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Measurement of scintillation efficiencies and pulse-shapes for nuclear recoils in NaI(Tl) and $CaF_2(Eu)$ at low energies for dark matter experiments

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Abstract

Measurements have been performed with a 2.85 MeV mono-energetic neutron beam of relative scintillation efficiency and pulse-shape for nuclear and electron recoils in Nal(Tl) and CaF₂(Eu). Scintillation efficiencies in Nal(Tl) relative to 60 keV gamma events were found to be 27.5 ± 1.8% for Na recoils (recoil energy $E_{rec} > 4$ keV) and 8.6 ± 0.7% for I recoils ($E_{rec} > 10$ keV). Relative scintillation efficiencies in CaF₂(Eu) for Ca and F recoils show some evidence for a fall with energy (17% to 8% for F) for 10 keV < $E_{rec} < 100$ keV. Pulse-shape analysis of NaI(Tl) data gives a mean photo-electron arrival time $\langle \tilde{t}_i \rangle$ of 263 ± 15 ns for Na events (visible energy E_{vis} in the range 2–8 keV and 272 ± 10 ns for I events (2 keV < $E_{vis} < 5$ keV). The $\langle \tilde{t}_i \rangle$ of 4 keV < $E_{vis} < 54$ keV electron events in NaI(Tl) is found to rise with energy (320 ns to 440 ns). Similar analysis of CaF₂(Eu) data shows $\langle \tilde{t}_i \rangle$ for 2 keV < $E_{vis} < 33$ keV Ca, F and electron events to rise with energy (660 ns to 850 ns) with no evidence for pulse-shape differences. © 1998 Elsevier Science B.V. All rights reserved.

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1. Introduction

Weakly Interacting Massive Particles (WIMPs) constituting the galactic Dark Matter may produce nuclear recoils in conventional matter through elastic scattering via spin-dependent or coherent interactions [1]. Several experiments searching for WIMPs make use of scintillation detectors sensitive to these low energy (< 100 keV) events [2–4]. In some scintillation materials the scintillation pulse-shape is a function of the dE/dx of the recoiling particle [5–7] and

this permits the use of Pulse-Shape Analysis (PSA) to identify nuclear recoil signal events and reject electron recoil background events [2,7]. NaI(Tl), with its high light-output and large pulse-shape differences is thus of considerable interest as a detector material and it has the additional feature that its principal isotopic constituents, ²³Na and ¹²⁷I, are sensitive to both spin-dependent and coherent WIMP interactions. The advantageous spin-coupling characteristics of ¹⁹F [8] make CaF₂(Eu) a further suitable material and several detectors incorporating this are

currently operating or under construction [9,10]. One particular proposal uses it as one component of a mixed scintillator detector capable of discriminating signal from background on the basis of recoil-range alone [11].

The performance of these detector materials must be accurately known in order to determine their sensitivity to Dark Matter particles. It is necessary to measure the efficiency with which the energy of nuclear recoils is converted into scintillation light relative to that for the electron recoils used in calibration and in addition it is essential when performing PSA to have a detailed knowledge of the characteristics of the scintillation pulse-shape for signal and background events. Measurements of these quantities may be obtained by studying nuclear recoil events caused by elastic scattering of neutrons from a mono-energetic beam [7]. In previous work [12-14] measurements of relative nuclear recoil scintillation efficiencies in several different materials, including Na and I in NaI(Tl) and Ca and F in CaF₂(Eu), were carried out at recoil energies > 15 keV with a 5.5 MeV neutron beam. In this work we extend these results to lower energies and in addition quantify the scintillation pulse-shapes for Na, I and electrons in NaI(Tl) and for Ca, F and electrons in $CaF_2(Eu)$.

2. Technique

The beam used in this work was an Activation Technology Corporation source producing 2.85 MeV mono-energetic neutrons through the d(d, ³He)n reaction, mounted in a $6m \times 5m \times 4m$ scatter chamber at the University of Sheffield. The energy of the beam was monitored throughout by observing the recoil spectrum of H scattering events in a neutron-discriminating Nuclear Enterprises NE213 liquid scintillation counter situated in the beam. No variations in energy were observed over the course of the experiments. The source produces ~ 10^9 neutrons per second isotropically and is heavily shielded with wax, lead and iron to absorb stray neutrons and

Fig. 1. Scintillation efficiency relative to 60 keV gammas for (a) Na, I and electron recoils in NaI(Tl) and (b) Ca, F and electron recoils in CaF₂(Eu).



gammas in an arrangement described in [12]. A narrow double-wedged collimator of mean diameter ~ 18 mm passes through the shielding to the beam head, from which it is separated by ~ 15 mm of Pb to reduce gamma background. The target crystals (25 mm $\phi \times 25$ mm) were mounted in the beam on an ETL 9266A 50 mm bi-alkali photomultiplier tube with differential voltage output. For the NaI(TI) tests a Hilger Analytical Ltd encapsulated crystal with 1000 ppm Tl doping was used while for the CaF₂(Eu) tests the crystal was unencapsulated and of doping 0.5% M Eu. Both crystals were wrapped in PTFE in order to optimise light collection. Neither crystal had been used in previous tests.

