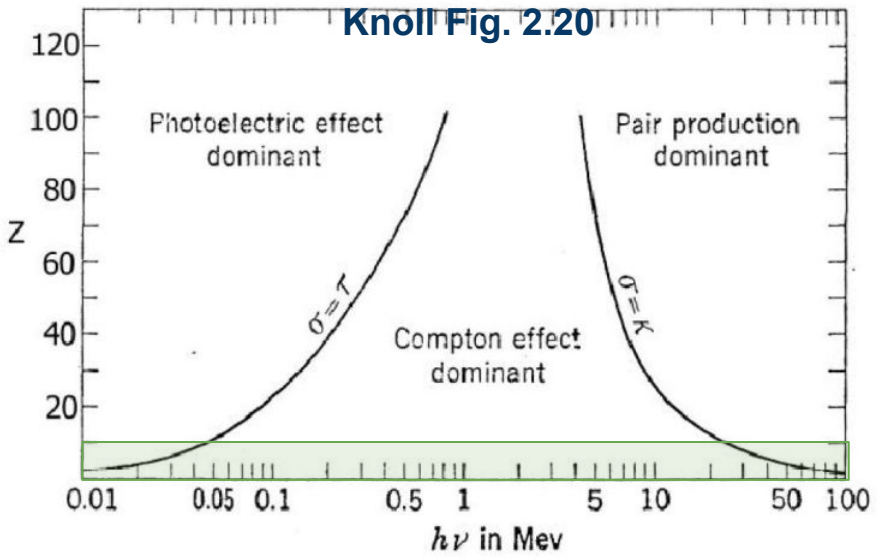
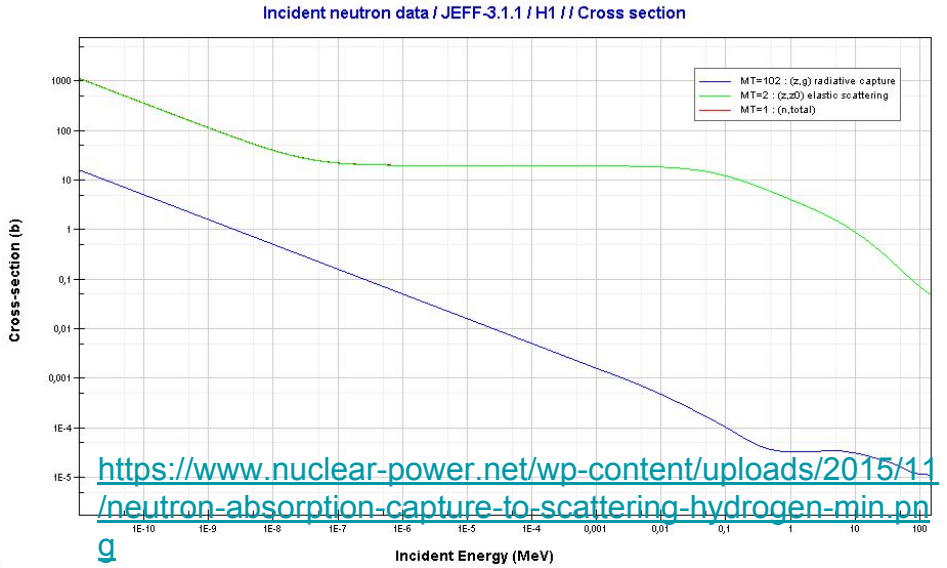
Organic Scintillation Detectors

- General Properties
  - Organic molecules
    - Low density, low Z
  - High Hydrogen content →sensitivity to fast ^{1}n via elastic scattering
  - Light output
    - Generally lower than inorganic scintillators
    - Large non-proportionality
      - Particle discrimination based on pulse shape (PSD)
  - Fast timing

Knoll Fig. 2.20

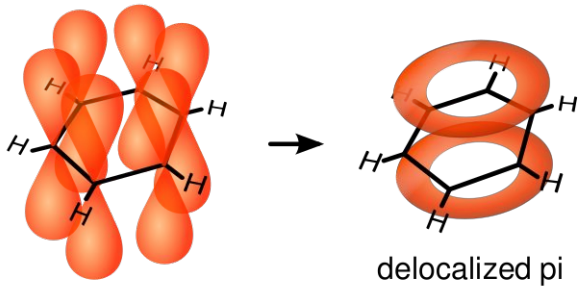




https://www.nuclear-power.net/wp-content/uploads/2015/11/neutron-absorption-capture-to-scattering-hydrogen-min.png

Excitation in Organics

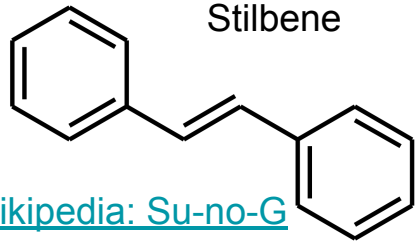
Wikipedia: Vladslinger



6 p_z orbitals

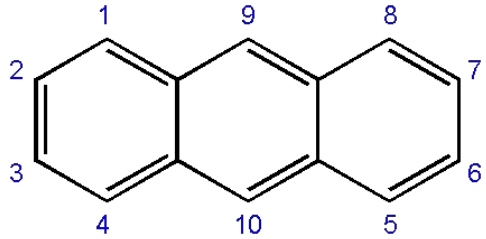
delocalized pi system

Stilbene



Wikipedia: Su-no-G

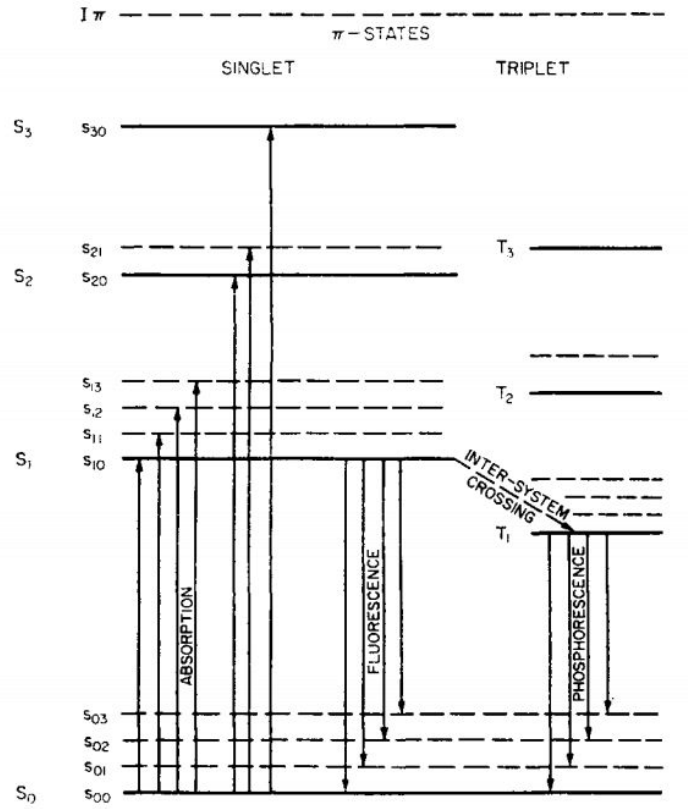
Anthracene



Wikipedia: Edgar181

p-terphenyl

Wikipedia: Edgar181



Knoll Fig 8.1 (from Birks)

- Orbital states result from **delocalized pi-bonds**
  - Transition between orbital states can result in luminescence (fluorescence, phosphoresence)

