Fluorescence of optical materials down to 4 K – acrylic, TPB and pyrene

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*Cryogenic Apparatus for Fluorescence Experiments:
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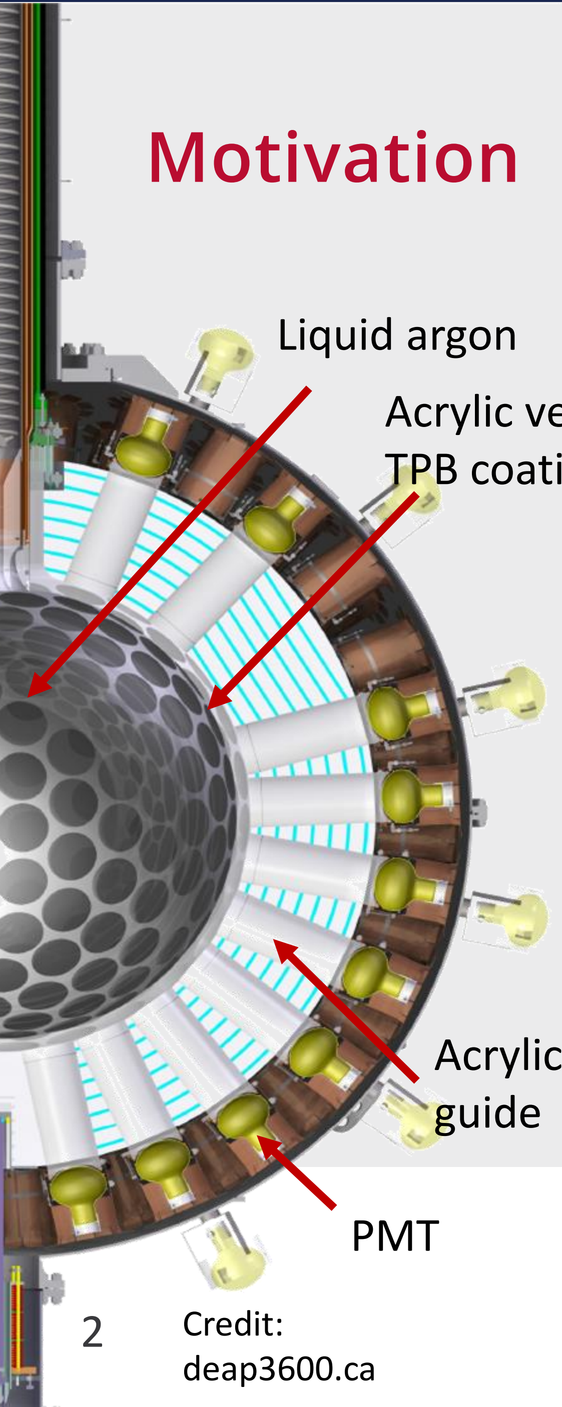
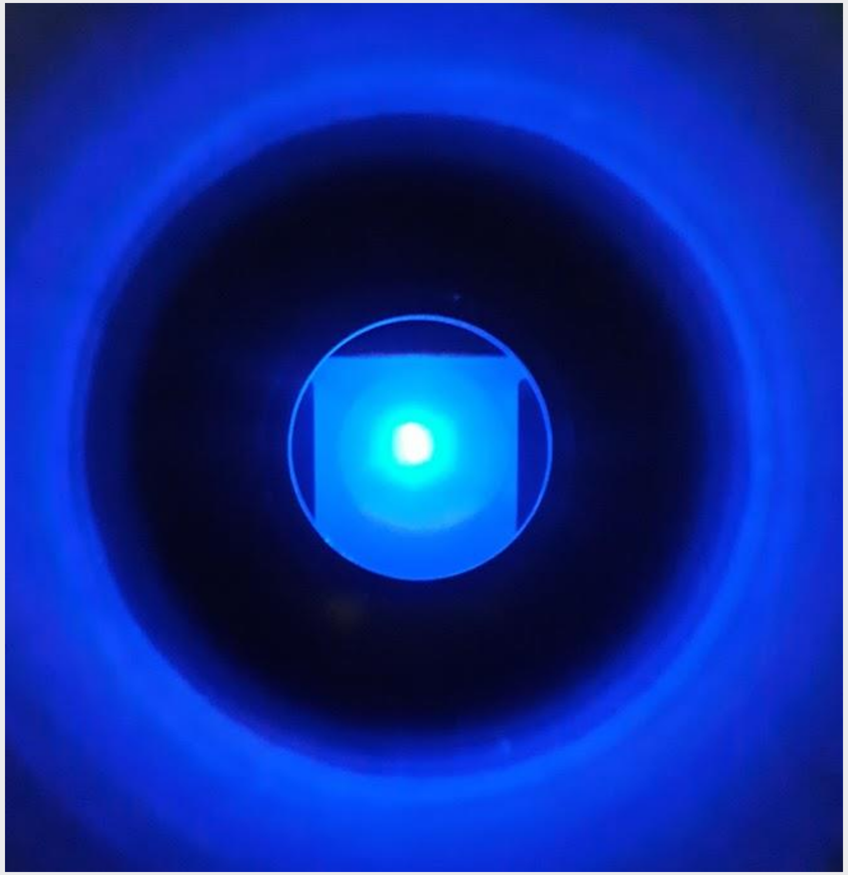
Motivation



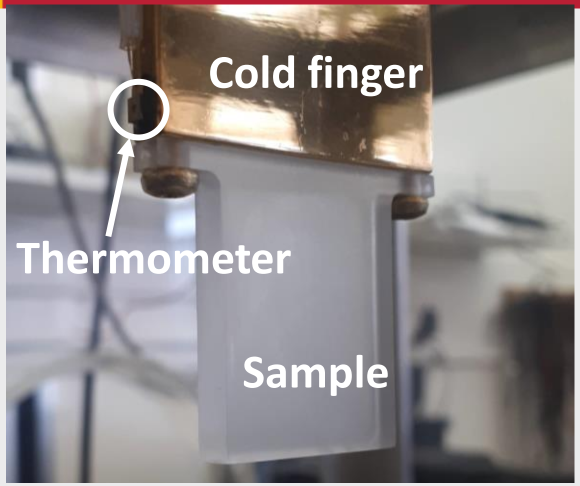
- Acrylic and applied wavelength shifters can fluoresce due to the UV scintillation light from particle interactions with noble liquids.
- Fluorescence properties of transparent materials used for these detectors and coatings applied to them, like wavelength shifters, can change with temperature.



Fluorescence from a TPB coated acrylic sample

Credit: deap3600.ca

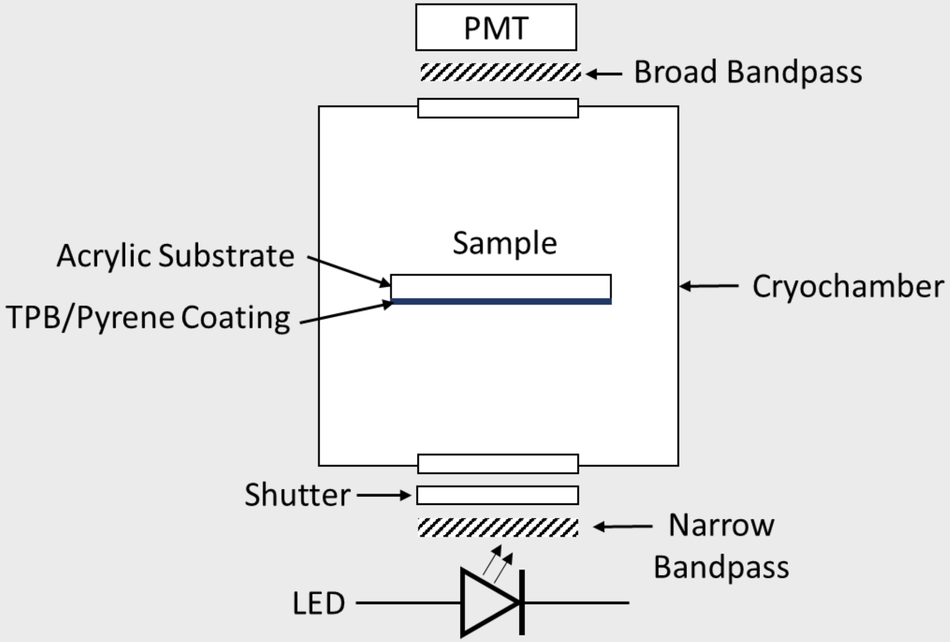
Optical Cryostat



- System containing a material sample capable of changing the temperature of the sample between 4 K and 300 K at <10^{-6} mbar.
- Two operating modes: Time-resolved and spectrometer mode

Time-resolved mode:

- Excited with a ~6 ns wide 285 nm LED pulse.
- Fluorescent light detected by a PMT read out by a digitizer.
- Broad bandpass filter with a 375 nm lower limit to eliminate stray UV LED light from reaching the PMT.

