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Scintillating Bolometer Crystal Growth and Purification for Neutrinoless Double Beta Decay Experiments

DOE Contract: DE-SC0015200, SBIR Phase IIA 5/28/2019 - 2/27/2022

RMD Principal Investigator: Michael R. Squillante

DOE Technical Contact: Michelle D. Shinn

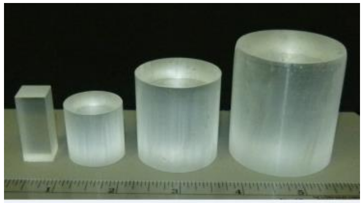
RMD Team: Josh Tower, Huicong Hong

MIT: Lindley Winslow, Joe Johnston

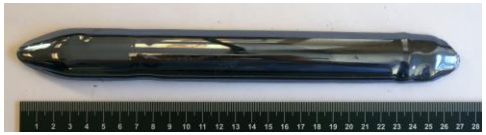
August 18, 2021

RMD Basic and Applied Research and Development

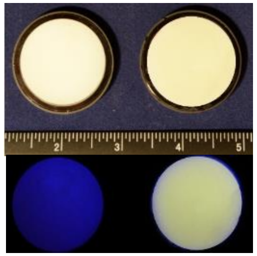
Materials Science



Scintillators



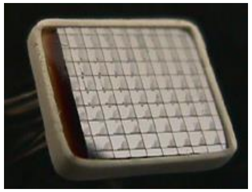
Semiconductors



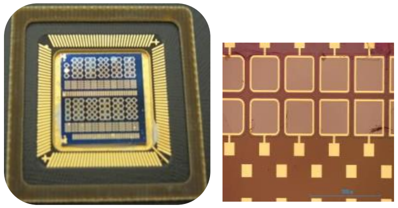
X-ray Imaging Screens

Ceramic Lasers and IR windows

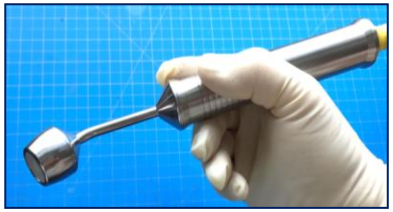
Sensors



APDs SSPMs Photosensors

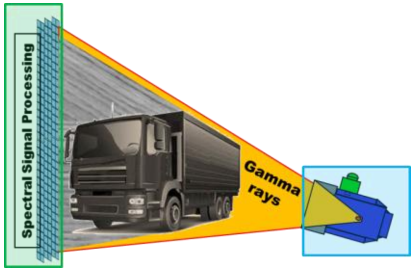


Wide Band Gap Geiger Photodiodes



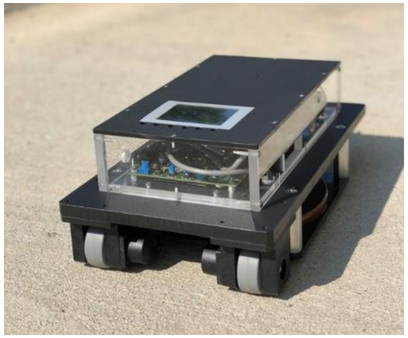
Surgical Beta-Probe

Instruments & Systems



HiRIS – High Resolution Imaging System

Hermes G/n w/ isotope ID



Robotic nuclear power plant concrete analyzer

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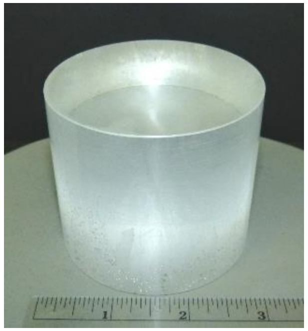
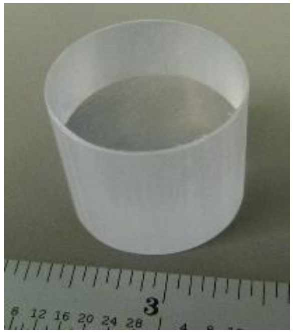
Commercial Products Based on RMD's Gamma-Neutron Scintillators

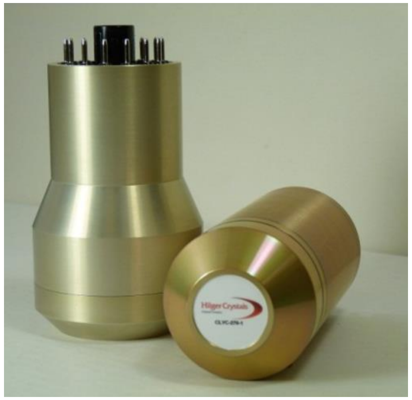


CLYC Crystal



CLLBC Crystal

Scintillation detectors



CLYC + PMT

CLLBC + SiPM

Commercial Instruments with RMD Detectors

Kromek D5 RIID

FLIR R440 RIID

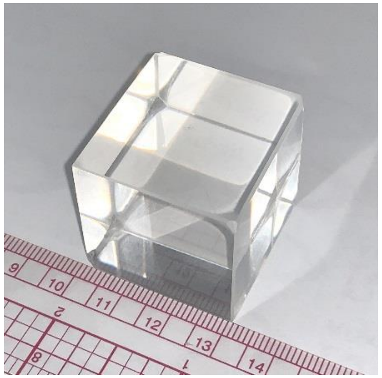
ANSTO CORIS360 Imager

Thermo-Scientific RIIDEye

Thermo-Scientific RadEye SPRD-GN

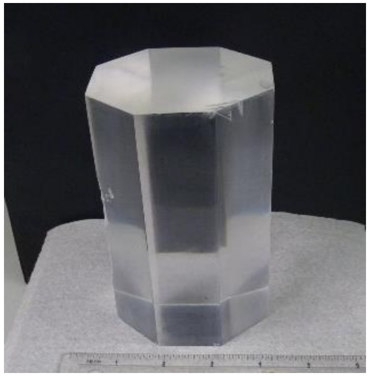
Low-Background Crystals for Nuclear and High-Energy Physics

