



RMD

A Dynasil Company



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

DOE Contract: DE-SC0015200, SBIR Phase IIA 5/28/2019 - 2/27/2023

RMD Principal Investigator: Michael R. Squillante

DOE Technical Contact: Michelle D. Shinn

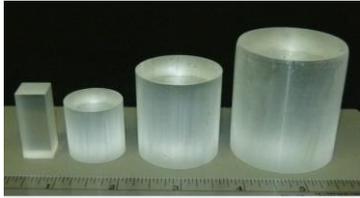
RMD Team: Josh Tower, Huicong Hong

MIT: Lindley Winslow, Joe Johnston

August 25, 2022

RMD Basic and Applied Research and Development

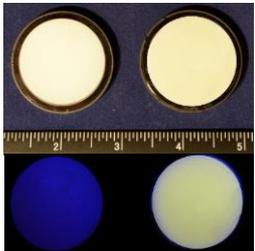
Materials Science



Scintillators



Semiconductors

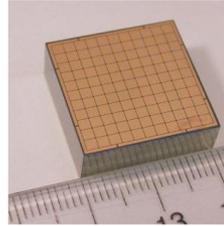


X-ray Imaging Screens

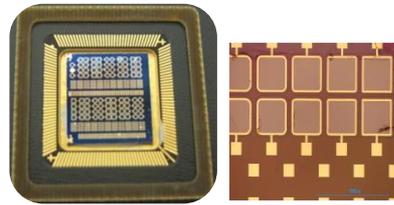


Ceramic Lasers and IR windows

Sensors



TI Br Gamma Imaging Array



Wide Band Gap Geiger Photodiodes



High Speed Digital Neutron Radiography

Systems



RadCam™

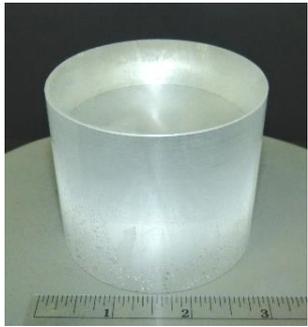


Space Weather monitor

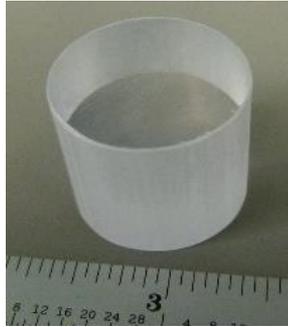


E2MH: Handheld RID

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



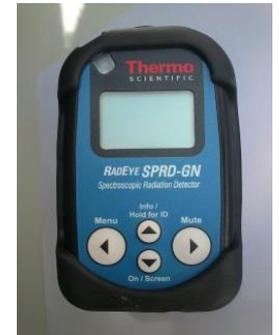
**FLIR R440
RIID**



**ANSTO CORIS360
Imager**



Thermo-Scientific RIIDEye



**Thermo-Scientific
RadEye SPRD-GN**

Low-Background Crystals for Nuclear and High-Energy Physics



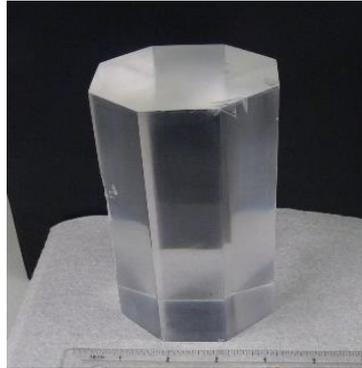
Scintillating Bolometer



for neutrinoless double-beta decay

NaI

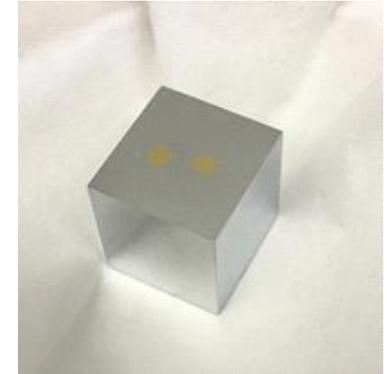
Room T Scintillator



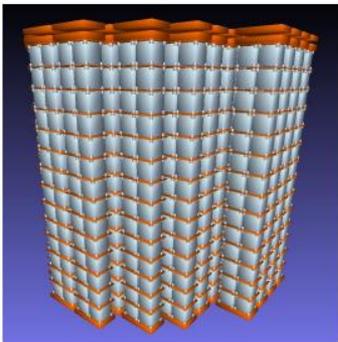
for dark matter search

Zinc

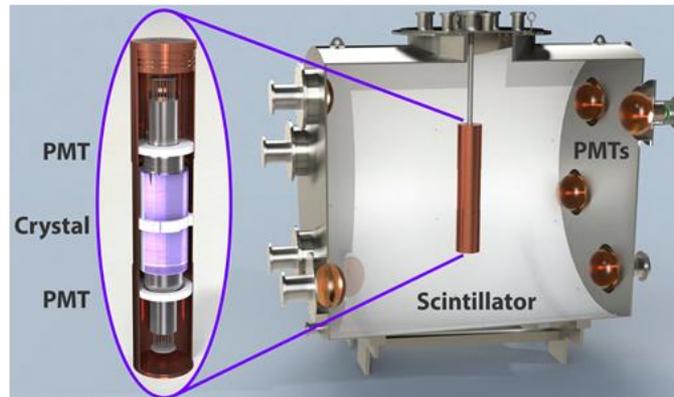
Superconducting Bolometer



for neutrino scattering



CUPID



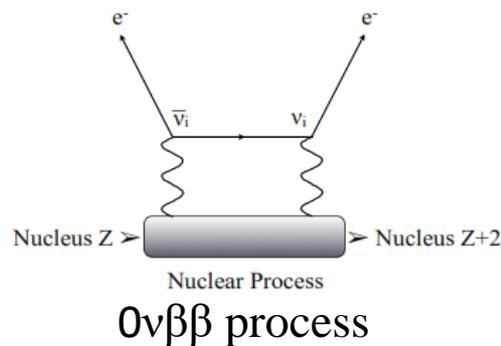
SABRE (now at LNGS)



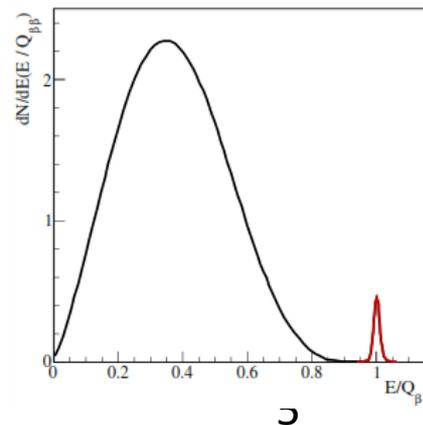
RICOCHET

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** ($0\nu\beta\beta$) is one of the highest priority experiments to answer these questions.
- A next generation experiment to search for $0\nu\beta\beta$ is **CUPID: CUORE Upgrade with Particle Identification**
 - CUPID will use Li_2MoO_4 (LMO) scintillating bolometers.



$0\nu\beta\beta$ spectrum



Selection of Isotopes with Double-beta decay

Candidate Isotopes for $0\nu\beta\beta$ 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 $0\nu\beta\beta$**
- Detects heat and light signals simultaneously.

Scintillating Bolometers are needed for better particle discrimination and background reduction

Phase IIA Technical Objectives

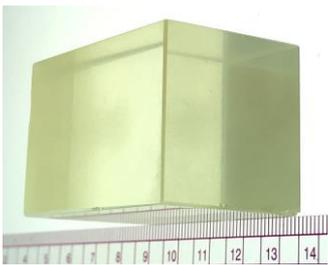
The goal is to complete the research and development needed to implement production of Li_2MoO_4 (LMO) scintillating bolometer crystals suitable for neutrinoless double-beta decay experiments.

- 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.

