Slow scintillation time constants in NaI(Tl) for different interacting particles

C. Cuesta,1, M.A. Oliván,1 J. Amaré,1 S. Cebrián,1 E. García,1 C. Ginestra,1 M. Martínez,1, a) Y. Ortigoza,1 A. Ortiz de Solórzano,1 C. Pobes,1 J. Puimédon,1 M.L. Sarsa,1, b) J.A. Villar,1 and P. Villar1

Laboratorio de Física Nuclear y Astrofísicas, Universidad de Zaragoza, Pedro Cerbana 12, 50009 Zaragoza, SPAIN
Laboratorio Subterráneo de Canfranc, Paseo de los Ayerbe s.n., 22880 Canfranc Estación, Huesca, SPAIN

Very large thallium doped sodium iodide crystals operated underground and in very low background environment in the context of a dark matter search experiment have been used to determine scintillation components in the tens of ms range in the light pulse induced by different interacting particles: $\gamma/\mu$ and $\alpha$.

PACS numbers: 29.40.Mc, 21.10.Tg, 23.60.+e, 95.85.Ry
Keywords: sodium iodide, scintillation, low radioactive background, underground laboratory

I. INTRODUCTION

Thallium doped sodium iodide scintillators have been widely used for radiation detection1–12 since they were proposed by R. Hofstadter in 1948.3 Because of their very high light yield, NaI(Tl) detectors became very soon one of the most convenient options for many applications, from radiology to environmental monitoring, as examples. NaI(Tl) detectors have other advantages, very large crystals can be grown and raw material is not expensive; also high efficiency for gamma ray detection is easily achieved. Nevertheless, NaI hygroscopic character makes difficult the manipulation of the crystals and its application in the very low energy x-ray regime. Main scintillation time constants of NaI(Tl) are well reported in the bibliography (see1 and references therein): the dominant decay time of the scintillation pulses is in the range 230–250 ns4–6, but slower components of 1.5 $\mu$s4 and 0.15 s7 have been also reported. Other possible phosphorescence components could be present at much longer timescales8, but data are scarce. Differences in the scintillation time constants for different particles are also well known and have been used for discrimination purposes: alpha vs. gamma interactions at high energy9–12 and nuclear recoils vs electron recoils at very low energies13–17. In the latter case, differences are quantified with an effective mean decay time, and are so small that only a statistical discrimination of events can be pursued. Dependence of the time constants with the energy of the particle has been also clearly established14,17.

NaI(Tl) detectors have been used since the 90’s in the search for the hypothetical Dark Matter (DM) particles filling our galactic halo and explaining an important part of the missing Universe mass16,18–23. Among these experiments, DAMA/LIBRA results have produced a large impact in the field by observing an annual modulation in the rates compatible with that expected for DM23. ANAIS (Annual modulation with NaI Scintillators) is an experiment to be carried out at the Laboratorio Subterráneo de Canfranc (LSC) in Spain, trying to confirm the presence of such a modulated signal using the same target and technique24,25.

As a byproduct of the ANAIS prototype operation phase, scintillation decay time constants in several large NaI(Tl) detectors have been studied in different temporal scales and for different interacting particles, profiting from the very low event rate and stability of the experiment due to the underground operation at the LSC and the ultralow radioactive background environment. Up to our knowledge, it is the first time such a thorough study of the scintillation time constants in NaI(Tl) in the millisecond range has been reported.

II. EXPERIMENTAL SET-UP

Four large NaI(Tl) crystals (see Tab. I) have been used to derive the results presented in this letter:

- a 10.7 kg hexagonal prism (distance between opposite vertices in the hexagonal face 15.94 cm, and 20.32 cm length) made by BICRON and stored underground at LSC since the late eighties. The original detector was opened at the University of Zaragoza (UZ) and re-encapsulated using OFHC copper;
- a 9.6 kg parallelepiped prism (10.16x10.16x25.40 cm3) grown by Saint Gobain Ltd. and encapsulated in ETP copper at the UZ after staying at a surface laboratory for several years, kept in dry atmosphere. Data used in this work were taken after almost three years of operation underground;
- two 12.5 kg cylindrical crystals (12.07 cm φ, 29.85 cm length) grown by Alpha Spectra Inc. from the same ingot, using low potassium content selected NaI powder, and encapsulated in OFHC copper. Data taking started immediately after taking them underground.

