STATUS OF TIMING WITH PLASTIC SCINTILLATION DETECTORS

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Timing properties of scintillators and photomultipliers as well as theoretical and experimental studies of time resolution of scintillation counters are reviewed.

Predictions of the theory of the scintillation pulse generation process are compared with the data on the light pulse shape from small samples, in which the light pulse shape depends only on the composition of the scintillator. For larger samples the influence of the light collection process and the self-absorption process on the light pulse shape are discussed. The data on rise times, fwhm’s, decay times and light yield of several commercial scintillators used in timing are collected.

The next part of the paper deals with the properties of photomultipliers. The sources of time uncertainties in photomultipliers as a spread of the initial velocity of photoelectrons, emission of photoelectrons under different angles and from different points at the photocathode, the time spread and the gain dispersion introduced by electron multiplier are reviewed. The experimental data on the time jitter, single electron response and photoelectron yield of some fast photomultipliers are collected.

As the time resolution of the timing systems with scintillation counters depends also on time pick-off units, a short presentation of the timing methods is given. The discussion of timing theories is followed by a review of experimental studies of the time resolution of scintillation counters.

The paper is ended by an analysis of prospects on further progress of the subnanosecond timing with scintillation counters.

1. Introduction

In many physics experiments a need arises to measure short time intervals with a good resolution and accuracy. Such experiments are mean life-time measurements of excited states, angular correlation function, time-of-flight measurements.

Physicists studying high-energy particles need high resolution fast coincidence systems to observe rare particles against a large background of other events. Sophisticated counter telescopes, employing a sequence of counters in coincidence and anticoincidence are used for particle, energy or event selection, and for studying the decay of fast particles in flight.

Several different nuclear radiation detectors are used in timing experiments: scintillation counters with organic and NaI(Tl) scintillators, Cherenkov detectors, multiwire proportional chambers, semiconductor detectors. Scintillation counters with organic scintillators, however, are still superior to produce the best time resolution, as well as to be used in a variety of different applications. Detection of $\gamma$-rays and $\beta$-particles in life-time measurements, neutrons in time-of-flight experiments, $dE/dx$ particle identification methods with thin scintillation foils, these are some of the possible applications.

Taking into account the great variety in size, shape and constitution, the possibility to work with a very high counting rate up to $10^7$ pulses/sec, it all makes them still very attractive and necessary in the many experiments in low- and high-energy nuclear physics.

The time resolution of scintillation counters is generally determined by the method used in the so-called delayed-coincidence technique which is widely applicable for the measurements of short nuclear life-times. The technique involves the measurements of time delay between the formation and subsequent decay of the nuclear state of interest. Fig. 1a shows the simplified block diagram of the basic time spectrometer. Time pick-off units are used to process the pulses provided by the detectors in such a way to define the time of occurrence of the detected events, as well as to optimise the time resolution of the system. The
time-to-pulse-height converter produces an output pulse with a height proportional to the time interval between input pulses from the detectors.

The time resolution of a timing system can be determined by measuring the distribution of the time delays between prompt events. A so-called prompt time spectrum is shown in fig. 1b. The figure of merit of a system is usually characterised by the full width at half maximum (fwhm) and the approximate logarithmic slope. The time resolution contributions can be associated with three phases of the timing process:

1) time variations in the interactions of the radiations with the detectors,
2) time variations in the response of the detectors to the radiations,
3) time variations associated with the electronic equipment.

Group (1) includes the variations in the source or target to detector flight times of the radiations, time characteristics of pulsed beam timing and time variation resulting from the interaction of the radiation over the detector dimensions. Although the minimisation of these time resolution contributions is rather straightforward, it very often produces a basic limitation of the obtainable time resolution.

Group (3) refers to the intrinsic time resolution of the electronic equipment, stability, time variation of time pick-off units and effect of electronic noise.

The contribution of group (2) relates to the time variations in the formation of the detector signal. This effect manifests itself in the shape of the detector signal, namely, the finite rise time, variation in the rise time and pulse height fluctuations, and it depends on the statistics of information carriers—photons and photoelectrons in the case of scintillation counters.

A process of nuclear radiation detection in the scintillation counter leads to several sources of time uncertainties and it can be summarised as follows:

1) The energy transfer from the detected nuclear particles to the optical levels in the scintillator.
2) The finite decay time of light emitting states in the scintillator and the light yield of the scintillator as a function of the energy of the detected radiation.
3) Variation of the time of light collection from the scintillator at the photocathode.
4) Efficiency of conversion of photons from the scintillator at the photocathode and the absolute number of photoelectrons.
5) Variation of transit times of photoelectrons from the photocathode to the first dynode.
6) Variation of the multiplication process in the electron multiplier.
Furthermore, the timing with the scintillation counter depends on:

7) Single electron response representing the frequency bandwidth of the photomultiplier.

8) Type of timing processing.

Processes (1) to (3) refer to the properties of the scintillator and they are reflected in the shape and intensity of the light pulse. Points (4) to (7) are related to the properties of the photomultiplier. Point (8) represents the method of electrical pulse collection from the photomultiplier. Fig. 2 shows the distribution of the current pulses from the scintillation counter, following ref. 1, calculated by a Monte Carlo method for a mean number of photoelectrons equal to 100. Note a large distribution of the pulse height and the rise times which reflects the influence of the above summarised processes on the time spread of the scintillation counters.

The various aspects of electronics timing with scintillation counters have been collected in the past by Bell, Schwarzschild, Bonitz, Bell, Ogata et al., Meling and Stary, Schwarzschild and Warburton, Williams and Gedcke, and Fossan and Warburton. The theory of timing has been reviewed by Gatti and Svelto.

In the following the present state of knowledge of the timing properties of scintillation counters with organic scintillators is given. In section 2 predictions of the theory of the scintillation pulse generation process are compared with the data on the light pulse shape from small samples, in which the light pulse shape depends only on the composition of the scintillator. For larger samples the influence of the light collection process and the self-absorption process on the light pulse shape are discussed. The data on rise times, fwhm, decay times and light yield of several commercial scintillators used in timing are collected. Section 3 deals with the properties of photomultipliers. The sources of time uncertainties in photomultipliers as a spread of the initial velocity of photoelectrons, emission of photoelectrons under different angles and from different points on the photocathode, the time spread and the gain dispersion introduced by electron multipliers are reviewed. The experimental data on the time jitter, single electron response and photoelectron yield of some fast photomultipliers are collected. As the time resolution of timing systems with scintillation counters depends also on the time pick-off units, a short presentation of the timing method is given in section 4. The discussion of timing theories in section 5 is followed by a review of experimental studies of the time resolution of scintillation counters. The paper is ended by an analysis of prospects on further progress of timing with scintillation counters.

2. Scintillators

2.1. Small samples

According to the theory of the light generation process in organic scintillators the detection of nuclear particles is equivalent to a δ-Dirac excitation of the lowest excited state of a crystal or a solvent in scintillating solutions. All preceding processes such as ion recombination, internal conversion from higher excited states of X, which yield molecules X in their lowest excited singlet states, are considered to be much faster than the final emission of the light.

In a unitary scintillator (a crystal, for example) which is transparent to its own fluorescence, the scintillation pulse is described by a single exponential function, as follows:

\[ i(t) = \frac{1}{\tau_M} e^{-t/\tau_M} \]  

where \( \tau_M \) is the molecular decay time constant. If the crystal absorbs a fraction (\( a \)) of its own fluorescence, it provides to increase the decay time constant and in eq. (1) \( \tau_M \) is replaced by \( \tau_M^a \), the technical decay time constant, which is given

\[ \tau_M^a = \frac{\tau_M}{1 - a} \]

1 Samples of scintillators in which the light pulse shape is not affected by the volume and only depends on the composition of the scintillator. Polished samples with thickness below 5 mm are regarded as small ones.
by\(^{12,14}\) 
\[
\tau'_m = \frac{\tau_m}{1 - a\phi}, 
\]
(2)

where \(a\) is the probability of re-absorption of fluorescence photons, \(\phi\) the quantum efficiency of the scintillator.

In the scintillating solutions the shape of the light pulse depends on the energy transfer process from the solvent \(X\) to the solute \(Y\) in binary systems, and to the wavelength shifter \(Z\), to provide a better matching to photomultiplier spectral sensitivity, in ternary solutions. Birks\(^2\) and Birks and Pringle\(^3\) have studied both theoretically and experimentally the light pulse shapes from several organic liquid and plastic scintillators. They have derived a general description of the light pulse from the binary and ternary liquid scintillators and the binary plastics in terms of molecular parameters.

In the binary liquid scintillators a complete "statistical mixing" of solvent molecules \(X\) and solute \(Y\) occurs owing to molecular diffusion and \(X\) excitation migration, so that the energy transfer rate is independent of time. The energy transfer process obeys Stern–Volmer kinetics\(^1\), and the scintillation light pulse according to Birks\(^2\) is described as follows:

\[
i(t) = \frac{\tau}{\tau - \tau_1} (e^{-t/\tau} - e^{-t/\tau_1})
\]
(3)

where
\(\tau\) is the fluorescence decay time constant of solute \(Y\),
\(\tau_1\) is the decay time of the solvent \(X\) in the presence of solute \(Y\).

In binary plastic scintillators molecules of the solute \(X\) and the solvent \(Y\) remain effectively stationary, apart from possible Brownian rotation during the energy transfer process, so that the transfer rate decreases with an increase in time. The energy transfer process is described by Forster kinetics\(^6\) assuming a long-range radiationless dipole–dipole interaction process. Birks\(^2\) has derived the Forster kinetics relations for the scintillation pulse shape of binary plastic solutions. At high molar concentration \([^1Y]\) of the solute, the pulse shape is of the form\(^{12,13}\):

\[
i(t) \sim e^{-t/\tau} - e^{-t/\tau_1} - 2e^{-(t/\tau_1)}1^{1/2}
\]
(4)

where \(\tau_1\) and \(\tau\) are the decay time constants of the solvent and solute, respectively.

\[
y = \frac{[^1Y]}{[^1Y]_0},
\]
and
\[
[^1Y]_0 = \frac{3000}{2\pi^{3/2} N R_0^3}
\]

(5)

is the "critical molar concentration" where
\(N\) is Avogadro's number,
\(R_0\) the critical energy transfer radius.

Typical scintillation pulse shapes evaluated from eq. (3) for binary liquid scintillators and from eq. (4) for binary plastics have been shown in ref. 12. A comparison of the fastest liquid and plastic scintillators containing the same solute shows that in the plastics a more rapid initial rise is expected. It reflects the difference between Forster and Stern–Volmer kinetics. The rate of the Forster energy transfer is initially rapid owing to the transfer to \(Y\) molecules from close lying molecules, but it decreases with an increase in time when transfer to more distant molecules of \(Y\) occurs.

