Using a Wavelength Shifter to Enhance the Sensitivity of Liquid Xenon Dark Matter Detectors

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***Abstract*—Liquid xenon dark matter detectors have been successfully employed to search for WIMPs, a proposed dark matter candidate. Increasing the scintillation wavelength from 175 nm to 300–400 nm can increase light collection and consequently improve sensitivity to dark matter. Such a shift increases the reflectivity of common reflector materials, decreases the amount of Rayleigh scattering and boosts the quantum efficiency of photomultiplier tubes. In this paper, we show that depositing p-terphenyl (p-TP) solid wavelength shifter on PTFE reflectors and the entrance window of a photomultiplier tube enhanced light-collection efficiency by 21%. We also observed an anti-correlation between the liquid xenon temperature and electron drift length, indicating that the p-TP dissolves into liquid xenon with a temperature dependent solubility. Possible ways to solve the drift length problem are discussed in the paper.**

***Index Terms*—Dark matter, liquid xenon, p-terphenyl, scintillation, time projection chamber, wavelength shifting.**

I. INTRODUCTION

RECENT measurements strongly support the hypothesis that much of the mass in the universe is cold, dark and non-baryonic [1]–[3]. Weakly interacting massive particles (WIMPs), believed to be relics from the Big Bang, are a primary candidate for dark matter, and their existence can be determined with a detector of sufficient mass and sensitivity [4], [5]. To search for WIMPs, several experiments (XENON, XMASS, and ZEPLIN) are using two-phase liquid xenon emission detector filled with 10–25 kg of liquid xenon (LXe), with intentions of expanding to 1 ton of LXe in the near future [6]–[8]. The performance of these types of detectors directly depends on the effective collection of UV photons (175 ± 10 nm original emission spectrum) and electrons [9], [10]. Detection of both the scintillation and ionization signals from the bulk LXe provided $> 99\%$ rejection of background events in the XENON10 experiment which recently demonstrated the best cross sectional limit for WIMPs [8]. The light collection from the bulk LXe can be improved by coating reflecting and transmitting surfaces with a high quantum efficiency wavelength shifter [11]. Shifting the emission spectrum to longer wavelengths improves the light collection efficiency in the following ways. First, the reflectivity of common reflectors increases as the wavelength is raised. For example, the reflectivity of PTFE at 175 nm has been reported as between 55% [12] and 95% [13], and is 98.5% at 350 nm [14]. This large variation at 175 nm can be explained by two known sources of degradation: exposure to high intensity UV and the presence of light absorbing contaminants [15]. Spectralon™ (a form of PTFE optimized for high reflectivity) has a reflectivity that rises from 98% at 300 nm to $> 99\%$ above 400 nm (and is 98.5% at 350 nm [14].) Secondly, the Rayleigh scattering length increases with wavelength as λ^4 [16]. Extending the Rayleigh scattering length decreases the total distance traveled by the photons, which decreases the likelihood for absorption by impurities in LXe (assuming the wavelength shifted photons are not more readily absorbed by impurities.) The scattering length in LXe is calculated to be about 30 cm at 175 nm wavelength, which agrees closely with the measured value (see Table 3.12 in monograph [17]). Finally, the light signal is increased by wavelength shifting because the quantum efficiency of photomultiplier tubes typically increases with wavelength. As an example, work at the Jefferson Lab concluded that the deposition of p-terphenyl (p-TP) onto glass window of *Burle* 8854 photomultiplier tubes increases their quantum efficiency in the blue range by about 40% [18].

A group exploring the benefits of using wavelength shifters in dark matter detectors has successfully operated a NaI detector with p-TP wavelength shifter at 160 K [19]. While a wavelength shifter has been used in two-phase argon-based detectors [20], [21], we are unaware of any published results in a LXe two-phase detector. In this paper, we report on the first such results using vacuum-deposited p-TP on both reflective and transmitting optical surfaces.