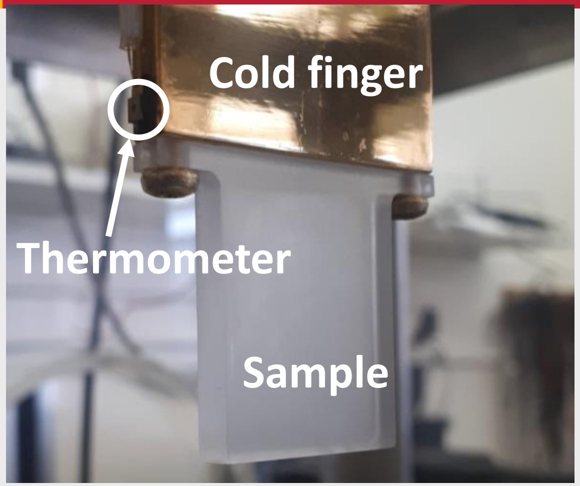


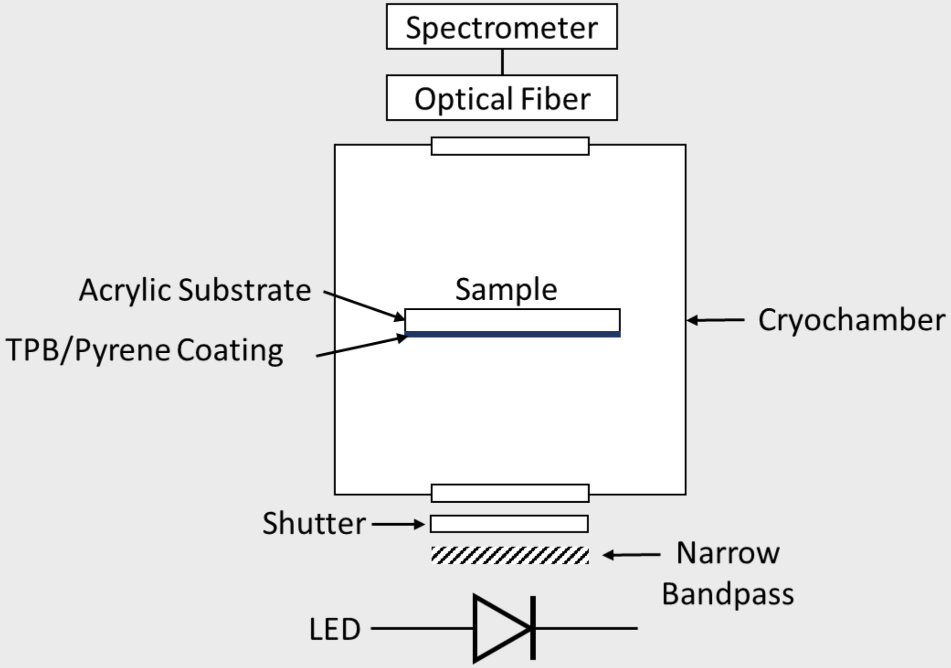
Optical Cryostat

- System containing a material sample capable of changing the temperature of the sample between 4 K and 300 K at $<10^{-6}$ mbar.
- Two operating modes: Time-resolved and spectrometer mode

Spectral mode:

- LED run continuously
- Spectrometer can also be attached at the same position as the PMT to study wavelength dependent fluorescence features.



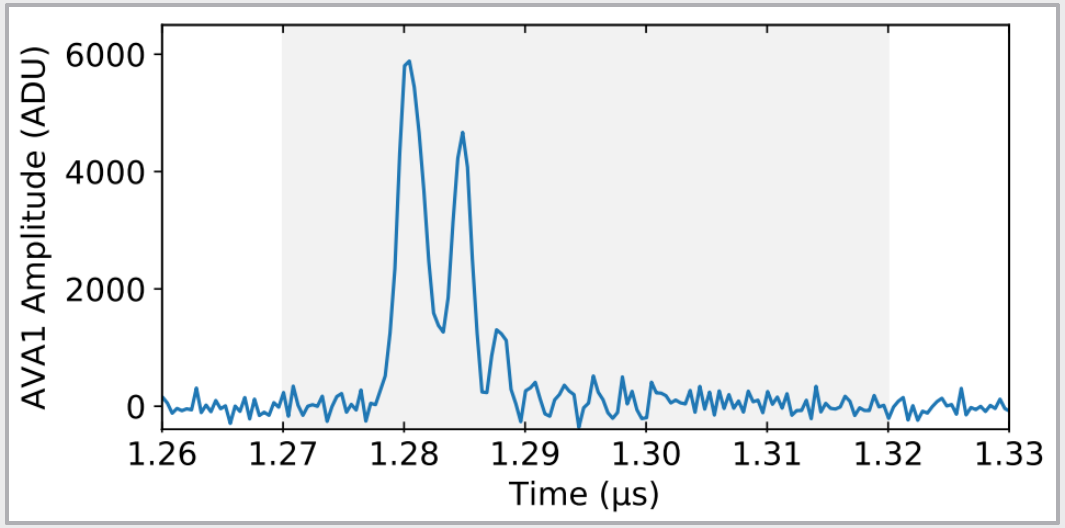


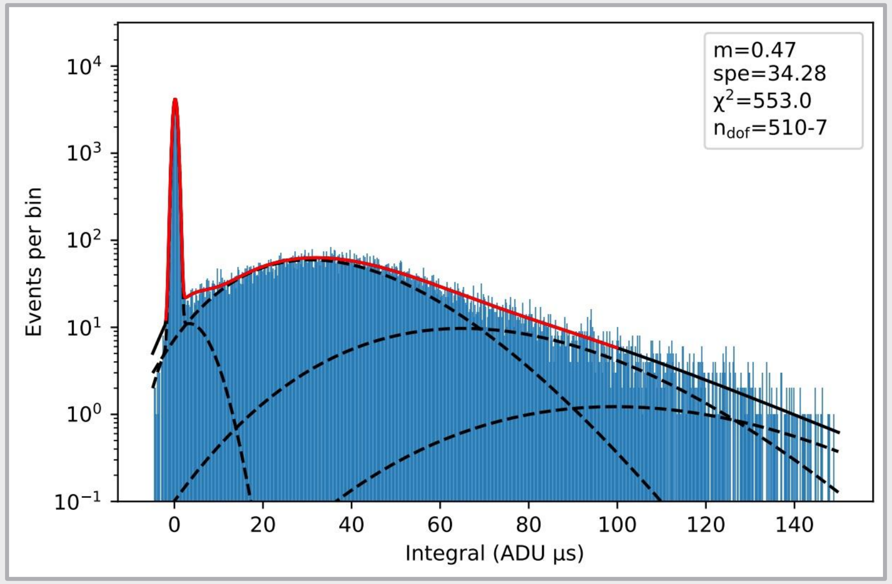
Acrylic and TPB Study

[1] E. Ellingwood et al., Ultraviolet-induced fluorescence of poly(methyl methacrylate) compared to 1,1,4,4-tetraphenyl-1,3-butadiene down to 4 K, Nuclear Instruments and Methods in Physics Research Section A, Volume 1039, 167119.

