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MEASUREMENT OF THE INSTRUMENT RESPONSE FUNCTION IN THE SINGLE PHOTOELECTRON TECHNIQUE

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Luminescence decay time measurements are generally performed using the single photoelectron technique. In the case of ionizing particle induced luminescence the determination of the true decay curve requires the precise knowledge of the instrument response function $R(t)$. In this paper the measurement of $R(t)$, using the Čerenkov effect is described.

1. Introduction

Besides the phase shift method the monophoton technique (photon sampling or single photoelectron technique) is now widely used to determine fluorescence decay time and measurements of values as low as 0.5 nsec have been reported[1]). In this method the experimental information $H(t)$ can be considered as a superposition integral :

$$H(t) = \int_0^t i(s)\, F(t-s)\, \mathrm{d}s, \qquad (1)$$

where $i(s)$ is the true decay function to be measured and $F(t-s)$ a function which depends on the instrument response function $R(t)$ and the excitation function $E(t)$ giving the number of molecules undergoing a transition to a excited level at time t. More precisely $F(t)$ can be written as a convolution between the functions $R(t)$ and $E(t)$:

$$F(t) = \int_0^t R(s)\cdot E(t-s)\,\mathrm{d}s\,.$$

If the luminescence of the studied medium is induced by the light emitted by a flash lamp, the true decay curve $i(t)$ can easily be obtained by the following procedure. After the measurement of $H(t)$, in a second stage, $F(t)$ is measured simply by replacing the fluorescent medium by a mirror. Extraction of $i(t)$ from the experimental curve $H(t)$ is then achieved by a process of synthesis described in several papers[2–3]).

However, if the luminescence is induced by ionizing particles the method just described is no longer applicable. In this case $E(t)$ reduces just to a Dirac delta function and the extraction of $i(t)$ necessitates knowledge of $R(t)$.

In this paper we describe a simple method, using the Čerenkov effect produced by β particles, of measuring $R(t)$. Application of this method will considerably improve the accuracy of scintillation decay measurements. Moreover, knowledge of the instrument response function can greatly facilitate progress toward better time resolutions with the monophoton technique.

In a first part of this paper, the principle of the monophoton technique of measuring short decay times is briefly recalled. The apparatus used to measure $R(t)$ is then described and finally some preliminary results are presented and discussed.

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2. The monophoton technique

Proposed by Thomas and Bollinger[4]) this sampling technique is essentially based on the use of fast photomultipliers. Let us first consider a photomultiplier used in a classical manner. If the light pulses produced by a luminescent medium, excited by a recurrent short excitation source (ionizing particles or short UV flashes) are detected by a photomultiplier the shape of the electrical pulses $h(t)$ delivered by this detector can be described by a convolution between $i(t)$ true decay function of the medium under study, $e(t)$ excitation function and $r(t)$ the photomultiplier response function:

$$h(t) = i(t) * e(t) * r(t)\,.$$

Hence, in order to measure $i(t)$, a simple method would be, $r(t)$ being known through an independent experiment, to deconvolute $h(t)$. Unfortunately there exist important fluctuations in the shape of the electric pulses and an average of numerous pulses has to be taken. Moreover their full width at half maximum is by no means negligible and hence very short decays are difficult to evidence.

There is however other information contained in the anodic electrical pulses of the photomultiplier which

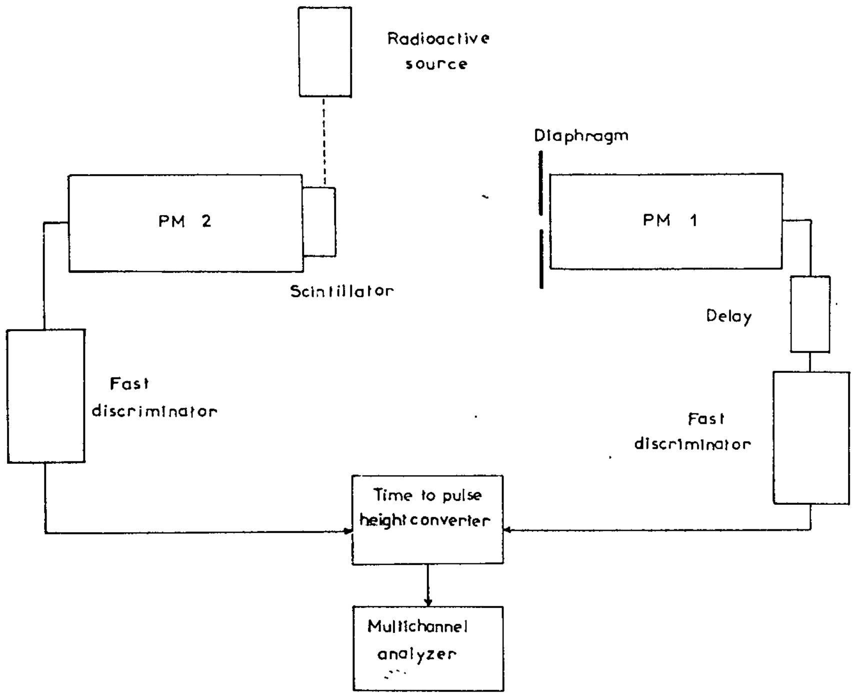

Fig. 1. Block diagram of the equipment used to measure the decay of ionizing particle induced luminescence.

