A Direct Search for the ^{229}Th Isomer Transition using the ALS Synchrotron: Measurements of Candidate Host Crystals

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Eric Hudson
Scott Sullivan

EMMI Workshop

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Method: VUV transparent crystal hosts

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Laser “Mössbauer” Spectroscopy

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- Mössbauer spectroscopy has told us what to expect
  - Lamb-Dicke regime (no Doppler or recoil)
  - Shifts/Linewidths

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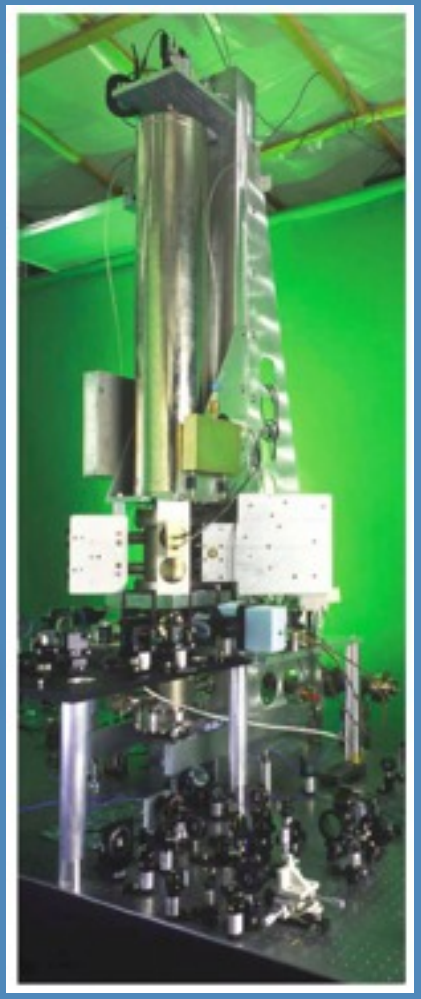

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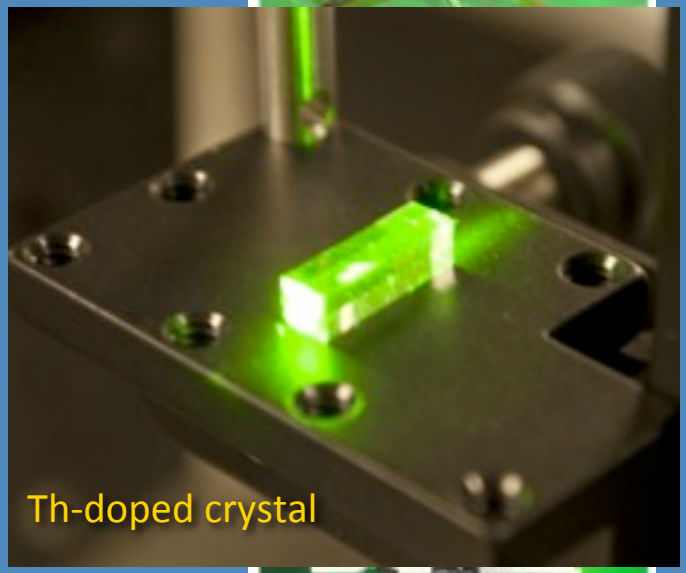
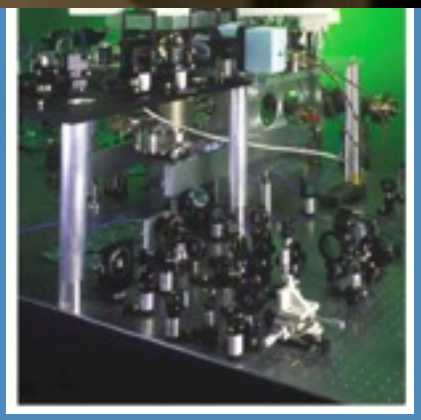




HFS Interaction summary

$$H_{HFS} = H_{E0} + H_{M1} + H_{E2} + \ldots$$

Shifts:

$$\Delta[\text{MHz}] = <100 + 0 + <100 \leq 200 \text{ MHz}$$

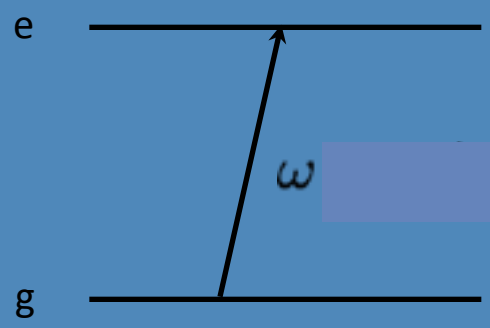
Broadening:

$$\delta[\text{kHz}] = 0.01 + <10 + 0 \leq 10 \text{ kHz}$$

Other effects:

- 2nd Order Doppler → 1 Hz/K
- Zero-phonon transition mode dependence → $T_D \geq 500$ K
  - Optical phonons frozen out (also possibly resolvable side-bands)
  - Acoustic phonons don't matter

Excitation Rate – 2 level system



Γ_N - Natural linewidth

Γ_b - Broadened linewidth

Δ - Light (laser) linewidth

$$\frac{dN_e}{dt} = +rN_g - \Gamma_N N_e - rN_e$$

$$r = \int \sigma(\omega)\frac{E_\lambda(\omega)}{\hbar\omega}d\omega \qquad \sigma(\omega) = \Gamma_N \frac{\lambda^2}{4} g(\omega)$$

$$g(\omega) = \frac{2}{\pi\Gamma_b}\frac{1}{1+4\left(\frac{\omega-\omega_o}{\Gamma_b}\right)^2} \qquad E_\lambda(\omega) = \frac{2I}{\pi\Delta}\frac{1}{1+4\left(\frac{\omega-\omega_L}{\Delta}\right)^2}$$

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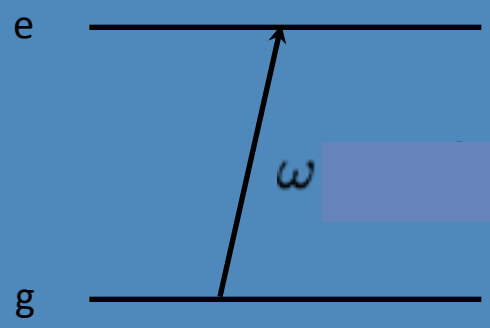

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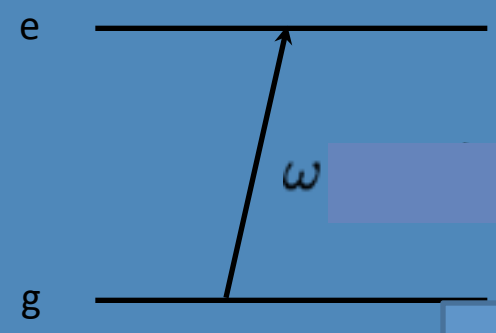

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N_g is HUGE!!!

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   (Schomburg *et al.* Appl. Phys. B **30**, 131-134 (1983))

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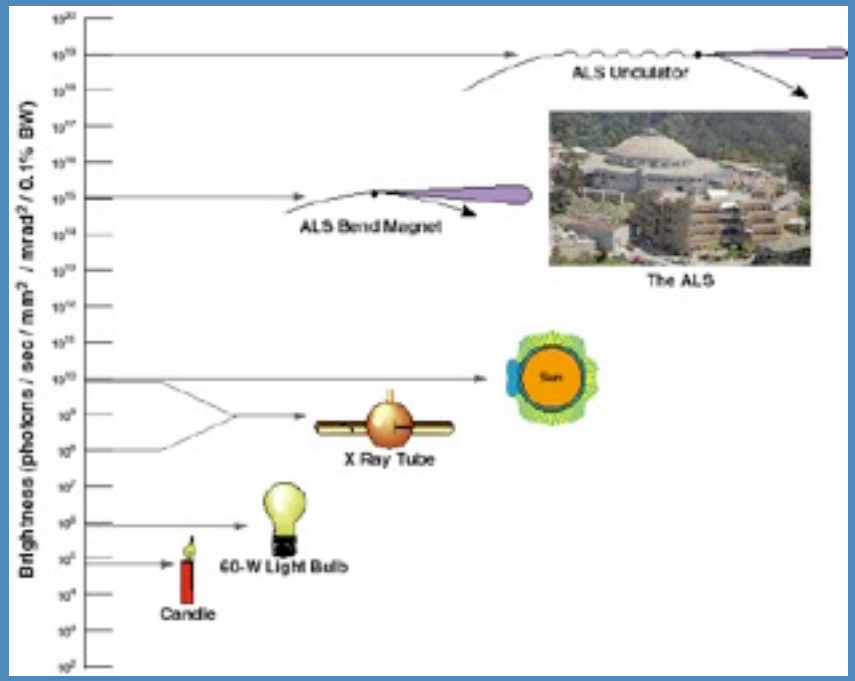

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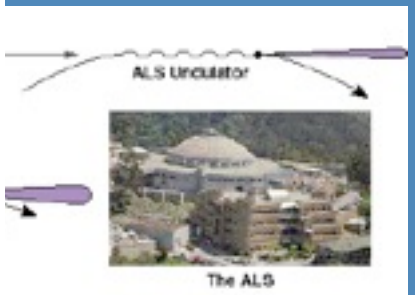

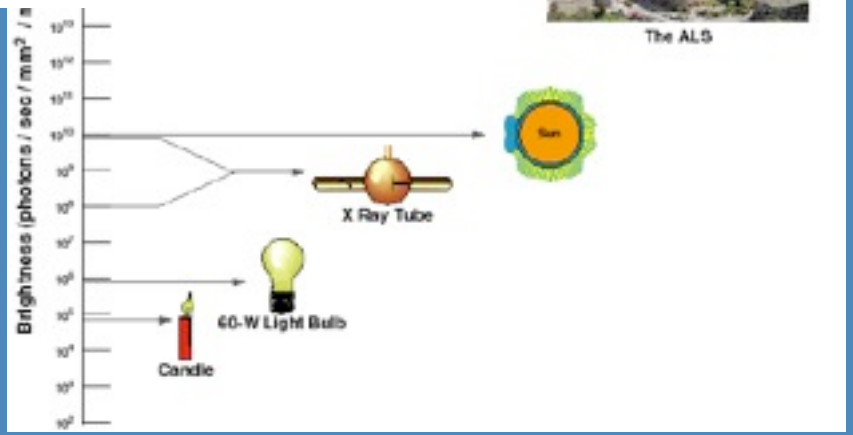

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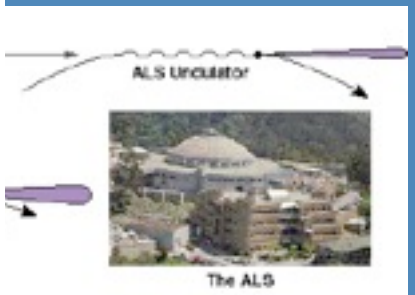