Calculating the Acrylic Light Yield

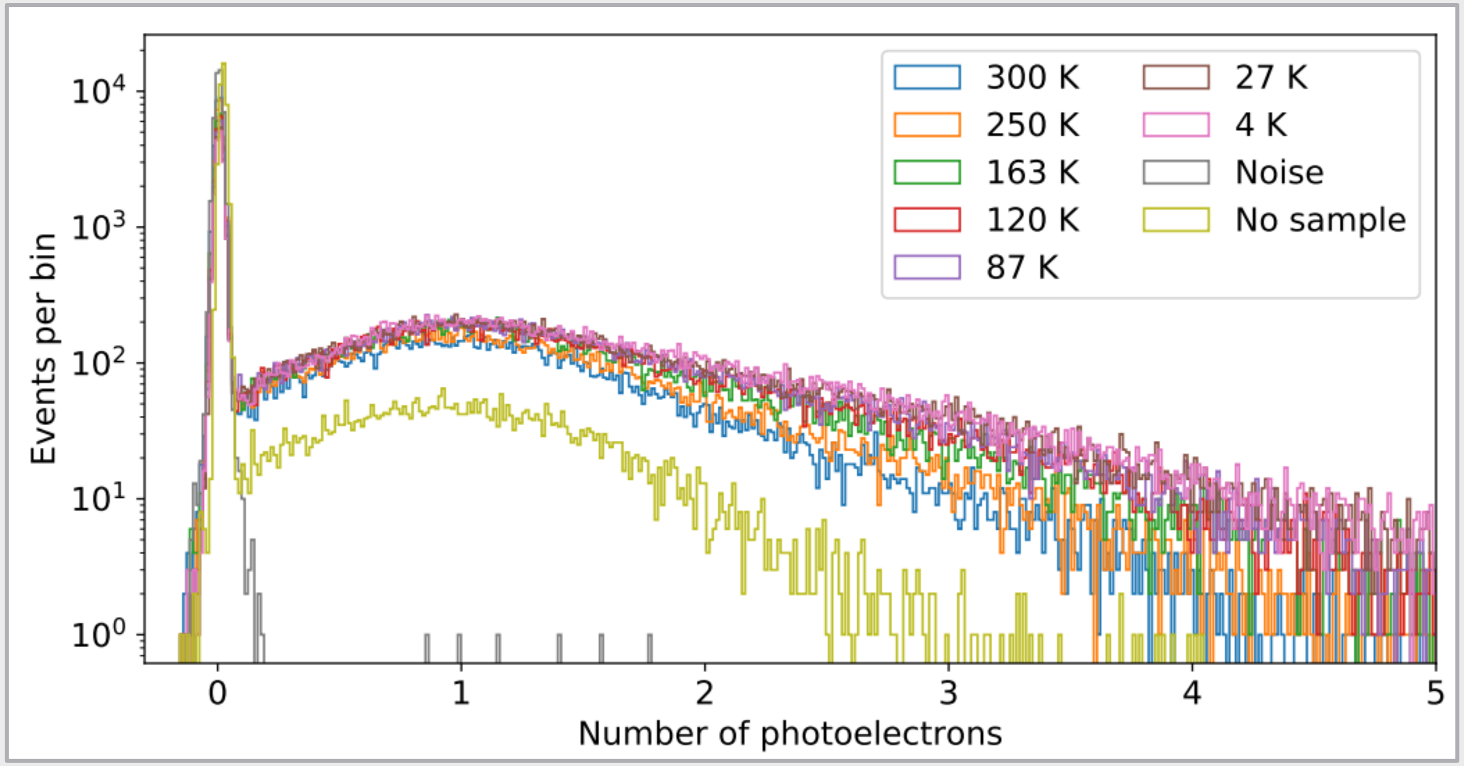
1. At each temperature record 45000 individual fluorescence pulses.
2. The light yield is the integral of each PMT pulse in a 50 ns window.
3. Build an integral distribution and fit with a model of single photoelectron distribution [2].
4. Light yield is the mean of this integral distribution.
5. Model also fits the SPE value the integral value per photoelectron.





Calculating the Acrylic Light Yield

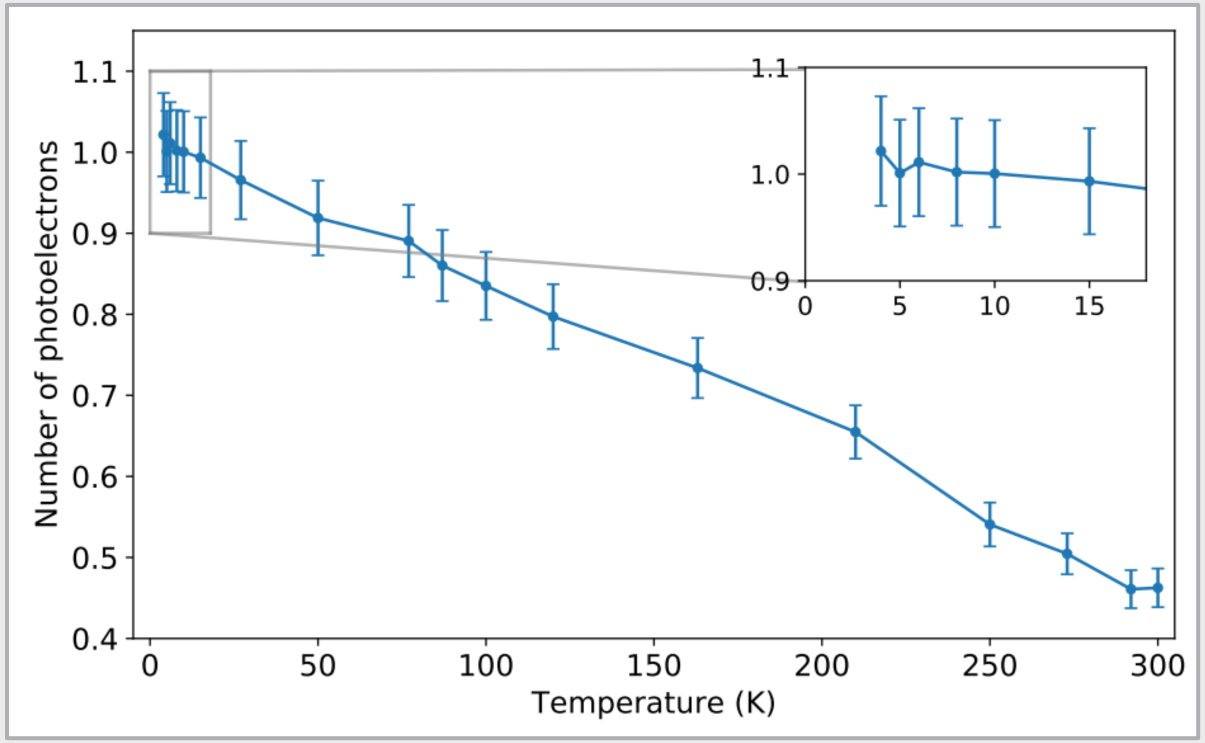
6. Repeat for different temperatures
7. Take noise run (@300 K) with shutter between LED and cryochamber closed
8. Take no sample run (@300 K) with LED pulsing as usual, but not sample installed.



