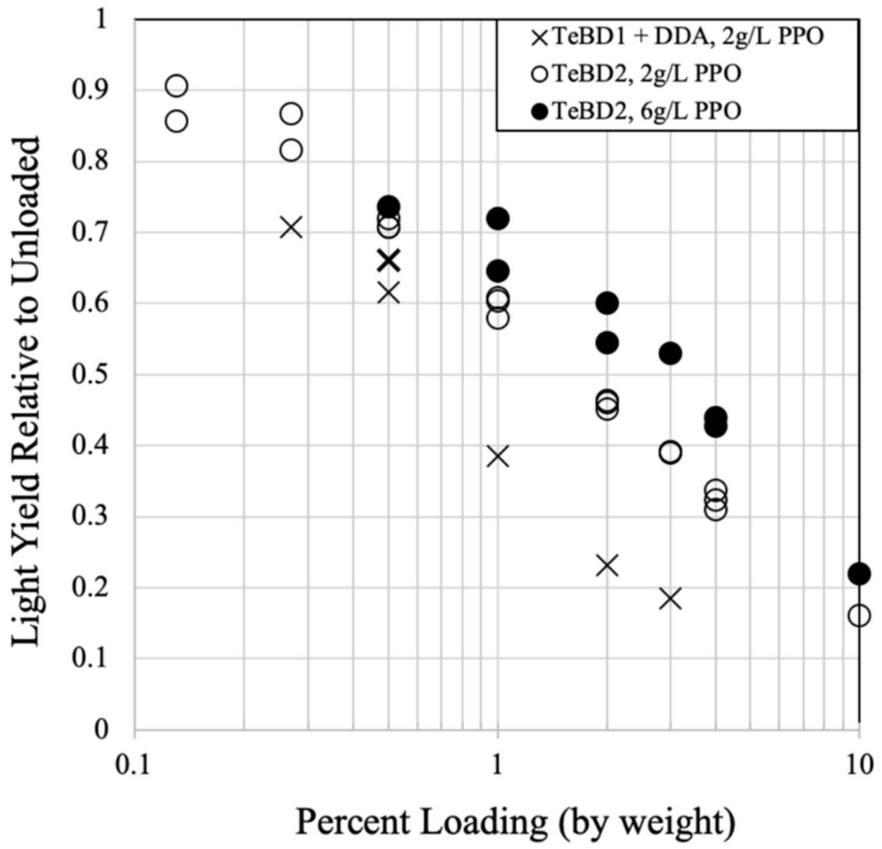
***Challenge-I**: addition of the inorganic metallic salt to the organic scintillator solvents*

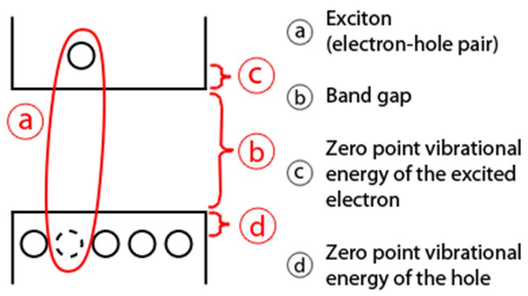
- **Conventional isotope loading techniques**
  - A mediator with high solubility for inorganic salt compounds as alcohols (Chooz)
  - Organometallic complex, which is soluble in the LS.
    - Carboxylates (Savannah River, LENS, Palo Verde, Daya Bay, RENO)
    - Diketone or phosphor-organic ligands (LENS, Double Chooz)
- **New isotope-loading techniques**
  - New organocomplex
  - Quantum dot
  - Metal-doped WbLS

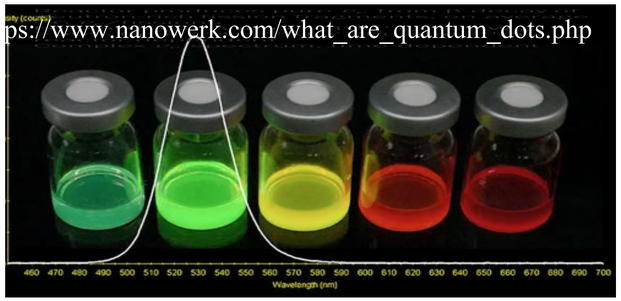
New Organocomplexing Ligands

- A method developed from 0νββ (SNO+) for higher loading of tellurium into liquid scintillator.
- An organocomplex using butanediol in conjunction with N,N-dimethyldodecylamine (DDA), which acts as a stabilisation agent.
- Stability of the loading has been demonstrated to be at least on the timescale of years; a highly scalable and economical approach.
- Further advances in purification techniques could provide a practical path to realising sensitivity to the non-degenerate normal mass ordering.



Quantum Dots



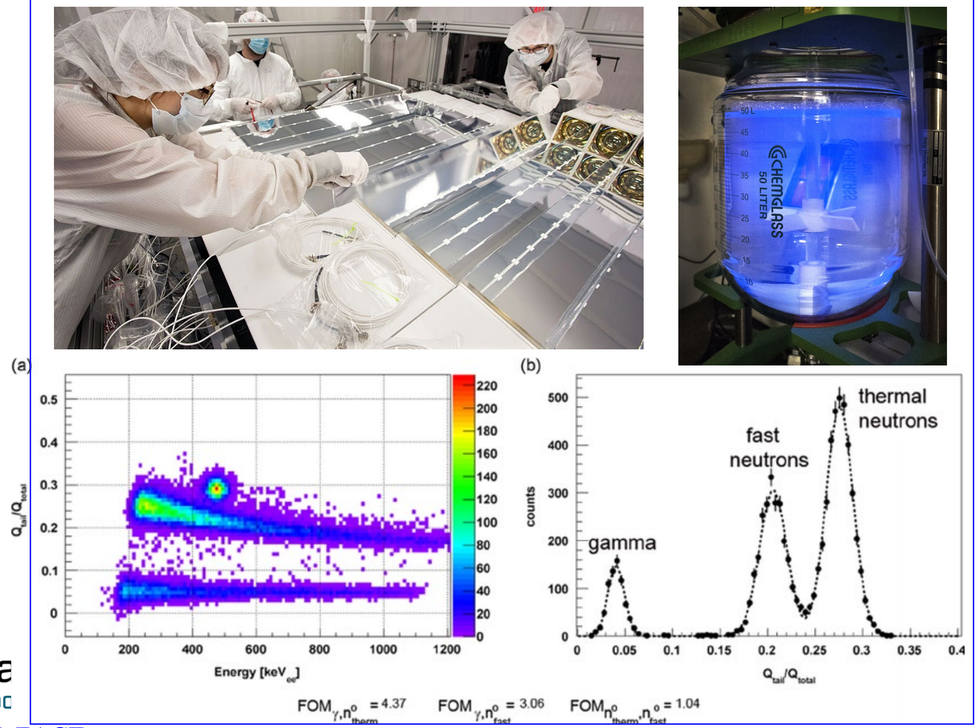


- Introduced from semiconducting nanocrystals in which the optical and electrical properties of the quantum dots are directly proportional to their size through resonance process (*tunable emission*).
- The most commonly used quantum dot cores are binary alloys such as CdS, CdSe, CdTe, and ZnS.
  - *neutron-enhanced isotopes (113Cd) and (106,116Cd) and Se, Te, and Zn, which are present in common quantum dot cores, $0\nu\beta\beta$ candidates.*
- Colloidal suspension instead of homogeneous mixing, which could cause aggregation in the concentrated solutions over long time scales.
  - *Mitigated by incorporating with chelating agents or WbLS surface active agents?*
- limitations in use for particle physics detectors are probably cost and availability in large quantity (ton).