Experimentally the light pulse shape from scintillators is studied by the so-called single photon method due to Bollinger and Thomas\(^7\). In this method the scintillator under investigation is coupled well to the photomultiplier in the reference counter to produce an accurate timing signal but very weakly to the other one so that only one photoelectron is produced for every \(\sim 50\) light flashes in the scintillator\(^7\). The time distribution spectrum observed at the output of the time-to-pulse-height converter combines the effect of the illumination function \(i(t)\) and the instrumental response function \(f_p(t)\) of the timing system, as follows:

\[
i^\ast(t) = i(t) * f_p(t).
\]
(7)

The function of the system response is mainly due to the time jitter of the photomultiplier detecting single photons. The fwhm of this function varies between 0.25 ns and 1 ns, depending on the quality of the photomultiplier used. The time jitter of fast photomultipliers and methods of determination are discussed in section 3.

Birks and Pringle\(^3\) have shown that the scintillation pulse shapes found experimentally for binary liquid scintillators agree well with those calculated using eq. (3). For plastic scintillators it is stated that because of the complexity of eq. (4) the generation of the synthetic pulse shapes and their convolution with the instrumental response func-
tion to fit experimental curves has been considered impractical.

As eqs. (3) and (4) differ weakly, eq. (3) is commonly used to describe the light pulses also from plastic scintillators. It has been proposed by Raviart and Koechlin and verified by Lynch and Kuchnir and Lynch for liquid and plastic scintillators based on the light pulse shape study by the single photon method.

The timing study with plastic scintillation counters has shown, however, a need to assume a slow initial rise of the light pulse to interpret the timing properties of the counter. As a consequence, the following convolution has been proposed to describe the light pulse shape from plastic scintillators:

\[ i(t) = f_G(t) * e^{-t/\tau}, \]

where

\[ f_G(t) \]

is the clipped Gaussian function shifted by 2.5\( \sigma \), which represents the rate of the energy transferred from the detected nuclear particle to the optical levels of the solute,

\[ \tau \]

is the decay time constant of the scintillator.

The above suggestion was confirmed in refs. 26 and 27 by the measurements of the arrival time of a fixed fraction of the total collected light of a scintillating pulse with respect to the impinging time of nuclear radiation represented by Cherenkov light generated in the photomultiplier window. The observed delays, characteristic for each scintillator, were about a factor of two larger than those expected from the shape of the light pulse according to eq. (3).

The direct test of the validity of eq. (8) to describe the light pulse shape from fast binary plastics has been performed in refs. 28 and 29. The light pulse shapes from the NE 111 scintillator and the modified NE 111 scintillators by adding quench agents have been measured by the single photon method using C31024 photomultipliers. A fit of eq. (8), convoluted with the prompt response of the system (fwhm = 250 ps), to the measured time distribution of the light pulses has shown that eq. (8) is able to reproduce in detail the shape of the light pulse from small \( \phi = 2.5 \text{ cm}, \ h = 5 \text{ mm} \) polished or black samples of NE 111. Similar fitting tests performed with eqs. (3) and (4) have not been successful to reflect the initial slow rise of the measured pulse (see fig. 3).

According to ref. 28, the initial slow rise, approximated by the Gaussian function in eq. (8), may come from the finite times of de-excitation of several higher levels of the solvent molecules excited by nuclear particles. This process precedes the intermolecular energy transfer which is considered in the theory of scintillators. As the intermolecular energy transfer process in binary plastics (such as Ne 111) is expected to be fast, it is not reflected in eq. (8). Thus the rise time of the light pulses seems to be controlled by preceding processes in the detection mechanism of nu-

![Fig. 3. Fits of eqs. (3), (4) and (8) convoluted with the prompt spectrum of the system to the light pulse measured with the polished 5 mm thick sample of NE 111 scintillator. Note that eq. (8) is only able to reflect an initial slope of the experimental pulse shape (from ref. 28).](image-url)
clear radiation in plastic scintillators. The observed slow initial rise of the light pulse, approximated also by eq. (8), from the sample of the PVT (polyvinyltoluene)\(^{29}\), commonly used as a solvent in plastic scintillators, as well as from stilbene and anthracene samples\(^{31}\) seems to confirm the above hypothesis. Note that for unitary scintillators the theory of light generation process\(^{12}\) predicts a light pulse shape of the form given by eq. (1).

The recently reported study of the light pulse shape in refs. 32 and 33 seems to be, however, in contrast to the above discussed eq. (8). A much faster rise time, equal to 130 ps, of the light pulse from the NE 111 scintillator than that found in ref. 28 (350 ps), as well as a significantly lower fwhm of the light pulses from quenched NE 111 plastics than those observed in ref. 29 have been measured. In the study presented in refs. 30 and 32 the samples of the scintillators were excited by the 50 ps wide pulses of electrons from the linear accelerator. Sampling techniques were used to record the signal from a fast vacuum photodiode. The fwhm of the system response, equal to 115 ps, was measured with Cherenkov radiation. In the study reported in ref. 33 the scintillators were excited by the 100–180 ps wide X-ray pulses generated by the laser, and the light pulses were recorded by a streak camera. Since the basic resolution of the streak camera was 20 ps, the measurements were limited by the fwhm of the X-ray pulse. The results of this experiment, however, were not consistent with those obtained in the Linac experiment\(^{32}\). The fwhm of the light pulse from the NE 111 scintillator, equal to 0.4 ns, was far too small compared to the observed fwhm (=1.2 ns) in refs. 28, 29 and 32. It shows that a significant internal disagreement exists in the results of all the discussed experiments from ref. 28, 32 and 33. A detailed comparison of the data from the above experiments and analysis of the experimental conditions suggest\(^{29}\) that a basic difference of the energy of the scintillators excitation should be pointed out. Eq. (8) is concluded from the study of the light pulse shape by the single photon method\(^{18,29}\), and the scintillators were excited by \(\gamma\)-rays or electrons of about 200–500 keV on average, typical for low-energy physics. In the Linac experiment the energy of the scintillator excitation may be estimated to be equal to about \(10^8\) MeV/Linac pulse\(^{32,41}\). In the case of the X-rays experiment the energy of the scintillator excitation is of the order of \(10^{10}–10^{12}\) MeV/X-ray pulse. It is difficult to predict at the moment what are the properties of scintillators at so high an energy of scintillator excitation.

The above discussed binary plastics have one drawback in that the emitted light is strongly absorbed inside the scintillators. Therefore to work with large samples one has to use ternary plastics. In earlier years it assured also a better matching of the emitted light to the spectral sensitivity of photomultipliers. The ternary plastics differ from the binary ones in that the energy is additionally transferred to the wavelength shifter before the light is emitted. For liquid scintillators, according to Birks\(^{12}\), the light pulse shape is described as follows:

\[
i(t) ≈ z^{-\frac{\tau_1 - \tau}{\tau_2}} e^{-\frac{t}{\tau_1}} + e^{-\frac{t}{\tau_2}} + e^{-\frac{t}{\tau}}, \tag{9}\]

where \(\tau_1\) and \(\tau_2\) are the decay time constants of the solvent in the presence of the primary solute, and primary solute in the presence of the wavelength shifter, respectively, \(\tau\) is the decay time constant of the wavelength-shifter.

In the case of the ternary plastics there is no equation describing the light pulse shape derived from the Forster kinetics\(^{12}\). It has been only verified by Benne\(^{34}\) that the Forster kinetics is applicable to the solute–solute transfer in organic solutions. Eq. (9) may be used also for the ternary plastics as a good approximation\(^{22}\). The light pulse shape of the ternary plastics was studied by Lynch\(^{20}\) and the two-exponential description according to eq. (3) was used to reflect the shape of measured pulses.

The study of the light pulse shape performed in ref. 28 leads, however, to the other equation to describe the scintillating pulse from the ternary plastics, as follows:

\[
i(t) = f_G(t) * (e^{-t/\tau} - e^{-t/\tau_1}), \tag{10}\]

where \(f_G(t)\) is the Gaussian function describing the rate of the energy transfer from the detected radiations to the primary solute, and the exponential functions describe the energy transfer process to the wavelength shifter (\(\tau_1\)) and final emission of the light (\(\tau\)).

The validity of eq. (10) was verified in ref. 28 by the study of the scintillation pulse from NE 104 and KL 236 scintillators (see fig. 4).
2.2. Thick samples

All the above considerations may be directly applied to thin samples of scintillators where the light pulse shape depends only on the composition of the scintillator. For larger samples the light collection process\(^{35-38}\) influences significantly the light pulse shape. Self-absorption and re-emission processes are considered to increase the decay time constant according to eq. (2). In ternary solutions, the energy transfer to the wavelength shifter occurs both radiationlessly and radiatively\(^{2}\). The efficiency of the last process depends on the dimensions of the sample and it is reflected in the shape of the light pulse. The influence of the primary, unshifted light from the ternary NE 102A scintillator has been clearly seen in the study of the light pulse shape from thin scintillator foils. The strong reduction of the fwhm and the decay time constant has been observed with foils thinner than 400 \(\mu g/cm^2\)\(^{39}\).

2.2.1. Light collection process

Theoretical investigations of the light collection process in cylindrical scintillators\(^{15,34}\) have shown that the probability density function of the transit time of photons is given approximately by the relation:

\[
f_L(t) = \frac{1}{\sigma_L} e^{-t/\sigma_L}
\]

(11)

with

\[
\sigma_L = \frac{\alpha h}{V_0},
\]

(12)

where

\(\alpha\) is a factor depending on the type of the reflecting surface and the optical matching at the...
scintillator exit ($\alpha=3$ for a non-specular surface),
h is the height of the scintillator,
$V_0$ the velocity of light.

The experimental study of the time spread of the light collection process was reported in refs. 38 and 40. Sipp and Miehe$^{40})$ have studied cylindrical liquid scintillators coated with aluminium foil and concluded that time spread associated with the light collection process is not of importance compared to the self-absorption and re-emission process. The study performed in ref. 38 for the samples of perspex coated with white reflecting paint has shown the contrary that the light collection process introduces a large time spread which influences strongly the light pulse shape$^{24)}$ and the time resolution of the scintillation counter$^{24)}$. Fig. 5 shows the time distribution spectra of the collected light measured with samples coated with NE 560 reflecting paint for the point light source placed in the centre of the top surface of the cylindrical sample. A pure exponential decay caused by the time spread of the collected light was observed with the decay time constant increasing with the height of the sample. Fig. 6 presents the standard deviation $\sigma_L$ of the transit times of photons collected from the samples coated with NE 560 reflecting paint in comparison to the Nutt$^{36)}$ and the Cocchi and Rota$^{35)}$ predictions. The whole study of ref. 38 confirms Nutt’s predictions$^{36)}$ that the probability density function of the transit time of photons is represented well by a single exponential function. The standard deviation $\sigma_L$ is proportional to the height of the samples for a fixed diameter and depends on the quality of the reflecting surfaces. To interpret the larger values of $\sigma_L$ found in the experiment compared to the calculated ones, back reflections from the boundary between the glass window of the photomultiplier and the photocathode itself were postulated.

2.2.2. Light pulse shapes from thick samples of scintillators

The influence of a larger volume of the scintillator on the light pulse shape has been observed by Lyons and Stevens$^{41)}$ and studied by Sipp and Miehe$^{40)}$ and Moszyński and Bengtson$^{28)}$. Fig. 7 shows the time distribution spectra of the light pulses from binary liquid scintillators (BIBUQ in xylene) of different height excited by $\beta$-rays as measured by Sipp and Miehe. The large widening of the decay time constant from 1.28 ns to 1.94 ns was observed when the height of the scintillator was increased from 0.2 cm to 5 cm.