Luminescence in Organics

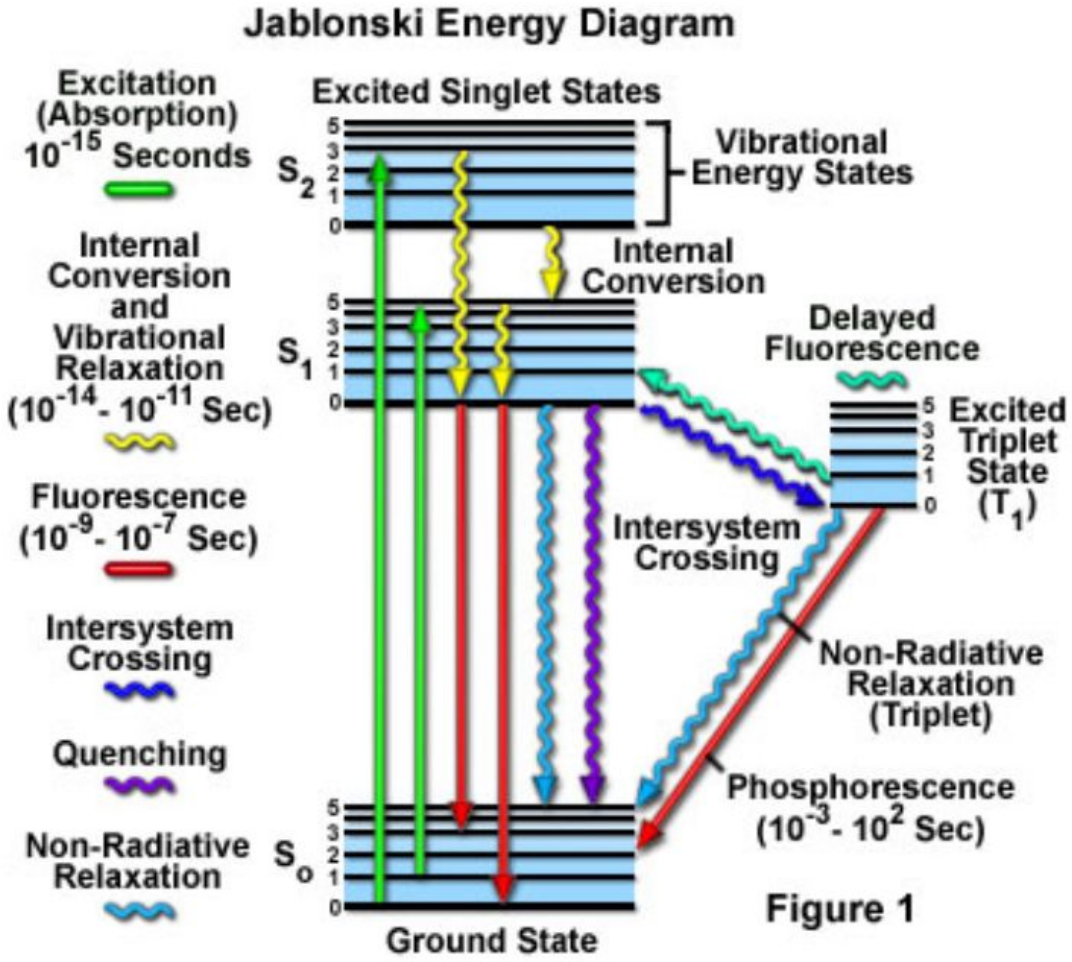


Figure 1

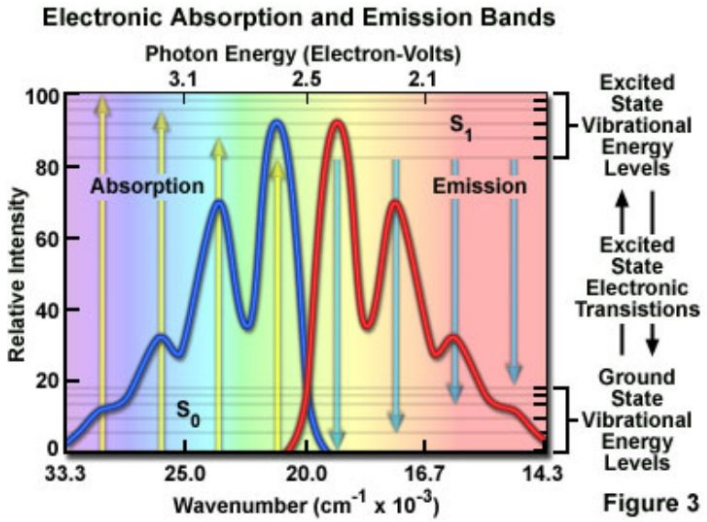
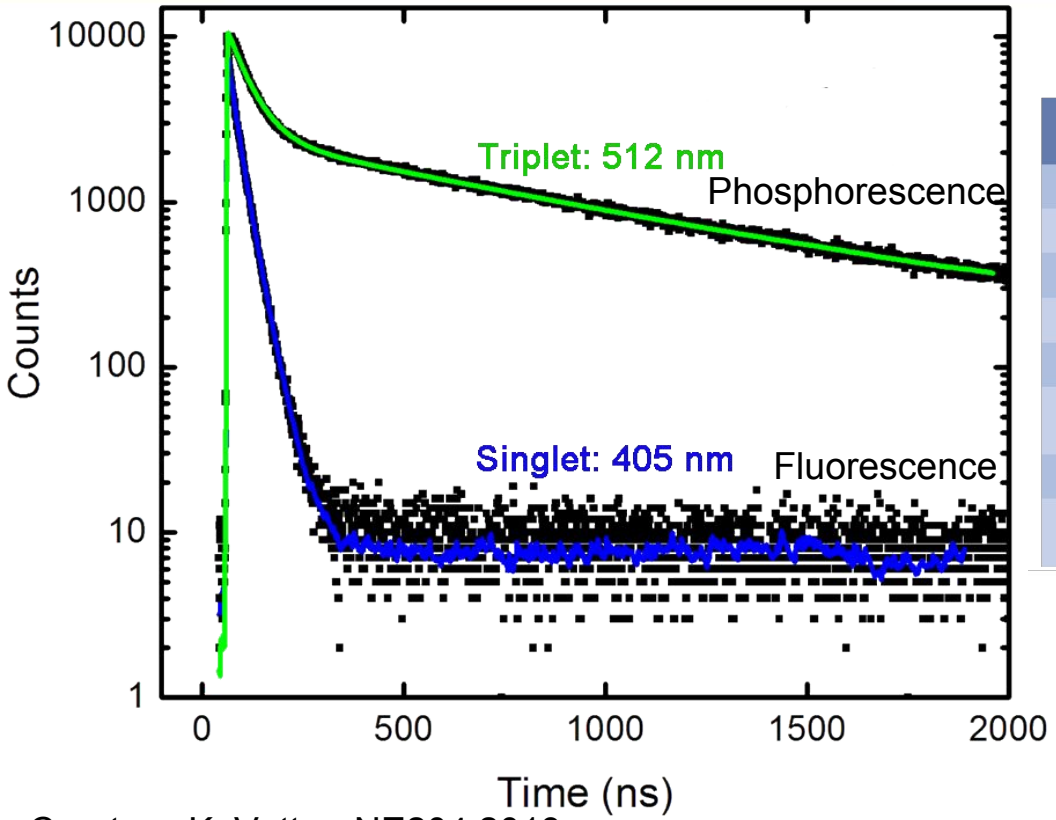


Figure 3

https://www.chem.uci.edu/~dmitryf/manuals/Fundamentals/Fluorescence%20Excitation%20and%20Emission%20Fundamentals.pdf

Luminescence in Organics



Courtesy K. Vetter, NE204 2013

Transition	Process	Rate Constant	Timescale (Seconds)
S(0) => S(1) or S(n)	Absorption (Excitation)	Instantaneous	10^{-15}
S(n) => S(1)	Internal Conversion	k(ic)	10^{-14} to 10^{-10}
S(1) => S(1)	Vibrational Relaxation	k(vr)	10^{-12} to 10^{-10}
S(1) => S(0)	Fluorescence	k(f) or Γ	10^{-9} to 10^{-7}
S(1) => T(1)	Intersystem Crossing	k(pT)	10^{-10} to 10^{-8}
S(1) => S(0)	Non-Radiative Relaxation Quenching	k(nr), k(q)	10^{-7} to 10^{-5}
T(1) => S(0)	Phosphorescence	k(p)	10^{-3} to 100
T(1) => S(0)	Non-Radiative Relaxation Quenching	k(nr), k(qT)	10^{-3} to 100

https://www.chem.uci.edu/~dmitryf/manuals/Fundamentals/Fluorescence%20Excitation%20and%20Emission%20Fundamentals.pdf