Teflon tape (about 2 mm thick) plus a reflecting multilayer foil (3M VikuitiF™), both wrapping the NaI crystals, are used for light diffusion and reflection in the two former detectors and only Teflon in the latter two. Quartz optical windows (3” diameter) in both sides of the encapsulations allow the coupling of two photomultiplier tubes (PMTs) per crystal. In the data used along...
Muons can only be tagged by the plastic scintillators veto signals for the MATACQ data (shortest timescale). For the oscilloscopes data, muon and gamma events are considered the same population; however, we can expect to find more muons than gammas above the $^{208}$Tl line at 2614.6 keV. Muons are found at a rate of about 45 events/day whereas for alpha events the corresponding rate is between 800 and 3000 events/day, depending on the crystal bulk contamination in uranium and thorium chains. $\alpha$ events are separated from $\gamma/\mu$ events by a very simple cut based on the different relationship between amplitude and area of the fast component of the pulse ($1\mu$s window), hereafter called fast pulse. Two populations can be clearly distinguished, estimating as 100% the efficiency of the discrimination above 2 MeVee and up to the saturation of the PMT signal. No assumptions about the timing constants are required to apply such a discrimination. Neither a difference between $\gamma$ and $\mu$ events has been found, as expected, nor a significant difference between fast pulses coming from the four studied crystals. Because of that, in sec. III B and III C only data from ANAIS-0 will be shown.

TABLE II. Parameters of the digitizers used in this work.

| Digitizer | sampling rate (MHz) | window (µs) | resolution (bits) |
|-----------|---------------------|-------------|------------------|
| MATACQ    | 2 GS/s              | 1.25        | 12               |
| TDS5034B-207 | 25 MS/s          | 320         | 8                |
| TDS5034B-208 | 250 MS/s         | 40          | 8                |

III. RESULTS

A. Events selection

Data acquisition in the very long timeline basis required the design of an special branch in the electronic chain. It consists of two Tektronix phosphor oscilloscopes (TDS5034B) in two different time scales and also different vertical ranges, having 8 bits of vertical resolution. Main characteristics of the oscilloscopes used in this work are: 350 MHz bandwidth, up to 5 GS/s sampling rate capability, 4 channels and between 500 and $8 \times 10^8$ sampled points. Trigger was done independently for the Tektronix oscilloscopes and the MATACQ board. The former were only triggered by very high energetic events (above 2 MeVee) and measurements were done for one detector each time. The most relevant configuration parameters of the two used oscilloscopes and the MATACQ board are summarized in Tab. II. TDS5034B-207 dynamic range allows to identify individual photoelectrons in the pulse tail, whereas TDS5034B-208 measures the whole of the pulse, whose area is taken as energy estimator.

B. Pulses in the microsecond range

Muon events can only be tagged by the plastic scintillators veto signals for the MATACQ data (shortest timescale). For the oscilloscopes data, muon and gamma events are considered the same population; however, we can expect to find more muons than gammas above the $^{208}$Tl line at 2614.6 keV. Muons are found at a rate of about 45 events/day whereas for alpha events the corresponding rate is between 800 and 3000 events/day, depending on the crystal bulk contamination in uranium and thorium chains. $\alpha$ events are separated from $\gamma/\mu$ events by a very simple cut based on the different relationship between amplitude and area of the fast component of the pulse ($1\mu$s window), hereafter called fast pulse. Two populations can be clearly distinguished, estimating as 100% the efficiency of the discrimination above 2 MeVee and up to the saturation of the PMT signal. No assumptions about the timing constants are required to apply such a discrimination. Neither a difference between $\gamma$ and $\mu$ events has been found, as expected, nor a significant difference between fast pulses coming from the four studied crystals. Because of that, in sec. III B and III C only data from ANAIS-0 will be shown.
to interactions of $\alpha$, $\mu$ and $\gamma$ in the crystal, and normalized to the same pulse area. The corresponding pulses can be seen in Fig. 1. No difference between gamma and muon events is noticed, and in the following we will consider both as a single population, as far as a different behavior in the longer timescales is not expected.

