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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 - 5/27/2021

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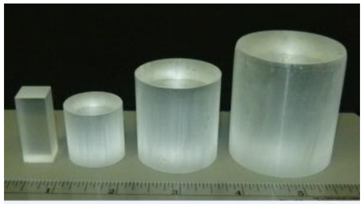
RMD Team: Josh Tower, Huicong Hong
MIT: Lindley Winslow, Joe Johnston

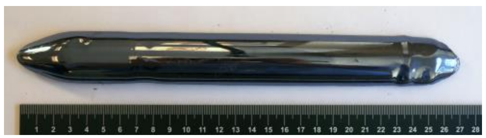
August 14, 2020

RMD Basic and Applied Research and Development

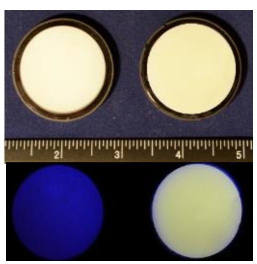
Materials Science



Scintillators



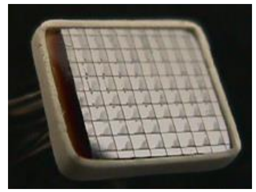
Semiconductors



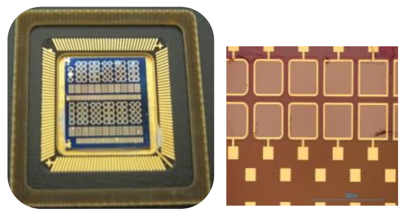
X-ray Imaging Screens

Ceramic Lasers and IR windows

Sensors



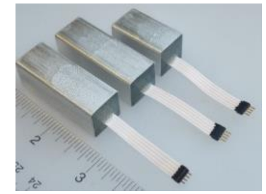
APDs SSPMs Photosensors



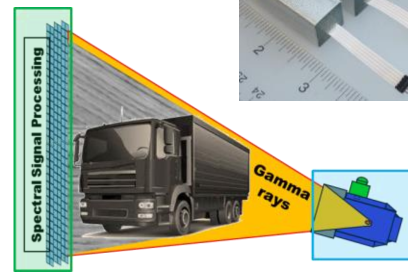
Wide Band Gap Geiger Photodiodes

Surgical Beta-Probe

Instruments & Systems



RadEye Detectors



HiRIS – High Resolution Imaging System

Hermes G/n w/ isotope ID

Robotic nuclear power plant concrete analyzer

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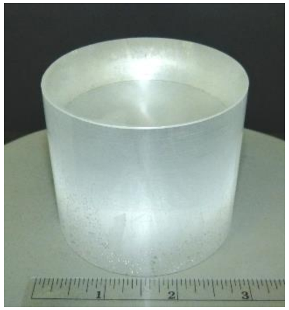
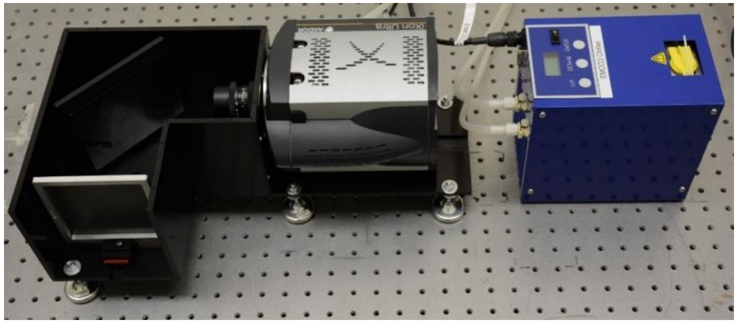
RMD Commercial Products



3" CLYC Crystals CLYC Pillars

Scintillation detectors

Thermo-Scientific



INL Neutron Imaging System

Zetec ECT power plant probe

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, and thus a Majorana particle?
- The question of the Majorana nature of neutrinos is one of the most important questions in physics today
- If the neutrino is a Majorana particle, the neutrino is responsible for the matter-antimatter asymmetry we observe in the universe.
- Searching for neutrinoless double beta decay (0νββ) one of the highest priority experiments to answer this question
- One such experiment is CUORE: **C**ryogenic **U**nderground **O**bservatory for **R**are **E**vents. CUORE uses 1 ton TeO_2 bolometers
- The next generation experiment will be CUPID: **CU**ORE with **P**article **Id**entification. CUPID will use scintillating bolometers

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 Li_2MoO_4 from the high purity raw materials
- Purify the Li_2MoO_4 further by zone refining to improve the radioactive background
- Grow single-crystal ingots using Czochralski for fabricating 200 – 250 gm detectors
- Develop processes for shaping and polishing crystals that maintain radio-purity
- Deliver detector crystals to MIT for cryogenic evaluation. Scintillating bolometer testing will include all operational characteristics, such as light output and radioactivity background.
- Demonstrate suitability for the CUPID neutrinoless double-beta decay experiment
- Grow LMO using isotope enriched ^{100}Mo and produce full-spec detectors to qualify as a supplier for the CUPID experiment.

