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BASIC LIMITATION OF SCINTILLATION COUNTERS IN TIME MEASUREMENTS

by

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BASIC LIMITATION OF SCINTILLATION COUNTERS IN TIME MEASUREMENTS*

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SUMMARY

The time resolution of a scintillation detector can be calculated from the scintillator illumination function, and the response of the photomultiplier to an instantaneous pulse of light. Such calculations using the parameters of present scintillators and photomultipliers agree with measured resolutions and predict improvements which can be achieved. Parameters of commercial and some promising experimental fast scintillators are given, as well as measurements on transit time spread of some fast photomultipliers.

Although scintillation counters have been supplanted to a large extent by semi-conductor detectors, they still have important applications which derive from their greater variety in size, shape and constitution, and the essentially noise-free amplification of the photomultiplier. Furthermore, compared to semiconductors they are less easily damaged and, at present, are less expensive. In time measurements, the noise-free character of the amplifier more than compensates for the smaller number of charge carriers produced, and results in superior time resolution for γ rays and β particles below a few MeV.

This paper considers the basic limitation on time resolution imposed by the characteristics of scintillator and photomultiplier. Much effort has been expended in the last 20 years to understand how the scintillator and photomultiplier affect the time resolution. Such information serves to guide the experimenter in the choice of scintillator and photomultiplier and in achieving optimum operating conditions. In addition, it helps him to know when he has approached the theoretical limit of time resolution.

In 1950 Post and Schiff[1] first discussed the limitations on resolving time that arise from the statistics of photon detection. They assumed that following excitation of the scintillator by an energetic event, the photomultiplier amplifies the primary photo-electrons without time spread, and that the resulting output pulses are fed into a discriminator that detects when when a definite number of pulses, say n, have accumulated. From Poisson statistics, they showed that the probability that the nth pulse occurs between t and t + dt is

$$P_n((t)dt = e^{-RF(t)} \frac{[RF(t)]^{n-1}}{(n-1)!} RF'(t)dt \qquad (1)$$

where RF(t) is the average number of photoelectrons detected in the interval between 0 and t, following the exciting event at t = 0, F(t) = dF/dt, and RF(∞) = R, the average number of photoelectrons detected per event. Although they assumed that there was no transit time spread in the photomultiplier, to include this factor requires only that F'(t) be redefined so the photomultiplier transit time spread, P(t), is folded in with the photocathode illumination function I(t), i. e.

$$F'(t) = P(t) * I(t) \qquad (2)$$

where I(t) is the probability density function, PDF, of arrival times of photons at the photocathode produced by an event at t = 0, and P(t) is the PDF of the time of detecting single electrons when an instantaneous flash of light weakly illuminates the cathode at t = 0, i. e., for I(t) = δ(t).

More elaborate calculations by Gatti and Svelto,[2] Hyman[3] and others[4] included the single electron response of the anode current, its variance in width, and the statistical variance in gain. When realistic assumptions were made concerning the transit time spread, all calculations indicated that there was a specific fraction, n/R, of the total pulse height which gave the best time resolution. This fraction varied from < 0.01 for NaI(Tl) to 0.3 for a fast organic scintillator. In agreement, experiment also demonstrated that there was an optimum fraction. However, attempts to make theoretical predictions fit experiment have been impeded by the lack of accurate information concerning the scintillator illumination function, I(t), the average number of photoelectrons per flash, R, and the function, P(t), which describes the overall photomultiplier discriminator response.

An example of the accuracy with which the performance of a scintillation counter can be predicted from the measured light intensity curve $F_{exp}(t)$ was given in a paper on pulse shape discrimination.[5] The left side of Fig. 1 shows the time dependence of the intensity of the scintillation light from Stilbene and 3 liquid scintillators excited by gamma rays and by neutrons as measured with the single photon sampling technique of Bollinger.[6] These curves are experimental values of the function F'(t) of Eq. 2. The curves at the right side of Fig. 1 represent integrals of the light-intensity curves plotted on a linear scale and normalized to 1 at t = 500 nsec., and correspond to F(t). The difference in scintillations caused by neutrons and gamma rays could be determined from a measurement of the time for the pulse to rise to some large fraction of its final value, with the aid of a fixed fraction discriminator.

