Response of parylene-coated NaI(Tl) scintillators at low temperature

M. Martínez, Fundacion ARAID, Univ. de Zaragoza – RPScint2013, 17 – 20 September 2013, Kyiv, Ukraine

Outline

1. NaI & NaI(Tl) scintillators revised
2. Parylene coating
3. Low temperature X-ray scintillation measurements
   – Light output vs T
   – Spectral response
4. Thermoluminescence
5. Response after thermal cycles
6. Summary

"Study of parylene-coated NaI(Tl) at low temperatures for bolometric applications", N. Coron et al., Astrop. Phys. 47 (2013) 31

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NaI & NaI(Tl) scintillators

- Well known at room and low temperature
  - NaI(Tl): 420 nm
  - NaI pure: 300 nm, better than NaI(Tl) at cold
  - Both increase light when lowering T . Maximum around 60 K (NaI) , 150-250 K (NaI(Tl)), then strong decrease
- Never tested at very low (mK) temperature

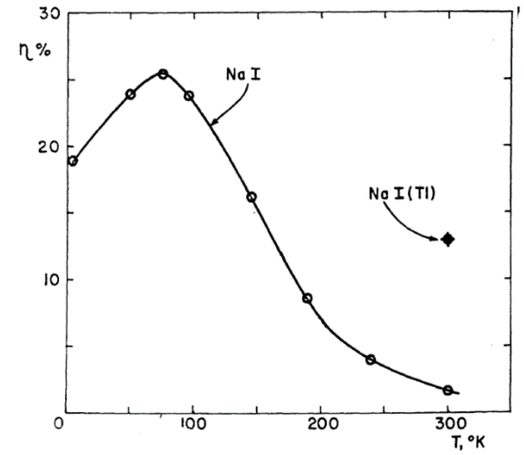


Fig. 3—Energy conversion efficiency, η, of unactivated NaI as function of temperature.

"Fundamental Studies of Scintillation Phenomena in NaI" W.J. Van Sciver and L. Bogart, IRE Trans. Nucl. Sci. 5 (1958) 90

NaI(Tl)

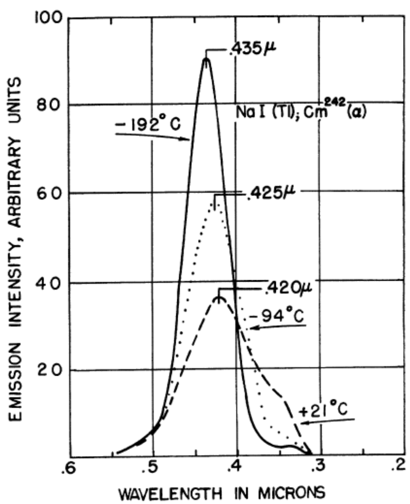


Fig. 5—Emission spectra of NaI(Tl) excited by Cm^{242} alpha particles.

NaI pure

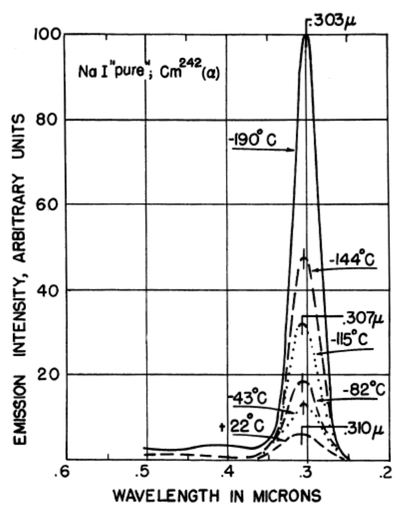


Fig. 9—Emission spectra of NaI ("pure") excited by Cm^{242} alpha particles.

"Alkali Halide Scintillators" W.J. Van Sciver, IRE Trans. Nucl. Sci. 3 (1956) 39.

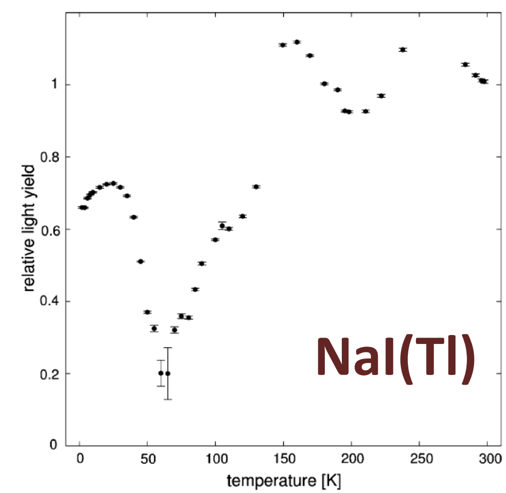


Fig. 4 Light yield of NaI(Tl) as function of temperature relative to the value at room temperature. Errors are dominated by systematics, see text for details

"Low temperature light yield measurements in NaI and NaI(Tl)" C. Sailer et al., Eur. Phys. J. C. 72 (2012) 2061

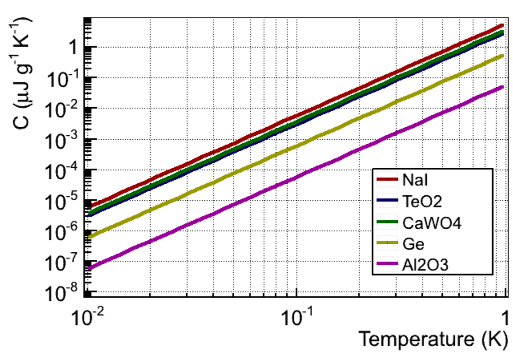
NaI/NaI(Tl) scintillators: pros and cons

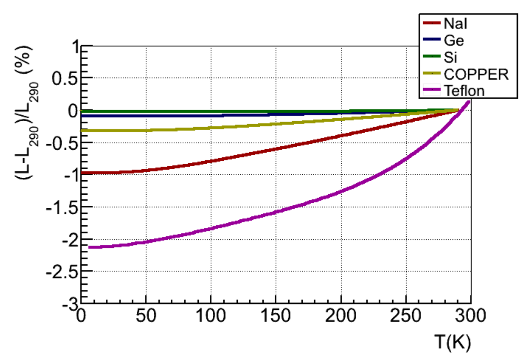
Pros

- ☺ Low cost and well-known technology (most widely used scintillator)
- ☺ High light yield
- ☺ NaI(Tl): $\lambda_{max} \approx 420$ nm, maximum efficiency region of bialkali PMTs
- ☺ Very radiopure crystals achievable by powder selection/purification
- ☺ Possibility to grow large mass crystals
- ☺ Particle discrimination by pulse shape analysis at high energy
- ☺ For Dark Matter applications:
  - 100% sensitive to SD-proton interaction
  - Sensitive to Light and heavy WIMPS
  - Target of DAMA/LIBRA

Cons

- ☹ **High hygroscopicity**
- ☹ For Dark Matter applications:
  - Low quenching factor NR/βϒ (Na ≈ 0.3, I ≈ 0.1)
  - No particle discrimination at low energy
- ☹ For bolometric applications:
  - Relatively high specific heat (θ_{debye}=164 K)
  - Large coefficient of thermal expansion (1% between 300 K and 4 K)





NaI coating

The higroscopicity complicates the crystal handling, specially for low T applications

NaI(Tl)

NaI pure

One solution could be to **coat the NaI crystal with an appropriate material acting as humidity barrier**

Look for coating materials:

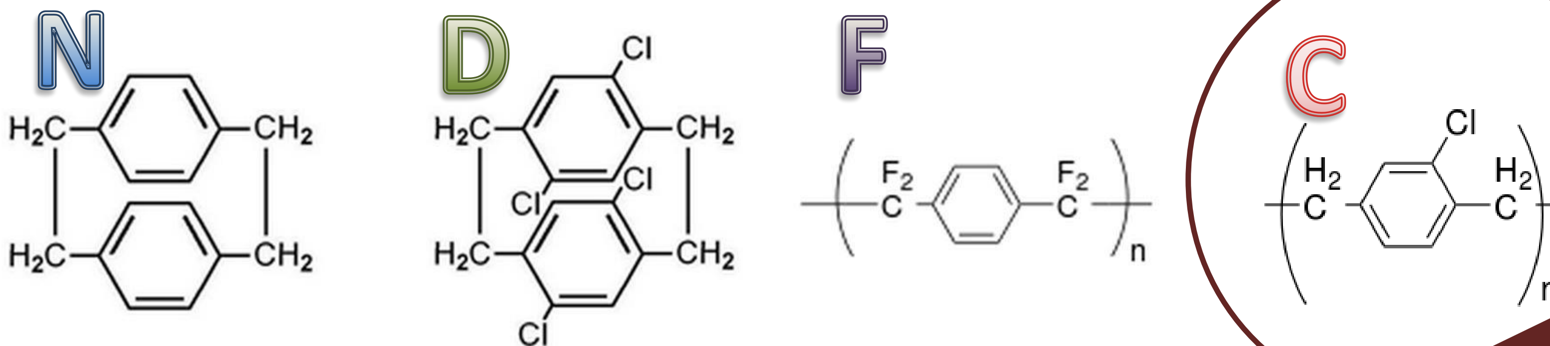
- Transparent in the wavelength of NaI/NaI(Tl) emission
- Radiopure
- Resistant to thermal cycles
- Low heat capacity (→ very thin films!)
- …

(Resistant to thermal cycles, low heat capacity, …: For low T applications)

A possibility: PARYLENE

Parylene

Polymer family based on poly-p-xylylene, commonly used in electronic and space industries as moisture/dielectric barrier



Good humidity barrier!