Acrylic Light Yield Results

Relative to the light yield at 300 K:

- ~85% light yield increase at 87 K
- ~120% light yield increase at 4 K



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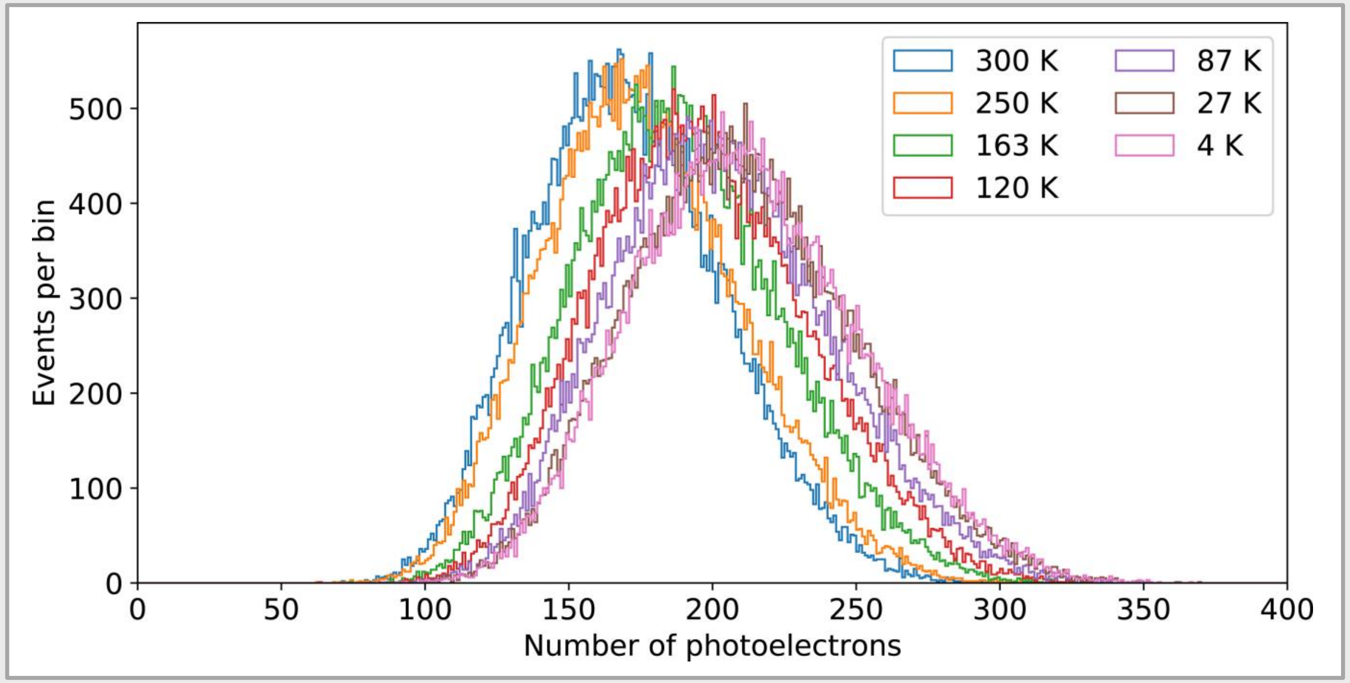
Multi-photoelectron analysis

- Exactly the same data acquisition procedure as acrylic only changing LED voltages or digitizer vertical range due to much larger pulses. These changes are accounted for in the analysis.

- Fit integral distribution with a skew normal function. [3]

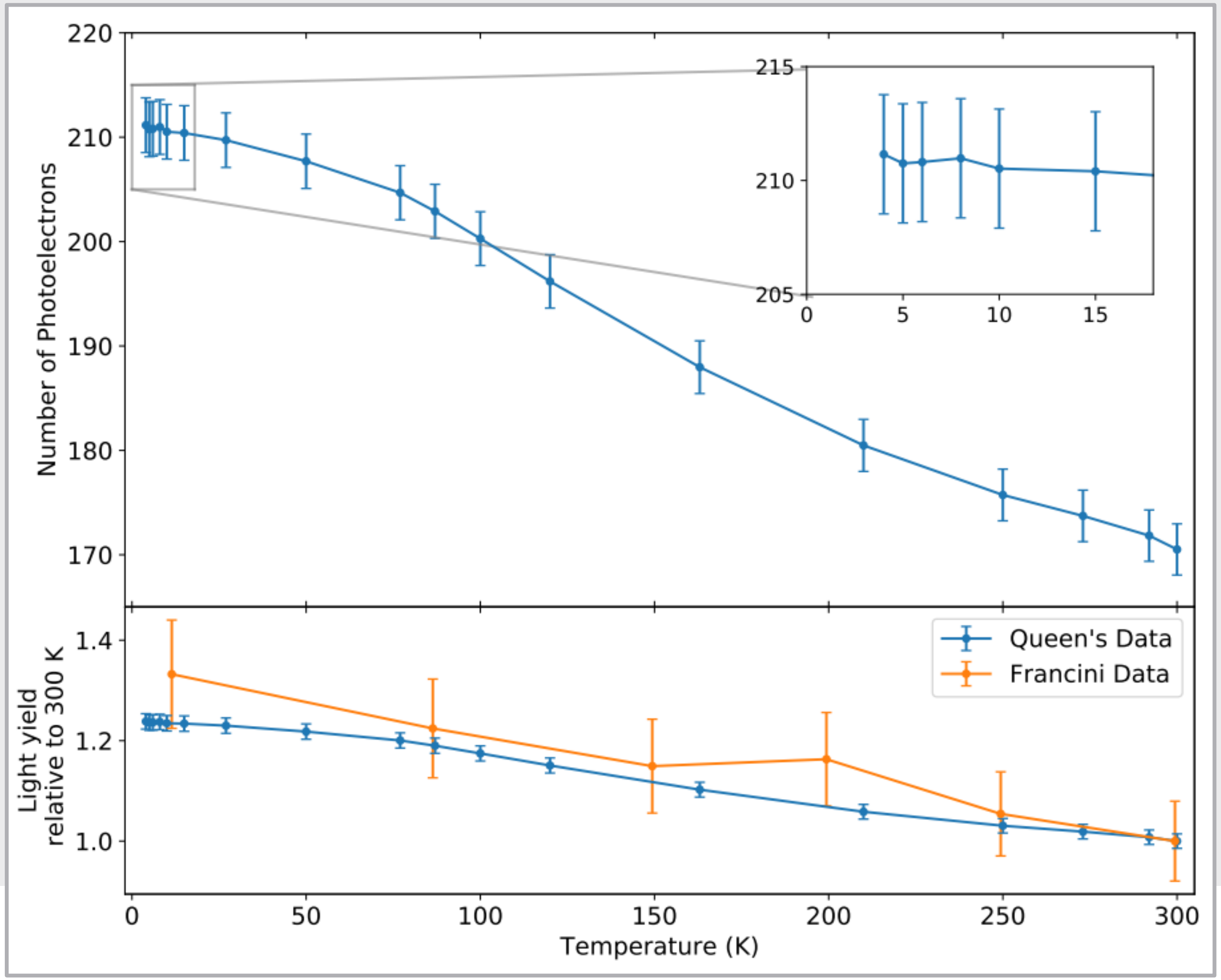
*
$$K(A,\xi,\omega,\alpha,s) = \frac{2A}{\omega\sqrt{2\pi}} exp\left(\frac{(\xi-s)^2}{2\omega^2}\right) \int_{-\infty}^{\alpha\left(\frac{\xi-s}{\omega}\right)} \frac{1}{\sqrt{2\pi}} exp\left(\frac{-t^2}{2}\right)\, dt$$

- The mean of the fit is the light yield.



