Modeling ionization and recombination from low energy nuclear recoils in liquid argon

M. Foxx^{a,b,c}, C. Hagmann^b, I. Jovanovic^a, A. Bernstein^b, T.H. Joshi^{d,b,e}, K. Kazkaz^b, V. Mozin^b, S.V. Pereverzev^b, S. Sangiorgio^b, P. Sorensen^b

^aDepartment of Mechanical and Nuclear Engineering, The Pennsylvania State University, University Park, PA 16802, USA

^bLawrence Livermore National Laboratory, Livermore, CA 94550, USA

^cPacific Northwest National Laboratory, Richland, WA 99352, USA

^dDepartment of Nuclear Engineering, University of California, Berkeley, CA 94720, USA

^ePresent address: Lawrence Berkeley National Laboratory, 1 Cyclotron Road, Berkeley, CA 94720, USA

Abstract

Coherent elastic neutrino-nucleus scattering (CENNS) is an as-yet undetected, flavor-independent neutrino interaction predicted by the Standard Model. Detection of CENNS could offer benefits for detection of supernova and solar neutrinos in astrophysics, or for detection of antineutrinos for nuclear reactor monitoring and nuclear nonproliferation. One challenge with detecting CENNS is the low energy deposition associated with a typical CENNS nuclear recoil. In addition, nuclear recoils result in lower ionization yields than those produced by electron recoils of the same energy. While a measurement of the nuclear recoil ionization yield in liquid argon in the keV energy range has been recently reported, a corresponding model for low-energy ionization yield in liquid argon does not exist. For this reason, a Monte Carlo simulation has been developed to predict the ionization yield at sub-10 keV energies. The model consists of two distinct components: (1) simulation of the atomic collision cascade with production of ionization, and (2) the thermalization and drift of ionization electrons in an applied electric field including local recombination. As an application of our results we report updated estimates of detectable ionization in liquid argon from CENNS at a nuclear reactor.

Keywords: liquid argon, ionization yield, neutrino, dark matter

1. Introduction

Dual-phase noble element detectors are extensively used in the search for dark matter [1, 2, 3, 4, 5, 6, 7], but they are also of great interest in the quest to detect coherent elastic neutrino-nucleus scatter (CENNS) [8, 9]. The reasons for their extensive use include the scalability, high sensitivity [10, 11], favorable electron drift properties [12, 13], and relatively low cost for a dual-phase xenon (Xe) or argon (Ar) detector. It is important to understand that we are nearing the absolute limits of sensitivity of those detectors, i.e. detection of very few electron-ion pairs and photons produced by primary interaction. In addition, recoiling nuclei (or ions) transfer a large fraction of their energy to heat, in contrast to ionization by electrons, where most of the energy is expended on ionization and photon production. Therefore, an understanding of physical processes involved in

detector operation at low energies is becoming increasingly important.

A variety of models have been used in the past in an effort to predict the ionization yield (defined as the number of electrons produced per energy deposited) at low energies (keV) in liquid noble element detectors. The Lindhard theory [14, 15] is successful in predicting the ionization yield at higher energies (MeV), but requires an independent knowledge of the ratio of energies expended to ionization and scintillation at lower energies [16], and an additional semi-empirical proportionality factor is needed to reconcile it with the measurements. Other semi-empirical models have been constructed, aimed at predicting the nuclear quench factors and scintillation yields in a variety of scintillator materials including noble liquids [17, 18, 19]. While capable of producing good fits to the experimental detector response data, those models ignore specific reconstruction of the spatial structure of ionization tracks at low ener-

Email address: Michael.Foxx@pnnl.gov (M. Foxx)

gies.

An atomistic Monte Carlo model has been previously developed to predict the nuclear quench factor in liquid argon (LAr) [8] at energies relevant for CENNS experiments conducted using antineutrinos produced by nuclear reactors. In this current work, we refine the atomistic Monte Carlo model [8] by including Ar^+ cross sections and the effects of local recombination in an applied drift field [20]. Though we do not estimate final uncertainties on quantitative predictions, our modeling accurately describes qualitative features of the ionization tracks at low energies which need to be taken into account in existing empirical models.

