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

Presented by: Joshua Tower, RMD Inc.

August 8, 2018

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

DOE Contract: DE-SC0015200
SBIR Phase II
Period of Performance: 4/10/2017 - 4/9/2019
RMD Principal Investigator: Michael Squillante
DOE Technical Contact: Michelle D. Shinn

DOE-NP SBIR/STTR Exchange Meeting

August 7-8, 2018

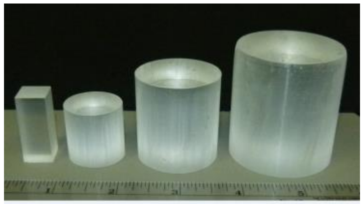
Rockville, Maryland

Outline of the Talk

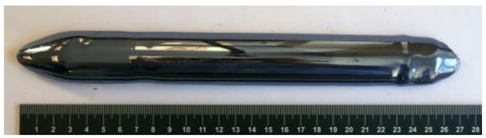
- Introduction
  - Radiation Monitoring Devices, Inc. (RMD) Research
  - Neutrinoless Double Beta Decay
- Phase I Summary
- Phase II Detailed Progress Report
- Plans for Continuing Work

RMD Basic and Applied Research and Development

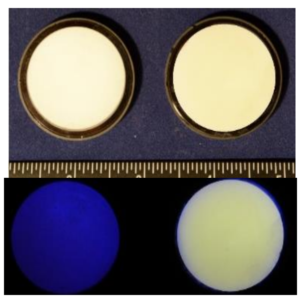
Materials Science



Scintillators

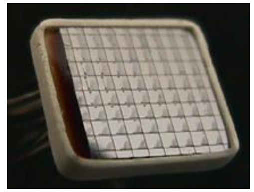


Semiconductors

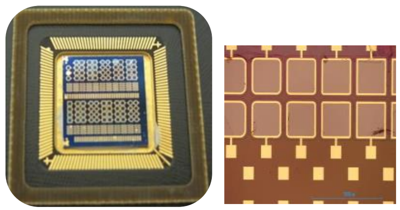


Imaging Screens

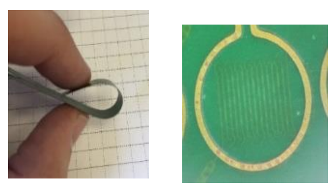
Sensors



APDs SSPMs
Photosensors

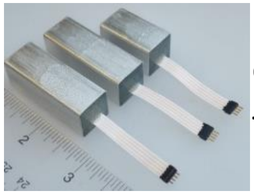


Wide Band Gap
Geiger Photodiodes



Flexible ECT Sensor
Array

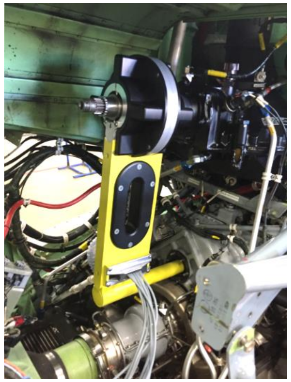
Instruments & Systems



CLYC detectors
for RadEye

HiRIS – High Resolution
Imaging System

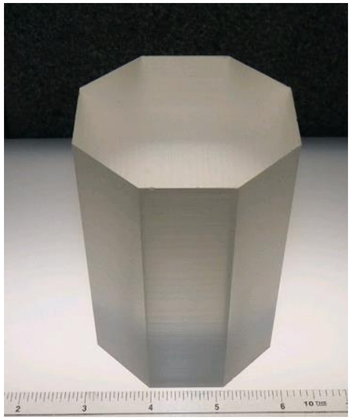
HeRMES

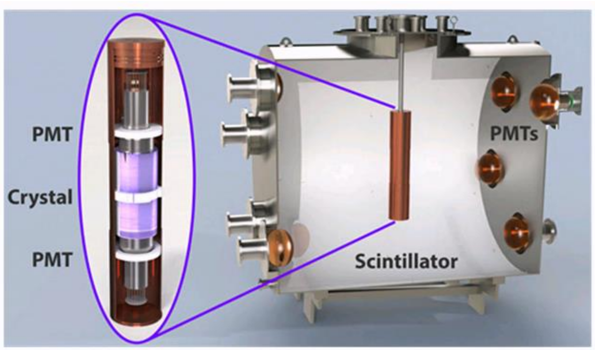


Fire Scout field prototype
maintenance tool*

RMD Low-Background Crystal Development for Rare Event Searches

Ultrapure NaI Scintillator for dark matter

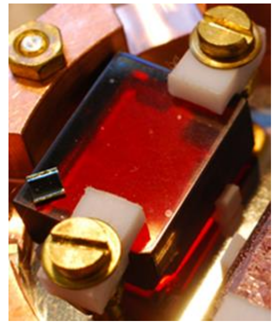




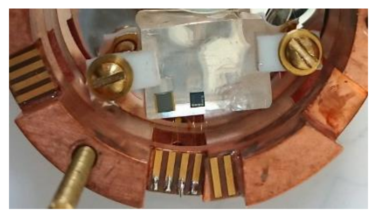
SABRE experiment which will use RMD's ultrapure NaI crystals

