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Demonstration of nuclear recoil discrimination using recoil range in a mixed CaF_2 + liquid scintillator gel detector for dark matter searches

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Abstract

We present first measurements on a prototype dark matter detector being developed to achieve event by event discrimination of nuclear recoils from electron recoils below 100 keV by utilising the difference in the recoil ranges of these particles. The detector consists of sub-micron scintillating grains of CaF_2 suspended in Dioxan gel scintillator with matched refractive index. We call this form of detector CASPAR (Cocktail of Alkali halide Scintillating PARticles). We present here results of monoenergetic neutron scattering tests on CASPAR and show how scintillation pulse shape analysis can be used as a powerful means of distinguishing Ca, F, C and H recoil events from electron recoils. > 90% discrimination of Ca and F recoils from electrons at 60 keV was observed for < 5% loss of signal. © 1997 Elsevier Science B.V.

1. Introduction

Discrimination of background electron recoils from nuclear recoils is a fundamental requirement of experiments aiming to detect WIMP dark matter by nuclear elastic scattering. This is because the electron background spectrum, arising ultimately from detector radio-impurities, can mimic the expected nuclear recoil event spectrum and yet is unlikely ever to be reducible sufficiently by radio-purification alone to render it unimportant [1]. Scintillation pulse shape analysis in NaI(Tl) has now been demonstrated as one route to discrimination and new limits for neutralinos have recently been obtained by this method [2,3]. However, the technique has not so far been powerful enough for full event by event discrimination at low energy (< 100 keV). Rather it has been of a statistical nature, requiring the accumulation of events such that, for instance, a histogram of pulse decay time constants can be obtained and compared with distributions for electron and nuclear recoils obtained using calibration gamma and neutron sources. The statistical significance with which a deviation of the distribution for the data away from that for electrons, towards that for nuclear recoils, is not observed is then used set limits on the WIMP flux [2].

An important objective for WIMP searches now is to improve scintillation detectors towards event by event discrimination while maintaining prospects for using target mass > kgs at relatively low cost. Such discrimination would enhance sensitivity but also allow this to improve at close to proportionality with detector mass × run time (Mt) rather than $(Mt)^{1/2}$ as

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provided by statistical discrimination [4]. Neutron and gamma calibrations would still be required but in this case mainly to determine where to place cuts to achieve event separation. Event by event discrimination has been demonstrated in low temperature Ge and Si detectors in which ionization and thermal signals are measured simultaneously. However, the mass of these detectors has so far been limited to < 100 g [5,6].

Improved pulse shape discrimination in NaI is possible by altering the operating temperature and Tl doping to optimize the intrinsic pulse shape characteristics [7]. Pulse shape discrimination in alternative scintillators, such as liquid Xe, has also been studied [8]. However, confirmation is still required that these techniques can sufficiently bring down the minimum energy at which event by event discrimination occurs. Therefore, new scintillator-based discrimination techniques have been sought. One possibility is to make use of the range of recoiling nuclei. In a solid this is typically ~ 10 times shorter than for electrons of the same energy [9] and so provides a potentially powerful alternative route to discrimination if a detector can be made sensitive to this difference.

2. The CASPAR detector

In an earlier paper we described how such a 'range discriminating' detector might be feasible using a scintillator cocktail of sub-micron inorganic scintillator grains suspended in a matrix gel of organic liquid scintillator [10]. We have termed this CASPAR – Cocktail of Alkali halide Scintillating PARticles. Certain characteristics are required for scintillator materials for a CASPAR detector, these are:

(i) high radio-purity ($<$ ppb U and Th, $<$ ppm K);

(ii) high scintillation efficiency ($> \sim 10$ photons/$\mathrm{keV_{electron}}$);

(iii) liquid and inorganic halide components with similar refractive indices;

(iv) scintillator components with very different scintillation decay constant τ_c – slow in the inorganic (> 100 ns) and fast in the organic (< 10 ns).