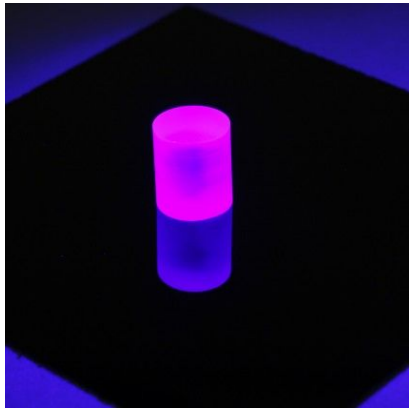
- Organic scintillator radiation detectors based on fluorescence
  - Phosphorescent component too slow for traditional pulse-mode operation

Organic Scintillators: Fundamental Processes

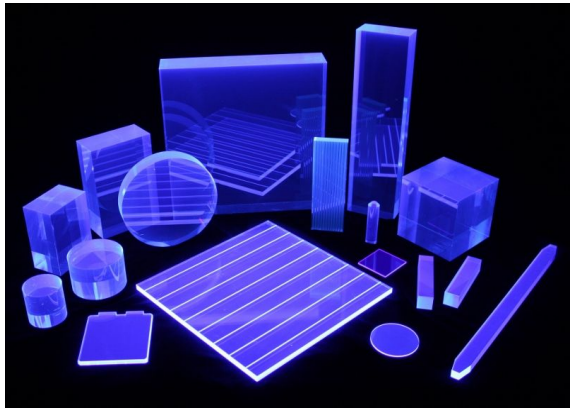
- Components needed for a organic-scintillation based radiation detector
  - Molecular energy states to convert energy deposition to scintillation
    - Favor radiative emission
    - Consider population of states & interaction of excited molecules → PSD
  - Efficient transfer of energy deposited via ionizing radiation to scintillation photons

Composition of Organic Scintillators

- Origin of scintillation mechanism → flexibility in scintillator materials
  - Does **NOT** require a pure crystalline structure
- Solid organic scintillators
  - Crystalline organics e.g. anthracene or stilbene
  - Plastics (scintillating dyes suspended in polymer)
- Liquid scintillators
  - Scintillating dyes suspended in organic solvents



LLNL Stilbene Crystal



Eljen plastics

Scintimax Liq. Scint.

Composition of Organic Scintillators

- Classification of Organic scintillators based on number/type of scintillating compounds they contain
- **Unitary compounds** e.g. pure monocrystals like anthracene or stilbene
- **Binary compounds:** inclusion of scintillating compound in organic solvent or polymer matrix
  - p-terphenyl in **liquid solvents** like xylene or toluene
  - p-terphenyl in styrene or PVT solvents → polymerization
- **Ternary compounds:** binary compounds that include a secondary solute (e.g. wavelength shifters)
  - E.g. 2,5-Diphenyloxazole (PPO)
- Ionizing radiation deposits energy in solute, emission from solvent - Efficient energy transfer required

Light Output

- Characteristics of light output depend on **energy** and **dE/dx** of deposition by charged particle
  - Scintillation efficiency (photons / MeV)
    - Total light liberated per deposited energy
    - Strong dE/dX dependence (type and density of excitation along track)
    - Typically degrades with increasing dE/dX
  - Timing characteristics of light curve
    - Fraction of prompt / delayed components of fluorescence also depends on dE/dX
    - Basis for particle identification based on timing characteristics of signal shape: **PSD**

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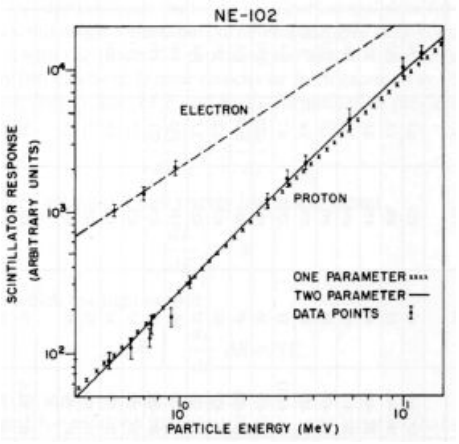
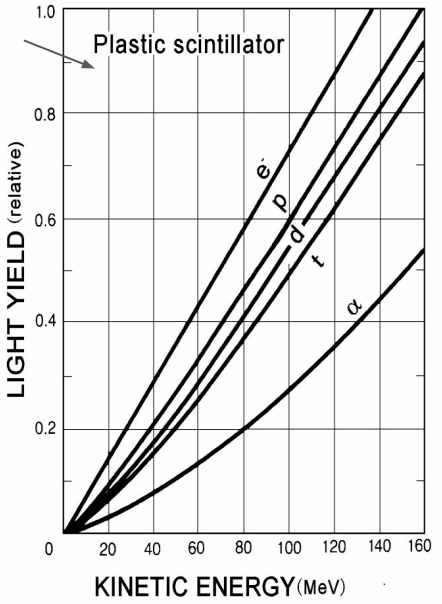
Scintillation Efficiency in Organic Scintillators

- Scintillation Eff. depends on particle type
  - e^- →linear resp >~100 keV
    - MIP → highest light yield
  - Describe light yield in terms of MeV_{ee} (electron equivalent)

M. Chen - fig derived from Gooding & Pugh



Knoll fig. 8.3



- Scintillation Eff. described by Birks' formula:

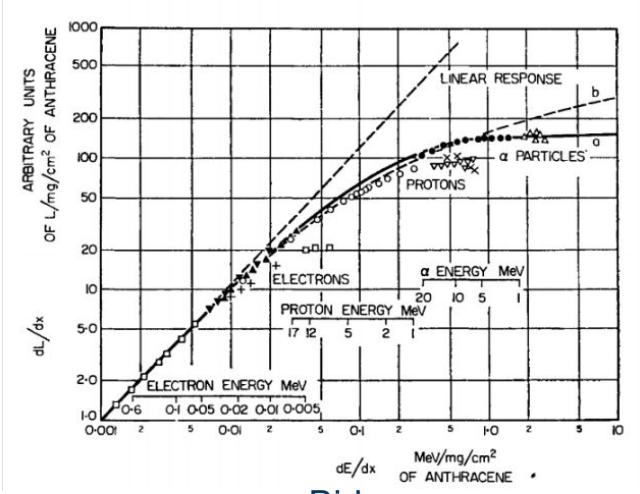
Fluorescence / unit path length

$$\frac{dL}{dx} = \frac{S\frac{dE}{dx}}{1+kB\frac{dE}{dx}}$$

Scint. Eff.

Density of damaged molecules

Quenching fraction



Birks

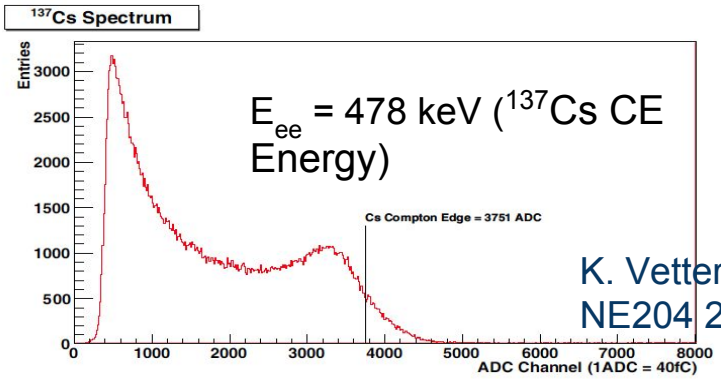
Light Output: MeV_{ee}

- Convert measured "energy" to energy deposited depending on particle type
- Some calibration models:
  - E_p = recoil proton energy, E_{ee} = energy associated with light prod