DOE-HEP

Scintillating Bolometers for 0νββ



$LiInSe_2$



$Na_2Mo_2O_7$

DOE-NP

Superconducting Bolometers for neutrino scattering

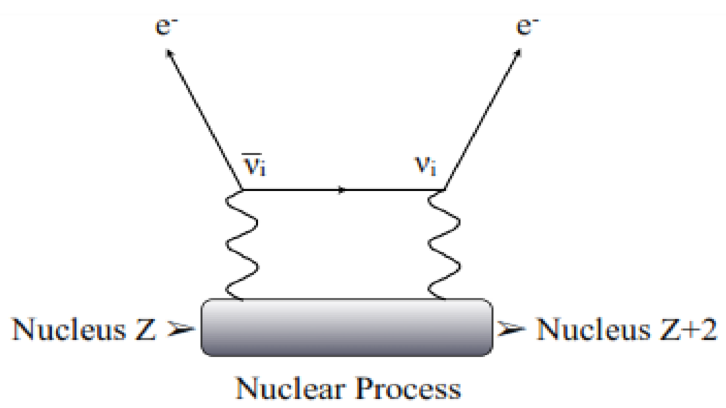
Zinc crystal in copper holder with NTD-Ge temperature detector

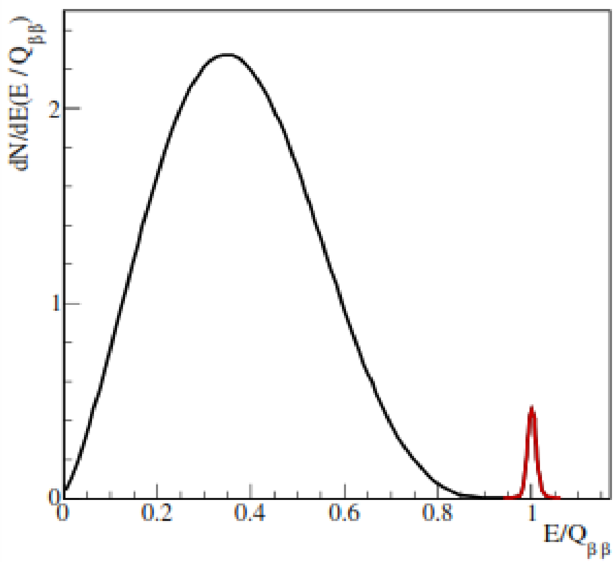
DOE-HEP

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)!

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.

^{100}Mo has promising properties!

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

SBIR Program Summary

- **Phase I**: Evaluate different crystal compositions for possible use in neutrinoless double beta decay experiments.
  - Samples were tested as scintillating bolometers through our collaborators at MIT*.
  - $Na_2Mo_2O_7$ was selected for further development in Phase II.
- **Phase II**:
  - Scale-up crystal growth to 2” diameter ($Na_2Mo_2O_7$).
  - Improve radio-purity.
  - Optimize surface preparation methods.
  - Provide sample crystals for cryogenic scintillating bolometer testing.

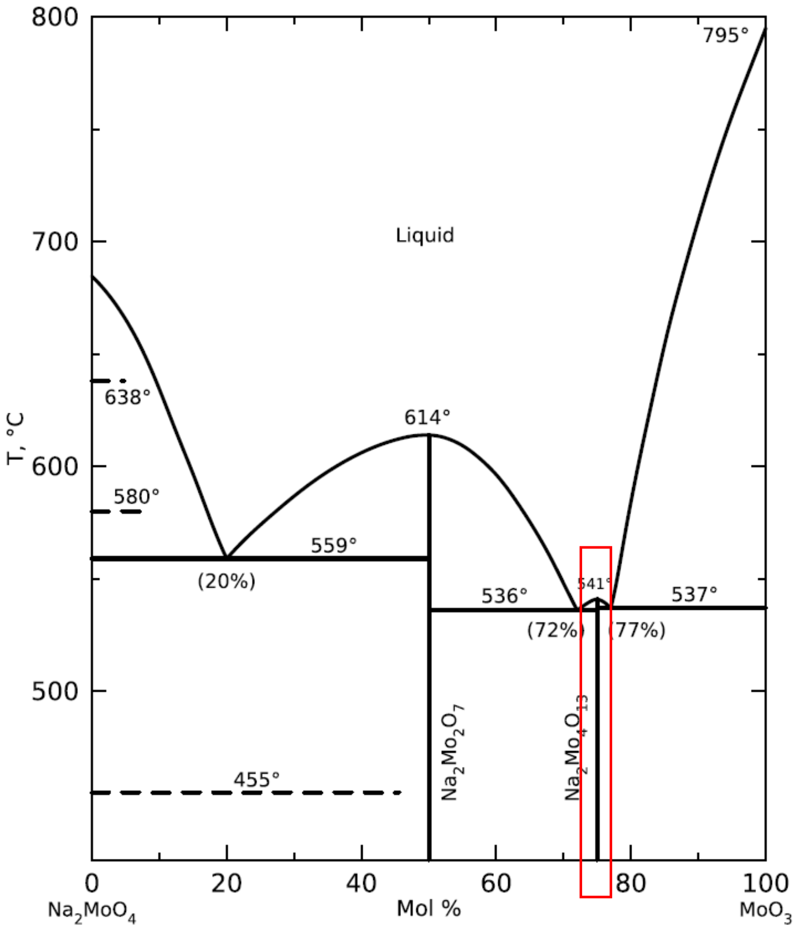
**RMD has teamed up with MIT (Prof. Lindley Winslow), who is part of the CUORE and CUPID collaborations.*