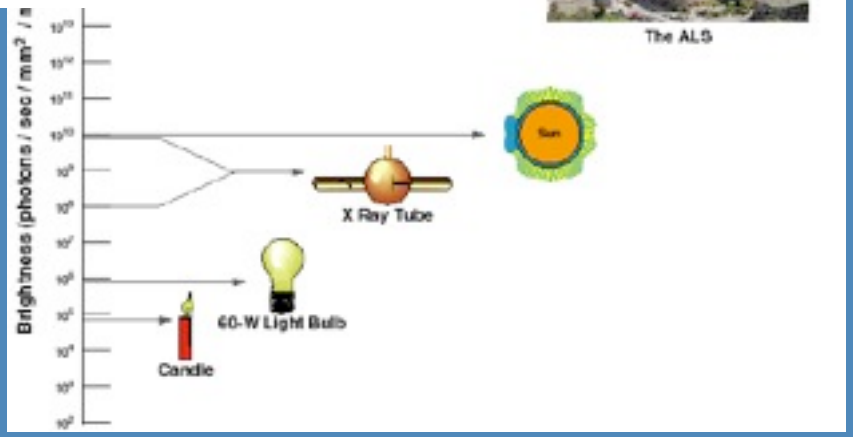

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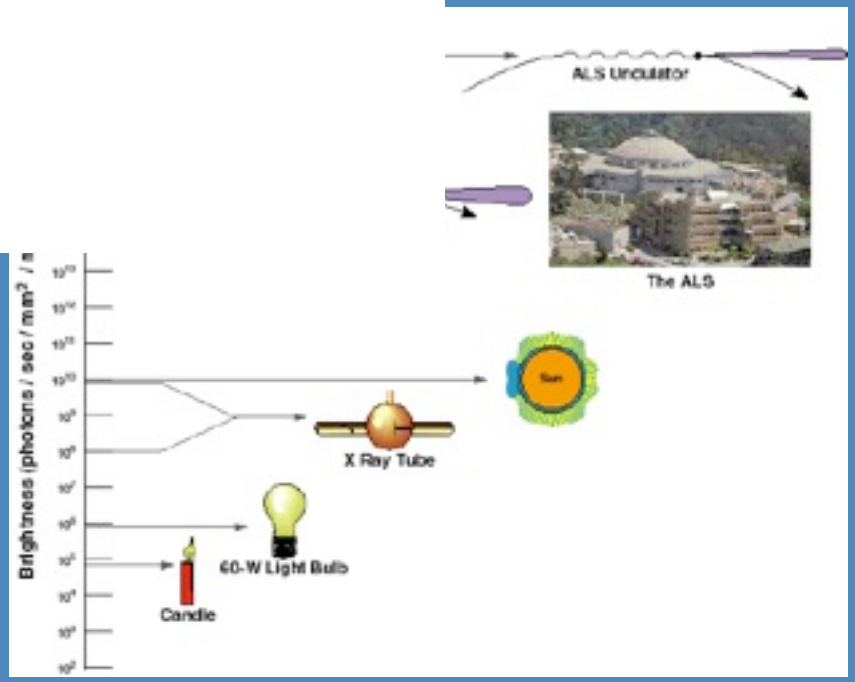

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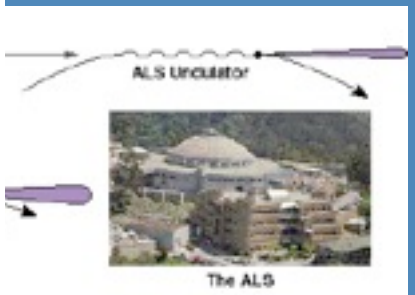

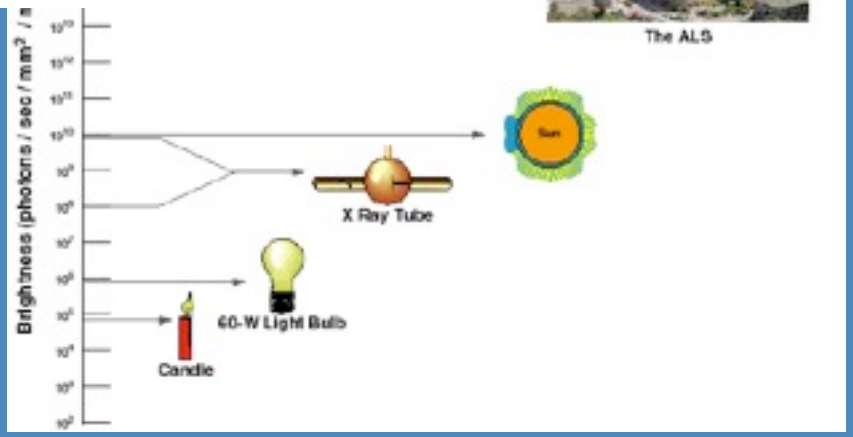

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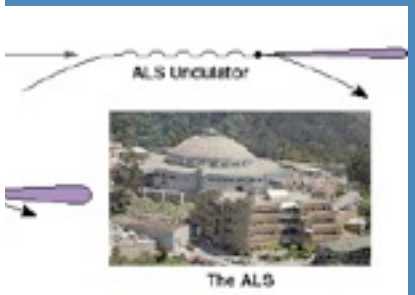

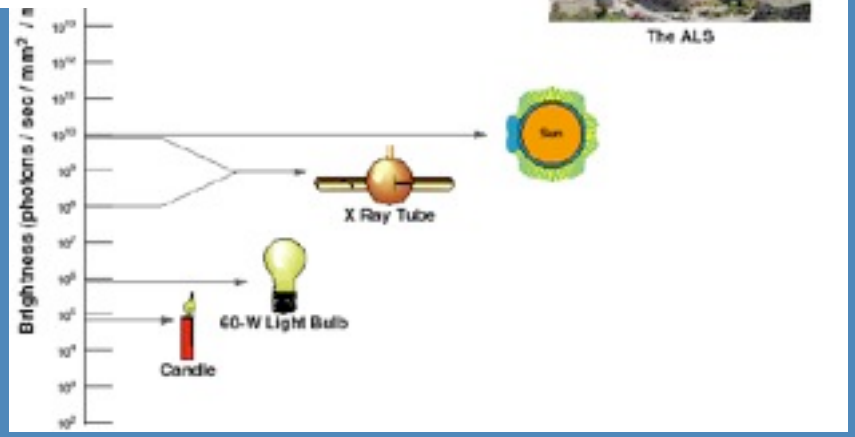

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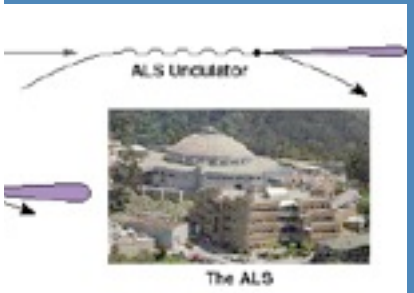
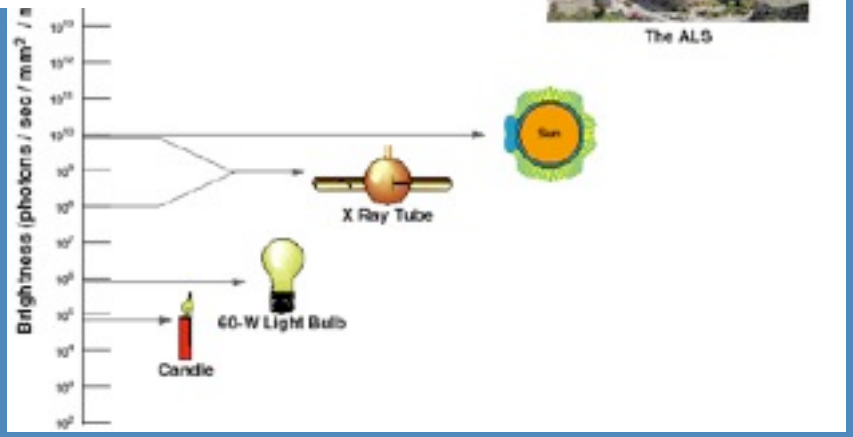
= 10^6 nuclei excited per second!!!





Expected Fluorescence Rate at ALS

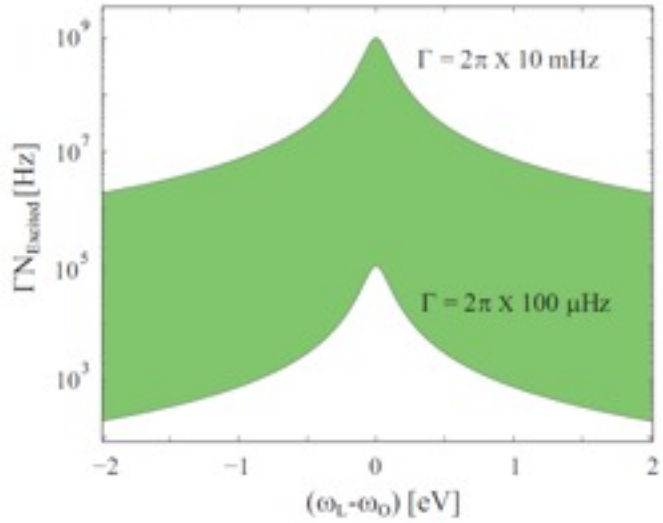


FIG. 1: (color online). Total fluorescence rate after sample illumination for 1000 s with the ALS is indicated by the shaded region. This region is bounded by the upper and lower limit expected for the excited state decay rate, Γ.

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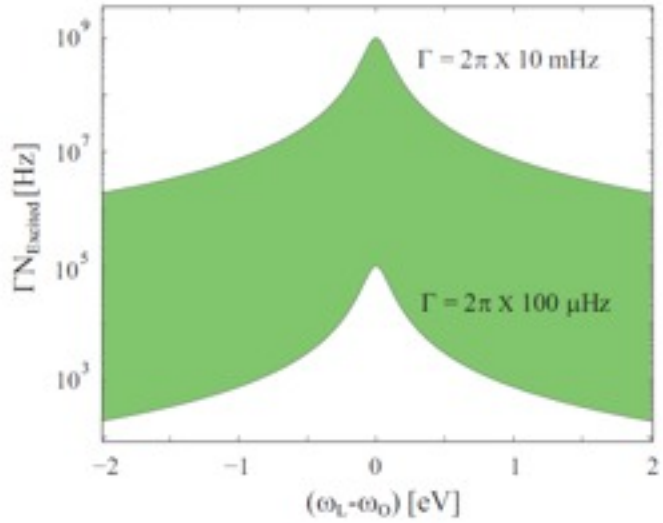

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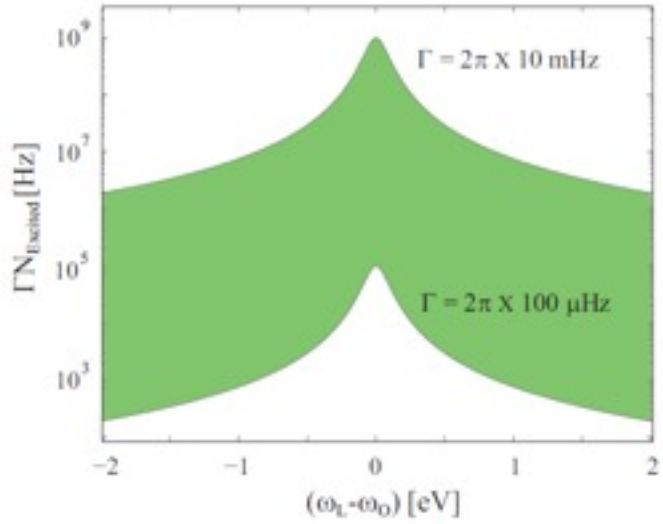

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Longer illumination times will allow the monochromator measurements narrowing range to 0.1 nm

Host Crystal Properties

- reasonably transparent in the VUV
  (down to ~160 nm)

- pure crystalline structure with all e^-'s paired
- must chemically accept ^{229}Th in the 4+ charge state
- resilient to radiation damage from alphas emitted by ^{229}Th (~100 uCi)
- low VUV induced fluorescent backgrounds

Crystals Tested To Date

- So far we have grown (w/ Th-232) and tested:
  - Th:LiCAF (also pure LiCAF)
  - Th:LiSAF (also pure LiSAF)
  - Th:NaYF
  - Th:YLF
  - Th:$BaMgF_4$
  - Th:Li_2ZiF_6
  - Na_2ThF_6

Thorium Doping Tests

Properly measuring doping concentration:

Variability for the same sample (Th:LiCAF) using different techniques.

Results for one growth run:

Rutherford back-scattering (LANL)	500 ppm	5×10^{18} cm^{-3}
Secondary Ion MS	10 ppm	1×10^{17} cm^{-3}
Gas discharge MS	300 ppm	3×10^{18} cm^{-3}
Neutron Activation	1.4(7) ppm	1×10^{16} cm^{-3}

Thorium Doping Tests

Properly measuring doping concentration:

Only Neutron Activation provides reliable results

EAI Project: 5876-12

Thorium by Instrumental Neutron Activation Analysis

Sample ID	Mass (g)	Thorium (ppm)
11-4-15	0.522	26000 (±1300)
12-4-1	1.993	<2
12-4-2	0.739	130 (±7)

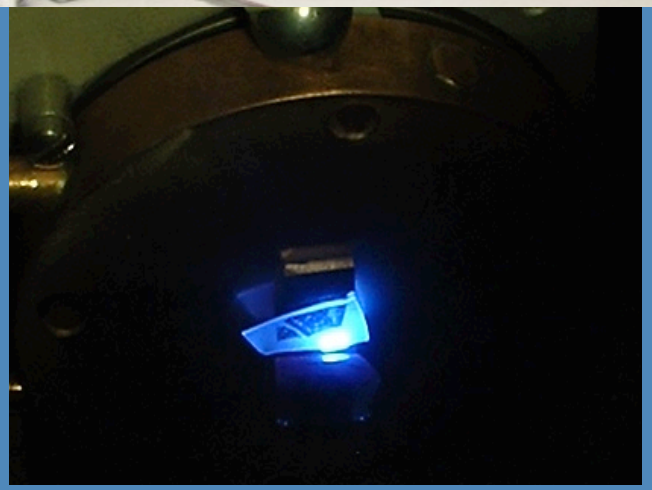
Note: Due to the nature of the samples, an irradiation of 1 minute was used.

