Scintillation time dependence and pulse shape discrimination in liquid argon

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(Dated: September 23, 2008)

Using a single-phase liquid argon detector with a signal yield of 4.85 photoelectrons per keV of electronic-equivalent recoil energy (keVee), we measure the scintillation time dependence of both electronic and nuclear recoils. We develop two methods of pulse shape discrimination to distinguish between electronic and nuclear recoils. Using one of these methods, we measure a background and statistics-limited level of electronic recoil contamination to be $7.6 \times 10^{-7}$ between 52 and 110 keV of nuclear recoil energy (keVr) for a nuclear recoil acceptance of 50% with no nuclear recoil-like events above 62 keV. Finally, we develop a maximum likelihood method of pulse shape discrimination based on the measured scintillation time dependence.

PACS numbers: 61.25.Bi, 29.40.Mc, 95.35.+d

I. INTRODUCTION

Recent years have seen an increase in the number of experiments using noble liquids as materials for detecting Weakly Interacting Massive Particles (WIMPs), a well motivated dark matter candidate [1]. The current best limit for the spin-independent WIMP-nucleon cross section for a 60-GeV WIMP mass is $4.6 \times 10^{-44}$ cm$^2$, set by the Cryogenic Dark Matter Search (CDMS) experiment [2]. Because many noble liquids have high scintillation yields, are easily purified of radioactive impurities, and are likely scalable to large masses with relative ease, they hold great promise for this application. The current best limit from a noble liquid detector is $8.8 \times 10^{-44}$ cm$^2$ at 100 GeV, set by XENON10 [3]. Noble liquid detectors with larger target masses will likely improve on these limits.

The key to the noble liquid dark matter detectors is discriminating between nuclear recoil events that constitute a WIMP signal and electronic recoil events that form the primary backgrounds. The XENON and ZEPLIN experiments are designed to collect both scintillation light and ionization from liquid xenon [4, 5]. These are dual-phase detectors that use both scintillation light and ionization charge collection to discriminate between event classes, as nuclear recoils and electronic recoils produce different ratios of charge to light. Liquid argon is an attractive alternative to liquid xenon due to the lower cost of natural argon and its simpler purification requirements. The WIMP Argon Programme (WARP) and Argon Dark Matter (ArDM) experiments employ dual-phase detectors that use liquid argon as the target [6, 7].

Alternatively, a single-phase detector collecting solely scintillation light might distinguish electronic and nuclear recoils using pulse shape discrimination (PSD). In 1977, Kubota et al. showed that the time dependence of scintillation light in liquid xenon and liquid argon is significantly different for heavy ionizers such as $\alpha$ particles and fission fragments when compared to light ionizers such as $\beta$ decay and Compton-scattered electrons [8]. This is because scintillation in liquid noble gases is produced by the decay of excimers that can exist in either singlet or triplet molecular states, which have very different lifetimes (Table I). The slow scintillation light emitted by triplet molecules can be suppressed in intensity by destructive triplet interactions, primarily Penning ionization and electron-triplet spin exchange; it is believed that these reactions are stronger for high excitation densities such as those produced by nuclear recoils, causing the observed time dependences. Therefore, the relative amplitudes of the fast and slow components can be used to determine which type of excitation occurred for a given event.

| Liquid | Singlet Lifetime (ns) | Triplet Lifetime (ns) |
|--------|-----------------------|-----------------------|
| Ne     | $< 18.2 \pm 0.2$      | $14900 \pm 300$       |
| Ar     | $7.0 \pm 1.0$         | $1600 \pm 100$        |
| Xe     | $4.3 \pm 0.6$         | $22.0 \pm 2.0$        |

TABLE I: Lifetimes of the singlet and triplet states for neon, argon, and xenon excimers [9, 10].

Pulse shape discrimination based on the timing of scintillation light has been studied for use with several noble liquids. In liquid helium, PSD has been studied in order to separate electronic recoil events from $^3$He($n,p)^3$H events in the search for the permanent electric dipole moment of the neutron [11]. PSD has also been used to suppress $\gamma$-ray backgrounds in liquid xenon [12, 13]. McKinsey and Coakley [14] pointed out that the much longer triplet lifetime in liquid neon should allow supe-
ior PSD, which has recently been verified experimentally 10. Following this observation, Boulay and Hime recognized that the similar properties of liquid argon could in principle achieve PSD with part per billion levels of electronic recoil contamination (ERC) 12. ERC is defined to be the probability of incorrectly classifying an electronic recoil event as a nuclear recoil event given a particular level of nuclear recoil acceptance. An ERC of $10^{-8}$ or better is required to perform a competitive WIMP search using liquid argon due to the presence of the radioactive isotope $^{39}$Ar, which produces about 1 Bq per kg of atmospheric argon 10, 17, 18. The WARP collaboration has used scintillation timing in combination with an ionization signal to reduce electronic recoil backgrounds in liquid argon 19. The Dark Matter Experiment using Argon Pulse Shape Discrimination (DEAP) has demonstrated a background limited ERC of $5 \times 10^{-6}$ using the DEAP-0 single phase detector for nuclear recoil energies above 1 MeV 20. At a given energy, ERC improves exponentially with scintillation light collection efficiency. For this reason, efficient scintillation light detection is the primary requirement for performing a sensitive WIMP search with negligible background at a suitably low energy threshold.

In this paper we describe measurements of scintillation in liquid argon due to low-energy nuclear and electronic recoils in the energy range relevant to a WIMP dark matter search. We measure the scintillation time dependence of liquid argon for both event classes. We develop two basic PSD methods and measure the level of discrimination in our apparatus. Finally, we use the measured time dependence to develop a maximum likelihood method of PSD.

