Time resolved fluorescence emitted by organic detectors under ions beams irradiation

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Context and objectives:

Our current work deals with the measurement of the fluorescence of solid organic scintillators under particles irradiation. The radio-induced fluorescence is strongly related with the Linear Energy Transfer (LET) of these particles. Our group in Strasbourg is involved in the inderstanding of the mechanisms which lead to the production of fluorescence. In this context we have developped original and unique experimental setup to measure the time resolved fluorescence at the nanosecond level. Indeed it is well known that the radio-induced fluorescence is composed by two parts (1): a “prompt part” and a “delayed part”. The prompt part is due to direct excitation, in a singlet state, of the scintillator. The delayed part is due to recombination phenomena of electron-hole pairs, called polaron in organic medium. The delayed part is intimately linked with ionisation and then increases with the LET particles. This delayed part is the key to develop new detectors with high energy discrimination.

Our previous work has been made under proton and alpha irradiation up to 4 MeV. To expand our database we need to irradiate our samples with heavier particles. Our goal is first to improve the models proposed to describe the radio-induced fluorescence by taking into account the prompt and the delayed parts. Almost no experimental data on these two parts exist in litterature for a given solid organic scintillator. Secondly one of our goals is to developp new solid organic detectors for specific applications in neutron-gamma discrimination (2), hadrontherapy and radioprotection dosimetry.

Radio-induced Fluorescence :

The total fluorescence intensity can be express as (1) :

$I_t(t) = I_p(t) \otimes I_d(t)$.

Where I_t is the observable, I_p and I_d are the prompt and delayed part respectively. We can express I_p as:

$$I_p(t) \propto \exp\left(\frac{-t}{\tau_s}\right).$$

where τ_s is the fluorescence decay time of the scintillating molecule. We assumed that the prompt part of the decay is due to direct excitation without pair creation.

For the delayed part, considering R(t) as the recombination rate of (e^--h) pairs and by ρ_s the probability of this recombination to be in a singlet state we can write:

$$I_d(t) \propto \exp\left(\frac{-t}{\tau_s}\right).R(t).\rho_s .$$

R(t) can be express as: $R(t) \propto At^{-3/2} + Bt^{-1} + Ct^{-2}$.

Where $At^{-3/2}$ represents the contribution of the diffusion process which mainly occurs in liquids medium. Bt^{-1} is a charge transfer term which occurs in the low energy deposition regions and can be assimilated to geminate recombination. Ct^{-2} is a bimolecular charge transfer term and can be assimilated to a triplet-triplet annihilation term. These two last terms are bounded to the charges mobility in the medium. Howard and co clearly showed that molecular structure is a main factor to explain the relative importance of these two last phenomenons (3).

Time correlated single photon counting (TCSPC) method

We present here, the experimental set up developed in our Institute at Strasbourg. The radiation source is a Van de Graaff particles accelerator, which allows us to get protons and alphas beam with an energy between 1 to 4 MeV. As shown in fig 1, we used a so called “flash lamp”, which allows us to obtain a nanosecond pulsed beam.

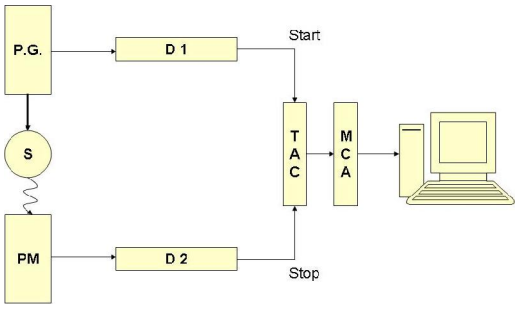

Fig. 1: Charged particles beam experiment : Block-diagram of time resolved fluorescence measurement apparatus (S : sample ; P.G. : pulse generator ; PM : photomultiplier (Hamamatsu R3235-01); D1 and D2 (ORTEC 584): discriminator 1 and 2 ; TAC : time-to-amplitude converter (TAC ORTEC 566) ; MCA : multichannel analyser).

The pulse of the flash lamp after discrimination (D1) opens a time gate at t_0 in the time-to-amplitude convector. The first fluorescence photon detected by the photomultiplier, working under single photon counting conditions, after discrimination (D2), stops the time gate at a time t. The time-to-amplitude converter yields signals with amplitude proportional to $\Delta t = t - t_0$, which are finally analysed by the multichannel analyser (MCA). Data are accumulated as long as necessary to get an histogram of Δt with a sufficient statistical accuracy. Using this experimental set-up, the flourescence intensity I(t) is analysed over a microsecond timescale, with a time resolution of 1 ns. This set-up can be adapted to any kind of accelerators.