In addition to the expected ionization yields for nuclear recoil energies up to ~ 10 keVr, we present a revised prediction for the CENNS ionization yields produced from reactor antineutrinos.

2. Primary Ionization Yield Using Atomic Collision Monte Carlo Simulation

The algorithm used in the TRansport of Ions in Matter (TRIM) [21]-based binary collision Monte Carlo model has been described in previous work [8]. It handles the elastic Coulomb deflections of the incident fast Ar atom in the medium during slowing down. For the energy range considered here, most of the energy in the slowing down process is transferred to other atoms in elastic Coulomb collisions, thus dominating the stopping power $-dE/dx$. The remainder of the energy is transferred through inelastic collisions.

In this work we have included the ionization and excitation of a neutral Ar atom by another Ar atom or Ar^+ ion and charge exchange between a fast Ar^+ ion and a neutral background Ar atom. Figures 1 and 2 show the inelastic cross sections compiled by Phelps [22] for incident Ar and Ar^+ projectiles, respectively. All ionization and excitation cross sections are small compared to the geometric cross section $\sigma_{geo} = n^{-2/3}$ for elastic Coulomb deflections, where n is the Ar number density [8, 21].

The excitation producing 811 and 795 nm photons is ignored for this simulation. The 811 nm and 795 nm cross sections are 10% and 2.5% of the ionization/UV cross section, respectively [22]. For a 10 keV recoil, it is estimated that <100 eV would be lost to the production of 811 and 795 nm photons.

We take the probability for ionization occurring in a Coulomb collision as $p_{ion} = \sigma_{ion}/\sigma_{geo}$, where σ_{ion} is the ionization cross section; an analogous expression is used for excitation. In every Coulomb collision the inelastic channels are sampled based on those

probabilities and are uncorrelated with the scattering angle. One of the outgoing particles is randomly chosen to be Ar^+ for ionization events and similarly Ar^* for excitations. Due to the large charge transfer cross section, σ_{CT} , few Ar^+ atoms survive to the next collisions event, with a survival probability estimated as $p_{sur} = \exp(-\sigma_{CT}/\sigma_{geo})$. The leading particle plus all secondaries with kinetic energies larger than the excitation or ionization threshold (~ 25 eV in the laboratory frame) are tracked as a part of the collision cascade.

Other approximations made include: (1) the inelastic cross sections of a fast Ar^* atom are taken to be the same as an Ar atom in the ground state due to the lack of data, and (2) the target atom in a collision is assumed to be a neutral Ar in the ground state. The additional energy lost in an inelastic collision is taken from the more energetic outgoing particle to eliminate the possibility of a particle acquiring negative energy.

Unfortunately, the experimental uncertainties of the cross sections used in this simulation are not known. Also, while the TRIM algorithm is often used for problems similar to the one considered here, we are unaware of a solid treatment in the literature of systematic uncertainties arising from neglecting three-body collisions in modeling ionization dynamics in liquid.

Figure 1: In a given neutral-neutral ($\text{Ar} + \text{Ar}$) collision, elastic scattering, ionization, or excitation can occur. Shown are the cross sections for ionization (red), $\text{Ar} + \text{Ar} \rightarrow \text{Ar} + \text{Ar}^+ + e^-$, and excitation (blue), $\text{Ar} + \text{Ar} \rightarrow \text{Ar} + \text{Ar}^*$, based on tabulated values in Reference [22]. The cross sections are nearly equal, with excitation cross section being slightly higher than ionization cross section, which results in similar rates of primary excitation and ionization.

The ionization electron spectrum is not well known and evidence exists that it depends on the collision energy [23]. For our studies of electron drift and recombination, we chose two discrete electron energies, $E_{elec} = 3$ and 10 eV, based on the electron spectrum for

Figure 2: Cross sections for charge transfer (black), $\text{Ar}^+ + \text{Ar} \rightarrow \text{Ar} + \text{Ar}^+$, ionization (magenta), $\text{Ar}^+ + \text{Ar} \rightarrow \text{Ar}^+ + \text{Ar}^+ + e^-$, and excitation (green), $\text{Ar}^+ + \text{Ar} \rightarrow \text{Ar}^+ + \text{Ar}^*$, based on tabulated values in Reference [22].