BARRIERE PROPERTIES*

PROPERTIES	ASTM METHOD	UNITS	PARYLENE C	N	D	F
Water Absorption (after 24h)						
	D570	%	< 0,1	< 0,1	< 0,1	
Water Vapour Transmission at 38°C						
	F1249-06	g.mm/(m^{-2}.j)	0,10	0,75	0,12	0,32
Gas Permeability at 25°C						
- N_2	D1434 F3985-05	cm^3.mm/(m^2.j.bar)	0,40	3,00	1,70	
- O_2	D1434 F3985-05	cm^3.mm/(m^2.j.bar)	2,90	15,20	12,10	85,00
- CO_2	D1434 F3985-05	cm^3.mm/(m^2.j.bar)	3,10	86,00	5,10	
- H_2	D1434 F3985-05	cm^3.mm/(m^2.j.bar)	420	220	90	

COMELEC S.A. Technical tables
http://www.comelec.ch/

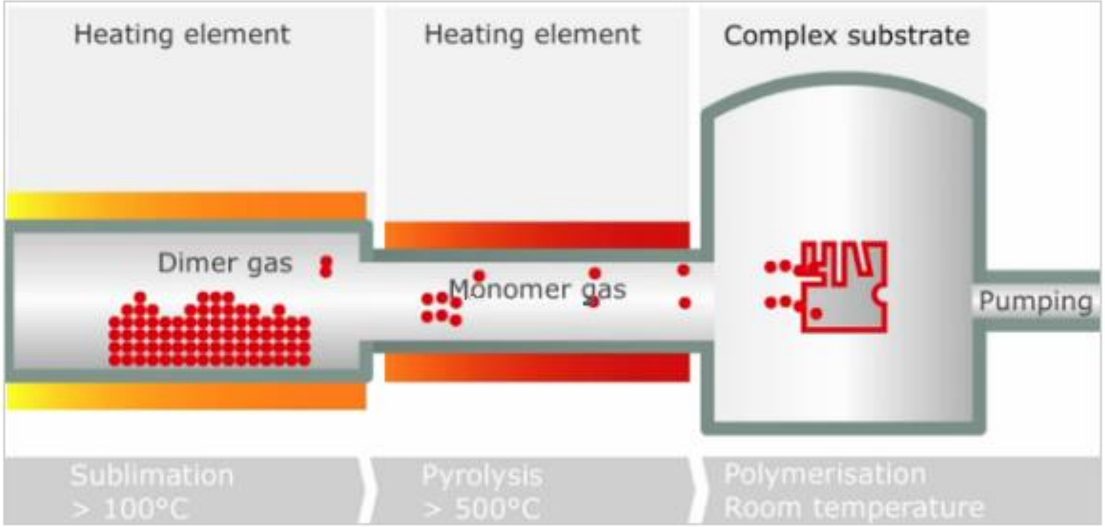
Parylene conformal coating

Parylene is deposited in very thin films by vapor-phase condensation polymerization

CONFORMAL COATING

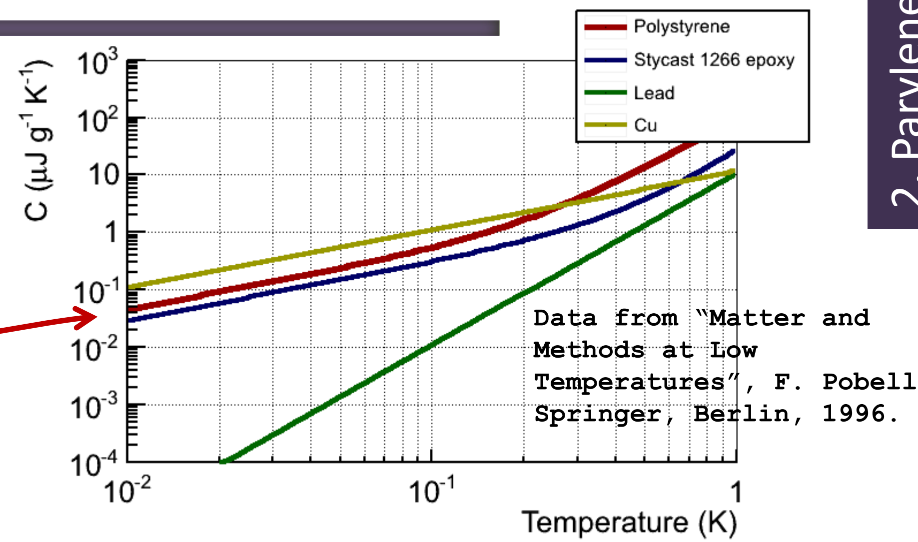
Monomers are adsorbed and simultaneously polymerizing on all the exposed surfaces.

- Thin layers (down to 0.1 µm)
- Pinhole-free
- Room temperature (avoiding thermal stresses on the sample)
- No solvents



Parylene heat capacity

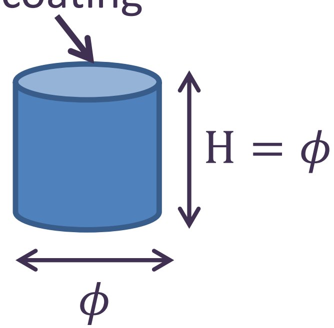
No data available at very low T, but it is expected to be similar to other aromatic polymers as polystyrene

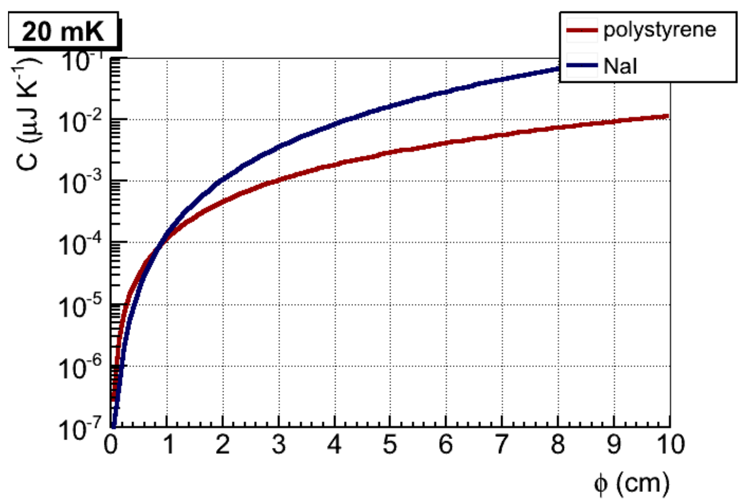


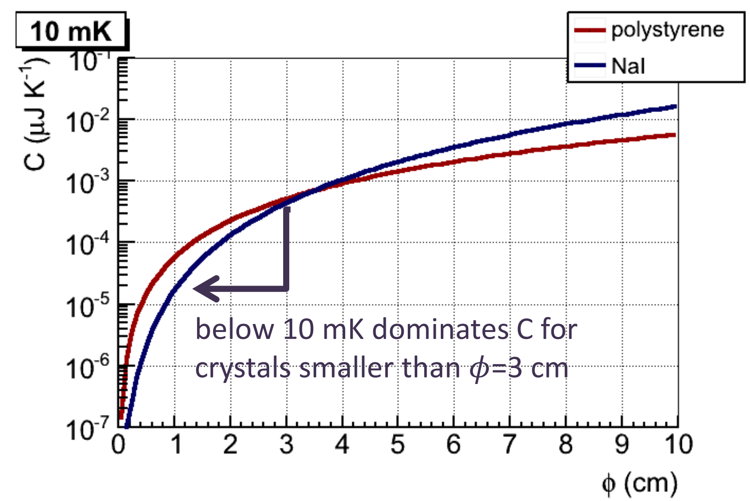
⇒Thin films!

(But at least several µm are needed to assure day-scale handling under normal RH conditions)

2 µm parylene coating







Parylene radiopurity

HPGe measurement at LSC on dimer (dichloro-p-cylophane) samples

Activity (mBq/kg)					
^{232}Th	^{226}Ra	^{238}U	^{40}K	^{60}Co	^{137}Cs
26 ± 8	<12	<370	<100	<4.5	<6.2

UPPER LIMITS AT 90% C.L.

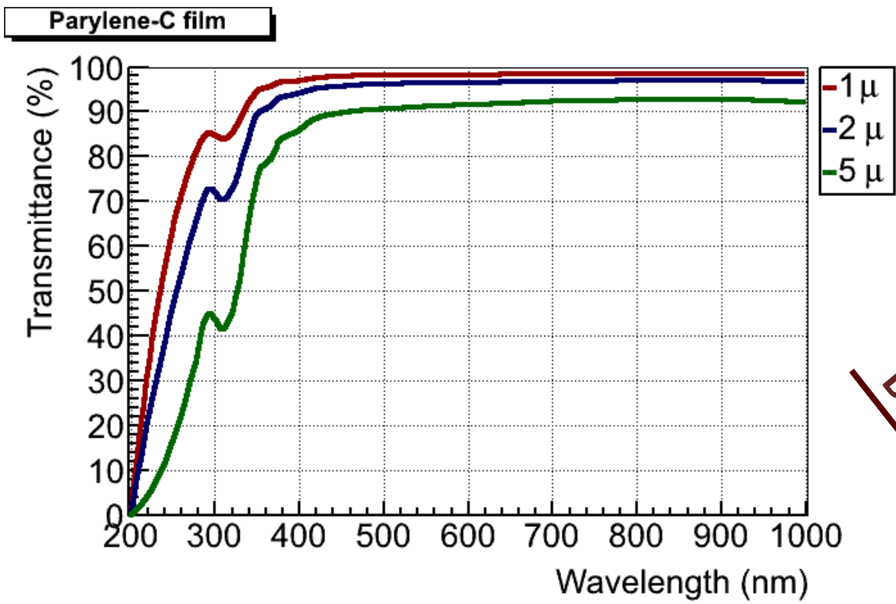
(From the measurements we cannot rule out the presence of out-of-equilibrium ^{210}Pb)

But cleaner parylene films are available

see for example: Loach, "Electronics and Cables for the MAJORANA demonstrator" , Talk at the 2010 Topical Workshop in Low Radioactivity Techniques (LRT2010)

→ less than 0.2 mBq/kg ^{232}Th

Parylene transmission in NaI/NaI(Tl) emission bands



Data adapted from "UV-visible and infrared characterization of poly(p-xylylene) films for waveguide applications and OLED encapsulation", Jeong et al., Synthetic Metals 127 (2002) 189

In order to keep an acceptable transmission in the UV region, films no thicker than 5 µm should be used.