A quantitative study of the light pulse shape from larger volume scintillators performed for the NE 111, NE 104 and KL 236 plastics is reported in ref. 28. The samples were coated with white reflecting paint and the separated scintillators tech-
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Fig. 8. Time distribution spectra of the light pulses measured with the 0.5 cm and 1 cm thick "white" samples and the 0.5 cm thick "black" sample of the NE 104 scintillator (points). Dashed lines were calculated convoluting eq. (10) with the measured time distribution spectra of the collected light. Full line on the pulse from the 0.5 cm "white" sample was calculated assuming larger time spread of the collected light than that measured experimentally. The full line on the pulse from the 1 cm thick "white" sample was calculated assuming a subsequent self-absorption and re-emission process according to eq. (13). The best fit for the black sample was obtained by lowering of the $\tau_1$ parameter in eq. (10) from 0.6 ns found for the polished sample to 0.5 ns for the black one (from ref. 28).

The evidence of the self-absorption and re-emission process is based on ref. 28 on the study of the light pulse shape from the thin and thick ternary samples of KL236 plastic with passband light filters. In the self-absorption and re-emission process the emitted light is shifted to longer wavelengths. Thus in the measurements with selected wavelengths, the longer-wave components are more affected by this process and the larger slowing-down of the light pulse was observed (see fig. 9).

2.3. DECAY TIME CONSTANT STUDY

The papers which deal with the light pulse shape study from organic scintillators were reviewed above. One should note also a number of papers studying the decay times of different plastic and liquid scintillators commercially avail-

$\text{i}(t) = i_s(t) \cdot e^{-t/\sigma_L} \cdot e^{-t/\tau_1}$, \hspace{1cm} (13)

where $i_s(t)$ is the light pulse from the small sample, according to eq. (10), $e^{-t/\sigma_L}$ the probability density function of the photon transit time in the scintillator, $e^{-t/\tau_1}$ a term describing the self-absorption and re-emission process.

The good fit was also observed using a larger $\sigma_L$ value than that measured experimentally, as follows:

$i(t) \sim i_s(t) \cdot e^{-t/\sigma_L}$. \hspace{1cm} (14)

In this equation the term $e^{-t/\sigma_L}$ represents the probability density function of the photon transit time in the scintillator, including the self-absorption and re-emission process.

The evidence of the self-absorption and re-emission process is based on ref. 28 on the study of the light pulse shape from the thin and thick ternary samples of KL236 plastic with passband light filters. In the self-absorption and re-emission process the emitted light is shifted to longer wavelengths. Thus in the measurements with selected wavelengths, the longer-wave components are more affected by this process and the larger slowing-down of the light pulse was observed (see fig. 9).
TABLE 1
Effective decay times of organic scintillator.

<table>
<thead>
<tr>
<th>Scintillator</th>
<th>Small samples $h \leq 0.5$ cm</th>
<th>Large samples $h \geq 1$ cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>NE 111</td>
<td>1.3c, 1.5b</td>
<td>1.65e, 1.7e, 1.66h</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.75f, 2.3f</td>
</tr>
<tr>
<td>Pilot U</td>
<td>1.38c</td>
<td>1.5f, 1.36h, 1.5h</td>
</tr>
<tr>
<td>Naton 136</td>
<td>1.64c</td>
<td>1.85f, 1.87f, 1.6m</td>
</tr>
<tr>
<td>KL 236</td>
<td>1.59b</td>
<td>2.3g, 1.7h</td>
</tr>
<tr>
<td>NE 102A</td>
<td>2.2c</td>
<td>5.1, 2.4, 2.5</td>
</tr>
<tr>
<td>NE 104</td>
<td>1.74c</td>
<td>5.6, 2.8, 1.9</td>
</tr>
<tr>
<td>NE 110</td>
<td>2.9c</td>
<td>3.1f, 3.3n</td>
</tr>
<tr>
<td>Pilot B</td>
<td>1.69b, 1.6m, 1.9f</td>
<td>1.27k, 1.67o</td>
</tr>
<tr>
<td>BIBUQ</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

a Ref. 28, $\Theta = 2.5$ cm, $h = 5$ mm, “black” sample.
b Ref. 28, $\Theta = 2.5$ cm, $h = 5$ mm, polished sample.
c Ref. 28, $\Theta = 12$ mm, $h = 2$ mm, polished sample.
d Ref. 40, $\Theta = 2.0$ cm, $h = 2$ mm, sides of the sample coated with aluminium foil.
e Ref. 28, $\Theta = 2.5$ cm, $h = 1$ cm, “white” sample.
f Ref. 28, light from the small sample $\Theta = 12$ cm, $h = 2$ mm, transmitted through white sample, $\Theta = 2.5$ cm, $h = 1$ cm.
g Ref. 13.
h Ref. 43, corrected for the contribution of the long component.
i Ref. 46.
j Ref. 45.
k Ref. 43, corrected for the contribution of the slow component.
l Ref. 42, $\Theta = 1/2"$, $h = 1/2"$ sample.
m Ref. 20, corrected for the contribution of the long component.
n Ref. 44.
o Ref. 40, $\Theta = 2$ cm, $h = 1$ cm, sides of the sample coated with aluminium foil.

The decay time constant of the scintillation pulses is measured by the single photon method\(^{17}\) and the effective decay time is determined from the linear part of the slope of the fast component of the measured pulse. In some cases\(^{20,21,43}\) the decay time constants of the fast component of the light pulse are corrected for the contribution of the long component. As the preceding discussion of the light pulse shape from the scintillators showed a significant dependency of the decay time constant on the size of the studied samples, the data from the literature given in table 1 are collected in two groups; the decay times measured for samples with the thickness less than 5 mm, and larger than 1 cm. In the past, however, the influence of the scintillator volume on the decay time constant was not considered and very often the size of the samples studied was not specified. It was shown in ref. 28 that the effective decay time constants measured with the small samples are systematically less compared to published ones, which in turn agree well in general with the measured ones for the larger samples.

In table 2 the data on the rise times from 10 to 90% of the pulse height and on the fwhm of the light pulses reported in refs. 28, 32 and 41 are given. In the timing application both these parameters are more meaningful than the decay time. Note that the binary NE 111 scintillator is superior. It shows the fastest rise time and the smallest fwhm of the light pulse. The timing experiments\(^{23-25}\) performed with the NE 111 scintillator have also given the best time resolution of the prompt time spectrum measured with the $^{60}$Co source.

2.4. LIGHT YIELD

One of the important parameters of the scintillation counter which depends on both the scintillator and the photomultiplier properties is the number of photoelectrons released from the photocathode per keV of energy lost in the scintillator by the nuclear radiation. In the past very often this figure was estimated to be very low. Recent measurements performed by Lynch\(^{20}\), Houdayer et al.\(^{47}\) and Bertolaccini et al.\(^{48}\) showed 1–2 pho-
TIMING WITH PLASTIC SCINTILLATION DETECTORS

TABLE 3
Light yield of some organic scintillators.

<table>
<thead>
<tr>
<th>Scintillator</th>
<th>Birks and Pringle$^{13}$</th>
<th>Lynch$^{20,21}$</th>
<th>Bertolaccini et al.$^{38}$</th>
<th>Bialkowski and Moszyński$^{39}$</th>
<th>Nuclear Enterprises$^{44}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anthracene</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td></td>
</tr>
<tr>
<td>Stilbene</td>
<td>80</td>
<td>64</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Naton 136</td>
<td>45</td>
<td>39</td>
<td>54, 58</td>
<td>60</td>
<td></td>
</tr>
<tr>
<td>Pilot B</td>
<td>60</td>
<td>42</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>NE 102A</td>
<td>58</td>
<td>70, 70</td>
<td></td>
<td>60</td>
<td></td>
</tr>
<tr>
<td>NE 111</td>
<td>50</td>
<td>40</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>NE 104</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>BIBUQ</td>
<td>60</td>
<td>58</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pilot U</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Photomultiplier</td>
<td>56UVP</td>
<td>RCA7585</td>
<td>56AVP</td>
<td>56DUVP</td>
<td></td>
</tr>
</tbody>
</table>

$^a$ Normalised to 100 for anthracene.

to electrons per keV, which depends on the type of scintillator and photomultiplier.

In table 3 the light yield of several scintillators used in the timing experiments is listed as a relative figure compared to the light yield of the anthracene crystal. As the measured light yield depends on the spectral response of the photomultiplier, the size of the studied sample and the method of reflecting surface preparation, a large spread in the data from different references is observed.

2.5. TIMING APPLICATION

The above review of the timing properties of the plastic scintillators leads to several conclusions concerning the choice of the optimal scintillator for given timing applications. For this purpose one has to consider:

- size of the scintillator,
- type,
- light yield.

For α- and β-rays the thickness of the sample may be easily limited to be below 5 mm; thus the light collection process and reabsorption process in the polished samples would not be of importance$^{38}$). A more serious problem is the case of γ-rays detection. The choice of the size of the sample would be the result of a compromise between the efficiency of γ-ray detection and the time spread introduced by the light collection process$^{38}$). Table 4 presents a comparison of the transit time jitter of some fast photomultipliers used in timing applications, with the time spread introduced by the light collection process in the 1 cm and 2.5 cm thick samples with the 2.5 cm diameter coated with white reflecting paint$^{38}$). Note that to utilise the rapidity of the C31024 photomultiplier it is necessary to reduce the height of the scintillators below 5 mm. The time spread of transit time of photons in the commonly used 1"×1" sample is larger than the time jitter of typical fast photomultipliers. The limited size of the sample has also the other meaning because it protects from the attenuation of the light inside the scintillators themselves. It is specially of importance for binary plastics which exhibit a large self-absorption.

TABLE 4
Comparison of the transit time jitter of some photomultipliers used in timing experiments with the time spread introduced by the light collection process (following ref. 38).

<table>
<thead>
<tr>
<th>Photomultiplier</th>
<th>$\sigma_{PM}$ (ns)</th>
<th>$\sigma_{L}$ (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\varnothing = 2.5$ cm</td>
<td>$\varnothing = 2.5$ cm</td>
</tr>
<tr>
<td></td>
<td>$h = 1$ cm sample</td>
<td>$h = 2.5$ cm sample</td>
</tr>
<tr>
<td>C 31024</td>
<td>0.13$^a$</td>
<td>0.2</td>
</tr>
<tr>
<td>RCA 8850</td>
<td>0.2$^b$</td>
<td>0.2</td>
</tr>
<tr>
<td>PM 2103</td>
<td>0.28$^c$</td>
<td>0.2</td>
</tr>
<tr>
<td>XP 1021</td>
<td>0.3$^d$</td>
<td>0.2</td>
</tr>
<tr>
<td>56 AVP</td>
<td>0.5$^d$</td>
<td>0.5</td>
</tr>
</tbody>
</table>

$a$ See ref. 62.
$b$ See ref. 40.
$c$ See ref. 59.
$d$ See ref. 57.
For time spectroscopy with the highest time resolution one has to choose a binary plastics such as an NE 111 scintillator and the smallest possible thickness. The light pulses from binary plastics are generally faster than from ternary ones, as the additional energy transfer to the wavelength shifter is omitted. Table 2 shows that at the moment the binary NE 111 scintillator is superior to any other on the market. When the efficiency of the detection of $\gamma$-rays, for example, is of importance, and one has to use samples of the scintillators thicker than 2 cm, ternary pastics would be recommended. The light collection process introduces so large a time spread, that the light yield and lower attenuation of the light inside the sample become of importance. For these purposes the Pilot U—the fastest ternary plastic (see tables 1 and 2)—would be the best one. The dependency of the time resolution on type and scintillator size will be discussed in section 6.3.