Pulses have been fitted to a combination of two exponential decays ($A_i e^{-t/\tau_i}$), independently for $\alpha$ and $\gamma/\mu$ events: one corresponds to the rise of the pulse, and the second, to the decay. However, the presence of slower components could affect the results of the fit in this region (see sec. III C). Results are given in Tab. III. In the case of $\alpha$ pulses, rise time is compatible with contributions coming from light propagation in the crystal and width of Single Electron Response (SER) of the PMT (gaussian shaped with FWHM of 12 ns for the R6956 PMT), hinting at a prompt light emission. Decay time for $\alpha$ pulses is compatible with that reported in the bibliography, although in the following we will consider both as a single population, as far as a different behavior in the longer timescales is not expected.

TABLE III. Results of the fits from 0 to 1000 ns after the pulse onset for alpha and gamma/muon events.

| Fit parameter         | $\alpha$ events | $\gamma/\mu$ events |
|-----------------------|-----------------|---------------------|
| $\tau_{rise}$ (ns)    | 17.98 ± 0.12    | 28.23 ± 0.14        |
| $\tau_{decay}$ (ns)   | 219.28 ± 0.52   | 287.35 ± 0.51       |
| $A_{rise}/A_{decay}$  | 1.223 ± 0.004   | 1.123 ± 0.003       |

For the possible presence of additional scintillation time constants in the few microseconds range. Clearly pulses undershoot the baseline, being not recovered in the 40 $\mu$s studied range, but in the first milliseconds. However, it can be noticed a much more important undershooting for $\alpha$ events than for those attributable to $\gamma/\mu$, pointing at a possible additional slow scintillation component in the latter that could partially compensate the undershooting. Similar effects have been observed for all the detectors studied and a modification of the PMT readout circuit would be required to further clarify this issue.

FIG. 1. Average pulses from $\alpha$ events and those produced by $\gamma$ and $\mu$, normalized to the same pulse area.

D. Pulses in the 320 milliseconds range

For the longest timescale, a different approach has been followed: photoelectrons (p.e.) have been identified individually, at a given position of the pulse, and an histogram has been produced with the corresponding temporal distribution for all the events in the ROI, separately for $\alpha$ and $\gamma/\mu$ events for each of the four NaI(Tl) detectors studied. The corresponding distributions can be seen in Fig. 3, conveniently normalized (although independently for each detector) to the same fast pulse area and averaged according to the number of events in the ROI for each population. Similar ROI have been studied for ANAIS-0 (see sec. III B), D0 and D1 detectors, but PIII ROI corresponds to higher energies (from 5 to 5.3 MeVee). Selection of individual p.e. is done very efficiently in the MATAQ data by applying a peak search algorithm, however, in the data from TDS5034B-207, we expect many p.e. to be lost because of the sampling rate algorithm, however, in the data from TDS5034B-207, we expect many p.e. to be lost because of the sampling rate used, and also we expect differences between detectors because of the different SER of each PMT model used; therefore, only relative values concerning the p.e. number in the slow scintillation components for each detector are considered and comparison with the fast component, or among detectors, is meaningless.

In Tab. IV the ratio between the average p.e. number for $\alpha$ and $\gamma/\mu$ events is shown for the four detectors studied, as well as the mean time, calculated only with the tail: from 4 till 304 ns after the pulse onset. The most relevant difference is the total number of p.e. identified in these slow components: $\gamma$ and $\mu$ events excite much more efficiently the long-lived states contributing

FIG. 2. Pulses from $\alpha$ and $\gamma/\mu$ events in a zoomed view of the pulse baseline to remark the pulse undershoot.
to them, whereas similar mean time constants are found for both types of interacting particles, being in all cases α events slightly faster. Because of the quenching factor for α vs β/γ events in NaI(Tl) and the fact that we have normalized to the fast pulse area and not to the energy deposited, the referred effect is still much more important in terms of photodetector performance per unit of energy. In spite of some similarities, significant differences among the studied crystals can be reported; hence, impurities or defects could play a role in this slow scintillation mechanism.