II. EXPERIMENT

A. Two-Phase Liquid Xenon Emission Detector

Two-phase emission detectors utilize the effect of effective extraction of ionization electrons from the bulk condensed noble gas in an electric field of a few kV/cm [9]. Modern emission detectors typically consist of several photomultiplier tubes (PMTs) viewing LXe and thin-wire electrodes for applying electric fields for collection and extraction of ionization electrons from LXe into the equilibrium gas phase. Detected particles interact with LXe and produce scintillation (prompt emission of photons from de-excitation and recombination at the point of interaction) and ionization electrons that escape recombination and collected in the gas by the applied electric field. The drifting electrons excite the xenon gas, releasing a second flash (electroluminescence) proportional to the number

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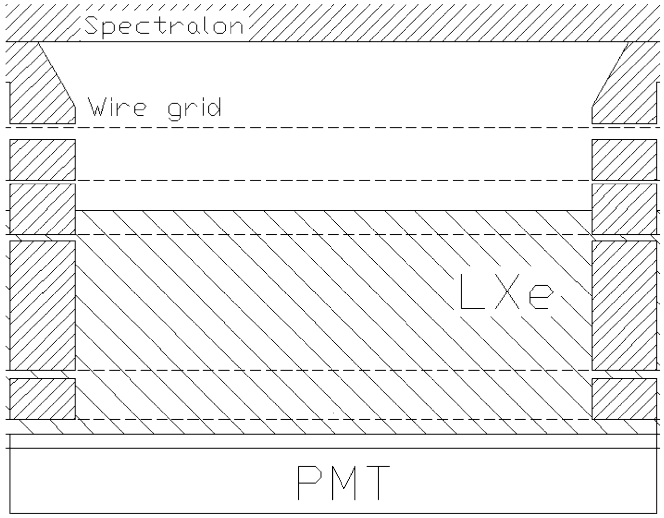

Fig. 1. Diagram of the detector in two-phase mode. In single-phase mode, the liquid is above the top Spectralon reflector.

of drifting electrons. The number of photons generated in the gas approximately follows the equation

$$N_{\rm ph} = a \cdot (E/P - b) \cdot x \cdot P,$$

where E (kV/cm), P (bar) and x (cm) are the electric field, gas pressure and track length, respectively; the empirical constants, a and b are equal to 70 photons/keV and 1 kV/cm-bar, respectively [10]. Both initial primary light (S1) and later proportional light (S2) signals are collected by the PMTs. The time difference between S1 and S2 provides a precise and unambiguous measurement of the depth of interaction. With a multiple-PMT array, the S2 hit pattern measurement gives the two-dimensional position of the interaction in the plane of the PMT photocathodes.

The detector used in this study consists of a single 50-mm diameter *Hamamatsu* R9288 PMT and five wire grids as shown in Fig. 1. The PMT is placed in the liquid for optimal light collection; most of the light undergoes internal reflection at the liquid surface due to the mismatch in gas and liquid refractive indices. The flat wire grids are formed by soldering stretched wires onto copper-patterned *Cirlex*™ polyimide rings. The wire grid just below the liquid level consists of 40 micrometer diameter beryllium-copper (BeCu) wires; the other four are made of 125 micrometer diameter gold-plated aluminum wires. All wire grids have a pitch of 2 millimeters. We call the grid just below the liquid surface the "gate"; the grid above and below this grid are the anode and cathode grids. The active volume is bounded by the cathode and gate grids and has a depth of 1.0 cm and diameter of 3.83 cm. The test setup includes Optical-Grade Spectralon reflector pieces and a PMT covered with the wavelength shifter. There are six reflector pieces—five rings and one circular plate. In the control setup, these parts are replaced with PTFE reflectors and an uncoated PMT. The quantum efficiencies of the coated and uncoated PMTs, as measured at room temperature by the manufacturer, are 17.32% at 350 nm and 16.27% at 175 nm, respectively.

The detector is housed in a liquid nitrogen coldfinger-cooled cryostat which maintained a LXe temperature stable to within 0.10 K. Patterned onto the *Cirlex*™ boards are three parallel plate capacitors for monitoring the liquid level between the gate and anode wire grids. We observed a liquid level stable to 40 micrometers over a period of six weeks.

A ^{57}Co source provided 122 keV gammas for calibration and measurement of light collection. An absolute calibration of the number of photons and electrons from these gammas was performed in an earlier experiment and is described briefly in [9]. The ^{57}Co source was placed beneath the stainless steel can, which contained the detector. The gammas were collimated to a 1.5 cm diameter region in the active volume.