Selection of Isotopes with Double-beta decay

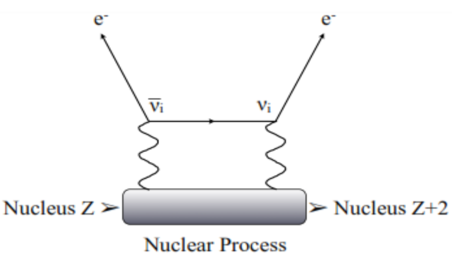
Candidate Isotopes for 0νββ Experiments

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

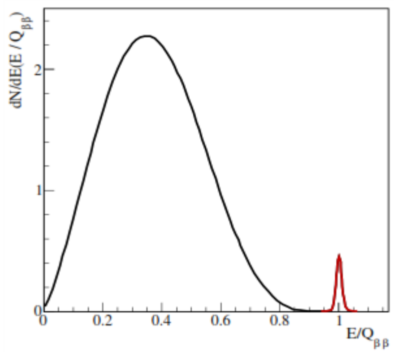
<u>Requirements for isotope</u>

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.



0νββ process

0νββ spectrum



Scintillating Bolometers *are needed for better particle discrimination and background reduction in next generation experiments.*

Li_2MoO_4 (LMO) Synthesis

1. MoO_3 99.9995% + Li_2CO_3 99.99% high purity powders
   - $MoO_3 + Li_2CO_3 \rightarrow Li_2MoO_4 + CO_2$
2. Mix powders in a plastic mixing bottle overnight on a roller
3. Press the mixture in a Teflon piston jig with a cold press to form a compact and dense mixture puck
4. Place and melt the puck inside a platinum crucible at 650C
5. Repeat steps 1-4 until crucible is sufficiently full

Cold Press

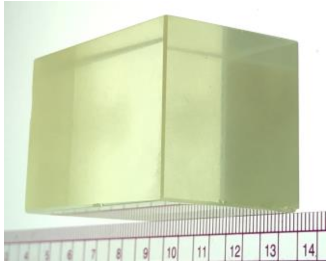
Puck generated

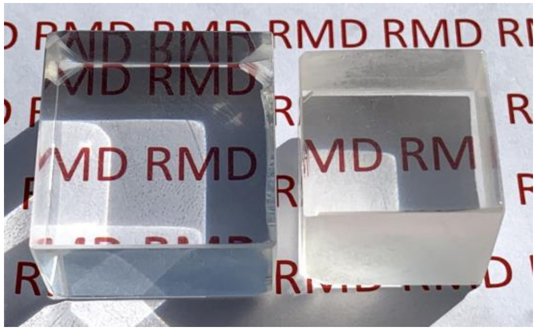
Puck melted in Pt crucible

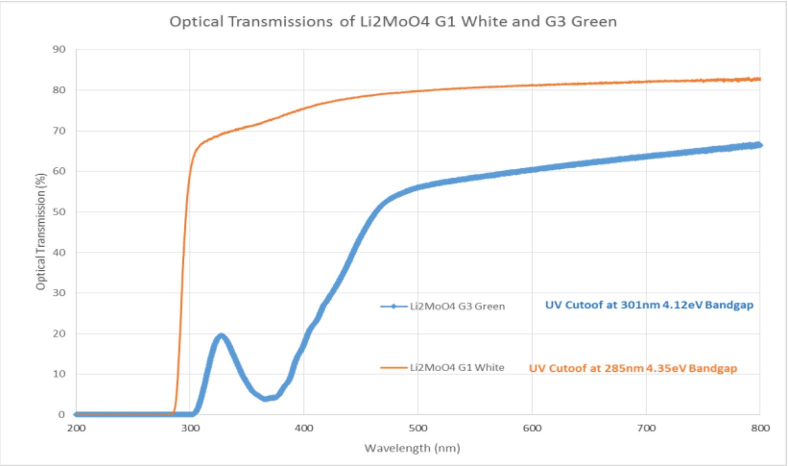
LMO Purification

- Start with good purity raw materials
  - Good sources identified in previous phase
  - Decent purity achieved without further purification
- Evaluating zone refining
  - Trying different crucible materials (Pt, carbonized quartz, etc.)
- MoO_3 (99.9995%) + Li_2CO_3 (99.99%) High Purity Powders