Li_2MoO_4
Scintillating Bolometer



for neutrinoless double-beta decay

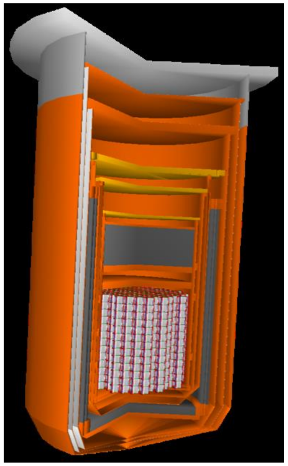
NaI
Room T Scintillator



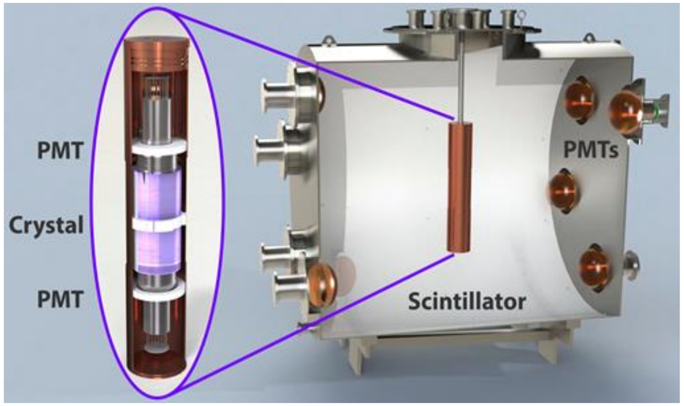
for dark matter search

Zinc
Superconducting Bolometer

for neutrino scattering



CUPID



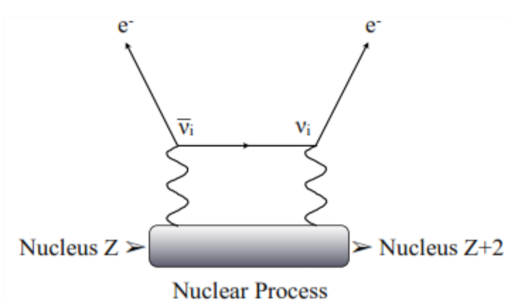
SABRE (now at LNGS)

RICOCHET

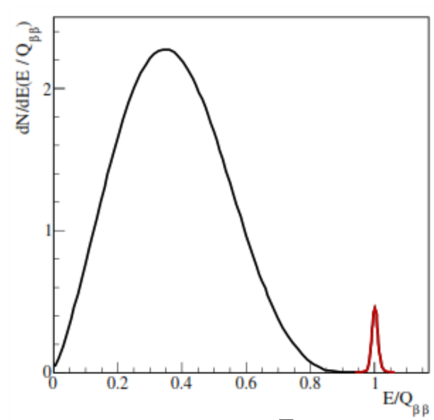
Understanding the Neutrino

- A key goal of Nuclear Physics is elucidating the nature of the neutrino
  What are the masses of the neutrino mass eigenstates?
  Is the neutrino its own antiparticle?
- The answers to these questions can change the Standard Model of physics.
- Searching for **neutrinoless double beta decay** (0νββ) is one of the highest priority experiments to answer these questions.
- A next generation experiment to search for 0νββ is **CUPID**: **CU**ORE with **P**article **Id**entification.
  - CUPID will use Li_2MoO_4 (LMO) scintillating bolometers.

0νββ spectrum



0νββ process



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Selection of Isotopes with Double-beta decay

Candidate Isotopes for 0νββ Experiments

element	isotope	end point energy (MeV)	% abundance
Ca	48	4.271	.187
Nd	150	3.367	5.6
Zr	96	3.35	2.8
Mo	100	3.034	9.7
Se	82	2.995	8.8
Cd	116	2.802	7.5
Te	130	2.527	24.6
Xe	136	2.457	8.9
Ge	76	2.039	7.8

^{100}Mo half-life = 7.8×10^{18} y
^{82}Se half-life = 0.97×10^{20} y

Requirements for isotope

1. Must decay by double beta process.
2. Good natural abundance and ability to enrich.
3. High endpoint energy (above 2.6 MeV ^{232}Th gamma ray).
4. Major constituent in a scintillating crystal.

Li_2MoO_4 Scintillating Bolometer

- Is both the source and detector of 0νββ
- Detects heat and light signals simultaneously.

***Scintillating Bolometers** are needed for better particle discrimination and background reduction*

Phase IIA Technical Objectives

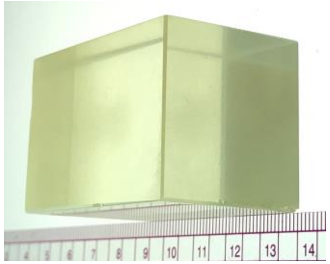
The goal is to complete the research and development needed to implement production of Li_2MoO_4 (LMO) scintillating bolometer crystals suitable for neutrinoless double-beta decay experiments.