$$E_p^{UM} = 0.035E_{ee}^2 + 0.1424E_{ee} - 0.0362$$ Pozzi et. al. 2004

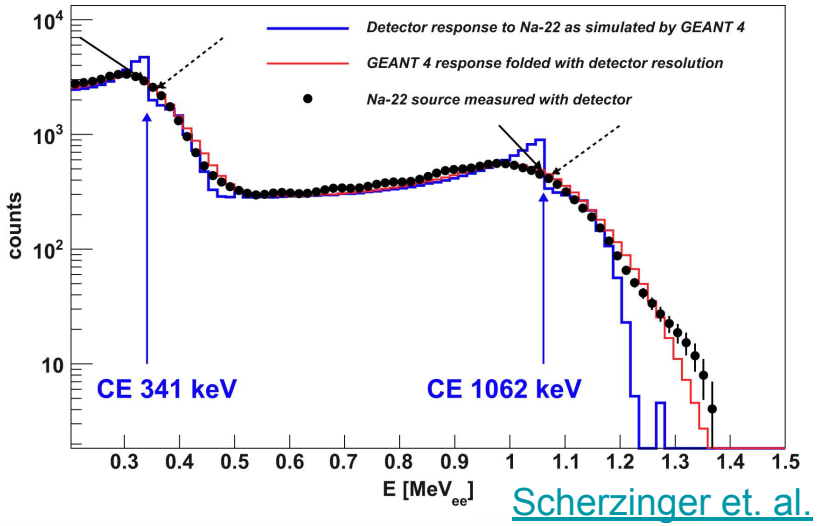
$$E_p^{Bachelor} = 0.028E_{ee}^2 + 0.215E_{ee}$$ Batchelor et. al. 1961

E [illegible] dge



E_{ee} = 478 keV (^{137}Cs CE Energy)

K. Vetter/P. Marleau NE204 2013



Scherzinger et. al.

Light Output

- Characteristics of light output depend on **energy** and **dE/dx** of deposition by charged particle
  - Scintillation efficiency (photons / MeV)
    - Total light liberated per deposited energy
    - Strong dE/dX dependence (type and density of excitation along track)
    - Typically degrades with increasing dE/dX
  - Timing characteristics of light curve
    - Fraction of prompt / delayed components of fluorescence also depends on dE/dX
    - Basis for particle identification based on timing characteristics of signal shape: **PSD**

Timing Components of Light Output

- 3 Main components to scint. Emission
  - Prompt fluorescence ($S_1 \rightarrow S_0$)
  - Delayed fluorescence ($T_1 \rightarrow S_1 \rightarrow S_0$)
  - Phosphorescence ($T_1 \rightarrow S_0$)

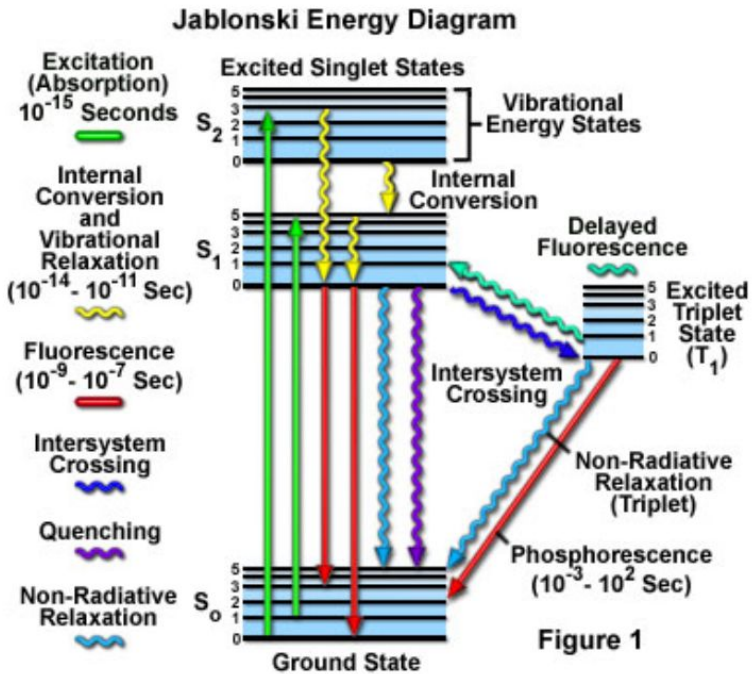
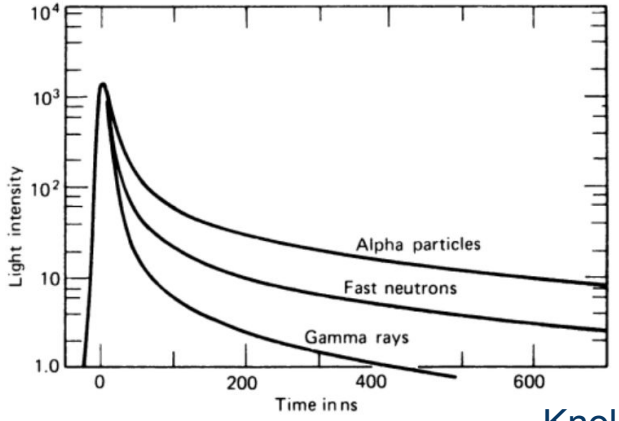


Figure 1

- Fraction of Prompt/Delayed fluorescence depends on excitation type/density
  - E.g. Triplet annihilation:
    - $T_1 + T_1 \rightarrow S_1 + S_0$; $S_1 \rightarrow S_0 + h\nu$



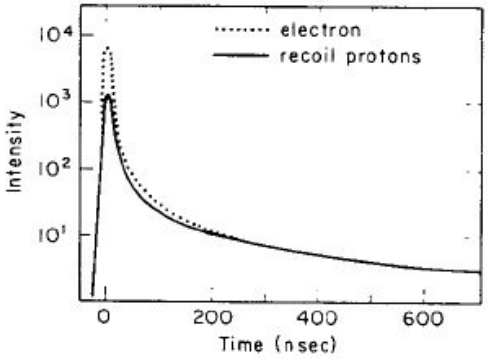
Knoll fig. 8.5

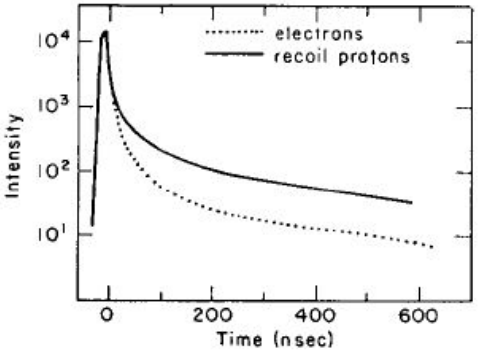
Delayed Fluorescence and Quenching

- Key points
  - Initial Singlet/Triplet population depends on density of states
  - Delayed fluorescence has same emission spectrum as prompt fluorescence, just delayed by triplet-annihilation process
    - Intensity of delayed fluorescence depends on concentration & diffusion of triplet excitons
    - Triplet & Singlet quenching processes have different dependence on dE/dx (triplet quenching less sensitive)
- Relative integrated intensities of prompt/delayed components therefore depends on dE/dx, thus particle type
  - Basis for particle identification via PSD

Pulse Shape Discrimination in Organic Scintillators

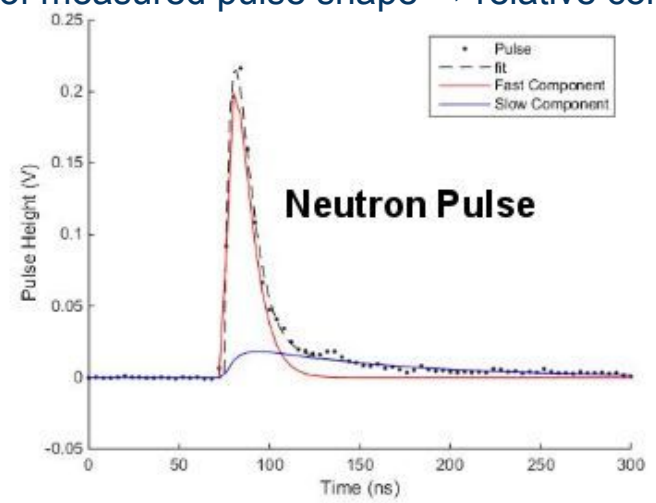
- We will focus on n,γ discrimination (cf. lectures from Drs. Marleau & Brubaker for motivation)