Phase I Crystal Growth and Characterization

Materials Evaluated in Phase I

- $ZnMoO_4$
  - Difficult to grow
- $PbMoO_4$
  - Can be grown well, but Pb not desired
- $Na_2Mo_2O_7$ & $Na_2Mo_4O_{13}$
  - Most promising!
- $LiInSe_2$
  - Good properties, but indium not desired
  - Related compound $LiGaSe_2$ might be considered

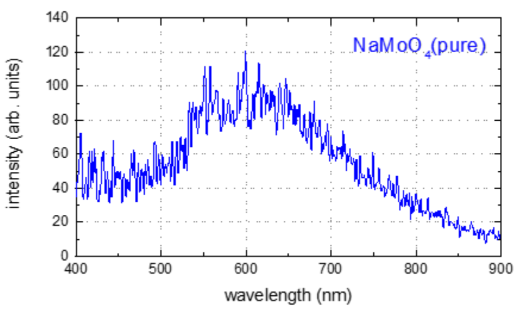
$Na_2Mo_4O_{13}$ Grown in Phase I



Summary:

- Initial small crystal grown by vertical Bridgman
- Good scintillation properties
- Low-T evaluation by MIT
- Promising for high purity

Radioluminescence at room T



Initial VB-Grown $Na_2Mo_4O_{13}$

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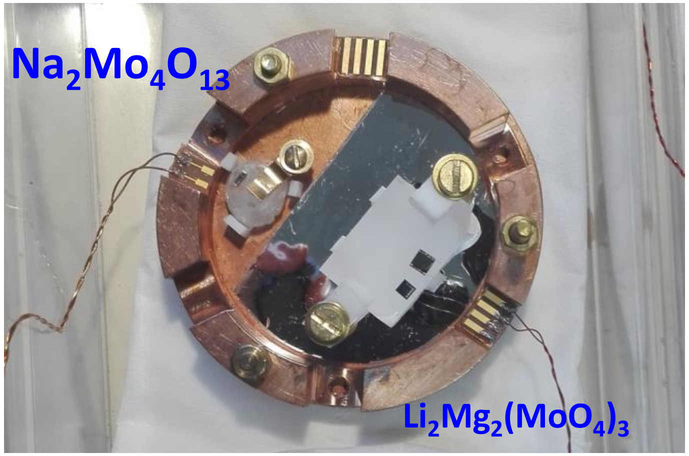
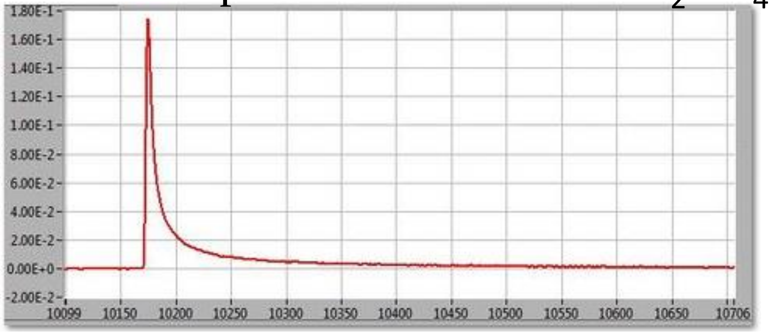
$Na_2Mo_4O_{13}$ Data from CSNSM in Orsay, France



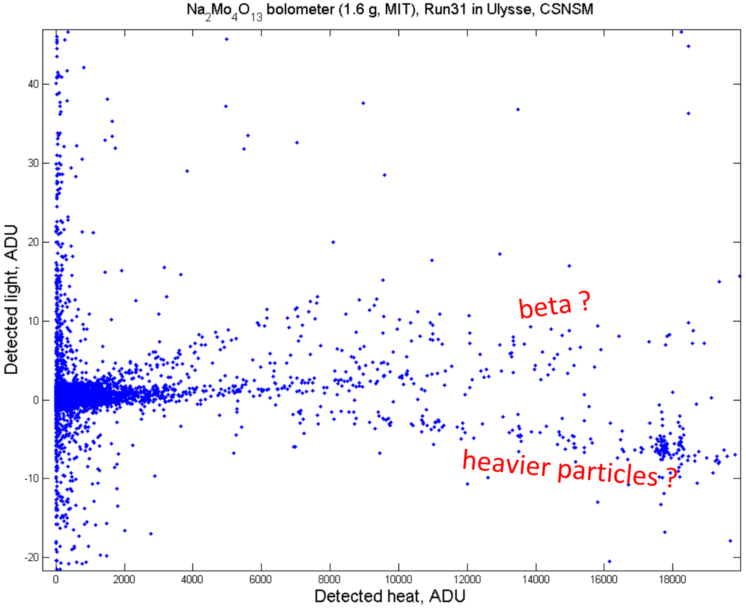
First scintillation pulse obtained from $Na_2Mo_4O_{13}$



“Ulysse” pulse-tube cryostat

Various crystals mounted in a tower

Background at Earth Surface



Phase II Progress Summary

- Developed synthesis process for $Na_2Mo_2O_7$
- Obtained and setup Czochralski (CZ) growth system
- Developed process for CZ growth of $Na_2Mo_2O_7$
  - Crystal orientation and seed fabrication
  - 1” process developed, 2” process underway
- Crystal cutting and polishing
- Basic optical and structural characterization
- Samples fabricated for cryogenic scintillating bolometer testing
- Scintillating bolometer testing was done at CSNSM