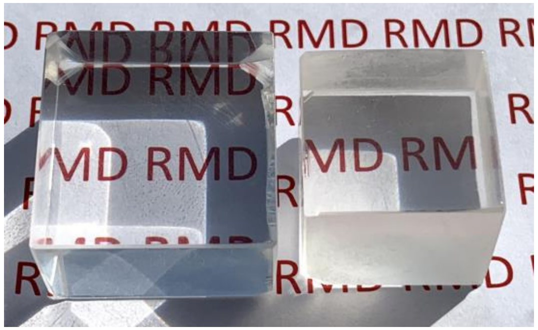
- Synthesize and purify Li_2MoO_4 from the high purity raw materials
- Grow single-crystal ingots using Czochralski to fabricate 45 mm cube detectors
- Develop processes for shaping and polishing crystals to maintain radio-purity
- Deliver detector crystals to the CUPID Collaboration for cryogenic evaluation. Scintillating bolometer testing includes all operational characteristics, such as light output and radioactivity background.
- Grow LMO using isotope enriched ^{100}Mo and produce full-spec detectors to qualify as a supplier for the CUPID experiment.

LMO Synthesis and Purification

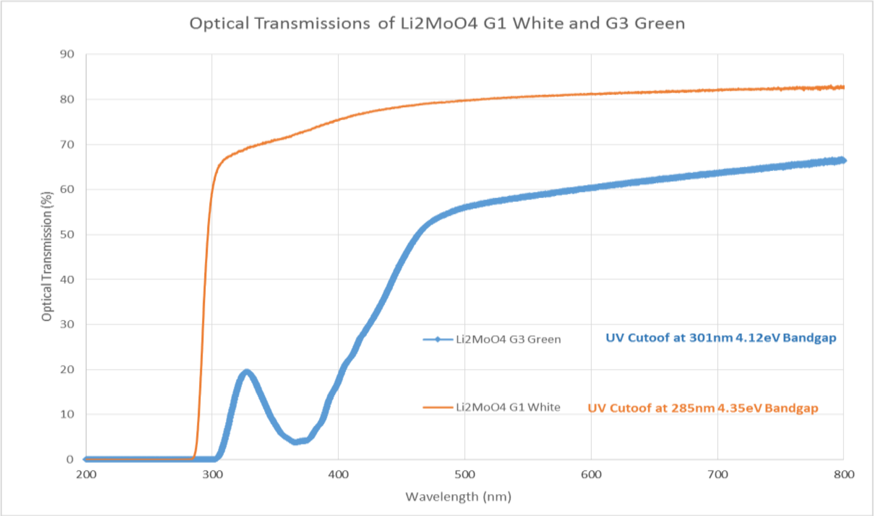
- Start with best purity raw materials
  - Good sources of the chemicals identified in previous phase
- MoO_3 (99.9995%) + Li_2CO_3 (99.99%) High Purity Powders
  - $MoO_3 + Li_2CO_3 \rightarrow Li_2MoO_4 + CO_2$

Greenish or brownish crystals can result if best quality materials are not used.





Optical transmission is good indication of crystal purity



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Czochralski Growth of Li_2MoO_4