Some Previous experimental work performed at Strasbourg:

We present here the work performed on 2 references organic scintillators :

- A scintillator based on polyvinyltoluene (PVT) developed by Bicron; trade name: BC-418
- Polycrystalline anthracene, produced by the “area fusion” technique.

Figures 2 to 3 present the results obtained for these two scintillators under proton and alpha particles of 2 MeV energy using our experimental set-up (Figure1)

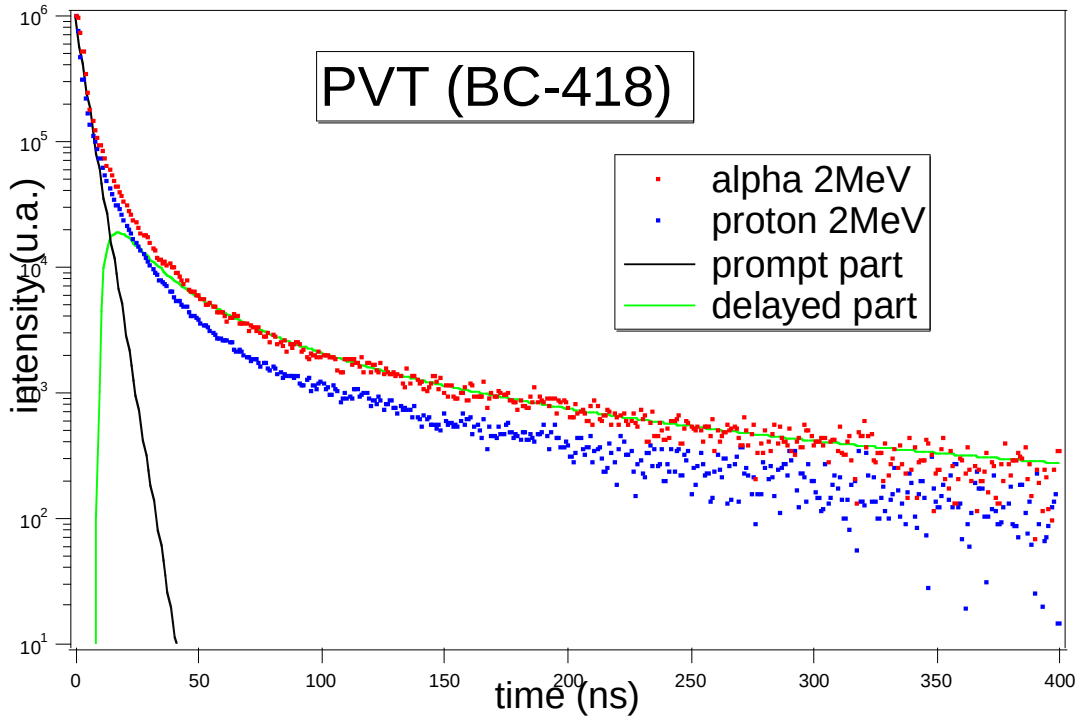

Figure2: Comparison of the fluorescence decay under alpha and proton excitation for a PVT sample. In green and black appear the deconvolution of the alpha decay in prompt part and delayed part.