Furthermore, fabrication of the detector requires addition to the cocktail of a second organic liquid of different refractive index (RI) used to match the net liquid RI to that of the grains. This is to ensure that there are no internal grain–liquid boundary reflections so that the mixture is transparent and will allow scintillations from both components out of the mixture, eventually to photomultiplier tubes.

The principle of the technique is that background electron recoil events will usually have a path length sufficiently long compared to the diameter of the grains that most, if not all, the scintillation produced from these will arise from the organic liquid. Hence the pulse shape for these events will have a large fast component. Conversely, nuclear recoil events in the inorganic scintillator grains, the nuclei of which form the WIMP 'target' in this detector, will have tracks typically $\sim \times 10$ shorter and so will remain largely within the grains to yield slow scintillation pulses. Any nuclear recoil events in the liquid would give fast events and so would also be distinguishable from the target nuclei events in the grains – the recoil events in the liquid being lost in the electron recoil background. Further design details and predictions of sensitivity can be found in [10].

The choice of materials available to construct a CASPAR detector are principally limited by item (iii) above. This essentially rules out the use of NaI as the inorganic component since this has a high refractive index (1.8) and no suitable organic scintillator with RI close to this appears readily available. An alternative is to use CaF_2. Although in undoped form this material has quite low scintillation efficiency ($\sim 7\%$ of NaI(Tl)), when doped with Eu it can reach scintillation efficiency comparable with NaI(Tl) ($\sim 50\%$) [11]. More importantly it has a very suitable RI (1.44) and is non-hygroscopic, making it easier to handle than NaI. The radio-purity of typical CaF_2 material is known to be poorer than NaI [12]. However, new purification techniques now being used for NaI are, in principle, applicable to CaF_2 so this problem should be surmountable in the future [13]. A further advantage of CaF_2 is the presence of fluorine which is thought likely to have an unusually high cross section for neutralino scattering [14].

The present tests made use of commercially available CaF_2 precipitate powder. This had a mean grain diameter of ~ 500 nm (measured using x-ray diffraction) and RI = 1.43. It was not possible to dope the CaF_2 grains with Eu at this stage. The

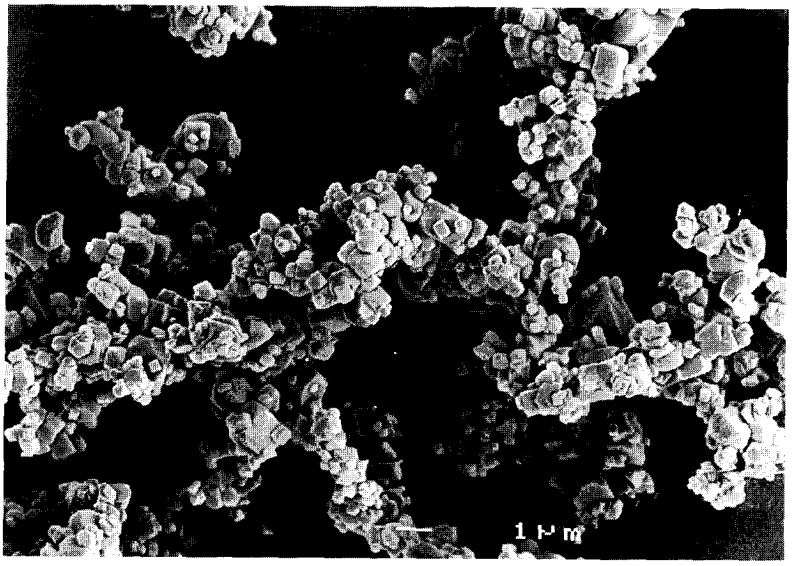

Fig. 1. Electron microscopy photograph of a typical CaF_2 precipitate sample.

single crystal nature of these sub-micron grains was confirmed using electron microscopy. Fig. 1 shows a typical sample. It can be seen that single crystals predominate which (out of suspension) occur in loose conglomerates. Tests with a UV laser were used to confirm that these grains do indeed scintillate and the time constant was subsequently found, using gamma sources, to be $\tau_c \sim 900$ ns. Fig. 2 shows the emission spectrum for a typical sample of precipitate powder grains obtained by uv stimulation. The expected emission for undoped CaF_2 at ~ 390 nm can be seen.