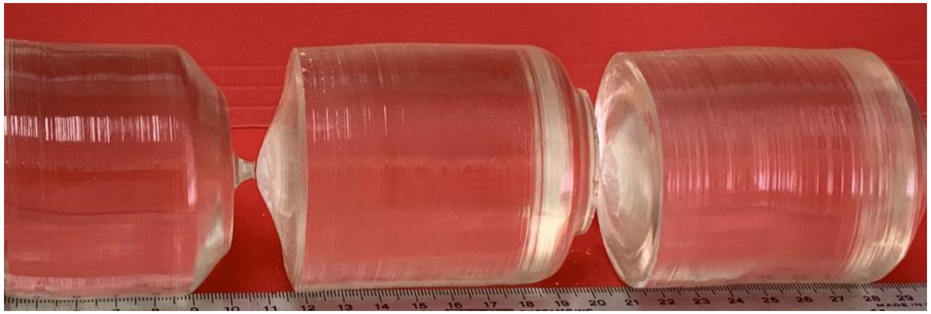
July 2019

~4 cm OD
~200 grams

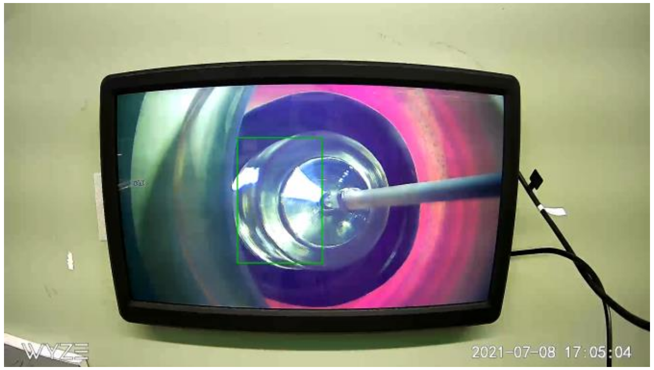
June2021

65mm OD x 65mm
813 grams

New CZ System for LMO



Repeatable growth process

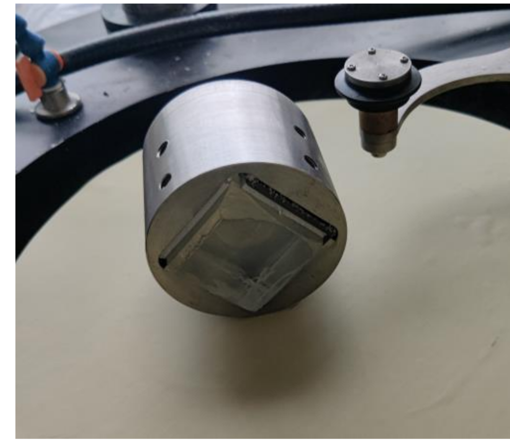


Growing crystal

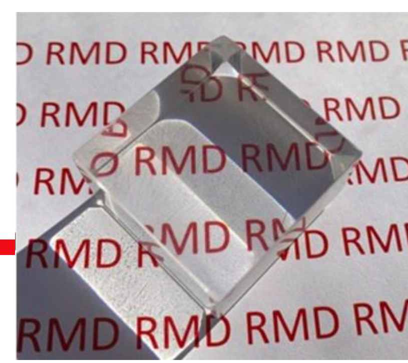
Fabrication of LMO Cubes for CUPID

Diamond wire saw for cutting 45 mm cubes

Special polishing fixture built for 45 mm cubes

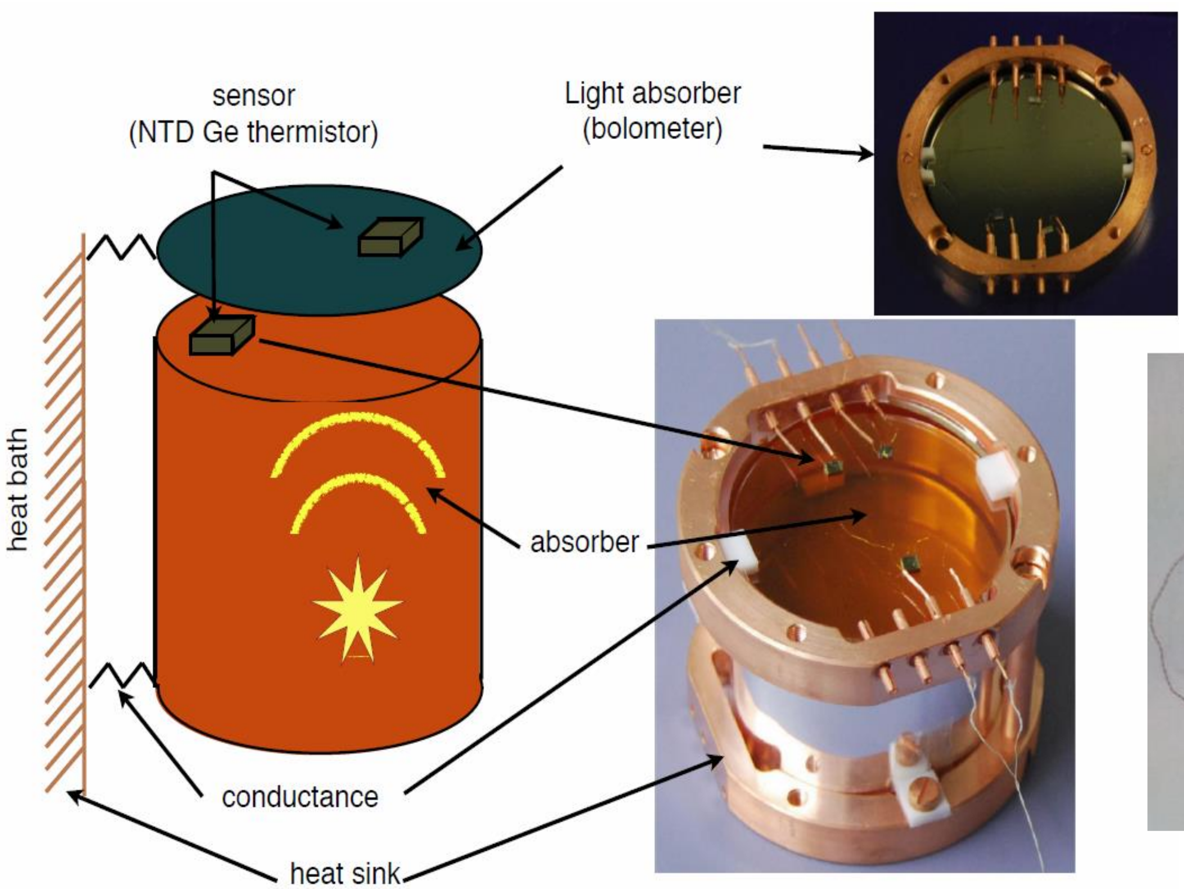


Final polishing and surface cleaning done in clean radon-free atmosphere using low-radioactivity materials.