II. EXPERIMENTAL DETAILS

A. Detector Design

The apparatus consists of a 3.14-liter active volume of liquid argon viewed by two 200-mm-diameter photomultiplier tubes (PMTs) 21, all contained within a stainless steel vessel and vacuum Dewar. Figure 1 shows a schematic of the central volume and PMTs. The active region is defined by a Teflon cylinder 200 mm in diameter and 100 mm in height with two 3-mm-thick fused-silica windows enclosing the top and bottom. The PMTs are held in place by Teflon rings above and below the central windows enclosing the top and bottom. The PMTs are all immersed directly in liquid argon and contained within a 25-cm-diameter by 91-cm-tall stainless steel vessel. The PMTs. Both windows are coated with $(0.20 \pm 0.01)$ mg/cm$^2$ of TPB, while the Teflon cylinder is coated with $(0.30 \pm 0.01)$ mg/cm$^2$. The Teflon cylinder, windows, and PMTs are all immersed directly in liquid argon and contained within a 25-cm-diameter by 91-cm-tall stainless steel vessel.

The stainless steel vessel is in turn housed inside a vacuum Dewar, and argon gas is introduced into the system through a tube on the top of the Dewar. The argon is liquefied in a copper cell mounted to the end of a pulse-tube refrigerator 22 inside the Dewar before flowing through a tube to the stainless steel vessel. All components that come into contact with the gas or liquid are baked to at least 60°C, and the ultra-high-purity argon gas (99.999%) is passed through a heated gas-purification getter 20 before entering the vessel. In addition, the argon is continually circulated through the getter and liquefied at a rate of at least 2.0 standard liters per minute (slpm) to ensure that high purity is maintained. The stability of the system is discussed further in Sec. II C.

The data acquisition system is custom-built around VME-bus waveform digitizers (WFDs); a sample WFD trace from a scintillation event in argon can be seen in Fig. 2. The PMT signals from the detector are divided three ways by a linear fan out with two copies of the signal sent to the WFDs and one sent to a triggering system. Each WFD has four channels that record eight-bit samples at 500 MHz. These samples are stored in a separate programmable-length memory buffer for each channel. For all data presented here, the record length is set to 26 µs. The two copies of the PMT waveforms are recorded separately at unity gain and at an attenuation of ten to increase the effective dynamic range of the eight-bit digitization. The buffer is continually filled but saved to disk only when the triggering system registers a fraction of a photoelectron in both PMTs within a 100-ns coincidence window. Once a trigger has been registered, the DAQ records the event for 22 µs, leaving

![FIG. 1: (Color online) Schematic representation of the scintillation cell.](image-url)
an additional 4 µs of baseline presamples in the data (see Fig. 2). The data are read to a computer via fiber-optic cable, and after the computer has recorded all 26 µs of data, it resets the system for the next event. The data collected by the DAQ software are saved in a ROOT-based file structure [27].

FIG. 2: Example of an electronic recoil event from a single PMT, digitized by the 8-bit WFD, sampling at 500 MHz. Four microseconds of presamples are recorded to measure the baseline.

**B. Data Collection**

All data collected are processed in software. First, for each PMT, the two gain scales are combined into a single waveform. Three µs of baseline presamples are averaged to obtain a baseline and baseline root-mean-square. The baseline is then subtracted from the trace. Ideally, we would count single photoelectrons in a pulse, but since we detect many photoelectrons that produce signals overlapping in time, we integrate the trace in order to determine the total number of photoelectrons. To mitigate the integration noise, we restrict the range of the integral to 50-ns regions in which the trace voltage crosses a threshold of approximately 2/5 the height of a photoelectron. This method is a hybrid of single photoelectron counting and pulse integration.

We then apply three cuts to all data. One cut removes events in which either PMT saturates due to excessive light exposure by rejecting events above an experimentally determined threshold of approximately 2000 times the single photoelectron pulse area. A second cut removes events for which the trigger time (defined as the time at which the voltage rises above 20% of its maximum value) differs by greater than 20 ns between the two PMTs. The third cut is designed to eliminate events that produce light in the windows or the glass of the PMTs. An asymmetry parameter \( A \) is defined as

\[
A = \frac{S_T - S_B}{S_T + S_B},
\]

where \( S_T \) and \( S_B \) are the signal areas in the top and bottom PMTs. For most data, we require \(-0.275 < A < 0.375\), due to a slightly larger gain in the top PMT. This cut is relaxed to \( |A| < 0.4 \) to improve the statistics of the nuclear recoil data described below.

We use a 10-µCi \(^{22}\)Na source to produce electronic recoils. In 90% of \(^{22}\)Na decays, a positron is emitted that immediately annihilates in the surrounding materials to produce 511-keV \( \gamma \) rays with equal and opposite momenta, or “back-to-back”. We use the second \( \gamma \) ray to tag electronic recoil events in the liquid by triggering on a coincidence within a 100-ns window between the PMTs in the argon and a NaI crystal scintillator placed back-to-back with our apparatus. This event tagging reduces backgrounds in our data from other radioactive decays and cosmic rays. To further decrease neutron backgrounds, we place one layer of water-filled containers above and around the sides of the dewar. These containers are cubes of side 30 cm in length and hold 20 liters of water.

In addition to the three universal cuts, we apply two additional cuts to ensure data purity. In software we narrow the coincidence window between the liquid argon PMTs and the NaI crystal to 30 ns. We also make an energy cut to the NaI energy spectrum to select 511-keV events in the NaI crystal. In the liquid argon, the majority of the 511-keV \( \gamma \) Compton scatter, producing events with a continuum of deposited energies.

To investigate the detector response to nuclear recoils, we use a portable deuterium-deuterium neutron generator [28] as a source of 2.8-MeV neutrons and a PMT viewing BC501A organic scintillator as a secondary detector. Both the generator and the organic scintillator are placed approximately 1.63 m from the center of the active volume. We require a detection of a scintillation event in the liquid argon, followed within 200 ns by an event in the organic scintillator. The experimental setup can be seen schematically in Fig. 3. By changing the scattering angle \( \theta \), we can choose the energy of the nuclear recoils observed in the liquid, \( E_{rec} \), using simple kinematics:

\[
E_{rec} = \frac{2E_{in}}{1 + M} \left[ 1 + M, -\cos(\theta) \sqrt{M^2 + \cos^2(\theta) - 1} \right],
\]

where \( E_{in} \) is the incident neutron energy and \( M \) is the atomic mass of the target.

