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

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Presented by Harish Bhandari

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This work has been supported by the US Department of Energy, Office of Nuclear Physics, SBIR grant No. DE-SC0015200

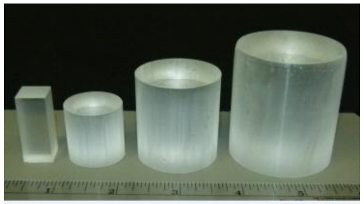
(Currently, we are between Phase II and Phase IIA)

Outline of Talk

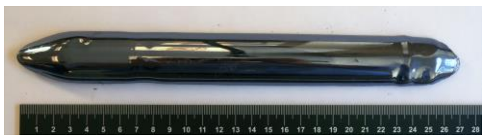
- **Radiation Monitoring Devices, Inc.**
- **Physics Motivation**
- **Crystal Growth (RMD)**
  - $Na_2Mo_2O_7$
  - Li_2MoO_4
- **Cryogenic Testing (MIT)**
  - $Na_2Mo_2O_7$
  - Li_2MoO_4
- **Plans**

RMD Basic and Applied Research and Development

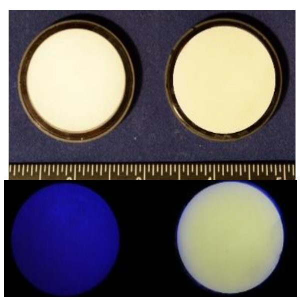
Materials Science



Scintillators

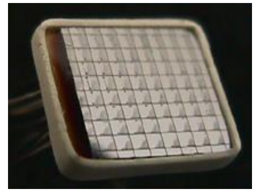


Semiconductors

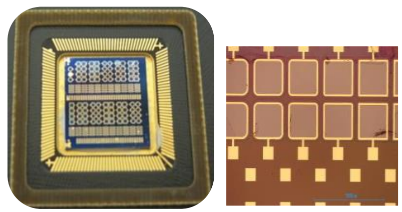


Imaging Screens

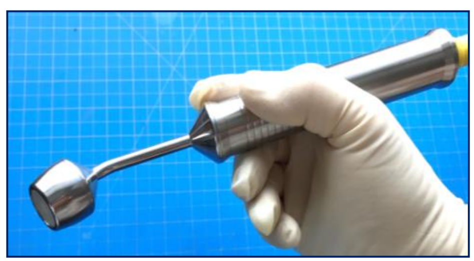
Sensors



APDs SSPMs
Photosensors

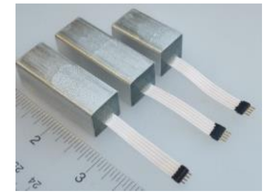


Wide Band Gap
Geiger Photodiodes

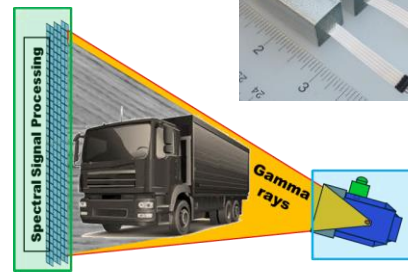


Surgical Beta-Probe

Instruments & Systems

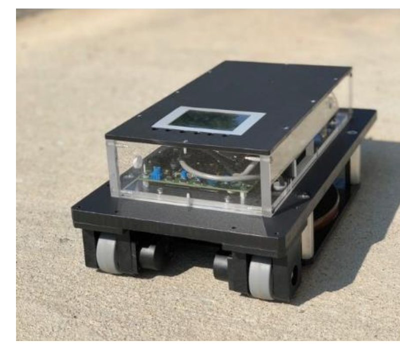


RadEye
Detectors



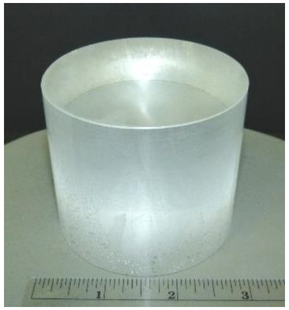
HiRIS – High Resolution
Imaging System

Hermes G/n
w/ isotope ID



Robotic nuclear power
plant concrete analyzer

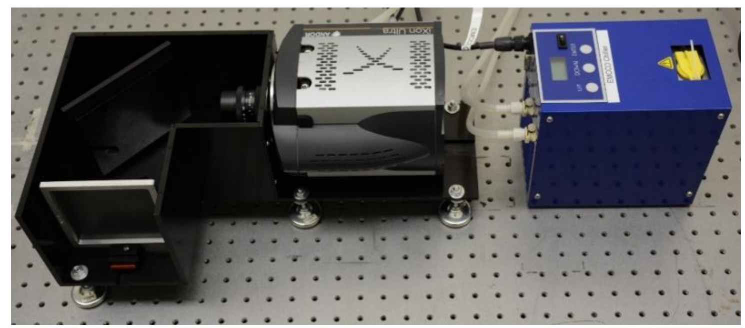
RMD Commercial Products



3” CLYC Crystals CLYC Pillars

Scintillation detectors

Thermo-Scientific

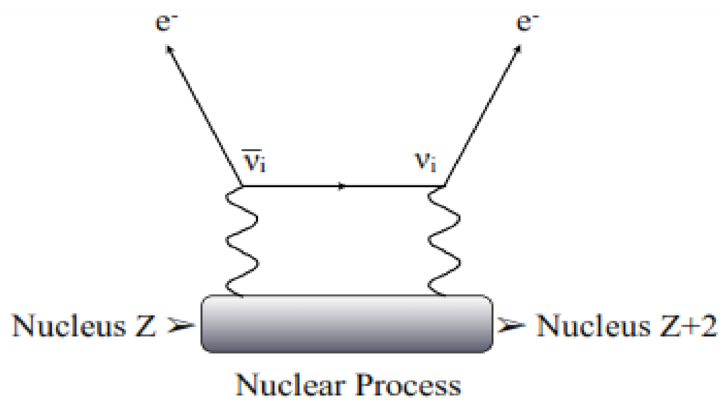


INL Neutron Imaging System

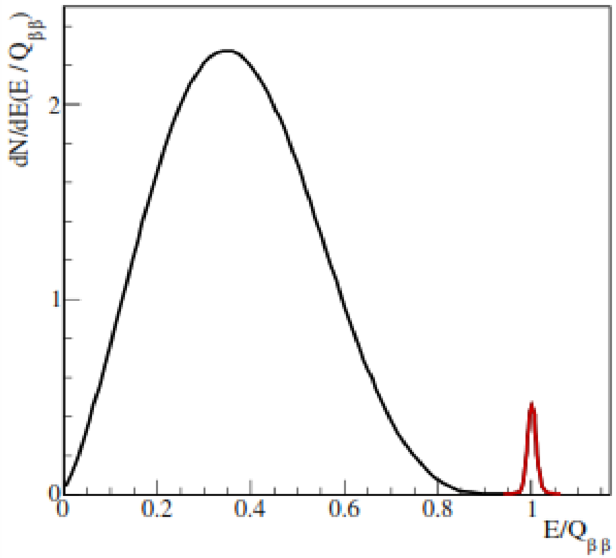
Zetec ECT power plant probe

Hypothesized Process of Neutrinoless Double Beta Decay ($0\nu\beta\beta$)

- Feynman diagram for neutrinoless double-beta decay through light Majorana neutrino exchange



- Spectrum of electron energies from double-beta decay.
- The red section at the endpoint Q indicates those from neutrinoless double-beta decay.



If $0\nu\beta\beta$ exists, then the neutrino must be a Majorana particle (its own antiparticle)!

- This would require changes to the Standard Model of Particle Physics

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.

^{100}Mo has promising properties!

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

$Na_2Mo_2O_7$ (NMO) and Li_2MoO_4 (LMO) Synthesis

1. MoO_3 99.9995% + (Na_2CO_3 99.997% or Li_2CO_3 99.99%) High Purity Powders
   - $2\ MoO_3 + Na_2CO_3 \rightarrow Na_2Mo_2O_7 + CO_2$
   - $MoO_3 + Li_2CO_3 \rightarrow Li_2MoO_4 + CO_2$
2. Mix powders in a plastic 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