Better match

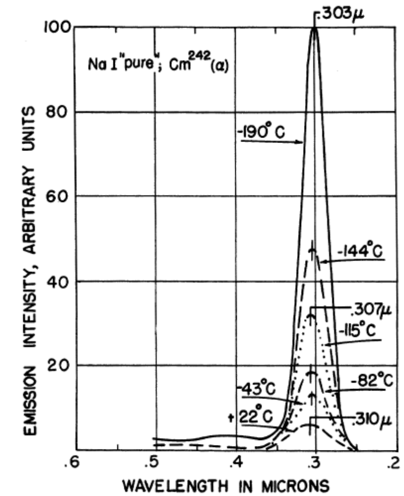
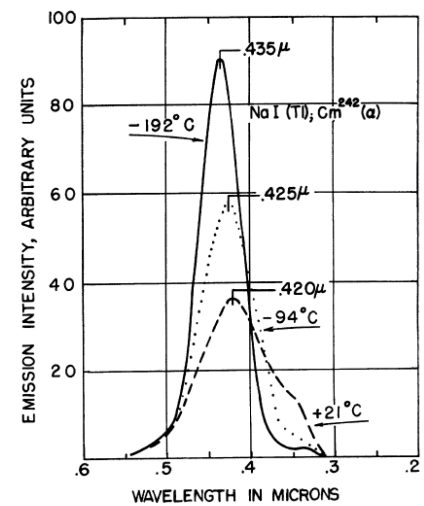


Fig. 9—Emission spectra of NaI ("pure") excited by Cm^{242} alpha

NaI pure



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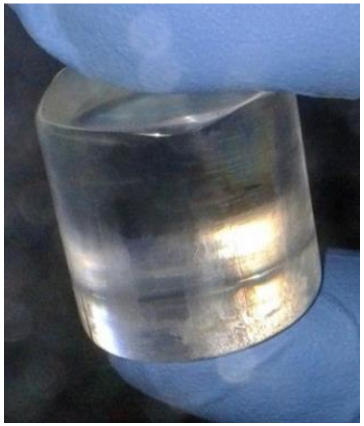
Na(Tl)

Sepctra from "Alkali Halide Scintillators" W.J. Van Sciver, IRE Trans. Nucl. Sci. 3 (1956) 39.

Parylene-coated NaI/NaI(Tl) samples



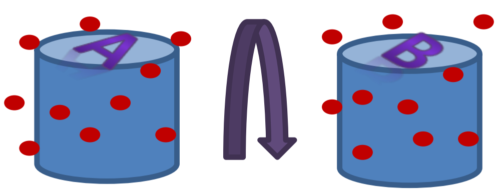
NaI(Tl) crystals provided by **Detect-Europe**
NaI crystals provided by **Hilger**

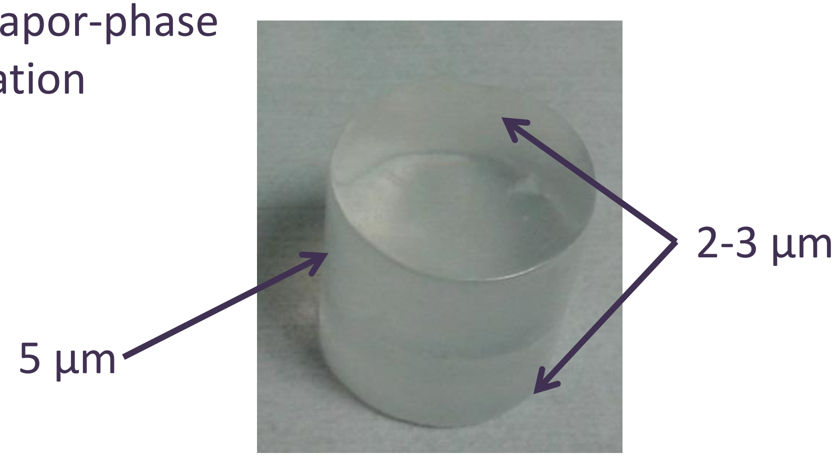
- cylindrical shape, H=ϕ=25 mm
- 45 g weight
- optically polished surfaces

comelec
PARYLENE COATING

Parylene deposition by vapor-phase condensation polymerization

Two consecutive depositions turning the crystals over in between





Low temperature X-ray scintillation measurements

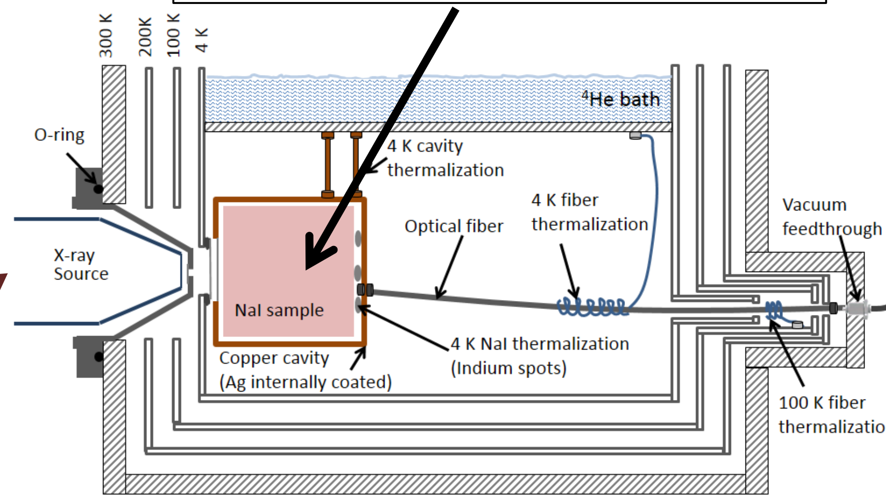
^{4}He cryostat

Thermometers
Pt100 (25-300 K)
RuO_2 (<25 K)

Optical fiber

Optical fiber

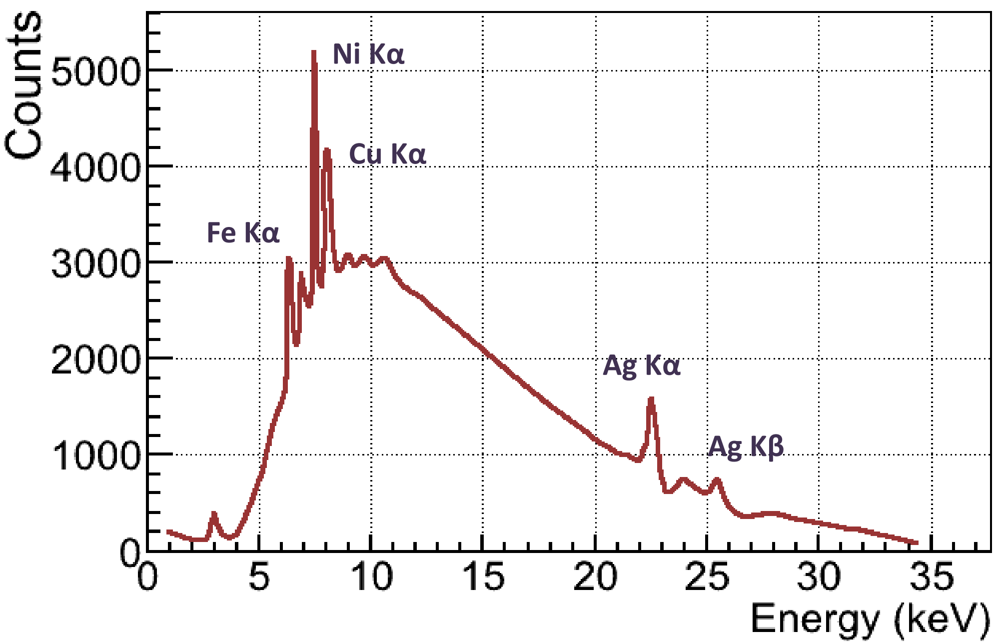
Sample inside Ag-coated cavity

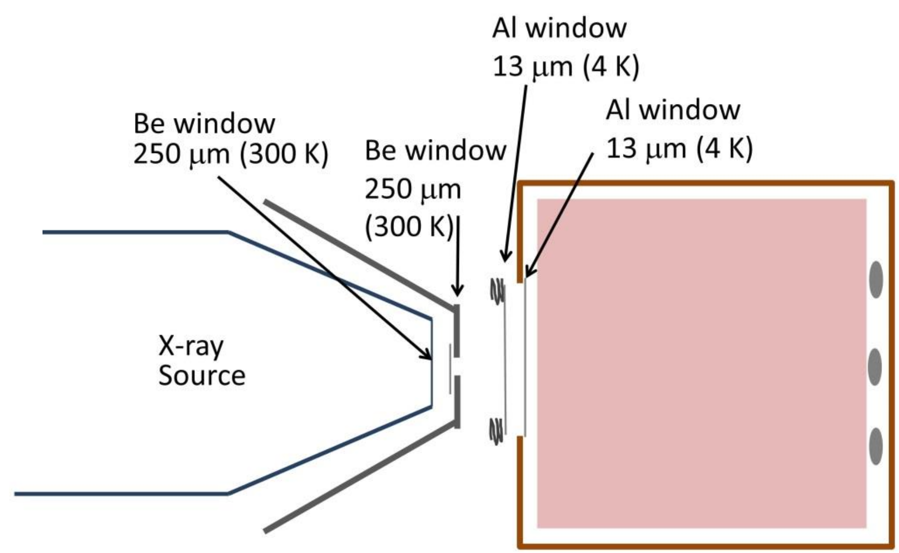


X-ray source (40 kV)