Fits have been carried out for α and γ/µ events for the four detectors studied, using the range from 4 to 304 ms after the onset of the pulse, trying to distinguish different scintillation components. Two exponential decays have been considered in the fit, except in the case of PIII data (affected by a poor statistics and higher energy of the selected events). Corresponding results are summarized in Tab. V. No clear conclusion can be drawn from the fits.

IV. CONCLUSIONS

Scintillation time constants in NaI(Tl) crystals from ns to 300 ms have been studied for α as well as for γ/µ events in the high energy regime. Very different behavior for both kind of interactions is confirmed, specially in the capability of exciting the slow scintillation mechanism (70-100 ms mean time constants). Significant differences have been observed among results derived from different NaI(Tl) crystals, pointing at an origin of such scintillation related to impurities or defects more than to the thallium doping. However, all the crystals studied showed such effects and further work is in progress to determine how this slow scintillation could affect the application of these detectors in DM searches.

ACKNOWLEDGMENTS

This work has been financially supported by the Spanish and European Regional Development Fund MINECO-FEDER under grant FPA2011-23749, Consolider-Ingenio 2010 Programme under grants MultiDark CSD2009-00064 and CPAN CSD2007-00042 and the Gobierno de Aragón.

1J.B. Birks, _The theory and practice of scintillation counting_. Pergamon Press Ltd., 1964.
2P. Lecoq, A. Annenkov, A. Gektin, M. Kozhirik, C. Pedrini, _Inorganic scintillators for detector systems_ (Springer-Verlag, 2006)
3R. Hofstadter, _Phys. Rev._ 75 (1949) 796.
4J.C. Robertson and J.G. Lynch, _Proc. Phys. Soc._ 77 (1961) 751.
5P.S. Eby and W.K. Jentschke, _Phys. Rev._ 96 (1954) 911.
6J.S. Schweitzer and W. Ziehl, _IEEE Trans. Nucl. Sci._ 30 (1983) 380.
7S. Kočička, A. Kočičk, and V. Ajdačič, _Nucl. Instrum. Meth._ 108 (1973) 297.
8C.R. Emigh and L.R. Megill, _Phys. Rev._ 93 (1954) 1190.
9P. Doll, et al., _Nucl. Instrum. Meth._ A 285 (1989) 464.
10J.C. Barton, _Appl. Rad. Isot._ 47 (1996) 997.
11K. Ichihara, et al., _Nucl. Instrum. Meth._ A 515 (2003) 651.
12C. Bacci, et al., _Phys. Lett. B_ 293 (1992) 469.
13R. Bernabei, et al., _Nucl. Instrum. Meth._ A 592 (2008) 297.
14R. Bernabei, et al., _Phys. Lett. B_ 389 (1996) 757.
15D.R. Tovey, et al., _Phys. Lett. B_ 433 (1998) 150.
16V. Kudryavtsev, et al., _Phys. Lett. B_ 452 (1999) 167.
17G. Gerbier, et al., _Astropart. Phys._ 11 (1999) 287.
18L. Miramonti, _Rad. Phys. Chem._ 64 (2002) 337.
19K. Fushimi, et al., _Phys. Rev. C_ 47 (1993) 425.
20K. Fushimi, et al., _Astropart. Phys._ 12 (1999) 185.
21M.L. Sarsa, et al., _Phys. Lett. B_ 386 (1996) 458.
22R. Bernabei, et al., _Nucl. Instrum. Meth._ A 375 (1996) 757.
23M.L. Sarsa, et al., _Phys. Lett. B_ 293 (1992) 469.