Table 1: Material specific properties of LAr used in the atomic collision model.

Property	Variable	Value	Units
Atom Density	n	2.11×10^{22}	cm^{-3}
Coll. Path Length	L_{geo}	3.62	$\text{\AA}$
Geo. Cross Section	σ_{geo}	1.31×10^{-15}	cm^2
Ionization Potential	I_p	14.3	eV
Excitation Energy	E_{exc}	12	eV
Ion. Elec. Energy	E_{elec}	3,10	eV

Ar-Ar collisions in Ref [23]. Material-specific input parameters taken into account in the simulation are listed in Table 1, and a summary of all treated interactions is provided in Table 2.

The range of Ar atoms with keV energies in LAr, shown in Figure 3, is short compared to the range of electrons of the same energy. The Ar ion recoil is simulated in this work, while electron recoils have been previously simulated in [20].

The primary ionization yields were obtained by averaging 10000 collision histories for each incident energy. The positions of the Ar ionization sites were recorded for the recombination stage of the simulation. Figure 4 shows the average number of produced ionization electrons as well as their dispersion which affects energy resolution. The ionization yield is only slightly ($\sim 2\%$) larger than the ionization yield obtained perviously [8] by tracking only neutral Ar atoms.

The primary scintillation and ionization yields in our current model are nearly identical, because ionization

Table 2: A summary of the possible interactions between neutral and charged Ar atoms as they slow down. Here, I_p is the ionization potential, E_{elec} is the ejected electron energy, and E_{exc} is the atomic excitation energy. ‘COM’ is center of mass.

Particle	Target	Collision Type	COM E Loss
Ar	Ar	Elastic	0
Ar	Ar	Ionization	$E_{elec} + I_p$
Ar	Ar	Excitation	E_{exc}
Ar^+	Ar	Charge Transfer	0
Ar^+	Ar	Ionization	$E_{elec} + I_p$
Ar^+	Ar	Excitation	E_{exc}

Figure 3: The range of the Ar atoms in LAr (red) is on the order of nm, which is much shorter than the range of electrons of the same energy (blue) [20]. Both ranges come from the respective simulations. The ionization electron thermalization length of $2.6 \mu\text{m}$ (green-dashed) is very weakly dependent on the secondary electron energy in the range of 1-5 eV [24].

and excitation collisions cross sections for Ar-Ar collisions are very nearly the same (Figure 1). The difference between inelastic cross sections for Ar and Ar^+ (Figures 1 and 2) becomes unimportant for the accuracy of the simulation due to the high probability of charge transfer. On the other hand, Figure 3 indicates that distances between exited Ar atoms of the compact ‘positive core’ became small and comparable to inter-atomic distances. At these conditions one should expect effective pair-recombination of excited states due to Penning ionization, without requiring direct collision of exited atoms, because electron tunneling is involved in the Penning process: $\text{Ar}^* + \text{Ar}^* \rightarrow \text{Ar}^+ + e^- + \text{Ar}$. As a result, at low recoil energies, ionization yield should be somewhat higher at the expense of primary scintillation. Unfortunately, data on Penning ionization in LAr is not available, in contrast to liquid He, where this pro-

Figure 4: The predicted electron yield including the Ar^+ collision cross sections and $E_{elec}=3$ eV (red) and dispersion (brown error band) for the recoiling Ar atoms. For comparison, the electron yield without the Ar^+ collision cross sections [8] is approximately 1 electron fewer ($\sim 2\%$) at a recoil energy of 10 keV.

Figure 5: Calculated number of collected electrons as a function of drift field for variable number of initial ionizations for $E_{elec}=3$ eV. The electron escape efficiency is near unity for all fields and $N < \sim 10$. For larger N , the Coulomb attraction between electrons and the dense core of positive ions becomes important resulting in substantial recombination. Little difference is observed when an ionization electron emission energy of 10 eV is used.

cess has been extensively studied both theoretically [25] and experimentally [26].