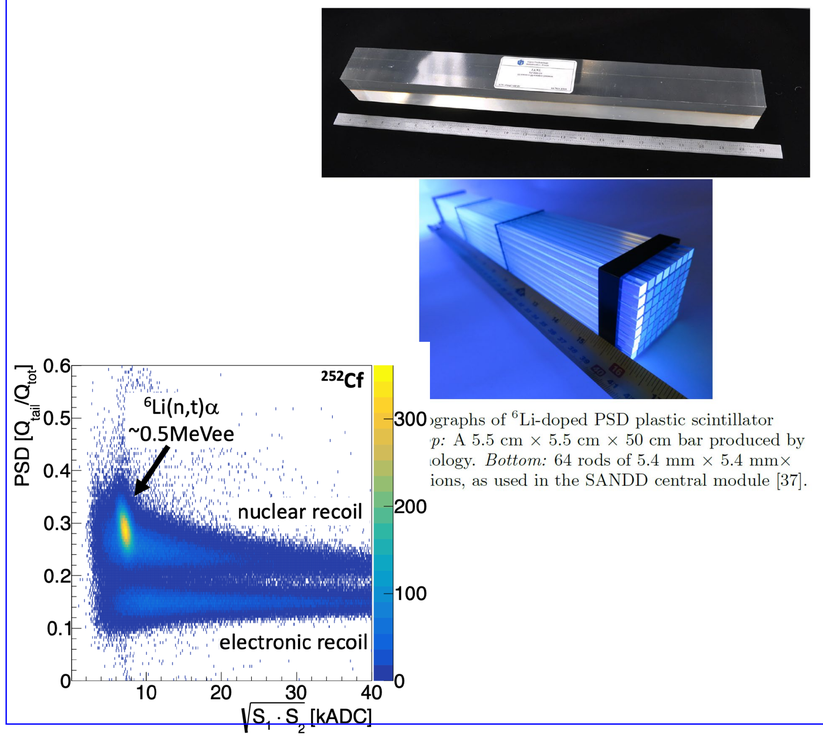
Metal-doped Water-based Liquid Scintillator

- A simpler approach to add the metal in aqueous solutions directly into liquid scintillators derived from WbLS principle
- User cases: Li, Gd, Te, K, Fe, W in several frontiers for neutrinos, nonproliferation, 0νββ, calibration, calorimetry
- A transformative technique for LSC cocktail (environmental and safeguard)

PROSPECT Li-doped LS



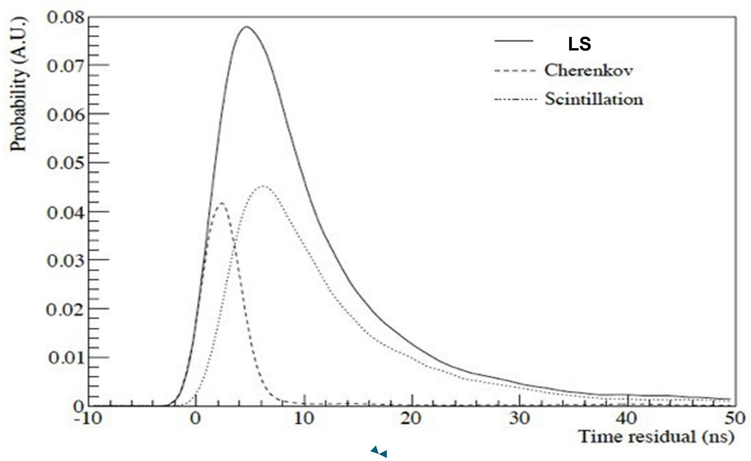
Li-doped Plastics

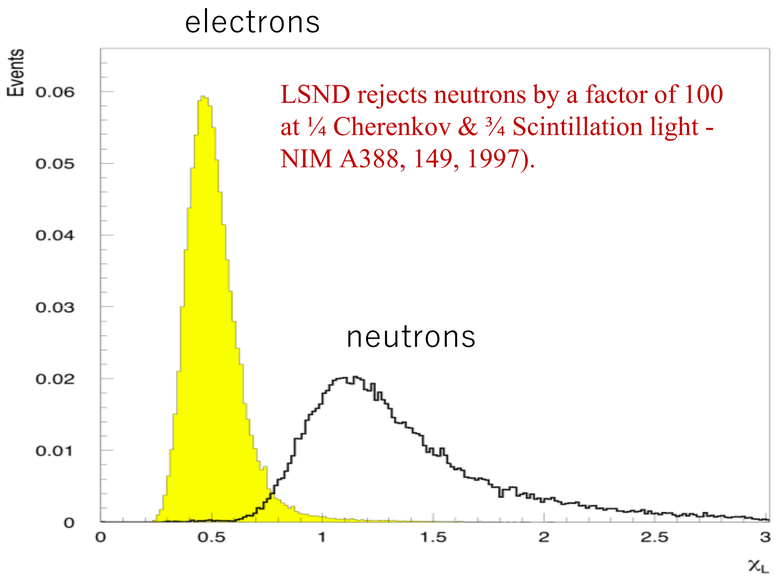


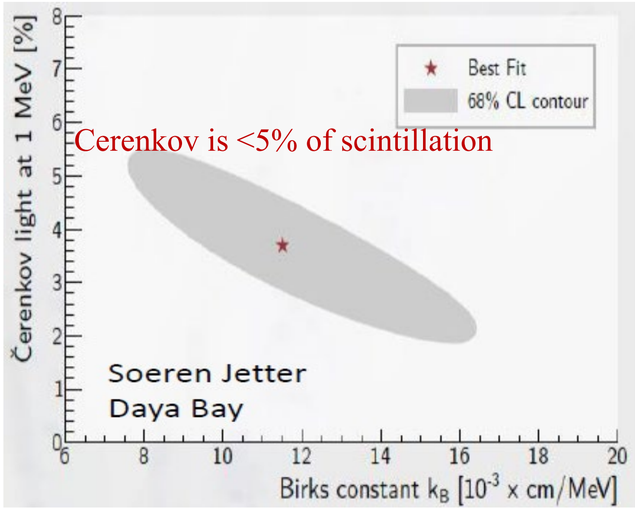
ographs of ^{6}Li-doped PSD plastic scintillator
p: A 5.5 cm × 5.5 cm × 50 cm bar produced by
ology. *Bottom:* 64 rods of 5.4 mm × 5.4 mm×
ions, as used in the SANDD central module [37].

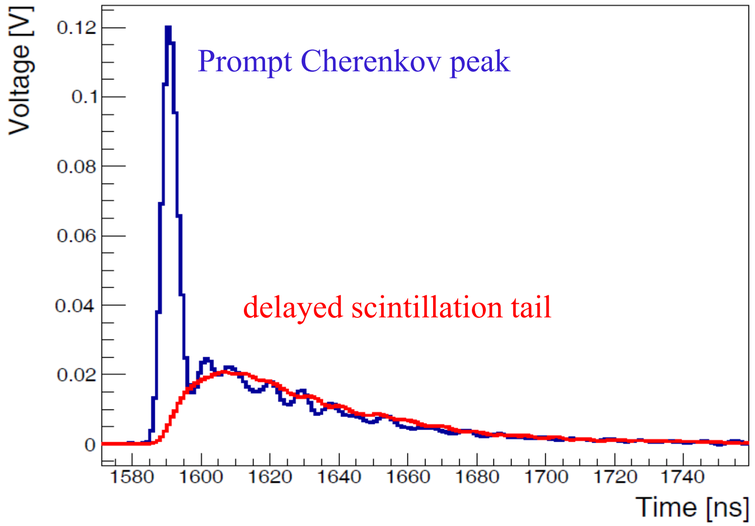
Challenge-II: Directionality

- Separation of Cherenkov from scintillation allows directional cut for particle ID to select/reject events
- Next generation LS detector development → directionality
  - ***Slow Scintillator**: Timing separation of slow scintillation from fast Cherenkov*
  - ***LiquidO**: Stochastic light confinement; lossless scattering*
  - ***Water-based Liquid Scintillator**: Cherenkov and scintillation detection*