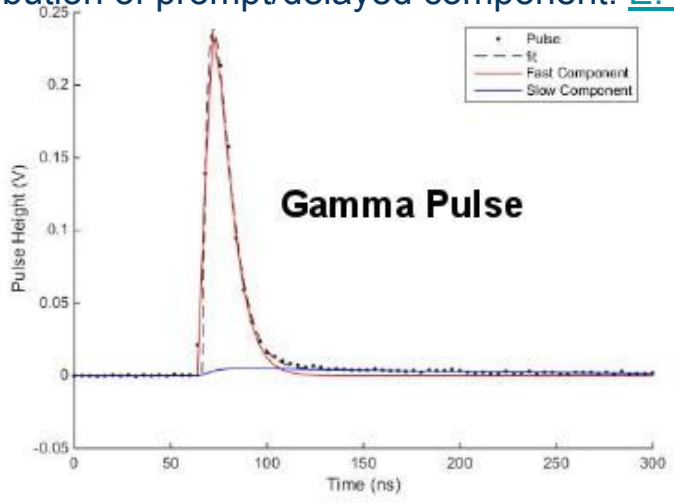




K. Vetter/P. Marleau
NE204 2013

Decomposition of measured pulse shape → relative contribution of prompt/delayed component. E. King et. al.



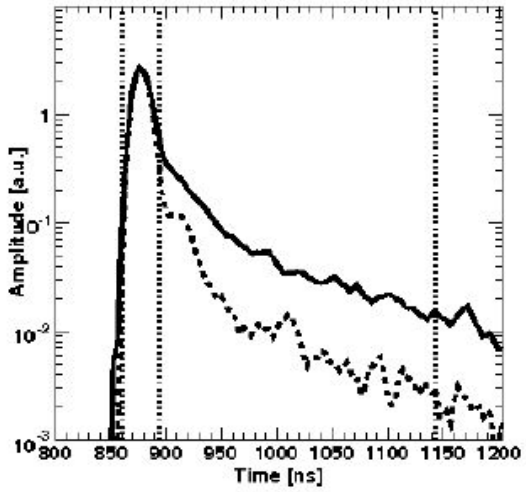


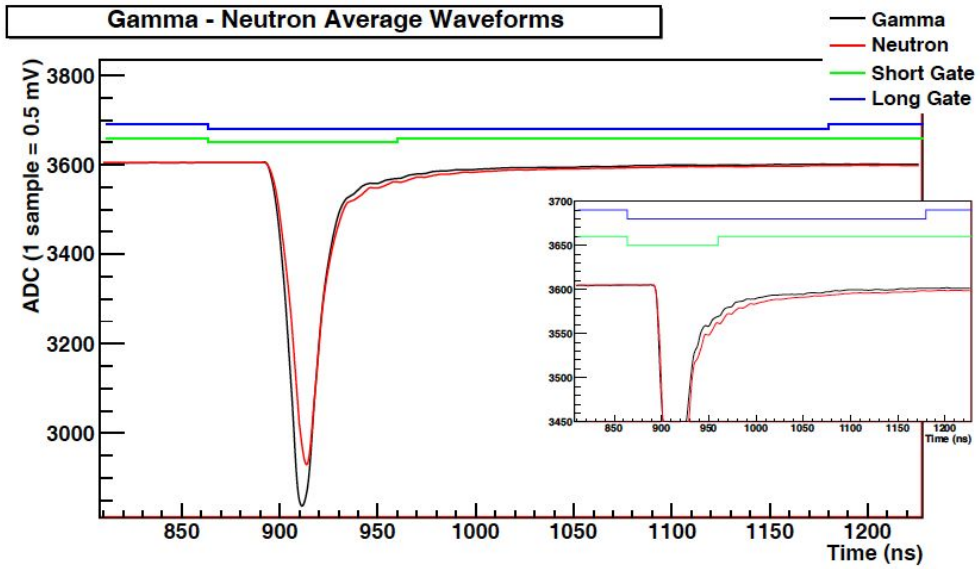
PSD Methods

- Integration windows applied to voltage signal
  - E.g. Prompt-to-tail or Tail-to-total ratio
- Risetime of integrated signal
- Pulse shape analysis
  - Template matching
  - Decomposition

PSD Methods

- Integration windows applied to voltage signal
  - E.g. PSD metric = Prompt-to-tail or Tail-to-total ratio





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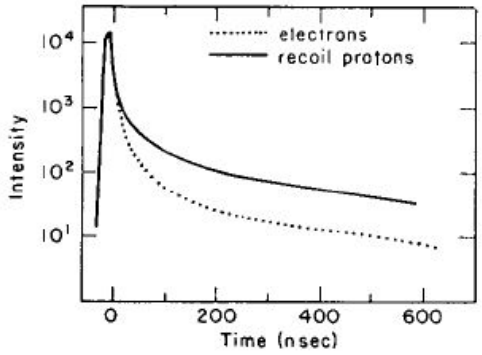
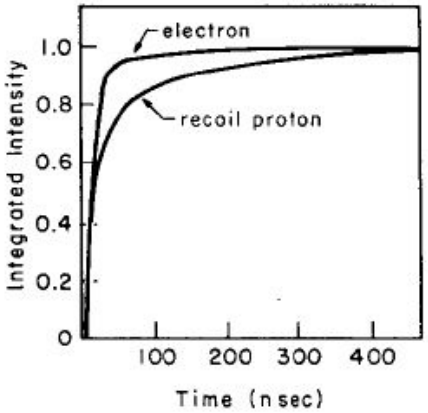
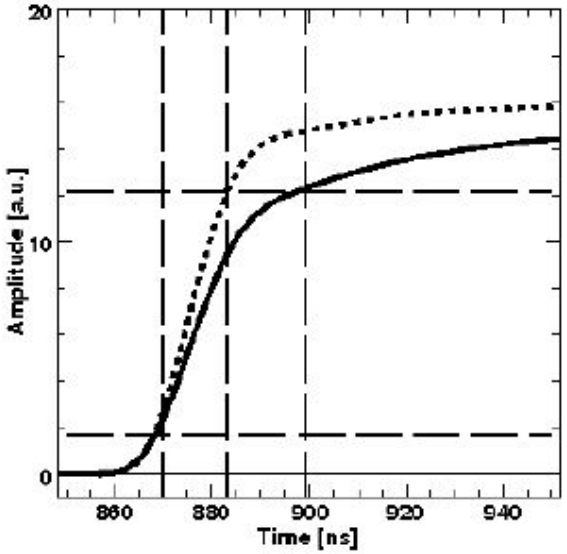
PSD Methods

- Risetime of integrated signal



Integrator





Derive PSD metric based on timing of integrated signal (e.g. t_{70}/t_{10} or t_{90}/t_{50})

K. Vetter/P. Marleau NE204 2013

PSD Methods

- Pulse shape analysis
  - Template matching, e.g. Gatti & Martini method

Starts by leading edge

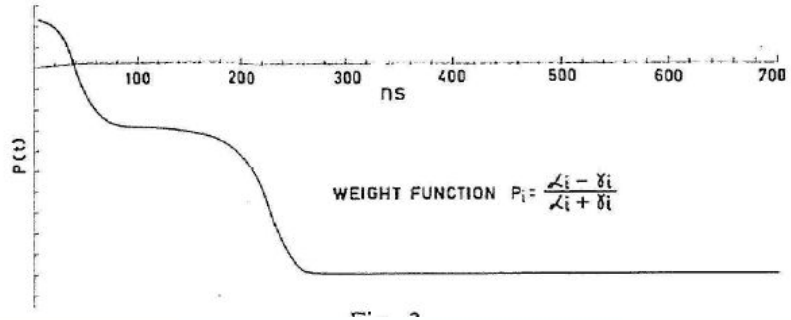
Integrate the entire pulse with a suitable weight