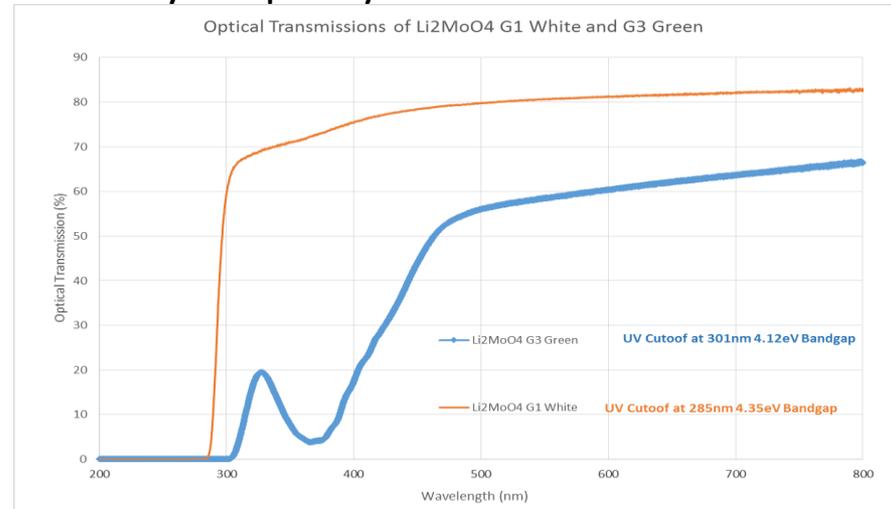
LMO Synthesis and Purification

- 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
 - $\text{MoO}_3 + \text{Li}_2\text{CO}_3 \rightarrow \text{Li}_2\text{MoO}_4 + \text{CO}_2$