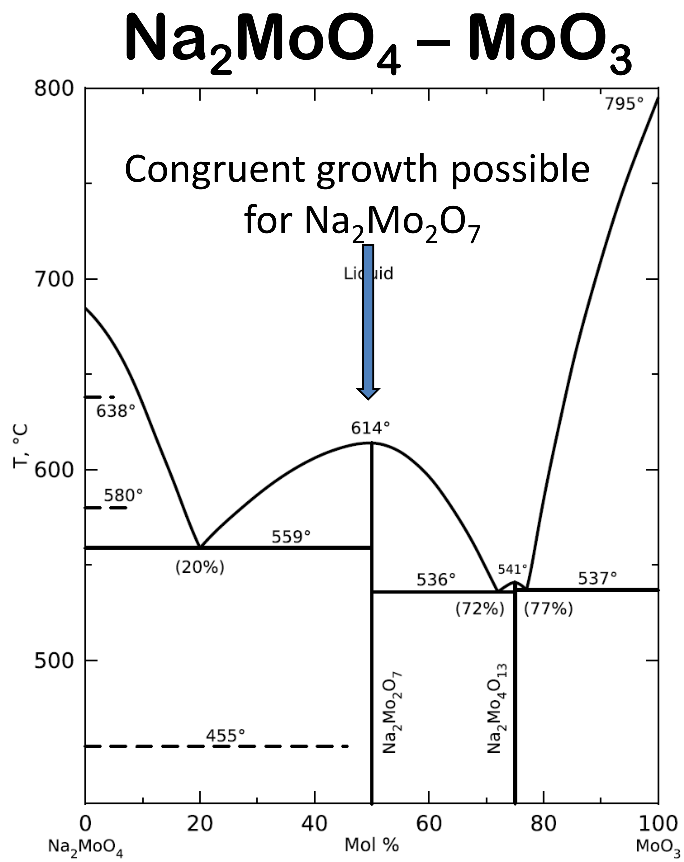
Cold Press

Puck generated

Puck melted in Pt crucible

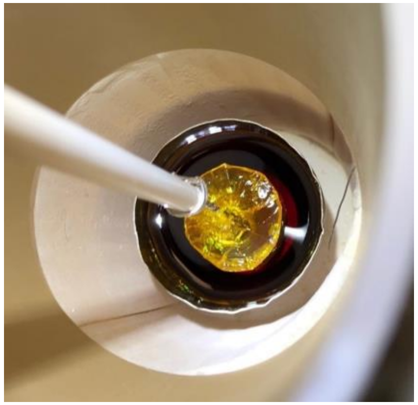
$Na_2Mo_2O_7$ Growth

Czochralski growth method used

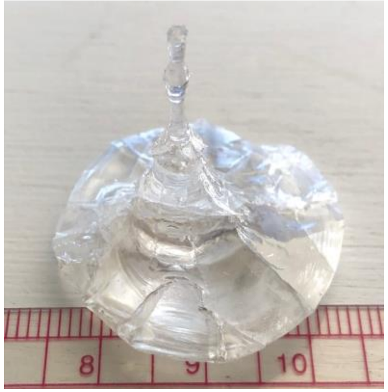


Structural phase change for Na_2MoO_4

Petrosyan *et al.*, *Russ. J. Inorg. Chem. (Engl. Transl.)*, **22** [10] 1542-1544 (1977).



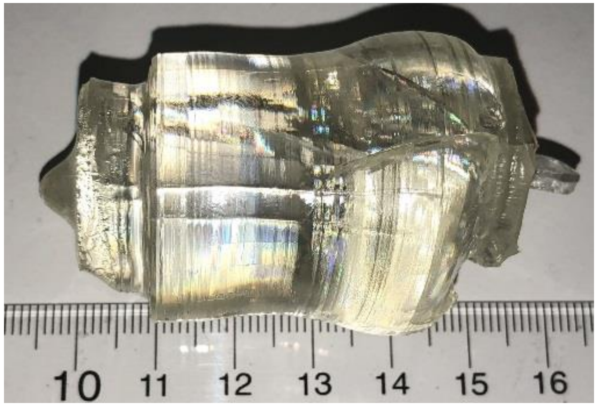
First growth run made using Pt wire as a seed.



