Evidence of delayed light emission of TetraPhenyl Butadiene excited by liquid Argon scintillation light

E. Segreto*

INFN - Laboratori Nazionali del Gran Sasso, Assergi, Italy

Abstract

TetraPhenyl Butadiene is the wavelength shifter most widely used in combination with liquid Argon. The latter emits scintillation photons with a wavelength of 127 nm that need to be downshifted to be detected by photomultipliers with glass or quartz windows. TetraPhenyl Butadiene has been demonstrated to have an extremely high conversion efficiency, possibly higher than 100% for 127 nm photons, while there is no precise information about the time dependence of its emission. It is usually assumed to be exponentially decaying with a characteristic time of the order of one ns, as an extrapolation from measurements with exciting radiation in the near UV. This work shows that TetraPhenyl Butadiene, when excited by 127 nm photons, reemits photons not only with a very short decay time, but also with slower ones due to triplet states de-excitations. This fact can strongly contribute to clarify the anomalies of liquid Argon scintillation light reported in literature since seventies, namely the inconsistency in the measured values of the long decay time constant and the appearance of an intermediate component.

1 Introduction

Liquid Argon (LAr) is a widely used active medium in particle detectors, especially in the fields of neutrino physics and Dark Matter direct search [1, 2, 3, 4]. It is often used in scintillation detectors thanks to its high photon yield (~ 40000 photons/MeV at null electric field for m.i.p.) and to the possibility of discriminating different ionizing particles through pulse shape discrimination techniques (see for instance [5]). The wavelength of the emitted radiation is around 127 nm, so in the vacuum UV (VUV). The most efficient and viable way of detecting LAr scintillation light is to downshift it to longer wavelengths, where common quartz or glass windowed photodevices are sensitive. The most popular wavelength shifter used in combination with LAr is TetraPhenyl Butadiene (TPB) [6, 7, 8], that has been shown to have an extremely high efficiency in converting VUV photons into visible ones (possibly higher than 100% [9]).

*E-mail: ettore.segreto@lngs.infn.it
On the other side there isn’t a precise knowledge of the TPB emission time spectrum when excited by 127 nm photons. It is usually described by a single decaying exponential with characteristic time in the range of 1 ns, as an extrapolation from measurements performed with exciting radiation in the range of the near UV (around 350 nm) [10] [11]. This is perfectly compatible with the photoexcitation of singlet states ($S_n$) of the II electrons of the TPB molecules. They decay via internal conversion to the first excited singlet state $S_1$ in less than one ns. The scintillation photon is produced by the radiative deexcitation of this state to the fundamental state ($S_1 \rightarrow S_0$) that typically has a characteristic time of the order of one ns [12] [13] [14].

The point never considered up to now is that VUV scintillation photons’ energy (9.7 eV) could very likely exceed the ionization potential of TPB. Actually there are no available data in the literature, but a calculation leads to a value of 5.4 eV [15]. This could appear an extremely low energy, but it is worth noticing that similar compounds like p-terphenyl and anthracene, both used as scintillators or wavelength-shifters, have ionization energies between 7 and 8 eV, not so far from that estimated for TPB. Furthermore it is not difficult to find examples of conjugated molecules with response similar to that of TPB in the UV-vis region with ionization energies in the range of 5-6 eV, as PTCDA, Alq3 or CuPc [16].

TPB molecules are very likely ionized by LAr scintillation photons and the emitted electron would have enough energy to excite singlet or triplet states of some of the surrounding molecules. Also the recombination of the electron-ion pair can lead to the population of triplet states. Excited singlet states produce the so called prompt fraction of scintillation within few ns through the de-excitation of the $S_1$ states to the ground level. Excited triplet states decay very fast to the lowest lying triplet state $T_1$ via internal conversion. These long lived states (the transition $T_1 \rightarrow S_0$ is forbidden by selection rules) are the precursors of the delayed fraction of scintillation in pure aromatic media through the triplet-triplet interaction process: $T_1 + T_1 \rightarrow S_1 + S_0$, where the scintillation photon is produced by the deexcitation of the $S_1$ state [12].