In addition to the decay curve of the scintillator, it was necessary to know the photo electrons/keV, P, obtained with the photomultiplier used. P was measured using the method of G. C. Kelley et al.[7] For calculating the distribution rise times to a fixed fraction of the final pulse height, Eq. (1) of Post and Schiff is no appropriate since it assumes a normal distribution of electrons around the mean value R. If the number of electrons arriving in time T is to be exactly R, it can be shown[5] that the probability that the nth electron arrives between t and t + td is

$$P_{n,R}(t)dt = \frac{e^{-R}R^{R}[1-F(t)]^{R-n}[F(t)]^{n-1}F'(t)dt}{(n-1)!\ (R-n)!} \qquad (3)$$

for a fixed fraction k, n = kR. [Equation (3) differs little from Eq. (1) at small fractions, but at large fractions, the difference is marked.] Figure 2 shows the measured rise time distribution for neutrons of 350, 500 and 750 keV and for gamma rays producing integrated pulses of the same height. The solid curves are calculated by substituting the measured values of F(t) and R into Eq. (3). As can be seen from Fig. 2 the experimental data fits the calculated curves closely. The only adjustment of parameter was that the fraction 0.895 was used since this gave a slightly better fit than the circuit design value, 0.85.

The rise and decay of the intensity of the fast component of scintillation light can be represented by $I(t) = (e^{-t/\tau_1} e^{-t/\tau_2})/(\tau_1-\tau_2)$ accurately for binary solution scintillators, and approximately for trinary solution scintillators. When the illumination function of an organic scintillator is measured using the photon sampling technique, it can be fitted by F'(t) in Eq. (2) after the slow components have been subtracted. F'(t) is evaluated from the convolution integral

$$F'(T) = \int_0^T P(t)[e^{(t-T)/\tau_1} - e^{(t-T)/\tau_2}]dt/(\tau_1-\tau_2) \qquad (4)$$

where τ_1 is the mean life for energy transfer from the bulk material to the fluor (in case of binary solution) and τ_2 is the mean life for decay of the fluoresence. P(t) is the instrument response function for Čerenkov light measured with the method of Kirkbride et al.[3] An example of a previously published measurement[9] for a solution of a substituted quaterphenyl[10] P4-G12, with τ_1 = 0.1 usec and τ_2 = 1.3 nsec. is shown in Fig. 3.

The measured instrument response function P(t) (shown in Ref. 5) could be approximated by a Gaussian function with σ = 0.4 nsec. Thus, it is realistic to substitute for P(t) a truncated Gaussian function in the manner of Hyman et al.[3]. Then Eq. (4) can be evaluated for any set of parameters τ_1, τ_2, σ and P. From Eq. (4) and its integral, the time resolution curve can be calculated using Eq. (3). The results of some of these calculations are presented in Fig. 4 and 5. Figure 4 shows the variation of full width at half maximum, FWHM, and the slope, $T_{1/2}$, with σ, the standard deviation of transit time spread; for parameters of NaI(Tl), P4-G12,[10] and Naton-136. For comparison with experiment, the asterisks show the best resolutions measured for NaI(Tl) and Pf-G12 with the RCA-8575. As can be seen from the slopes of the curves, substantial improvements in time resolution can be obtained with faster photomultipliers. The upper curves in Fig. 5 show calculations on the effects of varying τ_1 and τ_2 assuming σ = 0.6 nsec. The bottom graph of Fig. 5 shows the dependence of optimum fraction on σ, and its lack of independence on σ_1 (in this range of values).

It should be noted that these calculations apply to small scintillators where variation in light collection time and absorption are negligible.

Accurate information on the scintillation light intensity as a function of time is important to scintillator research and development, as well as to the estimation of time resolution. Table 1 shows a comparison of measurements of the mean lives for decay of the fast component of some commercial organic scintillators by 7 different experimenters using the single photon sampling technique of Bollinger and Thomas.[6] Not included in the table are the measurements of Lyons and Stevens[11] who reported their results on NE-111 and Pilot U in FWHM and 10 to 90% rise time, T_r, as follows:

Scintillator	FWHM	T_r
NE-111	1.23 nsec	0.35 nsec
Pilot U	1.8 nsec	0.5 nsec.

Their measurements were made with a 50 psec electron or bremstrahlen pulse, a vacuum photodiode and a sampling oscilloscope. They found the FWHM of NE-211 is about 2/3 that of Pilot U, whereas T. M. Kelley et al.[19] found the decay mean life shorter for Pilot U than for NE-211.

Although the constitution of some scintillators may vary from batch to batch as is certainly the case here with NE 111, there is evidence that differing techniques of measurement and reduction of data in various laboratories lead in some cases to poor agreement. For example, individual experimenters find Naton 136 and Pilot B to have the same mean life, but the highest value is 50% greater than the lowest value reported.

In making scintillation decay measurements, it is important to use small samples so that errors introduced by light collection time[11] and self absorption[12] are negligible. Also, corrections should be made for dead time and non-linearity of the time scale. After subtraction of the longer components, the residual fast component may be fitted with Eq. (4) using the measured system response function to obtain the best values of mean lives for transfer and decay.