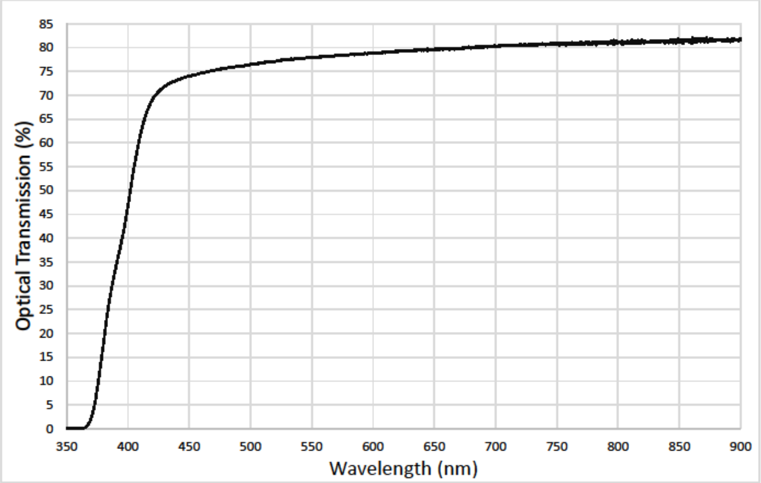
Initial crystal was cut into seeds for subsequent growth runs.

Seeded Cz Growth of $Na_2Mo_2O_7$

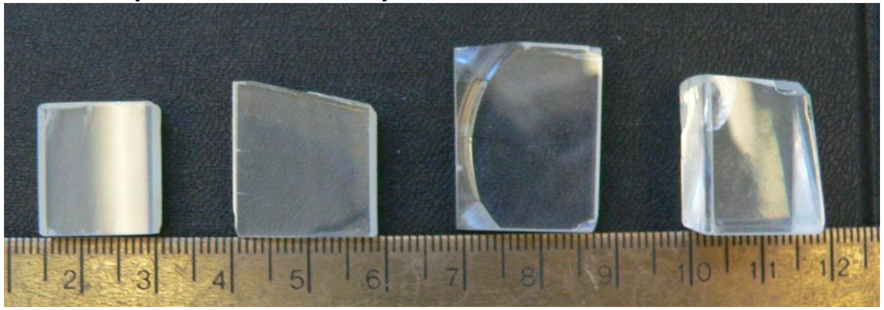
As-grown Ingot



Good Optical Transmission



Samples cut and polished for evaluation

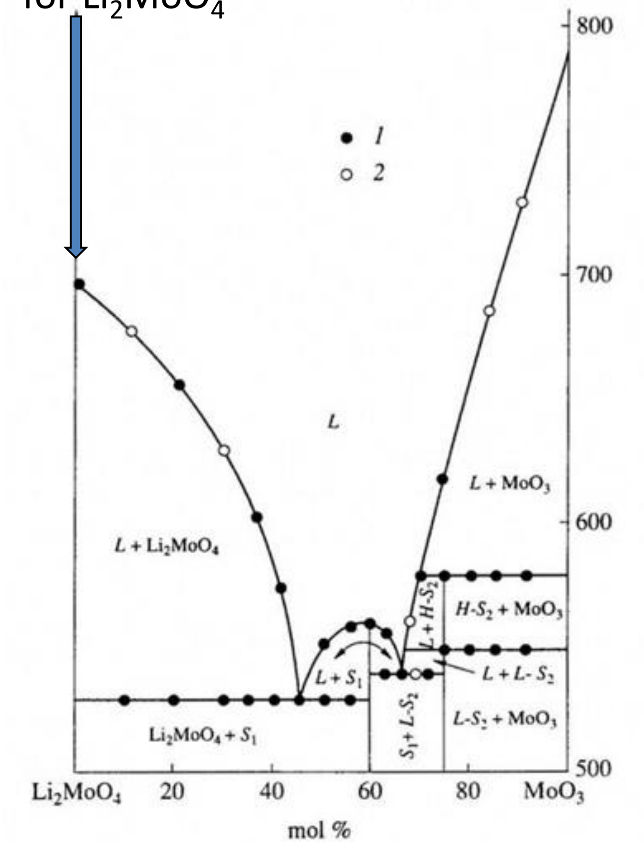


- Colorless transparent crystals are needed for best scintillation light yield.
- High purity and good stoichiometry are crucial for colorless crystals.