The ionization of TPB would also easily explain the observation of conversion efficiencies of LAr VUV photons higher that 100%, since each absorbed photon could excite more than one TPB molecule at a time. In this paper the experimental evidence of the existence of a delayed component of the scintillation light of TPB excited by LAr VUV scintillation photons is presented. A similar effect has already been reported, for example, for sodium salycilate and for p-terphenyl ([17] [18] and references therein). The proper consideration of this experimental fact can have a significant role in clarifying the inconsistency in the measured values of the long decay time constant and the appearence of an intermediate component of the LAr scintillation light found in literature [19] [23] [26] [27]. It also shows that the use of TPB and in general of wavelength-shifters may worsen the pulse shape capability of LAr.
since part of the fast photons are delayed.

2 Experimental evidence

The scintillation light of LAr proceeds through the deexcitation of the excited dimer Ar\(_2^*\) and shows two decay components: one very fast (~ 6 ns) originating from the decay of the lowest-lying singlet state - 1\(\Sigma^-\) and one very slow (~ 1.3 \(\mu\)s) from the decay of the lowest-lying triplet state - 3\(\Sigma^-\) [28, 29]. It has been clearly shown [19, 30] that N\(_2\) contaminations in LAr produce a quenching of the scintillation light, while no other emission phenomenon from N\(_2\) has been observed even at extremely high levels of contaminations (~ 10%). The quenching process is a collisional one and the net effect is that the decay times of LAr scintillation components are shortened according to:

\[
\frac{1}{\tau'_{f,s}([N_2])} = \frac{1}{\tau_{f,s}} + k_q \times [N_2]
\]

and consequently the relative abundances of the fast and slow components become:

\[
A'_{f,s}([N_2]) = \frac{A_{f,s}}{1 + \tau_{f,s} \times k_q \times [N_2]}
\]

where \(\tau_{f,s}\) and \(A_{f,s}\) are the decay times and amplitudes of the fast/slow component for uncontaminated LAr, \([N_2]\) is the nitrogen contamination in ppm and \(k_q\) is the reaction rate that has been measured to be \(k_q = 0.11 \pm 0.01 \mu\text{sec}^{-1}\text{ppm}^{-1}\) (19).

Taking into account that for \(\gamma/e^-\) excitations of uncontaminated LAr, \(A_f = 0.3, A_s = 0.7\), it can be easily found that for \([N_2] = 3000\) ppm one obtains \(\tau'_f \simeq 2\) ns, \(\tau'_s \simeq 3\) ns, \(A'_f = 0.1\) and \(A'_s = 1.4 \times 10^{-3}\). In practice the slow component is completely negligible.

The scintillation light in heavily N\(_2\) doped LAr results to be a very fast pulse that is ideal to study the TPB response to 127 nm photons.

In [19] the results of a test of the effects of nitrogen contaminations in LAr are very clearly presented. The detector was constituted by a PTFE cell containing about 0.7 l of LAr lined up with a highly reflective foil (VM2000 by 3M) covered by a thin film of TPB (surface density \(\sigma \simeq 450 \mu\text{gram/cm}^2\)) and observed by a single 2” photomultiplier. An injection system allowed to contaminate the ultra pure LAr with controlled amounts of N\(_2\). The details of the experimental set-up can be found in [19]. Contamination levels ranging from 1 ppm to 3000 ppm of N\(_2\) were explored. For each different contamination the LAr cell was exposed to a \(\gamma\) source of \(^{60}\text{Co}\). Scintillation light produced by electrons from \(\gamma\) interactions was wave-shifted on the surface of the cylinder and then detected by the photomultiplier.

\(^1\)It is assumed here that the probability density function for scintillation photons is \(A_f/\tau_f \exp(-t/\tau_f) + A_s/\tau_s \exp(-t/\tau_s)\) and \(A_f + A_s = 1\)
The average of the waveforms collected at 3000 ppm of N\textsubscript{2} contamination is shown in figure 1, left\textsuperscript{2}. According to the previous discussion this waveform should be regarded as the time response of pure TPB to LAr scintillation photons.