$Na_2Mo_2O_7$ Synthesis Process

1. MoO_3 99.9995% + Na_2CO_3 99.997% powders from Alfa Aesar
   - $2\,MoO_3 + Na_2CO_3 \longrightarrow Na_2Mo_2O_7 + CO_2$
2. Mix MoO_3 and Na_2CO_3 in a plastic bottle overnight on a roller
3. Press the mixture in a Teflon piston jig with a cold isostatic press to form a compact and dense mixture puck
4. Place and melt the puck inside a 100ml platinum crucible at 650C
5. Repeat steps 1-4 one more time to fill crucible

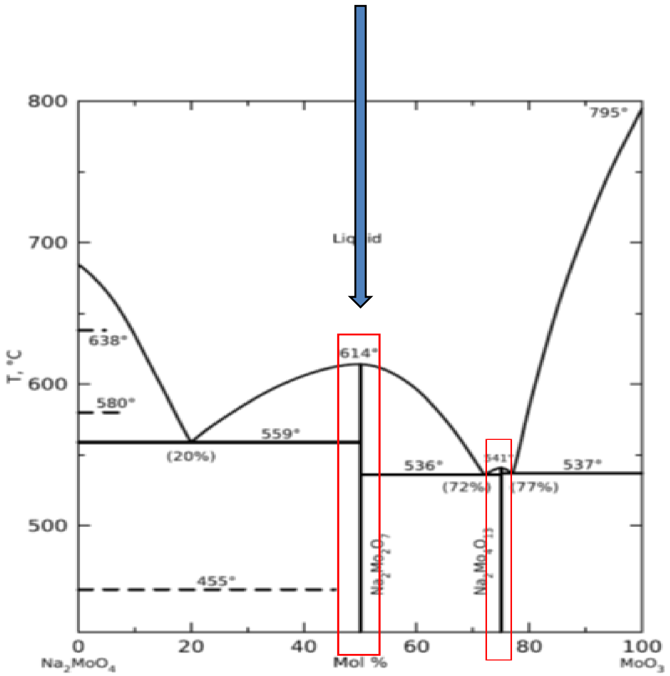
Cold isostatic Press

Puck generated

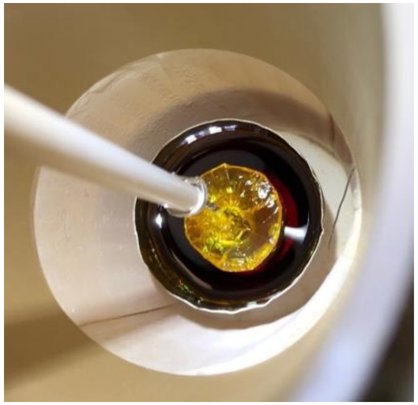
Puck melted in Pt crucible

$Na_2Mo_2O_7$ Grown in Phase II

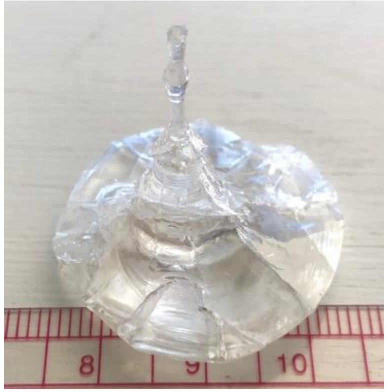
Congruent growth possible for $Na_2Mo_2O_7$



Czochralski growth method used



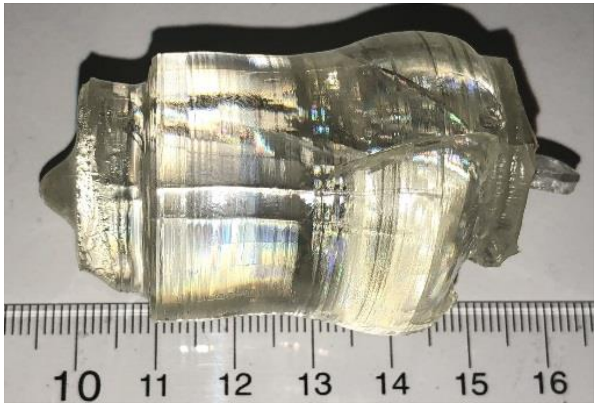
First growth run made using Pt wire as a seed



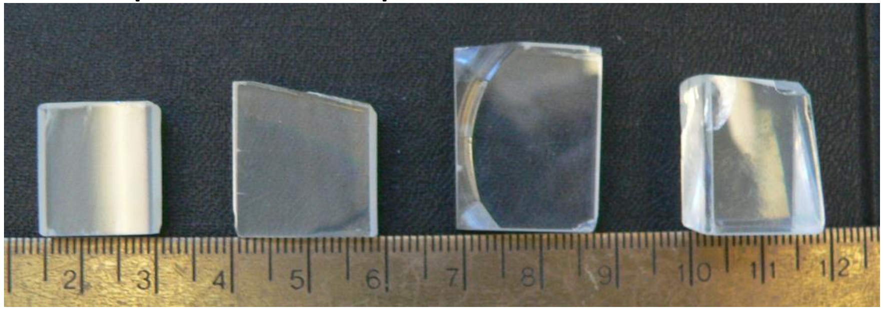
Initial crystal was cut into seeds for subsequent growth runs.