Events near the reflector surfaces can suffer charge loss. Also, events near the wire grids and walls may experience an electric field significantly different from that in the center. Use of Maxwell 2D, a electromagnetic simulation package, showed that the direction and magnitudes of electrical fields are uniform to within 5% except at regions within $\sim$1 millimeters from the cathode and gate grids or within $\sim$5 millimeters from the PTFE/Spectralon walls. Implementing conservative drift time cut and using the collimator ensures that all events occur in roughly the same drift electric field and do not suffer from loss of charge.

Before filling with xenon, the detector was heated to 50°C and pumped out for several days. The xenon is cleaned upon filling by passing the gas through a hot metal getter purifier. The xenon is further purified by continuously circulating the xenon through the purifier. In this procedure, the LXe is pulled from the fiducial volume and out of the cryostat where it boils before flowing through the purifier. A pump pushes the clean xenon gas back to the detector where it recondenses.

B. Selection of Wavelength Shifter

A number of solid wavelength shifters have been considered for noble gas detectors including trans-stilbene, tetraphenylbutadiene (TPB), sodium solicylate, diphenylstilbene and p-terphenyl [17], [22]. Among them p-TP has the best record in applications for noble gas detectors because of its reasonable stability in vacuum, low hygroscopy, chemical inertness, and extreme radiation hardness. Several studies have proven that p-TP does not contaminate pressurized xenon and helium with electronegative impurities and have demonstrated $> 90\%$ quantum efficiency for converting 175 nm photons into 340 nm range [23]. We selected p-TP because it has been used in construction of most effective high-pressure xenon scintillation and electroluminescence detectors (see, for example, [24] and references therein). Emission and absorption spectra of p-TP can be found in [25].

C. Vacuum Deposition of Wave Length Shifter

There are two basic ways of depositing p-TP: coating with solution of p-TP in toluene or with vacuum deposition of the sublimated p-TP vapor. We opt for the latter technique because of the inherent higher purity and finer control of thickness. A standard vacuum thermo-deposition system was used to deposit 0.5 mg/cm^2 of p-TP on Spectralon reflectors and the window of the PMT. Before beginning deposition, p-TP was baked at 50°C to remove absorbed water. The sublimation began at a pressure of 2e-7 torr and at a temperature of 160°C. The coated pieces were allowed to cool down in the nitrogen atmosphere before removal. The thickness of deposition is estimated by measuring the mass of the p-TP before and after deposition and calculating the setup geometry. Before usage, the coated pieces were stored in a dry box to prevent water absorption.

III. RESULTS

A. Light Collection Efficiency

The PMT signal in units of photoelectrons can be described as follows:

$$N_{\mathrm{phe}} = (E/w_{\mathrm{ph}}) \cdot \mathrm{LCE} \cdot \mathrm{QE}$$

where N_{phe} is the number of photoelectrons generated by the S1, E is the energy of the 122 keV gamma, w_{ph} is the energy required to generate one photon (15.2 ± 0.3 eV from the calibration measurement described in [9]), LCE is the light collection efficiency, and QE is the quantum efficiency of the PMT. The number of photons emitted is equal to E/w_{ph}. The quantity $N_{\mathrm{phe}}/\mathrm{QE}$ is the number of photons that strike the photocathode. To obtain N_{phe}, we measured the PMT single photoelectron and primary light (S1) spectrums at zero drift electric field in single-phase (liquid level above the top reflector.) The data was taken at 175 K. Only events that are 4 to 6 mm above cathode are included. Dividing the two Gaussian fit mean values to these spectrums yields N_{phe}. The setup with PTFE reflectors (no wavelength shifter) obtained 5.0 photoelectrons per keV while the wavelength shifter setup achieved 6.5 photoelectrons per keV. These two values have a statistical error of less than 1%. There are two potential sources of systematic error. The first is the nonlinearity in the PMT signal response to larger S1's due to current output limitations. This effect was determined to be negligible for experiment. The second is the possible change in quantum efficiencies when cooled to cryogenic temperature. Having observed no change in the QE's by changing the temperature from 165 to 185 K, we assume that these values are constant with temperature. The LCEs of the PTFE and Spectralon setup were $47 \pm 1\%$ and $57 \pm 1\%$, respectively—the use of the wavelength shifter enhanced the light collection by 21%. The uncertainty in light collection stems from the error in w_{ph}. A light Monte Carlo was used to determine the reflectivity of the PTFE given these results. Simulated photons undergo perfectly diffuse reflection off non-metal surfaces and shift in wavelength before reflecting if the surface is coated. Photons are propagated until absorption. Refractive indices of 1.68 [26] and 1.51 (given by the PMT manufacturer) were used for LXe and PMT glass, respectively, and values of 30 and 100 cm were used for the Rayleigh and absorption scattering length [17], [27]. Reflectivity values of 0.40 and 0.35 were used for scattering wavelength shifted photons off BeCu (assumed same as Cu) and Au, respectively [28]. The reflectively of BeCu and Au below 200 nm could not be found. Varying reflectivity of BeCu at 175 nm from 0 to 1 shifts the LCE by less than 1% because wires' small size; a value of 0 was arbitrarily chosen. The reflectivity of Au is 0.3 near 200 nm [28]. Assuming that the Au reflectivity at 175 nm is between 0.3 ± 0.3, the Monte Carlo can match the LCE of the PTFE setup with PTFE reflectivity of 0.85 ± 0.05. A higher LCE of 67 ± 2 % (with the error from the uncertainty in Au reflectivity) was expected for the Spectralon setup based on the Monte Carlo. A post-run inspection revealed that some of the wavelength shifter had migrated to other parts of the detector, which could account for the lower than expected LCE.

B. Electron Life-Time

To measure the effects of impurities on charge collection in LXe, datasets were taken with the detector running in a standard two-phase mode at 175 K with a 1 kV/cm drift electric field in the liquid and a 10 kV/cm field just above the liquid surface for production of proportional light (S2). The liquid level was set halfway between the anode and gate grids. At 10 kV/cm, the efficiency of electron extraction from the liquid is ~100% [29]. Here, we quantify the drifting charge loss with an e-folding drift length parameter, determined by fitting an exponential function to the charge signal (S2) as a function of depth for 122 keV gammas. The electron drift length, in separate measurements of the same system but with no p-TP present, has routinely been measured to be greater than ~100 cm. (At 1 kV/cm, a 1 cm drift length corresponds to a 4.4 μsec electron lifetime.) This level of purity is typically achieved after several days of recirculating the xenon gas through a commercial purifier at 2–3 liters per minute, and is usually stable for time scales of at least a few months, requiring no further purification.

However, in this recent work with the wavelength shifter present, the electron drift length stabilized at only ~0.4 cm after more than a week of recirculation. Various attempts at purifying the xenon did not improve the situation: recirculating purification at different flow rates; emptying the detector of LXe and purifying the xenon separately; and modifying the detector to improve the efficiency with which the liquid in central active part of the detector is passed through the purifier. Furthermore, no vacuum leaks were found in the detector or associated plumbing. The impurity problem is likely related to the presence of the wavelength shifter.

C. Influence of Electric Field Strength

At fixed recirculation flow and LXe temperature, we measured the electron drift length as a function of the drift electric field. The drift length drops with increasing electric field strength (Fig. 2.), indicating that the electron capturing impurity is not molecular oxygen, which has an electron attachment rate that reduces with increased field strength (see, for example, [17] and references therein).

D. Influence of Temperature

A potentially more important insight into the impurity issue was revealed by the fact that under conditions of no recirculating purification, the drift length did *not* decrease over time. This is in marked contrast to the common situation with such detectors early in a run when outgassing appears to be the dominant source of impurities. Moreover, in static conditions with no recirculating purification, the drift length is *inversely related*

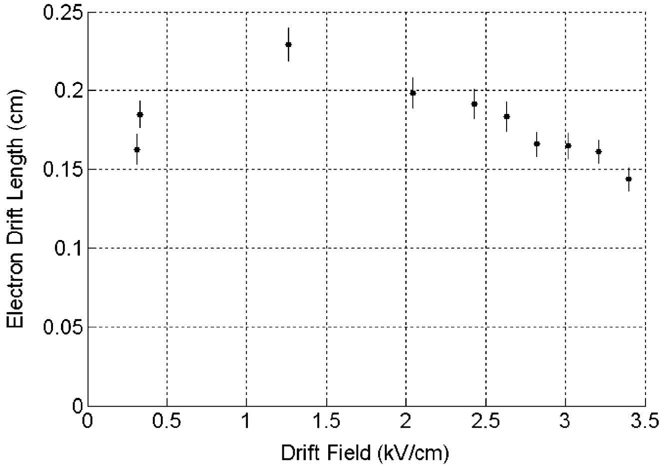

Fig. 2. Electron drift length in dependence on electric field strength in liquid xenon at 175 K. The vertical lines represent its statistical uncertainty.