This waveform clearly shows the expected very fast/instantaneous pulse, but also much slower components with decay times of the order of the µs, that are interpreted here as coming from the triplet-triplet interaction process in TPB. It has been clearly demonstrated that the delayed scintillation is not exponential \cite{12, 20, 21, 22}, but to have a simplified quantitative idea of the time evolution of the emitted light the waveform has been fitted with a function made of four decaying exponentials, convoluted with a gaussian function that accounts for the photomultiplier response and for the electronic noise. A more adequate treatment of the delayed scintillation will be presented in section 3. The result of the fit is shown in figure 1, left with a red line and the abundance and decay time of the components are reported in table 1.

The response function of TPB can thus be roughly thought as composed by the following exponential components:

- one very fast, compatible with an instantaneous emission of photons following the excitation. Its decay time can not be well determined with this experimental set up because of the gaussian smearing of the signal due to the photomultiplier response, the electronic noise and the propagation of the photons inside the chamber before

\footnote{This waveform is not shown in \cite{19}, but it has been kindly granted by the authors.}
detection. This component is the most abundant and accounts for about 60% of the shifted light;

- one with a decay time of the order of 50 ns accounting for about the 30% of shifted light. This component is consistent with what is observed by many experimental groups that used LAr in combination with TPB [19, 23, 24];

- a very slow one with a decay time of the order of 3 $\mu$s that contains about 8% of the shifted light. It can significantly alter the slow component of LAr scintillation light and can make extremely difficult the precise estimation of its time constant if not properly considered;

- a spurious component with a decay time around 300 ns and with only 1-2% of the light.

Table 1: *Decay times and relative abundances of the exponentially decaying components found in the response function of TPB to 127 nm photons. Only statistical errors from the fit are quoted.*

| Component                | decay time (ns) | abundance (%) |
|--------------------------|-----------------|---------------|
| Instantaneous component  | 1-10            | 60 ± 1        |
| Intermediate component   | 49 ± 1          | 30 ± 1        |
| Long component           | 3550 ± 500      | 8 ± 1         |
| Spurious component       | 309 ± 10        | 2 ± 1         |

In figure 1, right the relative integral of TPB response function versus time is shown. It is easy to see that it is necessary to integrate the waveform for at least 140 ns to accumulate the 90% of the signal, against the 15 ns that would be necessary in case of a simple exponentially decaying slope with a characteristic time of 6 ns.

Since during the N$_2$ contamination test described in [19], data were taken for many different values of N$_2$ concentration in LAr, a very stringent test of the hypothesis that the waveform shown in figure 1 left represents the response of TPB to LAr scintillation photons has been possible. For each level of [N$_2$] the average waveform shown in [19] has been fitted with a convolution of a double exponential, assumed to be the probability density function of LAr scintillation photons, with the response function of TPB. Some examples of fitted waveforms are shown in figure 2. In all the cases the fits are almost perfect and the average waveforms are reproduced very precisely along all the time interval considered (up to 9.7 $\mu$s after the onset of the signal).

The picture that emerges from this analysis is perfectly consistent and there is no need of invoking exotic mechanisms of LAr scintillation, different from the two excimer states deexcitation ($^1\Sigma$ and $^3\Sigma$), to explain all the features observed in the LAr scintillation waveforms. This is true in particular for one of the points deeply analyzed in [19].
without reaching definite conclusions, that is the existence of an intermediate decaying component between the singlet and the triplet deexcitation.

3 Test with $\alpha$ and $\beta$ interactions

Following the idea that the waveform of figure 1, left can represent the response of TPB to LAr scintillation photons, a dedicated experimental test has been performed. In order to check if the observed long tail is effectively related to TPB deexcitation, a sample of pure TPB has been directly irradiated with an ionizing radiation in a vacuum environment. This guarantees the formation of triplet states (through electron-ion recombination, secondary electrons excitations, $\delta$-rays...), that are in turn the precursors
of the delayed TPB scintillation, with a perfectly $\delta$ shaped excitation function and in a way completely independent from LAr scintillation light.

In general the scintillation of pure organic crystals excited by an ionizing radiation can be well described with the superposition of a prompt component and of a delayed one. The prompt component is found to be exponentially decaying with a time constant identical to the mean lifetime $\tau_S$ of the first excited singlet state $S_1$.