*A = scaling factor, ξ = location on distribution , s = scale of the distribution, α = shape of distribution

TPB Light Yield Results



Light yield relative to 300 K:

- 19.0% increase at 87 K
- 23.5% increase at 4 K

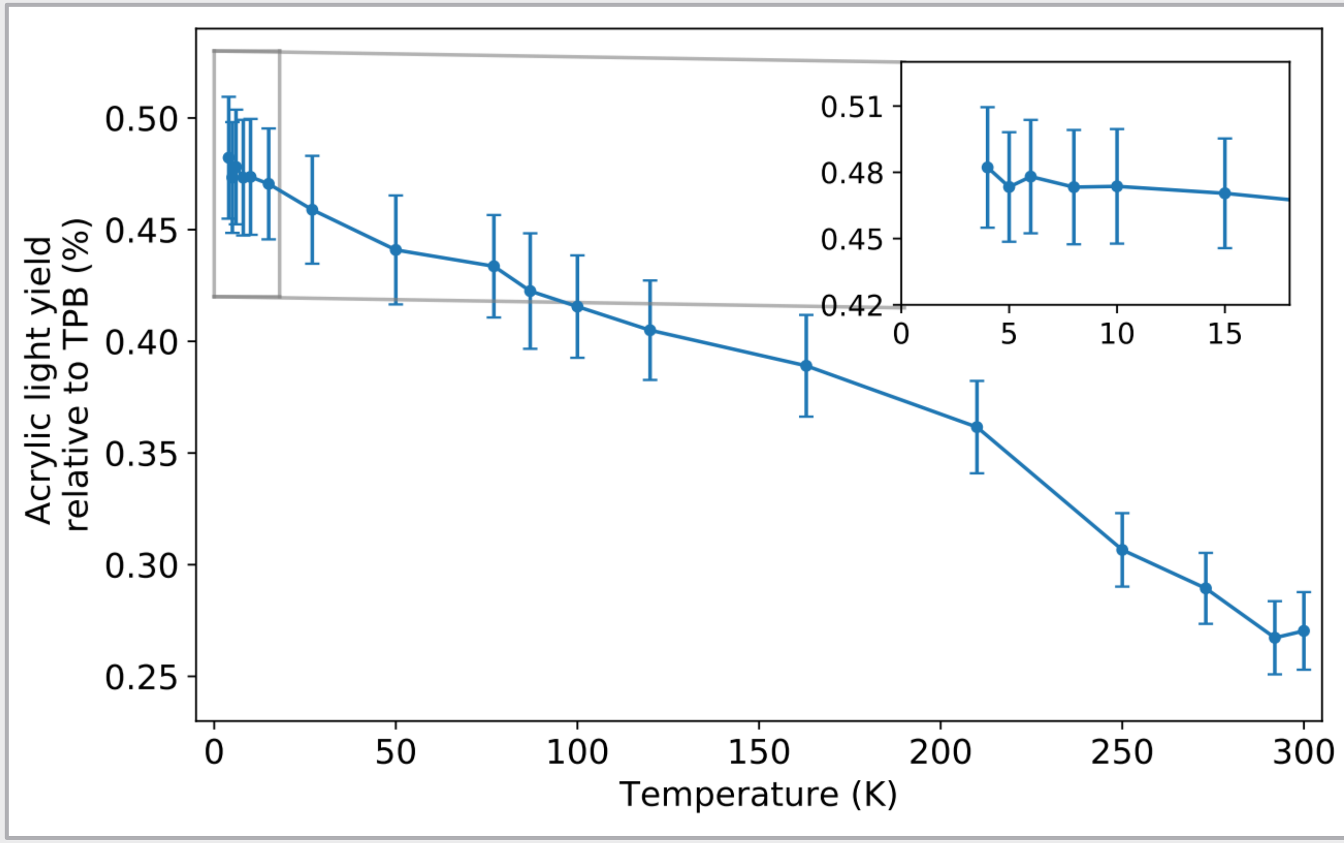
- Light yield relative to 300 K consistent within errors with previous characterizations of TPB (Francini, 2013) [4].

Acrylic-TPB Relative Light Yield

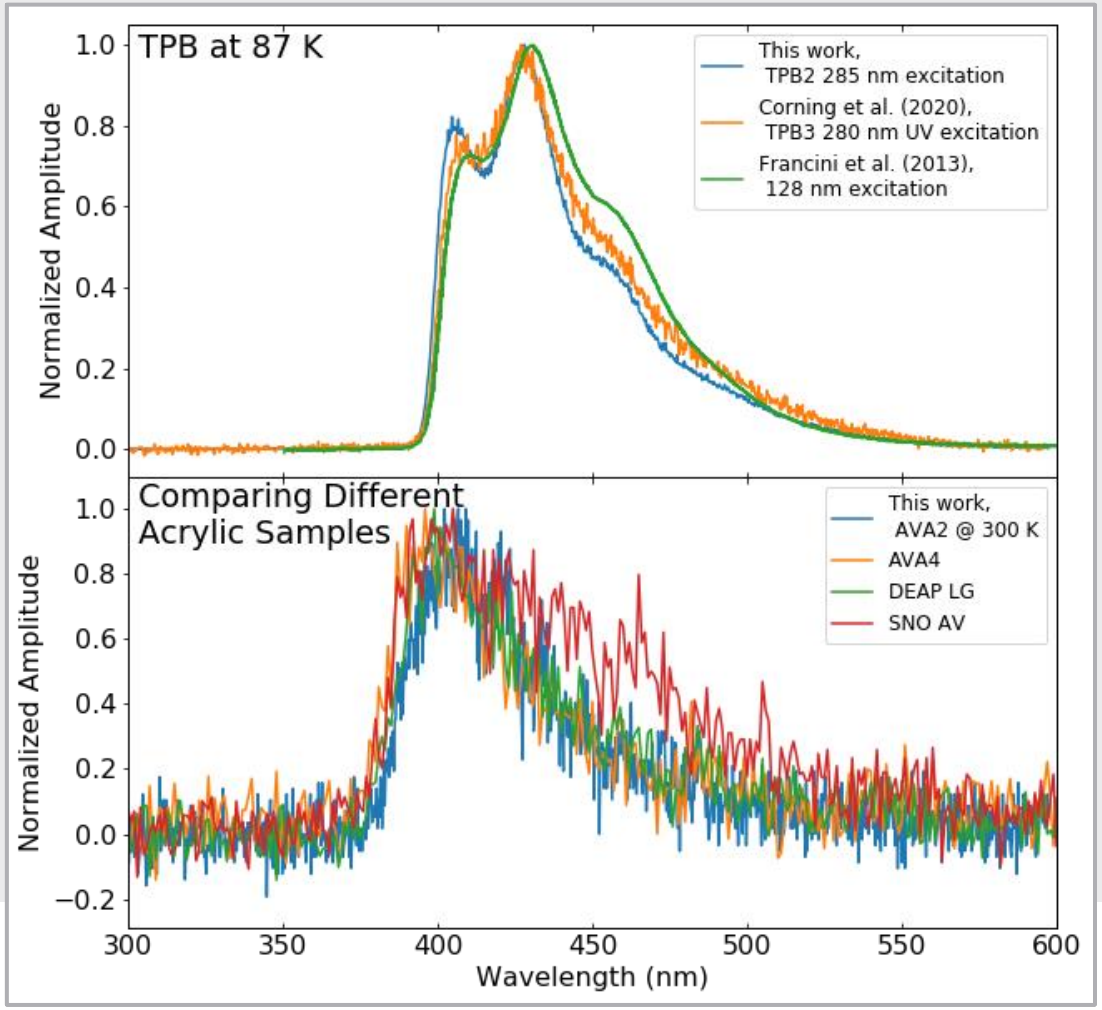
- Relative light yields are used to compare light yield results of different materials independent of the measurement method.
- Does still depend on the type of acrylic used

Relative light yield varies depending on temperature:

- 0.27% at 300 K
- 0.42% at 87 K
- 0.48% at 4 K



Comparison of TPB and Acrylic Spectra to Literature



- TPB and acrylic samples presented in literature were not all prepared or measured in the same way.
- The acrylic and TPB spectra look similar to spectra from literature at the same temperatures.
- Different formulations and measurement techniques can account for the slight differences in spectral features.

Pyrene Study

[5] H. Benmansour, et al., Fluorescence of pyrene-doped polystyrene films from room temperature down to 4 K for wavelength-shifting applications, J. Instrum. 16 (12) (2021) P12029. doi:10.1088/1748-0221/16/12/p12029.

[6] D. Gallacher, et al., Development and characterization of a slow wavelength shifting coating for background rejection in liquid argon detectors, Nuclear Instruments and Methods in Physics Research Section A, Volume 1034, 166683.