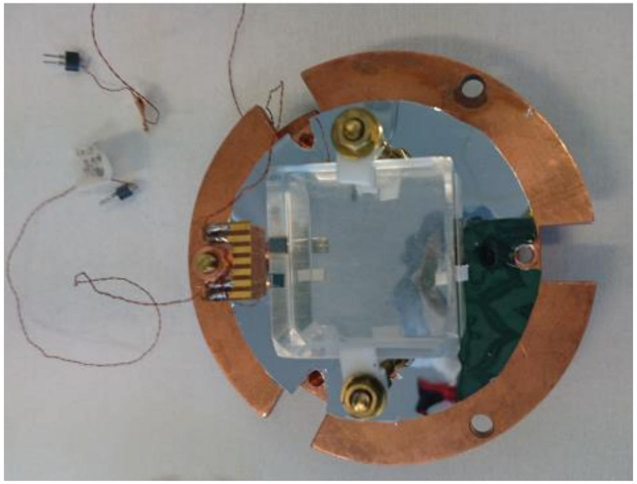


Cryogenic Testing of Scintillating Bolometers

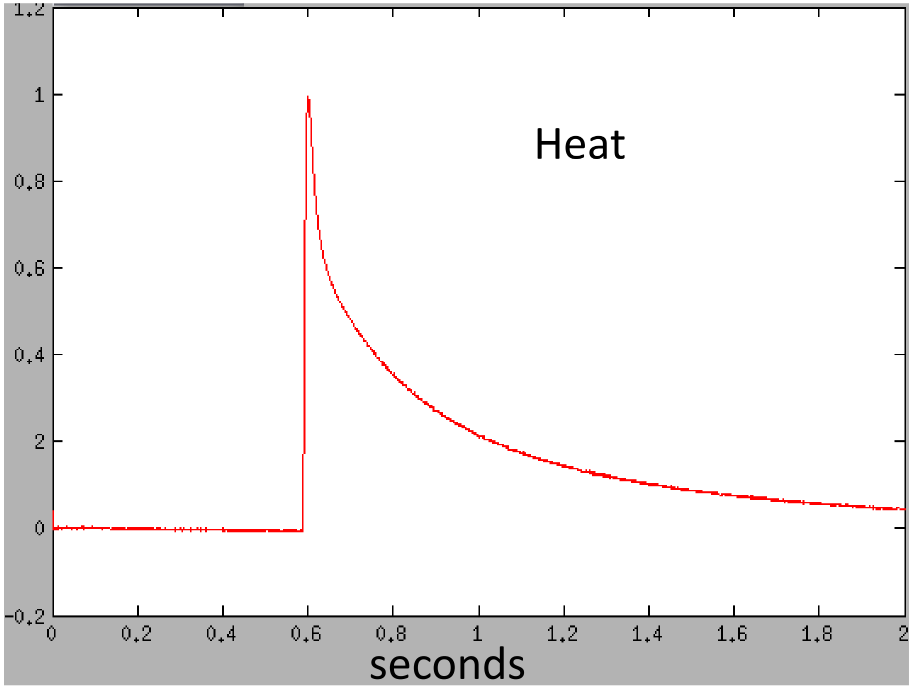
Above ground cryogenic testing by MIT at CSNSM

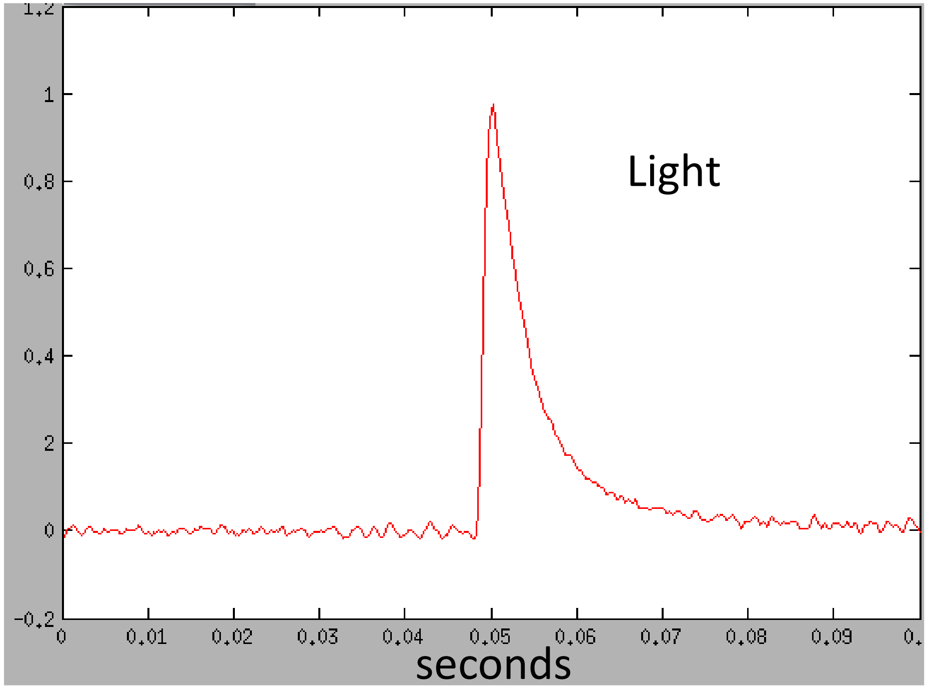


- Samples held at ~ 20 mK for multi-day testing.
- Light and heat pulses measured separately.



Mean light and heat pulses from LMO





Light pulse is ~ 100x faster than heat.