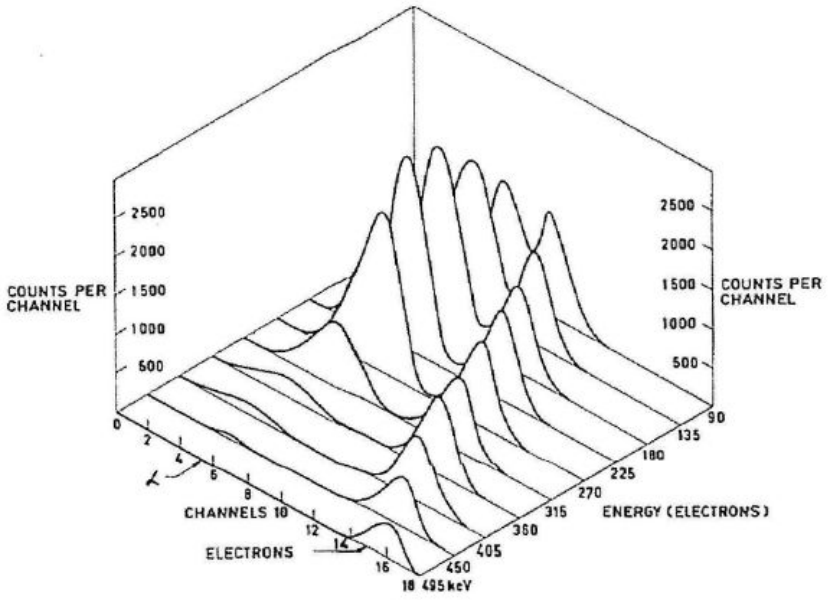
$$S = \int_0^T p(t)\,w(t)\,dt$$

The optimal weight can be shown to be

$$w(t) = (n(t) - \gamma(t))/(n(t) + \gamma(t))$$

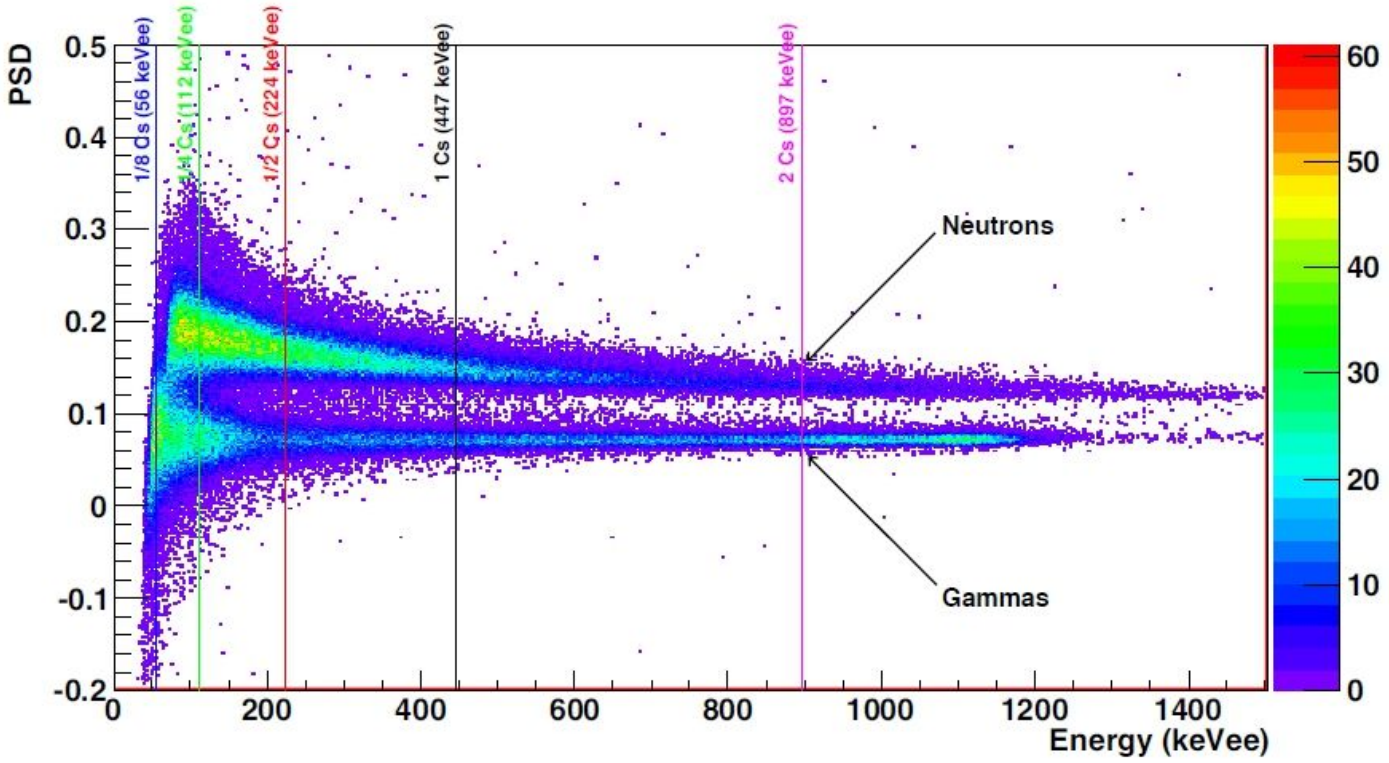
E. Gatti, F. de Martini, in: Nuclear Electronics, Proceedings of International Conference at Belgrade, Vol. II, IAEA, Vienna, 1962, p. 265.





PSD Plot

- 2D frequency distribution of PSD values vs. energy



2D plot of Energy versus PSD using an AmBe source at 2 kcounts/s; the two lobes of the neutrons and gammas are well separated. The data were acquired at Duke University (TUNL).

Image courtesy CAEN digitizer webpage

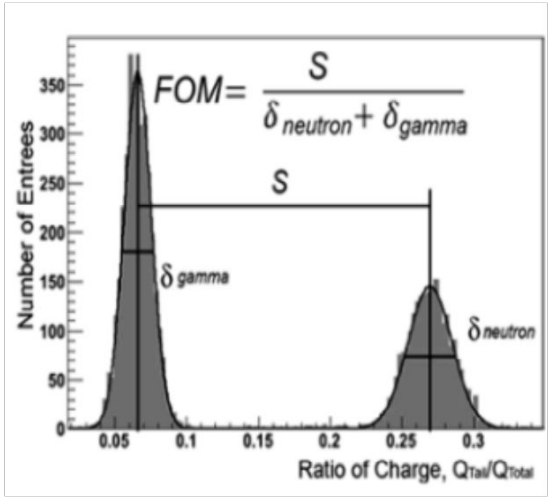
Quantifying PSD

- Evaluate PSD performance vs. MeV_{ee} with Figure Of Merit (FOM)

$$FOM = \frac{S}{\delta_{neutron} + \delta_{gamma}} \qquad \text{Equation 1}$$

$S \equiv$ the distance between the gamma ray and neutron peaks

$\delta \equiv$ the full width at half maximum (FWHM) of the peaks.