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



Optical transmission is good indication of crystal purity



Czochralski Growth of Li_2MoO_4

July 2019



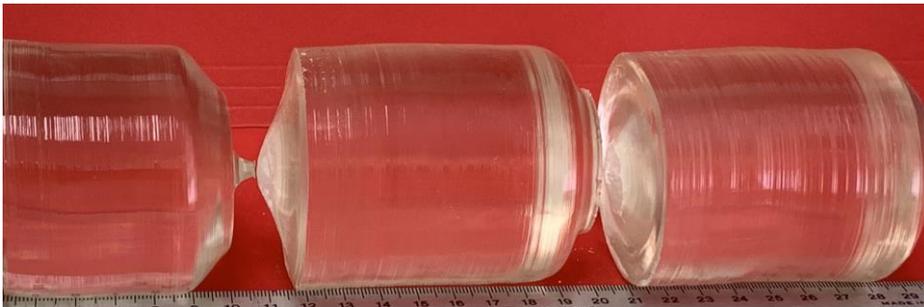
~4 cm OD
~200 grams



June 2021



65mm OD x 65mm
813 grams



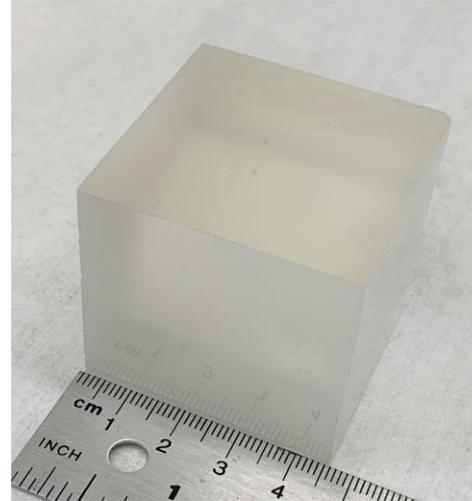
Repeatable growth process



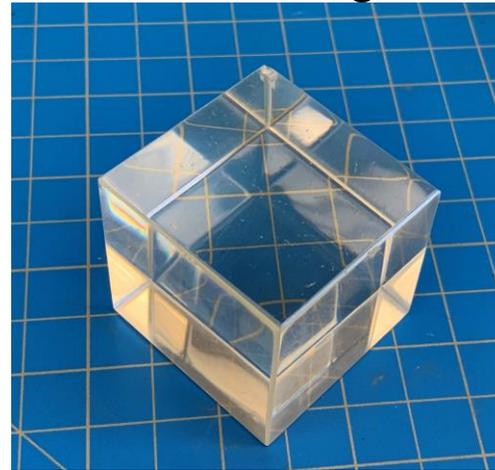
Czochralski Crystal Growth Furnace

Fabrication of Li_2MoO_4 to 45 mm Cubes for CUPID

Before Polishing



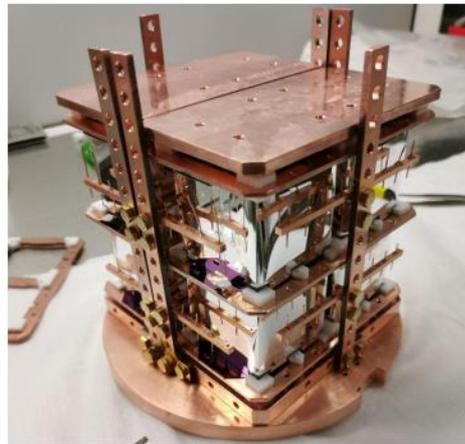
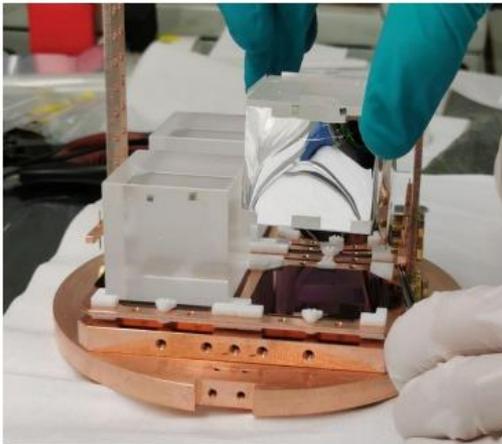
After Polishing