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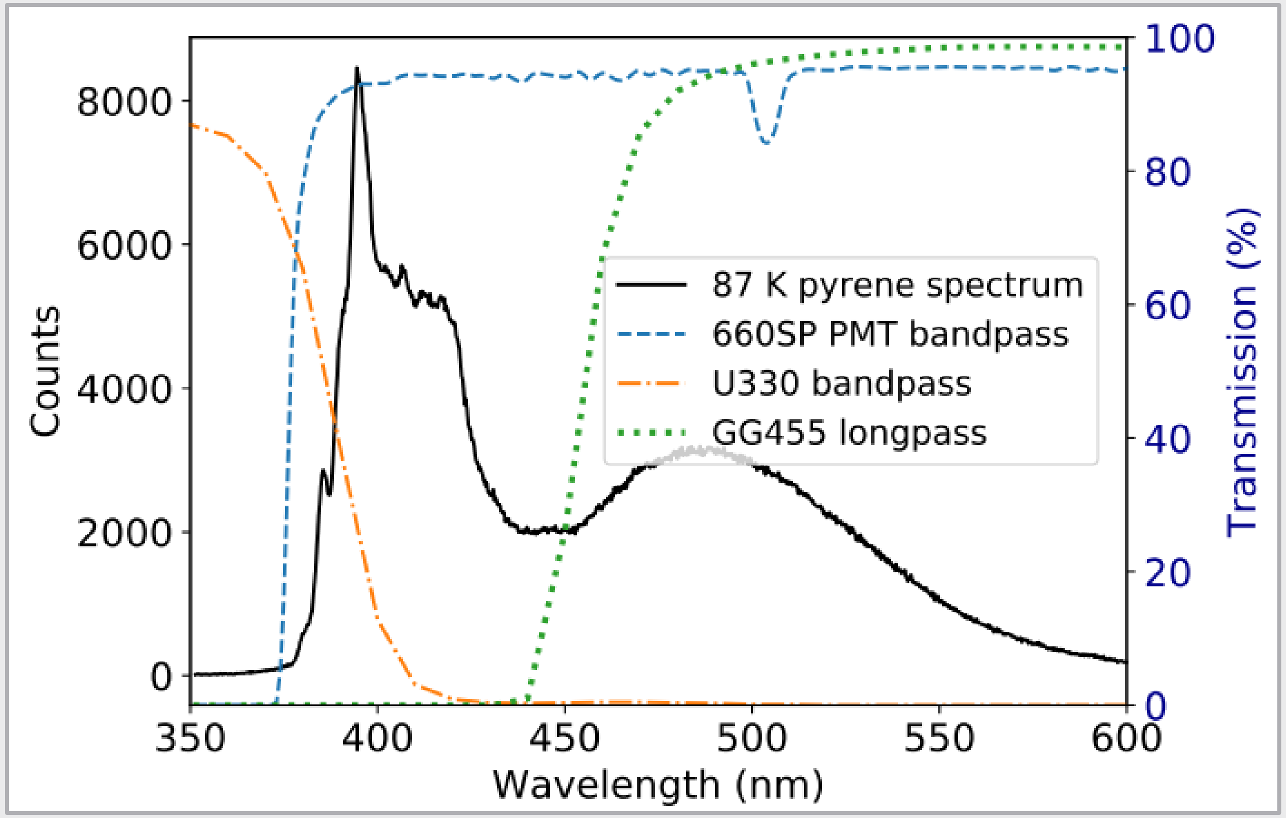
Pyrene Spectrum

Pyrene-polystyrene is used as a complementary WLS to TPB in DEAP-3600 to improve spatial pulse shape discrimination. [6]

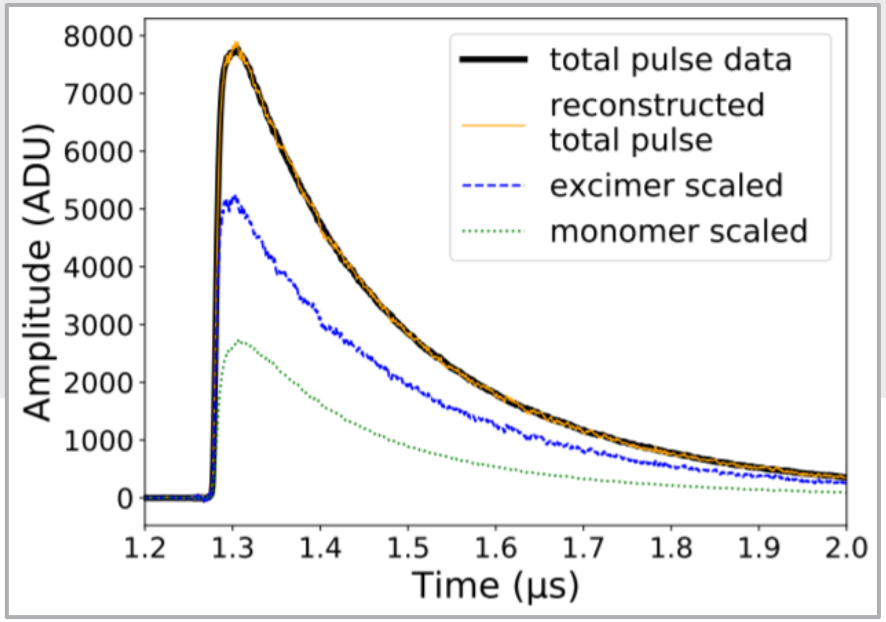
- Pyrene has a much longer time constant (~100s of ns) compared to TPB (~ns).
- The pyrene fluorescence mechanism consists of two components: monomer and excimer [7]
- Filters are used in time-resolved measurements to separately fit contributions from different emissions.



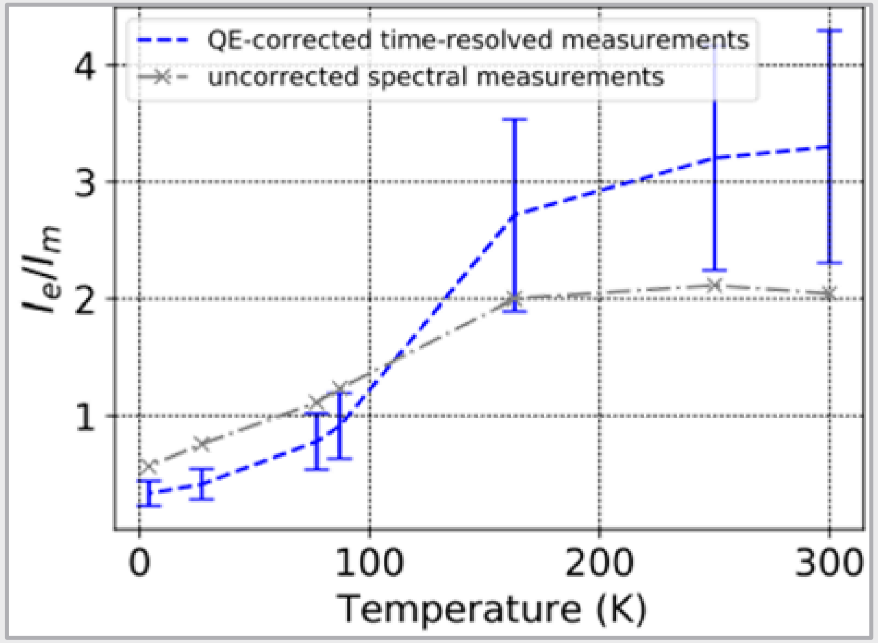
Monomer and excimer intensity

1. Take 3 data sets at each temperature:
   a. Monomer (with U330 filter)
   b. Excimer (with GG455 filter)
   c. Total pulse (no additional filter)

2. Scale monomer and excimer pulses such that their sum best reconstructs the total pulse data.



3. Integrate scaled monomer and excimer in a specific time window to determine proportion of light from monomer and excimer.