X-ray source

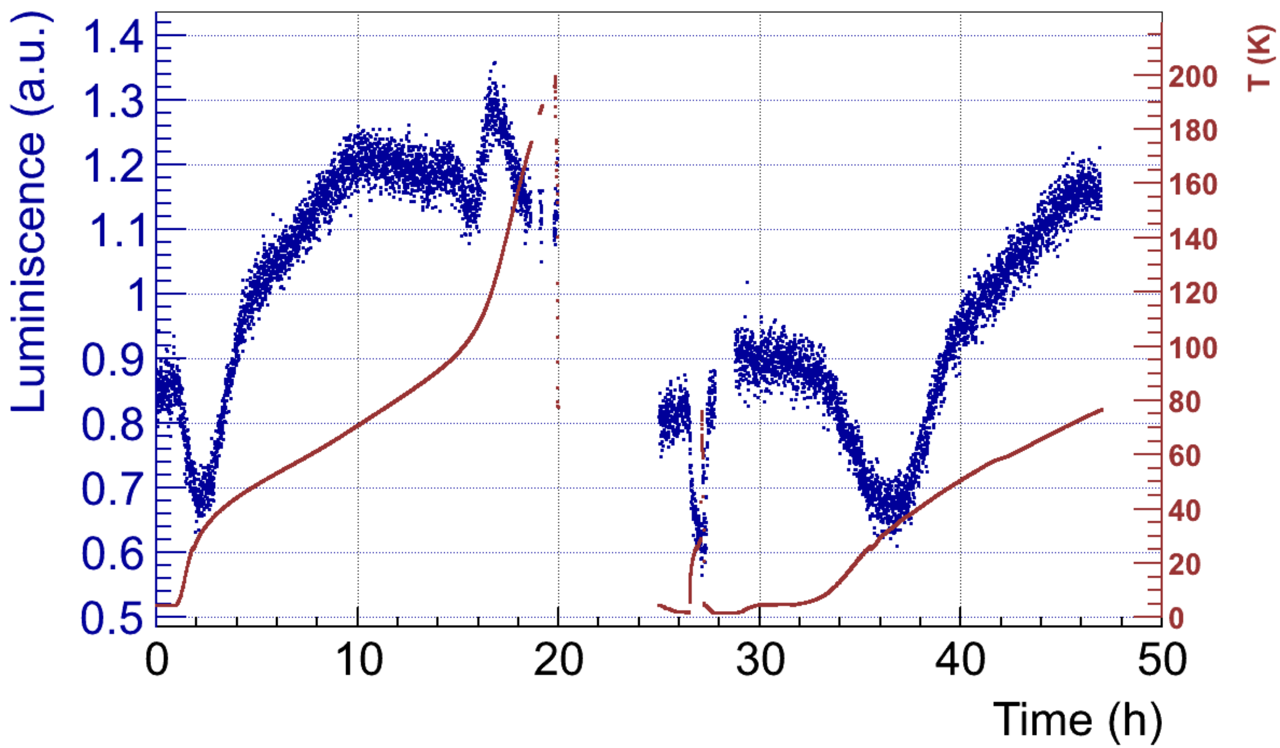
BULLET 40 kV X-ray source MOXTEK (silver anode)



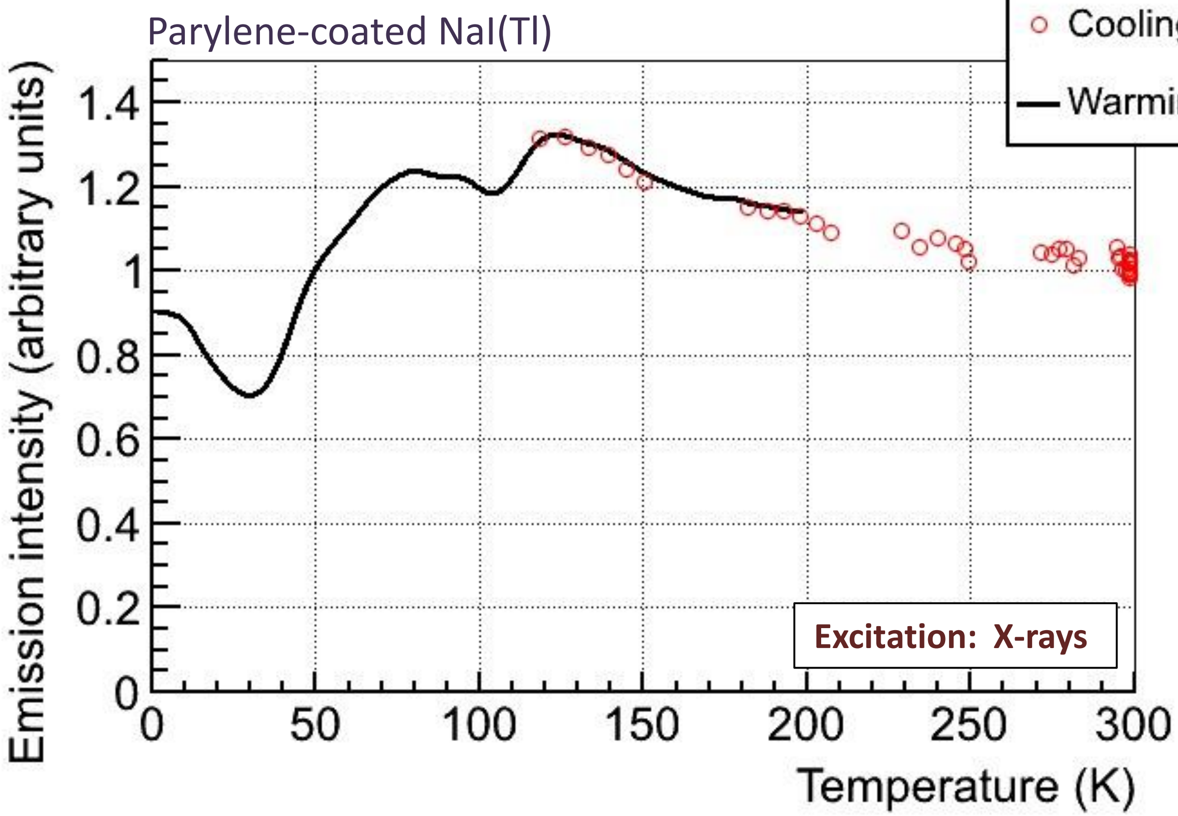


Light output as a function of temperature

- Detector: **Si photodiode** HAMAMATSU S1336-18BQ (detection range**: 190–1100 nm**)
- Fast cooling from 300 to 77 K (~20 min) and from 77 K to 4 K(~6 min)
- Measurement during two warming cycles (from 4 to 200 K and from 1.5 to 77 K):



Light output as a function of temperature



- Maximum at around 125 K
- Noticeable light decrease below 70 K
- Further increment of light below 30 K (light output @ 1.5 K= 90% light @ 300 K)

General behaviour similar to that found by Sailer et al. although relative intensities and temperatures of the maxima and minima of light emission differ (but different excitation…)

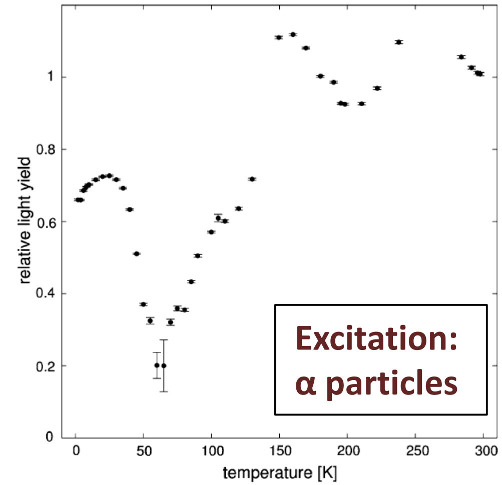


Fig. 4 Light yield of NaI(Tl) as function of temperature relative to the value at room temperature. Errors are dominated by systematics, see text for details

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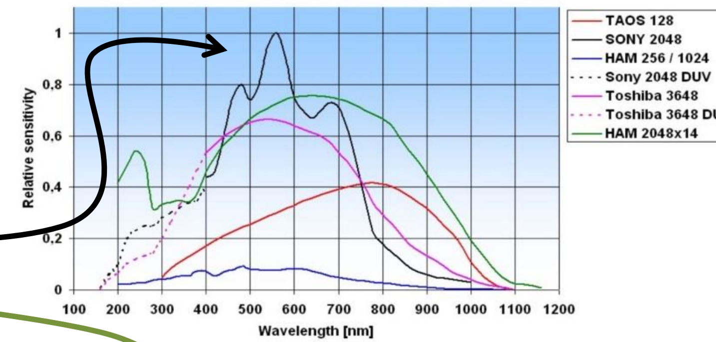
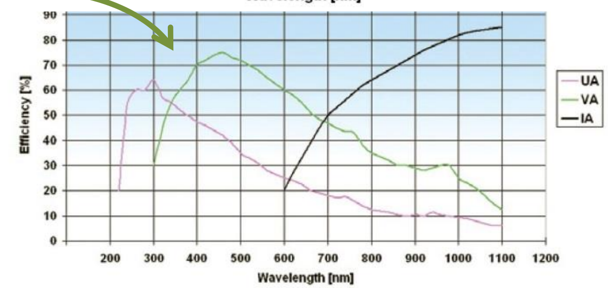
Spectral response: detector & fiber corrections

Avantes fiber spectrometer (range: 250-1100 nm)