To construct the detector the powder was added to a mixture of Napthalene (RI = 1.60) and 1,4-Dioxan (RI = 1.42) supplied by Zinsser Analytic (UK) Ltd. (the liquid components being based on Zinssers' Quickszint-368 scintillator and yielding a time constant $\tau_c \sim 4$ ns). Enough CaF_2 was added to give a volume packing fraction for the grains of 10%. The RIs of the liquid and inorganic scintillators were then matched by adding methanol (RI = 1.33) while the whole cocktail was simultaneously formed into a gel matrix using cab-o-sil. The cocktail was then transferred to a 50 mm × 50 mm diameter cylindrical quartz cell wrapped circumferentially in diffuse reflecting PTFE tape. The cell was then mounted on an

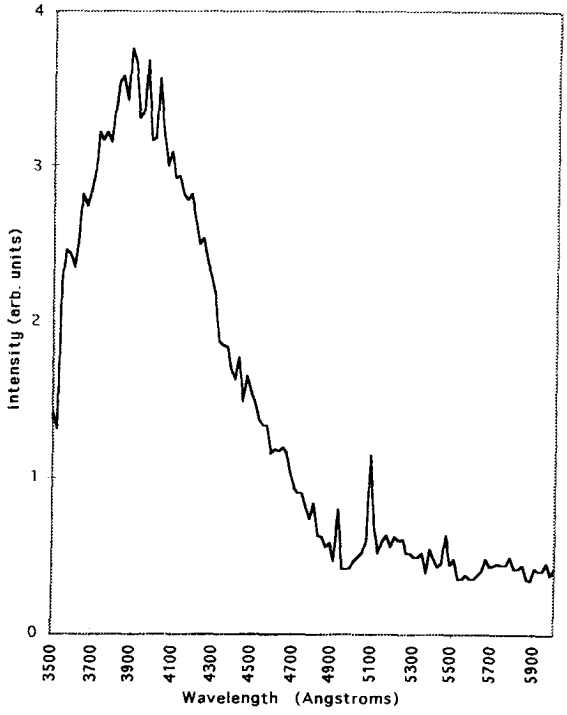

Fig. 2. Laser-corrected PL uv stimulated emission spectrum of a typical sample of precipitate CaF_2 grains.

Electron Tubes Ltd. 9266A 50 mm photomultiplier using silicone optical grease.

3. Neutron scattering tests

The optimum means of measuring the response of a detector to nuclear recoils is to use scattering by monoenergetic neutrons. For the present work neutrons were provided by the collimated 5.5 MeV mono-energetic neutron beam at the University of Birmingham Dynamitron facility, the CASPAR detector being mounted in the line of this beam. This allowed experiments to be performed in which neutron scattering events could be identified by registering scintillation pulses in the CASPAR detector in coincidence with events in an NE213 neutron sensitive detector placed at various known angles relative to the CASPAR-beam line, and at a fixed distance from the CASPAR detector (set to be 1 m). Knowledge of the neutron energy and the scattering angle allows the energy of individual nuclear recoil events to be calculated and compared with the observed scintillation pulse. The photomultiplier signals from CASPAR were passed into an integrating buffer and on into a Lecroy 9430 10-bit DSO. This was connected by GPIB to a Macintosh computer running in-house LabView data acquisition (DAQ) software. Incorporated into the DAQ system was neutron time of flight measurement used as an additional cut on the events. Further details of the techniques and apparatus can be found in [15,16].