- ***Good quality crystals can be grown, but cracking is common.***

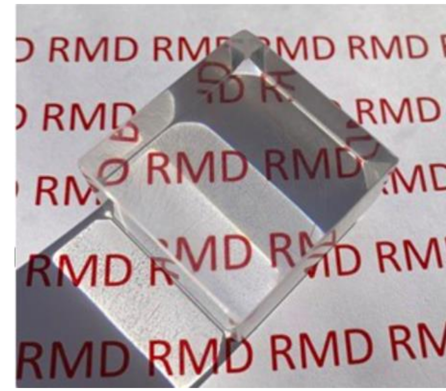
Cz Growth of Li_2MoO_4

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

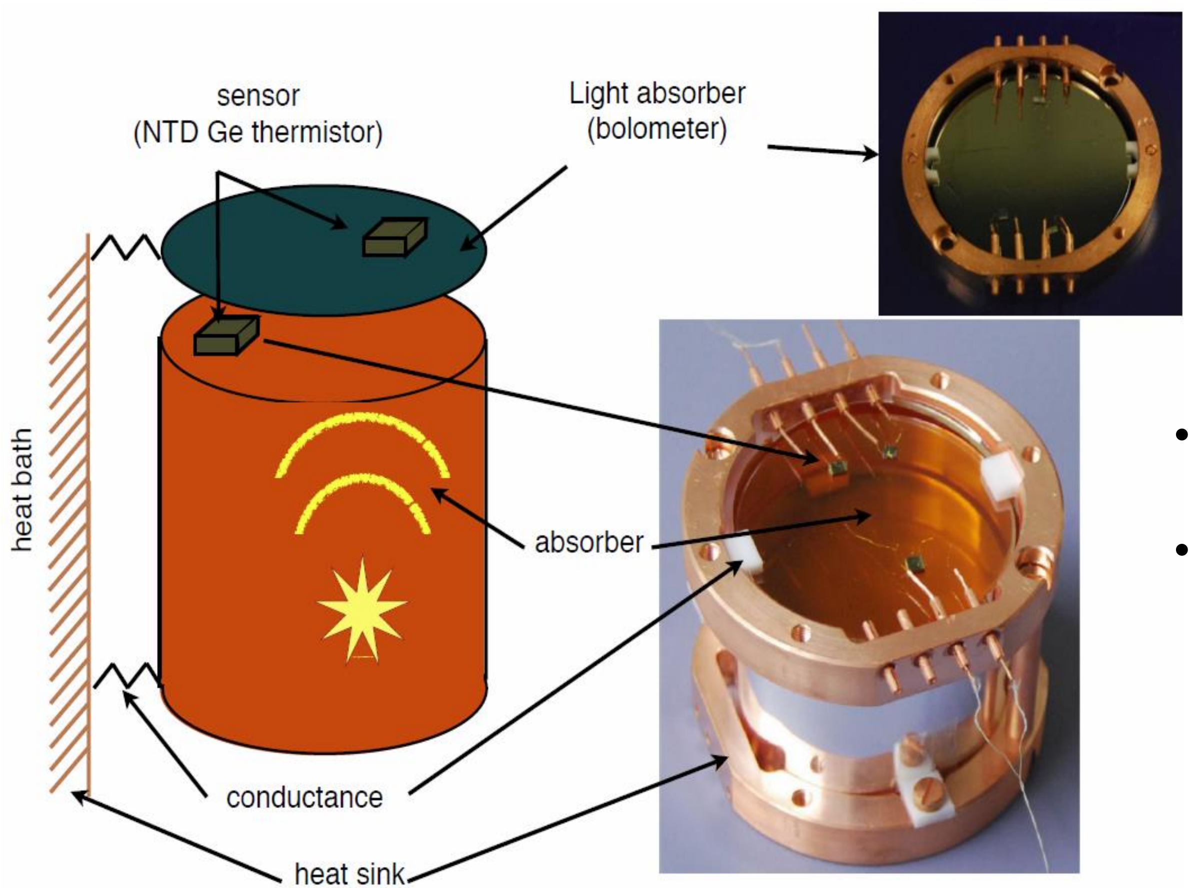


30 x 30 x 20 mm sample used for cryogenic testing

LMO is less prone to cracking more conducive to manufacturing, as compared to NMO.