3. Photomultipliers

The time resolution capabilities of photomultipliers are associated with:

a) the time jitter in the transit time of electrons travelling from photocathode to the anode,

b) numbers of photoelectrons released from the photocathode which are a function of the photocathode sensitivity and efficiency of photoelectrons collection on the first dynode,

c) spread in the gain of the electron multiplier which increases the pulse height distribution of output pulses.

The optimal conditions of timing with the photomultiplier depend also on the frequency bandwidth of the photomultiplier, reflected in the so-called single electron response\(^{11}\). It represents the shape of the output pulse of the photomultiplier generated by the single photoelectron which reached the first dynode.

3.1. TIME JITTER OF PHOTOMULTIPLIERS

The dispersion of the electron transit time in a photomultiplier is considered\(^ {11,50}\) to result from the following sources of time uncertainties:

a) variations of the transit time of photoelectrons between a photocathode and a first dynode due to a different initial velocity of photoelectrons and an emission of photoelectrons under different angles,

b) different transit times of photoelectrons emitted from different points at the photocathode,

c) electron time spread in the electron multiplier.

Transit time spread of photoelectrons is associated with their initial energy and direction of emission. If the initial velocity is represented as a spatial vector it can be resolved into two components, $W_n$ normal to the cathode and $W_t$ parallel to the cathode. To a first approximation $W_n$ determines the transit time of the electron and $W_t$ determines the path it will be travel.

A difference $\Delta t$ of the time of flight of photoelectrons emitted from a photocathode with the initial energy equal to $W_{ne}$ and the energy close to zero may be described approximately as follows\(^ {50}\):

$$\Delta t = \sqrt{\frac{2m}{e}} \sqrt{\frac{W_{ne}}{E}},$$  

where $m$ and $e$ are mass and charge of an electron, $E$ is the electric field intensity at the photocathode, and

$$W_{ne} = h(v-v_0),$$

where $h$ is Plank’s constant, $v$ the frequency of incident radiations, $v_0$ the frequency of the photocathode sensitivity threshold.

It can be seen from eq. (15) that the transit time spread resulting from the initial velocity distribution is decreased by increasing the voltage between the photocathode and the first dynode. It can not be, however, increased too much because of the influence of the electric field intensity at...
the photocathode on the noise level of photomultipliers. Eqs. (15) and (16) show also that the transit time spread is dependent on the wavelength of the incident light. For longer wavelengths of the light which are close to the threshold of the photocathode sensitivity a lower transit time spread may be expected. This was confirmed experimentally in ref. 51.

The tangential initial velocity component \( W_t \), which is parallel to the cathode, shifts the point of impact of photoelectrons at the first dynode by a certain distance (see fig. 10) and has generally negligible influence on the change of the transit time\(^5\). This shift depends also on the point on the photocathode from which the electron is emitted.

The transit time difference is purely geometrical in nature. It may be defined as the difference in transit time between an electron trajectory starting from the centre of the photocathode and other trajectories starting from other points on the surface of the cathode, it being assumed that all electrons have zero initial velocity. The transit time difference depends directly on the quality of the input electro-optics of the photomultiplier.

In spite of the fact that very often the time jitter of a photomultiplier is considered to be associated with the transit time spread of photoelectrons between photocathode and first dynode, the contribution of the next stages of electron multiplication can not be neglected. The same problems, which were discussed above for the input optics of photomultipliers, occur in the next stages of multiplication, but in an even more serious form\(^5\).

On the other hand, because of the multiplication process of electrons in the multiplier, the influence of the time jitter produced by the next stages is significantly reduced.

The variance \( \sigma_D^2 \) of the transit time of electrons through the multiplier may be written as follows\(^5\):

\[
\sigma_D^2 = \frac{\sigma_{D,1}^2}{g_1} (1 + g_2^2) + \frac{\sigma_{D,2}^2}{g_1 (g - 1)} (1 + g_2^2),
\]

where

- \( \sigma_{D,1}^2 \) is the variance of the transit time of electrons between first and second dynode,
- \( \sigma_{D,2}^2 \) is the variance of the transit time of electrons between two successive dynodes,
- \( g_1 \) and \( g \) the gain of the first and successive dynodes, respectively,
- \( g_{2,1} \) and \( g_{2} \) the variance of the gain of the first and successive dynodes, respectively.

It should be noted that variance of the transit times of electrons in the multipliers is strongly reduced when a high gain GaP(Cs) first dynode is applied in the photomultiplier. According to ref. 53 it may even be omitted altogether considering the total time jitter of photomultipliers with a first high gain dynode.

Finally, the total variance of the photomultiplier transit time may be written as follows:

\[
\sigma_{PM}^2 = \sigma_v^2 + \sigma_g^2 + \sigma_D^2,
\]

where

- \( \sigma_{PM}^2 \) is the total variance of the transit time of photomultipliers,
- \( \sigma_v^2 \) the variance of the transit time associated with the different initial velocity,
- \( \sigma_g^2 \) the variance of transit time due to different paths travelled by photoelectrons.

3.2. STUDY OF PHOTOMULTIPLIER TIME JITTER

Time jitter of fast photomultipliers is determined experimentally by the measurements of the single photoelectron time distribution spectrum. It is done by the illumination of the photocathode with subnanosecond light pulses of very low intensity to produce only single photoelectrons. The fwhm of the time spectrum of coincidences between the electrical pulses associated with the light pulse and single photoelectron pulses is a measure of the time jitter of the photomultiplier.

The measurements of the time jitter of a photomultiplier to be accurate enough need light pulses with a very short duration. Very often Cherenkov light generated in small radiators is used. The method of Kirkbride et al.\(^5\) applied also by Lynch\(^5\) employs single photon sampling of Cherenkov light excited by \(^{60}\)Co \( \gamma \)-rays which in coincidence excite a fast plastic scintillator coupled to a separate photomultiplier to produce a reference signal. Lamia et al.\(^54\) have proposed another method which uses the Cherenkov light generated in the common radiator. The single photoelectron time spectrum of two photomultipliers has been measured by simultaneous single photoelectron sampling of Cherenkov light. In ref. 29 the full Cherenkov light was used to produce a reference signal and attenuated light to release
single photoelectrons in the photomultiplier under investigation.

The other group of methods utilises artificial light pulse sources. Gas discharge light flash generators \(^7\) and light emitting diode generators \(^{35}\) are employed.

Recently reverse biased electroluminescence diodes driven by an avalanche transistor pulse have been used commonly as the source of accurately timed subnanosecond light pulses. It is estimated \(^{35}\) that the light pulse generated by the avalanche breakdown of a reverse biased light emitting diode fed by an electrical pulse having an fwhm of about 0.5 ns produces a light pulse with an fwhm below 200 ps. Unfortunately the emission spectrum of commonly used GaP light emitting diodes lies above 500 nm. Thus only a small overlap with the sensitivity spectrum of S-11 or bi-alkali photocathodes is achieved. It was shown in ref. \(^{51}\) that single photoelectron time resolution measured with the light emitting diode generator may differ significantly from that expected for 400 nm light wavelengths at the maximum of photomultiplier sensitivity.

The single photoelectron time resolution of fast photomultipliers was studied extensively in the past \(^{36,51-64}\). The transit time spread of photoelectrons and the time spread of electron multipliers have been determined by measuring the single photoelectron time jitter for a small spot at the centre of a photocathode \(^{51,53,57,59,62}\). A transit time spread difference has been studied with small spot scanning through the photocathode \(^{36,60,61}\). The influence of the electron multiplier alone may be only estimated from eq. (17). It was done in ref. \(^{24}\) for the XP 1021 photomultiplier. The standard deviation \(\sigma_D\), equal to 0.17 ns, has been found, assuming \(g_1 = g = 4\) and \(\sigma_D^2 = \sigma_{DD}\) calculated from the width of the single photoelectron response \(^{11}\).

The data on the time jitter of some fast photomultipliers used in timing experiments are collected in table \(^5\). The time jitter is expressed as the standard deviation found from the time spectra of the single photoelectrons. As the measured value may be affected by the type of light pulse used in the experiment the data are grouped depending on the type of light pulse source. The time jitter measured with the gas discharge light sources is corrected for a finite width of the light pulse. The data taken from the experiments with light emitting diodes are not corrected, as the fwhm of the light pulses is estimated to be smaller than 200 ps. Some photomultipliers showed the best single photoelectron time resolution for a focusing voltage different from that which gives the highest photoelectrons collection. This was observed for the RCA 8850 photomultipliers in ref. \(^{62}\) (see fig. 11). Fig. 12 shows an example of the time spectrum measured by Lescovar and Lo \(^{62}\) with the RCA 8850 photomultiplier for a 1.6 mm diameter spot in the centre of the photocathode illuminated by a light pulse (fwhm \(\leq 200\) ps) from the XP23 light emitting diode. Fig. 13 presents the time spectrum of single photoelectrons measured with the XP 22 light emitting diode generator for the C31024 photomultiplier \(^{53}\). The 6 mm diameter central part of the photocathode was illuminated.

![Fig. 11. Single electron time spread and relative collection efficiency of the RCA 8850 photomultiplier as a function of the voltage ratio between photocathode - focusing electrode and photocathode - first dynode. Full photocathode illuminated (from ref. 62).](attachment:image1)

![Fig. 12. Single electron time spread of the RCA 8850 photomultiplier with a 1.6 mm diameter area of photocathode illuminated (from ref. 62).](attachment:image2)
Table 5

Time jitter, $\sigma_{PM}(\text{ps})$, of some fast photomultipliers.

