Measurement of scintillation and ionization yield with high-pressure gaseous mixtures of Xe and TMA for improved neutrinoless double beta decay and dark matter searches

Y Nakajima, A Goldschmidt, H S Matis, D Nygren, C Oliveira, J Renner

Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

E-mail: YNakajima@lbl.gov

Abstract. Liquid Xe TPCs are among the most popular choices for double beta decay and WIMP dark matter searches. Gaseous Xe has intrinsic advantages when compared to Liquid Xe, specifically, tracking capability and better energy resolution for double beta decay searches. The performance of gaseous Xe can be further improved by molecular additives such as trimethylamine(TMA), which are expected to (1) cool down the ionization electrons, (2) convert Xe excitation energy to TMA ionizations through Penning transfer, and (3) produce scintillation and electroluminescence light in a more easily detectable wavelength (300 nm). These features may provide better tracking and energy resolution for double-beta decay searches. They are also expected to enhance columnar recombination for nuclear recoils, which can be used for searches for WIMP dark matter with directional sensitivity. We constructed a test ionization chamber and successfully measured scintillation and ionization yields at high precision with various Xe and TMA mixtures and pressures. We observed the Penning effect and an increase in recombination with the addition of TMA. However, many undesired features for dark matter searches, such as strong suppression of the scintillation light and no sign of recombination light, were also found. This work has been carried out within the context of the NEXT collaboration.

1. Introduction

Xenon TPCs are among the most popular and sensitive detector technologies to search for WIMP dark matter and neutrinoless double beta decay. While liquid Xe is more commonly used for such applications, gaseous Xe has intrinsic advantages over liquid Xe. For neutrinoless double beta decay search, gaseous Xe provides superior energy resolution and tracking capability. Those excellent capabilities are demonstrated in Refs. [1, 2] and being carried out within the NEXT experiment [3].

In addition, there is a potential capability of performing dark matter searches with directional sensitivity utilizing columnar recombination [4, 5]. To put this idea into practice, it is essential to efficiently cool ionized electrons. The directional sensitivity can be further enhanced by the Penning effect, which converts primary excitation into an ionization signal. Hence, doping with molecular additives would be essential to achieve enhancement of columnar recombination.

We are exploring possible performance improvements by adding trimethylamine(TMA) to

gaseous Xe. We report measurements of ionization and scintillation yields with a test ionization chamber that we constructed at Lawrence Berkeley National Laboratory.

2. Performance improvements with TMA

For typical Xe TPCs, observable signals are scintillation light from excited Xe ($Xe \rightarrow Xe^*, Xe^* + Xe \rightarrow Xe_2^* \rightarrow 2Xe(g.s.) + h\nu$) and ionized electrons ($Xe \rightarrow Xe^+ + e^-$). The relative size of those signals can be altered with the presence of sufficient recombination, $Xe^+ + e^- \rightarrow Xe^*$ and $Xe^* + Xe \rightarrow Xe_2^* \rightarrow 2Xe(g.s.) + h\nu$, which effectively converts ionization signals into the scintillation signals. A sufficient amount of columnar recombination would be required for a dark matter search with directional sensitivity. However, due to large electron diffusion, such recombination in a pure Xe medium is expected to be negligible.

TMA is known to have a large inelastic cross section for electrons due to various vibrational and rotational modes, and provides efficient cooling of ionization electrons, which is essential for the recombination processes. In addition, the ionization potential of TMA (7.85 ± 0.05 eV [6]) is well matched to the energy level of the first excited state of Xe (8.3 eV [7]). This allows a Penning transfer ($Xe^* + TMA \rightarrow Xe + TMA^+ + e^-$), which increases the intrinsic amount of ionization that would undergo columnar recombination. We also expect that the charge exchange process, $Xe^+ + TMA \rightarrow Xe + TMA^+$, would occur quickly. With efficient Penning and charge-exchange transfers, initial excited and ionized Xe would be largely converted into ionized TMA^+ and electron pairs. Finally, TMA produces fluorescence light at ~ 300 nm, which is much more easily detectable compared to the light from Xe (~ 170 nm) because of typically higher PMT quantum efficiency at 300 nm. If recombination of ionized TMA produces similar fluorescence light, via $TMA^+ + e^- \rightarrow TMA^* \rightarrow TMA(g.s.) + h\nu(300\text{ nm})$, then the detection efficiency for columnar recombination would be significantly increased.