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







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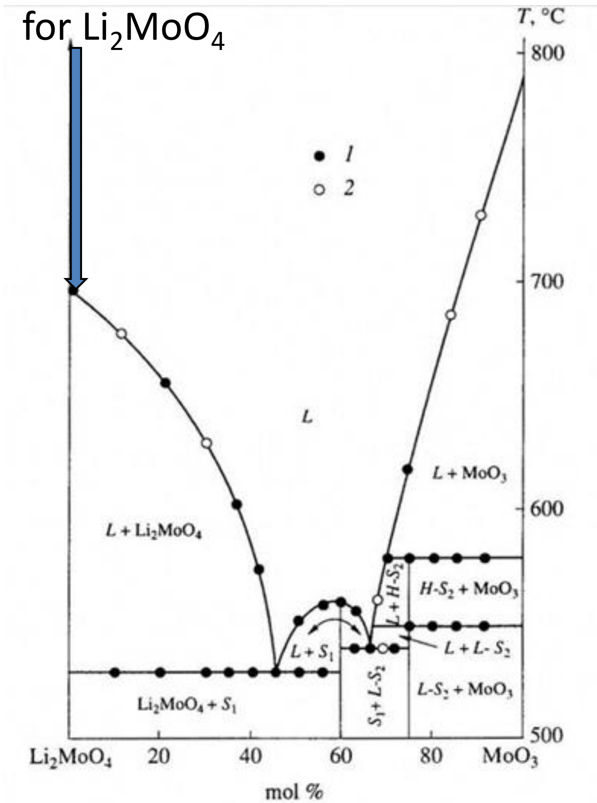
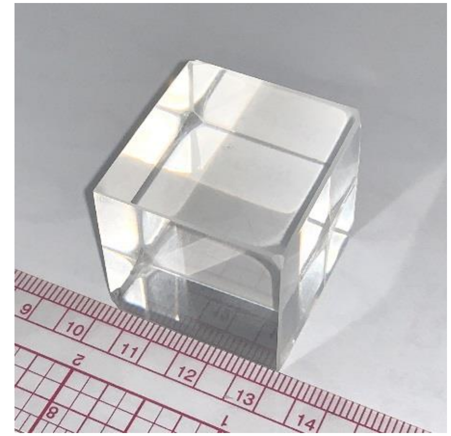
Czochralski Growth of Li_2MoO_4