With care, such time constants should be reproducible between different experimenters to within 0.1 nsec. The effect of self absorption and reemission on the scintillator mean life also should be evaluated as shown by Sipp and Miehe.[12] They found that the mean life for decay of a substituted quaterphenyl[10] in Xylene increased from 1.28 to 1.94 nsec when the scintillation light passed through 5 cm of the scintillator. If the fluorescence is efficient, there may be very little reduction in light associated with the increase in decay time, but the time resolution will be worsened.

In addition to the decay time constants of the scintillator, it is important to know the photoelectrons/keV, P. Which will be obtained with the photomultiplier to be used. It is common that values of P in time resolution calculations are estimated to

be unrealistically low. Although measured values of 1—2.5 electrons per keV for organic scintillators and 8—10 electrons per keV for NaI(Tl) have been reported by Bollinger,[6] Lynch,[9], Houdayer,[7] and others, much lower estimates of P continue to be found in the literature.

P can readily be determined using the method of G. C. Kelley et al.[7] in which the resolution obtained with a precision light pulser is used to estimate the average number of photoelectrons assuming Poisson statistics. A more direct measurement can be made of the number of electrons in a pulse by comparison with the average pulse height for a single electron when a photomultiplier with a high gain first dynode is being used. Houdayer, Mark and Bell connected the photomultiplier as a photodiode and measured the total charge with a charge sensitive amplifier.

There are certainly unrealized possibilities for better organic scintillators. Many molecules that have high fluorescence efficiency and short decay times but were relatively insoluble have been made soluble by substitution of alkyl or oxa-alkyl groups for hydrogen atoms at suitable locations in the molecule.[14] Although the substitution has only a small effect on the absorption and fluorescence of the molecule, there is usually some increase in stokes loss (the shift of the emission band away from the absorption band toward longer wave lengths). Also the substituted solutes exhibit less self quenching. At higher concentrations that become usable, the time for energy transfer from solvent to solute are shorter.

Among the solutes to which this solubilization technique has been applied, the p-oligophenylene series is particularly promising.[14] It has been shown that the decay time varies inversely with the number of phenyl rings in the molecule. In addition, the light output has been shown to be larger for the longer molecules.

Table 2 shows the results of measurements[9] of the time constants τ_1 and τ_2, and P, the number of photoelectrons/keV reported at the 11th SSCS. The materials were provided by H. Wirth in experimental quantities and, unfortunately, with the exception of QP-G12, have not become commercially available. Although these fluors look promising, to evaluate them as practical scintillators requires larger quantities than are available. When one considers that the conversion efficiency of the best organic scintillators is only about 3% there would appear to be room for improvement by further investigation of solvents as well as fluors.

Single electron transit time spread in photomultipliers can be measured in several ways. The method of Kirkbride[8] employs single photon sampling of Čerenkov light excited by ^{60}Co γ rays which in coincidence excite a fast plastic scintillator coupled to a second photomultiplier. Typical response functions are from 0.7—1.0 nsec FWHM for RCA 8850 and 8575 photomultipliers. Measurements of transit time spread by Lescover and Lo[15] using a GaP light emitting diode with a 200 psec current pulse in the backward direction yields the lowest values reported for RCA 8850 and C31024 as follows:

	Illumination Area	
	Center 1.6 mm	Full cathode
RCA 8850	330 psec FWHM	480 psec FWHM
RCA C31024	290 psec FWHM	410 psec FWHM

Prospects for much faster photomultipliers appear to rest on parallel plane structures employing channel electron dynodes or transmission dynodes with negative electron affinity. A photomultiplier, presently, could be made with an existing channel electron multiplier dynode closely spaced between a planar cathode and anode. Preliminary measurements by Leskovar and Lo[16] on an experimental version from Bendix in which the cathode was unnecessarily far from the dynode showed a transit time of 7.5 nsec, a rise time of 680 psec and a single electron transit time spread of 148 psec. FWHM. Peak current was 16 mA and gain was 10^7. Pietri[17] reports on a channel plate photomultiplier with 1 nsec transit time, transit time spread < 235 psec FWHM and impulse response < 300 psec FWHM. The useful gain was only 10^5. The transmission dynode with negative electron affinity has been under development at RCA for a number of years. Early efforts with silicon exhibited single-stage multiplication of several hundred, but because the conduction electron lifetime is long (~ 1 μsec) and they emerge by diffusion, the estimated rise time is 3.3 μsec and FWHM 12.6 nsec per stage. The silicon material was abandoned because of the excessive dark current; around 10^{-10} A/cm^2 at room temperature. Later efforts have been directed to GaAs which has a carrier lifetime of about 1 nsec, and estimated rise time of 37 psec and FWHM 135 psec. Because of the greater band gap the dark current is much lower. Much effort has been directed to providing an internal electric field in the crystal by graded doping. With a sufficient internal field the drift time across the dynode could be as low as 30 psec so that the transmission dynode photomultiplier could be very fast with the additional advantage of excellent single electron resolution associated with the extremely high stage gains.