Since we know the energy of the neutron, we can calculate the time-of-flight of a neutron that scatters in the liquid argon and the organic scintillator. We then apply a time-of-flight cut between the liquid argon cell and the organic scintillator to distinguish between \( \gamma \) rays and neutrons, as the neutron generator produces both. In general, the time-of-flight cut requires an event to occur in the organic scintillator 60–90 ns after the event in the liquid argon. In addition, we make a PSD cut in the organic scintillator data to further eliminate electronic recoils.
FIG. 3: (Color online) Schematic of the neutron scattering setup.

We also collect background data by looking at events in the liquid argon with no external source present. This background data provides an estimate of the accidental background rate that may be contaminating the γ-ray data sets, and it will be discussed further in Sec. III B.

C. Detector Calibration

We use a 10-µCi sealed $^{57}$Co source for daily calibrations. This source produces 122-, 137-, and 14.4-keV γ rays, with branching ratios of 86%, 11% and 9%, respectively. Any scintillation event in liquid argon produces a significant triplet component; since this component is spread out over many µs, it appears in the signal as many single photoelectrons well separated in time (for example, there are a number of single photoelectrons that appear after 7 µs in Fig. 2). Therefore, we measure the gain of the PMTs using the $^{57}$Co source by selecting single photoelectrons from the tail end of each pulse. The PMT traces are divided into 75-ns regions centered on times at which the trace crossed an experimentally determined threshold of roughly 1/3 of a photoelectron. These regions are then integrated to obtain the single photoelectron pulse area. The typical gain for the PMTs is approximately $4 \times 10^7$.

Figure 4 shows an example $^{57}$Co spectrum along with a simulation done with the Reactor Analysis Tool, a toolkit of Geant4 developed by the Braidwood collaboration [29, 30]. From the simulation, we find that the position of the primary peak is dominated by the 122-keV γ photoabsorption process. The simulation parameters describing absorption and reflection of the materials in the detector are tuned to match the observed signal yield, and the spectral shapes line up nicely.

By comparing the integrated signal corresponding to the 122-keV peak to that of a single photoelectron, we measure the signal yield of the detector to be 4.85 photoelectrons per keV electron equivalent (keVee), where keVee refers to the amount of energy deposited by an electronic recoil. The response of the detector was stable to within 5% during the four months of data acquisition.

FIG. 4: (Color online) Example spectrum of $^{57}$Co data, along with a simulation done using the Reactor Analysis Tool [30].

We use the 122-keV $^{57}$Co peak to provide a daily energy calibration. To check the quality of that calibration, we use the 511-keV γ rays produced by the $^{22}$Na source as a second point of reference. When calibrated using the $^{57}$Co source and the assumption that the signal scales linearly with deposited energy, the 511-keV absorption line appears in the $^{22}$Na spectrum as expected to within 1%. In addition, simulations of 511-keV γ rays are consistent with the data.

Impurities in the detector can build up over time via outgassing. These impurities can quench argon excimers or absorb emitted UV photons, which would lead to a decrease in light yield. Additionally, one would observe a decrease in the triplet molecule lifetime. Work by Himi et al. suggests that an impurity level of 0.5 atoms of nitrogen per $10^6$ atoms of argon in the liquid could decrease the observed triplet lifetime by as much as 0.1 µs [31]. Further experiments quantifying the reduction of the triplet lifetime due to nitrogen and oxygen impurities have recently been performed by the WARP collaboration [32, 33]. To avoid signal degradation, we continually circulate the argon through a getter before reliquefying back into the detector. We use daily measurements of both the light yield and the triplet lifetime to monitor the purity level.

We measure the light yield in the manner described earlier in this section, and we use the same $^{57}$Co data to measure the triplet lifetime. First, we select events in the 122-keV peak to make sure we use a similar data set for each individual measurement. The top and bottom PMT traces are normalized by the size of the single photoelectron and summed together. We align each pulse based on its estimated trigger time, defined as the time at which the trace first crosses 20% of its maximum value. At this trigger time, the relative time for each pulse is $t = 0$. 

### Number of photoelectrons

| Counts | 0 | 100 | 200 | 300 | 400 | 500 | 600 | 700 | 800 |
|--------|---|-----|-----|-----|-----|-----|-----|-----|-----|
| Cobalt data |  |  |  |  |  |  |  |  |  |
| Simulation |  |  |  |  |  |  |  |  |  |

#### Cobalt data

FIG. 4: (Color online) Example spectrum of $^{57}$Co data, along with a simulation done using the Reactor Analysis Tool [30].
Between 5000 and 10000 traces are averaged and the following model is fit to the average trace between 1 and 7 μs from the trigger:

\[ < V(t) > = A \exp(-t/\tau_l) + B, \]  

where \( < V(t) > \) is the expected trace, \( A \) is a normalization factor, \( \tau_l \) is the triplet lifetime and \( B \) is an additional baseline term that helps stabilize the fit over a range of fit windows. An example fit is shown in Fig. 5.

![Example fit of a single exponential to an average trace to measure the long time constant in liquid argon.](image)

We find two significant sources of systematic error in the fit parameters stemming from the voltage applied to the PMTs and the choice of fit window. Although we do not fully understand the PMT voltage effect, we estimate the systematic uncertainty to be 33 ns by changing the PMT voltages by ±75 V (equivalent to dividing or multiplying the gain by 2). We do not fit the data before 1 μs or after 9 μs, as the fit becomes less reliable due to contamination from the fast component and baseline noise, respectively. We estimate the systematic uncertainty associated with the fit window to be about 35 ns by both varying the end time by ±2 μs and by fitting the data within 5 μs windows ending at 6, 7 and 8 μs from the trigger. We combine the two sources of error into a single systematic uncertainty estimate of 50 ns.

During the four months of operation, we measure the long time constant and signal yield to be \( (1463 \pm 5_{\text{stat}} \pm 50_{\text{sys}}) \) ns and \( (4.85 \pm 0.08) \) photoelectrons/keVee. The uncertainty on the signal yield is statistical, and varying the PMT voltage by ±75 V has no apparent effect. For a fixed fit window and PMT voltage, the long time constant is stable to within 1%, while the signal yield is stable to within 5%.