Final polishing/cleaning is done in a clean inert atmosphere to prevent surface contamination.

CUPID: a next generation bolometric neutrinoless double beta decay experiment

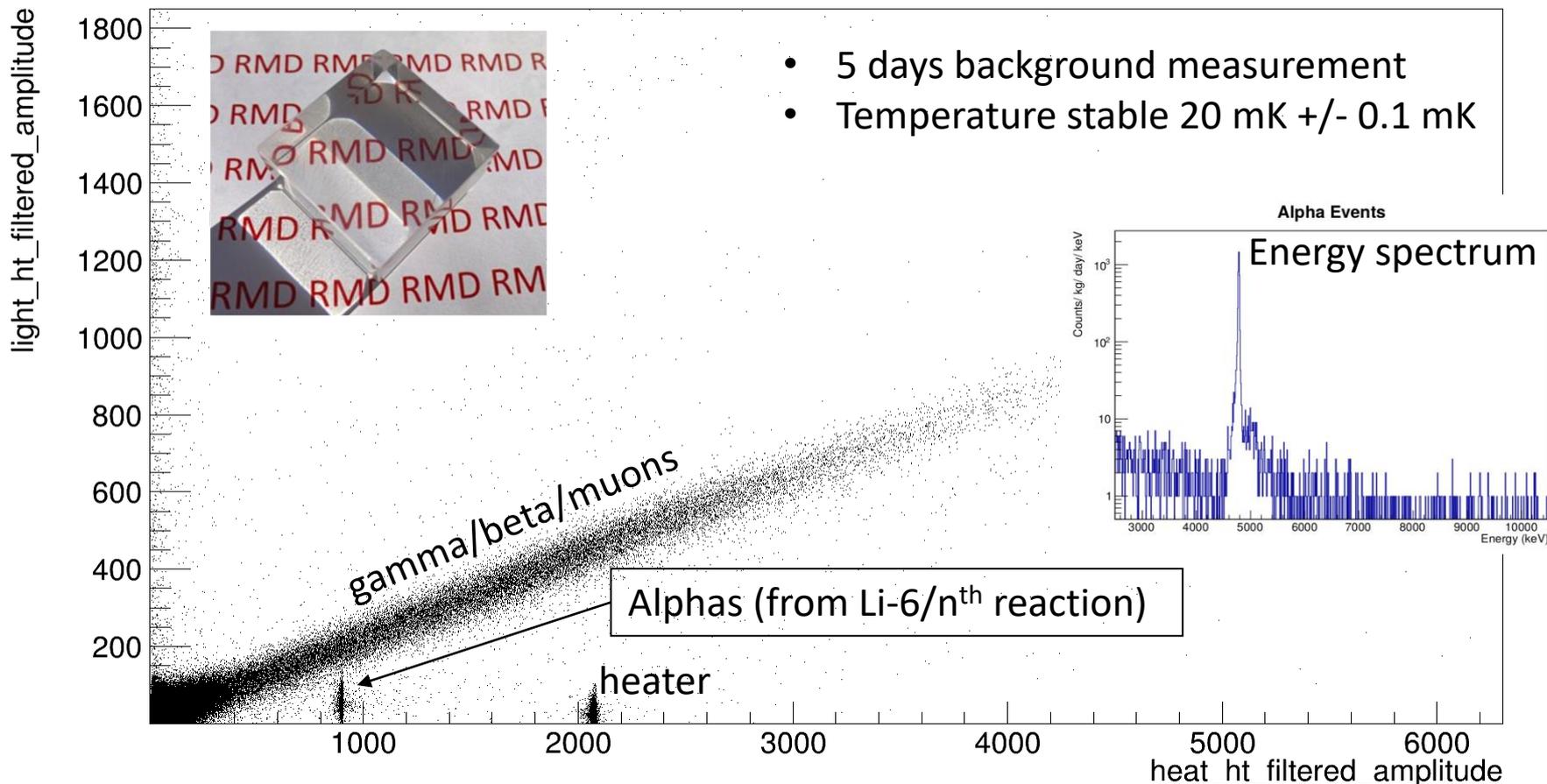
- **CUPID: CUORE Upgrade with Particle Identification**
- CUPID builds on the expertise and lessons of CUORE, CUPID-Mo, and CUPID-0.
- CUPID will consist of about 1500 hybrid heat-light detectors for a total isotope mass of 250 kg (^{100}Mo).
- The required performance in terms of energy resolution, alpha rejection factor, and crystal purity have been demonstrated.