- detector Sony 2048-DUV
- diffraction grating VA, 300 lines/mm

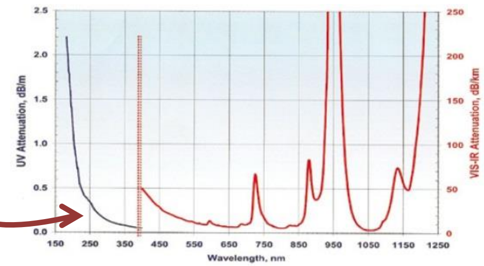




Plots taken from the Avantes manual

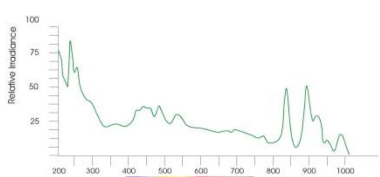
Avantes optical fiber

- 1 mm ϕ FC-UV 1000

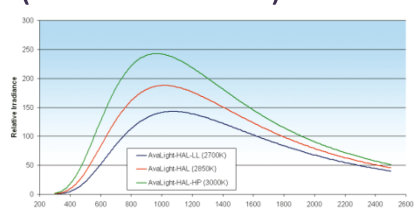


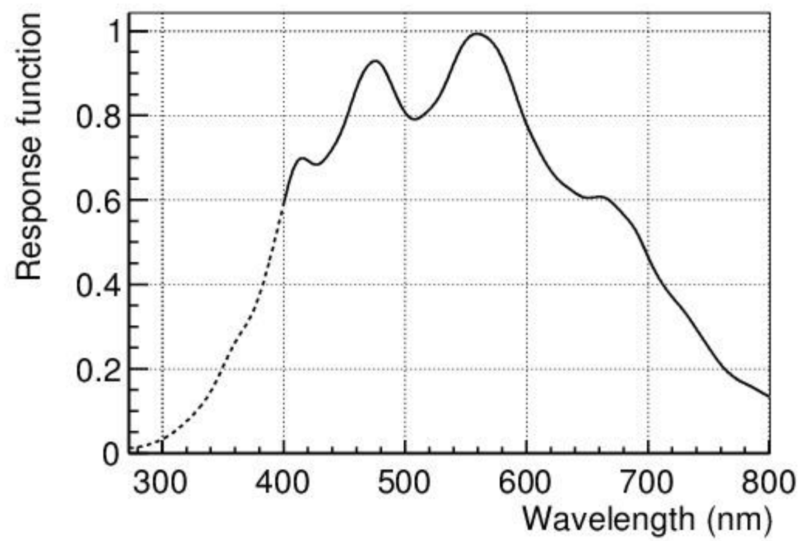
The transmission efficiency vs λ (detector + diffraction grating + optical fiber) was measured experimentally by comparing the known spectra of two light sources with the measured one

- Avalight-XE UV/Vis (250–400 nm)

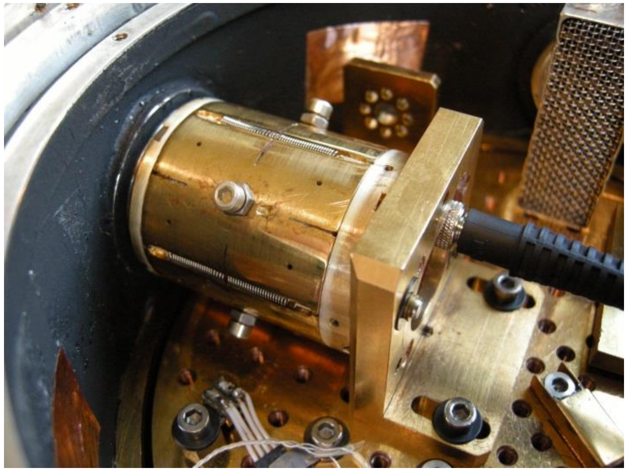


- Melles Griot 13 HLS 101 (400–1100 nm)

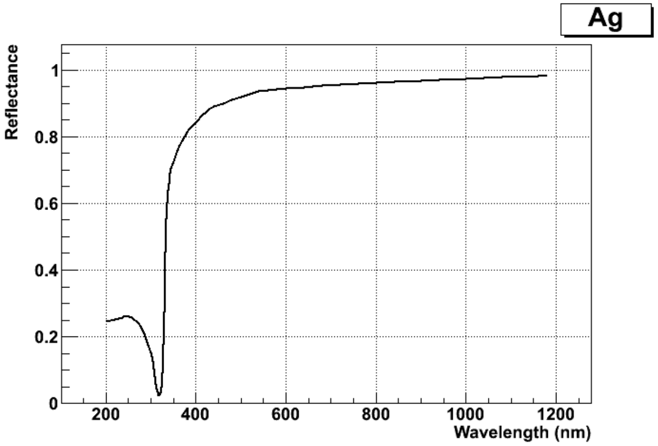




Spectral response: Cavity correction

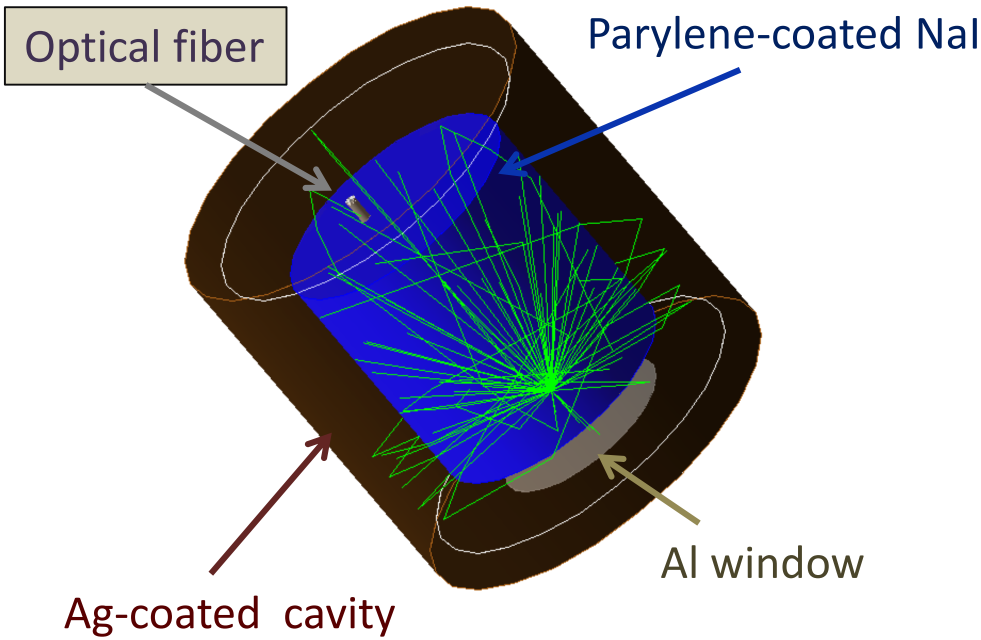


Ag internally-coated cavity



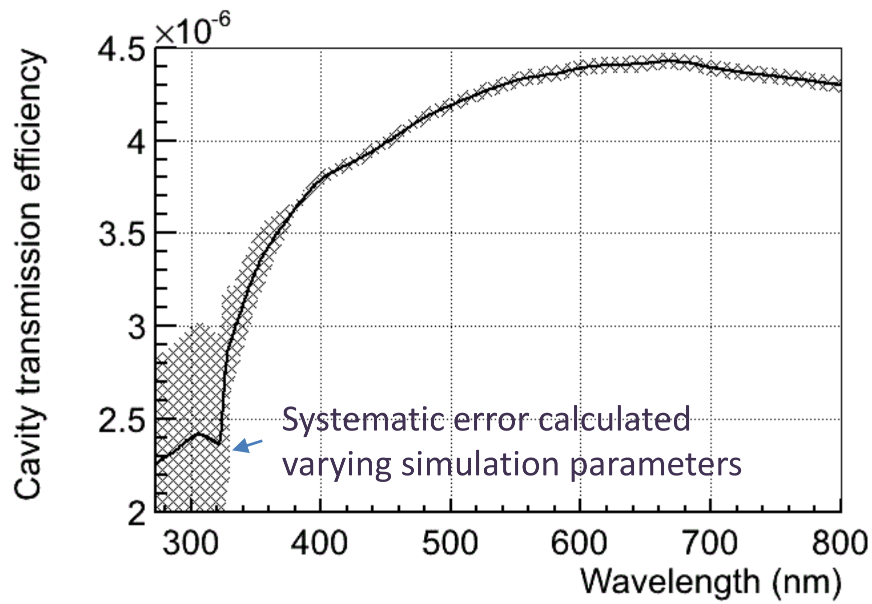
Cavity transmission efficiency depends on wavelength

Cavity correction. MC simulation

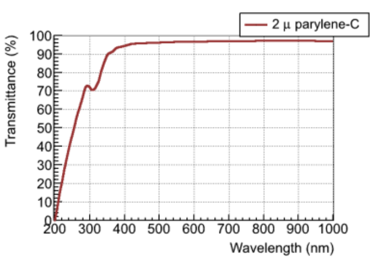


Geat4 MC simulation:

Launch opticalPhotons (white spectrum) and “count” those arriving to the fiber with correct angle as a function of λ

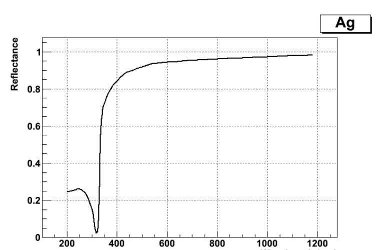


Parylene transmission:



Jeong et al., Synthetic Metals 127 (2002) 189

Ag reflectivity:



M. Bass, Handbook of Optics, second ed., 1995.

Cavity correction. Comparison with calculus

Approximate the cavity transmission efficiency ($\eta_c(\lambda)$) by:

"Energy partition in Sapphire and BGO scintillating bolometers", Y. Ortigoza et al., Astrop. Phys, 34 (2011) 603

$$\eta_c(\lambda) = \frac{I_{out}(\lambda)}{I_{in}(\lambda)} = x + R(\lambda)(1-x)\varphi \sum_{n=0}^{\infty} R^n (1-\varphi)^n = x + \frac{R\varphi}{1-R(1-\varphi)}(1-x)$$

Direct light fraction (x)