Calibrated sample, 10000 ppm thorium

1% Th:LiSAF sample
~1% doping efficiency

Alpha Induced Backgrounds

- ^{229}Th emits 4.8 MeV alphas with half-life of 7880 yr
- Irradiated Th:LiCAF sample with 30 nA beam of 5 MeV alpha particles at IBML at LANL (Richard Greco and Justin Torgerson)
- Detected photons with PMT (R7639, 115 nm – 230 nm)
- Observed 0.3 photons/alpha
- Equivalent to 2 MHz background over the entire PMT bandwidth



ALS Background Runs

- Since August 2008 we have been awarded and used 144 beamtime hours (8 hour shifts)
- We have characterized 6 potential host crystals, most doped with ^{232}Th:
  - Th:NaYF, Th:YLF, Th:LiCAF, Th:LiSAF, Th:$BaMgF_4$, Na_2ThF_6
- While crystals do show some VUV induced fluorescence, it is of a low level and short-lived

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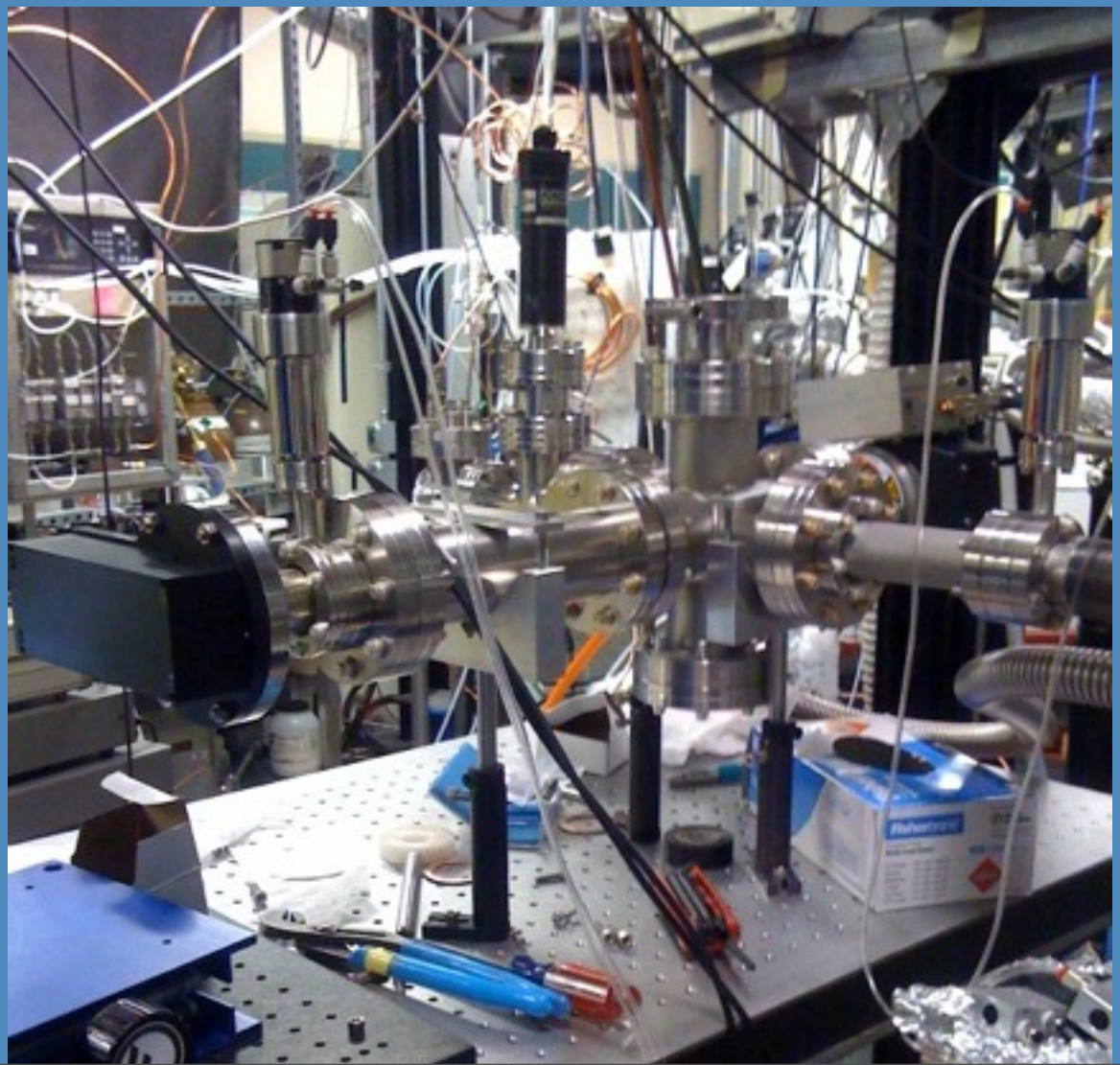

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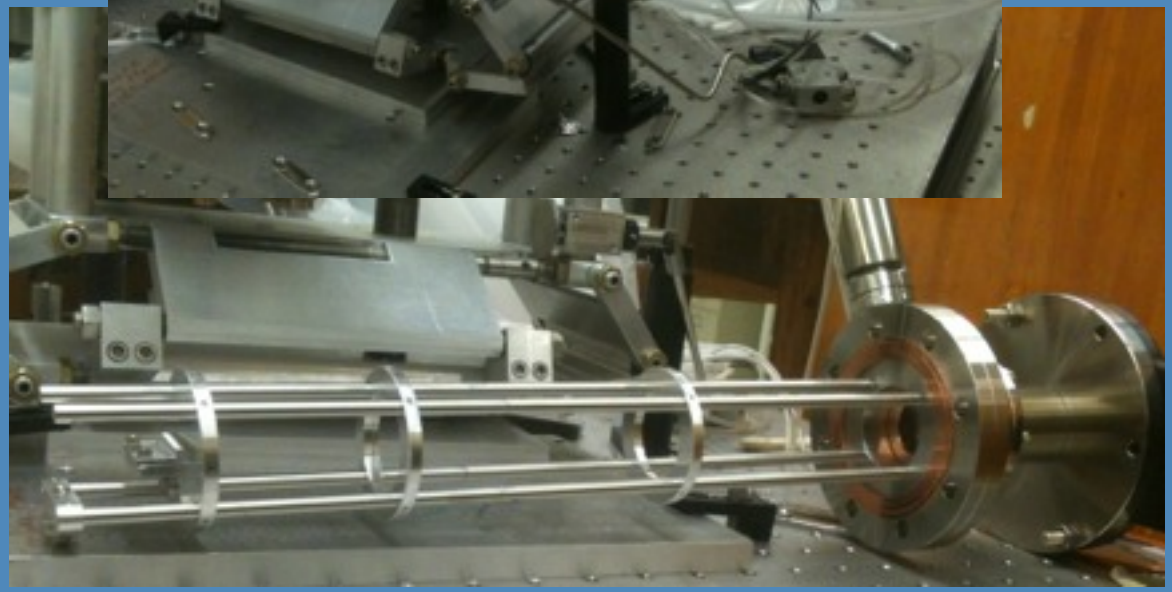
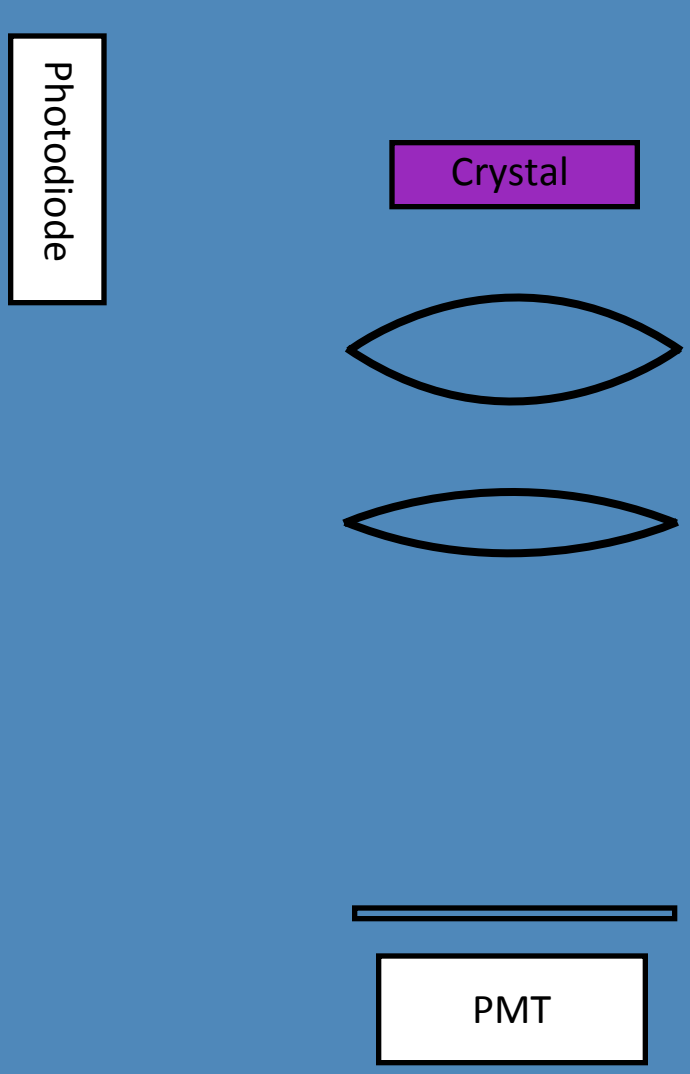


ALS - Crystal Measurement



Procedure:

1. Illuminate crystal (5-200 s)
2. Block ALS beam, open PMT shutter
3. Collect/count photons (5-100 s)
4. Repeat a few times
5. Change beam energy repeat

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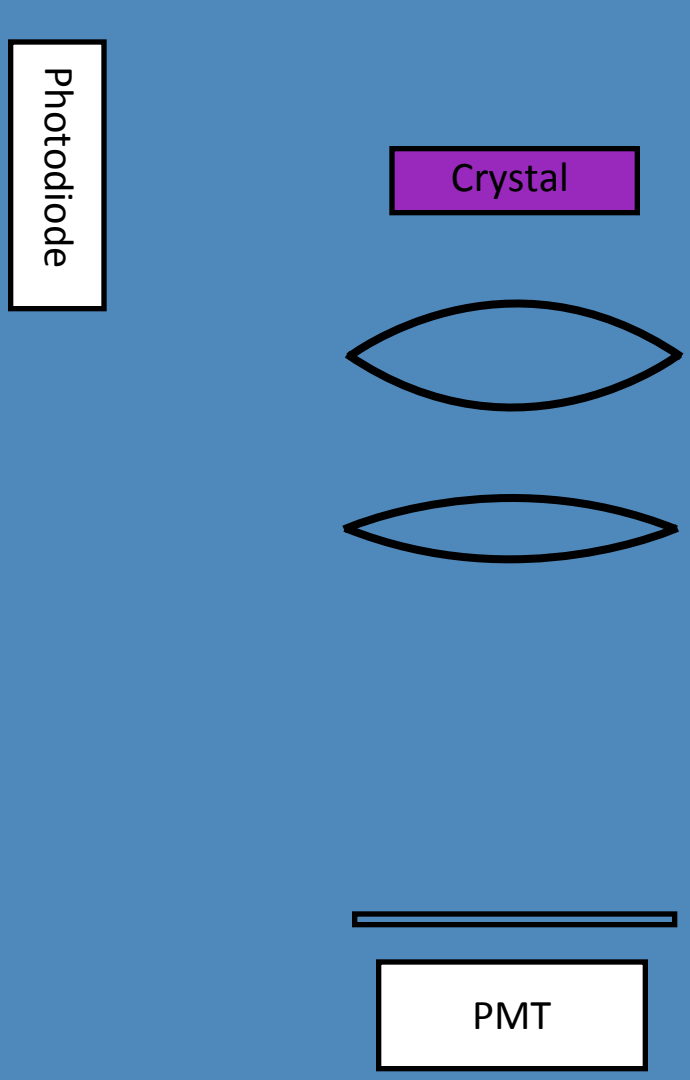