### III. EXPERIMENTAL RESULTS

#### A. Detection Time PDF Model

We measure the time dependence of scintillation light produced by electronic and nuclear recoil scattering events in our detector. For an event with energy deposited at time \( t_0 \), we model the temporal probability density functions (PDFs) for the emission times of scintillation photons as the weighted sum or mixture of two exponential PDFs:

\[ f(t - t_0) = q g(t - t_0, \tau_l) + (1 - q) g(t - t_0, \tau_s), \]  

where

\[ g(t, \tau) = \frac{1}{\tau} \exp \left( -\frac{t}{\tau} \right), \]

\( \tau_l \) and \( \tau_s \) are the long and short time constants for both nuclear and electronic recoil event classes at any given energy, and the probability parameter \( q \) takes different values for the two classes of events.

At each PMT, detected scintillation photons yield photoelectrons that produce observed voltage traces. For the small detector in this experiment, scintillation transit times are negligible. We assume that the duration of energy deposition and excimer formation is instantaneous compared to the time scales relevant to scintillation light emission. Hence, we model the expected voltage trace as a convolution of the impulse response function of the PMT and the PDF model for the emission of scintillation photons:

\[ < V(t) > \propto \int_{s=0}^{\infty} h_V(t - s) f(s - t_s) ds, \]  

where \( h_V(t) \) is the impulse response function of the PMT and \( t_s \) is an additional model parameter that relates the energy deposit time \( t_0 \) to the relative time scale we associate with our measurement of \( h_V(t) \). We estimate the impulse response function by averaging single photoelectron events observed in our calibration data. Neglecting additive noise and other instrumental systematic errors, the integral of the voltage trace is proportional to the number of photoelectrons produced by the event.

The data are divided into 15 non-uniform bins by photoelectron number, with the smallest signal bin including events consisting of 20–24 photoelectrons and the largest bin including events consisting of 240–279 photoelectrons. These bins define the region of interest. For each photoelectron bin, we generate template traces for both electronic and nuclear recoils by averaging all “tagged” events of a given type. The traces are averaged in the same manner as described in Sec. IIIC and normalized.

We use these average voltage traces to determine the model parameters: \( \tau_l, \tau_s, q_{\text{nuclear}} \) and \( q_{\text{electronic}} \). The observed and predicted fraction of a normalized trace in the
ith time bin are called $p_m(i)$ and $\hat{p}(i)$, respectively. We obtain the model parameters by minimizing the squared Matusita distance \cite{34,35,36} between $p_m$ and $p$:

$$|p - p_m|_M = \sum_i (\sqrt{p_m(i)} - \sqrt{\hat{p}(i)})^2,$$  \hspace{1cm} (7)

where negative values of $p_m$ are set to 0. For each bin, we fit our model to normalized mean voltage trace data in a time window that ends about 6800 ns after the trigger time. We determine $t_*$ to be 30 ns before the trigger time, and vary the offset parameter chosen to minimize the value of Eq. (7).

Table II shows the estimated model parameters for each photoelectron bin. Figure 6 shows the model prediction along with data for the 80–99 photoelectron bin.

Between 80 and 300 ns, there is a feature in both event classes that is not well predicted by the model. A scintillation component that decays as approximately $t^{-1}$ has been observed in liquid helium, attributed to diffusion-dominated excimer-eximer destruction \cite{11}. Unfortunately, a model that includes a third exponential component neither returns stable model parameters as a function of energy nor accurately predicts the trace behavior between 80 and 300 ns, so we prefer the two-component model. We assume that the sharp bump localized at 150 ns is caused by the cabling and electronics.

| Bin (pe) | $\tau_1$ (ns) | $\tau_2$ (ns) | $q_{\text{nuclear}}$ | $q_{\text{electronic}}$ |
|---------|-------------|-------------|----------------|----------------|
| 20–24   | 1634 ± 150  | 9 ± 3       | 0.378 ± 0.011  | 0.523 ± 0.015  |
| 25–29   | 1535 ± 128  | 10 ± 3      | 0.382 ± 0.011  | 0.573 ± 0.015  |
| 30–34   | 1478 ± 107  | 10 ± 3      | 0.357 ± 0.010  | 0.601 ± 0.014  |
| 35–39   | 1455 ± 102  | 11 ± 3      | 0.353 ± 0.010  | 0.627 ± 0.014  |
| 40–44   | 1461 ± 96   | 12 ± 3      | 0.344 ± 0.010  | 0.658 ± 0.014  |
| 50–54   | 1459 ± 92   | 12 ± 3      | 0.327 ± 0.009  | 0.681 ± 0.015  |
| 60–66   | 1439 ± 89   | 12 ± 3      | 0.315 ± 0.010  | 0.699 ± 0.015  |
| 70–76   | 1448 ± 89   | 13 ± 3      | 0.309 ± 0.010  | 0.710 ± 0.015  |
| 80–99   | 1447 ± 85   | 13 ± 3      | 0.298 ± 0.010  | 0.721 ± 0.015  |
| 100–119 | 1452 ± 84   | 13 ± 3      | 0.289 ± 0.010  | 0.733 ± 0.015  |
| 120–139 | 1447 ± 84   | 13 ± 3      | 0.284 ± 0.011  | 0.741 ± 0.016  |
| 140–159 | 1446 ± 84   | 14 ± 3      | 0.278 ± 0.012  | 0.747 ± 0.016  |
| 160–199 | 1450 ± 84   | 14 ± 3      | 0.272 ± 0.013  | 0.752 ± 0.016  |
| 200–239 | 1460 ± 84   | 15 ± 3      | 0.265 ± 0.015  | 0.760 ± 0.016  |
| 240–279 | 1467 ± 84   | 15 ± 3      | 0.258 ± 0.018  | 0.764 ± 0.016  |

Table II: Estimated model parameters and 1-sigma uncertainties for each photoelectron bin. The systematic errors described in the text are the dominant source of error.