In addition, Penning transfer increases the number of ionized electrons, which improves statistical precision of the energy measurement and track reconstruction. Tracking performance would also be improved with TMA by reducing the diffusion of ionization electrons [8]. These features would further benefit neutrinoless double-beta decay searches in gaseous Xe.

3. Experimental setup

In order to test the properties of the gas mixtures of Xe and TMA, especially Penning transfer efficiency and fluorescence light from recombination processes, we built a test ionization chamber as shown in Fig. 1.

A pair of parallel plate electrodes with gap width of 5 mm is placed in the chamber that can hold pressurized gas up to 8 bar. One of the electrodes is separated into a 2.5 cm-diameter inner electrode and an outer electrode. The signal induced in the inner electrode is used for the analysis, in order to avoid the effects from the charge produced outside the gap between the electrodes. Four PMTs are placed at the circumference of the electrodes to detect scintillation light from the gas between the electrodes. A long-pass filter that blocks light below 250 nm is installed in front of one of the PMTs, so that this PMT is only sensitive to the scintillation light from TMA, eliminating the Xe VUV light. We irradiated the gas between the electrodes with $\sim$60 keV gamma rays from a 10 mCi ^{241}Am radioactive source. Currents from electrodes and PMTs were read with pico-ammeters in DC mode. A more detailed description of the setup can be found in Ref. [9].

The gas flow in our system is illustrated in Fig. 2. Once the chamber is filled with the gas mixture, it is sealed and the gas inside is purified by continuous circulation through a SAES MC50-702F room temperature gas filter ("Cold getter" in Fig. 2). While this filter is essential for removing oxygen, which blocks the scintillation light from Xe, we found that the filter also takes a significant amount of TMA from the Xe + TMA mixture. We noticed that the TMA concentration in the gas mixture decreases shortly after introducing the gas into the system, and

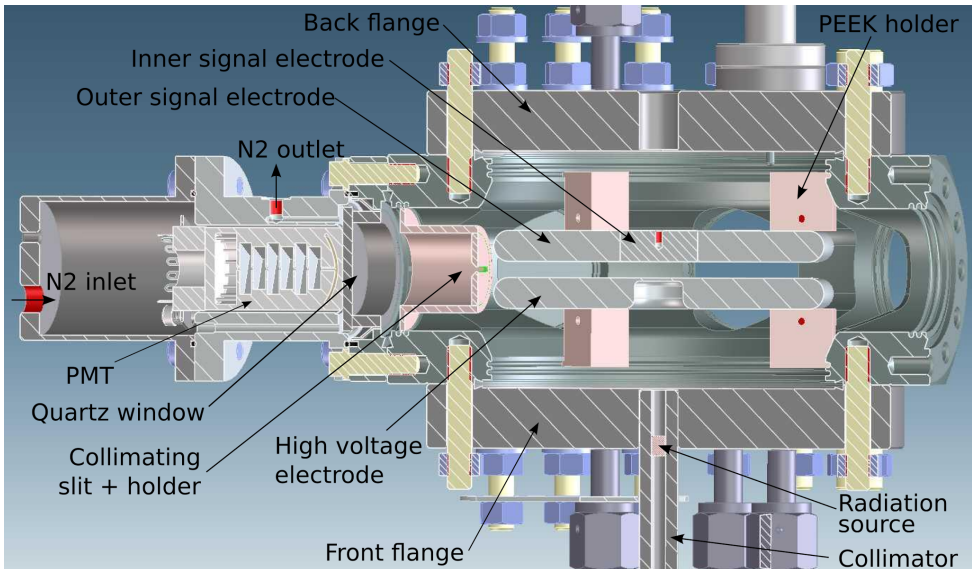

Figure 1. Schematic of the ionization chamber.