<table>
<thead>
<tr>
<th>Method of measurement</th>
<th>56AVP</th>
<th>XP1021</th>
<th>XP1210</th>
<th>Photomultiplier</th>
<th>RCA8575</th>
<th>RCA8850</th>
<th>C31024</th>
</tr>
</thead>
<tbody>
<tr>
<td>Čerenkov light</td>
<td>480$^b$</td>
<td>290$^f$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gas discharge light sources$^a$</td>
<td>510$^c$</td>
<td>220$^c$</td>
<td>190$^c$</td>
<td>280$^g$</td>
<td>380$^g$</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>610$^d$</td>
<td>390$^d$</td>
<td>220$^d$</td>
<td>310$^h$</td>
<td>300$^i$</td>
<td>440$^h$</td>
<td></td>
</tr>
<tr>
<td>LED generators</td>
<td>590$^e$</td>
<td>280$^e$</td>
<td>160$^e$</td>
<td>200$^j$</td>
<td>140$^k$</td>
<td>115$^k$</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>200$^l$</td>
<td>170$^c$</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>106$^i$</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>160$^m$</td>
<td>64$^p$</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>200$n$</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>140$o$</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^a$ Corrected for the width of the light pulses.
$^b$ Ref. 54.
$^c$ Ref. 63.
$^d$ Ref. 57.
$^e$ Ref. 58.
$^f$ Ref. 28.
$^g$ Ref. 59, small spot at the centre of the photocathode.
$^h$ Ref. 59, full photocathode.
$^i$ Ref. 66.
$^j$ Ref. 51.
$^k$ Ref. 62, small spot.
$^l$ Ref. 62, full photocathode.
$^m$ Ref. 61.
$^n$ Ref. 51, full efficiency of photoelectrons collection.
$^o$ Ref. 51, reduced efficiency of photoelectrons collection.
$^p$ Ref. 51, corrected for the contribution of the light pulse, fwhm = 200 ps.

The earlier discussed dependency of the transit time jitter of photoelectrons on the wavelength of the incident light was studied in ref. 51 for the 56 CVP photomultiplier having an S-1 photocathode. A significant increase of the time resolution above 30% was observed when the wavelength of the incident light was changed from 790 nm to 580 nm (see fig. 14). This gave experimental evidence that the time jitter resulting from the spread of the initial velocity of photoelectrons is proportional to the square root of the maximal initial energy of photoelectrons [see eq. (15)]. Based on this conclusion the measured time jitter of C31024, RCA8850 and XP2020 photomultipliers with the use of the XP22 light emitting diode at 560 nm light wavelength was recalculated to estimate the time jitter at 400 nm, near the maximum of the photocathode sensitivity (see table 6).

The influence of the transit time difference on the total time jitter of photomultipliers can be observed by measurements of the single photoelectron time resolution for the small spot at the centre of the photocathode and the full photocathode open for the incident light. It shows that this influence is larger for the photomultipliers with a first high gain dynode (see table 5). About a 30% increase of the time jitter is observed for the RCA8850 and C31024 photomultipliers when the full photocathode is illuminated$^{62}$.

The electron transit time difference, measured for the C31024 photomultiplier by Lescover and Lo$^{66}$) as a function of the position at the photocathode surface for non-optimised and optimised values of voltage ratio between the photocathode-focusing electrode and the photocathode-first dynode, is presented in fig. 15.

3.3. Pulse response of photomultipliers

The above discussed transit time spread of the electrons has also another consequence: it widens the response function of the photomultiplier to
very short light pulses; thus it introduces the frequency bandwidth limitation. The rise and the width of the output pulse can be determined by summing the variances in the transit time pro-
duced by the dispersion phenomena at the various points of the photomultiplier.

**Table 6**
The estimation of the time jitter at 400 nm of some fast photomultipliers from the measured time resolution at 560 nm (following ref. 51).

<table>
<thead>
<tr>
<th>Photomultiplier</th>
<th>fwhm(_a) (ns)</th>
<th>fwhm(_b) (ns)</th>
<th>fwhm(_c) (ns)</th>
<th>(E/k) (_d) (V/m)</th>
<th>(W_{hv}) (_d) (eV)</th>
<th>(\lambda = 400) nm fwhm(_b) (ns)</th>
<th>fwhm(_PM) (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C31024</td>
<td>0.25</td>
<td>0.15</td>
<td>0.15</td>
<td>13000</td>
<td>0.28</td>
<td>0.28</td>
<td></td>
</tr>
<tr>
<td>RCA8850</td>
<td>0.48</td>
<td>0.44</td>
<td>0.44</td>
<td>4400</td>
<td>0.84</td>
<td>0.84</td>
<td></td>
</tr>
<tr>
<td>XP2020</td>
<td>0.33</td>
<td>0.26</td>
<td>0.26</td>
<td>0.33</td>
<td>1.2</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td></td>
<td>0.51</td>
<td>0.47</td>
<td>0.24</td>
<td>7400</td>
<td>0.5</td>
<td>0.46</td>
<td>0.61</td>
</tr>
</tbody>
</table>

\(a\) Measured with the XP22 LED generator.
\(b\) Corrected for the contribution of the light pulse, fwhm = 200 ps.
\(c\) For the XP2020 corrected for the contribution of the time jitter introduced by the electron multiplier, fwhm = 2.36 \times 0.17 ns (following ref. 24).
\(d\) The threshold wavelength of the bialkali photocathode \(\sim 660\) nm.
\(e\) See eq. (15), fwhm = \(kd\Delta t\).
\(f\) Focused for the highest current pulse.
\(g\) Focused for the best time resolution of the single photoelectron time spectrum.
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In the timing theory\(^\text{11}\) the speed of the photomultiplier response is characterised by the single electron response which presents the shape of the photomultiplier output pulse generated by the single photoelectron reaching the first dynode. The variance \(\sigma_{\text{SER}}\) of the single electron response may be calculated assuming equal variances \(\sigma_{\text{DD}}^2\) of transit times of electrons between two successive dynodes, as follows\(^\text{11}\):

\[
\sigma_{\text{SER}}^2 = (n-1) \sigma_{\text{DD}}^2,
\]

where \(n\) is the number of stages of the photomultiplier.

Experimentally the shape and width of the single electron response can be determined by observing the shape of the noise pulses, or output pulses under single photon illumination at the sampling scope. Typical fast photomultipliers have \(\sigma_{\text{SER}}\) equal to \(\sim 1\) ns (see table 7).

The definition of the single electron response omits the influence of the variance of the transit time of photoelectrons between photocathode and first dynode on the output pulse, which is crucial when the time jitter of the photomultiplier is considered. In standard photomultipliers with 10–14 dynodes the speed of the output pulse is weakly affected by this variance. It is more pronounced in the case of the 5-stage C31024 photomultiplier. The rise time, equal to 0.8 ns, and fwhm = 1.8 ns of the single photoelectron pulse are increased to 1.2 ns and 2.7 ns, respectively, for multielectron pulses\(^\text{68}\).

The speed of the larger output pulses can be affected by the space charge effect in the last stages of multiplication. This effect was well observed for the C 31024 PM\(^\text{68}\). The fwhm of the output pulse, equal to 5 ns at 4 mA of the peak current, was increased to 8 ns at 7 mA. An influence of the space charge effect on the transit time jitter of photomultipliers at higher levels of photomultiplier illumination can also be expected\(^\text{11}\).

### 3.4. PHOTOELECTRON YIELD

The time resolution of the scintillation counter depends essentially on the number of photoelectrons released from the photocathode\(^\text{11}\). It is obvious that this number depends on both the light yield of the scintillator and properties of photomultipliers, as well as on the geometry of the scintillator or coupling with the photomultiplier. The relative light yield of several scintillators was discussed in section 2.4.

The properties of photomultipliers which influence the number of photoelectrons include the quantum efficiency of the photocathode and the overall collection efficiency of photoelectrons at the first dynode. The quantum efficiency of the typical S-11 photocathode (Sb–Cs–O) used in fast photomultipliers is equal to \(\sim 20\%\) at 420 nm light wavelength. Important progress has been made recently due to the application of bialkali photocathodes (Sb–K–Cs) with a quantum efficiency higher than 30\% at 400 nm. The bialkali photo-

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**Fig. 15.** Photoelectron transit time difference as a function of the position at the photocathode sensing area in RCA C31024 photomultiplier (from ref. 60).

**Table 7**

<table>
<thead>
<tr>
<th>Photomultiplier</th>
<th>(\sigma_{\text{SER}}) (ns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>56AVP</td>
<td>1.06(^a)</td>
</tr>
<tr>
<td>XP1021</td>
<td>1.2(^b)</td>
</tr>
<tr>
<td>XP1210</td>
<td>0.85(^c)</td>
</tr>
<tr>
<td>KP2020</td>
<td>0.93(^c)</td>
</tr>
<tr>
<td>C31024</td>
<td>1.55(^d)</td>
</tr>
<tr>
<td>X-P31024</td>
<td>1.0(^e)</td>
</tr>
<tr>
<td>X-P2007</td>
<td>0.75(^f)</td>
</tr>
</tbody>
</table>

\(^a\) See ref. 58.
\(^b\) See ref. 25.
\(^c\) See ref. 24.
\(^d\) See ref. 64.
\(^e\) See ref. 66.
\(^f\) See ref. 68.
TABLE 8

<table>
<thead>
<tr>
<th>Photomultiplier</th>
<th>NE 111 N/keV</th>
<th>Naton 136 N/keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>56AVP</td>
<td>0.44&lt;sup&gt;a&lt;/sup&gt;</td>
<td>0.73&lt;sup&gt;b&lt;/sup&gt;, 0.65&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>56DUVP</td>
<td>1.34&lt;sup&gt;c&lt;/sup&gt;</td>
<td>1.63&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>XP1021</td>
<td>0.9&lt;sup&gt;d&lt;/sup&gt;, 0.87&lt;sup&gt;c&lt;/sup&gt;</td>
<td>1.34&lt;sup&gt;c&lt;/sup&gt;, 1.04&lt;sup&gt;b&lt;/sup&gt; 1.2&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>XP1023</td>
<td>0.9&lt;sup&gt;c&lt;/sup&gt;</td>
<td>1.18&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>RCA8575</td>
<td></td>
<td>1.4&lt;sup&gt;e&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

<sup>a</sup> See ref. 67.
<sup>b</sup> See ref. 48.
<sup>c</sup> See ref. 49.
<sup>d</sup> See ref. 25.
<sup>e</sup> See ref. 22.

The cathode is characterised also by a low dark current. The collection efficiency of recent photomultipliers is about 90%. The pulse height and time fluctuations are also influenced by the homogeneity of the photocathode and the collection efficiency from different points on the photocathode. It was studied by Lescovar and Lo<sup>69</sup>) for several RCA photomultipliers. The results showed that the relative anode pulse height varies within ±20% and −5% for a distance ±18 mm measured from the centre of the photocathode of RCA 8850, RCA 8852 and C 31024 photomultipliers. The largest anode pulse variation occurred at the edges of the photocathode, due primarily to reduced collection efficiency.

The photoelectron yield of the photomultiplier for a given scintillator may be measured directly by the comparison of the mean value of the pulse height distribution of the single photoelectrons which determine the gain of the photomultiplier and the characteristic point at the energy spectrum of radiation detected in the scintillator: edge of the Compton spectrum for γ-rays<sup>48</sup>), full energy peaks of characteristic X-rays or conversion electrons<sup>49</sup>). As the measured number of photoelectrons characterise a given scintillation counter a rather large spread is found in published data (see table 8).

### 3.5. GAIN DISPERSION OF PHOTOMULTIPLIERS

The pulse height distribution of the output pulses from photomultipliers is caused to a large extent by the statistical fluctuations of the multiplier gain. The statistical properties of the multiplier may be evaluated by the analysis of the shape of the single photoelectron pulse height distribution<sup>50</sup>). In this way one can eliminate all contributions to the distribution of the output pulse originating from the photocathode and the input optics.