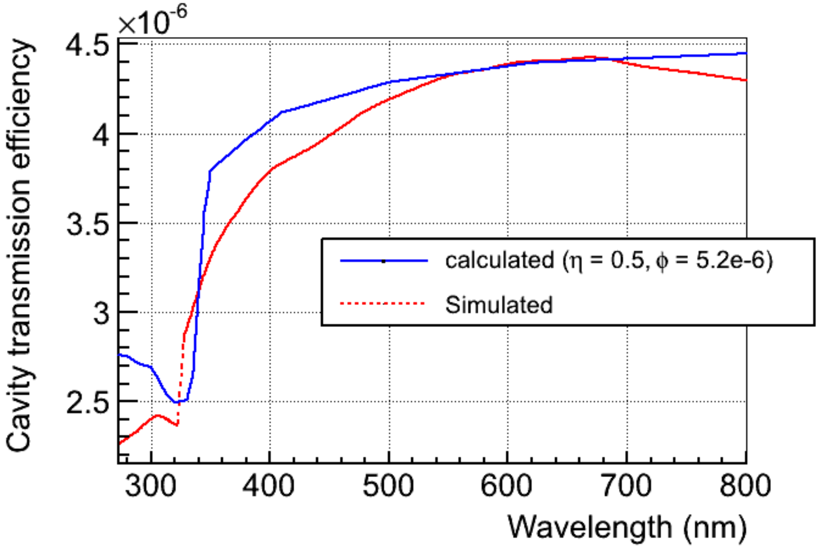
Ag reflectivity ($R(\lambda)$)

Multiple reflections ($\sum_{n=0}^{\infty} R^n (1-\varphi)^n$)

Probability to reach the fiber with correct angle (φ) $\approx \frac{S_{fiber}}{S_{cavity}} \frac{\Omega_{fiber}}{2\pi}$

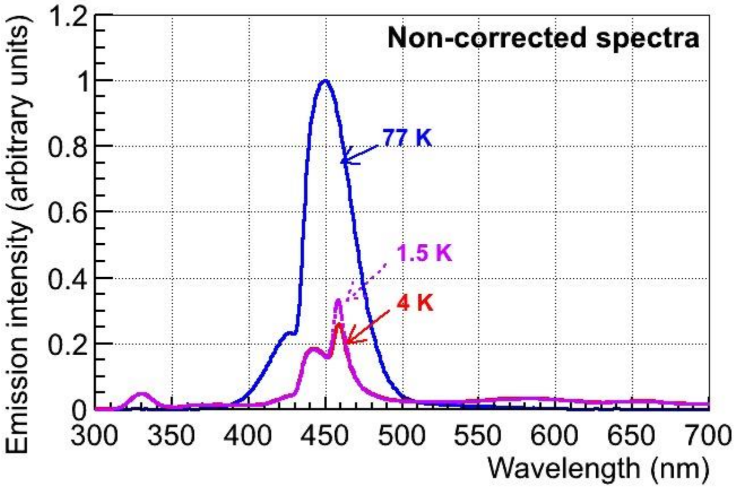
If we consider a factor of absorption in the crystal (η) ⇒

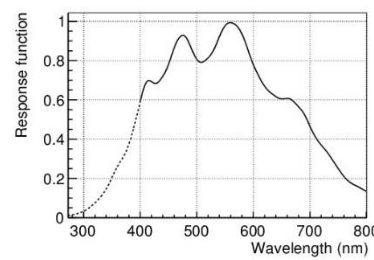
$$\eta_c(\lambda) = \eta x + \frac{R\eta\varphi}{1-R\eta(1-\varphi)}\eta(1-x)$$



Spectral response at several temperatures

Parylene-coated NaI(Tl)





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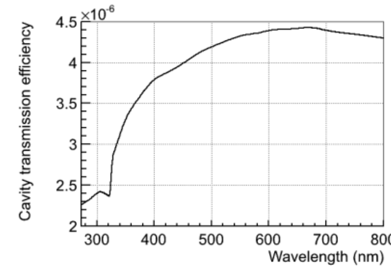
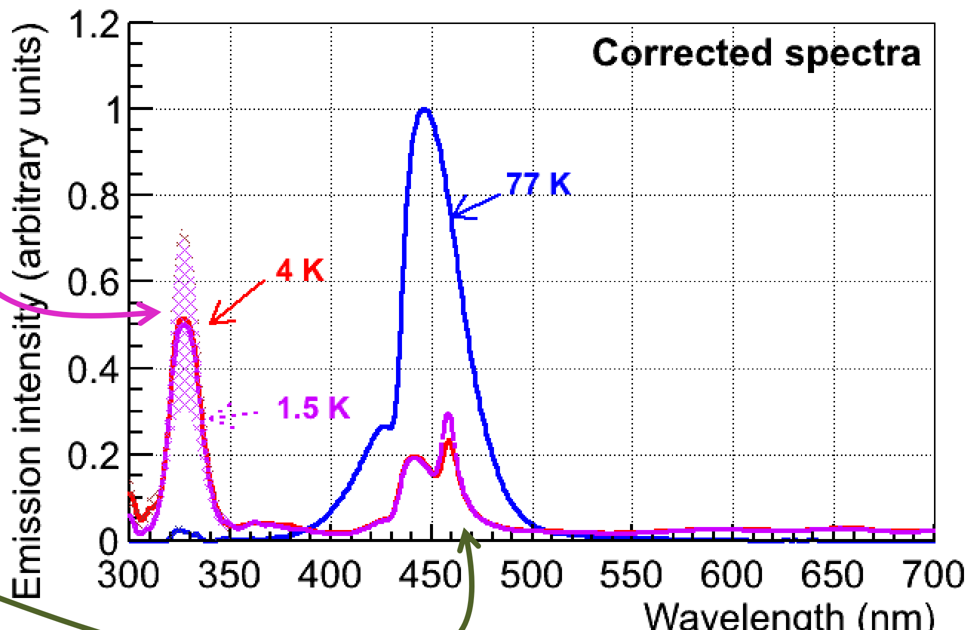