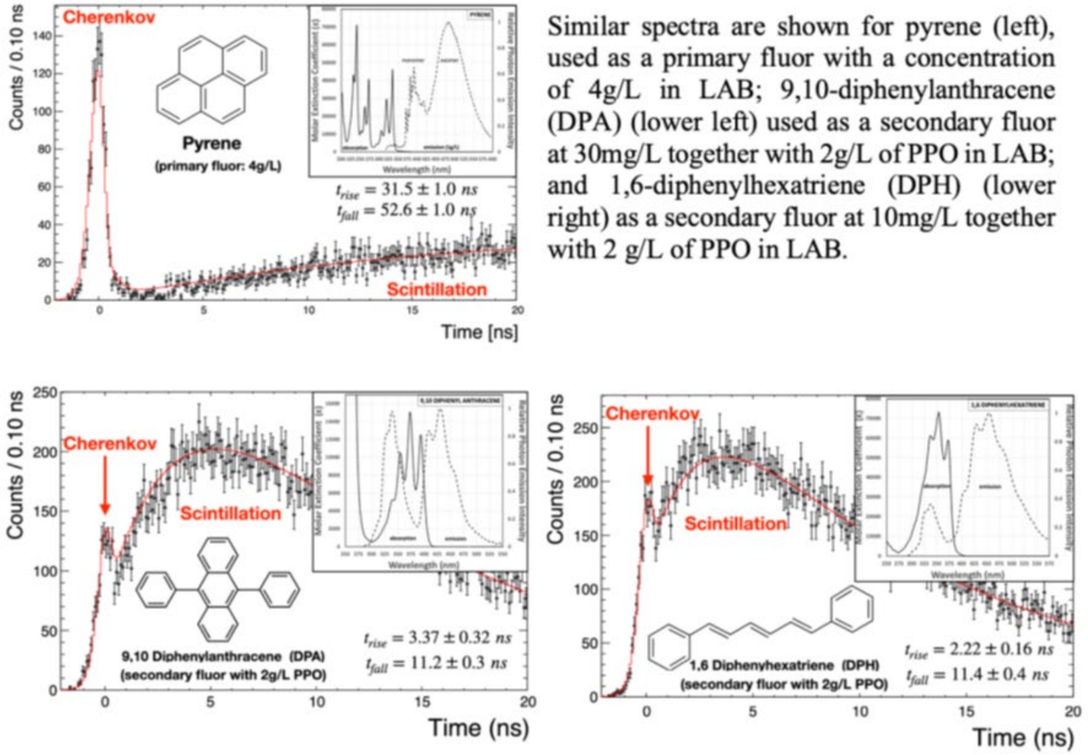






Slow Scintillator

- The properties of slow fluors or wavelength shifters to provide a means to separate Cherenkov light in time from the scintillation signal which allows for directional and particle ID information while also maintaining good energy resolution.
- Readily applied to existing and planned large-scale liquid scintillator instruments without the need of additional hardware development and installation.



Similar spectra are shown for pyrene (left), used as a primary fluor with a concentration of 4g/L in LAB; 9,10-diphenylanthracene (DPA) (lower left) used as a secondary fluor at 30mg/L together with 2g/L of PPO in LAB; and 1,6-diphenylhexatriene (DPH) (lower right) as a secondary fluor at 10mg/L together with 2 g/L of PPO in LAB.

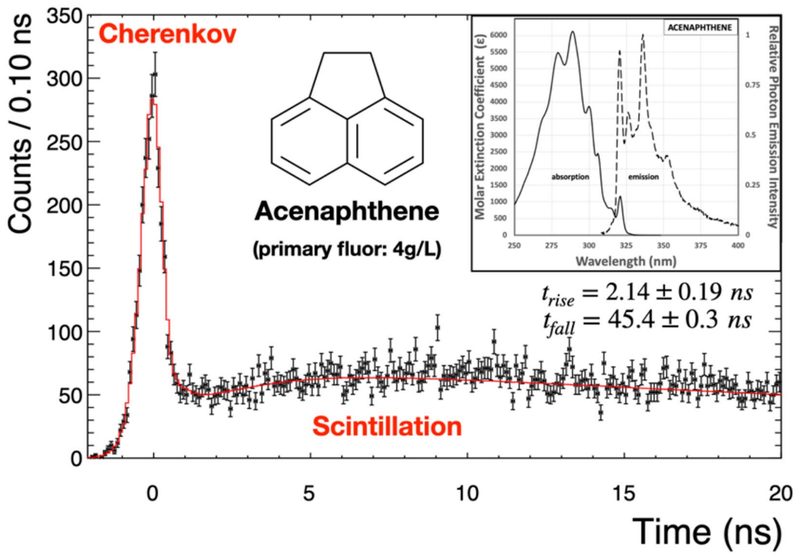
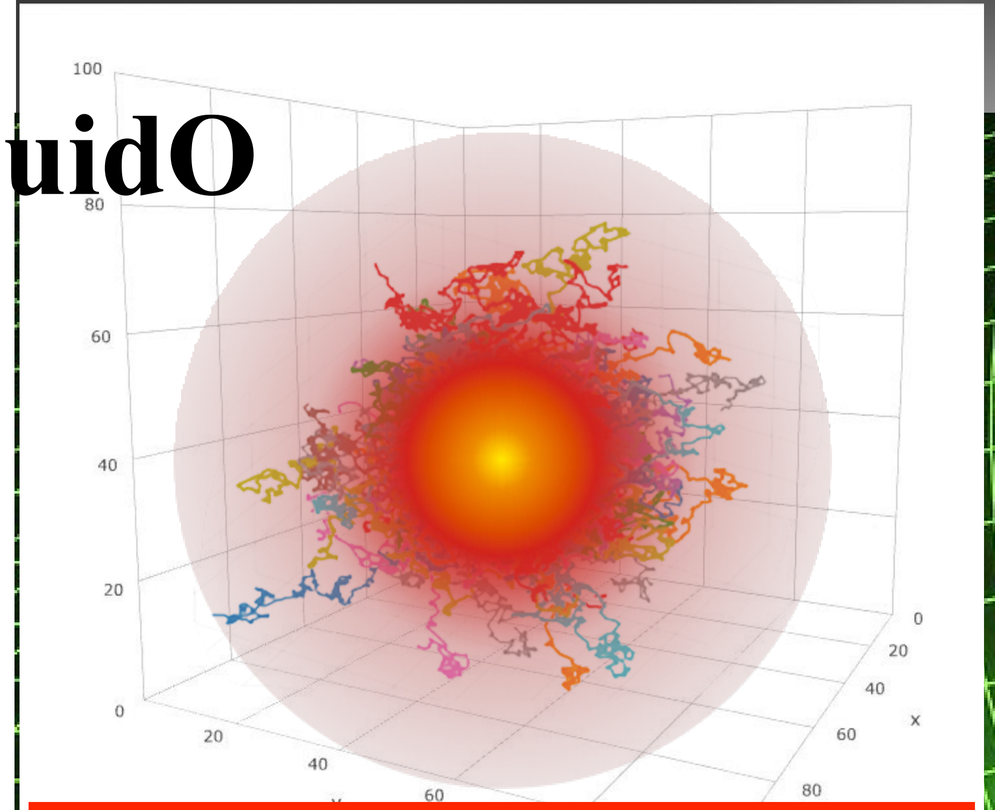


FIG. 1: Zoomed in time spectrum for 4 g/L acenapththene in LAB with clear Cherenkov peak.