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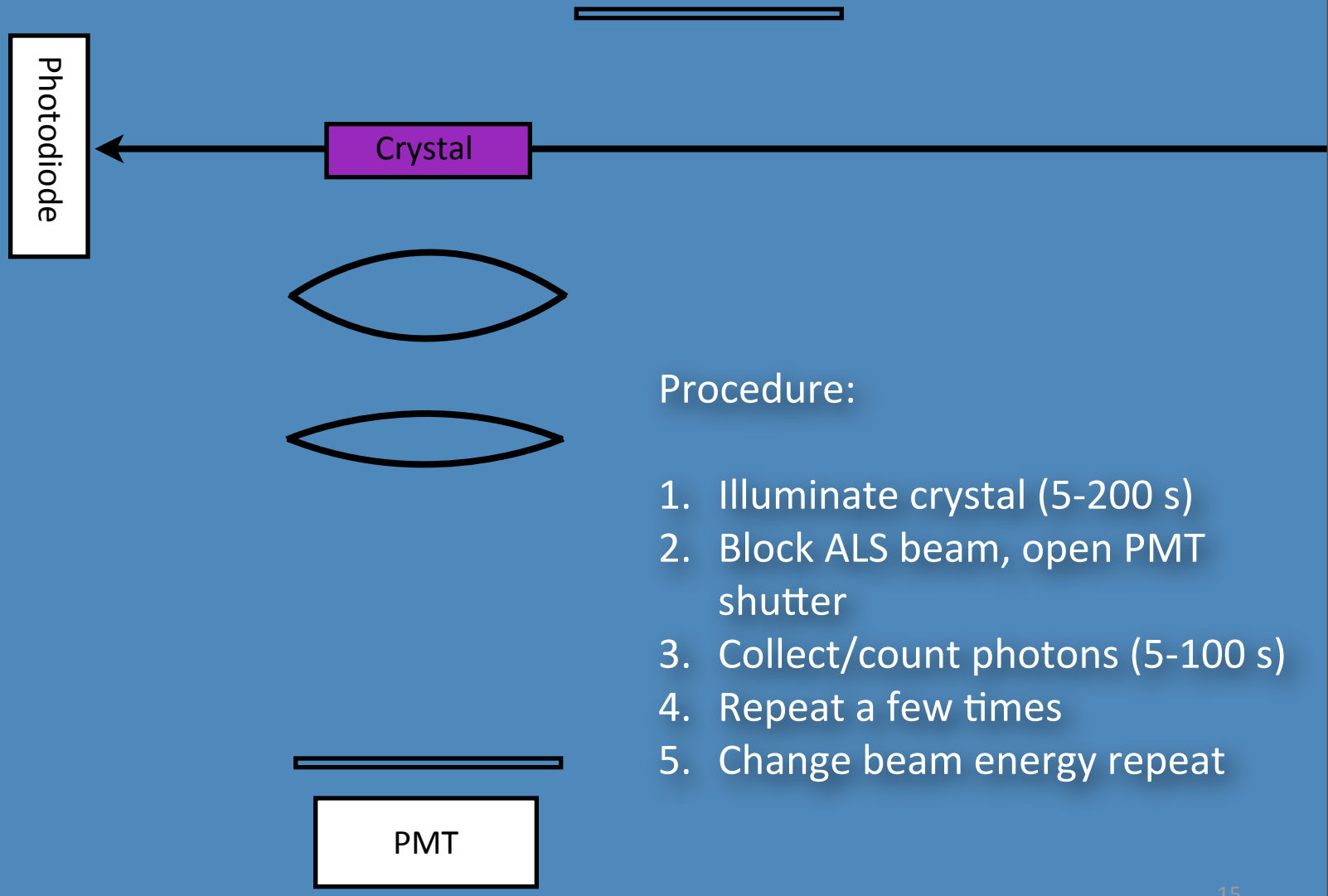

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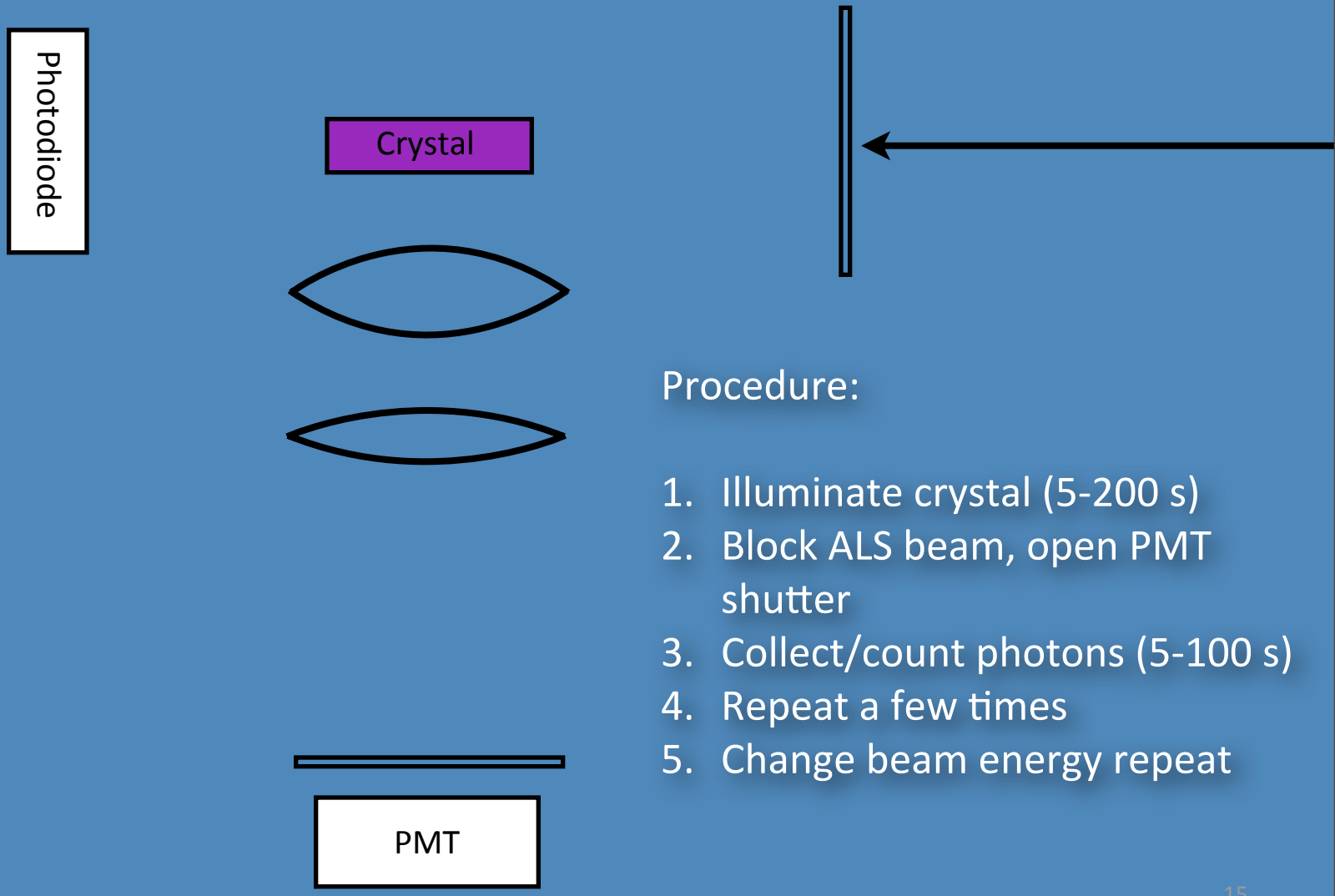

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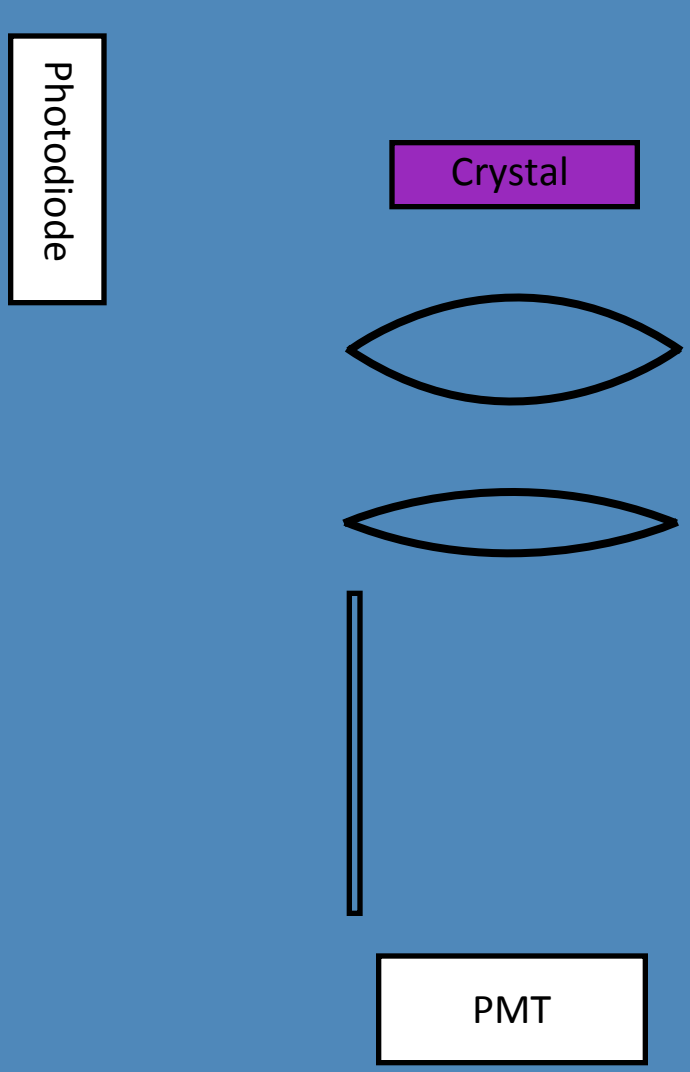