We determine 1-sigma random uncertainties for the model parameters with a nonparametric bootstrap resampling scheme \cite{37}. A rigorous quantification of systematic uncertainty arising from the fit window, the voltage applied to the PMT, and variation in the response of each individual PMT is difficult. Nonetheless, we get approximate estimates of systematic uncertainties from these sources, and these systematics are the dominant source of error. We estimate 1-sigma systematic uncertainties in the parameters $\tau_i$ (for $i = l, s$) as $0.5 \times (\tau_{i,\max} - \tau_{i,\min})$, where $\tau_{i,\max}$ and $\tau_{i,\min}$ are the maximum and minimum values taken by $\tau_i$ for different choices of time windows. We refit the model to data using time windows of approximately 5000 ns and 8600 ns from the trigger time, and vary the offset parameter $t_*$ by ± 2 ns. We also check for differences between the response of each PMT in five different photoelectron bins. We include the voltage-dependent errors described in Sec. 11 in the systematic uncertainty of $\tau_i$, and we estimate the voltage-dependent systematic variation of the $q$-values. We find that the time constants are most influenced by choice of fit window, while the $q$-values are most influenced by PMT effects. We combine all sources of error to obtain the estimates of systematic uncertainty shown in Table II.

As a consistency check on the quality of fit, for each photoelectron bin we use our model to predict the prompt fraction, a discrimination statistic described in detail in the next section. We compute the difference between

![FIG. 6: (Color online) Observed and predicted mean voltage traces for nuclear and electronic recoil events of 80 to 99 photoelectrons.](image-url)
the predicted and observed prompt fractions, and the root-mean-square values of this prediction error across all bins are respectively 0.007 and 0.003 for the nuclear and electronic recoil event classes. The fractional root-mean-square value of this prediction error is about 1\% for both event classes.

B. Prompt Fraction Method

The prompt fraction method is a simple approach to pulse shape discrimination. For each trace, we define the prompt fraction \( f_p \) as

\[
 f_p = \frac{\int_{T_0}^{T_f} V(t) \, dt}{\int_{T_0}^{T_f} V(t) \, dt},
\]

where \( V(t) \) is the voltage trace from the PMT, \( \xi \) is an integration time determined to optimize the ERC, \( T_0 = t_0 - 50 \text{ ns}, T_f = t_0 + 9 \mu s \), and \( t_0 \) is the trigger time as defined in Sec. II B. The measured discrimination does not significantly improve by extending \( T_f \) to 20 \( \mu s \). Fig. 7 shows a scatter plot of \( f_p \) versus energy for both electronic and nuclear recoils. The two populations of events represent the tagged data remaining in the neutron generator data set and the \( ^{22}\text{Na} \) data set after the selection cuts described in Sec. II B have been made. We choose \( \xi = 90 \text{ ns} \) by estimating the ERC based on a simple Gaussian model for values of \( \xi \) from 50 to 250 \text{ ns} over a variety of different photoelectron bins. Although the value of \( \xi \) has only a weak effect on the predicted discrimination, a choice of \( \xi = 90 \text{ ns} \) provides the best results across the widest range of energies in the region of interest, and that value is used for the analysis presented in this paper.

![FIG. 7](Color online) A scatter plot of \( f_p \) vs. energy for tagged electronic and nuclear recoils, where \( \xi = 90 \text{ ns} \).

For energy bins of width 1 keVee between 5 and 32 keVee, we form histograms of the electronic and nuclear recoil \( f_p \) statistics. To estimate the expected value of \( f_p \), \( \hat{f}_p \), we fit a Gaussian function to the empirical distributions. In Table III and Fig. 8 we present the estimated mean \( f_p \) for both classes of events in the energy range of interest. We estimate the systematic uncertainty on the values in Table III to be 3\%. This uncertainty estimate comes from changing the PMT voltage by \( \pm 75 \text{ V} \) and from variations in the measured signals between the two PMTs. The mean values of the \( f_p \) distributions for the two event classes are closer at low energies than at high energies, possibly because \( dE/dx \) for nuclear recoils decreases at low energies while increasing for electronic recoils. Therefore, the PSD improves at higher energies both because of increased photoelectron statistics and because of increased separation between the mean \( f_p \) values.

| Energy (keVee) | \( f_{p,\text{electronic}} \) | \( f_{p,\text{nuclear}} \) |
|---------------|-----------------------------|-----------------------------|
| 5–6           | 0.391 ± 0.012               | 0.566 ± 0.018               |
| 6–7           | 0.376 ± 0.011               | 0.595 ± 0.018               |
| 7–8           | 0.361 ± 0.011               | 0.607 ± 0.019               |
| 8–9           | 0.349 ± 0.011               | 0.625 ± 0.019               |
| 9–10          | 0.339 ± 0.010               | 0.638 ± 0.020               |
| 10–11         | 0.334 ± 0.010               | 0.640 ± 0.020               |
| 11–12         | 0.328 ± 0.010               | 0.649 ± 0.020               |
| 12–13         | 0.322 ± 0.010               | 0.663 ± 0.020               |
| 13–14         | 0.319 ± 0.010               | 0.658 ± 0.020               |
| 14–15         | 0.314 ± 0.009               | 0.675 ± 0.020               |
| 15–16         | 0.311 ± 0.009               | 0.683 ± 0.021               |
| 16–17         | 0.309 ± 0.009               | 0.678 ± 0.021               |
| 17–18         | 0.304 ± 0.009               | 0.685 ± 0.021               |
| 18–19         | 0.302 ± 0.009               | 0.682 ± 0.021               |
| 19–20         | 0.299 ± 0.009               | 0.684 ± 0.021               |
| 20–21         | 0.297 ± 0.009               | 0.690 ± 0.021               |
| 21–22         | 0.295 ± 0.009               | 0.695 ± 0.021               |
| 22–23         | 0.292 ± 0.009               | 0.699 ± 0.021               |
| 23–24         | 0.290 ± 0.009               | 0.690 ± 0.021               |
| 24–25         | 0.288 ± 0.009               | 0.688 ± 0.021               |
| 25–26         | 0.289 ± 0.009               | 0.695 ± 0.021               |
| 26–27         | 0.288 ± 0.009               | 0.696 ± 0.021               |
| 27–28         | 0.285 ± 0.009               | 0.696 ± 0.021               |
| 28–29         | 0.284 ± 0.009               | 0.701 ± 0.021               |
| 29–30         | 0.283 ± 0.009               | 0.708 ± 0.021               |
| 30–31         | 0.281 ± 0.009               | 0.701 ± 0.021               |
| 31–32         | 0.282 ± 0.009               | 0.689 ± 0.021               |

TABLE III: This table presents estimated mean values of \( f_p \) versus energy, where \( \xi = 90 \text{ ns} \). The main sources of uncertainty are systematic, stemming from voltage effects and differences in the measured signals between the two PMTs.