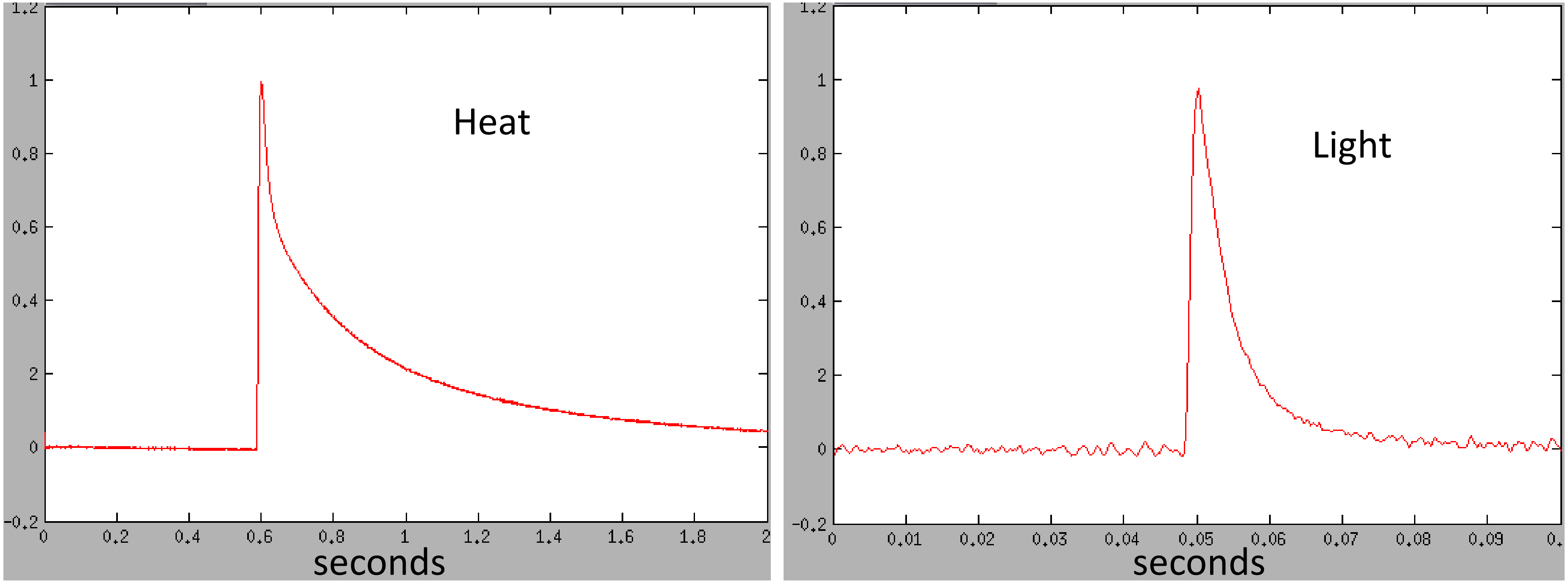
Cryogenic Testing of Scintillating Bolometers

Above ground cryogenic testing at CSNSM in Orsay, France



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

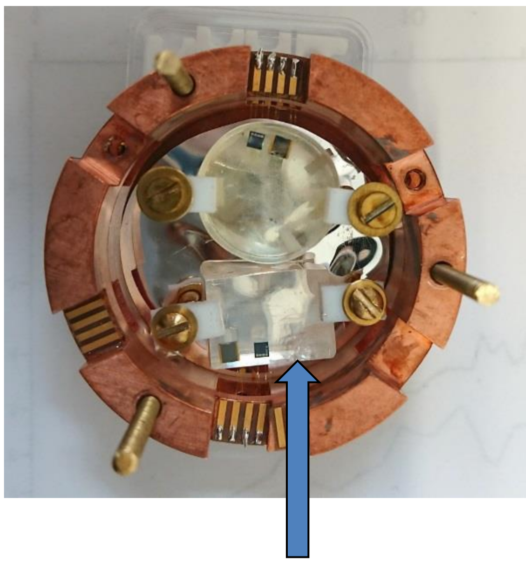
Mean light and heat pulses from LMO



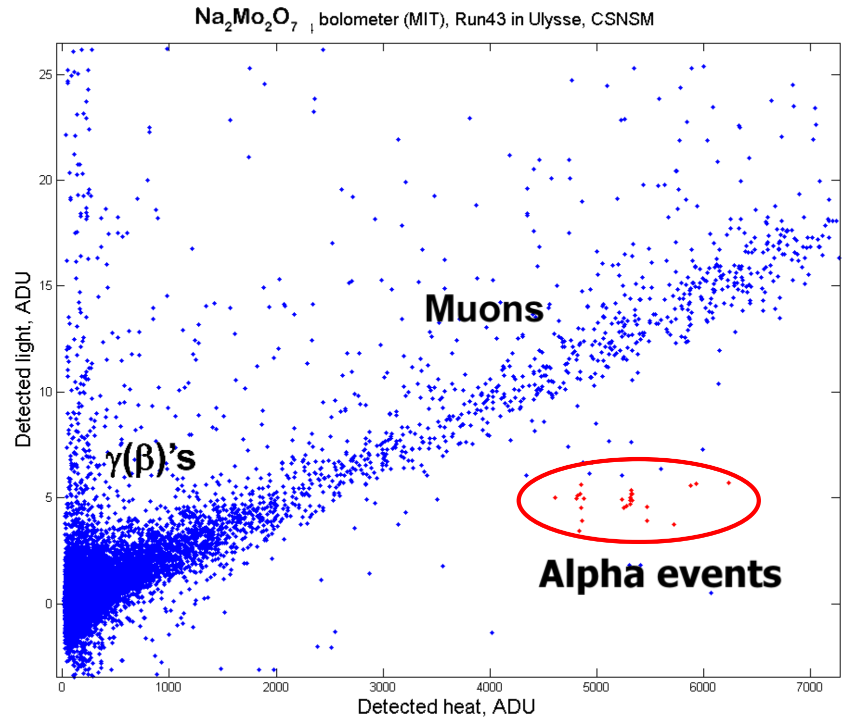
Light pulse is ~ 100x faster than heat.