Seeded Cz Growth of $Na_2Mo_2O_7$

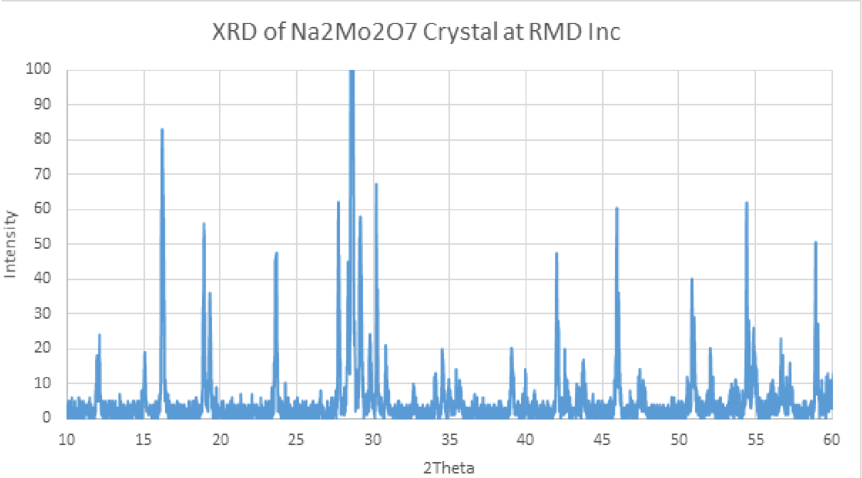
As-grown Ingot

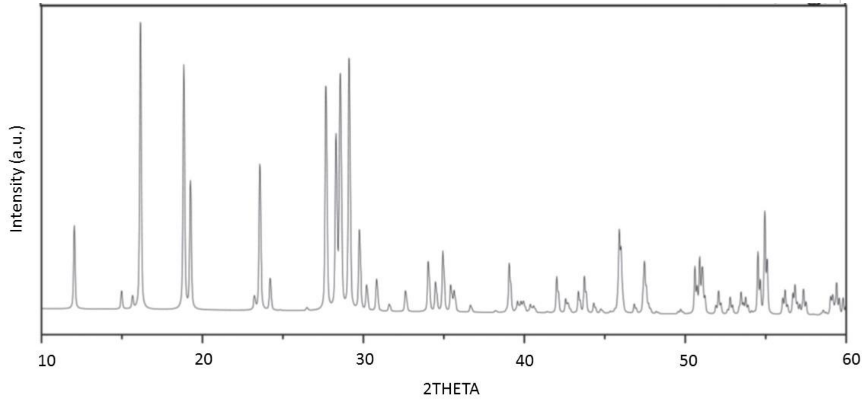


Samples cut and polished for evaluation



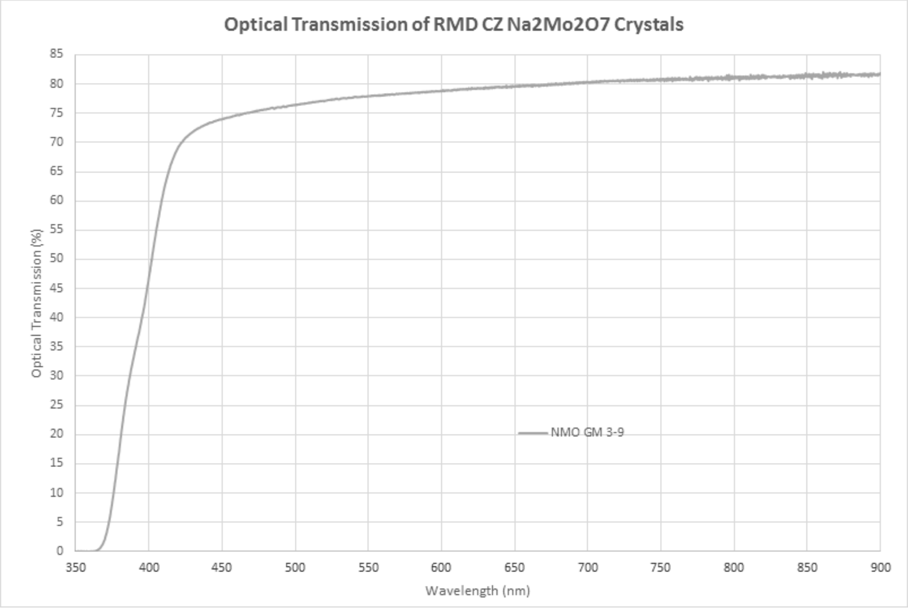
Powder XRD confirmed composition





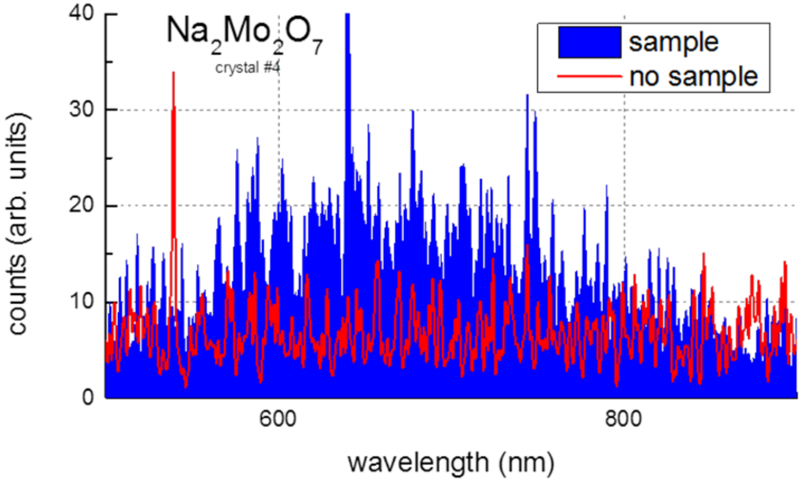
XRPD Simulated Pattern of Na2Mo2O7

Colorless Crystal with Excellent Transparency



Good transparency in indicative of high purity