As scintillators and photomultipliers become faster and more efficient, the variation in light collection time with position of the exciting event become more significant. This effect has been calculated by Cocchi and Rota[18]. At some point it may be profitable to average the timing signal from 2 photomultipliers placed at opposite ends of the scintillator. This method has been used for very large scintillators, but could be applied to smaller scintillators.

References

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Figure Captions

Fig. 1. Left: Measured light intensity vs time for scintillations produced by γ rays and neutrons. Right: Integrals of measured curves on left.

Fig. 2. Rise time distributions for γ rays and neutrons. The experimental points represent distribution in times between the firing of fixed fraction discriminators set at ~10% and ~85% of the final pulse height for γ rays and neutrons. The curves were calculated from Eq. (3) using the measured illumination functions for stilbene shown in Fig. 1, and the measured value of P (2.3 electron/keV of electron energy).

Fig. 3. Analysis of a decay time measurement. Left: The measured decay curve (dotted) for P4-G12 in toluene (65 g/liter) and the calculated slow component adjusted to equal area of the measured curve at long times. Right: Experimental data (dotted) after subtraction of the slow component, and as calculated (solid) from Eq. (4) with measured P(t) and τ_1 = 0.1 nsec, τ_2 = 1.3 nsec.

Fig. 4. Calculated values of FWHM, and $T_{1/2}$ vs σ for 3 scintillators, showing mainly the effect of transit time on time resolution.

Fig. 5. Upper curves show calculated values of $W_{1/2}$ (FWHM) and $T_{1/2}$ vs τ_1 (upper left) and vs τ_2 (upper right). Lower curves show calculated dependence of optimum triggering fraction on σ and τ_1.

TABLE 1

COMPARISON OF MEASURED MEAN LIVES, τ.

Scintillato-	Kirkbride[a]	Lynch[b]	Kunze[c]	Birks[d]	Binkert[e]	Kelley[f]	Sipp[g]
Pilot B[h]	1.90	1.6		2.4		1.69	
Naton 136[k]	1.87	1.6		2.3		1.70	
NE-111[j]			2.3	1.7	1.75	1.66, 2.27	
Pilot U[h]						1.36	
P4-G12[m]		1.27					1.28

(a) Reference 8.

(b) Reference 9.

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(g) Reference 12.

(h) Pilot B, Pilot U manufactured by Nuclear Enterprises, Ltd.

(j) NE-111 manufactured by Nuclear Enterprises, Ltd.

(k) Naton 136 manufactured by Thorn Electronics, Ltd.

(m) Reference 10.

TABLE 2

RELATIVE LIGHT OUTPUT AND TIME CONSTANTS

The relative light output and the number of photoelectrons per keV were measured with a bialkali photocathode (RCA-8575). The values shown for τ_1 and τ_2, the mean lives for energy transfer and decay, are those for which Eq. (4) best fits the experimental data. The mean life τ_3 for the longer component was estimated from the slope of the semilogarithmic plot at long times (10—30 nsec).

Scintillator liquids	Mole fraction	Relative pulse height	P (p. e. /keV)	τ_1 (nsec)	τ_1(min) (nsec)	τ_2 (nsec)	τ_3(est) (nsec)
PPO		100	1.7			1.55	9
PBD		112	1.9				
P4-G12	$5 \cdot 10^{-3}$	124	2.1	0.15	0.07	1.27	8
P5-B9	$2.5 \cdot 10^{-3}$	130	2.2	0.2	0.1	1.22	9
TMSP	$1.8 \cdot 10^{-3}$	112	1.9	0.15	0.15	1.02	6.5
Glasslike							
TMQP	1.0	92	1.6	<0.05	<0.05	1.08	11.5
Plastics							
Pilot B		90	1.5	0.4		1.6	9.5
Naton 136		80	1.4	0.4		1.6	10
Crystals							
Stilbene		140	2.4	0.1		4.05	
NaI(Tl)		520	9.0				

Liquids are dissolved in toluene with oxygen removed.

PPO: diphenyl oxazole

PBD: phenyl-biphenyl-oxadiazone

P4-G12: 1^1, 4^4-bis (2-butyloctyloxy)-P-quaterphenyl

P5-B9: 1^2, 5^3-bis (3-ethylheptyl)-P-quinquiphenyl

TMSP: 1^2, 2^3, 5^2, 6^3-tetramethyl-P-sexiphenyl

TMQP: 1^2, 2^3, 3^2, 4^3-tetramethyl-P-quaterphenyl

Pilot B: Pilot Chemical Co.