The multiplication of electrons in the multiplier is assumed to be a stochastic process, leading to a certain distribution of electrons at the anode characterised by<sup>50</sup>:
- a mean value of the multiplier gain,
- a relative variance.

If one assumes that the first stage of the multiplier has a gain <em>g</em><sub>1</sub> and that all the other stages have the same gain <em>g</em>, the overall gain and its variance may be represented by the following expression (for <em>n</em> ≫ 1 and <em>g</em> > 1):

<math>
\bar{G} = g_1 g^{n-1}
</math>

and

<math>
\sigma^2_\lambda = g_a \sigma_1 + \frac{g_2 \sigma}{g_1(g-1)},
</math>

where <em>g</em><sub>a</sub> and <em>g</em><sub>0</sub> are the variance of the gain of the first and successive dynodes.

Fig. 16 presents the example of a single photoelectron pulse height spectrum of a photomultiplier with the standard dynodes<sup>50</sup>). The mean pulse height of the single photoelectron is marked on the spectrum. For comparison, fig. 17 shows the pulse height spectrum of a few first photoelectrons from a photomultiplier with a first high gain dynode<sup>69</sup>). Note the well-defined peaks up to 5 photoelectrons. Owing to the high gain of the first dynode, typically <em>g</em><sub>1</sub> ∼ 50, the variance <em>\sigma^2_\lambda</em> of the single photoelectron distribution is strongly reduced.

![Fig. 16. Single photoelectron pulse height spectrum from a photomultiplier with the standard dynodes. The mean height of the single photoelectron is marked on the spectrum.](image-url)
4. Timing methods

The time resolution obtainable with a scintillation counter depends also on the method of derivation of the standard timing pulse from the output signal of the counter. The time derivation unit or time pick-off unit permit to minimise the time spread introduced by the scintillation counter. The time resolution contribution coming from the electronics circuits of the time pick-offs and the time-to-pulse-height converter (see fig. 1), is generally very small and will be omitted in the later discussion.

The most popular timing methods are the following:

a) leading-edge timing,
b) crossover timing,
c) constant fraction timing.

Leading-edge timing with a fixed threshold discriminator on the anode current pulse from the photomultiplier is the simplest time pick-off method. The leading-edge discriminator triggers at a fraction \( h = V_T / V_s \) for a signal pulse height \( V_s \). This fraction varies over the accepted range of signal to pulse height \( V_s \). Owing to its simplicity, leading-edge timing has a wide application in measurements with the narrow dynamic range of input pulse height. For a larger dynamic range the time walk effect may dominate the time resolution of the system.

To define the time walk consider a fast discriminator with a threshold \( V_T \) operating on detector signals of finite rise time. Although the two signals in fig. 18 were caused by events occurring at the same time \( t_0 \), the larger signal \( V_1 \) crosses the discriminator threshold before the smaller one \( V_2 \). This shift in the crossing time causes the output pulse of the discriminator to walk along the time axis as a function of pulse height. The walk is most noticeable for pulse heights close to the discriminator threshold.

The second walk effect is due to the charge sensitivity of practical discriminators. Even though the discriminator threshold has been exceeded, a finite amount of charge \( q \) is required to trigger the discriminator. The additional walk time \( \Delta t q \) needed to collect this charge increases as the signal decreases, and becomes very large for signals with amplitudes \( V_s \sim V_T \).

The fast crossover method (clipping stub technique) was developed to overcome the serious walk effect inherent with wide dynamic range use of leading-edge timing. In this method the anode current pulse is clipped with a short delay line to produce a bipolar pulse with a zero-crossing time. A fast crossover discriminator is used as time pick-off device. Since the zero-crossing time represents the same phase point for all pulse heights, the walk effect is almost entirely canceled. This system reduces walk very effectively and it provides better time resolution than leading-edge timing for large dynamic ranges. For a narrow dynamic range the time resolution is worse than with leading-edge timing because the discriminator fraction of crossover timing is usually 50%, which gives a large statistical time jitter of the scintillation counter.

A need to work with the optimal fraction in leading-edge timing and the excellent reduction of walk provided by fast crossover timing have stimulated design of a circuit which would trigger at...
5. Timing theories

The timing properties of the scintillators and photomultipliers discussed above, as well as the influence of the timing methods used to process the output signal, are considered and approximated in mathematical form by the timing theories\(^1,4,11,19,21,22,24,37,73,76-86\) to explain and predict the time resolution of the scintillation counter.

In 1950 Post and Schiff\(^76\) first discussed the limitations on resolving time that arise from the statistics of photon detection. They assumed that following the excitation of the scintillator by an energetic event the photomultiplier amplifies the primary photoelectrons without time spread and that the resulting output pulse is fed into a discriminator that detects when a definite number of photoelectrons, say \(n\), have accumulated. From Poisson statistics, they showed that the probability that the \(n\)th photoelectron occurs between \(t\) and \(t + dt\) is as follows:

\[
P_n(t) dt = e^{-NF(t)} \left[ NF(t) \right]^{n-1} NF'(t) dt \frac{n!}{(n-1)!},
\]

where \(NF(t)\) is the average number of photoelectrons detected in the interval between 0 and \(t\), following the exciting event at \(t=0\), \(F'(t) = dF/dt\), \(NF(\infty) = N\), 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 that the photomultiplier transit time spread \(P(t)\) is folded in with the photocathode illumination function \(I(t)\), i.e.,

\[
F'(t) = P(t) \ast I(t),
\]

where \(I(t)\) is the probability density function of arrival times of photons at the photocathode produced by an event at \(t=0\), \(P(t)\) is the probability density function of the transit time spread of electrons in the photomultiplier. The modified Post and Schiff theory was successfully applied by Lynch to predict the shape of the time spectra in a pulse shape discrimination circuit\(^21\) and it is used to predict the time resolution of a scintillation counter\(^22\).

Gatti and Svelto\(^11,77,78\) and Hyman et al.\(^1,19\) analysed independently the timing properties of scintillation counters and they obtained very similar analytical formulations. The model describing
the system under study can be characterised in the following way:

The timing properties of the scintillator are reflected by the probability density function of the photoelectrons generated at the photocathode, \( I(t) \). \( N \) is the mean number of photoelectrons per radiation impact on the detector. Hyman\(^{19}\) has assumed an \( I(t) \) of the following form:

\[
I(t) \sim e^{-\gamma t} - e^{-\gamma t},
\]

where \( \tau \) is the decay time constant of the scintillation pulse, \( \gamma = \tau / \tau_1 \), and \( \tau_1 \) is the rise time constant of the scintillation pulse.

The properties of the photomultiplier, according to Gatti and Svelto\(^{11}\) are contained in the single electron response function \( SER \). This function is characterised by the number \( A \) of electrons at the photomultiplier output, time \( h \), defined as the time between the emission of the photoelectron from the photocathode and the centroid of the output pulse, and by the mean square width \( \lambda \) of the SER pulse. The mean value of \( A \) is expressed as follows:

\[
A = g^{n-1},
\]

where \( g \) is the mean gain per stage of amplification, and \( n \) the number of dynodes.

The mean value of \( h \) is the sum of the mean transit times between successive dynodes:

\[
h = h_0 + h_1 + \ldots + h_{n-1},
\]

where \( h_0 \) is the transit time of photoelectrons between photocathode and first dynode, and \( h_1 - h_{n-1} \) are the transit times between successive dynodes.

The mean value of \( \lambda^2 \) is given by:

\[
\lambda^2 = \sigma_1^2 + \sigma_2^2 + \ldots + \sigma_{n-1}^2,
\]

where \( \sigma_i^2 - \sigma_{i+1}^2 \) are the variances of transit times between successive dynodes. If \( \sigma_i^2 \) are equal and denoted by \( \sigma_{dp}^2 \), then:

\[
\lambda^2 = (n-1) \sigma_{dp}^2.
\]

According to Hyman\(^{1,19}\) the single electron response can be described analytically by a clipped Gaussian function as follows:

\[
f_a(t) = \frac{1.1125}{(2\pi)^{1/2} \lambda} \left[ \frac{35}{32} \exp \left[ -\frac{(t-2.2166\lambda)^2}{2\lambda^2} - \frac{3}{32} \right] \right],
\]

for \( 0 \leq t \leq 4.332 \), and \( f_a(t) = 0 \), for \( t < 0 \) and \( t > 4.433 \). The single electron response is considered in the theories\(^{11,19}\) as rigid in shape but variable in height and time.

Applying Poisson statistics to the photomultiplier gain fluctuation\(^{79}\) one can find the variance \( \varepsilon_{\lambda}^2 \) is given by

\[
\varepsilon_{\lambda}^2 = \frac{g^2 g_o^2}{g-1^2},
\]

where \( g_o \) is the variance of the gain of the single stage.

A more elaborate equation for \( \varepsilon_{\lambda}^2 \) when the gain of the first dynode is larger than the other ones is presented in section 3.5 [see eq. (21)]. Hyman\(^{1,19}\) used the quantity \( r \) to described the gain dispersion as follows:

\[
r = (1 + \varepsilon_{\lambda}^2)^{1/2}.
\]

The statistical fluctuation of the transit time jitter of the photomultipliers was discussed in detail in sections 3.1 and 3.2. To describe analytically the probability density function of the time jitter of the photomultiplier Hyman used the clipped Gaussian function of the same form as for the single electron response [see eq. (29)], but with the standard deviation \( \sigma \).

All the statistical properties of the photomultiplier are included in the so-called “equivalent illumination function” \( I^*(t) \) of the first dynode as follows:

\[
I^*(t) = I(t) \ast f_o(t),
\]

where

\( I(t) \) is the probability density function of the photocathode illumination,

\( f_o(t) \) the probability density function of the transit time jitter of electrons in the photomultiplier.

The output current pulse from the photomultiplier is represented by the convolution integral as follows:

\[
i(t) = I^*(t) \ast f_a(t),
\]

where

\( I^*(t) \) is the equivalent illumination function [see eq. (32)],

\( f_a(t) \) single electron response function.

Finally, the theory of timing considers the type of pulse processing to define the so-called machine time at which the standard timing pulse is generated by an electronics unit. The most important methods of pulse processing are as follows:
a) Straight response—the time signal is generated when the photomultiplier current pulse exceeds a preselected threshold \( h \), which is measured as a fraction of the maximum height of the mean pulse of the current.

b) Integral response—the time signal is generated at the crossing of the integrated current pulse through a preselected threshold \( h \), which is measured as a fraction of the total charge of the mean pulse collected at the anode.

c) Centroid response—the time signal is referred to the centroid of a preselected amount of charge, collected at the anode. The fraction \( h \) is measured with reference to the total charge of the mean pulse collected at the anode.

Hyman\(^{19}\) has presented predictions of his theory as a series of plots representing the time resolution of the scintillation counter as a function of the triggering fraction \( h \) of the output pulses. The plots were calculated for different response types as well as for different sets of parameters characterizing the scintillation counter.