Good Particle Discrimination in Light-vs-Heat Plot

$Na_2Mo_2O_7$ mounted in cryogenic sample holder with Ge NTD devices.

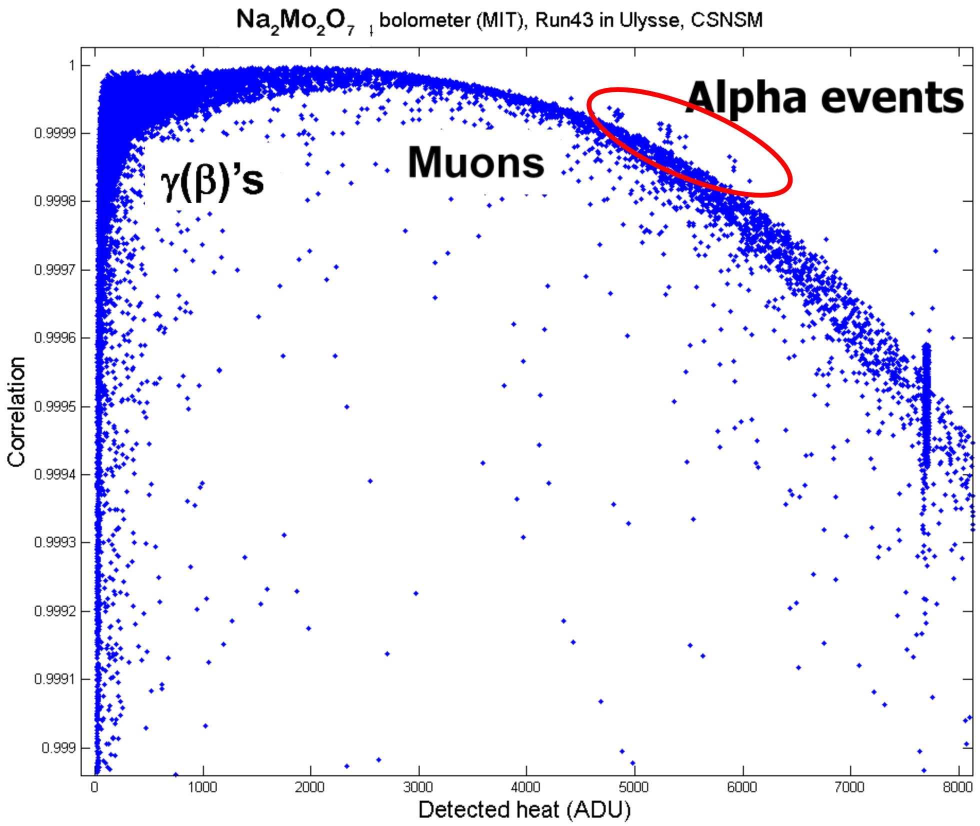


$Na_2Mo_2O_7$



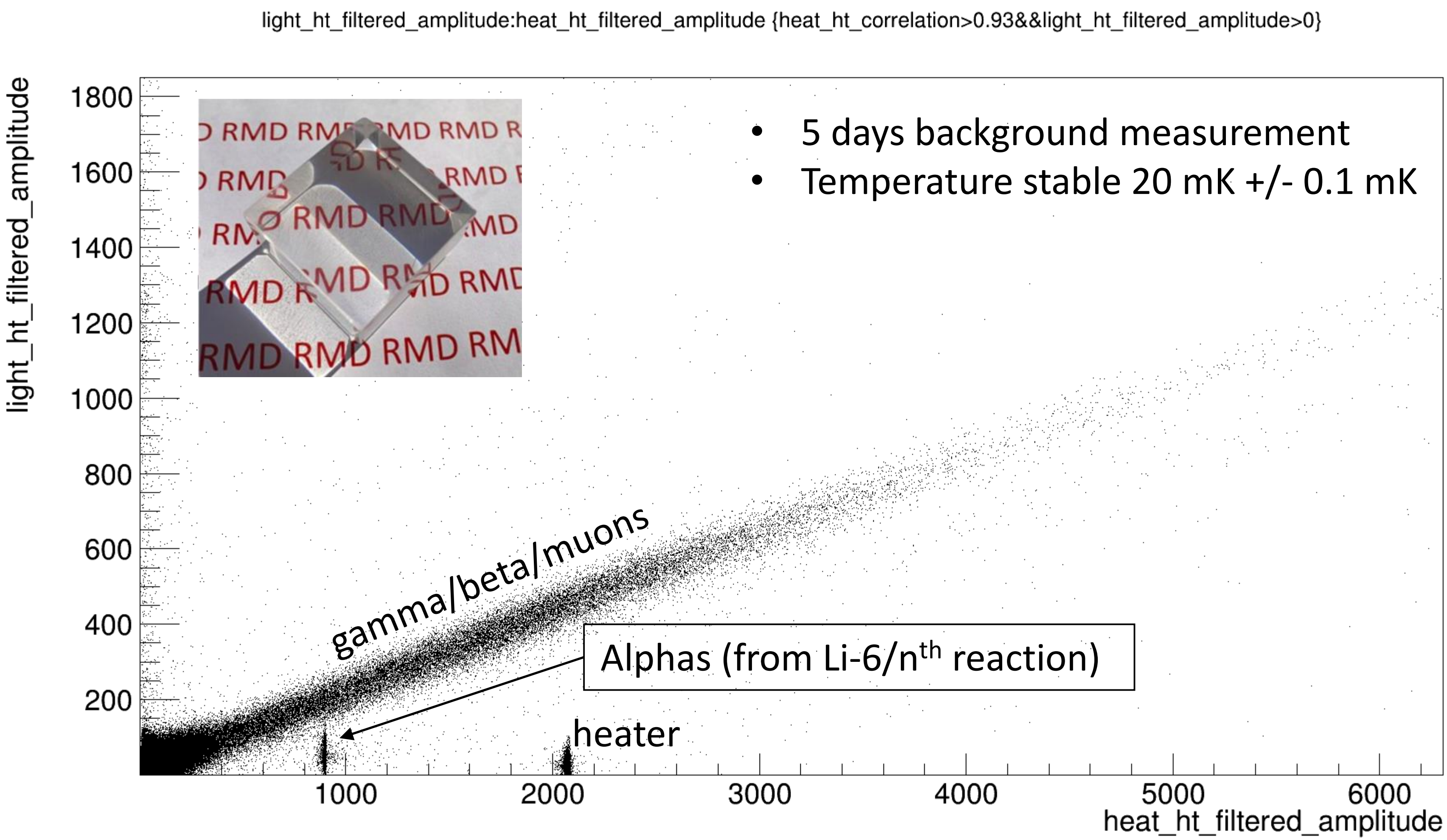
- Alpha events come from U and Th decay chains from internal crystal background.
- Alphas are at energy similar to expected 0νββ decay, so discrimination is crucial.
- Muon events will be shielded in underground laboratory.