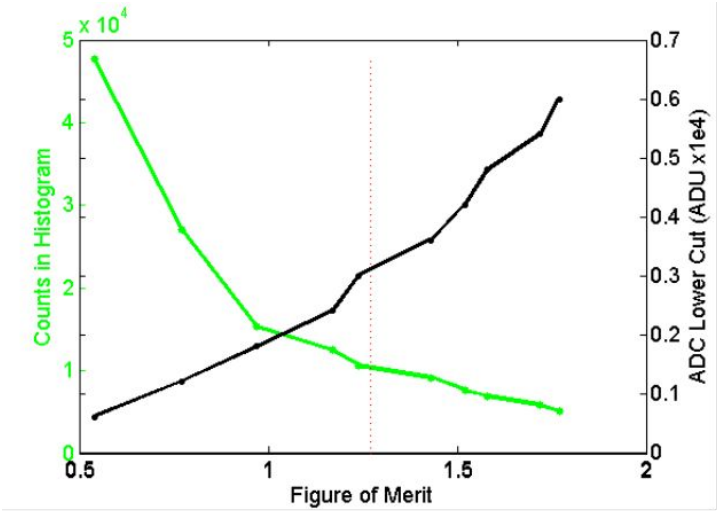


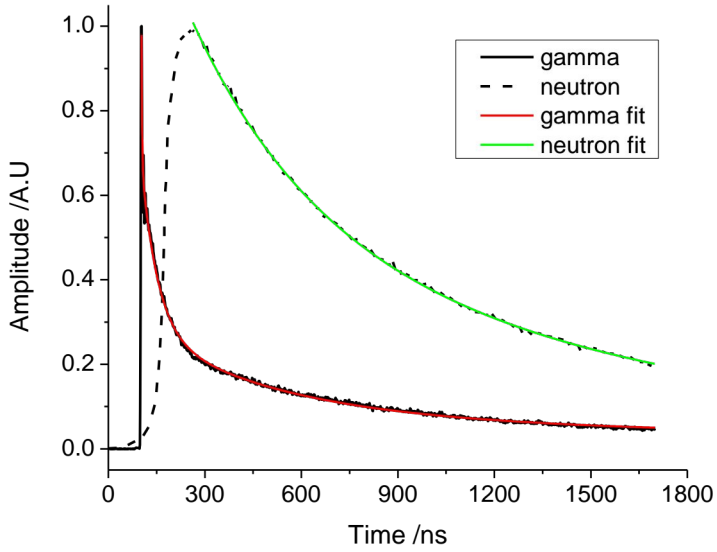
Figure 1: Definition of the FOM for gamma ray-neutron separation based on PSD (adapted from Figure 1, "Pulse Shape Discrimination in Impure and Mixed Single-Crystal Organic Scintillators," N. Zaitseva et al., 2011, *IEEE Transactions on Nuclear Science, 58:6,* p. 3411).

Images from this nice overview from PNNL

Aside: n,γ PSD with Elpasolites

- Elpasolites: new(ish) type of **inorganic** scintillator
  - Commercially "available" examples: CLLB(Ce), CLYC(Ce)
  - Some exhibit excellent PSD capabilities, e.g. **CLYC**:
    - Based on variations in scintillation mechanisms

Types	CVL /ns	Ce^{3+} /ns	V_k /ns	STE /ns
Gamma-ray	2	50	420	3400
Neutron	-	-	390	1500



Kui-nan et. al Characterization of CLYC

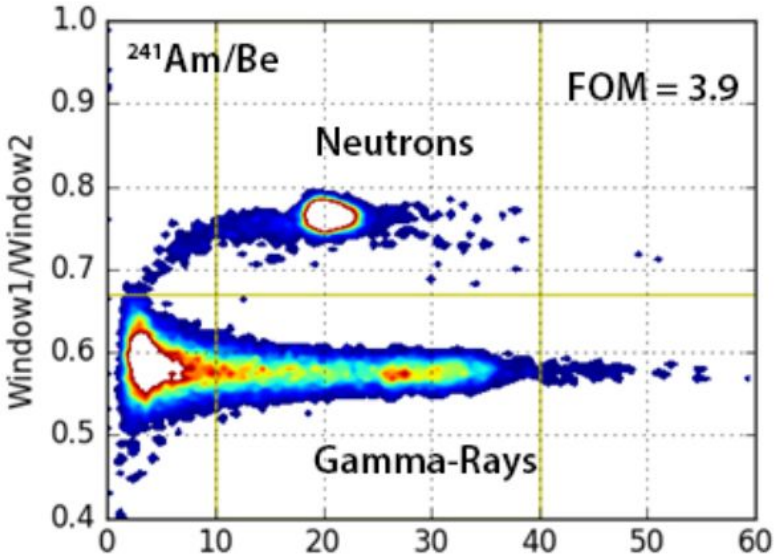


Figure from RMD website

Loaded Organics

- Can “load” liquid & plastic scintillators with certain elements to add or enhance certain capabilities
  - Load with high concentrations of dyes to enhance PSD capabilities (e.g. PSD plastics)
  - Load with high-Z (Pb, Tin) to enhance PE-abs. for low energy gamma-ray spectroscopy
  - Load with ^{10}B or ^{6}Li to enhance thermal neutron sensitivity

Example: High-Z loaded Plastics

- Example dopants: Sn, Pb, Bi, Gd
- Enhance PE absorption probability for low-energy gamma-rays
  - Potentially even get full-energy peaks!
- Tradeoff: Scintillation eff. tends to decrease with increasing wt% high-Z material
- Commercially available (e.g. EJ-256)

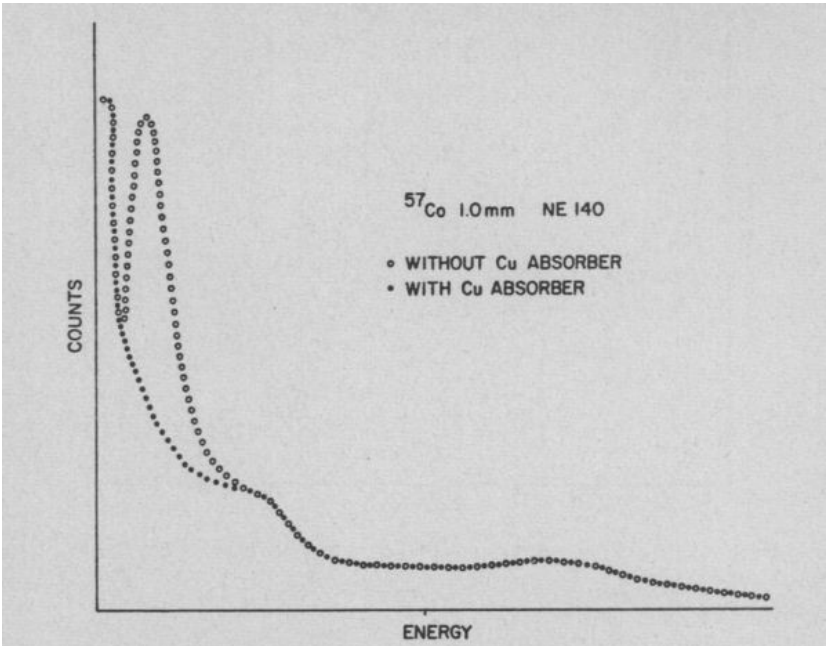
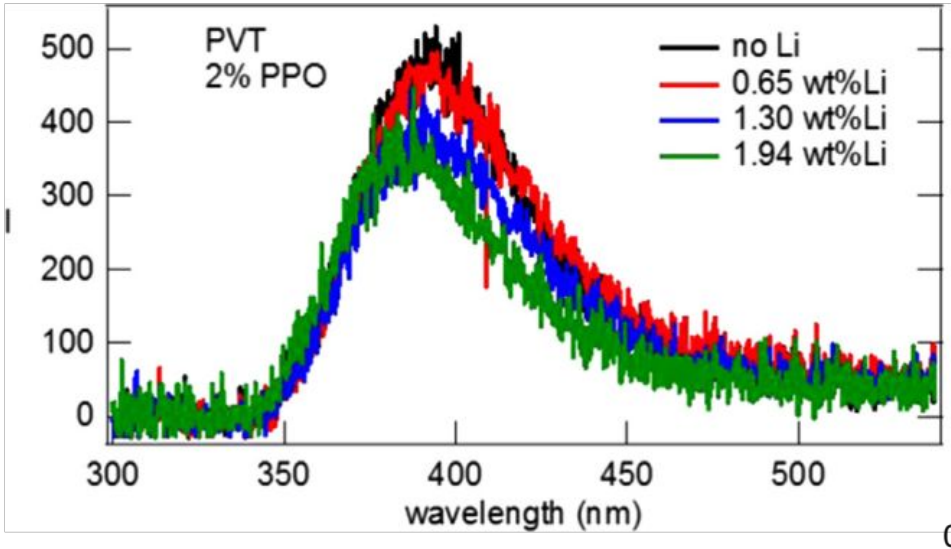