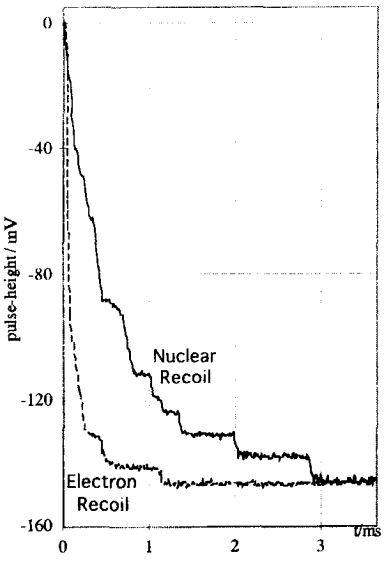

Fig. 3. Example pulses from CASPAR for: (a) nuclear recoils within grains and (b) electron recoils.

Experiments were performed with sealed gamma sources and with neutrons scattered at various angles. In all cases pulse shapes were recorded and analysis performed using IDL software routines. Fig. 3 shows some example pulses. The basis of the analysis was the characterisation of the shape of each pulse in terms of a parameter R defined as the ratio of the amplitude of the slow component to the total pulse-height [17]. In principle this parameter can be extracted by fitting the sum of two exponentials to the integrated scintillation pulses using decay time constants appropriate for the liquid and CaF_2, in this case 4 ns and 900 ns respectively. In practice the actual time constants as measured were lengthened by the rise time associated with the electronics chain, in particular the buffer circuit, which resulted in a minimum measurable time constant of ~ 20 ns. The pulse shape analysis procedure differs from that used previously with NaI where each integrated pulse was fitted by a single exponential and the value of the time constant obtained used as the discrimination parameter [2].

The results of measurement of R versus pulse-height are shown in Fig. 4 for neutrons with scattering angles of 120°, 90° and 60° respectively (Fig. 4b–d) together with a calibration run from an ^{241}Am source (60 keV gammas) used to determine the response of the detector to electron recoils (Fig. 4a). In these plots the area of the squares is proportional to the number of counts. The ^{241}Am 60 keV photopeak is clearly seen with a low value of $R \sim 0.25$. This is consistent with only a small fraction of each electron recoil track passing through the CaF_2 grains. The R value is slightly higher than would naively be expected from the known grain packing fraction ($R \sim 0.11$). The difference arises from pile up of the pulses with later arriving single photoelectrons which can mimic a small slow component. This produces an effective DC offset in the R value. From the data the scintillation efficiency of the liquid mixture was found to be ~ 0.45 photoelectrons/keV.

It can be seen that in all the neutron runs (Fig. 4(b–d) the distribution of events is quite different

from that found for the electron events. Considering Fig. 4b first (neutrons scattered by 120°), the diffuse cluster of events lying between pulse height amplitudes of 0.05–0.15 V have the high R value of 0.7–1.0. That is, they are dominated by a very large slow (~ 900 ns) component (or are entirely slow) and so can be interpreted as events from the CaF_2 grains. Since any electron recoils have track lengths typically much greater than the measured grain diameters, and should anyway be rejected by the coincidence and TOF requirements, these events are interpreted as due to Ca and F recoils. The recoil scintillation efficiency for Ca and F in large single crystals has previously been measured as 8% and 12% respectively [16]. Assuming these values apply for the micro-crystals here then the expected electron equivalent energy deposited at this neutron scattering angle would yield 31 keV and 94 keV which for 0.05–0.15 V pulse height corresponds to a light collection efficiency for the grains of ~ 0.5 photoelectrons/keV. This is in agreement with that expected assuming the grains also have approximately the same gamma scintillation efficiency as a larger single crystal of the same material. The light collection efficiency was not expected to be sufficient to resolve separate Ca and F peaks. Note that because the grains are so small it is not possible accurately to measure their electron scintillation efficiency with gamma sources directly because the induced electron track lengths are rarely short enough to produce scintillation totally within a given grain and those that would produce all slow pulses would be below threshold. However, some estimate can be made by analysing the form of the pulse height spectrum of the slow portion of gamma pulses. These estimates were found to be consistent with the value of ~ 0.5 photoelectrons/keV.