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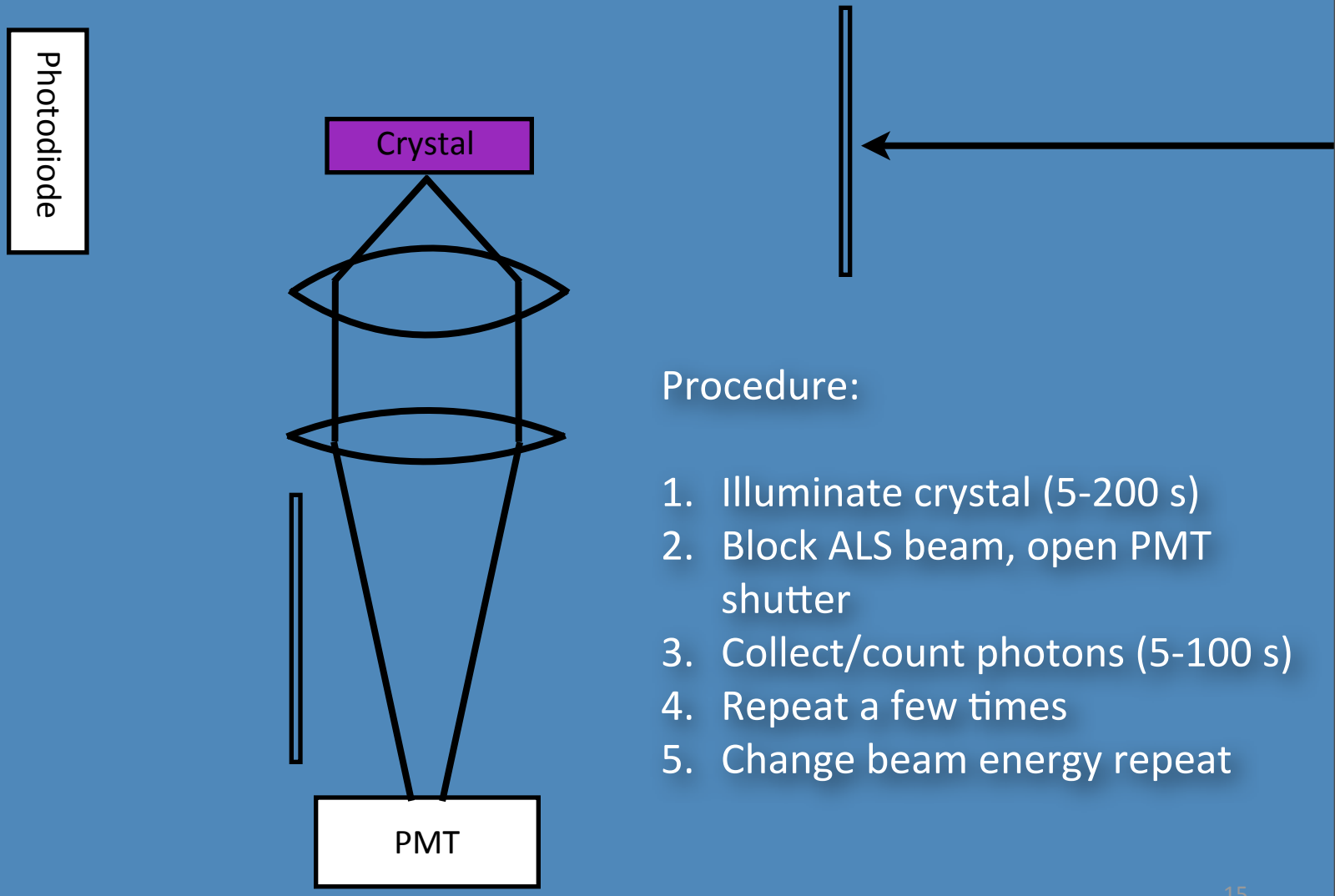
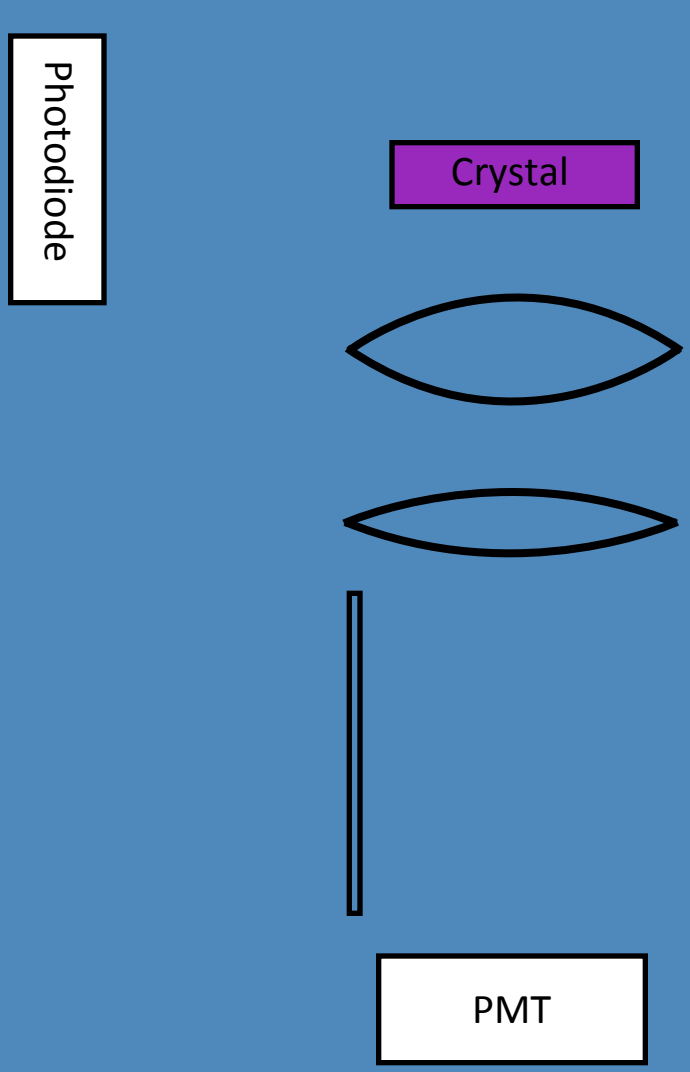
ALS - Crystal Measurement



Procedure:

1. Illuminate crystal (5-200 s)
2. Block ALS beam, open PMT shutter
3. Collect/count photons (5-100 s)
4. Repeat a few times
5. Change beam energy repeat

ALS - Crystal Measurement



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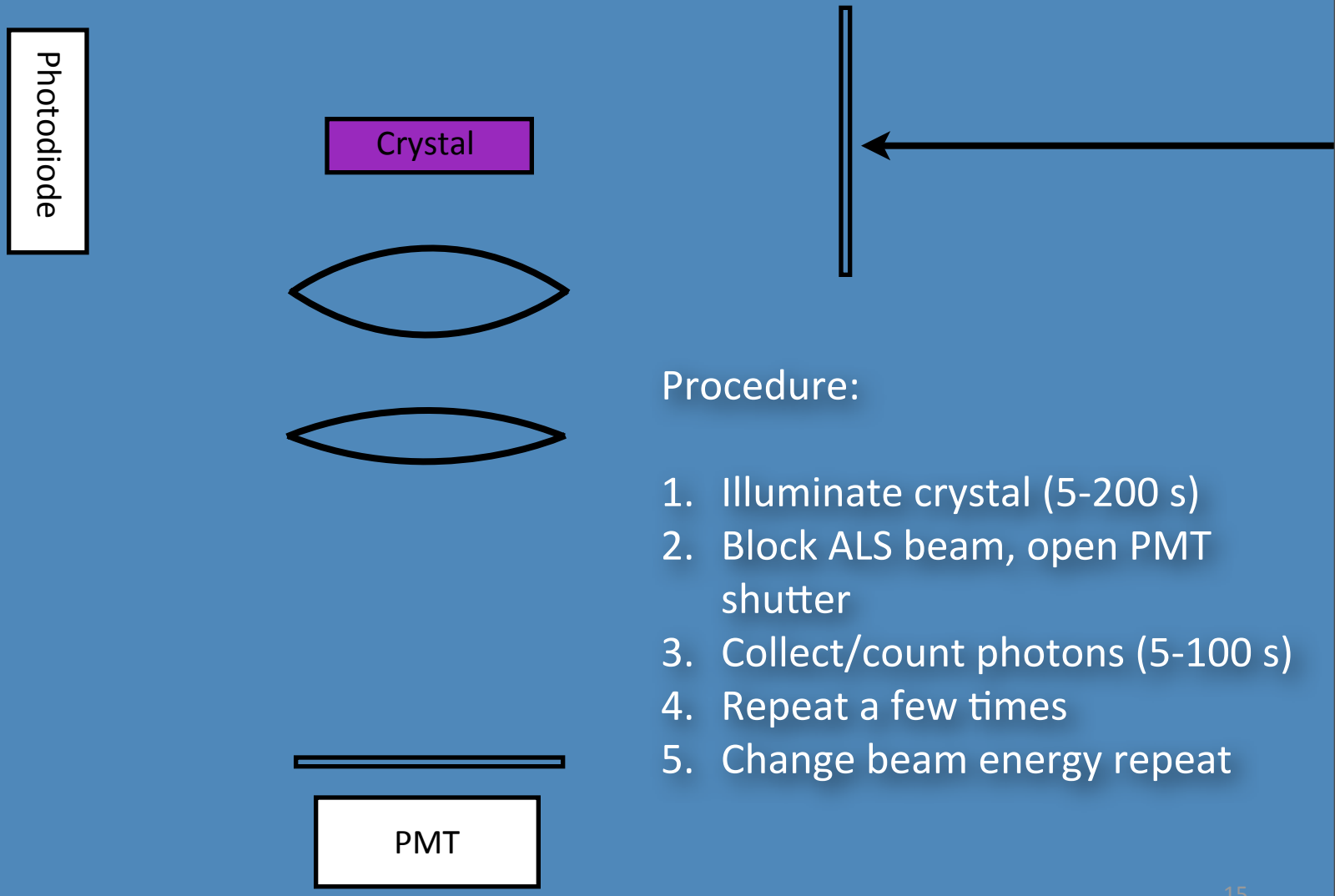
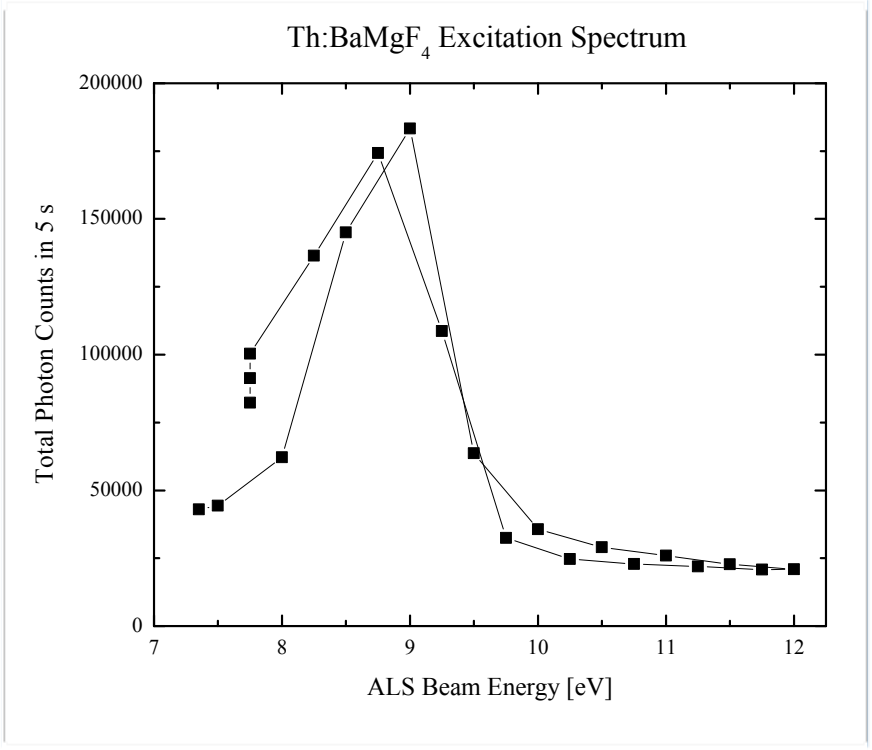
ALS - Crystal Measurement

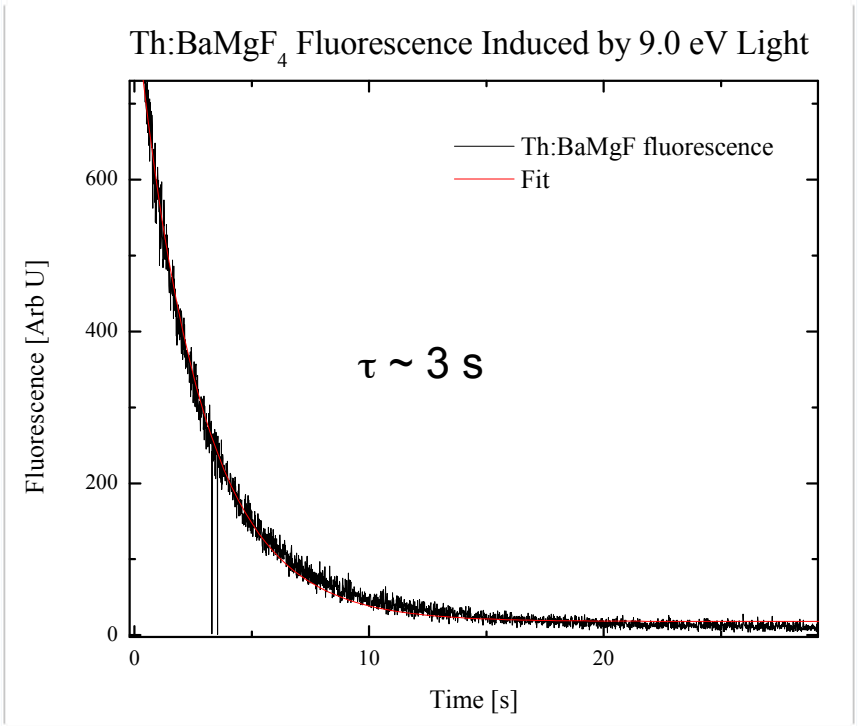


Procedure:

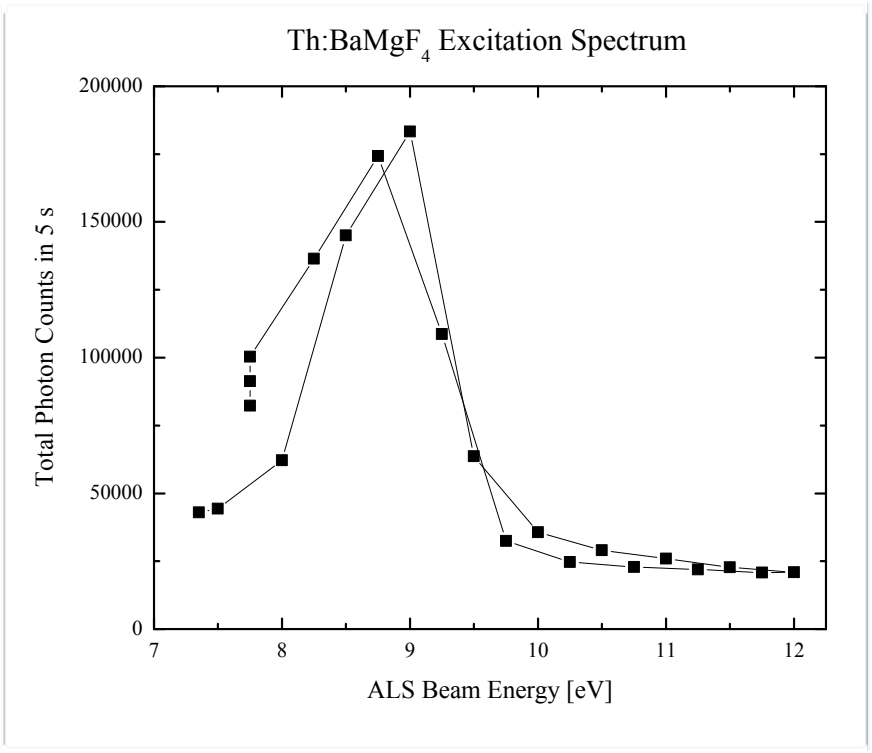
1. Illuminate crystal (5-200 s)
2. Block ALS beam, open PMT shutter
3. Collect/count photons (5-100 s)
4. Repeat a few times
5. Change beam energy repeat

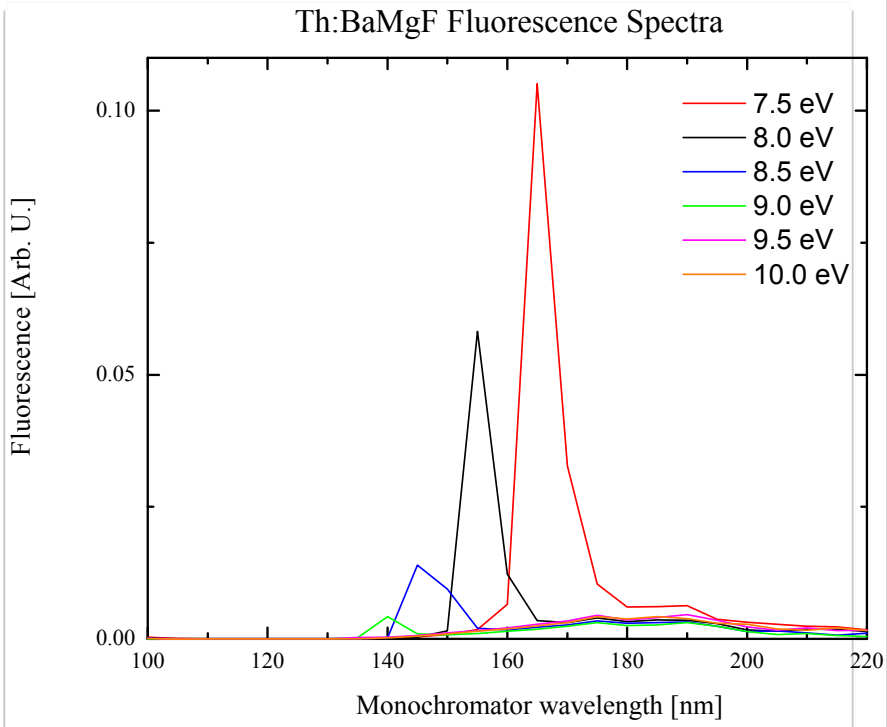
Th:BaMgF$_4$


Th:BaMgF4 Excitation Spectrum
Total Photon Counts in 5 s
ALS Beam Energy [eV]
200000
150000
100000
50000
0
7
8
9
10
11
12

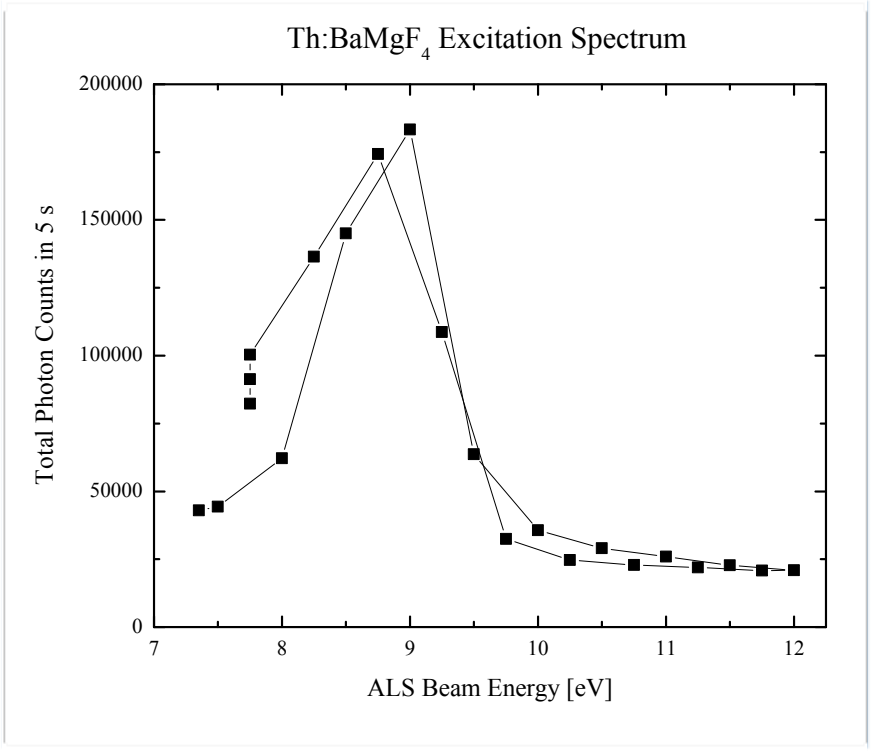

Th:BaMgF4 Fluorescence Induced by 9.0 eV Light
Th:BaMgF fluorescence
Fit
τ ~ 3 s
Fluorescence [Arb U]
Time [s]
600
400
200
0
0
10
20

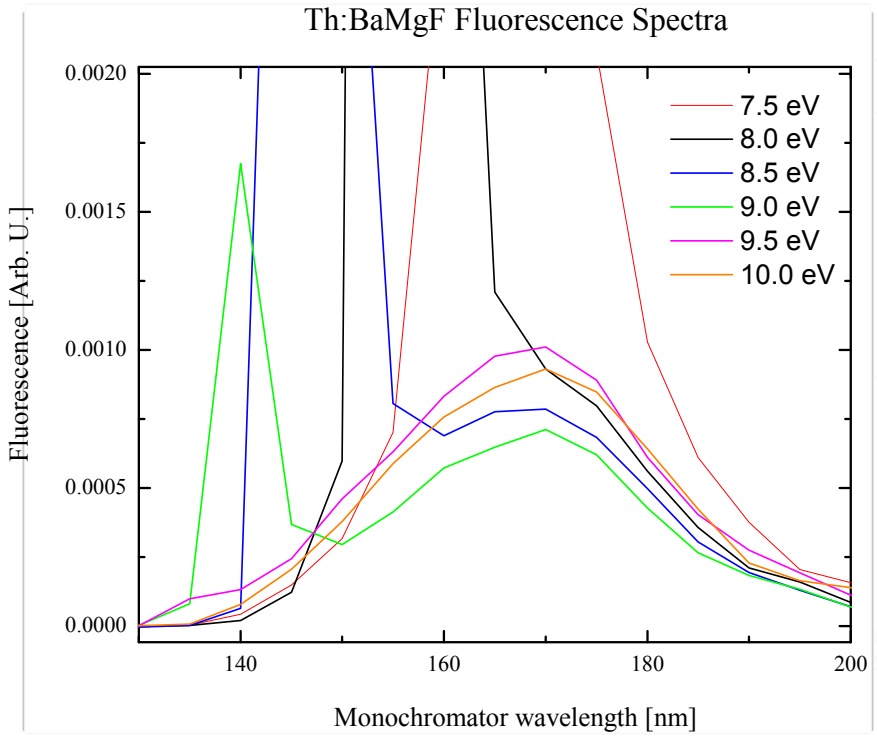
Th:BaMgF$_4$


Th:BaMgF4 Excitation Spectrum
Total Photon Counts in 5 s
ALS Beam Energy [eV]
200000
150000
100000
50000
0
7
8
9
10
11
12

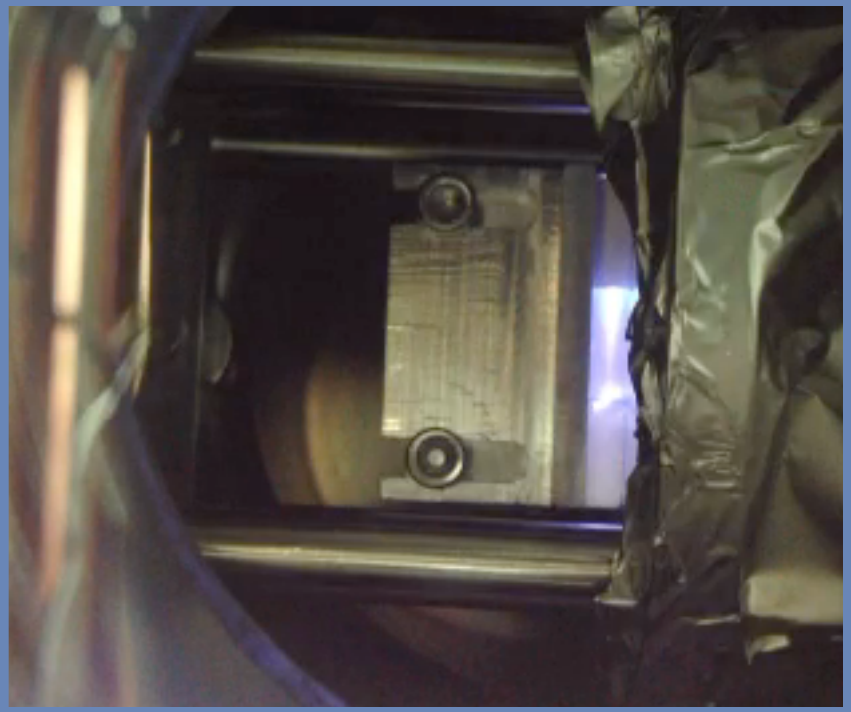

Th:BaMgF Fluorescence Spectra
Fluorescence [Arb. U.]
Monochromator wavelength [nm]
7.5 eV
8.0 eV
8.5 eV
9.0 eV
9.5 eV
10.0 eV
0.10
0.05
0.00
100
120
140
160
180
200
220