Pulse Shape Discrimination Possible with $Na_2Mo_2O_7$



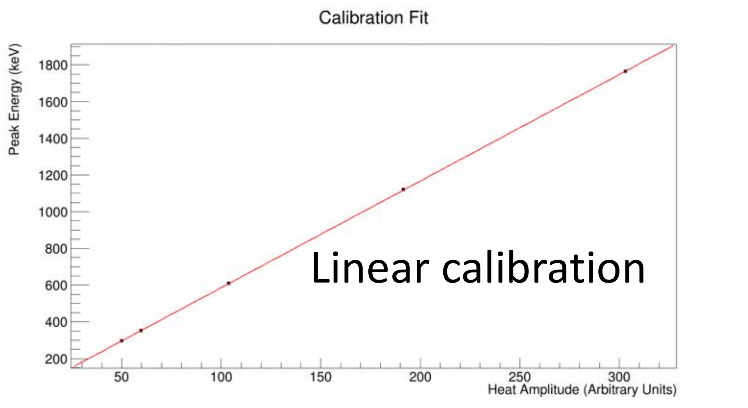
Light versus Heat Chart for LMO

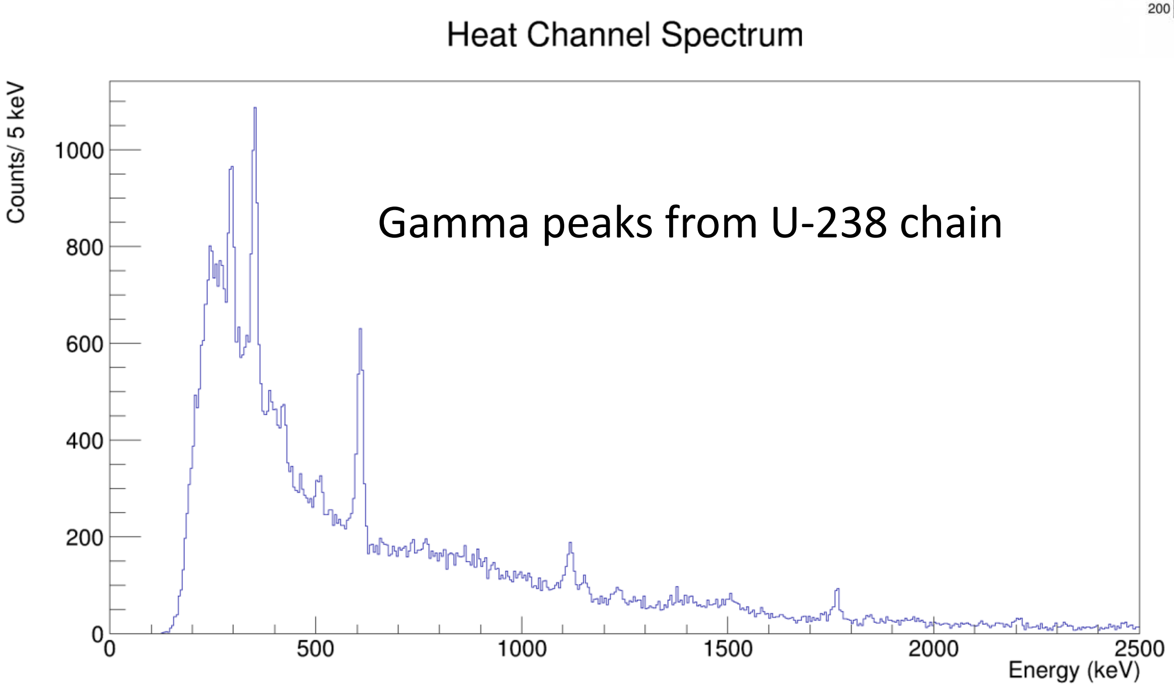
Good separation of alphas!