The cluster of events with pulse-height of ~ 0.37

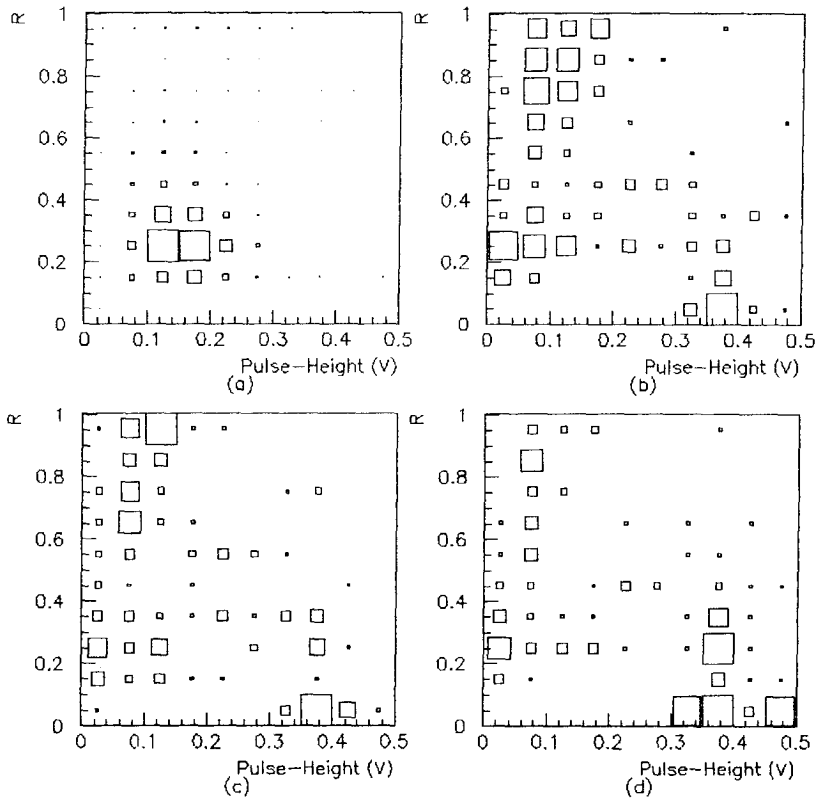

Fig. 4. R versus pulse height for: (a) ^{241}Am gamma events, and for neutrons with scattering angles of (b) 120°, (c) 90° and (d) 60° respectively.

V and low R (see Fig. 4(b–d)) are consistent with H recoils which in this case are being cut off by the maximum measurable pulse-height of $V_{max} = 0.4$–0.5 V and appear as different events with amplitudes lying around this maximum. The measured proton recoil scintillation efficiency of liquid scintillators is typically 10% relative to electrons [18] which, given the measured scintillation efficiency for gammas in this detector, is consistent with pulse-heights for these events of > 0.4 V. The long H recoil track length and large pulse height amplitude allowed only the first part of these pulses to be recorded within the dynamic range of the DAQ, consistent with the extremely low value of R found (though there is some straggling to higher R values arising from lower energy H recoils and this increases as the scatter angle drops as expected).

Also seen in Fig. 4b is a cluster at pulse-height of ~ 0.05 V with an R of ~ 0.25, similar to the electron events of Fig. 4a. However, the requirement in these tests for events in CASPAR to be in coincidence with neutron detection in the NE213 and with a well characterised time difference corresponding to the neutron time of flight (TOF) means this cluster is unlikely to result from gammas. They are instead interpreted as C recoils in the liquid scintillator and indeed the pulse height is consistent with the carbon recoil efficiencies typically measured in organic (C_nH_m) scintillators of 2–4% in this energy range [19].