Th:BaMgF$_4$

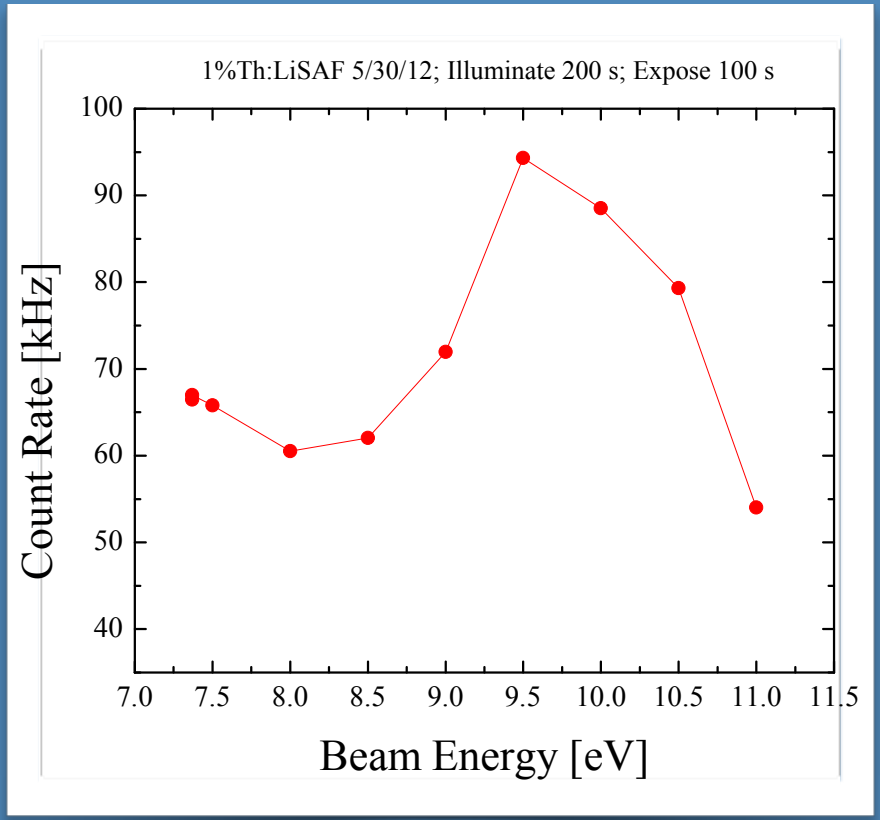

Th:BaMgF4 Excitation Spectrum
Total Photon Counts in 5 s
ALS Beam Energy [eV]
200000
150000
100000
50000
0
7
8
9
10
11
12


Th:BaMgF Fluorescence Spectra
Fluorescence [Arb. U.]
Monochromator wavelength [nm]
0.0020
0.0015
0.0010
0.0005
0.0000
140
160
180
200
7.5 eV
8.0 eV
8.5 eV
9.0 eV
9.5 eV
10.0 eV

Th:$BaMgF_4$



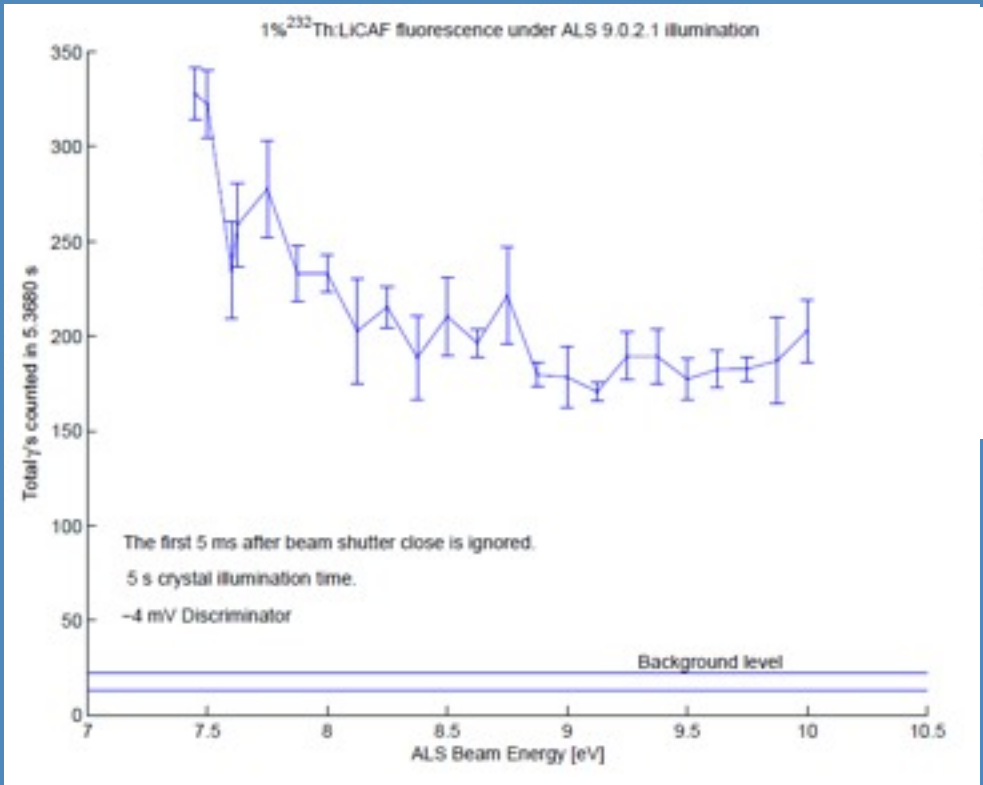
Th:LiSAF

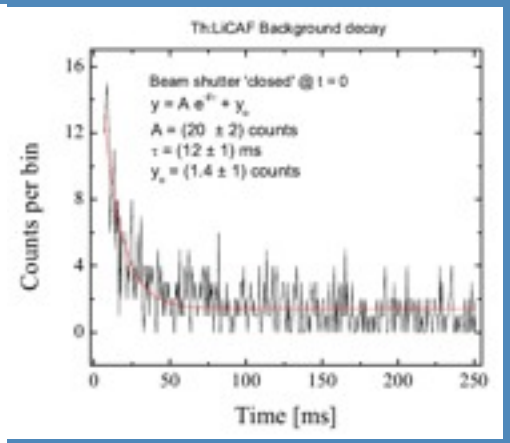


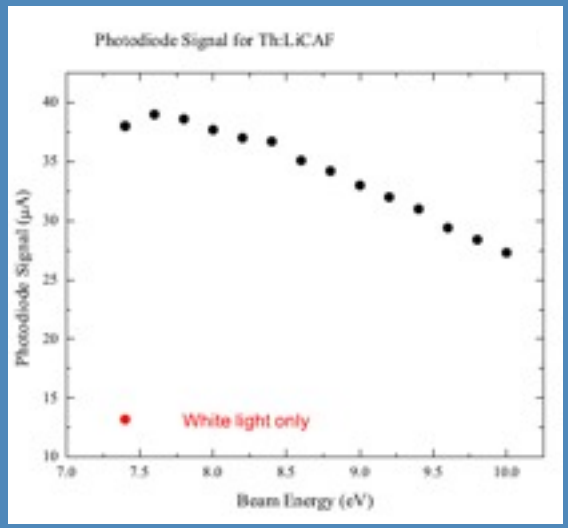
- 200 s Illumination, 100 s photon collection (50 ms delay)
- Maximum fluorescence rate detected ~100 kHz
- Fluorescence lifetime at 9.5 eV measured to be 500 ms

TH:LISAF

Th:LiCAF

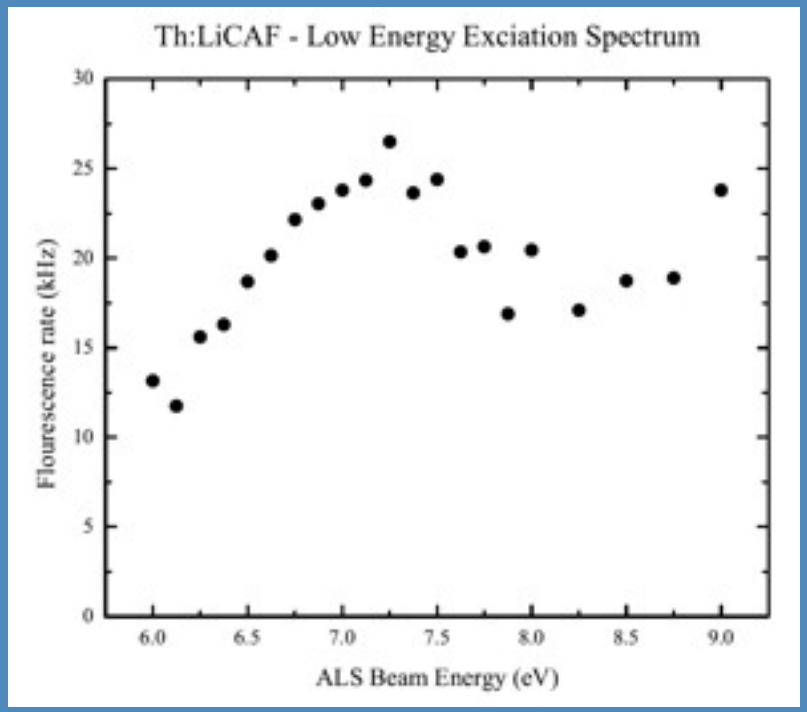




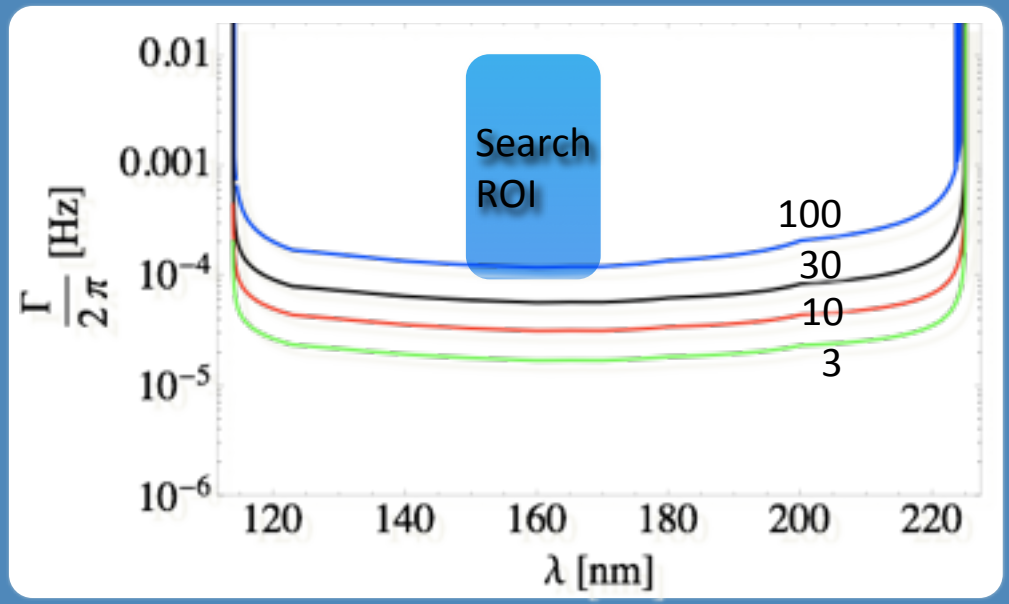


- Target doping 1% molar
  5 s Illumination, 5 s photon collection (5 ms delay)
- Highest value (at 7.4 eV) corresponds to 350 kHz, 12 ms lifetime

Mock Search for Isomer

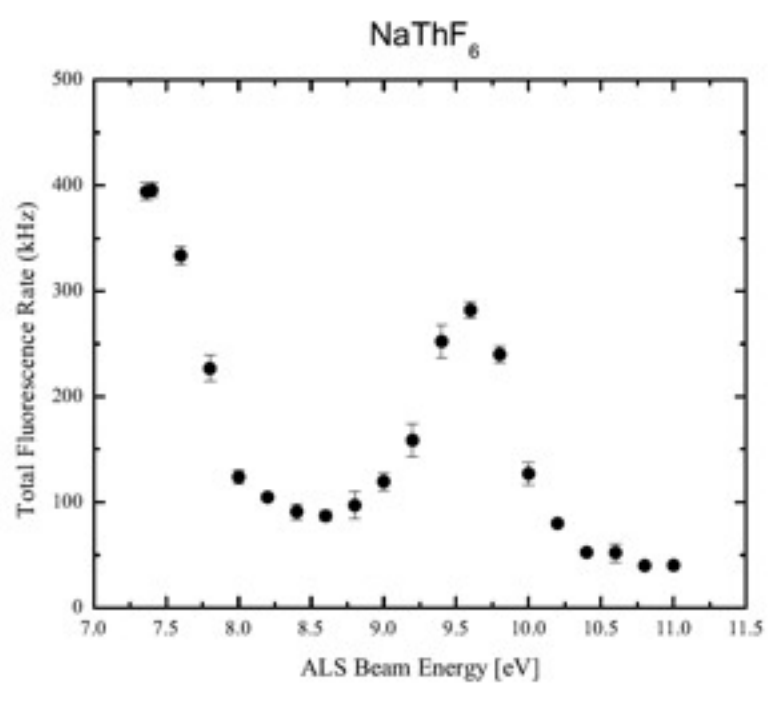


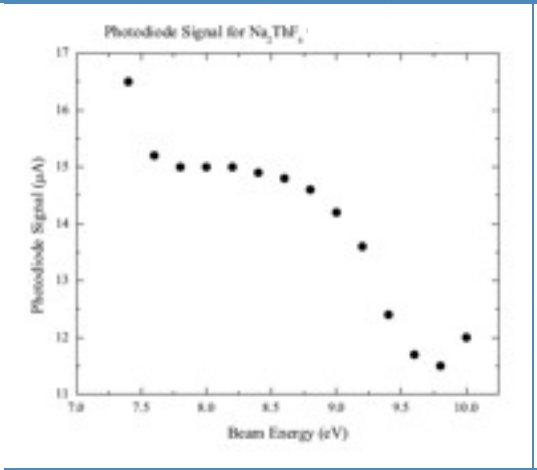
ALS Search Signal to Noise taking all data for Th:LiCAF into account