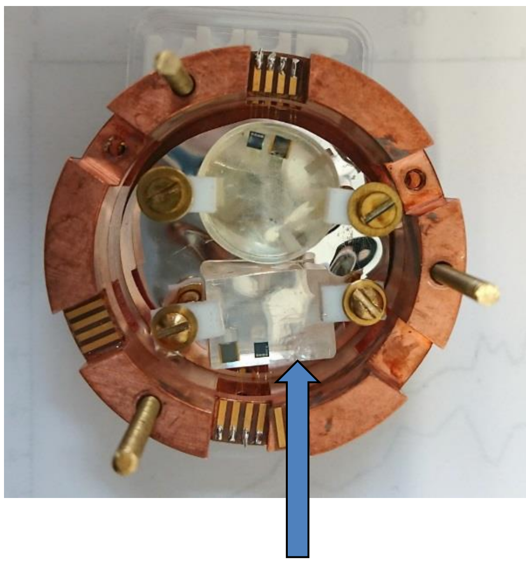
Room temperature emission spectrum of a $Na_2Mo_2O_7$ crystal



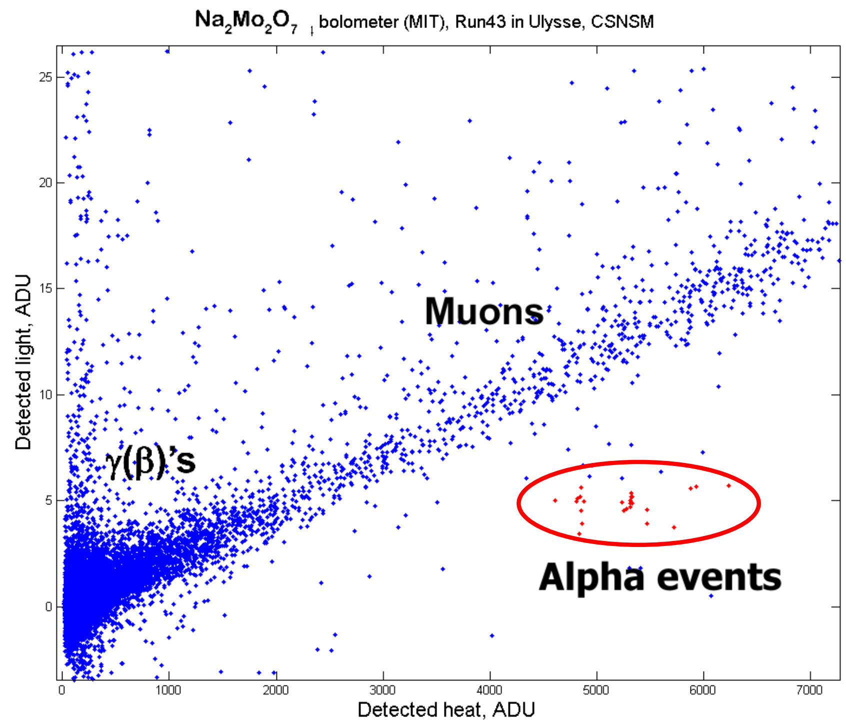
Emission is very weak at room T, but increases by many times at low T.

Good Particle Discrimination in Light-vs-Heat Plot

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

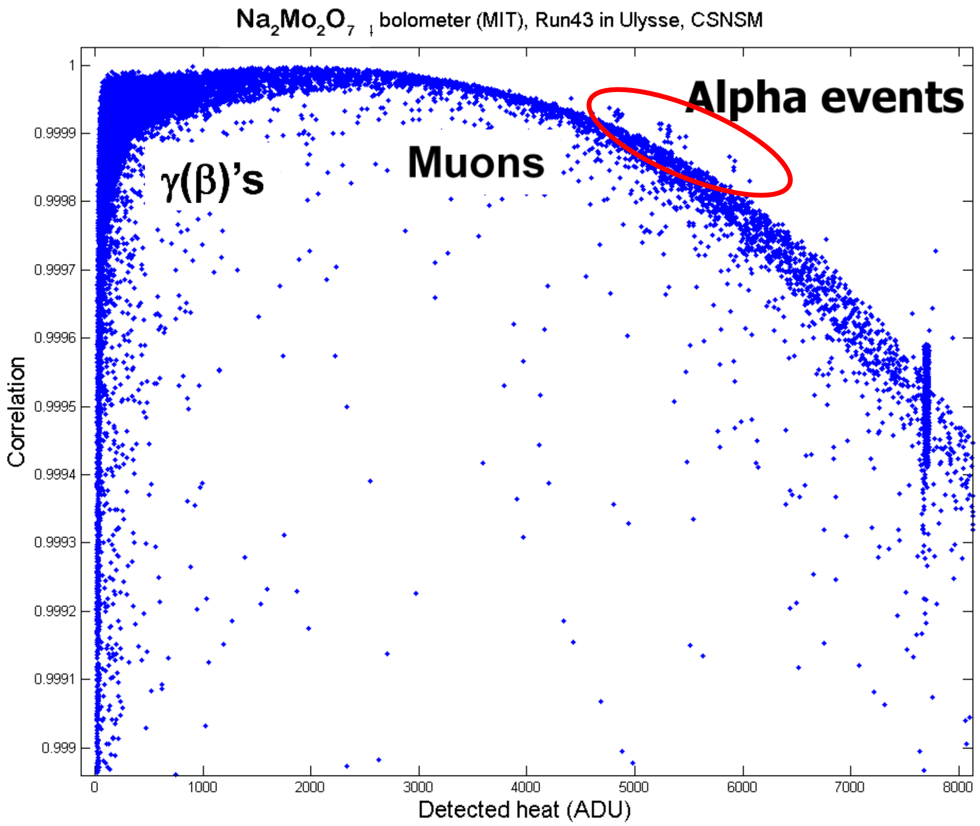


$Na_2Mo_2O_7$



- 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.