Nuclear recoils of a particular energy were selected by looking for coincidences between events in the target detector and those in a 75 mm ϕ NE213 counter situated ~ 0.5 m from the target at an angle θ from the beam direction. Kinematics then uniquely specify the nuclear recoil energy. The NE213 counter was shielded from neutrons scattered from the walls of the chamber by borax blocks mounted on all sides except that facing the target. The output from the counter was fed into a LINK systems 5020 pulseshape discrimination unit providing a neutron trigger signal. A TAC module was used to compare the arrival times of events in the target and those in the NE213 unit. Valid event pulses within a 1 μ s coincidence window were digitised with a Lecroy 9430 DSO and passed to a Macintosh computer running custom DAQ software for storage on disk. Also stored were the TAC amplitudes to permit further cuts during off-line analysis. Sharp TAC peaks (width < 50 ns) were observed indicating good signal-tobackground performance. Further details of the experimental procedure may be found in [12,13].

3. Results

Data was taken at a variety of neutron scattering angles between 105° and 15° and in addition a 1 μ Ci 60Co source was used to produce low energy recoils

Fig. 2. Scintillation pulse mean time for (a) Na, I and electron recoil events in NaI(Tl) and (b) Ca, F and electron recoil events in CaF₂(Eu).



for comparison with the nuclear recoil data. Fig. 1 shows the results of the measurements of relative scintillation efficiencies for nuclear recoils in NaI(TI) and $CaF_2(Eu)$. The mean visible energies of the nuclear recoils at each scattering angle were determined by Gaussian χ^2 fits to the appropriate peaks in the energy spectrum. Calibration of the detectors in terms of visible energy was performed with the 59.57 keV gamma line from a 10 μ Ci ²⁴¹Am source. All scintillation efficiency results are hence calculated relative to this high energy electron recoil calibration point. This mirrors the procedure used in operational Dark Matter experiments where all energy calibration is carried out with high energy sources [2]. Although this technique does not take into account non-linearities in crystal response or electronics at low energies it is thus the appropriate technique for comparison with data from operational detectors. Non-linearities in the response to low energy gamma events were nevertheless investigated with foil X-ray sources and these results are also presented in Fig. 1. In all cases the total error in the relative scintillation efficiency is dominated by systematic effects due to the finite sizes of the target crystals and NE213 coincidence counter. The error in beam energy (< 50 keV) has a negligible effect. A potential source of error in the Na recoil data at $E_{rec} > 100$ keV was due to a contamination with < 20% gamma events due to the 57 keV inelastic I peak, however these events were removed using PSA. For comparison both statistical errors and total errors are displayed on data points in Fig. 1. Statistical errors are on average a factor of 6 smaller than systematics in Fig. 1(a) and a factor of 7 smaller in Fig. 1(b).

The results for NaI(Tl) are consistent with previous measurements at higher energies [12] and indicate average Na and I relative scintillation efficiencies of $27.5 \pm 1.8\%$ and $8.6 \pm 0.7\%$ in the recoil energy ranges 4-252 keV and 10-71 keV respectively. The CaF₂(Eu) results on the other hand show some evidence for a fall in the relative scintillation efficiencies for Ca and F in the energy range 10

Fig. 3. Photo-electron distribution for scintillation pulses (a) from Na and electron recoils in NaI(Tl), visible energy 10-20 keV, and (b) F and electron recoils in CaF₂(Eu), visible energy 10-25 keV.



t (ns)

 $\rm keV < E_{\rm rec} < 100\,$ keV. The low values at higher energies are consistent with earlier results [13]. This behaviour is suggested by the inverse dependence of the relative scintillation efficiency upon the dE/dx of the recoiling nucleus [6], which will fall at very low energies. A rise in relative scintillation efficiency at low energies would have important consequences for Dark Matter experiments since it would imply lower recoil energy thresholds and hence sensitivity to a greater part of the Dark Matter energy spectrum.