Naton 136: Nash-Thompson Ltd.

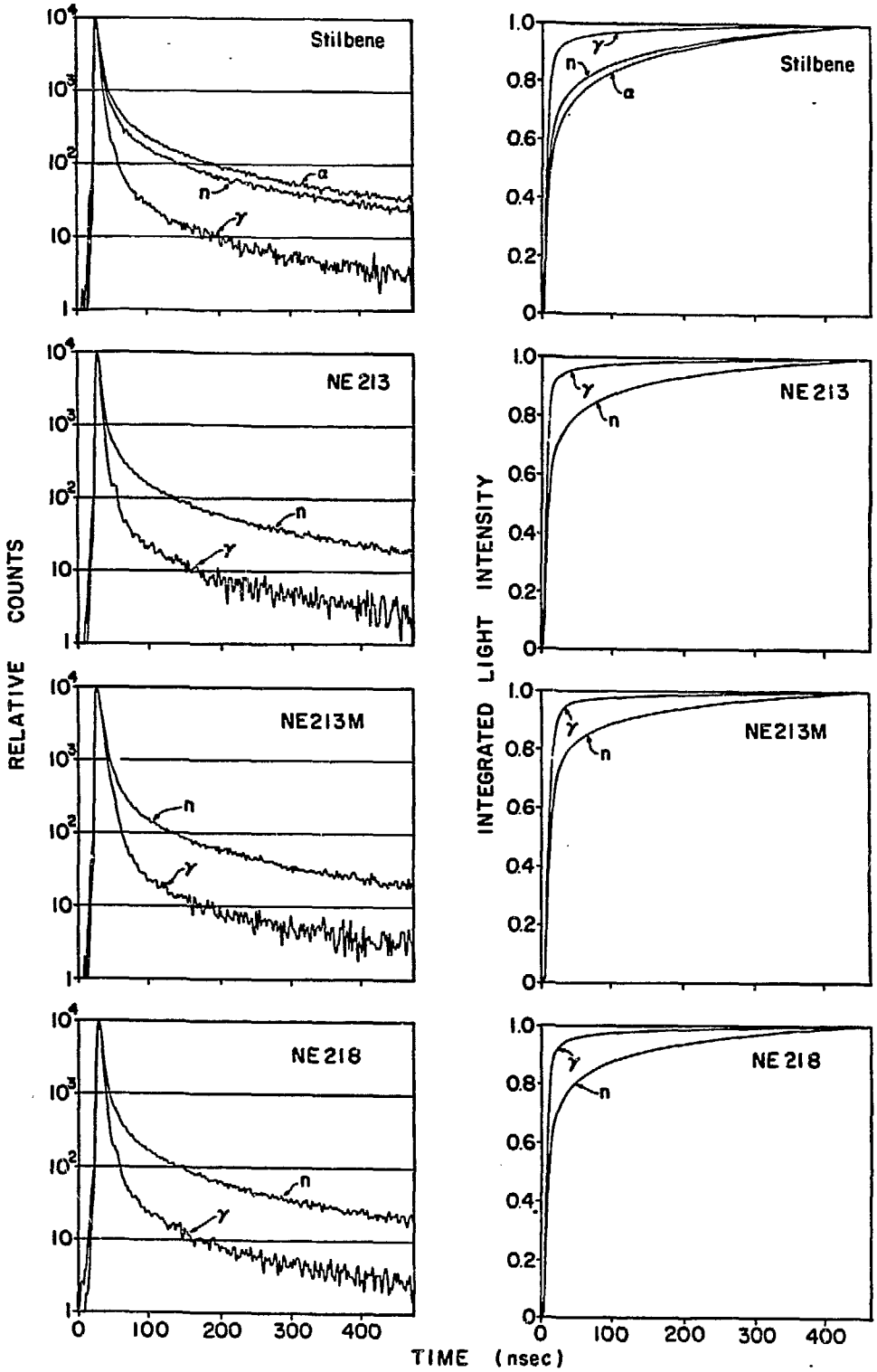
Stilbene
NE 213
NE213M
NE 218
RELATIVE COUNTS
INTEGRATED LIGHT INTENSITY
TIME (nsec)
n
γ
α