* *GiovanniBenato_TIPP2021(2).pdf*

Light versus Heat Chart for RMD-Grown LMO

light_ht_filtered_amplitude:heat_ht_filtered_amplitude {heat_ht_correlation>0.93&&light_ht_filtered_amplitude>0}

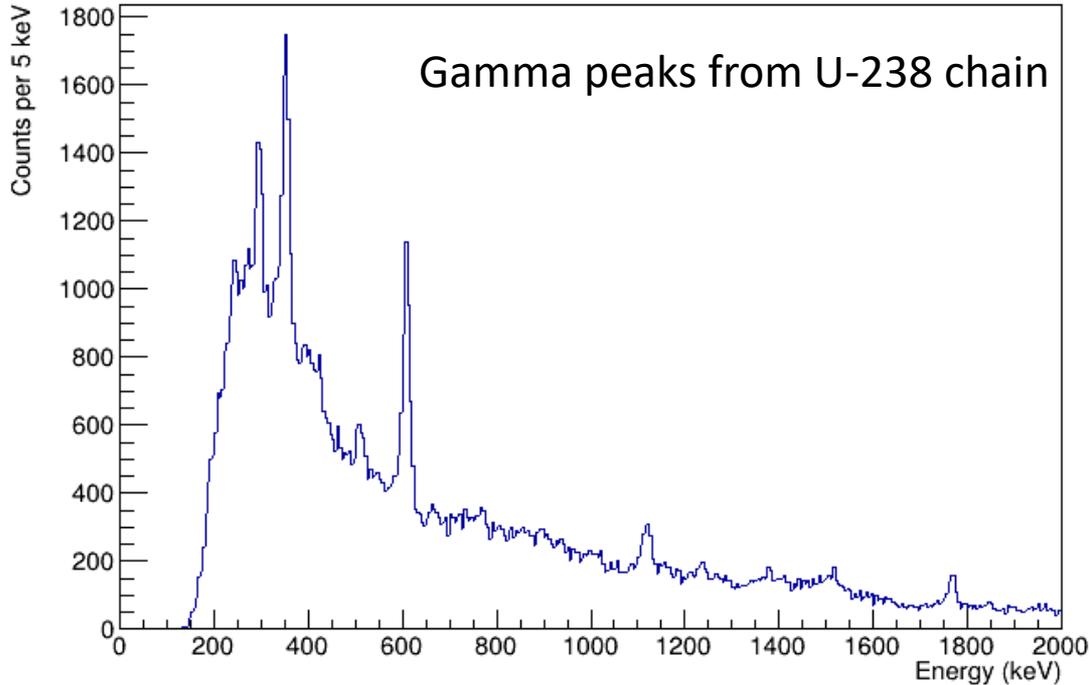


- 5 days background measurement
- Temperature stable 20 mK +/- 0.1 mK

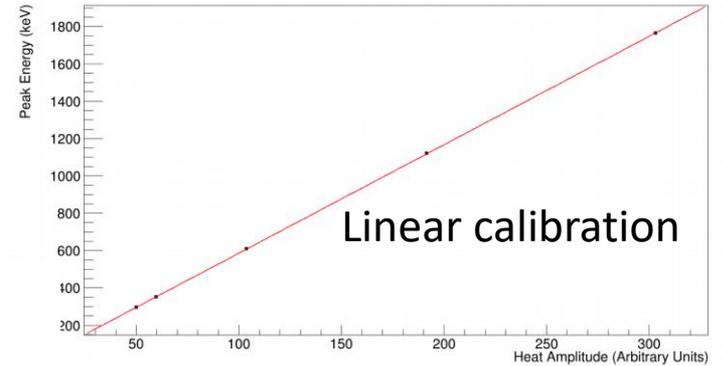
Good separation of alphas!