Assuming the events in Fig. 4b are due to nuclear recoils then as the neutron scatter angle is reduced we would expect all but the H cluster to move to lower energy, consistent with the reduced recoil energies of the nuclei. As indicated above, the high energy H events pile up in the upper bins so would not be expected to move. This is shown in Fig. 4c and d where data are plotted using identical procedures as for Fig. 4b but at recoil angles of 90° and 60° respectively. The expected recoil energies at these angles, using the assumed efficiency factors for Ca (8%) and F (12%) [16] are 21 keV and 63 keV respectively at 90° and 10 keV and 31 keV at 60°. The clusters interpreted as C, Ca and F events are seen to move to lower energy by the predicted amount within the statistics. As expected they are also seen to de-populate relative to the H events due to a reduction in detection efficiency as the distributions approach the low energy threshold of the detector, set here at 2 photoelectrons, and more events are lost below threshold.

Thus there is clear evidence that Ca and F events are observed in CASPAR and can be distinguished from gammas. It is the Ca and F recoil events observed in the detector which are of interest in dark matter searches as it is the Ca and F that would provide the best WIMP target. Comparing the Ca + F cluster of Fig. 4b with the gamma calibration of Fig. 4a and optimizing a simple cut yields a level of electron/nuclear recoil discrimination of $> 90\%$ at 60 keV (electron equivalent), with a corresponding loss of signal of $< 5\%$.

4. Discussion and conclusions

The preliminary tests here indicate that range-based nuclear recoil discrimination in a CASPAR type detector can give significant discrimination below 100 keV ($> 90\%$ at 60 $keV_{electron}$) and has the potential for event by event discrimination. In a practical WIMP search experiment, requiring exploration of events at < 50 keV_{recoil}, an energy threshold below ~ 5 $keV_{electron}$ would be needed together with detector radio-purity at least as good as NaI(Tl). Although the present detector did not achieve a low threshold (it was not specifically designed to) a clear route towards this has been identified. Firstly, the precipitate CaF_2 grains need to be doped with Eu. This is known to increase the scintillation efficiency in large single crystals by $\sim \times 8$. Growth of doped sub-micron crystals should, in principle, be feasible using reactions with $EuCl_2$ or $EuCl_3$ and these are now under investigation. Secondly, the grain size needs to be reduced (to < 500 nm) to improve discrimination in the low energy (shorter recoil track length) region [17]. This appears possible by careful control of the precipitate growth using ultrasound (a standard chemical technique [20]). The radio-purity of CaF_2 is known to be generally poorer than NaI [12]. However, efficient extraction of actinides from NaI powder has recently been demonstrated using specialised chealating agents [13] and there is no practical reason why this technique can not be applied to CaF_2 purification. Several further improvements are possible, in particular, a reduction in the

minimum measurable time constant. An upgrade to incorporate non-integrating buffer electronics and faster photomultipliers will rectify this.

Not only may CASPAR open the possibility of a new form of WIMP detector with powerful recoil discrimination but there are several other important advantages over existing techniques. Firstly, it has the advantage of simplicity and low cost. Secondly, scintillation detectors based on intrinsic pulse shape discrimination show a reducing intrinsic discrimination power towards lower energies [2]. This should not be a problem in CASPAR since the electron and nuclear recoil path lengths used for discrimination remain sufficiently different even at low energy ($<$ 10 keV). Thirdly, the presence of H nuclei in CASPAR may make it feasible to monitor any remnant neutron flux (such as from alpha or fission background in the detector) which would otherwise not be rejected by the discrimination. That is, although such neutron events would not be rejectable it is feasible that their presence or otherwise might be determined. This arises because any population of background neutrons is likely to have a population at low energy that would result in H recoils with a continuous energy distribution. These would have a fast scintillation component (from the liquid) making them distinguishable from those of Ca and F. In practice, the possibility of H recoiling into grains means the events may be visible by a 'washing out' effect on the event distribution in R with no isolated cluster observed at $R \sim 1.0$. Finally, in the CASPAR technique the signal pulse is slow and therefore well removed from most photomultiplier noise events which are fast (< 10 ns). This is not the case for alternative scintillator-based dark matter detectors based on pulse shape analysis such as the UVIS technique [4] or liquid xenon [8].

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