Figure 1

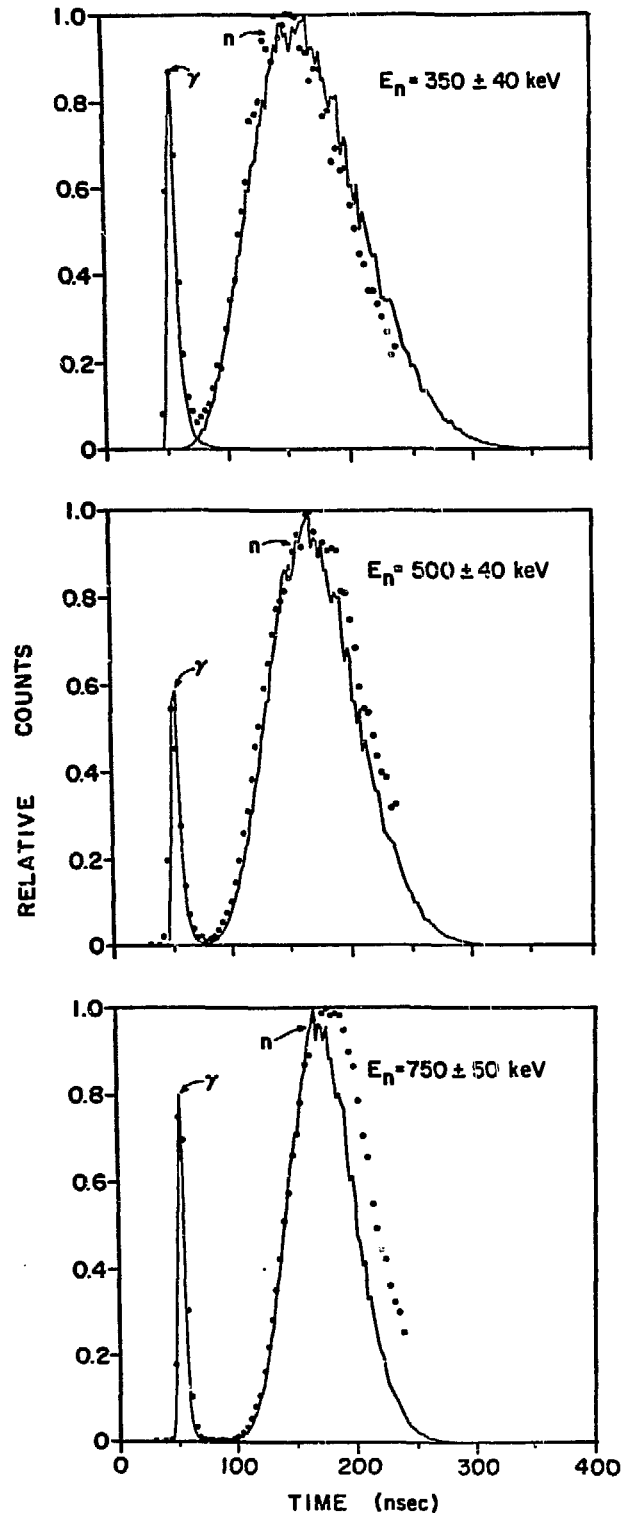
1.0
0.8
0.6
0.4
0.2
0
n
γ
E_n = 350 ± 40 keV
E_n = 500 ± 40 keV
E_n = 750 ± 50 keV
RELATIVE COUNTS
0
100
200
300
400
TIME (nsec)