is much lesser subject to fluctuations: the time t_1 connected to the first order moment of the pulse and defined by the following relation:

$$t_1 = \int_{-\infty}^{+\infty} t \cdot h(t)\,dt \Big/ \int_{-\infty}^{+\infty} h(t)\,dt.$$

The monophoton technique takes advantage of this property of the fast photomultipliers. The method consists in measuring the time delay between a monitoring signal, synchroneous of the excitation of the luminescence and the detection of a single photon, taken at random among those forming the light pulse. Depending on the mode of excitation — UV light flash or ionizing particle — the monitoring signal is either the electric pulse accompanying the discharge in the flash lamp or the pulse delivered by a second photomultiplier (P.M. 2) coupled to a scintillator. The conditions of detection of a single photon by the first photomultiplier are easily achieved by attenuating sufficiently with a diaphragm or neutral filters the intensity of light reaching the photocathode. By simple probability considerations it can be shown[5]) that the histogram of these time intervals is related to the true decay function $i(t)$ through eq. (1).

The experimental set-up used in the case of the ionizing particle induced luminescence is shown on fig. 1. The scintillation produced by the ionizing particle in the luminescent medium is detected by two photomultipliers one of which is shielded by a diaphragm in order to release only single photoelectrons on the cathode (P.M. 1). The electrical pulses delivered by the two detectors are shaped by fast discriminators. The leading edge of the resulting pulses is then connected, in one manner or another depending on the type of discriminator used, to the time t_1 defined above. The spread in transit time of the electrons in the photomultiplier 1 and the fluctuations of the starting time of pulses in the discriminators are the two main factors which limit the time resolution of the apparatus. The time intervals are measured with a time to pulse height converter and the results analyzed with a 400 channel analyzer.

3. The Čerenkov effect as a very short recurrent light source

The Čerenkov effect, i.e. the light emission produced by a charged particle whose velocity is greater than the phase velocity of light in the medium, has been widely used to detect nuclear particles[6]). Its use as a very short light source is less common because of the very low intensity of the emission. However this fact is of no inconvenience in the case of the monophoton

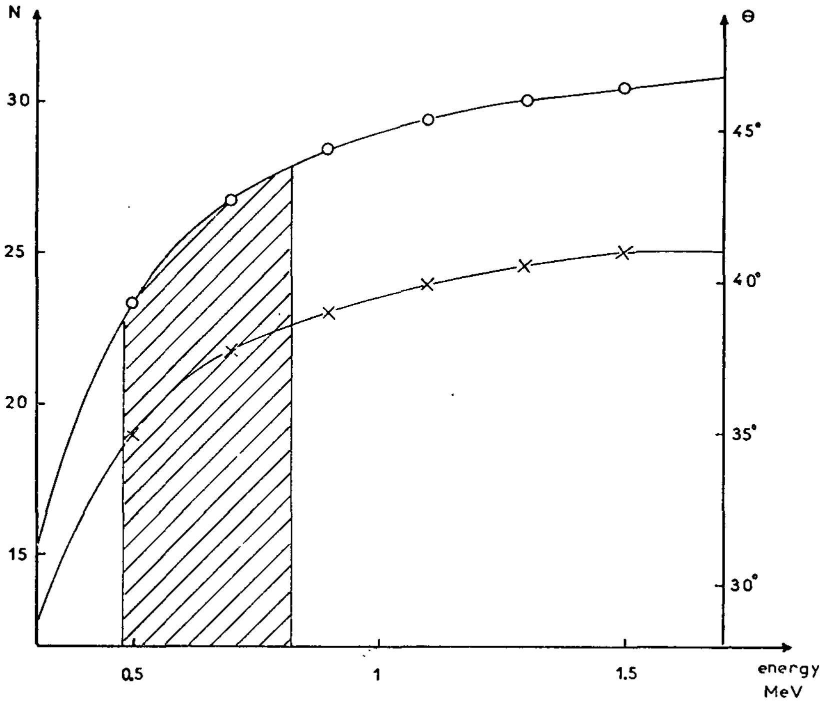

Fig. 2. The variations of the Čerenkov angle θ (○) and the radiation yields N (×) in plexiglass (thickness: 1 mm, spectral region: 350–550 nm) as functions of the kinetic energy of electrons. The values to be considered with our equipment are located in the shaded area.