Scintillation pulse-shapes in NaI(Tl) and $CaF_{2}(Eu)$ are generally taken to consist of a single exponential decay component [2,3]. PSA makes use of the difference in the mean time constant of this decay between nuclear and electron recoil events. This effect is strongly present in NaI(Tl) but much less so in $CaF_{2}(Eu)$. In the case of a single exponential decay this time constant corresponds to the mean arrival time of the photo-electrons in the pulse relative to the pulse start-time. Since this start-time is not known *a priori* it must be approximated by the arrival time of the first photo-electron, valid for large numbers of photo-electrons. For a given energy range the distribution of these mean photo-electron arrival times $\langle t_i \rangle$ will be a log-normal distribution [2] with mean $\langle t_i \rangle$ characteristic of the recoiling particle, be it nucleus or electron. For this statistical property of the scintillation pulses to form the basis of PSA it is clearly essential to know $\langle \bar{t}_i \rangle$ for nuclear and electron recoil events in the energy range of interest.

In order to measure the $\langle \bar{t}_i \rangle$ at each scattering angle the $\langle t_i \rangle$ of events in 1σ energy regions about the recoil peaks were calculated. Log-normal functions were then fitted to the distributions of these $\langle t_i \rangle$ and values for $\langle t_i \rangle$ extracted, as in [2]. These results are presented in Fig. 2 for both NaI(Tl) and CaF₂(Eu). The errors on the $\langle \bar{t}_i \rangle$ in Fig. 2 include statistical fluctuations in the $\langle t_i \rangle$ distributions and systematic effects due to the variation in temperature of the crystals over the course of the experiments. This temperature was monitored continually and was found to vary from a mean of 11°C by no more than $\pm 3^{\circ}$ C. For NaI(Tl) a 1°C temperature change leads to a 3 ns shift in decay time [2] and consequently gives a contribution to the error of ± 9 ns. For $CaF_2(Eu)$ the effect is smaller and leads to an additional error of conservatively no more than ± 5 ns.

Also presented in Fig. 2 for comparison are the $\langle t_i \rangle$ for electron recoils generated by the ⁶⁰Co source.

The overall mean $\langle \bar{t}_i \rangle$ for nuclear recoil events in NaI(Tl) was found to be 281.0 + 5.1 ns, while the values for electron recoil events rise rapidly with energy from ~ 320 ns (E_{vis} ~ 7 keV) to ~ 440 ns $(E_{vis} \sim 52 \text{ keV})$. The mean $\langle \bar{t}_i \rangle$ for Na events (2) $keV < E_{vis} < 8 keV$) and I events (2 $keV < E_{vis} < 5$ keV) were found to be 263 + 15 ns and 272 + 10 ns respectively and hence there is no evidence for significant pulse-shape differences between them. This supports the premise [2] that coherent I scattering events in Dark Matter detectors can be simulated with Na recoil events induced by exposure to a fission neutron source (e.g. 252 Cf). In CaF₂(Eu) the $\langle \bar{t}_i \rangle$ for both nuclei and electrons rise from ~ 660 ns ($E_{vis} \sim 6$ keV) to ~850 ns ($E_{vis} \sim 30$ keV). In this case no evidence is found for significant differences in $\langle \bar{t}_i \rangle$ for either electron. Ca or F events.

The lack of electron contamination in the event samples obtained with this technique allows direct examination of the scintillation pulse-shapes to investigate deviations from pure exponentials. The photo-electron arrival time distributions for 10 keV $< E_{vis} < 25$ keV F and electron recoil events in $CaF_2(Eu)$ are plotted in Fig. 3(b) and show no differences within errors. These may be compared with the equivalent distributions for 10 keV $< E_{vis} <$ 20 keV Na and electron recoil events in NaI(Tl) plotted in Fig. 3(a). Although the nuclear recoil pulse-shape in this case is well approximated by a single exponential the electron recoil pulse-shape displays a more pronounced flattening at the beginning of the pulse, as reported in [5,7]. This contributes significantly to the slower overall $\langle \bar{t}_i \rangle$ of electron recoil events. A Likelihood Ratio test with these two distributions would make optimal use of this additional pulse-shape information when performing PSA of data from NaI(Tl) Dark Matter experiments and this technique is currently under study. This should lead to significantly improved discrimination and hence Dark Matter sensitivity.

4. Conclusions

Measurements have been performed at low energy of relative scintillation efficiency and pulse-shape for nuclear and electron recoils in NaI(Tl) and CaF₂(Eu). We conclude that while the relative scintillation efficiencies of Na and I and NaI(Tl) remain constant with energy at 27.5 ± 1.8% ($E_{rec} > 4$ keV) and 8.6 ± 0.7% ($E_{rec} > 10$ keV) respectively, those of Ca and F in CaF₂(Eu) show some evidence for a rise towards lower energy ($E_{rec} < 100$ keV). Significant pulse-shape differences between Na and electron recoil events were found in NaI(Tl), beyond those assumed in current Dark Matter experiments. No differences in pulse mean time between Na and I recoil events for $E_{vis} < 5$ keV were seen however. No pulse-shape differences of any kind were found in CaF₂(Eu).

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