3. The Model of Electron Recombination and Electron Separation Efficiency

In addition to calculating how many electrons are created in a given recoil, the fraction of electrons which escape recombination also needs to be calculated. The simulation of the electron drift is based on the Cohen-Lekner theory [27], as described by Wojcik [28]. The method for simulating electron drift and recombination for incident electron energies up to 10 keV has been presented in [20]. The position of the secondary electrons is obtained from the atomistic Monte Carlo, and the subsequent transport is performed using the framework described in Ref [20].

Tallying the number of electrons which survive local recombination after thermalization and drift in the externally applied field yields the electron separation efficiency ('electron extraction efficiency' is usually used to describe the process of extraction of electrons from the liquid into the gas phase in dual-phase detectors). This calculation is performed for various electric fields and for two representative electron emission energies from the Ar atom: 3 eV and 10 eV [23], Figure 5. Little difference is seen in the charge separation efficiency for different electron emission energies. This is because the electron thermalization length is only weakly dependent on initial electron energies for the large (>1 eV) electron energies; calculations for the range of 1-5 eV were performed in Ref. [24]. Thus, we can disregard the

real distribution of emitted electron energies for the energy range 1-10 eV. All the electrons should thermalize away from the dense positively charged core of the track; the size of this core is given by the range of Ar recoils in Figure 3. As a result, a small number of positive ions in the core require only a weak external field to prevent recombination. In contrast, ionization electrons produced with very low energies, 'near zero energy electrons', should thermalize in close proximity to positive core and a larger electric field will be required to separate them from the core. Ionization electron energies below 1 eV were inaccessible in the experiment performed in Ref. [23], and the relative value of 'near zero energy electrons' is unknown.

As the number of produced electron-ion pairs increases ($\gtrsim 10$), even for 'distant' (long thermalization length) electrons, Coulomb attraction to positively charge core becomes comparable to the force of the external electric field, which tends to increase trapping and recombination. With typical drift fields ~ 1 kV/cm, however, the recombination effect is negligible over the range of recoil energies studied.

The dependence of the escape efficiency as a function of drift field is very similar for Ar and electron recoils [20] for the same number of released electrons. The reason for this are the short track lengths of both projectiles compared to the thermalization distance of the ionization electrons, as shown in Figure 3.

Figure 6: Ionization yield curve for LAr below 10 keV before (red) and after (blue) recombination, as calculated by the Monte Carlo simulation, for 10 eV ionization electrons in an electric field of 1 kV/cm.

Figure 7: The number of collected electrons shown in Figure 5 in terms of nuclear recoil energy. The green data points are the 6.7 keV nuclear recoil data measured in Ref [29]. The errors on the measured data are too large to distinguish the two ionization yield scenarios shown in Figure 6 (with versus without recombination).

4. Comparison with Experimental Data

To predict the detectable ionization yield from nuclear recoils, the primary ionization yield is combined with the electron separation efficiency. The atomic collision model is used to perform calculations at a given energy, and the number of ions produced and their respective positions in each trial are used as an input to the electron recombination model. The resulting number of electrons which do not recombine with initial ion cloud is then used to calculate the drift field dependent ionization yield. For recoil energies below ~ 1 keV (~ 4 electrons), electron recombination does not have a significant effect on the ionization yield for drift fields > 0.1 kV/cm. For larger energies, recombination becomes significant and results in a reduction in the detectable electron yield, as shown in Figure 6. The primary scintillation and ionization yields are nearly identical, as both of them are dominated by the neutral Ar-Ar cross sections (shown in Figure 1).

A comparison of the collected number of electrons from the model with those measured for 6.7 keV nuclear recoils in Ref [29] is shown in Figure 7. For currently available data and attained experimental accuracy, the model reproduces both the magnitude and trend of the experimental data points. Moreover, taking into account Penning ionization should bring the ‘field dependence saturation’ value to a larger total charge, while the existence of ‘near zero energy electrons’ in ionization electron energy distribution should increase the ‘saturation field’ to a higher value. The fraction of ‘near zero energy electrons’ is unknown due to experimental thresholds in Ref [23]. We do not introduce additional fitting parameters to account for the mentioned effects. To im-

prove the model predicting power, Penning processes cross sections and ionization electrons energy distribution need to be measured in an independent experiment or theoretically estimated.