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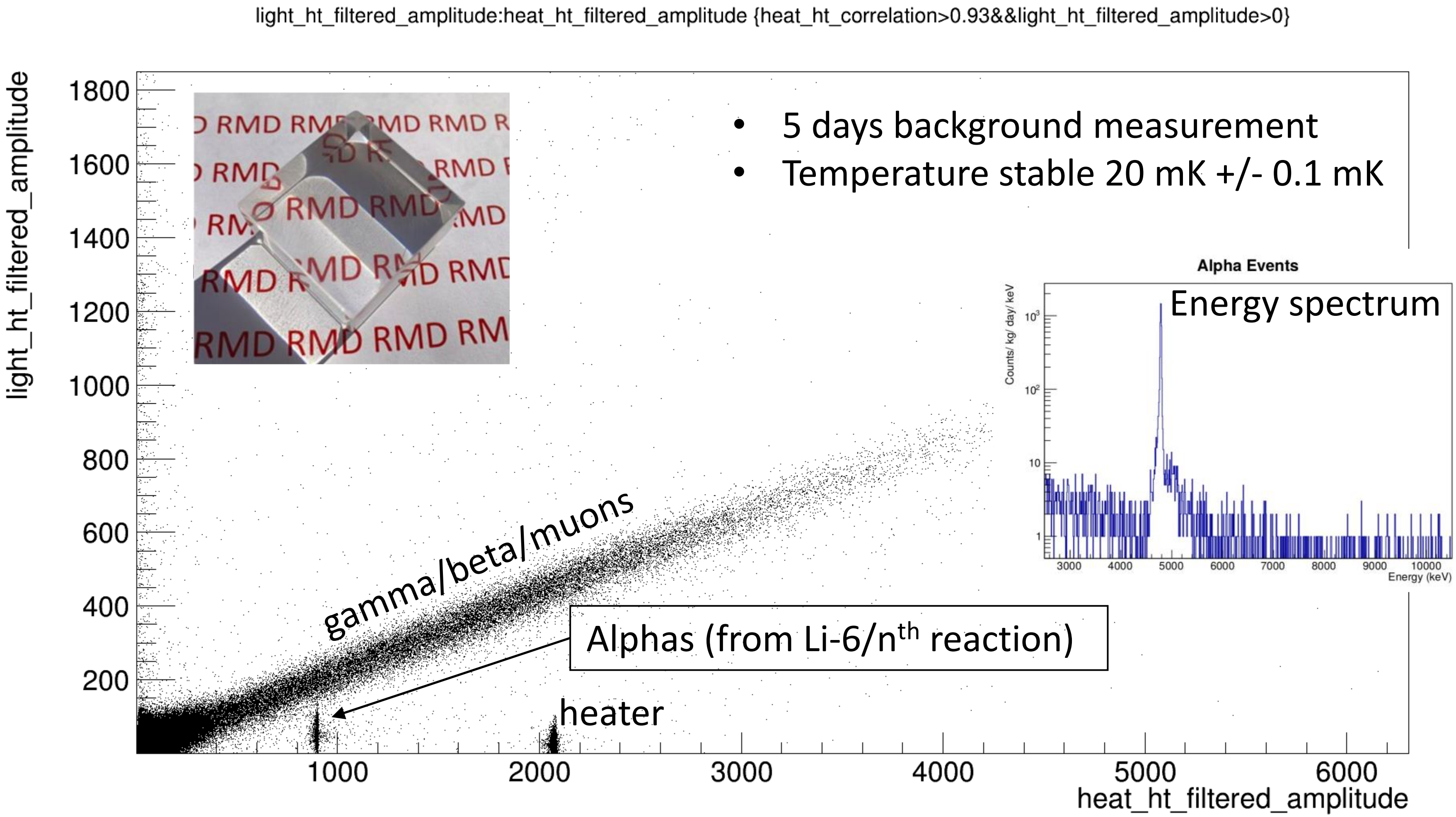
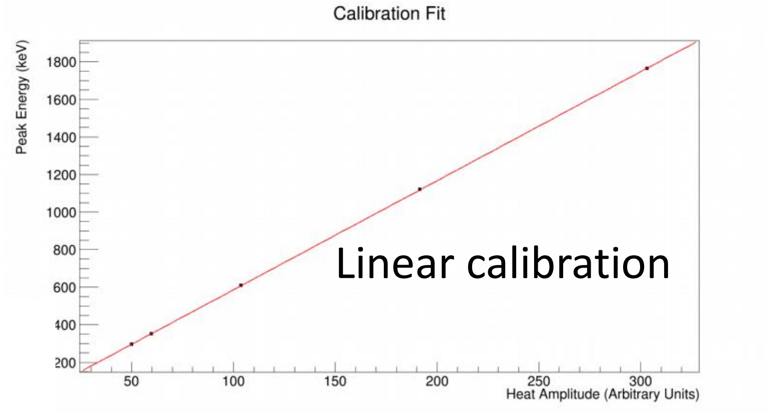
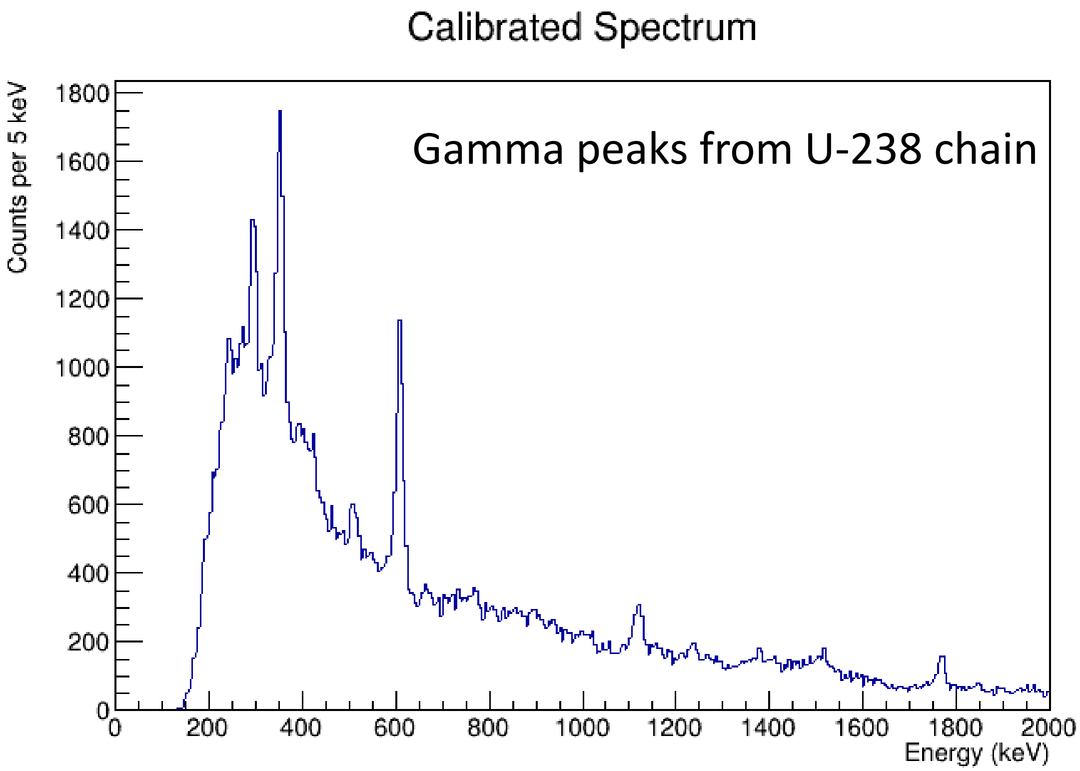
Light versus Heat Chart for LMO