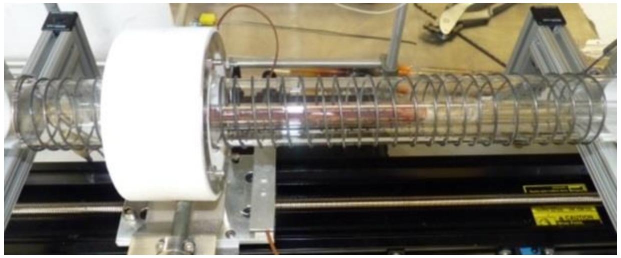
Pulse Shape Discrimination Possible with $Na_2Mo_2O_7$



Material Purification

- Based on the initial cryogenic testing, the current level of crystal "bulk" purity is believed to be suitable for 0νββ experiments!
  – Nevertheless, we plan to test zone refining for further purification and use it as needed.

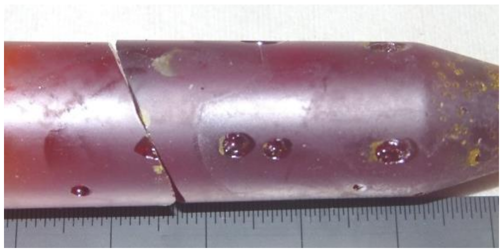
RMD Zone Refiner



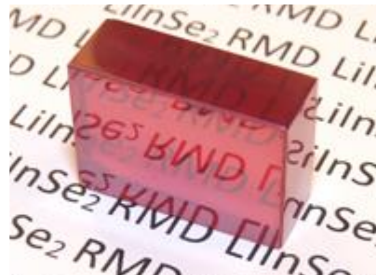
- Further attention will be on Surface Purity
  – Final polishing and cleaning in a radon-free clean environment is crucial.

LiInSe$_2$ (Phase I)