5. Ionization Yield from CENNS at a reactor

Let us consider the CENNS signal resulting from interactions with antineutrinos produced in a nuclear reactor. The antineutrino spectrum emitted from a nuclear reactor depends on the relative contributions of the fissioning nuclides to fission power, since the antineutrino spectrum produced in fission varies with the parent nucleus [30, 31]. In the remainder of this discussion, we assume a 3 gigawatt thermal (GWth) reactor at a 25-m standoff (antineutrino flux is approximately $6 \times 10^{12} \text{ cm}^{-2}\text{s}^{-1}$), and with the same isotopic content as used in Ref. [8].

The net spectrum of detectable electrons is obtained by folding the expected Ar recoil spectrum [8] with the ionization yield and the ionization electron separation efficiency. For our estimate we assumed $E_{elec}=10$ eV and a drift field of 1.0 kV/cm. Most of the recoils ($\sim 70\%$) produce no detectable ionization, with the remaining events yielding up to ~ 5 electrons. The total expected CENNS rate of both ionizing and non-ionizing events is 56 events per kg of Ar per day [8]. Figure 8 shows the updated prediction of the CENNS event rates, along with internal backgrounds.

The primary internal background is ^{39}Ar , which decays via β -decay with an end-point energy of 565 keV and a half-life of 269 years. It has been measured

that the activity of ^{39}Ar is ~ 1 Bq/kg in atmospheric Ar [32, 33, 34]. Assuming the beta spectrum given in Ref. [35] and a W-value of 23.6 eV, we expect the spectral contribution of ^{39}Ar to be flat in the low-energy (“few electron events”) region with an amplitude of ~ 4.1 events $\cdot(e^-)^{-1}\text{kg}^{-1}\text{day}^{-1}$, see Figure 8. Efforts have been made to collect Ar from exhaust streams of underground mining facilities, which are depleted in ^{39}Ar by $\sim 150\times$ that of atmospheric Ar [36]. With an assumed depletion factor of 150, the background ^{39}Ar rate drops to just 0.027 events $\cdot(e^-)^{-1}\text{kg}^{-1}\text{day}^{-1}$, $\sim 100\times$ lower than the predicted CENNS rates. Due to the low-energy region of interest, there is not enough primary scintillation produced to further reduce the electronic recoil background through pulse shape discrimination.

Both Ar and Xe dual-phase detectors demonstrate detector-dependent low-energy background at low part of the energy spectra (events where 1-10 electrons were detected), and the number of these events increases with decreasing energy [9, 37]. Single-electron events can be related to trapping of electrons on liquid-gas boundary after “previous” events and subsequent “delayed evaporation”. This effect should be stronger for Xe detectors, because LAr has lower electron work function than liquid Xe (0.21 and 0.61 eV respectively [38]). Another mechanism responsible for delayed electrons and photon production could be associated with trapping of positive ions in thin physisorbed solid Ar (Xe) layers on the surfaces of metal electrodes and their subsequent delayed recombination. This question requires additional detailed investigation and is well beyond the scope of the present paper.

6. Discussion

Interpretation of our results is strongly influenced by the sensitivity we are demanding of current and future noble element detectors. We would like to point out that the basic physics behind both charge and light yield at low energies has never been fully explored with respect to the relevant atomic and condensed matter physics. This gap in understanding has not prevented obtaining robust results from noble element detectors, but only because previous experiments have limited their analysis to higher energies where the uncertainty of a single quantum of excitation was overshadowed by larger systematic uncertainties. The state-of-the-art has reached the point, however, where ionization / scintillation detectors are sensitive to the single quantum, and indeed we have already embarked on searches for signals in this realm. Modeling the behavior of these materials

Figure 8: Predicted CENNS event rates from reactor neutrinos for pure LAr (blue), with the backgrounds from ^{39}Ar overlaid. The ^{39}Ar background for atmospheric Ar (dashed-magenta) is reduced by a factor of ~ 150 [36] when underground Ar is used (solid-green). The event rate assumes a neutrino flux of $6 \times 10^{12} \bar{\nu} \text{ cm}^{-2}\text{s}^{-1}$ at a distance of 25 m from a 3 GWth nuclear reactor core.

down to the single quantum level therefore becomes increasingly important as we push to this absolute limit of detector sensitivity. Understanding the source of backgrounds and parasitic effects is equally important, as their rates and energies compete with the sought-after signals of interest. Just one example of a background to single-quantum signals may come from positive ions trapped in solid noble element layers inside the cryogenic detectors [39].