Figure 2

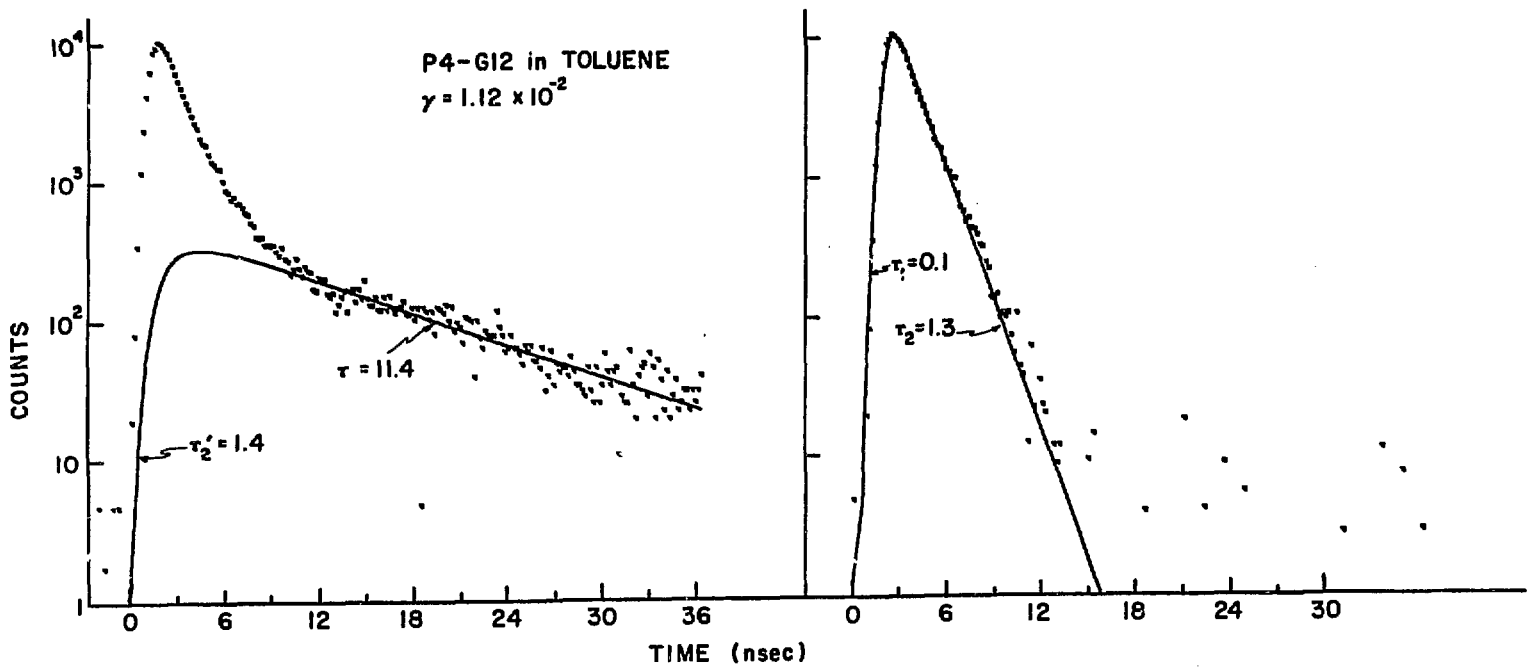
P4-G12 in TOLUENE
$\gamma = 1.12 \times 10^{-2}$
$\tau = 11.4$
$\tau_2' = 1.4$
$\tau_1 = 0.1$
$\tau_2 = 1.3$
COUNTS
TIME (nsec)

Figure 3

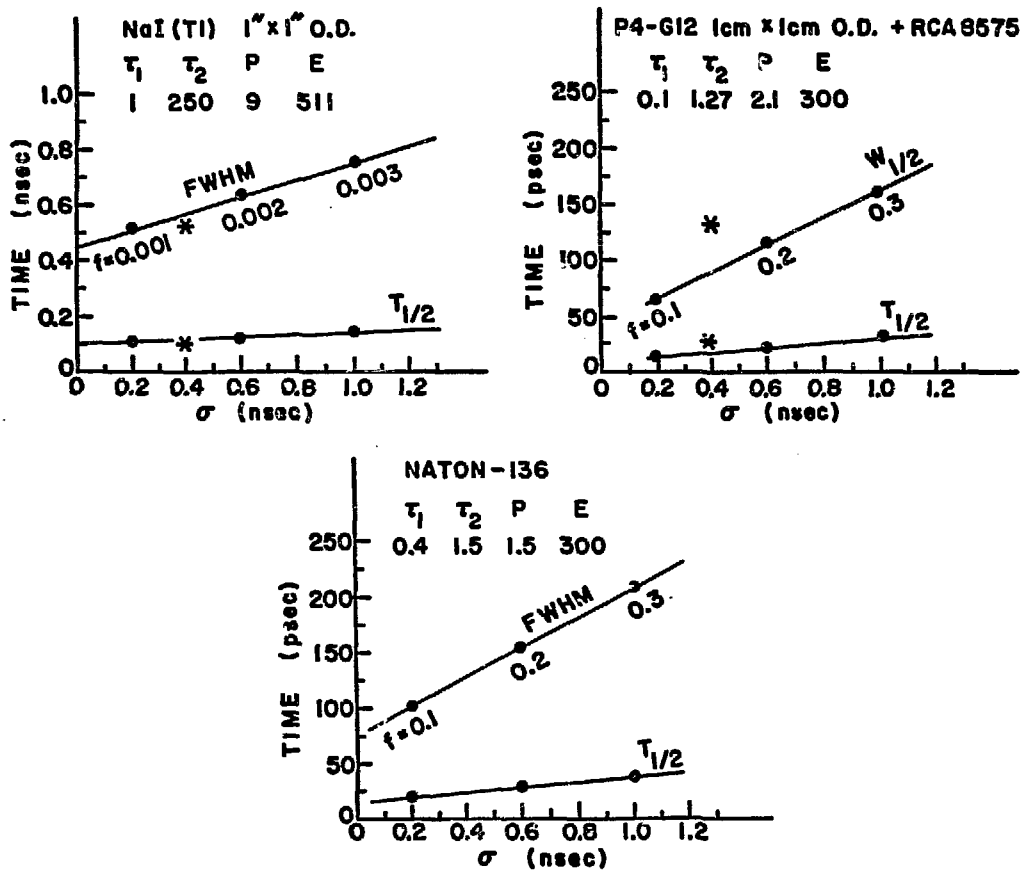
NaI (Tl) 1"x1" O.D.
τ1 τ2 P E
1 250 9 511
TIME (nsec)
FWHM
f=0.001
0.002
0.003
T1/2
σ (nsec)
P4-G12 1cm x 1cm O.D. + RCA 8575
τ1 τ2 P E
0.1 1.27 2.1 300
TIME (psec)
W1/2
f=0.1
0.2
0.3
T1/2
σ (nsec)
NATON-136
τ1 τ2 P E
0.4 1.5 1.5 300
TIME (psec)
FWHM
f=0.1
0.2
0.3
T1/2
σ (nsec)