As-Grown Ingot

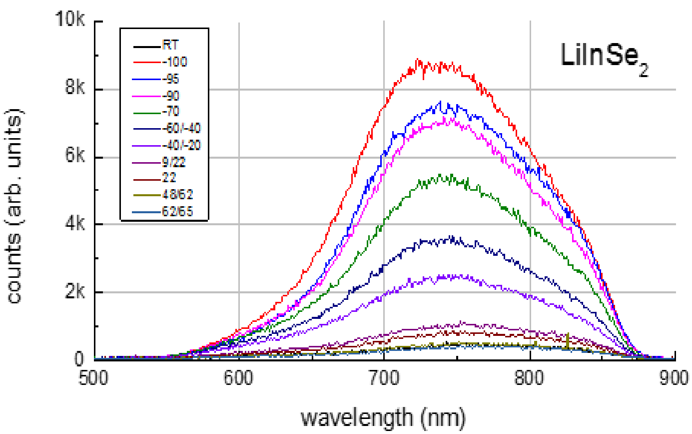


Cut and polished

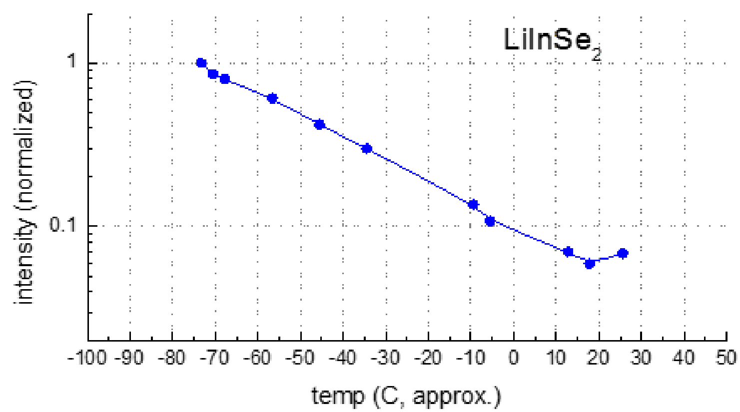


In cryostat sample holder

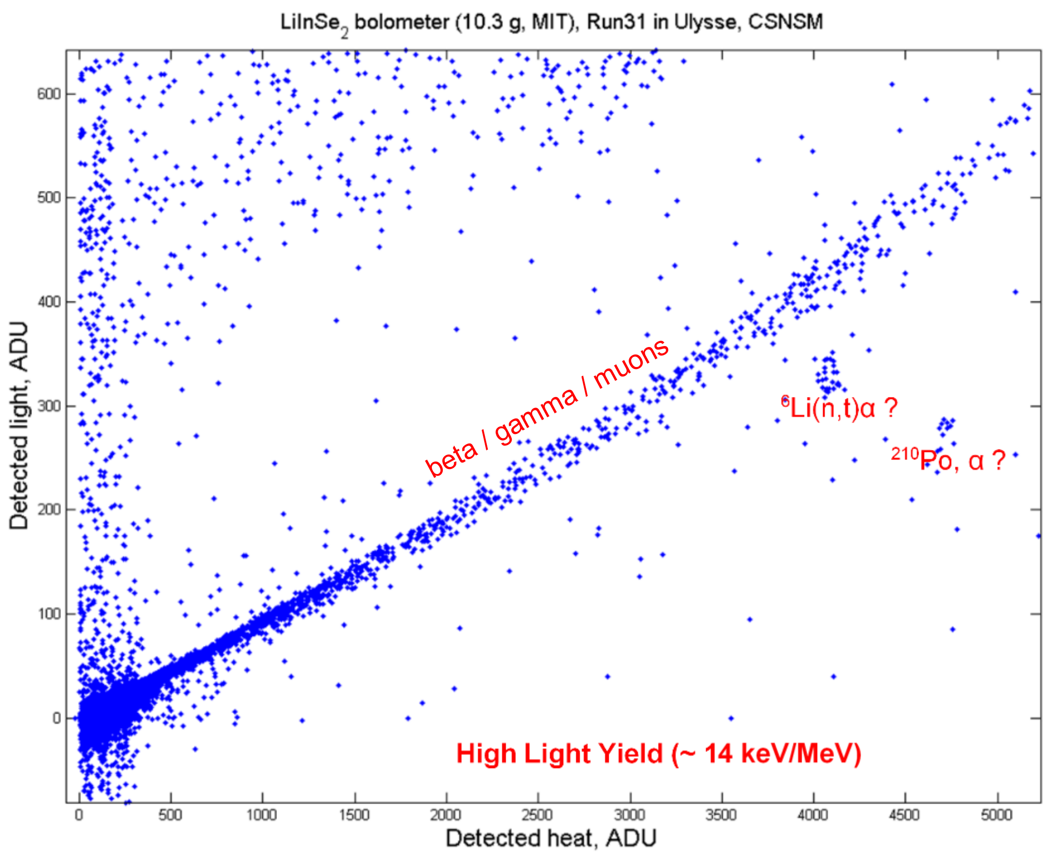
Emission Spectra at Different Temps



Emission Intensity vs Temperature



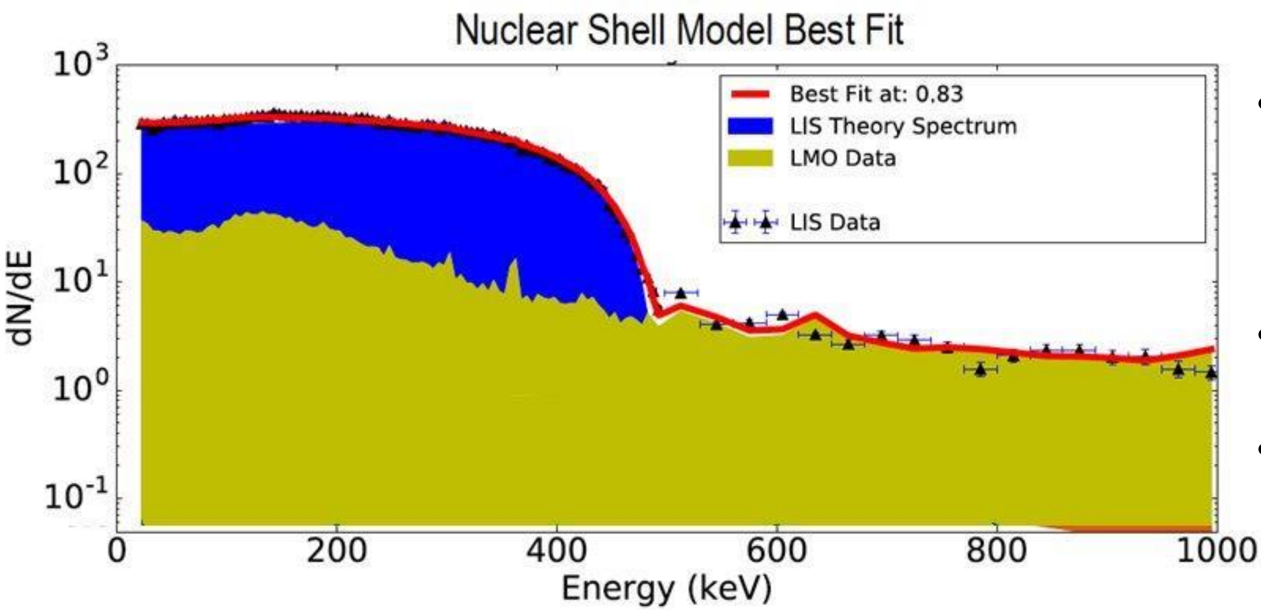
Light vs Heat Plot for $LiInSe_2$



- Good particle discrimination
- High light yield
- Neutrons detected through ^6Li reaction

Recent Beta Decay Spectrum of ^{115}In Measured in $LiInSe_2$

Scintillating Bolometer data measured ~ 20 mK in Ulysse cryostat



- Indium beta decay spectrum is significantly above the background, even on the Earth's surface.
- Indium Half life = 4.4×10^{14} years.
- Background measured by $LiMoO_4$ bolometer.

This is one of the first demonstrations of a precision beta decay measurement using the "source-equals-detector" setup that is the key to many rare event searches.

On-Going Work Plans for Year 2

- Scale-up $Na_2Mo_2O_7$ crystal size to 2" diameter
  - New heaters and crucibles have been ordered.
- Perform zone refining purification and evaluate results
- Complete the development of clean surface finishing process
- Deliver 1" and 2" diameter samples to MIT for cryogenic testing and purity evaluation