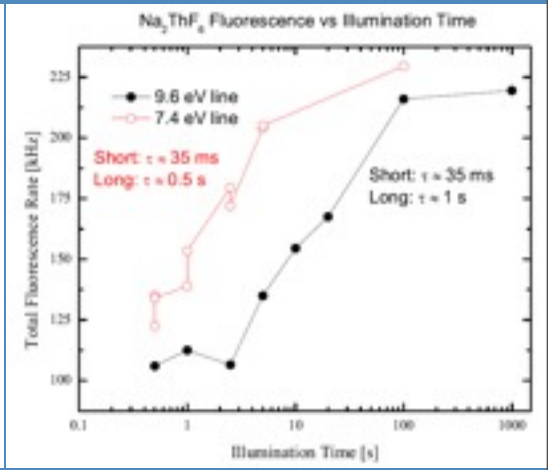


- 200 s Illumination, 100 s collection (5 ms delay)
- Largest value corresponds to ~25 kHz

Na_2ThF_6



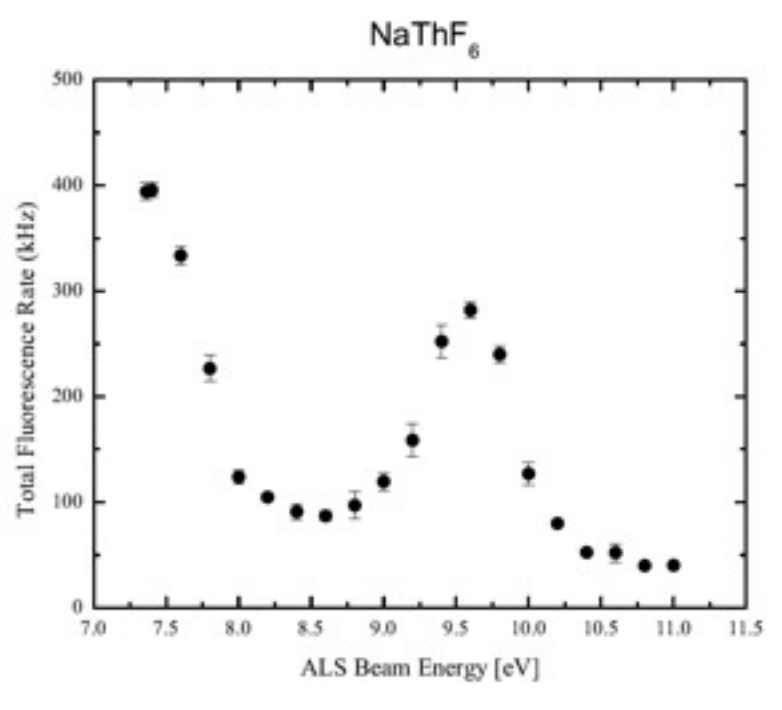


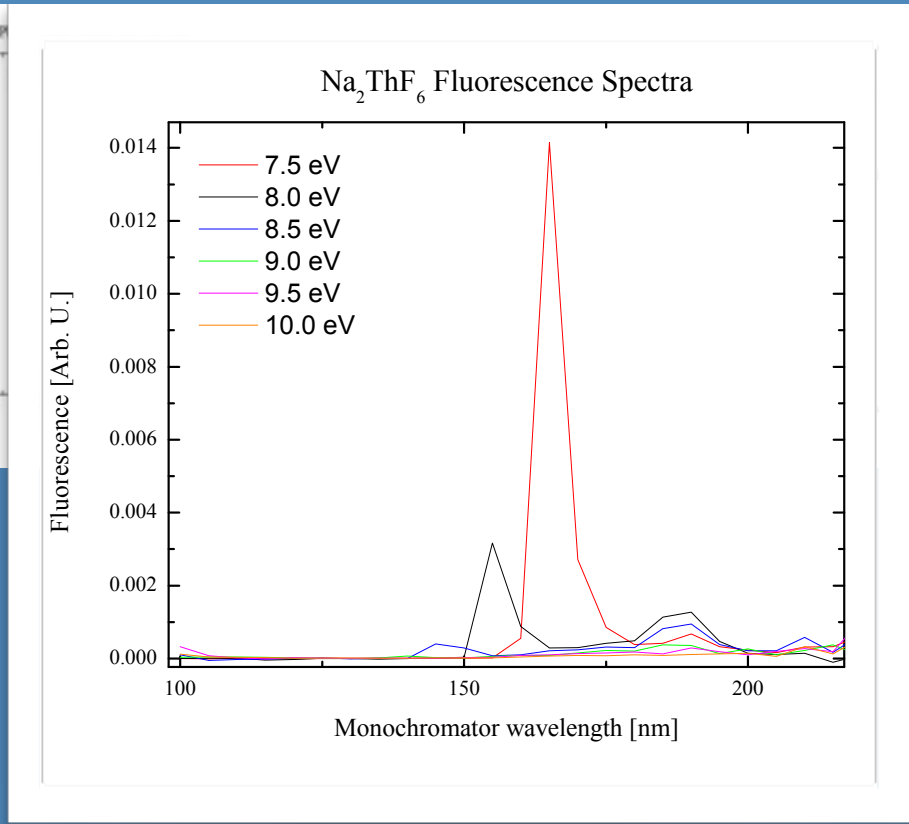


Peak	τ_1	τ_2
7.4 eV	~35 ms	~0.5 s
9.6 eV	~35 ms	~1 s

- 5 s Illumination, 5 s photon collection
- Maximum fluorescence rate ~400 kHz, but mostly with ~35 ms lifetime
- Showed dark spot/radiation damage due to VUV beam

Na_2ThF_6





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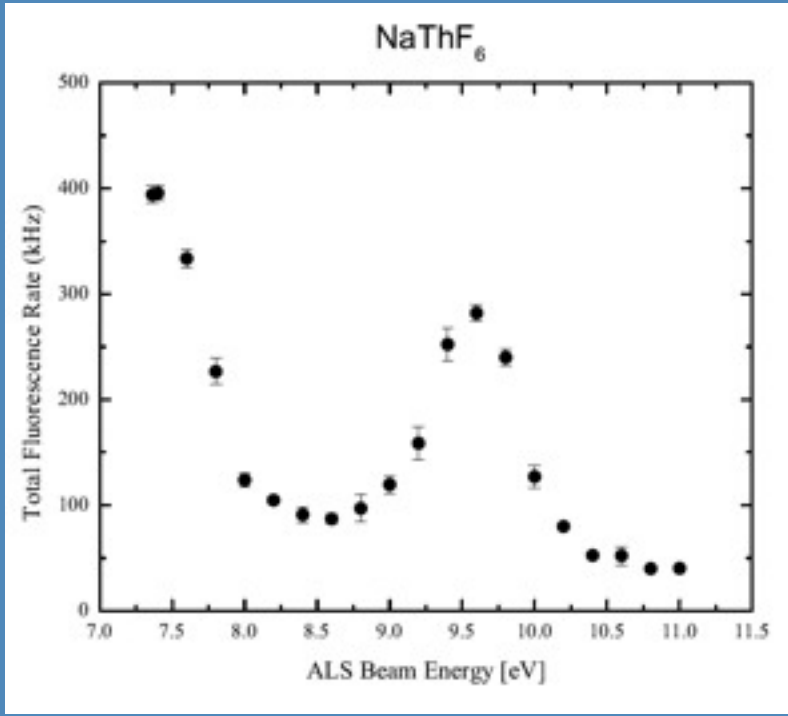
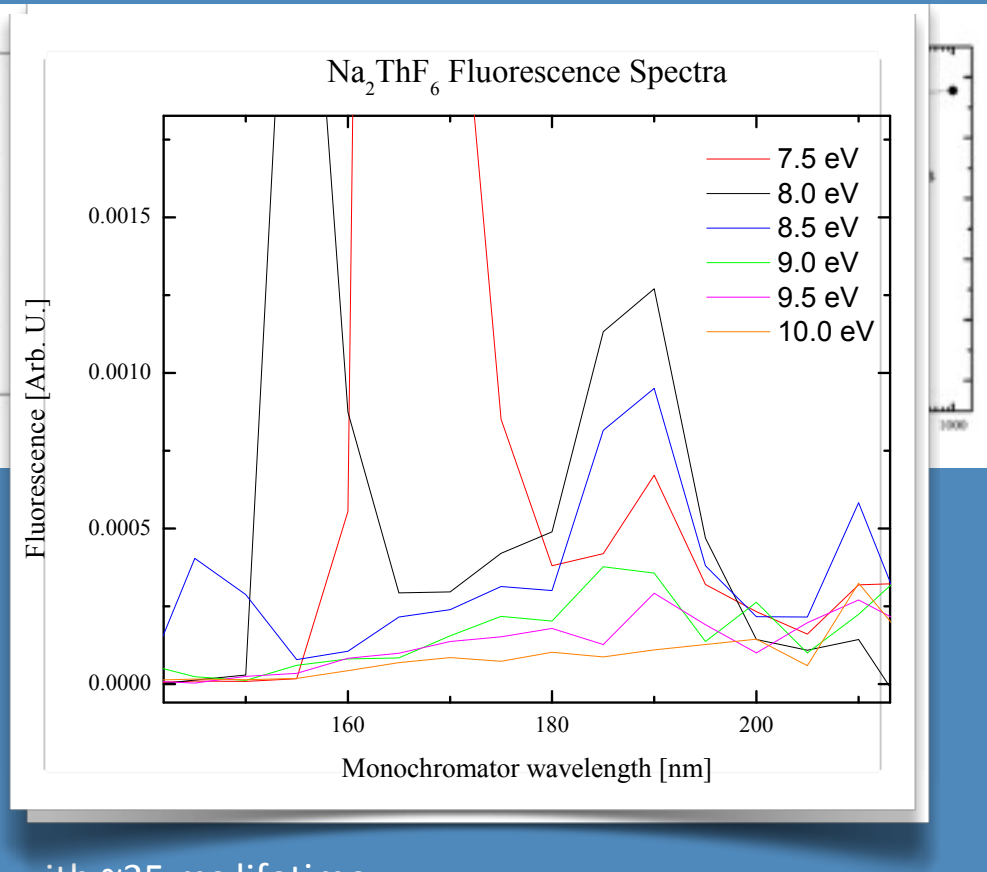
Na_2ThF_6





- 5 s Illumination, 5 s photon collection
- Maximum fluorescence rate ~400 kHz, but mostly with ~35 ms lifetime
- Showed dark spot/radiation damage due to VUV beam

Summary

- We've characterized 6 potential host crystals.
  - Doping looks very promising
  - All six appear to be okay in terms of VUV induced fluorescence (short-lived and broadband), but Na_2ThF_6 and Th:NaYF show signs of radiation damage (possibly impurities)

- Th:LiSAF and Th:LiCAF exhibited lowest fluorescence levels
  - BG Fluorescence rates of ~10-100 kHz, but short lived (<500 ms)
  - Compare to expected rates of 15 kHz with lifetime of 10,000 s or 1 GHz with lifetime of 100 s!