We estimate the ERC as the number of tagged electronic recoil events with \( f_p > f_{p,\text{nuclear}} \) divided by the total number of electronic recoil events, where \( f_{p,\text{nuclear}} \) is the estimated mean \( f_p \) for nuclear recoils of that energy. This restriction sets a nuclear recoil acceptance level of approximately 50\%. Since the shielding, coincidence, and timing cuts do not eliminate all neutron backgrounds in
Data analyzed with prompt fraction method

the detector, a background estimation \( N_{bg} \) is made by measuring the rate of background neutrons \( R_{bg} \) and assuming that the background is dominated by these neutrons hitting in accidental coincidence with \( \gamma \) rays \( (R_{\gamma}) \) in the liquid scintillator during the time allowed by the time-of-flight cut:

\[
N_{bg} = R_{bg} \times R_{\gamma} \times \text{TOF} \times T_a,
\]

where \( T_a \) is the acquisition time of the data. To enable comparison with the measured ERC, we divide \( N_{bg} \) for each energy bin by the total number of electronic recoil events in that bin.

Figure 8 shows the ERC observed using the prompt fraction method. We also plot the background estimation, two PSD projections based on the statistical model described below, and the ERC observed by applying a multibin method of PSD described in the next section. We convert the energy axis in Fig. 9 to keV of nuclear recoil energy (keVr) from keVee by dividing all electron equivalent energies by a constant nuclear recoil scintillation efficiency of 0.29. This value was obtained by measurements using the same apparatus described in this paper and will be discussed in an upcoming publication [28]. We present the PSD results in keVr because that is the unit of interest for a dark matter detector. Using the prompt fraction method, for a nuclear recoil acceptance level of approximately 50%, we measure a background- and statistics-limited level of ERC in our detector of \( 8.5 \times 10^{-6} \) between 52 and 110 keVr (11 contamination events). We observe no nuclear recoil-like events above 69 keVr. For comparison, there is an uncorrelated neutron background rate of \( \sim 6 \) mHz between 69 and 110 keVr, corresponding to 0.25 expected background counts in that energy range.

Following work done by members of the DEAP collaboration [29, 30], we model our estimates of the number of photoelectrons in the prompt and late time windows, \( N_p \) and \( N_l \), as normally distributed, independent random variables with means \( \mu_p \) and \( \mu_l \) and variances \( \sigma_p^2 \) and \( \sigma_l^2 \). The estimated total number of photoelectrons, \( N_{tot} = N_p + N_l \), is also a random variable, with mean \( \mu_{tot} = \mu_p + \mu_l \) and variance \( \sigma_{tot}^2 = \sigma_p^2 + \sigma_l^2 \). We express \( \mu_p \) and \( \mu_l \) in terms of \( \mu_{tot} \) and \( \tilde{f}_p \) and we decompose the variances into two components:

\[
\begin{align*}
\mu_p &= \tilde{f}_p \mu_{tot} \\
\mu_l &= (1 - \tilde{f}_p) \mu_{tot} \\
\sigma_p^2 &= \mu_p + \sigma_{p,add}^2 \\
\sigma_l^2 &= \mu_l + \sigma_{l,add}^2
\end{align*}
\]

where \( \sigma_{p,add} \) and \( \sigma_{l,add} \) represent additional sources of random variability beyond what we expect from Poisson counting statistics (for example, integration noise).

Hinkley [31] has described in detail the probability density function of the ratio of two normally distributed, correlated random variables. For simplicity, we present here an approximation [given by Eq. 9 of Ref. [31]] to the PDF of \( f_p = N_p / N_{tot} \):

\[
g_{f_p}(x) = \frac{\sigma_p^2 \mu_p x + \sigma_l^2 \mu_l (1-x)}{\sqrt{2\pi(\sigma_p^2 x^2 + \sigma_l^2 (1-x)^2)^3/2}} \times \exp\left[\frac{-(\mu_p x - \mu_l (1-x))^2}{2(\sigma_p^2 x^2 + \sigma_l^2 (1-x)^2)}\right].
\]

where we have used the fact that the correlation, \( \rho \), between \( N_p \) and \( N_{tot} \) is

\[
\rho = \frac{\sigma_p}{\sqrt{\sigma_p^2 + \sigma_l^2}}.
\]
For the analysis below, we use the exact PDF [Eq. 1 in Ref. 41], although in practice the approximation is extremely good down to the lowest energy bin examined.

We fit the electronic recoil data with the statistical model in each energy bin by fixing \( N_{\text{tot}} \) according to our measured light yield and treating \( f_p \), \( \sigma_{p,\text{add}} \), and \( \sigma_{l,\text{add}} \) as free parameters. We assume that the statistical distribution of \( f_p \) does not strongly depend on the value of \( N_{\text{tot}} \) for events of the same energy. For any particular energy deposit, \( N_{\text{tot}} \) is a random variable, and events due to many different energy deposits can contribute to prompt ratio data in any one bin in \( N_{\text{tot}} \) space. The probability density function for prompt ratio data is a mixture of energy dependent PDFs, and we neglect this energy blurring effect. Simple Monte Carlo studies suggest that the ratio-of-Gaussians model breaks down for idealized Gaussian data due to the constraint on \( N_{\text{tot}} \) resulting from the binning of data, and this effect has not been taken into account in our analysis.