Congruent growth for Li_2MoO_4

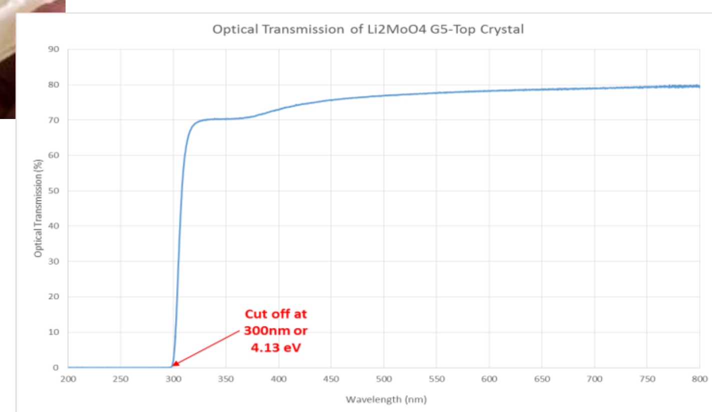


No structural phase change for Li_2MoO_4

Solodovnikov et al., Russ. J. Inorg. Chem., Vol. 44, No. 6 1999



30 x 30 x 20 mm sample used for cryogenic testing



Optical Transmission of Li2MoO4 G5-Top Crystal

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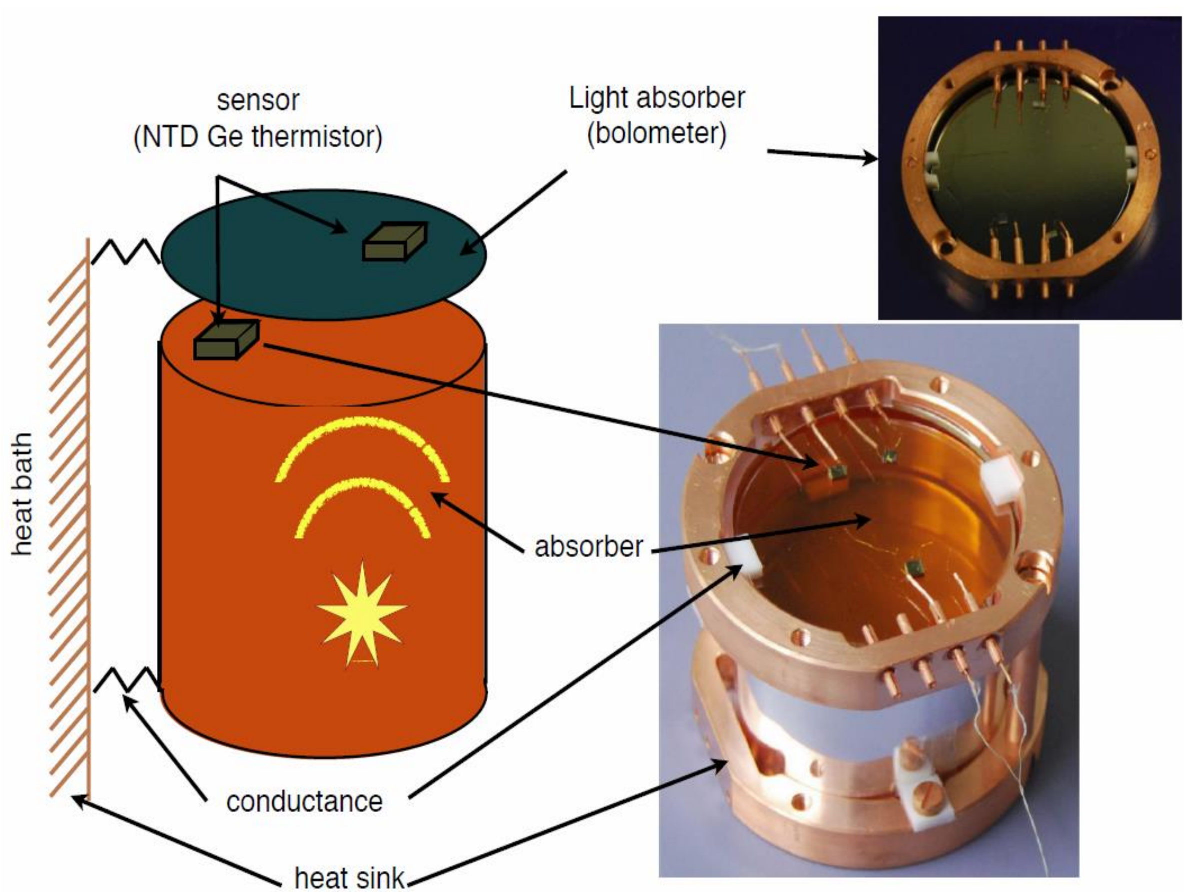
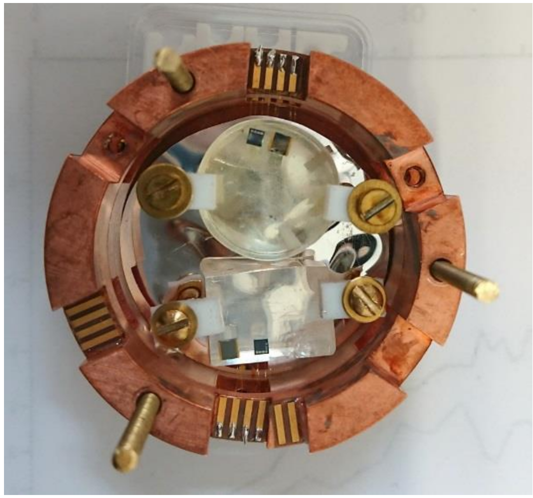
Cryogenic Testing of Scintillating Bolometers

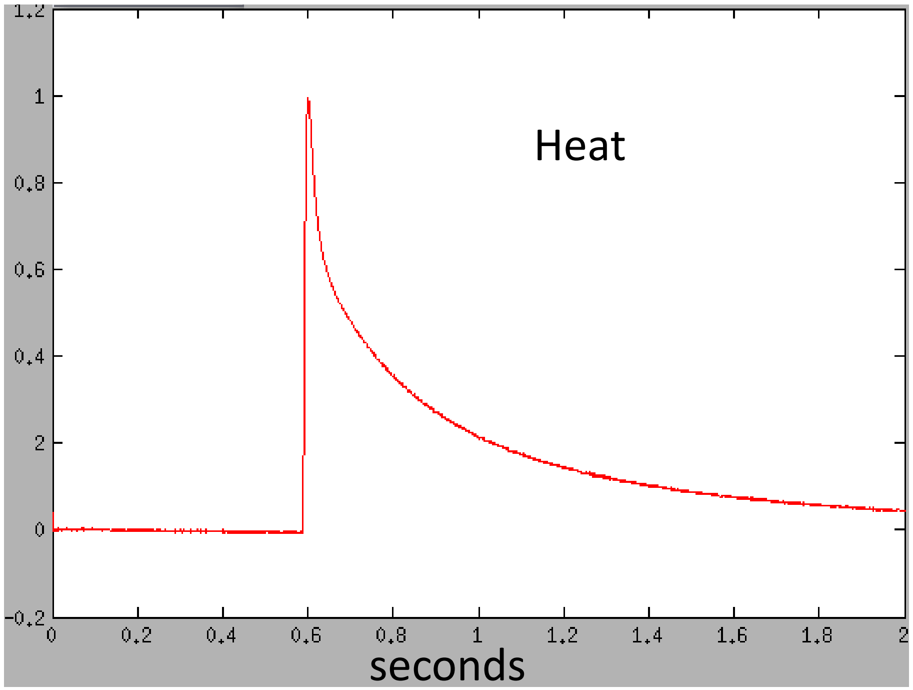
Above ground cryogenic testing by MIT at CSNSM