The standard deviation \( \langle \Delta t \rangle \) of the time measurement was normalised in the following way:

\[
\frac{\langle \Delta t \rangle N^{1/2}}{\tau \cdot r} = H \left( h, \frac{\lambda}{\tau}, \frac{\sigma}{\lambda}, \gamma \right).
\]  

(34)

Fig. 20 shows a family of curves according to eq. (34) for the integral response.

The Hyman theory\(^{19}\) in its original form was not successful to explain the time resolution of the scintillation counter\(^{11}\). The comparison with experiment was not completely reliable as the theory does not account for all the physical and geometrical characteristics of a scintillator\(^{87}\). This theory was modified in ref. 24 by the introduction of a new description of the photocathode illumination function, as follows:

\[
I(t) = f_G(t) \cdot e^{-t/\tau},
\]

(35)

where \( f_G(t) \) is the clipped Gaussian function describing the generation and collection of the light in the scintillator. As a consequence, the \( \sigma \) parameter in the theory representing the time jitter of the photomultiplier was redefined in the following way:

\[
\sigma^2 = \sigma_{PM}^2 + \sigma_{SC}^2,
\]

(36)

where \( \sigma_{PM}^2 \) and \( \sigma_{SC}^2 \) are the variance of the time jitter in the photomultiplier and the scintillator, respectively.

A discussion of experiments\(^{34}\) testing the modified Hyman theory is given in section 6.

Apart from the work of Gatti and Svelto\(^{11,77,78}\) and Hyman et al.\(^{11}\) and Hyman\(^{19}\), other calculations of the time resolution of the scintillation counter have been published based on others methods and assumptions. Euling\(^{80}\) and Donati et al.\(^{81}\) viewed the statistical events in photomultipliers as time-dependent branching process. El-Wahab and Kane\(^{82}\) calculated the differential probability of the arrival of the \( C \)th photoelectron under the assumption of exponential decaying light emission of the scintillator and of an exponential function of the transit time spread in the photomultiplier. More sophisticated calculations were obtained by adding two further exponential functions for the finite rise time of a scintillator light pulse and for the transit time spread\(^{83-85}\). They can be well fitted to experimental values for the dependency on triggering fraction without having to consider the various machine times. Sigfridsson\(^{86}\) used a similar statistical method.

The inductive method based on a Monte Carlo calculation was proposed by Cocchi and Rota\(^{37}\). It suggests a significant deviation from the theory of Gatti and Svelto and of Hyman when the number of photoelectrons released from the photocathode is reduced below 100. The influence of the pulse height selection on a real timing system was predicted by the calculations.
6. Time resolution study

The time resolution of scintillation counters has been studied experimentally for about thirty years. The progress in the best time resolution obtainable with scintillation counters is given in table 9. It was achieved by development of better scintillators and photomultipliers, by progress in the design of fast electronic circuits as well as by better understanding of the timing properties of scintillation counters.

During several years three problems were studied experimentally:
- the best time resolution obtainable with scintillation counters and its relation to predictions of the timing theories,
- dependency of the time resolution on the energy lost in the scintillator by nuclear radiation,
- the optimum of the pulse height fraction used to trigger electronics, or more generally the shape of the time resolution dependency on the pulse height fraction.

The problem of interpretation of the time resolution of the scintillation counter was not easy to solve in the past as the timing parameters of the counter were difficult to measure by straightforward methods. The large value of the optimal pulse height fraction observed experimentally was hard to understand in the light of the timing theories. Several experiments (3) concerning the time resolution versus the energy lost in the scintillator deviated from that predicted by the theory (11).

The recent study of the time resolution of scintillation counters presented in refs. 24 and 25 seems to explain several misunderstandings. The better knowledge of the light pulse shape from plastic scintillators has permitted to modify the Hyman theory and to get reasonable agreement between theory and experiment.

6.1. Time resolution versus energy

The dependency of the time resolution versus energy is essentially contained in $N$ — the average number of photoelectrons emitted from the photocathode. Due to the statistical nature of the emission process the number of photoelectrons is distributed around the average $N$ with a spread which is reflected in the pulse height resolution $\delta E$, which to a good approximation is proportional to $\sqrt{E}$.

When one investigates the dependency of time resolution on absorbed energy one must be careful to select the same fraction of the distribution of photoelectrons for all energies. This means that the energy window settings $\Delta E$ must be proportional to $\delta E$ which in turn is approximately proportional to $\sqrt{E}$.

The time resolution versus energy was studied in refs. 24 and 25 according to the above considerations using NE 111 scintillators and XP 1021 photomultipliers. Results of the experiment are shown in fig. 21. For the correct choice of the window settings the time resolution is proportional to $1/\sqrt{E}$ and gives an excellent agreement with the

![Fig. 21. Time resolution versus energy for two types of pulse height selection for leading-edge and constant fraction timing (from ref. 24).](image-url)
theoretical predictions given by Gatti and Svelto\(^{11,77,78}\) and Hyman\(^{1,19}\). For comparison, the results for a window \(\Delta E\) proportional to \(E\) are also shown which have been used by many authors\(^3,90\). Note also that the time resolution versus energy is independent of the energy window when constant fraction timing is used in the experiment.

6.2. Time Resolution Versus Pulse Height Fraction Used to Trigger the Electronics

The determination of the dependency of time resolution on pulse height fraction used to trigger has an essential meaning for the optimisation of the time resolution of scintillation counters. The measurements consist of a determination of the full width at half maximum (fwhm) of the time spectrum; the threshold of the fast discrimination is generally constant and variation of the pulse height fraction is affected by a calibrated attenuator\(^3\).

The value of the optimal pulse height fraction and more generally the shape of the time resolution dependency on the pulse height fraction determined for leading-edge timing may be directly compared with those predicted by timing theories. Thus it gives a sensitive test of the validity of theory. A number of experiments\(^3,24,25,40,87-90\) for various scintillators and photomultipliers have been performed to check the theoretical predictions regarding the time resolution.

In ref. 24 NE 111 and Naton 136 scintillators of different dimensions coupled to XP 1021 photomultipliers were studied. The experiments showed that the shape of the dependency of the time resolution on the pulse height fraction, and in particular the optimum value of the pulse height fraction, depend strongly on the type and dimensions of the scintillator. The comparison of the experimental and theoretical curves according to Hyman theory is given in fig. 22. The shape of the experimental curves measured for the different scintillators follows the shape of the theoretical curves calculated for different parameters of photomultipliers. All the curves are normalised according to eq. (34). As a conclusion of the above study the modification of the Hyman theory was proposed (see section 5), which was able to give agreement with the results of the experiments.

In ref. 25 the additional aspects of the comparison of the experimental and theoretical time resolution versus the pulse height fraction curves are considered. As was discussed in section 6.2, the experimentally observed time resolution is dependent on the statistical distribution of the average number \(N\) of photoelectrons at the output of the photomultiplier; therefore, the theoretical calculations are performed for the quantity

\[
H = \frac{\langle \delta \tau \rangle}{\tau} D^{-1}
\]

rather than for \(\langle \delta \tau \rangle\) itself,

![Graphs showing time resolution dependency on pulse height fraction for different scintillators and photomultipliers.](image-url)
where
\[ D \] is the standard deviation of the pulse height distribution,
\[ \langle \delta t \rangle \] the standard deviation of the time distribution.

In the Hyman theory (19) the \( D \) factor is defined as
\[
D = r/\sqrt{N},
\]
where \( r \) is a quantity describing the gain dispersion [see eq. (31)].

In experimental conditions the coefficient \( D \) can be determined from the energy resolution \( \delta E/E \) (fwhm) of the scintillation counter measured for the nuclear radiation, as follows:
\[
D = 1/2.36 \frac{\delta E}{E}. \tag{39}
\]
Recent data (20, 47, 48) on the photoelectron yield given by plastic scintillators coupled to photomultipliers lead to the conclusion that the dispersion \( D \) is not only determined by the average number \( N \) of photoelectrons, as in eq. (38), but also depends on other factors. So the values of \( D \) determined by eqs. (38) and (39) may differ substantially, even by factor of 1.5. In the conclusion of the study of ref. 25 it was shown that a normalisation to the energy resolution of the scintillation counter seems to be more reasonable, as it takes into account all properties of the scintillation counter.

The above problem is strongly related to the choice of the energy window width in the side channel when one makes a comparison of the experiments with the theory predictions. The theory considers the average number \( N \) of photoelectrons emitted from the photocathode which produces the Gaussian pulse height distribution at the output of the photomultiplier. Therefore, for the continuous energy distributions (Compton spectra, \( \beta \)-rays spectra), it can be approximated by a window width for the gating channel equal to the energy resolution of a monoenergetic peak of energy equal to the average one accepted in the window.

A study (25) of the time resolution as a function of the pulse height fraction performed following the above conditions for a 2 mm thick NE 111 scintillator coupled to a XP 1021 photomultiplier showed a quantitative agreement with the predictions of the modified Hyman theory. Fig. 23 presents a fit of calculated and measured curves of the time resolution versus the pulse height fraction. The experimental points were averaged for energies between 50 and 200 keV (from ref. 25).

### 6.3. The best time resolution

The experimental tests of the timing theories were generally performed for leading-edge timing, although the best time resolution was obtained with constant fraction timing. Fig. 24 presents a comparison of the time resolution versus pulse height fraction for leading-edge and constant fraction timing (24). It shows the important improvement of the time resolution even for the narrow dynamic range and the shift of the optimal pulse height fraction for the constant fraction timing. The geometrical difference of the curves shows the "walk" component which is cancelled by the constant fraction timing.

The best time resolution for the scintillation counters to date is from refs. 24, 49 and 106 (see table 9). The fwhm = 132 ps and slope \( t_{1/2} = 18 \) ps is shown on the prompt time spectrum in fig. 25, measured with 1 cm thick and 2.5 cm diameter NE 111 scintillators coupled to XP 1021 photomultipliers for 15% energy windows at 930 keV (\( ^{60}\)Co source) (24). Fig. 26 shows the effectiveness of the constant fraction timing for a very wide dynamic range.
Fig. 24. Comparison of the time resolution versus pulse height fraction curves for leading-edge and constant fraction timing. The geometrical difference of the curves shows the "walk" component.

Table 9

Progress in the best time resolution of the scintillation counters measured with the $^{60}\text{Co}$ source.

<table>
<thead>
<tr>
<th>Year</th>
<th>Time resolution$^a$ (ps)</th>
<th>Scintillators</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1952</td>
<td>2000</td>
<td>Stilbene</td>
<td>103</td>
</tr>
<tr>
<td>1963</td>
<td>320</td>
<td>Naton 136</td>
<td>3</td>
</tr>
<tr>
<td>1964</td>
<td>240</td>
<td>Naton 136</td>
<td>56AVP 3</td>
</tr>
<tr>
<td>1964</td>
<td>210</td>
<td>Naton 136</td>
<td>56AVP 104</td>
</tr>
<tr>
<td>1965</td>
<td>225</td>
<td>Naton 136</td>
<td>XP1020 90</td>
</tr>
<tr>
<td>1966</td>
<td>160–200</td>
<td>Naton 136</td>
<td>RCA7850 105</td>
</tr>
<tr>
<td>1966</td>
<td>185</td>
<td>Naton 136 XP1020</td>
<td>XP1020 87</td>
</tr>
<tr>
<td>1970</td>
<td>132</td>
<td>NE 111</td>
<td>C70045A 94</td>
</tr>
<tr>
<td>1971</td>
<td>130</td>
<td>NE 111 XP1021</td>
<td>XP1021 24</td>
</tr>
<tr>
<td>1972</td>
<td>130</td>
<td>NE 111 XP1021</td>
<td>XP1021 49</td>
</tr>
</tbody>
</table>

$^a$ For two counters.