Here, we use the statistical model to estimate the expected fraction of electronic recoils that are misclassified as nuclear recoils:

\[
\text{ERC} = \int_{\eta}^{1} g_{f_p}(x) \, dx. \tag{13}
\]

Here, we choose \( \eta = f_{p,\text{nuclear}} \) to set the nuclear recoil acceptance level to approximately 50\%, and we choose the parameters of \( g_{f_p} \) according to the fits to the electronic recoil \( f_p \) distribution. Fig. 3 shows the predicted ERC versus energy according to this model. We also plot the idealized case where \( \sigma_{l,\text{add}} \) and \( \sigma_{p,\text{add}} \) are set to 0. In general, we expect that the normal distribution model for \( N_p \) and \( N_l \) is an approximation for any energy of interest. For the idealized case where \( \sigma_{p,\text{add}} = \sigma_{l,\text{add}} = 0 \), \( N_p \) and \( N_l \) would be Poisson random variables rather than Gaussian random variables. Hence, the ERC predicted by the ratio-of-Gaussians model for the idealized case should be interpreted with caution particularly at lower energies where the accuracy of a normal distribution model for a Poisson random variable can be very poor.

Figure 10 shows an example of the model fit for 14–15 keVee and 30–31 keVee electronic recoil events. There is a deviation from the model at low \( f_p \) values that we attribute to pile-up and noise triggers. There is also an excess of events in the high \( f_p \) region. This excess might be caused by edge effects in our detector coming from \( \gamma \) tracks going into the walls or the TPB layer, producing extra prompt light. A larger detector with position reconstruction capability might be able to eliminate such edge effects. There could be some unknown phenomena at work in the production and decay of argon molecular states, yielding a small fraction of events with anomalously large \( f_p \) values. A third possibility is approximation error stemming from incorrect model assumptions, such as the Gaussianity of \( N_p \) and \( N_l \) or the effect of data binning. These possible effects can be better investigated by detectors with improved neutron shielding, and future studies will require more work to better calibrate the ERC predictions made by the ratio-of-Gaussians model.
C. Multibin Method

The prompt fraction method of PSD is based on binning the voltage trace into two time bins. We generalize this approach by representing a normalized voltage trace as a $K \times L$ dimensional matrix. In this representation, we categorize the data by the number of photoelectrons in the event, and $L$ refers to this division of data into photoelectron bins. We also partition the voltage trace into $K$ time bins. We choose $K = 10$ for ease of computation, but there may be a better choice of $K$. With the exception of the four smallest signal bins, which have been combined to form bins of 20–29 and 30–39 photoelectrons, we use the same average voltage traces obtained in Sec. III A as templates.

We partition each template into $K = 10$ time bins. The upper and lower endpoints of each time bin are selected so that the fractions of the template trace for 80–99 photoelectron electronic recoil events that falls in each time bin are approximately equal. The initial bin starts 50 ns before the trigger, and the endpoints of each bin are as follows, measured in nanoseconds from the trigger: 8, 18, 56, 200, 440, 750, 1180, 1800, 2950, and 8000. We do not adjust these endpoints for different photoelectron number but use the 80–99 photoelectron based time binning scheme for all cases.

The $k$th component of the normalized template for an event that falls in the $l$th photoelectron bin for the nuclear and electronic recoil classes is denoted $p_n(k, l)$ or $p_e(k, l)$, respectively. For instance, $p_n(1, 1)$ represents the fraction of the 20–29 photoelectron nuclear recoil template trace that falls in the first time bin (between 50 ns before the trigger and 8 ns after the trigger).

To assign an event to either the nuclear recoil or electronic recoil class, we first compute a discrimination statistic based on analysis of an idealized experiment in which we could observe the absolute detection time of each photoelectron without error. In this ideal case, for a fixed number of detected photoelectrons, the observed number of photoelectrons in the time bins is a multinomial random variable.

Given that the fraction of detected photoelectrons in the $k$th bin is $p_m(k, l)$, the multinomial log-likelihood statistics for the nuclear and electronic recoil classes are

$$
\ln Y_n = N_{\text{tot}} \sum_{k=1}^{K} \sum_{l=1}^{L} \delta_{ll'} p_m(k, l) \ln p_n(k, l') + \text{const} \quad (14)
$$

and

$$
\ln Y_e = N_{\text{tot}} \sum_{k=1}^{K} \sum_{l=1}^{L} \delta_{ll'} p_m(k, l) \ln p_e(k, l') + \text{const}, \quad (15)
$$

respectively [42]. Here, $N_{\text{tot}}$ is the total number of detected photoelectrons, and $p_n(k, l)$ and $p_e(k, l)$ are the expected values of the fraction of detected photoelectrons in the $k$th time bin for the nuclear and electronic recoil classes, respectively. For this idealized case, a natural choice for the discrimination statistic is the log-likelihood ratio statistic, $\ln R_m$:

$$
\ln R_m = \ln Y_e - \ln Y_n. \quad (16)
$$

In our experiment, the observed data is not multinomial, primarily because we observe a noisy voltage waveform rather than discrete detection times. Nonetheless, we compute a discrimination statistic using Eqs. (14) (15) where we estimate $N_{\text{tot}}$ and $p_m(k, l)$ from any voltage trace of interest, and determine $p_n(k, l)$ and $p_e(k, l)$ as described earlier.

Figure 12 shows a scatter plot of $\ln R_m$ versus energy, analogous to Fig. 7. From this point the analysis parallels the prompt fraction method, as we form histograms of $\ln R_m$ by energy bin and fit a Gaussian function to the observed $\ln R_m$ statistics to estimate the mean of $\ln R_m$. Fig. 13 shows an example of the fitted projections for 14–15 keV ee and 30–31 keV ee events.

For each energy bin, we estimate the mean values of $\ln R_m$ for nuclear recoils based on the Gaussian fits to the observed distributions, and this estimated mean value determines an approximate 50% nuclear acceptance threshold. We then determine the fraction of events in the tagged electronic recoil data set that have discrimination statistics less than this mean value to determine the observed level of ERC using the multi-bin method. If we determine the 50% nuclear acceptance threshold by finding the median values of the nuclear recoil distributions, the observed ERC is not significantly affected. Figure 14 in Sec. III B shows the ERC using the multibin method. The multibin method outperforms the prompt fraction method by as much as an order of magnitude. For a nuclear recoil acceptance of approximately 50%, we have

![Figure 11](color online) Estimated $\sigma^2$ and $\sigma^2_e$ parameters from the statistical model plotted against the estimated number of photoelectrons in the late and prompt components, respectively. Each distribution is fitted by a straight line, and the ideal case where $\sigma^2 = \mu$ is also shown.
FIG. 12: (Color online) A scatter plot of $\ln R_m$ vs. energy for both electronic and nuclear recoils.