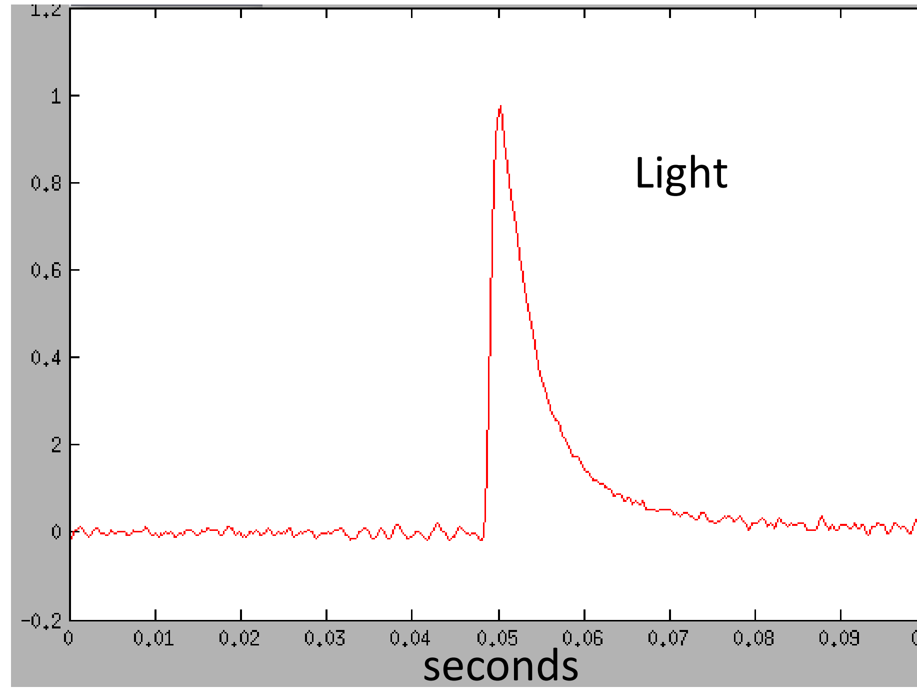


- Samples held at ~ 20 mK for multi-day testing.
- Light and heat pulses measured separately.



Mean light and heat pulses from LMO





Light pulse is ~ 100x faster than heat.