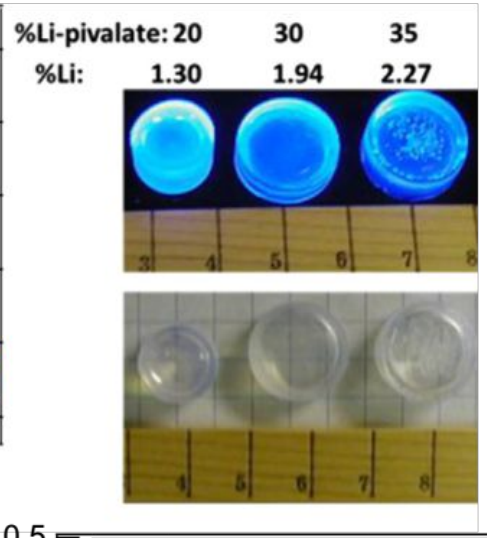
Fig. 10. Energy spectrum obtained with a 20(dia) x 1 mm NE 140(5% Sn) scintillator for 14.4 and 122 keV (^{57}Co) with and without a 0.1 mm Cu absorber.

Cho et. al. 1975

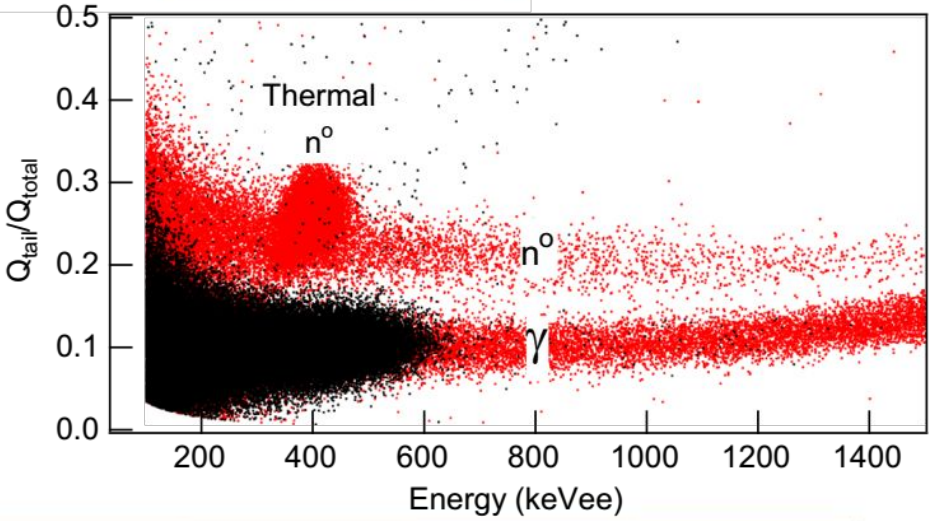
Example: Li-Loaded Plastics

- Load plastic with Li to enhance response to thermal neutrons



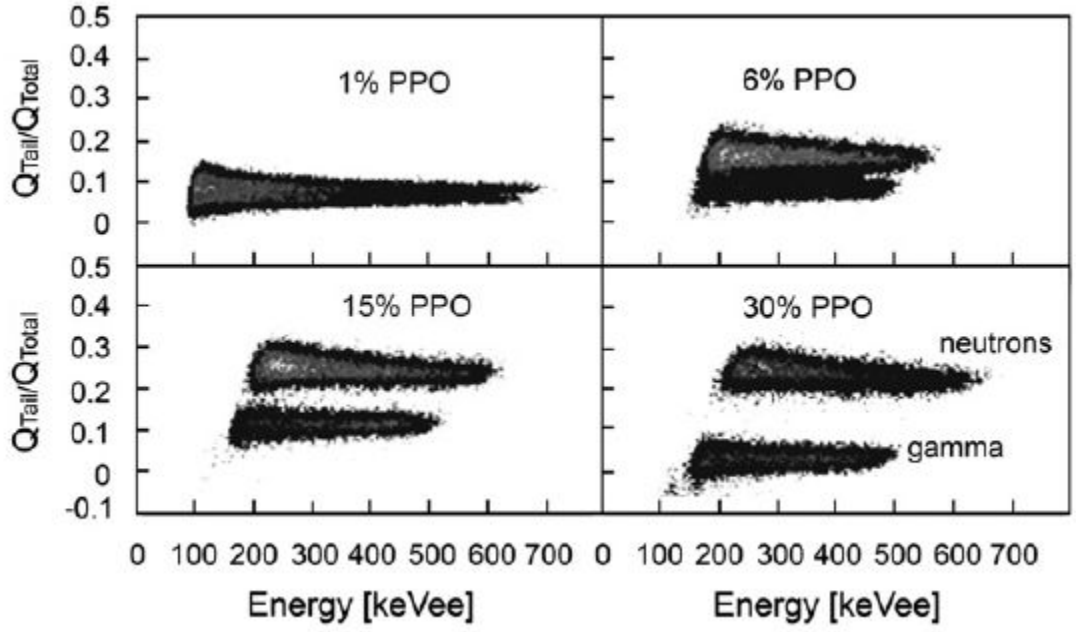


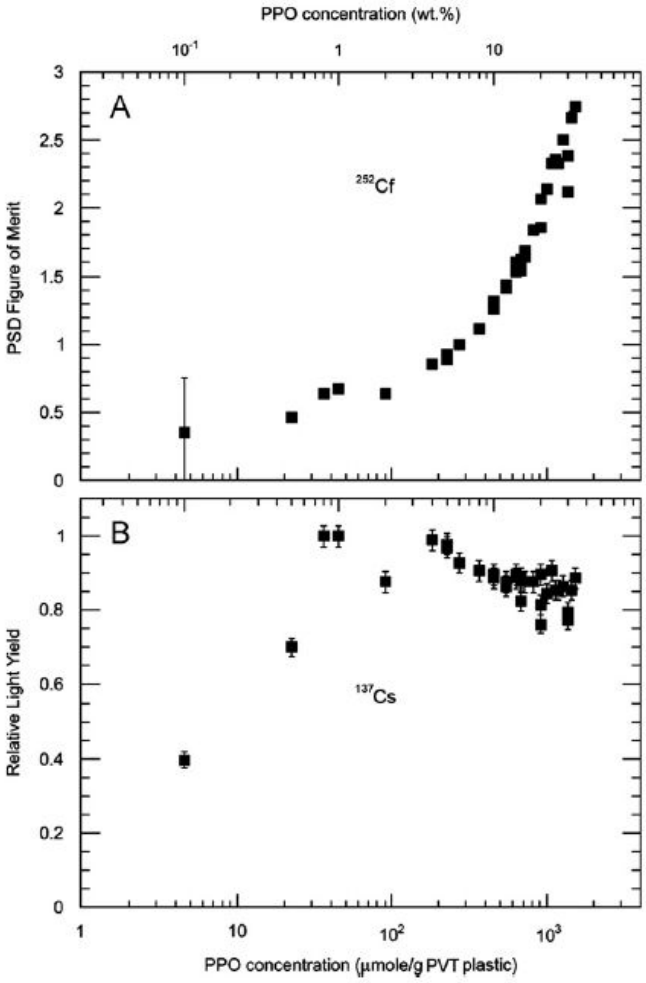
Images from [Cherepy et. al. 2018](#) (N.B. Bi-loaded plastics also discussed in this paper)



Example: PSD-Enhanced Plastics

- Increased dye-loading to enhance PSD capabilities





Images from [Zaitseva et. al. 2012]

Organic Scintillator Sources

- One advantage of organic scintillators is that they tend to be *relatively* cheap
- Some providers (with links to their product pages) include:
  - Eljen, St. Gobain, Scintimax, etc.