LiquidO



LiqudiO→ photon's "random walk" (self-confinement)

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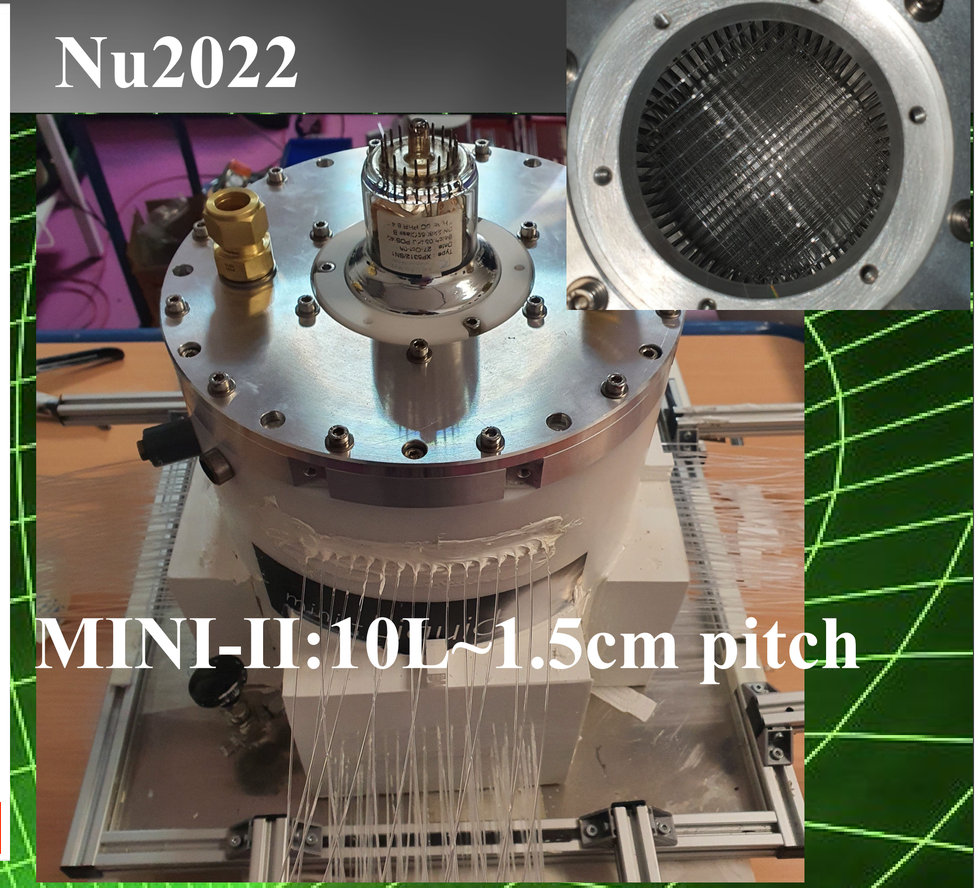
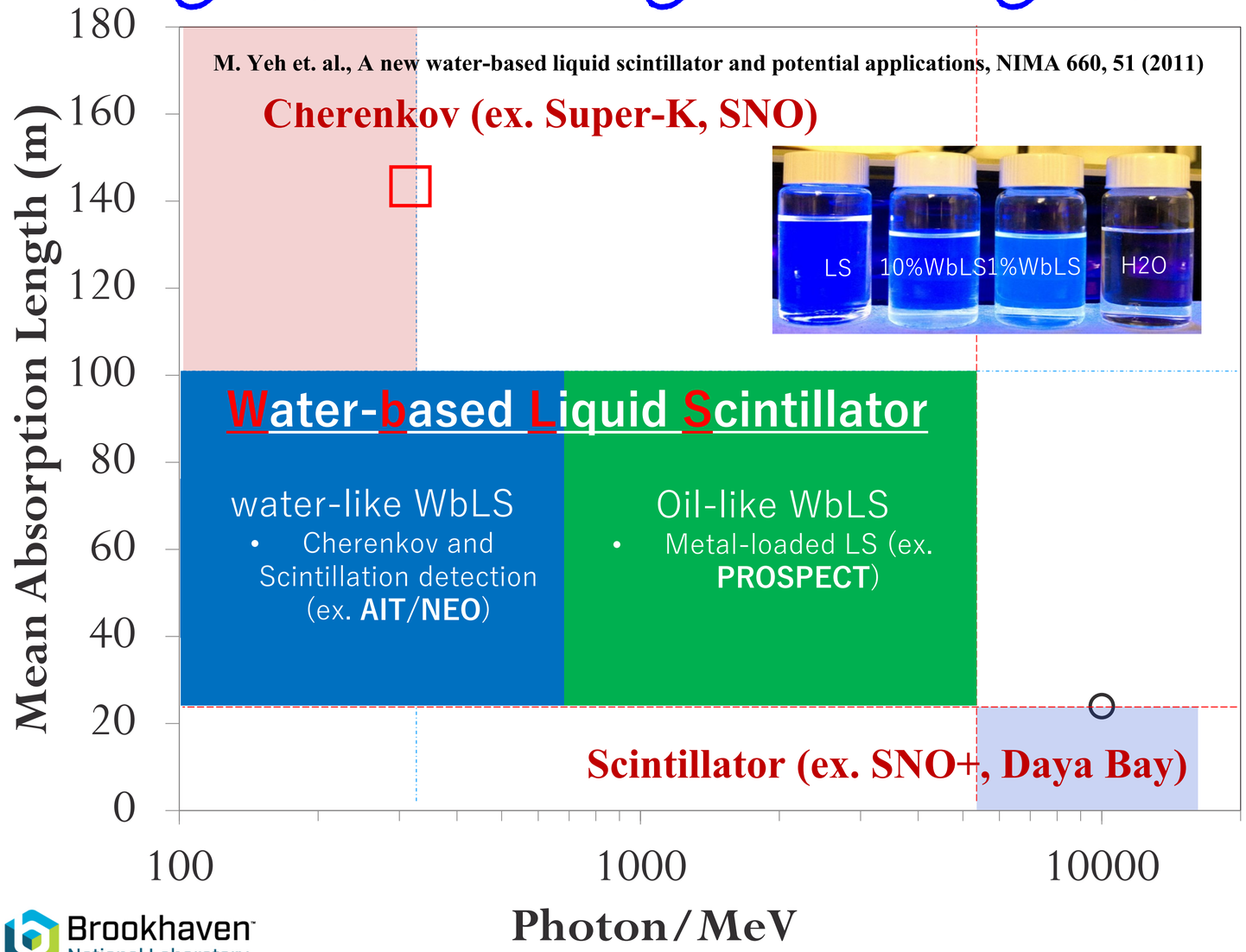


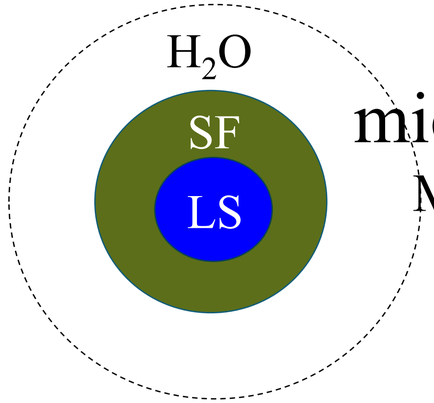
Transparency
λ(scattering)≥10m

Rayleigh & Mie Scattering
λ(scattering)≤1cm

Inducing light to a point…

If you always do what you always did, you will always get what you always got. -Albert Einstein





micelle introduces Mie + Rayleigh scattering

Stability and Scattering determined by

- dynamics equilibrium

  → Hydrophilic-Lipophilic Balance (HLB)