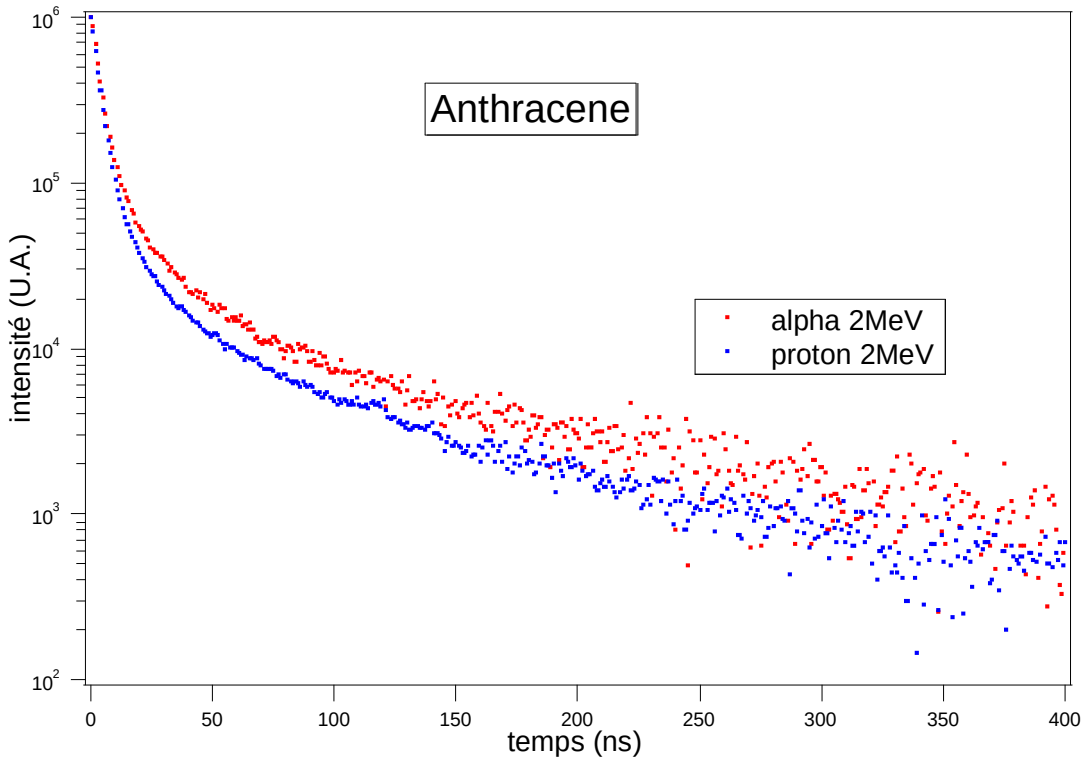

Figure 3 : Comparison of the fluorescence decay under alpha and proton excitation for an Anthracene cristal obtained via a multiarea fusion technique.

Ion beam request at Tandem accelerator in Bucharest:

We have collected data for protons and alpha up to 4 MeV. It is critical for us to irradiate these scintillators with heavier particles in order to study the effect of LET on the prompt and delayed parts of the radio-induced fluorescence. One of the aim of this study is to build a unique set of data. Ion beams must be pulsed with a frequency of the order of some tens of kHz. The width of pulses has to be of the nanosecond order.

Fisrt test (june 2011)

In june a first experiment was conducted with carbon and sulfur ions. The scheme of the experiment is identical as the one presented Fig. 1. For the time correlated single photon counting method the start signal is now given by the pulsing system of the 9 MV tandem. Figure 4 presents a picture of the system.

Fig. 4 : Irradiation cell for the time correlated single photon counting method. One can observe the photomultipler (black cylinder) at the top of the irradiation cell.

Fig. 5 and 6 present the first results obtained for carbon ions (7 MV- 1to 2 nA) and sulfur ions (6.2 MV- 1to 2 nA) for two scintillators ; anthracene and BC418. Anthracene is present as monocristal of 5 to 200 µm thick. BC418 is manufactured by Bicron. This is a ternary scintillator. Polyvinyltoluene (PVT) constitutes the base of the detector where two fluorescent dyes are dissolved. BC148 is present as a 10 µm plastic film thick. Small thicknesses are necessary to study the effect of LET on the fluorescence yield.