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Pyrene Time Constants

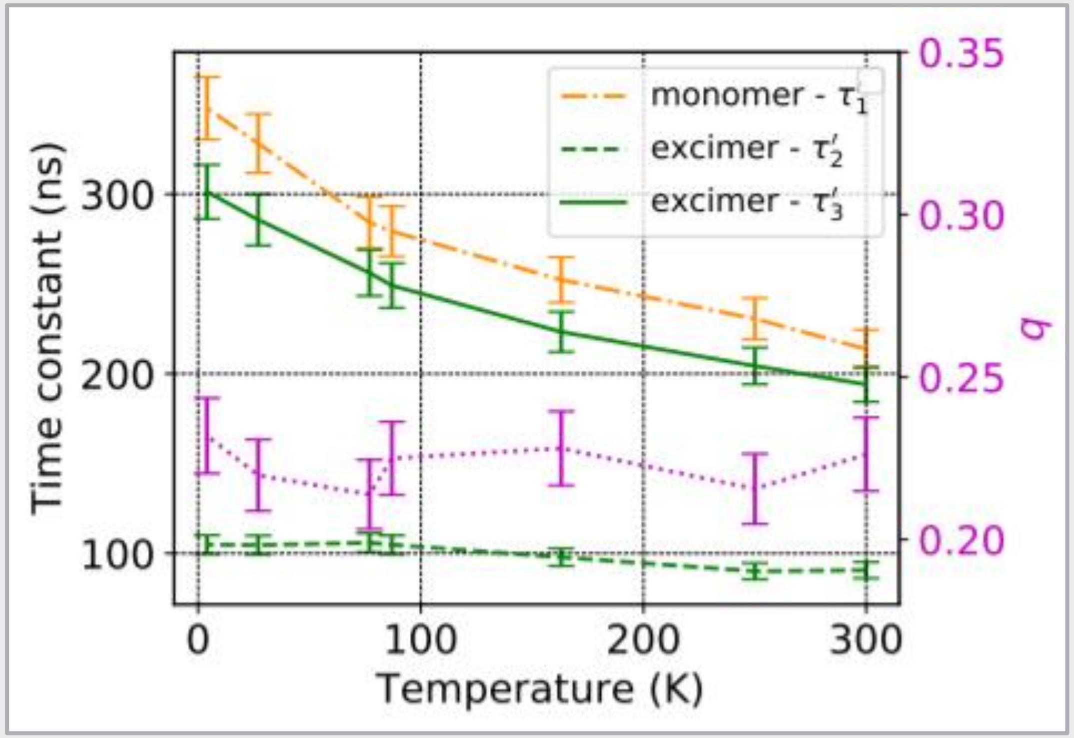
- Fit the monomer and excimer time constants separately.
- The monomer model [8]

$$i_m(t) = \frac{N_1'}{\tau_1'} e^{-\frac{t}{\tau_1'} - 2q\sqrt{\frac{t}{\tau_1'}}}$$

- The excimer model [8]

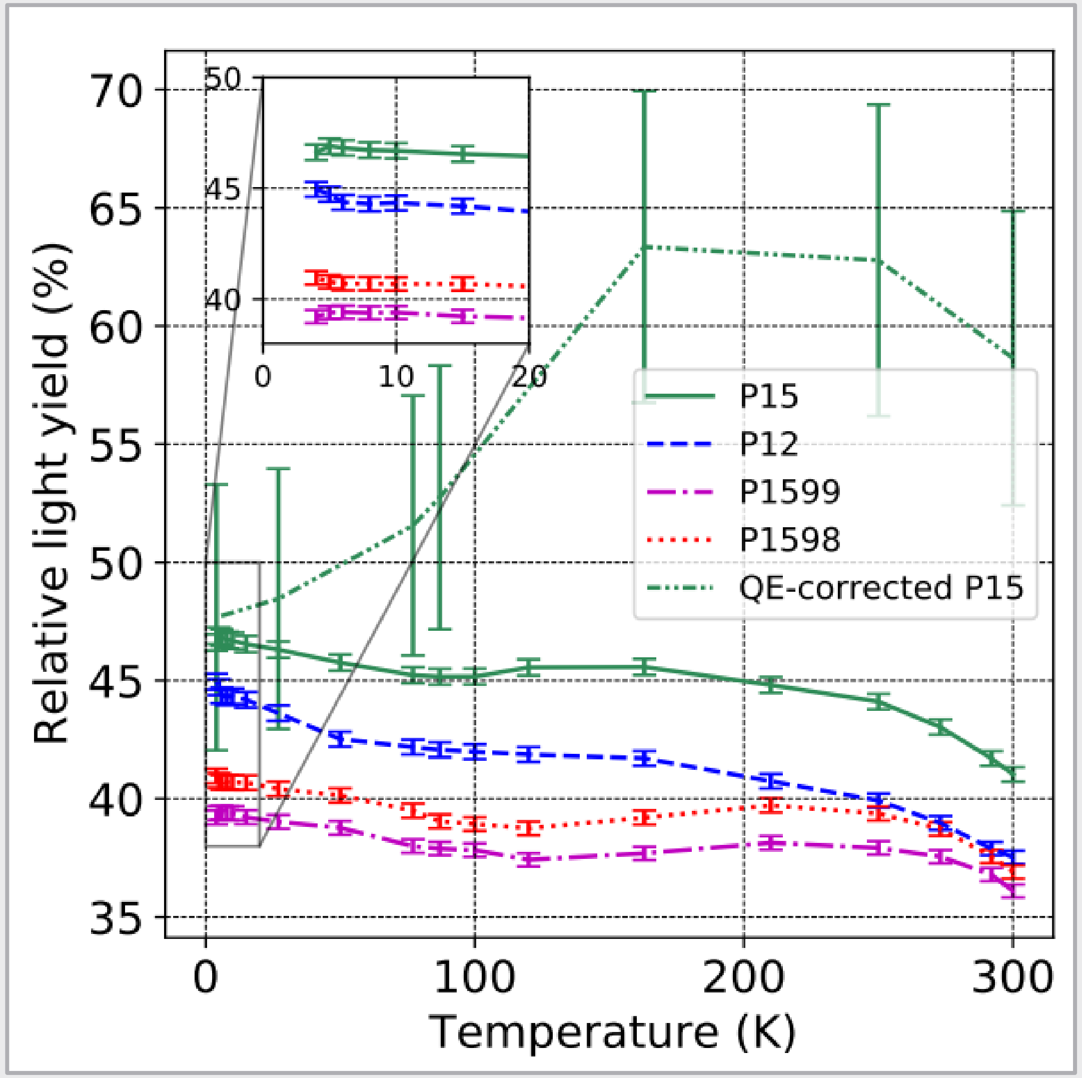
$$i_e(t) = -\frac{N_{\text{rise}}'}{\tau_{\text{rise}}'} e^{-\frac{t}{\tau_{\text{rise}}'}} + \frac{N_2'}{\tau_2'} e^{-\frac{t}{\tau_2'}} + \frac{N_3'}{\tau_3'} e^{-\frac{t}{\tau_3'}}$$

Pyrene time constants change with temperature



Pyrene Light Yields

- Like the previous study this is the light yield relative to TPB. (solid green line labelled P15)
- Actually studied four different pyrene-polystyrene coatings
  - **P15: 15% conc. , 99.9% f.g.***
  - P12: 12% conc., 99.9% f.g.
  - P1599: 15% conc., 99% f.g.
  - P1598: 15% conc., 98% f.g.



* f.g. = fluorescence grade

Future Cryostat Plans

- Currently working on a study of the fluorescence of Clevios coated acrylic samples.
- In addition to the usual 285 nm LED we also have 267 nm with other wavelength LEDs arriving soon.

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Conclusions

The fluorescent light yield from acrylic, TPB and pyrene increases with decreasing temperature.

Acrylic produces a low level of fluorescence (compared to TPB). The light yield and spectrum could depend on the type of acrylic.

The flowguides of DEAP-3600 will be coated with pyrene. The measured optical properties of this coating will help to determine the expected signal from events through pyrene.

Plans for using updated system with different wavelength LEDs to measure the temperature dependent fluorescence of different materials.