- parity match (functional group)

hundreds of surfactants available in market

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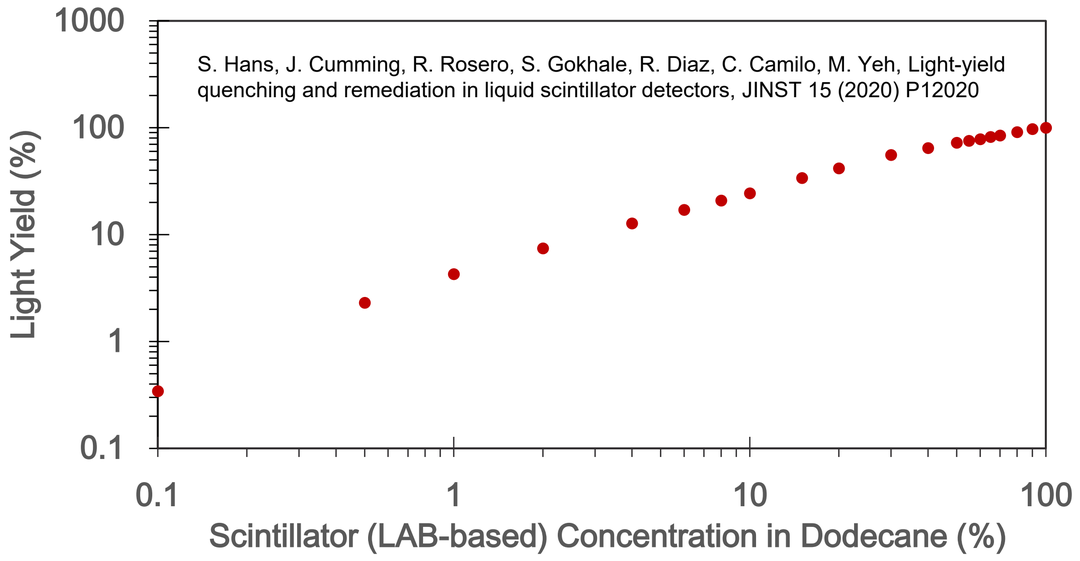
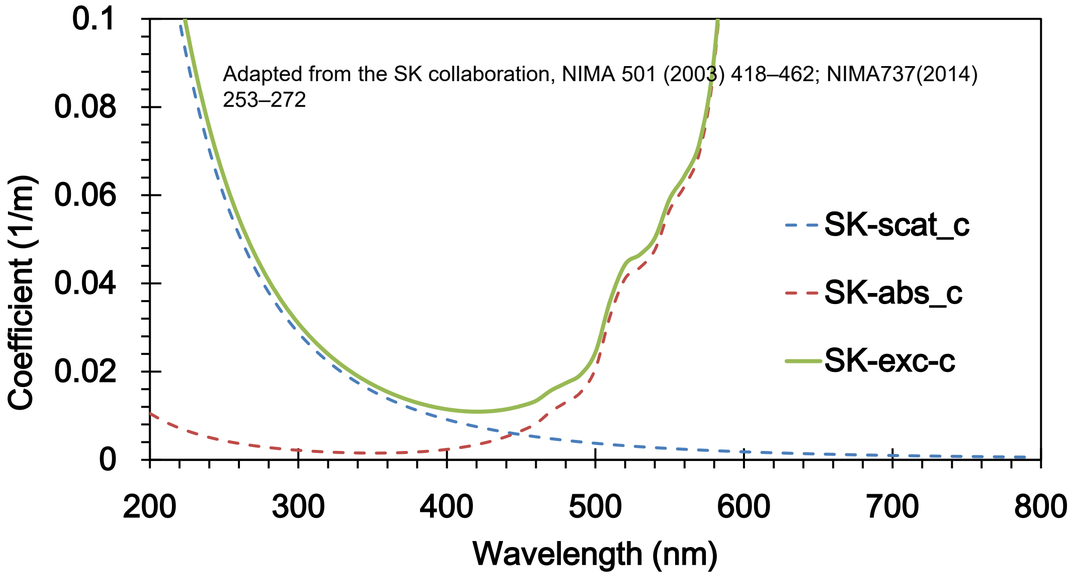
Water-based Liquid Scintillator

- a novel low-energy detection medium, bridging scintillator and water, to explore physics below Cherenkov threshold
- tunable scintillation light from ~pure water to ~organic.
- a directional scintillator detector capable of Cherenkov and scintillation detections
- viable to load a variety of metallic isotopes for varied physics applications
- enabling a rich physics program spanning topics in nuclear, high-energy, and astrophysics, and across a dynamic range from hundreds of keV to many GeV (ANNIE, WATCHMAN, THEIA25/100)





Theia: multipurpose neutrino detector

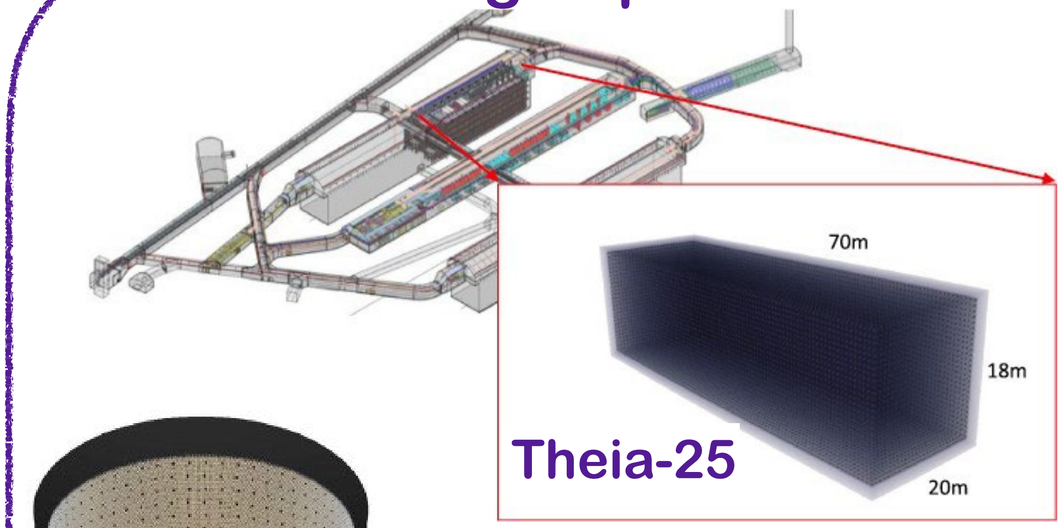
solar neutrinos (CNO, ^{8}B)

geoneutrinos

diffuse supernova neutrinos (DSNB)

supernova burst neutrinos

Design options



Large scale, multipurpose detector

- Baseline: 25ktonne (17kt FV)
- Ideal: 100 ktonne (70kt FV)

Scintillator fraction tunable depending on the physics goal ->staged approach

neutrino mass ordering

neutrino CP-violating phase δ

neutrinoless double beta decay

nucleon decay

- 10 countries
- 38 institutes
- 80+ collaborators

THEIA: An advanced optical neutrino detector Eur. Phys. J. C 80, 416

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Steps toward a WbLS detector at 10s ktons scale

- *High-energy Cherenkov + Low-energy Scintillation*
- *Reduced chemical hazards, not combustible*
- *Increased affordability*