FIG. 13: (Color online) Projections of Fig. 12 onto the y-axis for 14–15 keVee (top) and 30–31 keVee (bottom) events, with Gaussian fits to both the electronic (left) and nuclear (right) recoil distributions.

measured a background- and statistics-limited ERC of $7.6 \times 10^{-7}$ between 52 and 110 keVr (1 contamination event). We observe no contamination events above 62 keVr using the multibin method.

IV. MAXIMUM LIKELIHOOD PSD

We predict the PSD achievable in a large detector with many PMTs by assuming we can measure discrete times for each photoelectron detected in an event. We simulate events in this detector using the PDFs measured in Sec. III A and assume that the detection times of photoelectrons are measured without error. We develop a maximum likelihood PSD method and apply it to the simulated data.

For an event that generates $N$ photoelectrons, we denote the detection times as $t = (t_1, t_2, ..., t_N)$. We define the log-likelihood function, $\ln L$, of this data following Ref. [43] as

$$\ln L = -m + \sum_{i=1}^{N} \ln(r_{bg} + \lambda f(t - t_0)),$$

(17)

where

$$m = \int_{T_b}^{T_e} (r_{bg} + \lambda f(t - t_0))dt,$$

(18)

t_0 is the time at which the energy deposit occurs, $f(t-t_0)$ is the PDF for the observed photoelectrons [Eq. 4], $\lambda$ is the expected number of detected photoelectrons generated by the event, $T_b$ and $T_e$ are the start and end time of the observation, and $r_{bg}$ is the background rate.

In our simulations, we set $r_{bg} = 0$ and $t_0 > T_b$. The maximum likelihood estimates of $t_0$ and $\lambda$ are therefore $t_1$, the first detection time, and $\hat{\lambda}$, where

$$\hat{\lambda} = \frac{N}{qF(T_e - t_1, \tau_1) + (1-q)F(T_e - t_1, \tau_2)}$$

(19)

and $F(T, \tau) = 1 - \exp(-T/\tau)$. Consequently, $m = N$. Additionally, the time window in our simulations is 40 $\mu$s, so $\int_{T_b}^{T_e} f(t-t_0)dt \approx 1$. Thus, for our case the likelihood function of the observed data is well approximated as

$$L(t) \approx \exp(-N) N^N \prod_{i=1}^{N} f(t_i - t_1).$$

(20)

Following Ref. [13] and the discussion above, for each simulated event we determine $L(t)$ for both the electronic and nuclear recoil event classes as modeled in Sec. III A. These values are denoted $L_e(t)$ and $L_n(t)$. We define the log-likelihood ratio, $\ln R$:

$$\ln R = \ln L_e(t) - \ln L_n(t).$$

(21)

Figure 14 shows this log-likelihood statistic for simulated electronic and nuclear recoil events that yield 50 photoelectrons. The event is assigned to the nuclear recoil class if $\ln R$ is less than an adjustable threshold that can be varied to increase the discrimination against electronic recoils at the cost of decreasing nuclear recoil acceptance. The Monte Carlo estimate of the distribution of $\ln R$ has
prominent ripples even though the Monte Carlo estimates of \( L_n \) and \( L_e \) do not. In our Monte Carlo study, we simulate events that yield a fixed number of photoelectrons according to PDFs with the assumption of perfect knowledge. Furthermore, we neglect dark current noise. In an actual experiment, we expect that imperfect energy resolution due to variability in counting statistics and dark current effects would attenuate the ripples in the \( \ln R \) distribution.

To illustrate the maximum likelihood method, we simulate a detector with a signal yield of 6 photoelectrons/keVee, as might be possible in a detector with full PMT coverage. We neglect dark current and we set the discrimination threshold to accept 50% of nuclear recoils. For comparison, we simulate prompt fraction data by assuming the number of prompt photoelectrons is a binomial random variable with an expected value determined by the measured PDFs, and the total number of photoelectrons is assumed to be known without error. This idealized binomial model predicts a much lower ERC than the statistical model discussed in Sec. III B.

The maximum likelihood discrimination method outperforms the idealized prompt fraction method, as shown in Fig. 13. We also expect that the maximum likelihood method will be more robust than the prompt fraction method to background noise. For the prompt fraction case, we select a threshold in prompt photoelectron space to yield a nuclear recoil acceptance probability as close to 0.5 as possible. Due to quantization effects, the actual nuclear recoil acceptance probability varies about 0.5, and there are sawtooth-like artifacts in the prompt fraction ERC curve shown in Fig. 15.

V. CONCLUSION

Using a detector with a signal yield of 4.85 photoelectrons/keVee, we measured the scintillation time dependence of electronic and nuclear recoils in liquid argon down to 5 keVee or 20 keVr. We developed a prompt fraction method of PSD in liquid argon, and for a nuclear recoil acceptance level of 50%, we measured a background- and statistics-limited level of ERC to be \( 8.5 \times 10^{-6} \) between 52 and 110 keVr with no contamination events above 69 keVr. We also developed a multibin method of PSD, improving on the prompt fraction method by as much as an order of magnitude. With this method, we measured a background- and statistics-limited level of ERC of \( 7.6 \times 10^{-7} \) between 52 and 110 keVr for the same nuclear recoil acceptance of 50%. We modeled the observed prompt fraction data as the ratio of two normally distributed, correlated random variables, where we assumed \( N_p \) and \( N_l \) were uncorrelated; we discussed discrepancies between observed and predicted prompt fraction results. Finally, we developed a maximum likelihood method of PSD for a detector capable of measuring a discrete detection time for each observed photoelectron in an event.

Acknowledgments

We thank M. Boulay, C. Jillings, B. Cai, and J. Lidgard for useful discussions and for developing the ratio-of-Gaussians model used in this analysis. We acknowledge S. Seibert and J. Klein for their contributions to the Monte Carlo simulations software package. This work was supported by the David and Lucille Packard Foundation, the Los Alamos Directed Research and Development Program, and the U.S. Department of Energy.
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