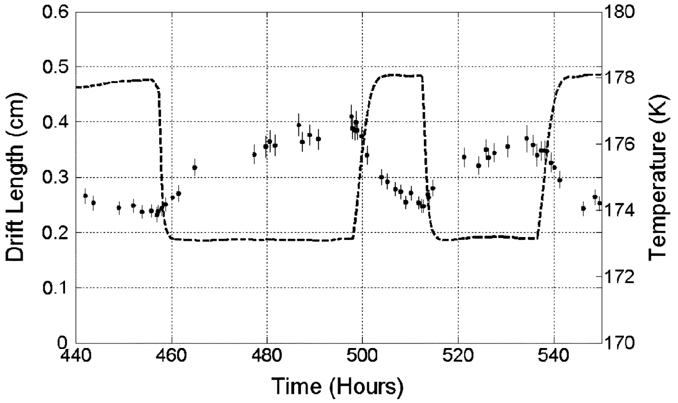

Fig. 3. Electron drift length (dots) and temperature (dashed line) versus time in the wavelength shifter test setup. The vertical lines represent the statistical uncertainty in the drift lengths.

to temperature with an apparently fixed relationship that is reversible over several cycles of temperature; this is shown in Fig. 3.

IV. DISCUSSION

This strong and reversible correlation between drift length and temperature suggests that the p-TP (or possibly an impurity in the p-TP) is dissolved in the LXe at a level determined by a temperature-dependent solubility. Molecules of p-TP may be acting as acceptors themselves or affecting the rate of electron capture by residual electronegative impurities by cooling the drifting of electrons. The fact that the drift length is reduced at elevated electric field strength indicates that the major acceptor is not molecular oxygen and may be associated with the presence of molecules of N_2O type [30]. Such kind of dominating acceptors is specific to LXe purified with hot getters [17].

The effect of dissolving organic substances in LXe from near room temperature to about $-100°$C was observed more than 25 years ago (see [31], [32]). Since that time, studies have shown in many ways that xenon behaves like the first member of the n-alkane family and thus has been used as an effective anesthetic [33].

TABLE I
PROPERTIES OF P-N-PHENYLS

	p-3P	p-4P	p-5P	p-6P
M.W.	230.1	306.41	382.50	458.59
M.P, °C	212	>300	381	475.16

M.W = Molecular Weight; M.P. = melting Point; p-3P = para-terphenyl; p-4P = para-terphenyl; p-5P = p-Quinquephenyl; p-6P = p-Sexiphenyl

Compounds with higher molecular weight are likely to have a lower solubility in LXe. From this point of view, it looks reasonable to repeat the experiment with heavier members of *p-N-phenyl* family such as p-quaterphenyl (p4P), p-quinquephenyl (p5P), and p-sexiphenyl (p6P)—see Table I. Indeed, p4P has been used as an effective extreme-ultra-violet wavelength shifter in the past (see, for example, [34]) and has demonstrated emission and adsorption spectra similar to that of p-TP [35]; p-N-phenyls have demonstrated high chemical stability and high luminescence quantum yield in the blue range [36]; and all of them can be deposited by evaporation with the sublimation temperature elevated for heavier members. These organic substances attract attention because of their potential demonstrated in organic light-emitting diodes, organic field-effect transistors, and solar cells [37].

In conclusion, we have demonstrated that the presence of wavelength shifter can increase the light collection of a LXe detector with Spectralon used as a reflecting surface. Our first choice of p-TP as the wavelength shifter results in unacceptable levels of charge loss, but other promising wavelength shifter materials with potentially lower solubility in LXe remain an attractive option.

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