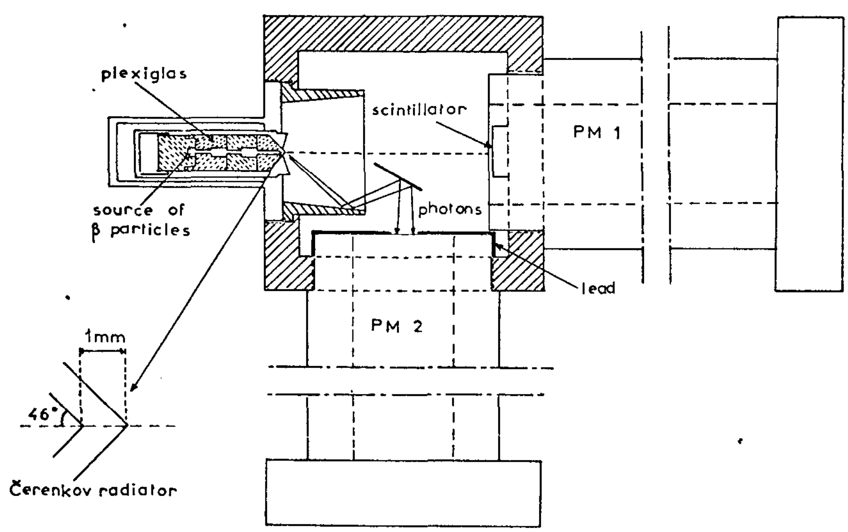

Fig. 3. Experimental set-up used to measure the instrument response function.

technique which requires only a very low photon flux.

Let us recall some characteristic properties of the phenomenon. The radiation is only observed at a particular angle θ with respect to the track of the particle, whose value is given by the Čerenkov relation:

$$\cos\theta = 1/\beta n\,, \qquad \text{with } \beta = v/c\,,$$

where v represents the velocity of the particle and c the phase velocity of the light in the medium. The radiation yield, i.e. the number of photons N emitted by an electron within a spectral region defined by wavelengths λ_1 and λ_2 is equal to:

$$N = 2\pi\alpha l\left(\frac{1}{\lambda_2}-\frac{1}{\lambda_1}\right)\sin^2\theta\,,$$

where α is the fine structure constant ($e^2/\hbar c = 1/137$), n the average refractive index of the medium and l the thickness of the radiator used. Fig. 2 shows the variation of N and θ as a function of the energy of electrons in the case of a plexiglass radiator (thickness $= 1$ mm, $\lambda_1 = 550$ nm, $\lambda_2 = 350$ nm).

As regards the duration of the light pulse two effects must be considered. First the variation of the refractive index with the wavelength, the effect of which can be evaluated by the following expression giving the duration Δt of the light pulse observed along a line parallel to the axis of the particle, at a distance ρ from this axis:

$$\Delta t = (\rho/\beta c)(\tan\theta_2 - \tan\theta_1)\,, \qquad (2)$$

where θ_1 and θ_2 are the Čerenkov angles corresponding to the frequency limits of the light receiver. In the present experimental conditions ($\rho = 8$ cm, $\theta_1 = 44°\,15'$, $\theta_2 = 43°\,50'$):

$$\Delta t = 1.4\times10^{-12}\,\text{sec}\,.$$

A second effect to be considered is the Coulomb scattering during the passage of the particle through the medium. It can be characterized by a spread in the observed angular distribution of the Čerenkov effect. The mean square angle of scatter is given by[6]):

$$\langle\theta^2\rangle = (E_s^2/p^2\beta^2)t\,,$$

where p is the momentum of the particle in MeV/c, E is a constant ($= 21$ MeV) independent of the mass of the particle and the nature of the medium and t is the path length measured in units of radiation lengths X_0 ($X_0 \approx 43$ g/cm^2 for plexiglass). Considering a particle of 0.8 MeV and a radiator of plexiglass of 1 mm thickness one finds:

$$\langle\theta^2\rangle^{\frac{1}{2}} \approx 52°\,.$$

In view of this result one is tempted to use a relation of type (2) and conclude to a great value for Δt. However considering the geometry of the experimental set-up used, only photons corresponding to Čerenkov angles θ between 39° and 44° can reach the photocathode. Consequently the duration of the light pulse cannot exceed 5×10^{-11} sec which is small compared to the estimated time resolution of the monophoton apparatus.