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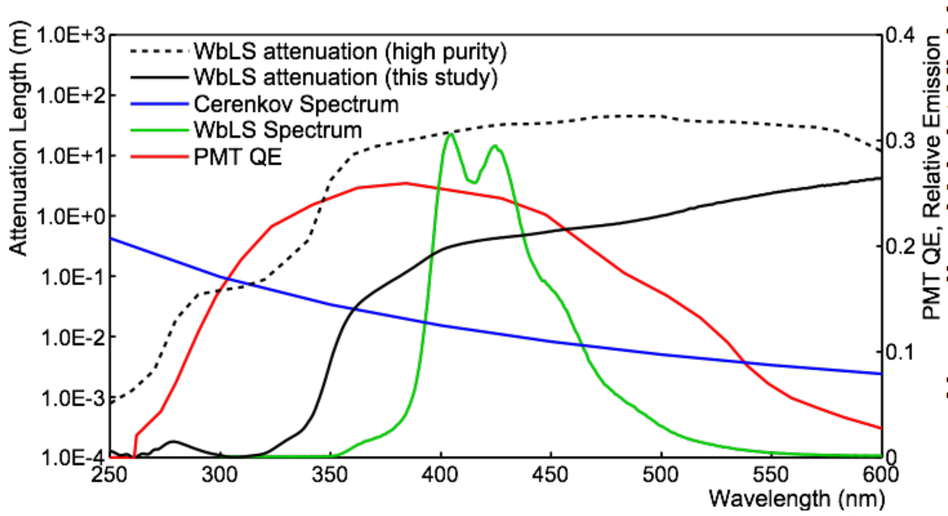
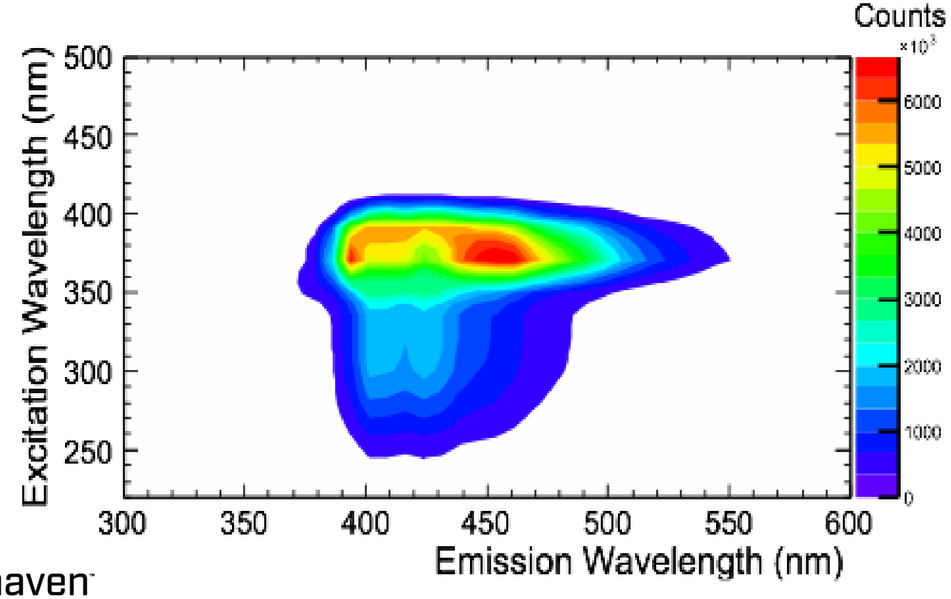
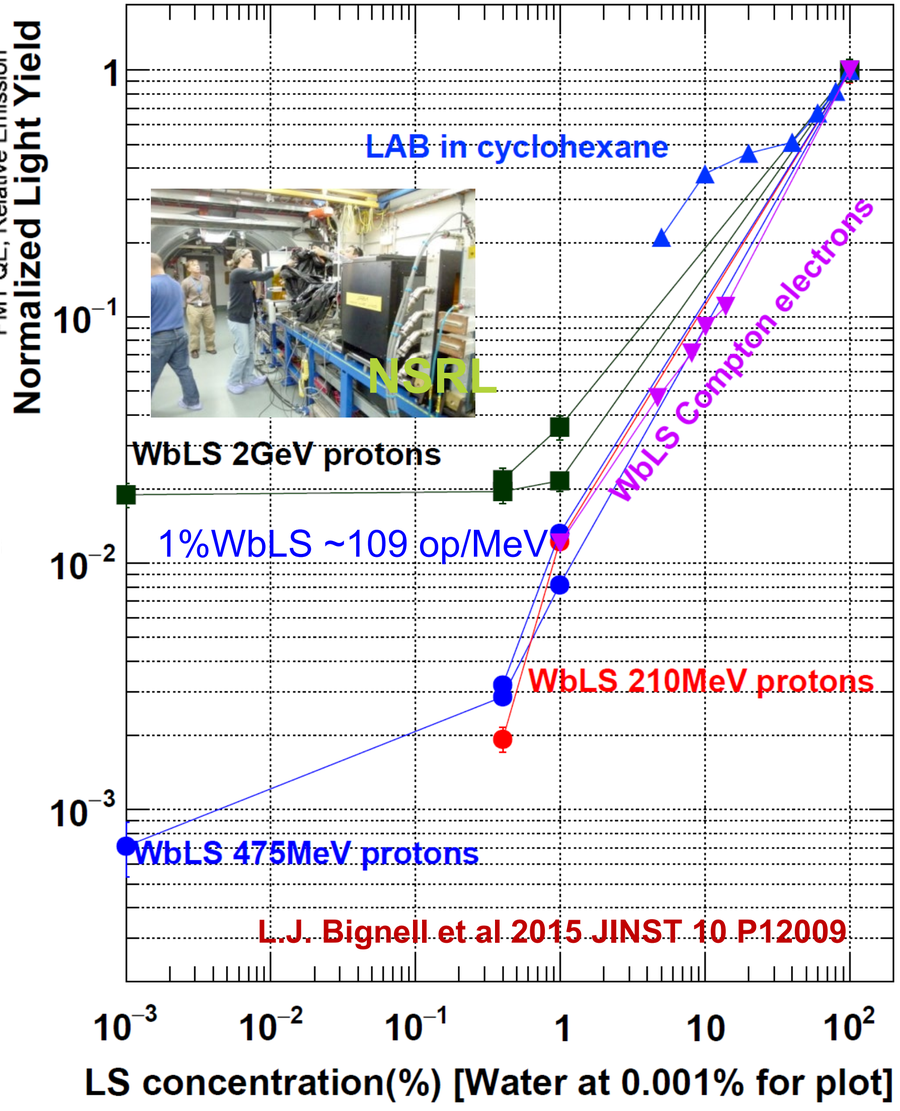
WbLS fundamentals







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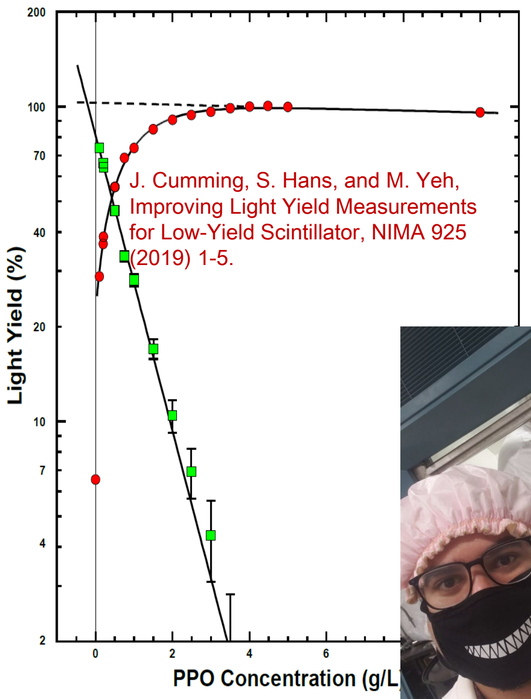
M. Yeh for NuFACT

Principle proven with characterizations

Scintillator Yield, *Č/S* Separation, Timing Structure, optical (absorption and scattering),…,etc.