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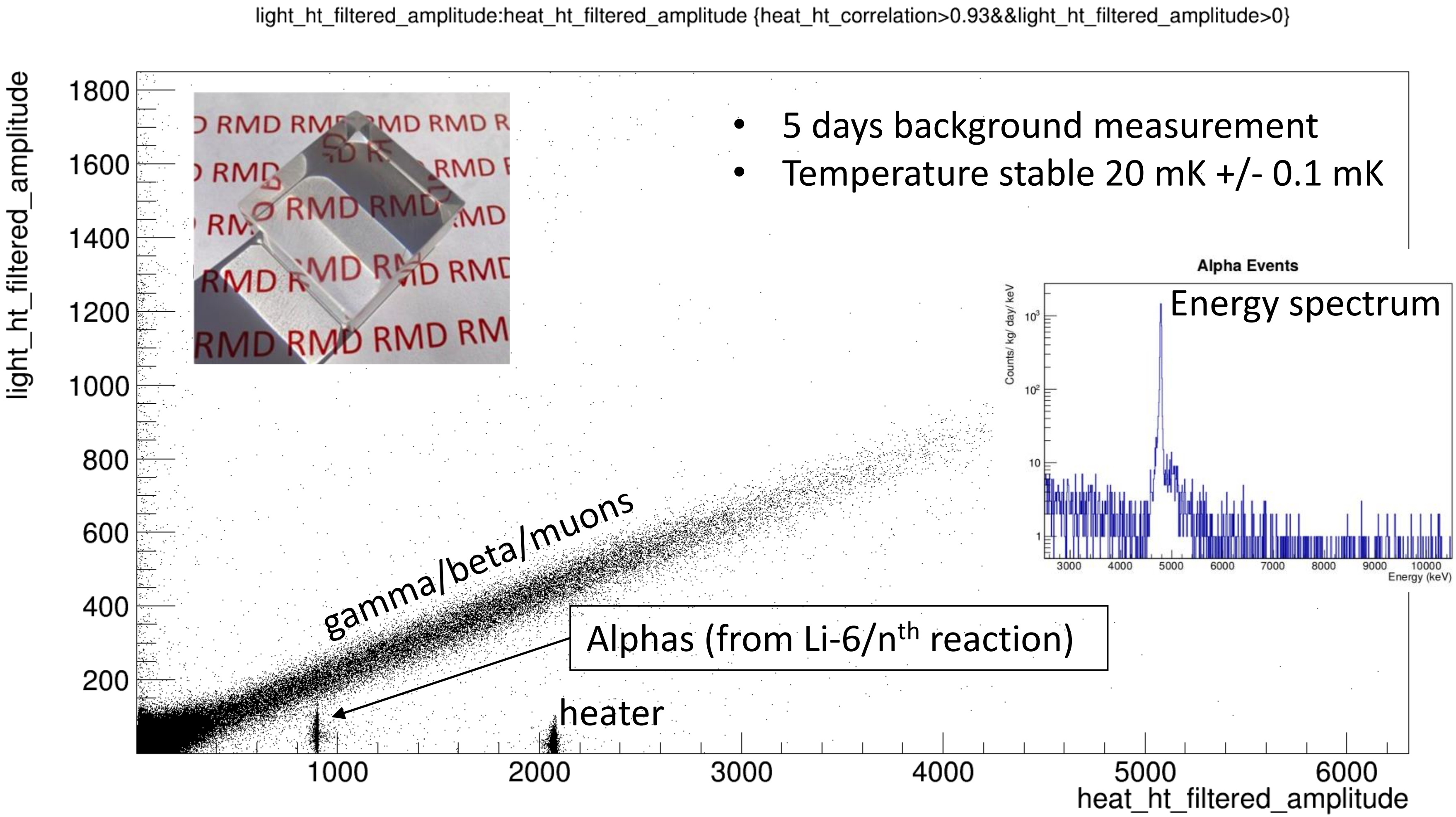
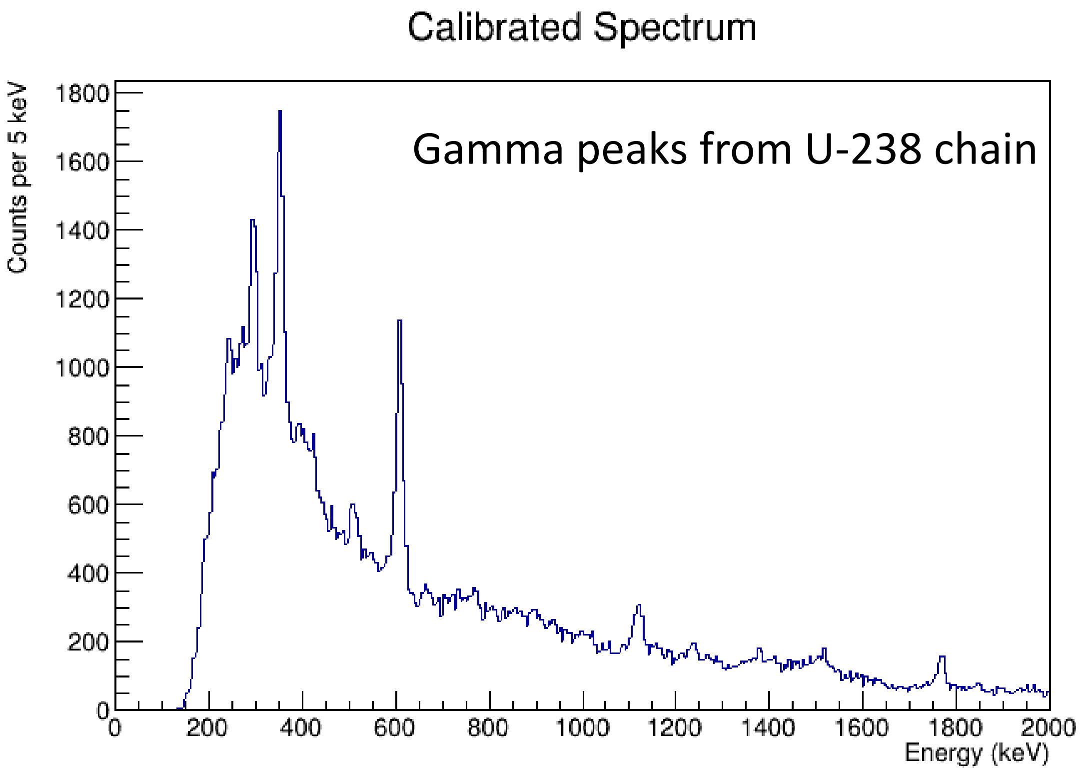
Light versus Heat Chart for LMO