4. Description of the experimental set-up

The experimental set-up is represented on fig. 3. A beam of β particles emitted by a collimated source of ^{32}P (pure β emitter, $E_{max} = 1.7$ MeV, activity: 3–30 mCi) passes through a conical plexiglas radiator (see details on fig. 3) of 1 mm thickness. The photons produced by Čerenkov effect are collected on the photocathode of a 56 UPV Amperex photomultiplier (P.M. 2) by means of a conical and a plane mirror. Detection of bremsstrahlung X rays or scattered β particles is avoided by use of a lead shield (thickness: 2 mm). Only the half of the light beam was used as

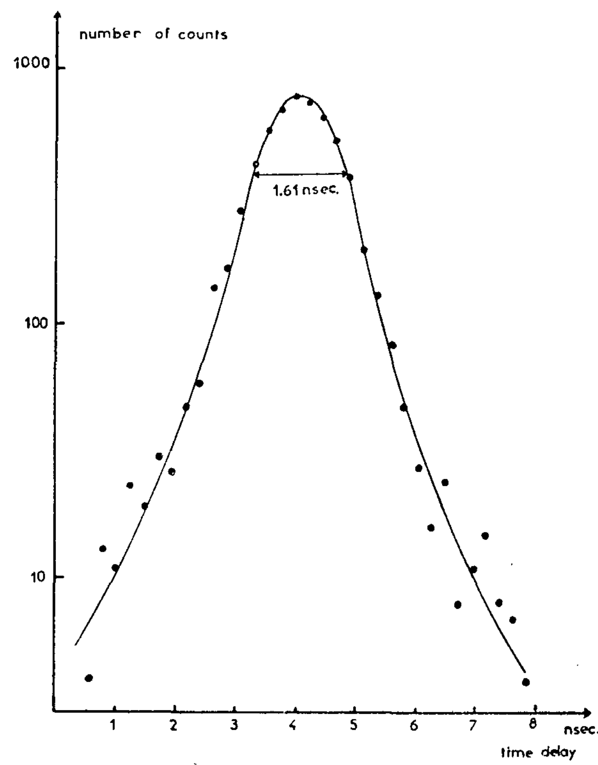

Fig. 4. Instrument response function.

can be seen on fig. 3. After their passage through the radiator the β particles are detected by a plastic scintillator NE 102 coupled to another photomultiplier (P.M. 1). In order to prevent scintillation photons from reaching P.M. 2, the plastic scintillator is covered by a thin opaque vacuum deposited aluminium film. The electric signals delivered by the two photomultipliers after being shaped by fast discriminators (chronetics, model 101) are fed in a time to pulse height converter (chronetics, model 105) whose pulses are analyzed with a 400 channel pulse height analyzer (RIDL).

5. Results

Fig. 4 shows the instrument response function $R(t)$ obtained by accumulating the pulses in the multichannel during 4 h. The shape is approximately Gaussian as can be deduced from a plot of log $R(t)$ versus $(t-t_0)^2$, t_0 being the abscissa corresponding to the peak of the instrument response function. More precisely, in the case of the considered apparatus, $R(t)$ can be described by the relation:

$$R(t) = C\exp[-(t-t_0)^2/K^2],$$

with $K = 1.15$ nsec, C being a constant. The full width at half maximum is 1.61 ± 0.05 nsec. This value in good agreement with a result of Pfeffer[7]), which measured the response function (fwhm = 1.6 nsec) using a mercury contact relay (Clare) and assumed a very short duration for the light emission of this source. Our result confirms thad indeed this must have been the case.

It must be remarked that without the lead shield of P.M. 2, $R(t)$ is no longer symmetrical, the part corresponding to the positive values of $t-t_0$ being enhanced. This effect is probably due to scattered β particles or bremsstrahlung X rays reaching the P.M. 2.

The number of Čerenkov photons detected is surprisingly low. In a typical experiment, 10^5 β particles per minute reached the plastic scintillator. During the same time interval 437 pulses delivered by P.M. 2 and in coincidence with the monitor signals, were recorded. Assuming that 10% of the β particles passing through the radiator possess an energy above the Čerenkov threshold and considering the quantum yield of the photocathode ($\approx$ 10%), a value of 0.43 for the number of Čerenkov photons detected per incident β particle, is obtained. This number is to be compared to the expected mean value of 5.4 photons reaching the photocathode of P.M. 2 per β particle. The latter value is obtained, taking in account the effect of the losses of light due to absorption in plexiglass (transmission coefficient $\approx$ 0.7), the reflection on the mirrors (reflectance $\approx$ 90%) and the plexiglass-air interface (losses $\approx$ 4%), on the initial mean light output of 20 photons/β particle. Thus an efficiency of 8% for the light collection can be deduced. The main cause of this low yield seems to be the light losses at the plexiglass-air interface which has to be very thoroughly polished. Further work devoted to the improvement of the light collection is in progress.

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