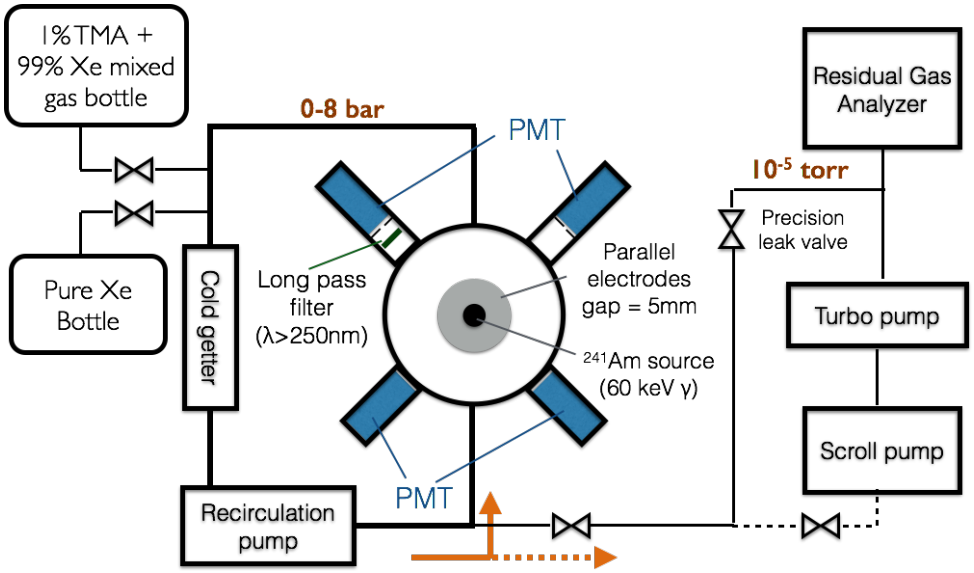

Figure 2. Schematic diagram of the gas flow system.

then stabilizes after about one hour of recirculation. This makes it difficult to precisely control the TMA fraction in the chamber, and hence we continuously monitored the gas composition using a residual gas analyzer (RGA). As shown in Fig. 2, a small fraction of the gas inside is continuously introduced into the RGA through the precision leak valve. The gas pressure around the RGA was adjusted to $\sim 10^{-5}$ torr. Figure 3 shows a typical mass spectrum of the Xe+TMA mixture, obtained with the RGA. Singly and doubly ionized Xe, as well as TMA mass peaks, can be seen. We monitored the relative height of Xe and TMA pressures, and extracted the TMA fraction in the main chamber. The absolute calibration of the TMA fraction was done by introducing 1% TMA and 99% Xe mixture to the RGA directly, without the filter.

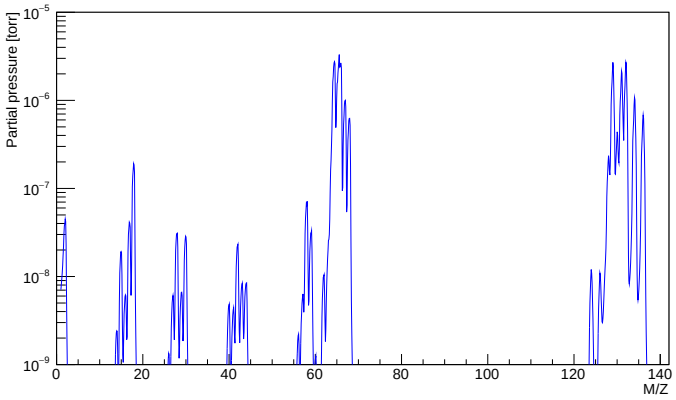

Figure 3. Example output of an RGA analog mass scan for a Xe+TMA gas mixture. The horizontal axis presents the mass divided by the unit charge. Peaks around $M/Z = 130(65)$ correspond to singly(doubly) ionized Xe atoms. Peaks around M/Z of 58, 42 and 30 are mostly from TMA and its fragments. Residual components of H_2(M/Z=2), H_2O(M/Z =18) and N_2(M/Z=28) are also shown.