Good separation of alphas!

Calibrated Heat Spectrum for LMO





- Baseline FWHM: 10.4 keV
- Sensitivity: 11 nv/keV

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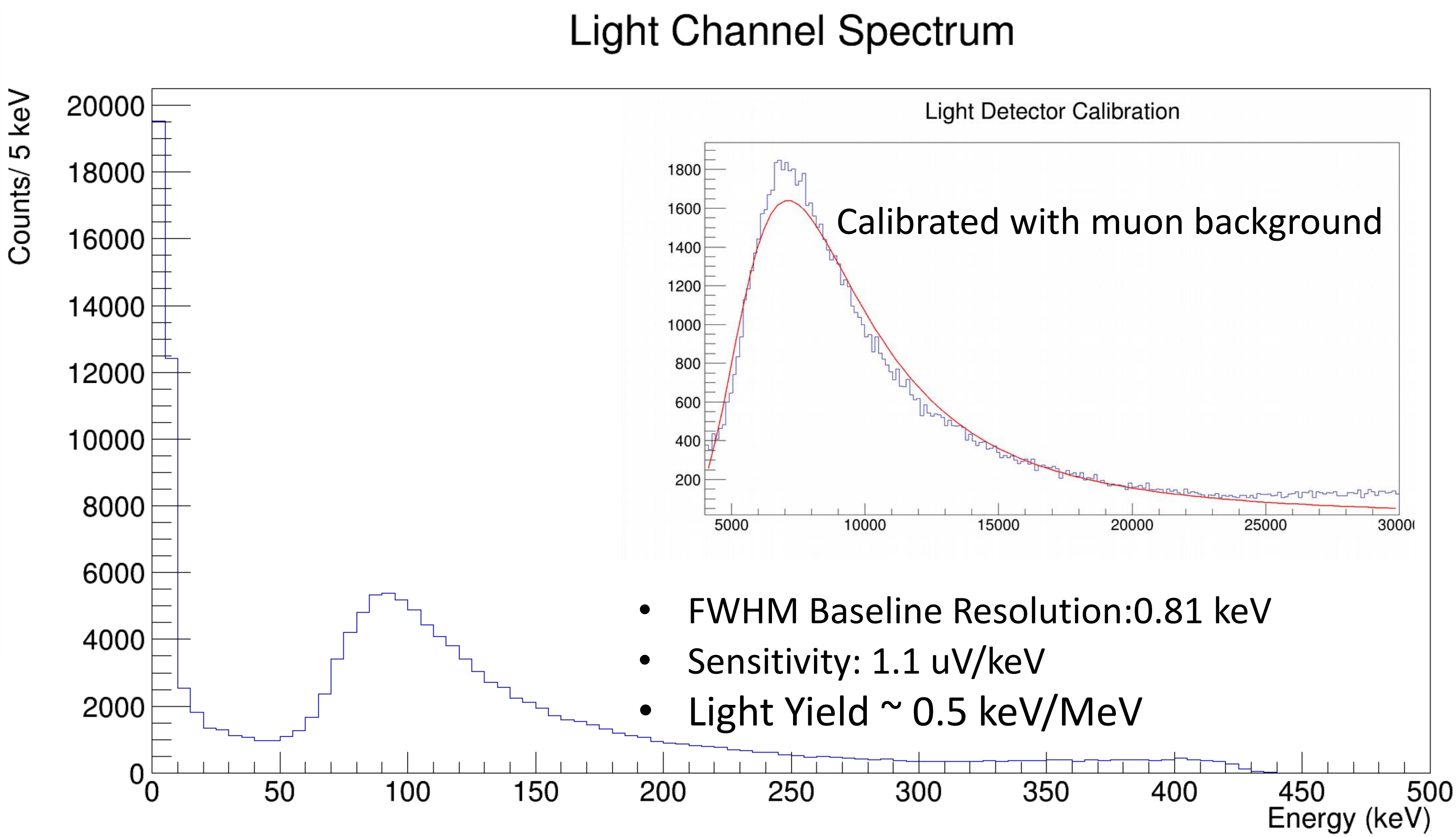
LMO Light Channel Spectrum