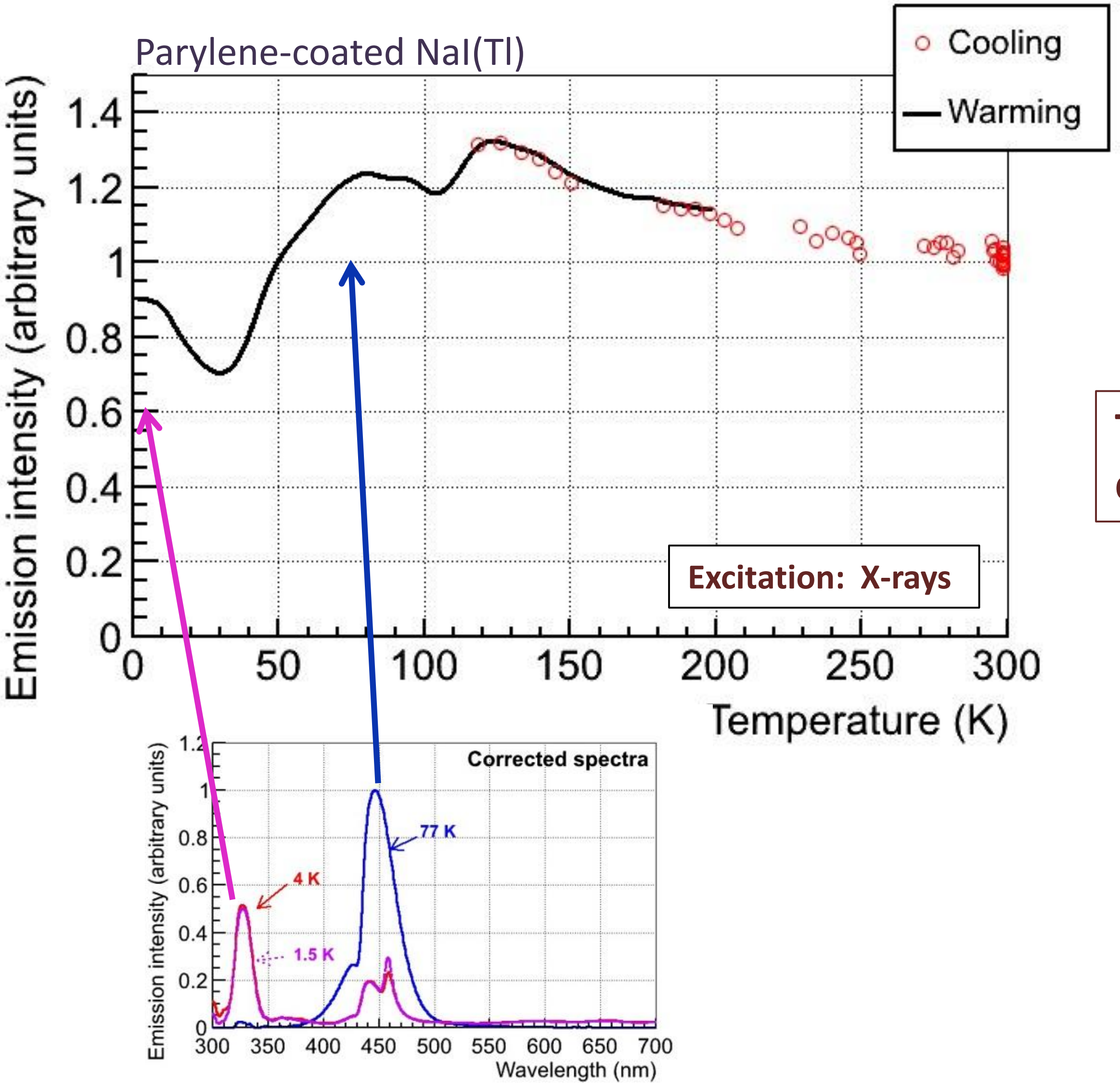


40% systematic error below 350 nm due to detector & cavity corrections

320 nm line dominates the emission at very low temperature

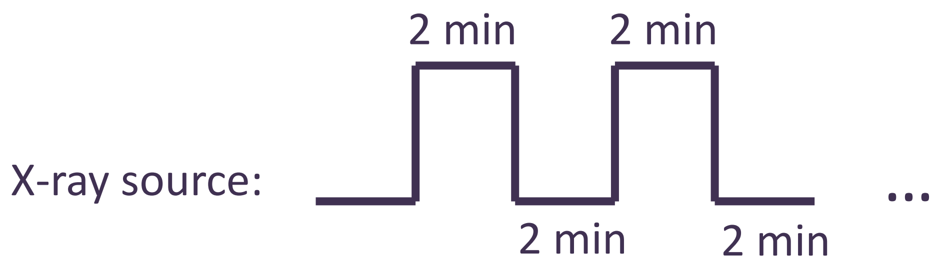
Double peak @ 440 nm and 460 nm

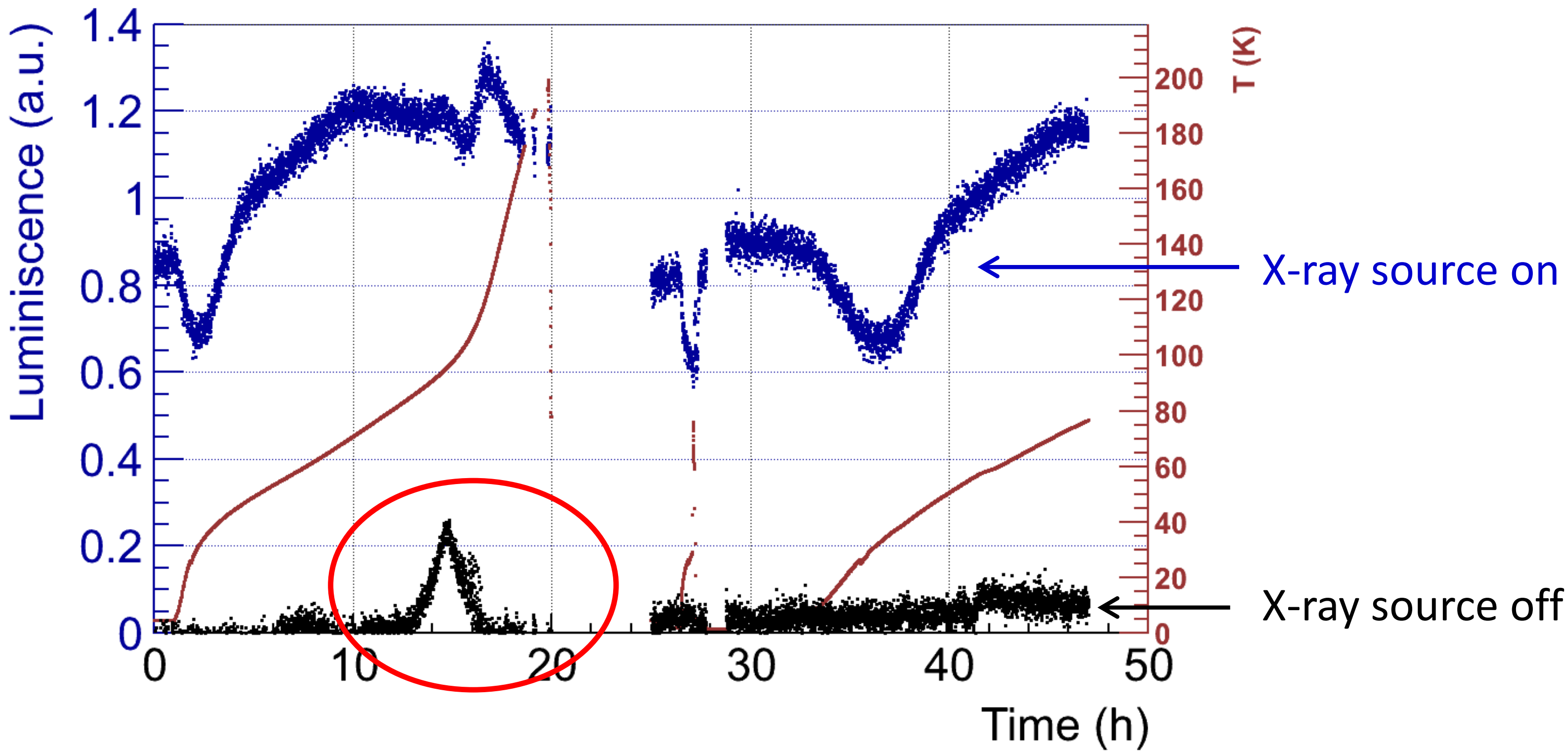
Spectral response at several temperatures



The light increase below 30 K is due to the 320 nm emission

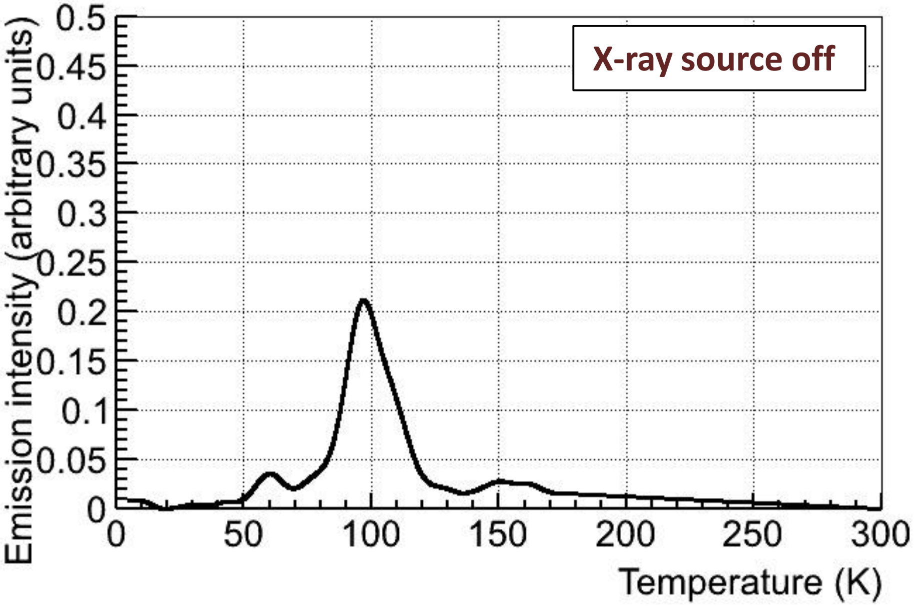
Thermoluminescence

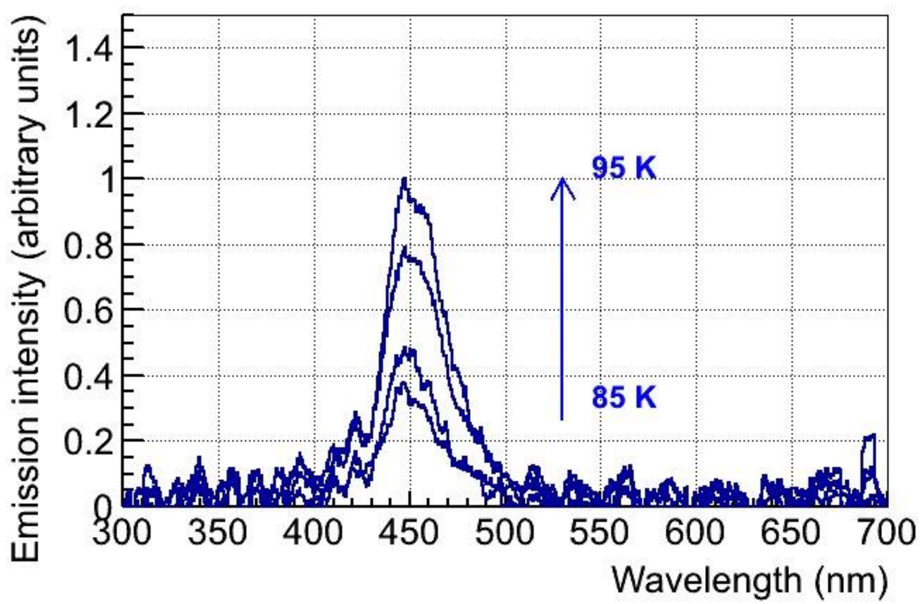




Thermoluminescence

- A thermoluminescence peak at around 95 K and two smaller ones at 60 and 150 K.
- Spectral response (taken at regular intervals between 84 and 95 K) shows a single emission band centered at 450 nm.