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

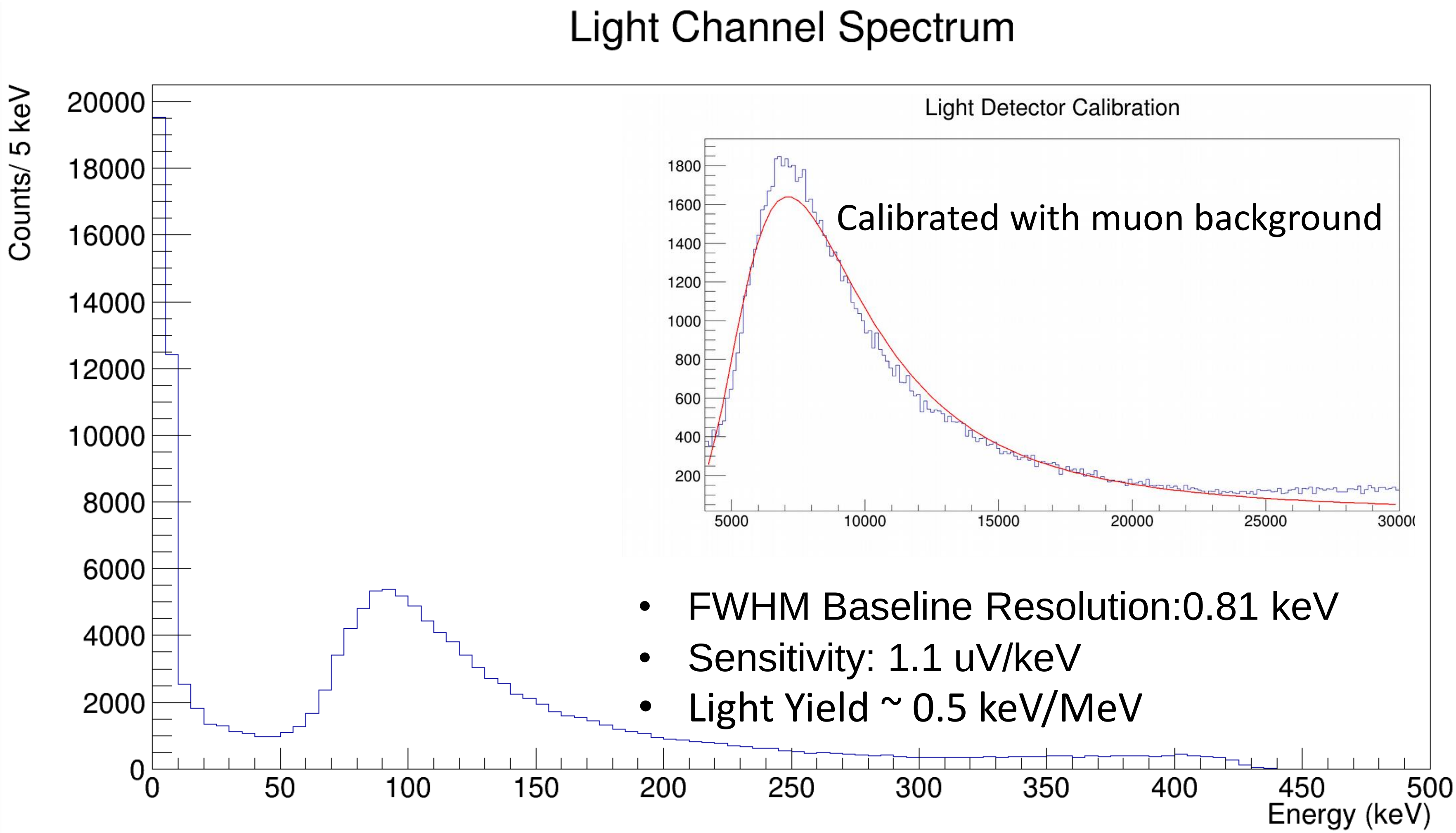
Calibrated Heat Spectrum for LMO





- 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

LMO Internal Background Limits

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	4	<0.24	<0.5
	Th-228	5520.08 ± 0.22	2	<0.12	<0.55
U-238	U-238	4269.7 ± 2.9	5	<0.31	<0.72
	Ra-226	4870.62 ± 0.25	-	<0.12	<0.50
	Rn-222	5590.4	2	<0.12	-
	Po-218	6002.4	1	<0.07	-
	Po-210	5407.45 ± 0.07	6	<0.38	<1.7
Pt-190	Pt-190	3252 ± 6	2	<0.12	-

- 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]

Summary and Plans

- Single crystals of $Na_2Mo_2O_7$ and Li_2MoO_4 were grown by Czochralski.
- Colorless transparent crystals were obtained.
- Cryogenic testing of scintillating bolometers showed good light output, good alpha separation, and low internal radioactivity.
- On-going work will be focused on increasing sample size, reducing internal background, and incorporating enriched ^{100}Mo for LMO Crystals.
  - Objective to become a fully qualified supplier for the CUPID project.