- FWHM Baseline Resolution:0.81 keV
- Sensitivity: 1.1 uV/keV
- Light Yield ~ 0.5 keV/MeV

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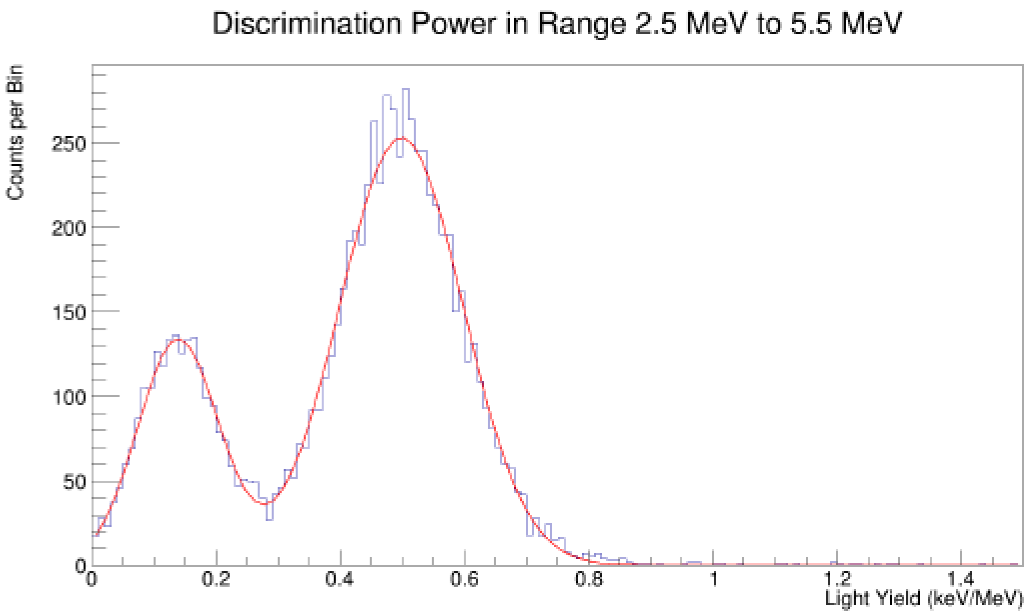
Alpha Particle Discrimination

Alpha Particle Discrimination Power = 3.0



mu_1 = 0.145
sigma_1 = 0.0682
mu_2 = 0.520
sigma_2 = 0.104

DP = |mu_1-mu_2|/sqrt(sig_1^2+sig_2^2)
→
DP = (0.520-0.145)/
sqrt(0.682**2+0.104**2)

DP = 3.0

LMO Internal Alpha Background Limits

Alpha Contamination: Limits are 0.08 to 0.3 mBq/kg

– Comparable to the CLYMENE crystal

Alpha Contamination Limits

Chain/ Contamination	Nuclide	Q-Value (keV)	Counts	Limit on Activity (mBq/kg)	CLYMENE LMO-Small (mBq/kg)
Th-232	Th-232	4081.6 ± 1.4	5	<0.24	<0.5
	Th-228	5520.08 ± 0.22	8	<0.10	<0.55
U-238	U-238	4269.7 ± 2.9	9	<0.12	<0.72
	Ra-226	4870.62 ± 0.25	-	<0.21	<0.50
	Rn-222	5590.4	13	<0.21	-
	Po-218	6002.4	7	<0.08	-
	Po-210	5407.45 ± 0.07	8	<0.10	<1.7
Pt-190	Pt-190	3252 ± 6	15	<0.25	-

- Feldman-Cousins tables are used to set 90% limits
- Count limits are converted to activity limits with the exposure of **0.22 kg*days**
- Ra-226 limit is set by assuming secular equilibrium with Rn-222
- Comparison is to CLYMENE (Exposure 0.039 kg*days)
- Accounting for different exposures, the two sets of limits are comparable (arXiv:1801.07909 [physics.ins-det])

Manufacturing Plan

Supply of ^{100}Mo

- The molybdenum, supplied as $^{100}MoO_3$ powder, will be purchased from ISOFlex in a quantity and purity sufficient for the prototype objective of the Phase IIA project
- The enriched $^{100}MoO_3$ is by the far the most expensive component of the LMO detectors planned for CUPID, at > $50,000 per kg for the ^{100}Mo

Production Schedule

- 600 crystal cubes 4.5 cm on a side by 2025. We plan to begin delivering crystals to CUPID in 2022
- One Czochralski crystal puller can produce up to 100 crystals per year
- Three pullers will be needed to complete the delivery on time

Summary and Plans for the Remainder of Phase IIA

We will continue follow the original plan described in the proposal.

- Provide unenriched 45 mm cube crystals to MIT/CUPID for cryogenic evaluation.
- Incorporate enriched ^{100}Mo and produce a prototype detector crystal fully suitable for CUPID.
- Finalize and document the production process.
- Prepare to produce full size crystals and transition to Phase III production to supply ~ 600 of the crystals to CUPID 2023-2025.
    - Purchase additional CZ furnaces
    - Set up manufacturing scale purification, synthesis, and fabrication facilities