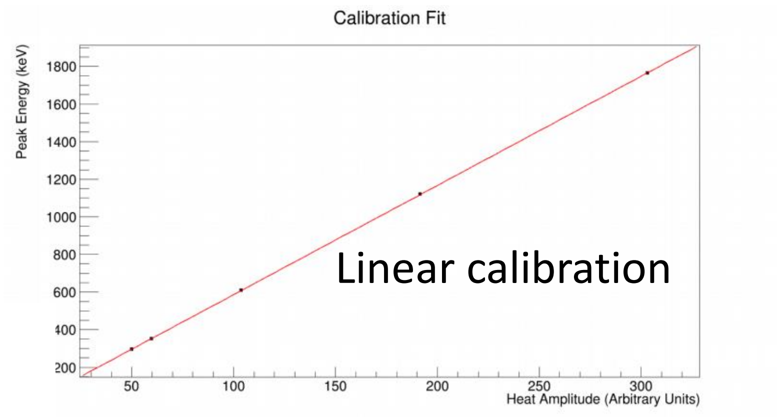
- 5 days background measurement
- Temperature stable 20 mK +/- 0.1 mK



Good separation of alphas!

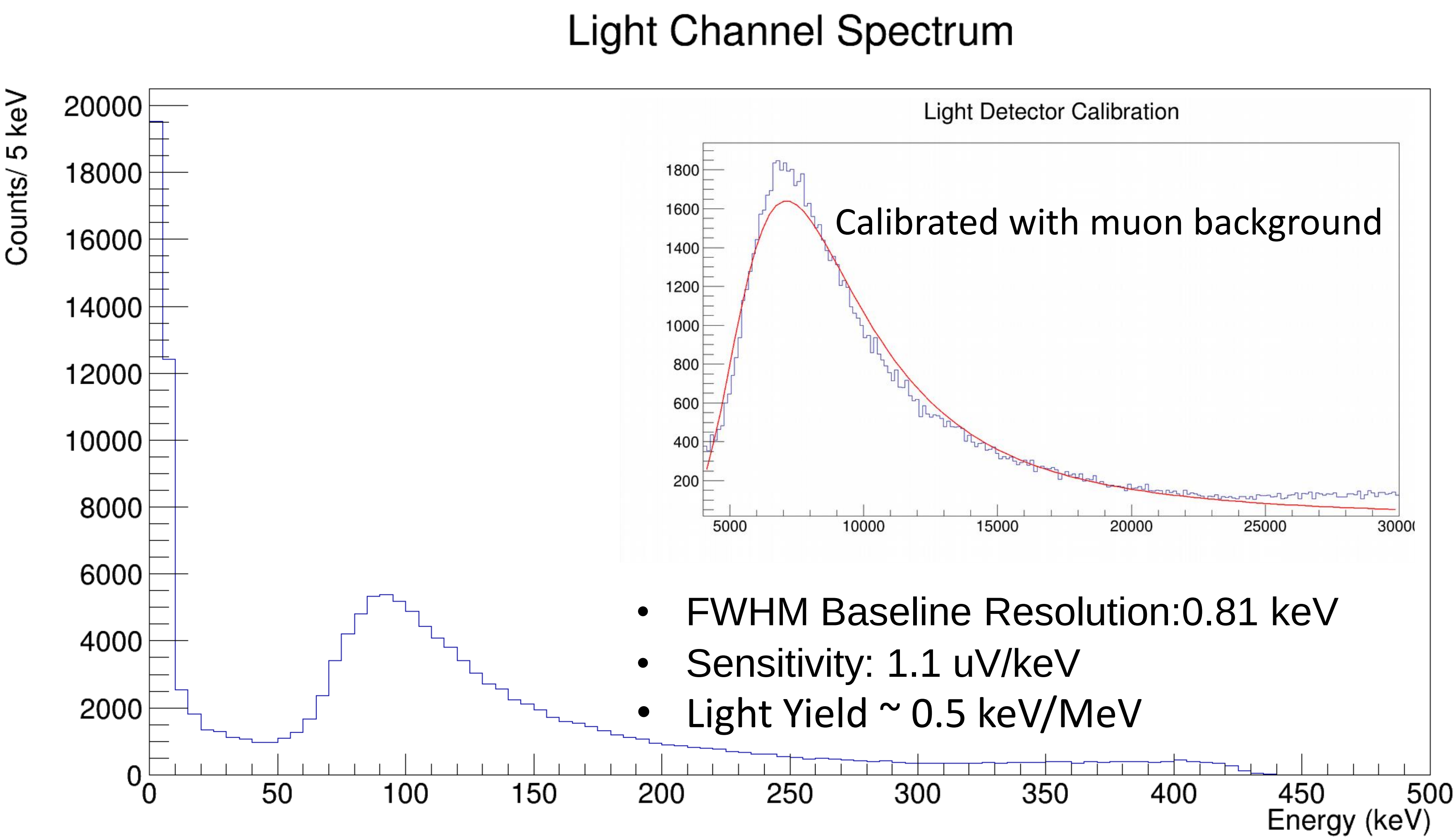
Calibrated Heat Spectrum for LMO





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

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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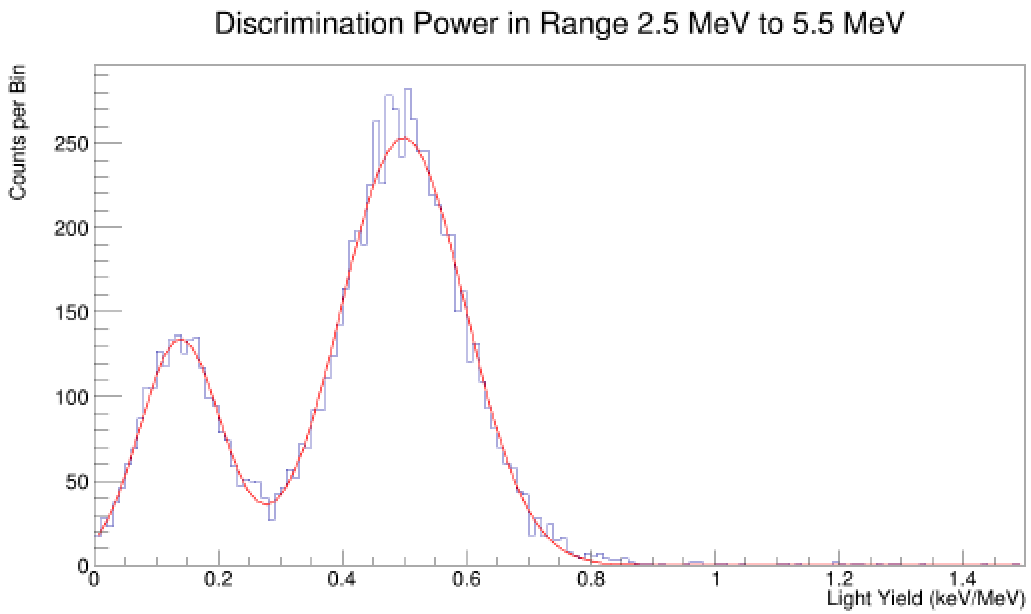
Alpha Particle Discrimination

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 $^{100}MoO3$ powder, will be purchased from ISOFlex in a quantity sufficient for the prototype objective of the Phase IIA project
- The enriched $^{100}MoO3$ is by the far the most expensive component of the LMO detectors planned for CUPID, at approximately $69,000 per kg for the ^{100}Mo
- There is a choice of 1 or 2 stages of centrifugation
- The 2-stage centrifugation cost more but has significantly better purity and will be utilized for this project

Production Schedule

- The required shape size and delivery schedule for LMO crystals to meet the US contribution to CUPID
- 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

Purity of Enriched ^{100}Mo from ISOFlex

1 stage of centrifugation