References

[1] E. Ellingwood et al., Ultraviolet-induced fluorescence of poly(methyl methacrylate) compared to 1,1,4,4-tetraphenyl-1,3-butadiene down to 4 K, Nuclear Instruments and Methods in Physics Research Section A, Volume 1039, 167119.

[2] I. Chirikov-Zorin, et al., Method for precise analysis of the metal package photomultiplier single photoelectron spectra, Nucl. Instr. Meth. A 456 (3) (2001) 310–324.

[3] R. Cheng, Non-Standard Parametric Statistical Inference, Oxford University Press, 2017.

[4] R. Francini, VUV-Vis optical characterization of Tetraphenyl-butadiene films on glass and specular reflector substrates from room to liquid Argon temperature, J. Instrum 8 (2013) 09006.

[5] H. Benmansour, et al., Fluorescence of pyrene-doped polystyrene films from room temperature down to 4 K for wavelength-shifting applications, J. Instrum. 16 (12) (2021) P12029.

[6] D. Gallacher, et al., Development and characterization of a slow wavelength shifting coating for background rejection in liquid argon detectors, Nuclear Instruments and Methods in Physics Research Section A, Volume 1034, 166683.

[7] F.M. Winnik, Photophysics of preassociated pyrenes in aqueous polymer solutions and in other organized media, Chem. Rev. 93 (1993) 587.

[8] G.E. Johnson, Effect of Concentration on the Fluorescence Spectra and Lifetimes of Pyrene in Polystyrene Films, Macromolecules 13 (1980) 839.

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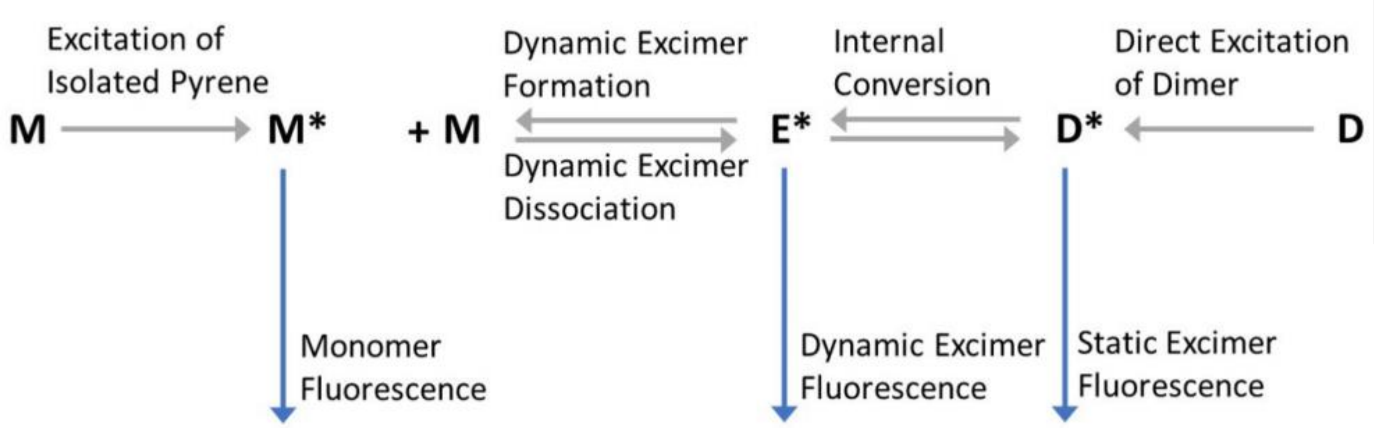
Extra Slides

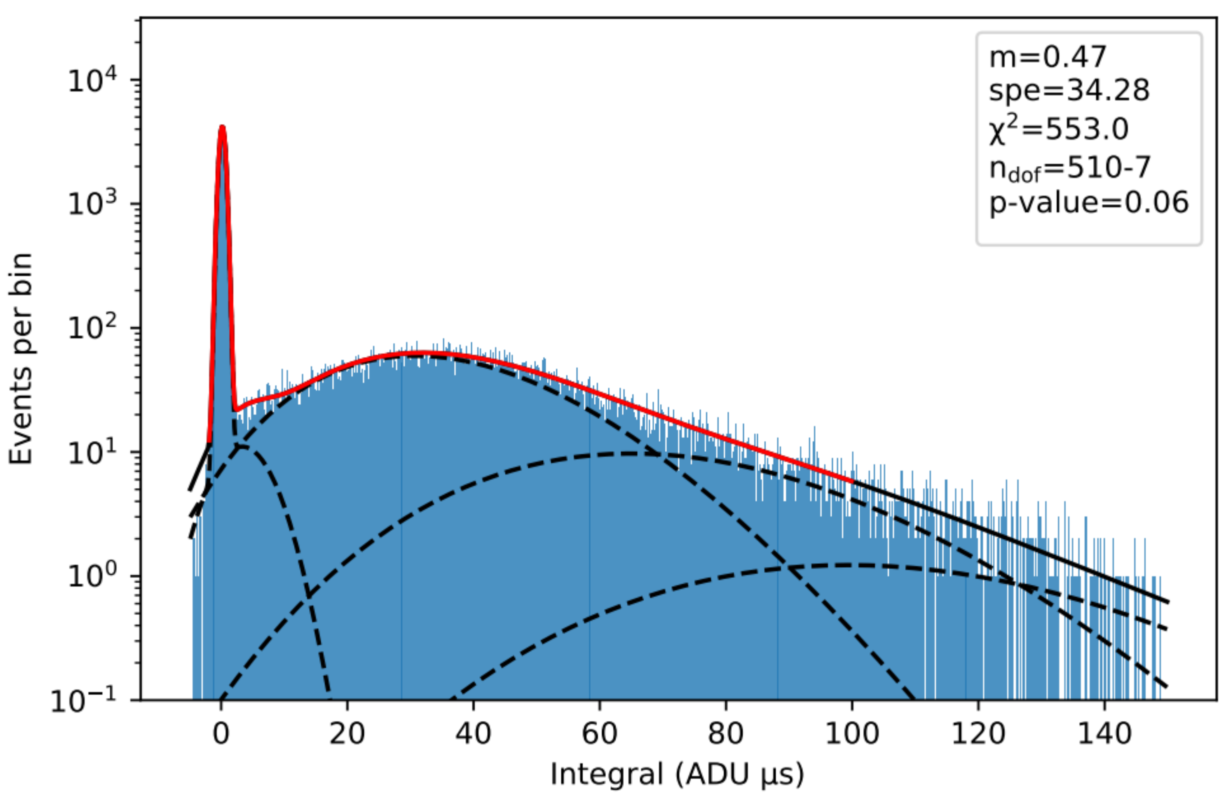
Pyrene Mechanism

- Adapted from: F. M. Winnik. **Photophysics of preassociated pyrenes in aqueous polymer solutions and in other organized media**. [7]
- Monomer emission occurs when an isolated pyrene molecule absorbs light, then de-excites radiatively.
- Dynamic excimer emission takes place when an exciton migrates from an excited monomer to a preassociated dimer, creating an excimer (on the scale of ~ns) which de-excites radiatively.
- Static excimer emission happens when a preassociated dimer is excited (on the scale of ~ps), then de-excites radiatively and due to their proximity, the dimer have perturbed absorption and excitation spectra.



Fitting Single Photoelectron Integral Distribution

- Integral distribution of acrylic luminescence at 300 K. Red curve is overall fit to model in fit range, solid black is overall model outside of fit range, dashed black curves are individual model components.
- Parameter m is the average number of photoelectrons per pulse
- spe is the average integral of a single photoelectron.
- The single photoelectron integral distribution from [7].



Fitting Multi-Photoelectron Integral Distribution

- Integral distribution of pyrene fluorescence shown at 300 K, 87 K and 4 K.
- 300 K data shown here fit with a skew normal distribution as the red solid curve as described in [8]
- Red curve is the skew normal fit to the distribution. The mean of the distribution is the light yield.