The time evolution of the delayed component depends on the dynamics of the triplet-triplet interaction process. It can be predicted by solving the diffusion-kinetic equation for the triplet density along the ionizing track, assuming a Gaussian shape, with scale parameter $r_0$, for the initial triplet distribution function $[12, 21, 22]$. The asymptotic time dependence of the delayed light ($t \gg \tau_S$) is found to be:

$$I(t)_{\text{delayed}} \simeq \eta_S \frac{N}{[1 + A \ln(1 + t/t_a)]^2(1 + t/t_a)}$$

where $N$ and $A$ are constants depending on the nature of the scintillator, $\eta_S$ is the fluorescence yield and $t_a$ is a relaxation time that is linked to the diffusion coefficient of triplet states in the scintillator, $D_t$, through the relation: $t_a = r_0^2/4D_t$.

The time evolution of the delayed light is independent on the particle type, but only on
the dynamics of the triplet-triplet interaction process. Only the relative abundance of fast and delayed components is expected to depend on the linear energy transfer, and consequently on the particle type.

The experimental set-up that has been built is essentially constituted by a stainless steel vacuum tight chamber hosting a 2” photomultiplier (ETL D745UA), an holder for the TPB sample and one for the radioactive source. A schematic view of the set-up is shown in figure 3. The TPB sample is a film with a surface thickness \( \sim 10^3 \mu \text{gram/cm}^2 \) evaporated on a highly reflective plastic foil (3M VM2000) circular in shape with a diameter of 8 cm. The choice of having a reflective substrate below the TPB has been done to maximize the amount of light that can be collected by the photomultiplier. A drawback of using VM2000 is that it is weakly emitting light when irradiated by ionizing particles. The film has been produced at LNGS with a dedicated evaporation system. More details can be found in [25]. The distance between the sample and the photomultiplier is 5 cm. Two sources have been alternatively used:

- an \( \alpha \) source made of an alloy of Uranium and Aluminum emitting \( \alpha \) particles with a continuous spectrum with end point around 5 MeV;
- a \( ^{90}\text{Sr} \) \( \beta^- \) source with a Q value of 546 keV.

During each measurement the stainless steel chamber has been evacuated down to a pressure of \( 5 \times 10^{-5} \) mbar to allow \( \alpha \) particles and electrons to hit the TPB film without being captured by air. Scintillation signals detected by the photomultiplier have been sent to a fast Waveform Recorder (Acqiris, DP235 Dual-Channel PCI Digitizer Card). The signal waveforms passing a threshold, set at a level corresponding to few photoelectrons, have been recorded with sampling time of 1 ns over a full record length of 10 \( \mu \text{s} \).

All the measurements have been performed at room temperature, since it has been shown in [31] that the time dependence of the late components of TPB fluorescence excited by \( \alpha \) particles does not change appreciably at LAr temperature.

3.1 Data analysis and comparison

The average waveforms for the \( \beta \) and \( \alpha \) particles tests have been calculated applying simple cuts to eliminate waveforms that presented saturations, afterpulses or multiple signals and the result is shown in figure 4. The red curve refers to \( \beta \) irradiation and the blue one to \( \alpha \), while the black curve is the one obtained with LAr scintillation light, already shown in figure 1 reported here for visual comparison. Beta and VUV photons curves are nicely overlapped as is observed also for sodium salicylate [17]. The small differences found around 300-400 ns after the onset of the signal could be ascribed to a small effect of fluorescence of the plastic substrate of the TPB film, since the electrons have enough energy to reach and traverse it. Alpha particles, instead, show a much higher abundance of delayed component.
Figure 4: Black line: average waveform obtained with the 0.7 l LAr scintillation chamber with 3000 ppm of N₂ when irradiated with ⁶⁰Co γs. This waveform should be regarded as the time response of TPB (see text). Red line: average waveform obtained by irradiating a TPB film with electrons. Blu line: average waveform obtained by irradiating a TPB film with α particles.