Element	Permissible Abundance	Element	Permissible Abundance
U-238	<0.01 ppm	Co	<30 ppm
Th-232	<0.01 ppm	Cu	<30 ppm
W	>1000 ppm*	Zn	<30 ppm
Sr	<30 ppm	Zr	<30 ppm
Ba	<30 ppm	Nb	<30 ppm
Si	<50 ppm	Cd	<30 ppm
Al	<30 ppm	Sn	<30 ppm
Sc	<30 ppm	Sb	<30 ppm
Ti	<30 ppm	Hf	<30 ppm
Cr	<30 ppm	Ta	<30 ppm
Mn	<30 ppm	Pb	<30 ppm
Fe	<30 ppm	Bi	<30 ppm
Ni	<30 ppm		

2 stages of centrifugation

Element	Permissible Abundance	Element	Permissible Abundance
U-238	<0.001 ppm**	Ni	<5 ppm
Th-232	<0.001 ppm**	Co	<5 ppm
Ra-226	< 10 mBq/kg *	Cu	<5 ppm
W	<50 ppm	Zn	<5 ppm
Sr	<5 ppm	Zr	<5 ppm
Ba	<5 ppm	Nb	<5 ppm
Si	<15 ppm	Cd	<5 ppm
Al	<10 ppm	Sn	<5 ppm
Sc	<5 ppm	Sb	<5 ppm
Ti	<5 ppm	Hf	<5 ppm
Cr	<5 ppm	Ta	<5 ppm
Mn	<5 ppm	Pb	<5 ppm
Fe	<5 ppm	Bi	<5 ppm

Summary and Plans for the Remainder of Phase IIA

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

- Scale up the crystal growth to 3” diameter by 3” long
- Optimize purification methods, especially for MoO_3
- Continue to provide crystal samples to MIT for cryogenic evaluation.
- Incorporate enriched ^{100}Mo and produce a prototype detector crystal fully suitable for CUPID.
- Finalize and document the production process
- Produce full size crystals and transition to Phase III production