Figure 4

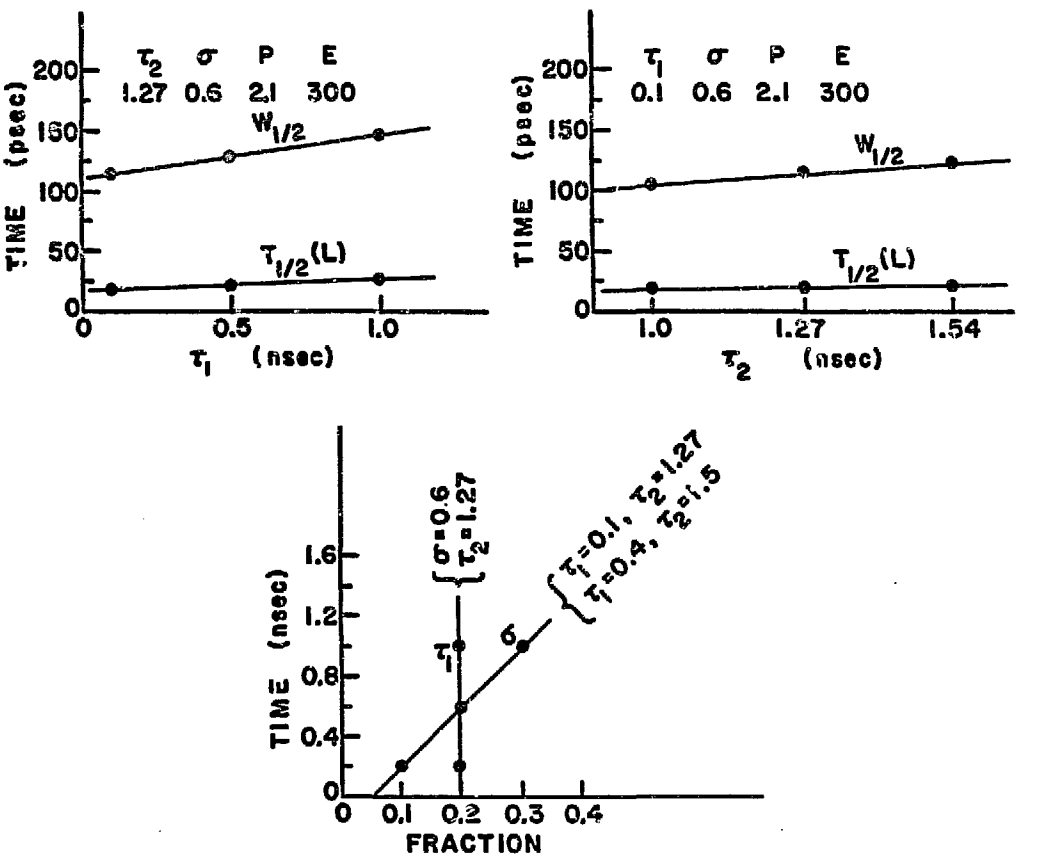
τ2 σ P E
1.27 0.6 2.1 300
W1/2
T1/2(L)
TIME (psec)
200
150
100
50
0
0
0.5
1.0
τ1 (nsec)
τ1 σ P E
0.1 0.6 2.1 300
W1/2
T1/2(L)
TIME (psec)
200
150
100
50
0
1.0
1.27
1.54
τ2 (nsec)
σ=0.6
τ2=1.27
τ1
δ
τ1=0.1, τ2=1.27
τ1=0.4, τ2=1.5
TIME (nsec)
1.6
1.2
0.8
0.4
0
0
0.1
0.2
0.3
0.4
FRACTION

Figure 5