In order to investigate the details of the tails of the three curves and to check if they are compatible with each other a single photon counting like procedure has been adopted. The classical Coincidence Single Photo-electron Counting technique has been used many times in scintillation lifetime measurements [30, 32, 33]. The recorded waveforms allow to implement an offline version of this technique. Starting at 170 ns after the onset of a triggered signal, a single photo-electron finding algorithm is run through the waveform and for each photo-electron pulse (defined by appropriate cuts) the arrival time is stored. In order to minimize the pile-up of single photo-electrons, that is a time dependent effect, only waveforms with a total integral below 40 photo-electrons in the case of electron and photon excitations and 20 in the case of αs have been considered. In this way the pile up probability in the first 30 ns results to be below 5% in all three cases.

The time of arrival of photo-electrons has been accumulated in three histograms, shown in figure 5. They are almost perfectly overlapped. The one related to VUV pho-

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3The pile-up probability has been estimated assuming as probability density functions the waveforms shown in figure 4.
Figure 5: Histograms of the arrival time of single photons for photon (black curve), $\alpha$ (red curve) and electron (green curve) excitations of TPB. Histograms are normalized to unit area. The blue line represents a fit of the histogram related to VUV photons with the function of equation 5.

tons has been fitted with the function of equation 3 where $A$, $t_a$ and the product $\eta S N$ are left as free parameters. The result of the fit is shown in figure 4 with a blue line. The best fit values for $A$ and $t_a$ are 0.22 and 51 ns respectively, not so different from what is found for anthracene [34] - $A=0.25$ and $t_a=40$ ns - and for stilbene [35] - $A=0.25$ and $t_a=80$ ns.

The perfect compatibility of the delayed scintillation of TPB excited by LAr scintillation photons with that induced by ionizing particles clearly demonstrates that it is generated by a triplet-triplet interaction mechanism, started by the ionization of TPB by 127 nm photons. The TPB response function to LAr scintillation light is not a fast decaying exponential, as it is found for near UV excitation, but has a much more complex structure with a delayed component that has a non-exponential shape and that accounts for about the 40% of the emitted radiation.
4 Conclusions

This work shows the experimental evidence of the existence of a delayed scintillation component of TPB when excited by the VUV radiation of LAr. The production of the triplet states, that are the precursors of the delayed light, is made possible by the high energy of LAr scintillation photons that can ionize the organic molecules of TPB. Its time dependence has been measured with an experimental set-up that uses LAr scintillation light quenched by nitrogen contaminations to excite TPB. It has been compared to the time behavior of the delayed light of TPB when excited by $\beta$ and $\alpha$ particles and they have been found to be perfectly compatible among each other. The time shape of the light emission has also been found to be consistent with what expected from the delayed luminescence of a unitary organic scintillator as described in literature.

This experimental fact sheds some light on the most relevant incongruities that have been reported in the past years concerning the time dependence of LAr scintillation light. Namely the presence of an intermediate component with a decay time in the range of 50-100 ns and the ambiguity in the determination of the decay time of the slow component, for which values ranging from 800 ns to 1600 ns have been reported. The use of TPB, of another shifter, or of no shifter can influence the time shape of the detected scintillation signal in different ways. In this respect the most reliable value for the slow decay time appears to be the one of $1300 \pm 60$ ns reported in [26] measured without shifter and in [19] with sophisticated deconvolution techniques.

LAr scintillation is often used for particle discrimination since the relative abundance of the fast and slow components strongly depends on the particle type. The use of TPB tends to worsen this feature of LAr since a consistent part of the prompt light is delayed and the two populations are more mixed. TPB is an exceptionally efficient shifter for the VUV scintillation light of LAr and also a convenient one for its emission wavelength around 430 nm matching the quantum efficiency of many standard photomultipliers, but it has some drawbacks when the time features of the scintillation signals are used since they result to be slightly distorted.

acknowledgments

The author acknowledges Prof. F. Cavanna for his contribution to this work with discussions and ideas; Prof. R. Francini for the discussions on molecular processes; Dr. A. A. Machado for her help and patience and Dr. N. Canci for his contribution to the measurements with $\alpha$ and $\beta$ particles. This paper is dedicated to the memory of Antonio Di Filippo.

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