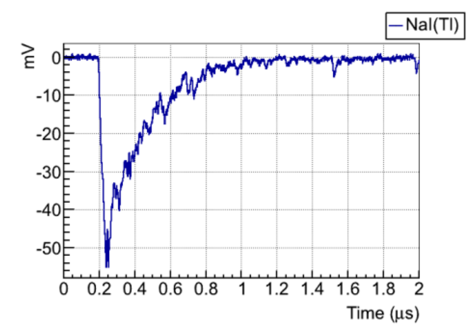


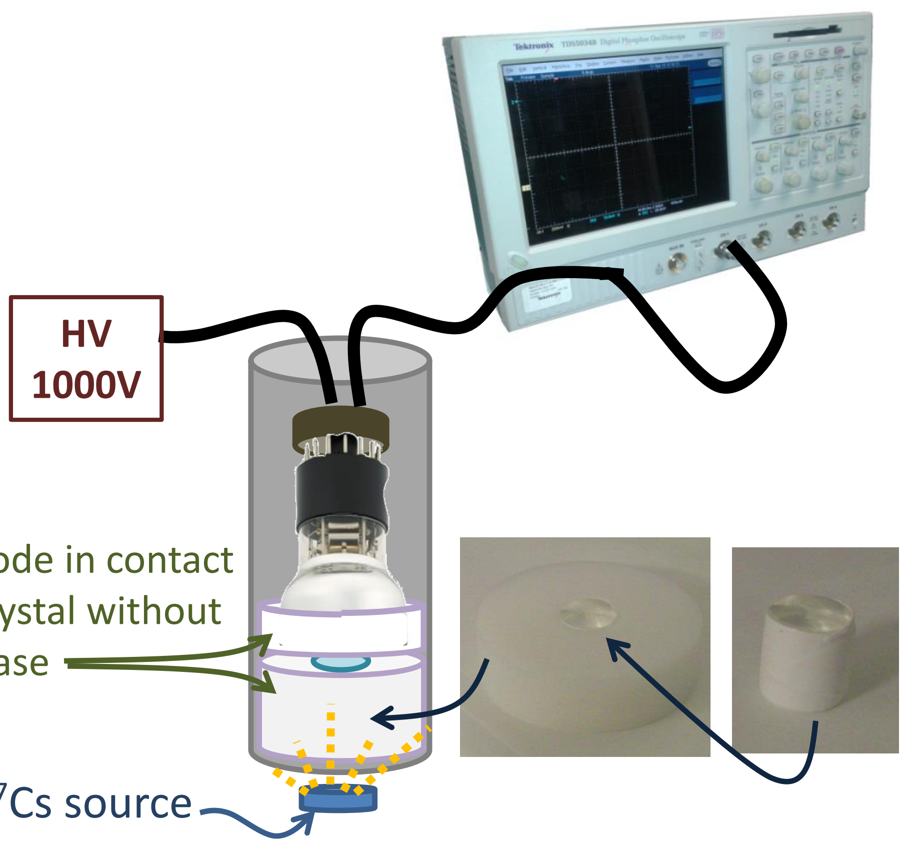
Response after thermal cycles

Light output measurement **before** and **after** the thermal cycle

- Light detector: **PMT HAMAMATSU R6233**-100SEL , spectral response: 300- 650 nm
- Excitation: ϒ 662 keV (^{137}Cs)

PMT pulses are digitized with a Tektronik TDS500 scope (2 µs window, SR=1.25 GS/s) and integrated by software





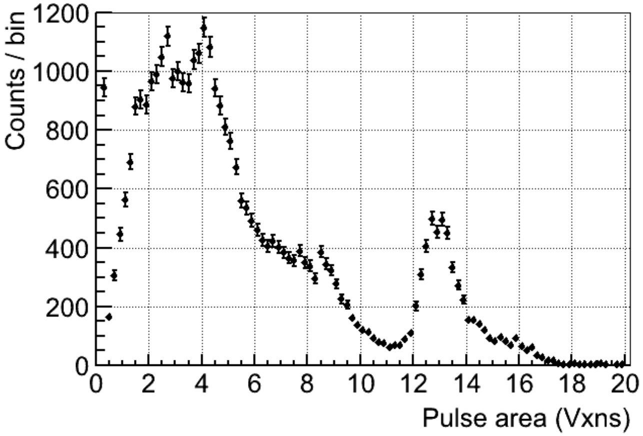
photocathode in contact with the crystal without optical grease

Crystal wrapped with a Teflon strip inside a cylindrical polyethylene holder

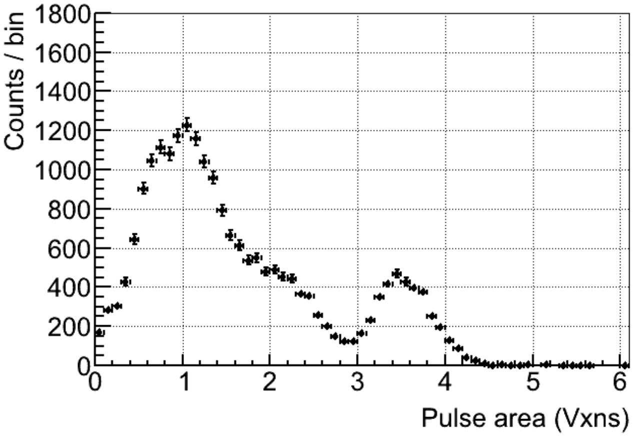
Parylene resistance to thermal cycles

Light output measurement before the thermal cycle

- **Parylene-coated NaI(Tl)**



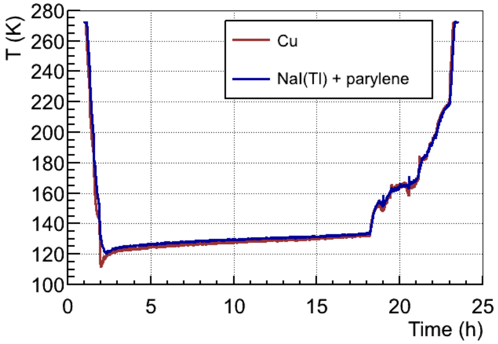
- **Parylene-coated NaI pure**

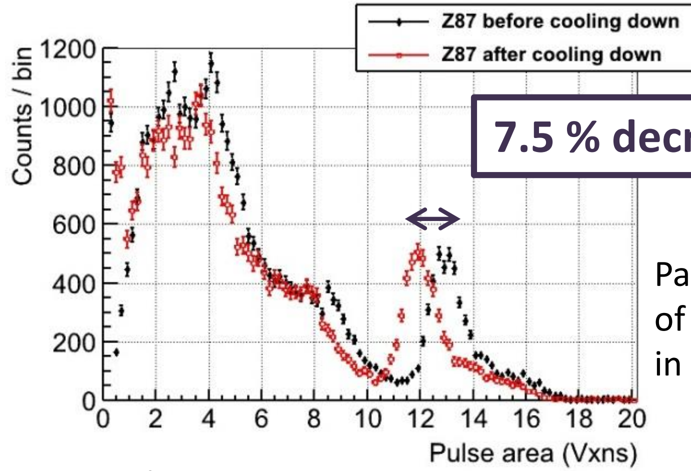


Parylene resistance to thermal cycles

Light output measurement before and after the thermal cycle

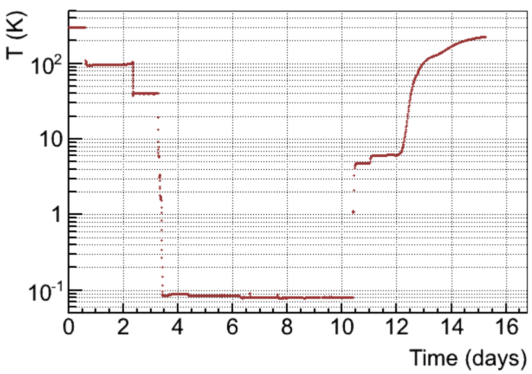
- **Parylene-coated NaI(Tl):**
  Cooling down to 100 K (with a N_2 bath)

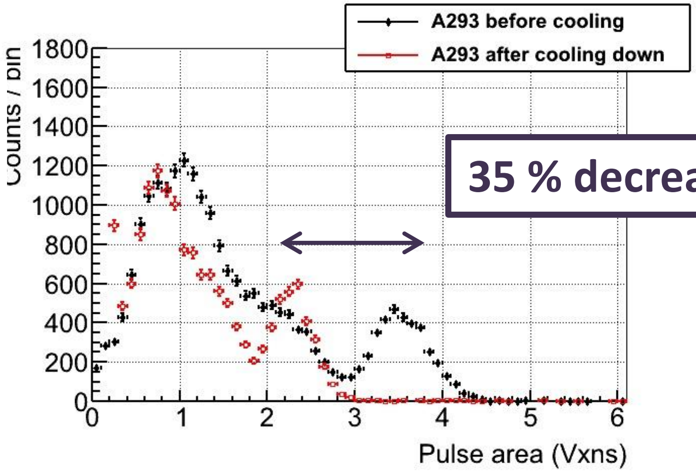




Parylene loss of adherence in certain areas

- **Parylene-coated NaI pure:** Cooling down to 80 mK (with a dilution unit) (Mounting time one week)

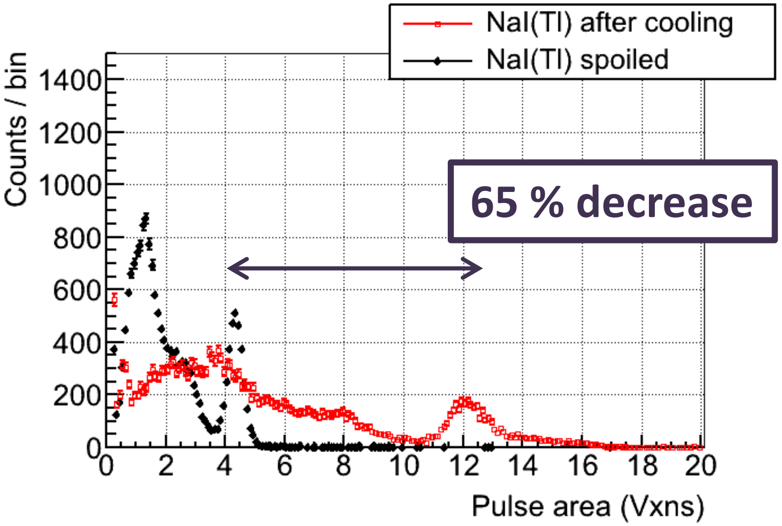




Parylene resistance to ambient moisture

2-5 μm parylene allow handling under normal RH conditions at several-days scale, but it is not a permanent coating against ambient moisture

After one month at RH 30-50% parylene coating was found to be ineffective, with large areas of the surface white and almost opaque, and a reduction in light output of around 65%



Summary

- NaI/NaI(Tl) hygroscopicity limits their application for many purposes, especially at low temperature. To avoid this problem we study parylene-coated NaI and NaI(Tl) crystals.
- The response of a parylene-coated NaI(Tl) under X-ray excitation have been studied from 1.5 to 300 K:
  - Maximum of emission is found to be at around 125 K
  - Notable decrease in light output below 70 K
  - Further increment of light below 30 K (light output @ 1.5 K= 90% light @ 300 K)
  - At 1.5 K the wavelength of maximum emission of NaI(Tl) is observed at 320 nm
- A thermoluminescence peak has been found at around 95 K, with a single emission band centered at 450 nm. Two smaller thermoluminescence peaks have also been observed at 60 K and 150 K
- We have studied the mechanical resistance of the coating under thermal cycles, observing a degradation of the optical appearance and the light output after cooling down to about 100 mK, which compromises the reusability of the samples.