Fig. 25. Prompt time spectrum from $^{60}\text{Co}$ source obtained with the NE 111 scintillators on XP 1021 photomultipliers (from ref. 24).
range of the energy; the fwhm = 195 ps was measured with 90% of the $^{60}$Co Compton spectrum with the NE 111 scintillators and the XP 1021 photomultiplier (from ref. 49).

The obtainable time resolution of a scintillation counter depends also on the size of the scintillator. The light collection process introduces a time spread which could be larger than the time jitter of fast photomultipliers (see sections 2.2.1. and 2.5). The self-absorption and re-emission process affect the decay time of scintillating pulses and according to ref. 40 cause deterioration of the time resolution. The influence of all these processes on the light pulse shape is reviewed in section 2.2. Table 10 presents the time resolution normalised to the energy measured with NE 111 and Naton 136 scintillators of different thickness. Note that the time resolution growth is by a factor 1.5 when the height of the scintillator is increased from 2 mm to 2 cm.

7. Further prospects on better timing characteristics of scintillation counters

The discussed above properties of scintillation counters showed that at present a time resolution slightly larger than 100 ps for high-energy $\gamma$-rays ($^{60}$Co source) was achieved using XP 1021 photomultipliers and 2.5 cm diameter and 1 cm high NE 111 scintillators. For lower energies one can note the 330 ps time resolution for 50 keV electrons measured in coincidence with high-energy $\gamma$-rays. All the studies showed that both scintillators and photomultipliers are the source of time uncertainties. Besides, the time resolution can be strongly limited by the size of the scintillator because of the light collection process and the time spread of the nuclear radiation interaction in the volume of the scintillator. To improve the time resolution of scintillation counters one has to expect further development of faster scintillators and photomultipliers.

7.1. SCINTILLATORS

According to the theory of the light pulse generation process in organic scintillators the speed of the light pulse depends on the composition of the scintillators as follows:

- for a given solvent and solute of the binary system the speed of the light pulse improves with the increase in the solute concentration owing to a faster energy transfer,
- the solute should have a high solubility, a short fluorescence lifetime and a high fluorescence quantum yield to increase light yield,
- the solvent should have a high yield of the excitation of the lowest singlet state $^1X$,
- binary scintillators are generally faster than ternary ones.

<table>
<thead>
<tr>
<th>Scintillator</th>
<th>Dimensions</th>
<th>fwhm $E^{1/2}$ a</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>diam. (cm)</td>
<td>height (cm)</td>
</tr>
<tr>
<td>Naton 136</td>
<td>2.5</td>
<td>2.0</td>
</tr>
<tr>
<td></td>
<td>2.5</td>
<td>1.0</td>
</tr>
<tr>
<td></td>
<td>2.5</td>
<td>0.2</td>
</tr>
<tr>
<td></td>
<td>1.25</td>
<td>0.2</td>
</tr>
<tr>
<td>NE 111</td>
<td>2.5</td>
<td>2.0</td>
</tr>
<tr>
<td></td>
<td>2.5</td>
<td>1.0</td>
</tr>
<tr>
<td></td>
<td>2.5</td>
<td>0.2</td>
</tr>
<tr>
<td></td>
<td>1.25</td>
<td>0.2</td>
</tr>
</tbody>
</table>

a Time resolution of one counter measured with constant fraction timing.
Based on the above criterion the NE 111 plastic scintillator was developed and is the fastest one on the market. It contains 40 g/l of PBD dissolved in polyvinyltoluene (PVT). A further increase of the solute concentration in the NE 111 solution was not possible because of the solubility limit of the PBD in PVT. Lyons and Hocker\textsuperscript{108} have proposed to exchange PBD in NE 111 scintillator for butyl-PBD which has a larger solubility in PVT. The study\textsuperscript{33} of the speed of the light pulse from 50 g/l, 100 g/l and 150 g/l of butyl-PBD in PVT samples seems to suggest an improvement of the speed of the scintillator response. A similar study performed in ref. 29 by the single photon method showed no difference between the light pulses from the above samples and NE 111 plastic.

The method used by Lyons and Hocker\textsuperscript{108} reflects new possibilities for better organic scintillators. Many molecules with a high fluorescence efficiency and short decay times but which were relatively insoluble have been made soluble by a substitution of alkyl or oxa-alkyl groups for hydrogen atoms at suitable locations in the molecule\textsuperscript{109}. Among the solutes to which this solubilization technique has been applied, the p-oligophenylene series is particularly promising\textsuperscript{109}). It has been shown that the decay time varies inversely with the number of phenyl rings in the molecule and that the light yield has been larger for the longer molecules. The BIBUQ solution in toluene or xylene is an example of this family of scintillators\textsuperscript{20}).

The studies reported in refs. 24 and 25 seem to suggest another limitation of the scintillation pulse speed which is associated with the solvent itself. The relatively slow initial rise of the light pulses from unitary and binary scintillators was considered to result from de-excitation of several higher levels of the solvent before the excitation of the lowest singlet state \( ^1X \). It suggests to analyse the solvent properties in the light of this hypothesis.

As the conversion efficiency of even the best organic scintillators is only about 3\% one would expect improvements by further investigation of solvents and fluors.

### 7.2. PHOTOMULTIPLIERS

The last years have brought important progress in better constructions of fast photomultipliers. The bialkali photocathode with \(~\sim 50\%\) larger quantum efficiency was introduced. High gain dynodes (mainly the first one) reduce strongly the contribution of the electron multiplier to the total time jitter of the photomultiplier. The best time resolution published up till now is still obtained, however, with the standard XP1021 photomultipliers\textsuperscript{24,49,106}). Sipp and Miehe\textsuperscript{40} used the RCA 8850 photomultiplier with a high gain first dynode but the time resolution study was performed mainly with the common scintillator coupled to both photomultipliers. Thus the time resolution, equal to 176 ps measured with \( \beta \)-rays of energy about 290 keV (equivalent to Compton edge of 511 keV annihilation \( \gamma \)-rays from a \(^{22}\text{Na} \) source), can not be compared directly to the results obtained in the coincidence experiments. In such an experiment with \( \gamma \)-rays from a \(^{22}\text{Na} \) source they got fwhm = 220 ps, thus comparable to that of the XP 1021 photomultiplier\textsuperscript{24}). Very preliminary experiments\textsuperscript{110} performed with the C31024 photomultiplier seem to suggest that a better time resolution can be obtained for energies lower than 100 keV. For higher energies the space charge effect may limit the speed of the photomultiplier.

A further progress of fast photomultipliers will probably be associated with the application of microchannel plates as the electron multiplier.

Although, photomultipliers with channel plates are still in the development stage the microplates themselves are already used in several timing experiments to detect heavy ions or electrons\textsuperscript{111,112} showing excellent timing capabilities. Girard and Bolore\textsuperscript{122} studied an arrangement which consisted of two microchannel plate multipliers detecting electrons from a carbon foil. Fission fragments of \( \alpha \)-particles from a \(^{252}\text{Cf} \) source impinge on the foil at an incidence of 45\°. The time resolution of the arrangement was 87.5 ps for fission fragments and 117 ps for \( \alpha \)-particles. Owing to the basic symmetry of the experimental device one can estimate the time resolution of one counter to be equal to 62 ps and 85 ps for fission fragments and \( \alpha \)-particles, respectively.

Development photomultipliers with channel plates were studied by several authors\textsuperscript{57,113-116}). Recently Lo et al.\textsuperscript{115} have performed a general study of performance characteristics of two development photomultipliers with the microchannel plates LEP HR350 and HR 400. These photomultipliers have curved high gain microchannel plates to avoid ion feedback. Proximity focusing is used for the input and collecting stages. The HR 350
photomultiplier has an S-20 photocathode with a useful diameter of 13 mm and it incorporates a coaxial anode. The HR 400 has a 15 mm diameter S-20 photocathode and a simple plate as anode. The gain of each tube was $10^6$. The measured timing characteristics showed better figures for the HR 350 photomultiplier. The SER fwhm equal to 1.25 ns and 1.4 ns, and rise time equal to 0.64 ns and 0.9 ns, were determined for HR 350 and HR 400 photomultipliers, respectively. The measured single photoelectron time spread fwhm $\leq 200$ ps was limited by the width of the light pulse from the light emitting diode.

Taking into account a high quantum efficiency equal to $\sim 20\%$ and anode pulse peak current linear up to 260 mA, all together show that the microchannel plates photomultipliers exhibit very good timing capabilities, superior to the conventional ones with discrete dynodes.

7.3. OTHER LIMITATIONS OF THE TIME RESOLUTION

Faster scintillators and photomultipliers which might be expected in the future will improve the time resolution, but still it will be limited by the light collection process and the spread in time of flight of nuclear radiation. It was discussed in section 2.2.1 and reported in refs. 24 and 38. Therefore the problem to choose a scintillator size, a small one for high time resolution or a big one for larger detection efficiency, will be even more severe than today.

The low efficiency of organic scintillators for $\gamma$-rays makes a need to find other ones based on materials with a high Z. For X-rays and low-energy $\gamma$-rays ($<100$ keV) detection the standard plastics and liquid scintillators are loaded with lead or Sn admixtures. The effective light may be larger, however, because of detection of X-rays in the loaded plastics due to the photoeffect. The usefulness of the loaded plastics is limited to low-energy $\gamma$-rays and unfortunately nothing is gained for high-energy $\gamma$-rays. A need for fast inorganic scintillators arises at the moment. The NaI(Tl) and NaI scintillators give a time resolution $36,118-120$ several times worse than that measured with the best plastics.

By arranging plastic and NaI(Tl) detectors so that the Compton scattered $\gamma$-rays escaping from the plastic are absorbed in the NaI(Tl) it is possible by external summing to obtain well-resolved full energy peaks as demonstrated in ref. 121. Kluge and Thomas$^{122}$ have performed measurements in the picoseconds region by centroid shift self-comparison using combined plastic–NaI/Tl detectors. Because of geometry effects, the delay of light collection in the plastic detector is energy dependent and this effect was carefully studied. One could imagine future systems based on microchannel photodetectors with mosaic anodes to compensate for light collection problems.

A very fast response of a multicrystalline ZnO(Ga) scintillator was reported by Luckey$^{123}$. A decay time equal to 0.34 ns and a fwhm equal to 250 ps were recently measured by Tirsell et al. $^{22}$. However, because of the multicrystalline structure of this scintillator very thin samples for $\alpha$-particle detection must be used at present. The light yield for 5 MeV $\alpha$-particles is larger than that of plastics, but there is no well-defined energy peak in the spectrum$^{27}$.

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