LS Samples	τ_{rise} [ns]	τ_1 [ns]	f_1 [%]	τ_2 [ns]	f_2 [%]	τ_3 [ns]	f_3 [%]
0.5 g/L PPO	0.98 ± 0.04	7.0 ± 0.6	61	$16. \pm 1.$	34	80 ± 5	4.4
1 g/L PPO	0.92 ± 0.07	5.5 ± 0.3	72	13.7 ± 0.9	25	84 ± 2	3.7
2 g/L PPO	0.88 ± 0.05	4.3 ± 0.2	82	13.4 ± 0.8	15	86 ± 1	3.7
3 g/L PPO	0.83 ± 0.06	3.6 ± 0.1	84	13.4 ± 0.9	12	83 ± 2	4.0
5 g/L PPO	0.69 ± 0.03	2.81 ± 0.07	84	13.0 ± 0.8	12	80 ± 2	4.4
10 g/L PPO	0.49 ± 0.03	2.06 ± 0.04	77	11.2 ± 0.4	18	72 ± 2	5.1
30 g/L PPO	0.33 ± 0.12	1.15 ± 0.02	55	9.9 ± 0.5	38	64 ± 4	7.0
70 g/L PPO	0.26 ± 0.03	0.58 ± 0.02	38	10.5 ± 0.4	55	69 ± 4	7.1
90 g/L PPO	0.24 ± 0.02	0.48 ± 0.01	35	11.1 ± 0.1	58	72 ± 3	7.4

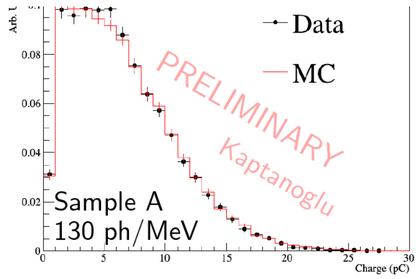
Table 1 Rise times (τ_{rise} in ns), decay times (τ_i in ns), and decay component fractions (f_i in %) are displayed for each pure LS sample, varying PPO concentration in LAB, from excitation with a pulsed X-ray excitation source, as shown in Figures 3 and 5. The error bounds listed are 95% confidence intervals measured by repeating this experiment; thus they account for any random error of sample positioning, data acquisition, and component fitting.

WbLS Samples	τ_{rise} [ns]	τ_1 [ns]	f_1 [%]	τ_2 [ns]	f_2 [%]	τ_3 [ns]	f_3 [%]
1% LAB/PPO	0.23 ± 0.06	2.00 ± 0.03	87	12 ± 1	6.8	110 ± 10	6.2
5% LAB/PPO	0.23 ± 0.04	2.00 ± 0.02	88	10.0 ± 0.6	6.6	106 ± 6	5.7
10% LAB/PPO	0.29 ± 0.03	2.22 ± 0.03	89	10.7 ± 0.9	6.0	102 ± 9	5.5

imes (τ_{rise} in ns), decay times (τ_i in ns), and decay component fractions (f_i in %) are displayed for each WbLS sample, from excitation X-ray excitation source, as shown in Figure 4.

D.R. Onken, F. Moretti, J. Caravaca, M. Yeh, G.D. Orebi Gann, E. Bourret, Time Response of Water-based Liquid Scintillator from X-ray Excitation, accepted by Materials Advances (2020).

- *Principal demonstrated in laboratory environment; characterization and analysis tools, developed over the past years*
- *WbLS-rhttps://orcid.org/**0000-0003-2244-0499**elated pubs:*



	Preferred LY [ph/ MeV]
0.1% GdWbLS	60
Sample A	130
Sample B	140
Sample C	160
Sample 4	140
Sample 5	160
Sample 6	130
Sample 7	~ 100

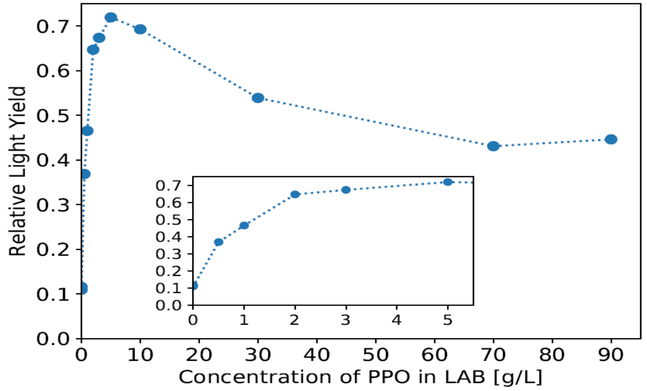


Fig. 6 Relative light yield for the pure LS samples, varying as a function of PPO concentration in LAB. Light yield values are presented as a fraction of the EJ-301 light yield. The inset focuses on concentrations ≤5 g/L PPO.

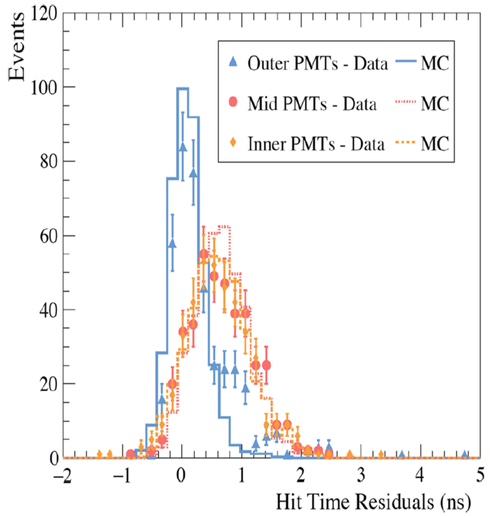
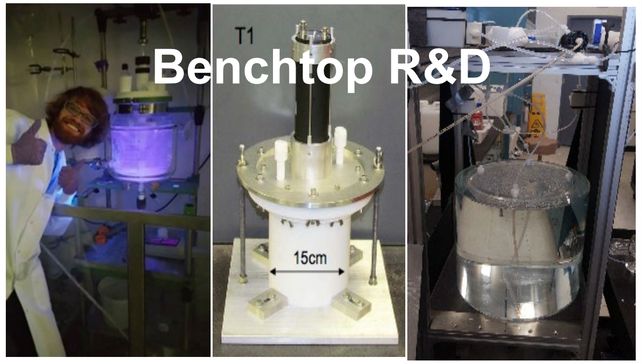
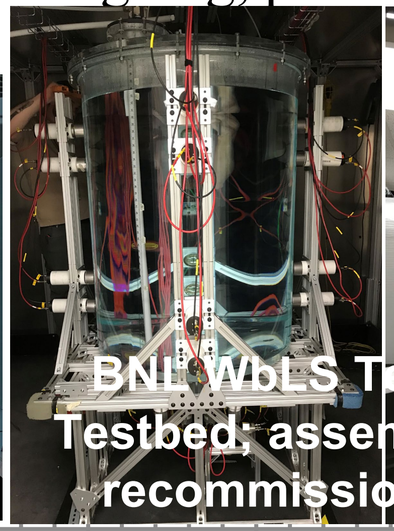


Fig. 10 Hit-time residual distributions in LAB/PPO for data and MC
J. Caravaca, F.B. Descamps, B.J. Land, M. Yeh, G.D. Orebi Gann, Cherenkov and scintillation light separation in organic liquid scintillators, Eur. Phys. J. C (2017) 77:811.