Calibrated Heat Spectrum for LMO

Calibrated Spectrum



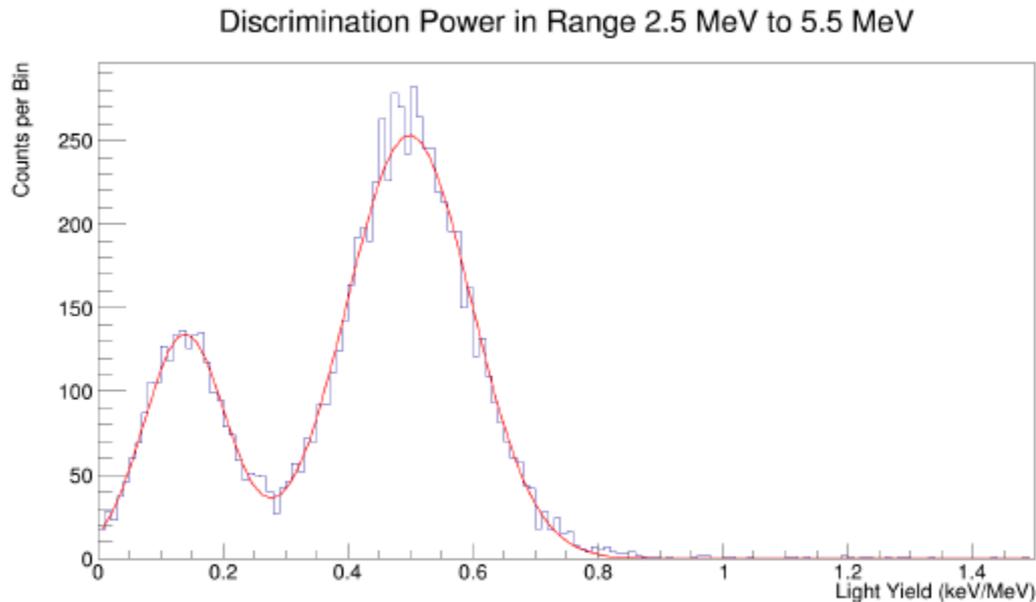
Calibration Fit



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

Alpha Particle Discrimination

Alpha Particle Discrimination Power = 3.0



$$\begin{aligned}\mu_1 &= 0.145 \\ \sigma_1 &= 0.0682 \\ \mu_2 &= 0.520 \\ \sigma_2 &= 0.104\end{aligned}$$

$$\begin{aligned}DP &= |\mu_1 - \mu_2| / \sqrt{\sigma_1^2 + \sigma_2^2} \\ &\rightarrow \\ DP &= (0.520 - 0.145) / \\ &\quad \sqrt{0.0682^2 + 0.104^2}\end{aligned}$$

$$DP = 3.0$$

Summary and Plans for the Remainder of Phase IIA

Phase IIA Summary

- Scale up LMO crystal growth to 3" diameter by 3" long. Growth process optimized for production.
- Post-growth processing optimized to make the 45 mm cubes needed for CUPID.
 - Annealing, cutting, polishing, cleaning
- Samples of 45 mm cubes with natural-Mo produced and provided to MIT for cryogenic evaluation.
 - 2 or 3 more LMO cubes remain to be delivered to MIT for eval.
- Incorporation of enriched ^{100}Mo not done yet due to difficulty in obtaining the required $^{100}\text{MoO}_3$.

Future Work – Supply LMO Crystals for CUPID

LMO Production

- CUPID requires ~ 1600 LMO 45 mm cubes. RMD would increase production capacity to supply 40% of them.
- A crucial aspect of the LMO crystal manufacturing will be recovery of the valuable ^{100}Mo from the crystal scraps (tailings and residual melt).
 - Development of the chemical methods to reconstitute the scrap LMO back into MoO_3 and purify it sufficiently.
 - It might also be necessary to further purify the initial enriched $^{100}\text{MoO}_3$, depending on its condition as-received from the enrichment facility.