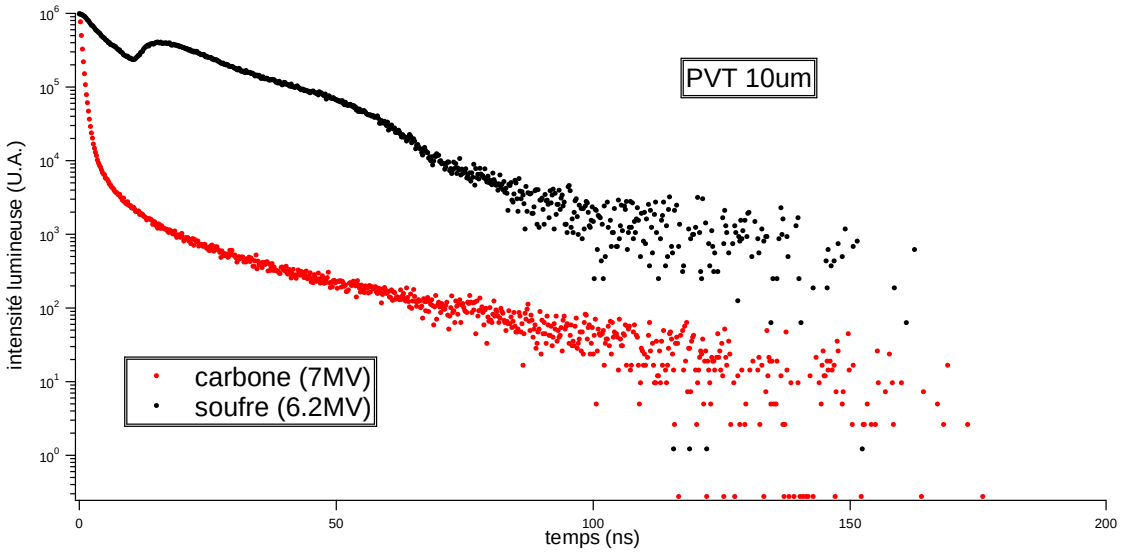

Fig. 5 : Time resolved fluorescence (intensity is given in arbitrary units) induced in a BC418 (PVT) of 10 µm by carbon ions (red curve) and sulfur ions (black curve).

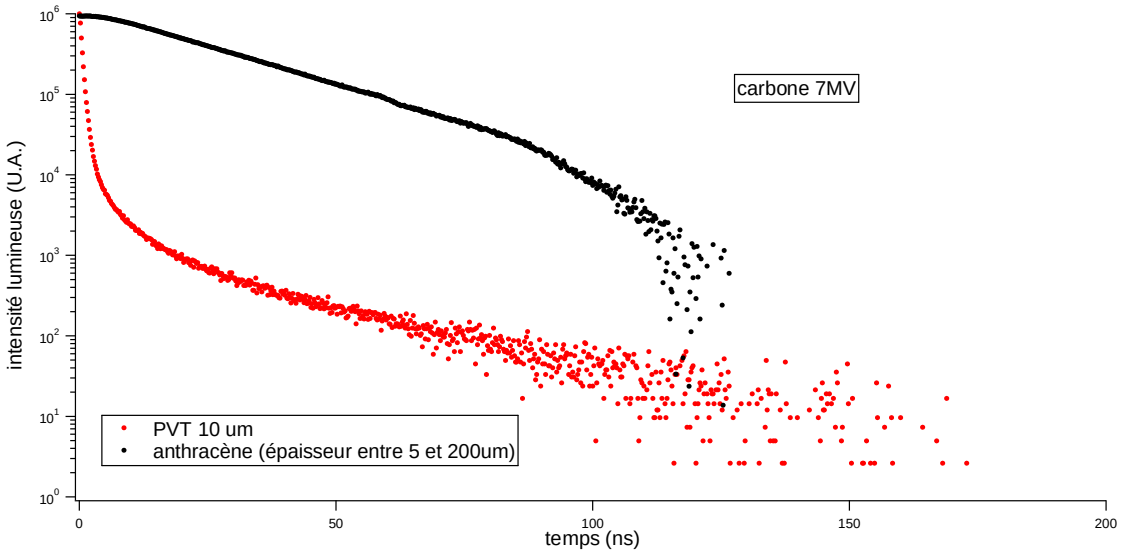

Fig. 6 : Time resolved fluorescence (intensity is given in arbitrary units) induced by carbon ions (7 MV) in BC418 (red curve) and in anthracene (black curve).

It is important to point out that Fig. 5 and Fig. 6 present only first qualitative results. These figures clearly show that, as expected, the delayed fluorescence increases with the ion's LET and that anthacene exibits a larger delayed fluorescence than BC418. The time resolution seems nice for carbon ions (3-4 ns). This is not the case for sulfur ions, for wich obviously the excitation pulse is composed of different peaks. This lack of resolution could be solved by improving our irradiation cell to enable a better focussing of the pulsed ion beam on the scintillator. We propose to equip the cell with a diaphragm at its entrance, and to set up a faraday cup just behind the sample's holder. Using a thin rapid scintillator, enabling ions to going through, we will be able to observe simultaneously the shape of the pulsing (with the fluorescence of the scintillator) and the current of the pulsed ion beam received by the scintillator.

Futur experiments

These very first tests prove that the facilites provided by the tandem accelerator at the Nuclear physics department in Bucharest are definitively adapted to the objectives of our studies.

In Future, our demand concerns ion pulsed beams from proton to manganese to perform experiment on a large scale of LET. In each case we ask for a fluence of about 1 nA . cm^{-2}.

For further information you can contact us at:
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