Advancing WbLS R&Ds: BNL development

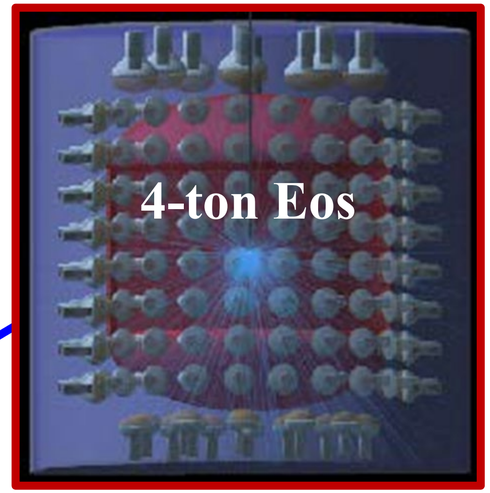
- LS research laboratories with state-or-art equipment
  - Bench top R&Ds and mid-scale (10-100 liter) study
  - **Host students and collaborators >5 per year**
- Ton-scale liquid scintillator production facility (commissioned in FY19; user cases → PROSPECT and LZ)
- 1-ton Testbed (1TBNL, commissioned in FY22)
- 30-ton Demonstrator (30TBNL, renovation ongoing, plan to commission in FY23)

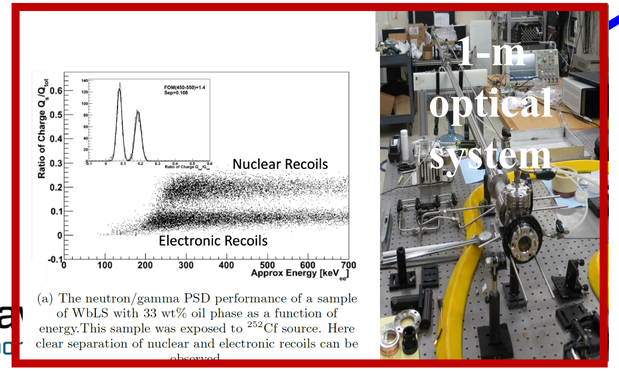




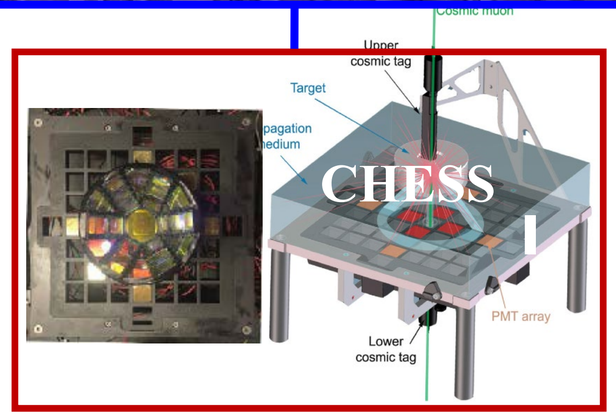
2010 2011 2012 2013 2014 2015 2016 2017 2018 2019 2020 2021 2022 2023 2024

Advancing WbLS R&Ds: other national labs & universities





(a) The neutron/gamma PSD performance of a sample of WbLS with 33 wt% oil phase as a function of energy.This sample was exposed to ^{252}Cf source. Here clear separation of nuclear and electronic recoils can be observed.

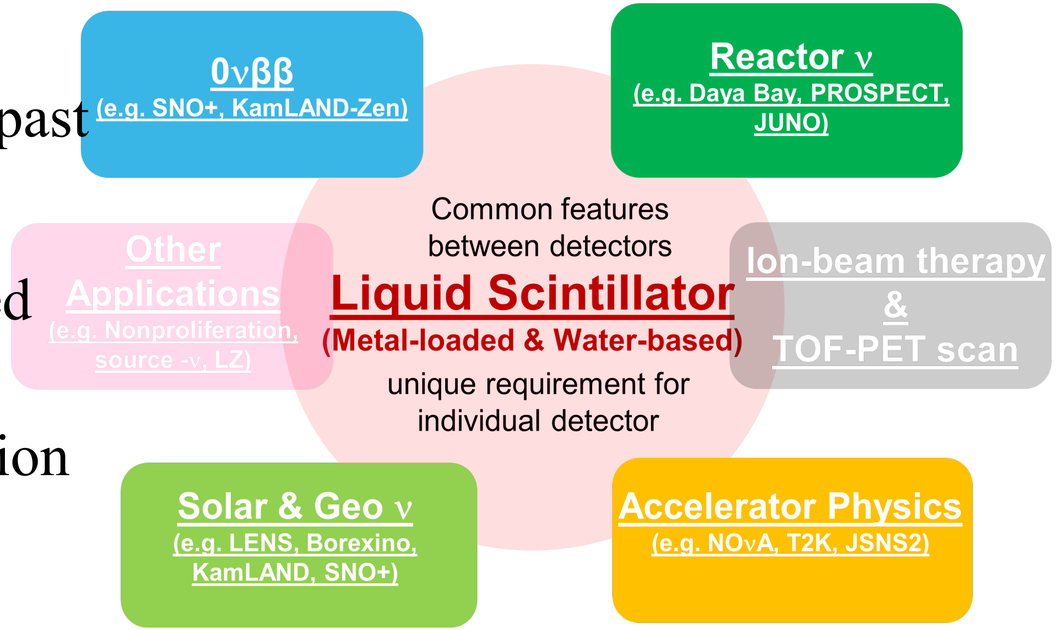


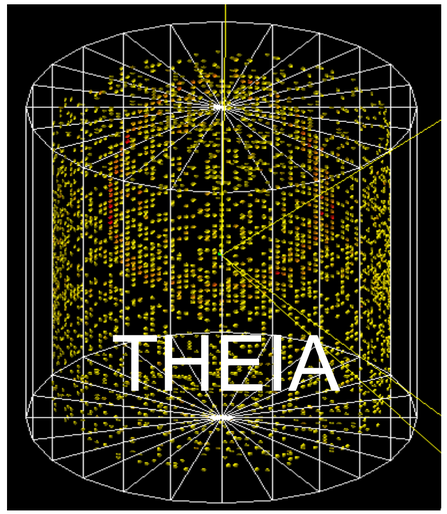
WbLS Research, Operation, and Progress

- Complete WbLS mixing and deployment in ONE step within the detector vessel
  - In-situ sequential mixing scheme
    - *Direct injection of scintillator materials into water*
    - *Remove on-site complications to save labor & space*
  - Proven at 100-liter scale; towards prototyping tests
- In-situ circulation and purification system
  - Nanofiltration system separates oil from water and sends each to purification
  - Benchtop test successfully demonstrates >95% (~ppm) efficacy; ready for prototyping tests
- Metal doping in WbLS successfully deployed/demonstrated
  - PROSPECT (Li), WATCHMAN (Gd), 30%W for calorimetry?
- Low material cost; affordable for 10s kton detectors:
  - 10-ton materials ($4k per ton) for a 1000-ton detector (99% water)

Summary

- Scintillator research has been largely advanced over the past decade
- WbLS is a new scintillation material along with advanced development of instrumentation, such as dichroicon, nanofiltration, LAPPD…, etc., building for next-generation experiments (flexible doping, LS% or Metal, allowing a phased program, i.e. THEIA)
- WbLS R&D is advancing with several ton-scale prototypes
  - 1TBNL, ANNIE in 2022
  - 4TEos, 30TBNL in 2023/24
- Readiness of deployment for a kiloton scale WbLS detector in 2024





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