For a pure Ar detector, the ionization yield for the average expected nuclear recoil energy from CENNS of reactor neutrinos (250 eV) is predicted to be $\sim 2e^-/\text{keV}$. With equal number of excitation and ionization events, there is potential to increase the ionization yield slightly through the utilization of the Penning mechanism in which excited Ar atoms ionize a Penning substance such as Xe [8, 40]. For 1.6% Xe doping, the Penning mechanism has a measured conversion efficiency of $\sim 60\%$ [40]. Due to the short range of recoiled Ar atoms, the Penning ionization of Xe must compete with the Penning ionization of another excited Ar atom or molecule, and thus for low-energy recoils the optimal concentration of Xe may be higher to maximize ionization yield. Even in pure Ar, the short Ar recoil track length—and hence large exciton density—may increase ionization due to bi-excitonic quenching [41]. At present the conversion coefficient is known for LHe [26] but not for LAr.

The predicted ionization yields suggest that LAr is a plausible candidate for detecting CENNS using reactor antineutrinos. The ^{39}Ar background can be reduced by

using underground Ar as the detector medium. Since it is possible that the CENNS signal could be observed with atmospheric Ar in the reactor on/off scenario, one must consider the ‘total-cost’ comparison of constructing a larger detector utilizing atmospheric Ar versus a smaller detector utilizing underground Ar. In addition to the search for CENNS with reactor neutrinos, LAr could also be utilized in the search for other interactions producing similar low-energy depositions. In any case, based on our predictions, it does not appear that local electron recombination has a large effect ($\lesssim 5\%$) on the very low energy ($\lesssim 5$ electrons) signals.

The source of low-energy backgrounds in dual-phase noble element detectors has not been conclusively identified. Multiple experiments have handled this lack of understanding by applying a lower limit to the analysis threshold (see, e.g., [29], though it is useful to note that all recent searches for weakly interacting massive particles have also applied analysis thresholds). In particular, Ref. [37] shows that the unattributed background in a dual-phase argon detector is rising quickly through 270 eV, and this background is in direct competition with the 250-eVee (electron equivalent) CENNS signal from reactor neutrinos. This rapid rise in ionization background at low energy is also seen in dual-phase xenon detectors [42, 43], leading to the hypothesis that all dual-phase noble element detectors might share a common effect contributing to the low-energy background.

7. Future Work

While our model represents the first step to develop an understanding of the intrinsic LAr detector response at low (<50 keV) energies, more work is needed. We have focused solely on the microscopic behavior of Ar ionization and recombination, but there are detector-specific effects that must be included in the individual detector response function, such as the effect of trapped ions on detector surfaces, which are beyond the scope of this paper.

The authors note that there may be other approaches to calculating the yield such as a first-principle atomic physics calculation [44]. Regardless of the theoretical approach, experiments in the corresponding energy range must be conducted for validation. Future experiments should not only measure yields and cross sections of relevant interactions such as Ar-Ar, Ar-Ar⁺, and Ar^{*}-Ar, but also measure the rate of Penning ionization as a function of material composition. Other fundamental measurements should include the spectra of ionization (secondary) electron energies in different collisions, and

the role of multi-body collisions. The authors recommend that the highest priority for calculations and measurements be given to Ar and Xe, given these materials’ prevalence in searches for dark matter and CENNS.

Experiments have been recently reported focusing on the measurement of the ionization and scintillation yields in LAr at <50 keV nuclear recoil energies [29, 45]. The framework presented here should be applicable to other noble elements such as Xe, and a prediction of the ionization yield for CENNS on Xe will be possible when Xe-Xe inelastic cross sections become available.

Other important potential applications for the model exist, related to the operation of dual-phase noble element detectors. The use of a stopped pion neutrino source, such as the spallation neutron source (SNS), would result in greater recoil energies than those produced from reactor antineutrinos [46]. In the search for dark matter, keV-scale energy depositions are anticipated from low-mass weakly interacting massive particles (WIMPs) [47].

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