4. Results with pure Xe

The system was initially tested with pure Xe gas [9]. Figure 4 shows results of light yield measurements at various electric fields and pressures. The amount of energy deposition in the active region, which changes with pressure, is corrected using a GEANT4-based Monte Carlo simulation. Primary scintillation light for $E/\rho < 10^5$ [Vcm^{2}g^{-1}] and proportional amplification at higher electric fields (electroluminescence light) are clearly seen. The results are consistent among the different pressures up to 8 bar. Figure 5 shows the current induced in the electrode due to ionization processes. Again, good agreement between different pressures was seen. The charge yield is relatively flat for $E/\rho > 10^3$ [Vcm^{2}g^{-1}], which indicates an accurate charge correction efficiency at various conditions. In addition, this flatness which extends to higher fields of $E/\rho \approx 10^5$ [Vcm^{2}g^{-1}] suggests that the increase in scintillation yield at the same higher field region is due to proportional amplification without charge amplification (avalanche). The reduction of the charge yield at the low field region of $E/\rho < 10^3$ [Vcm^{2}g^{-1}] is likely due to electrons diffusing out. Those results are in good agreement with the previous measurements [10, 11]. A more detailed description of test results with pure Xe can be found in Ref. [9].

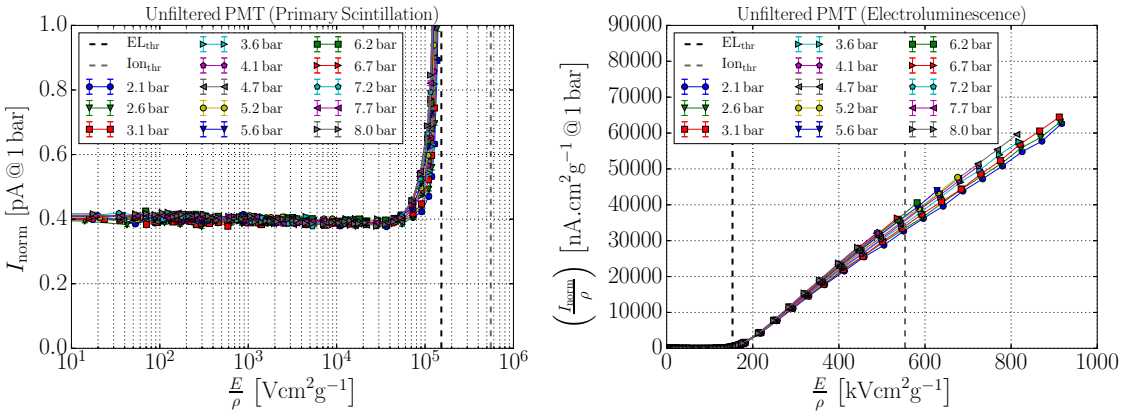

Figure 4. Light yield of pure Xe gas as a function of external electric field for different total pressures. (Left) Results in the primary scintillation region. (Right) Results in the Electroluminescence region.

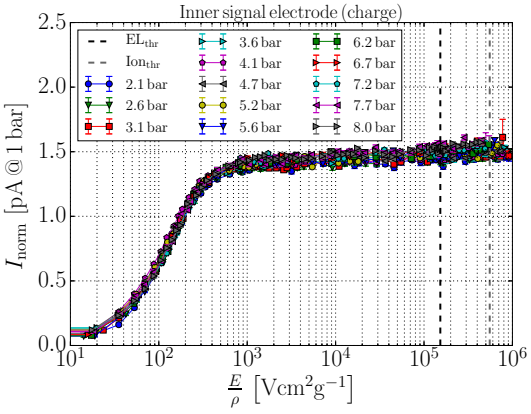

Figure 5. Charge yield of pure Xe gas as a function of external electric field for different total pressures.

5. Results with Xe and TMA mixture

5.1. Charge yield

Figure 6 shows the charge yield observed at the inner electrode at approximately 4 bar total pressure and for various TMA concentrations. We found that the slope of charge yield vs. electric field at $10^3 < E/\rho < 10^5$ (Vcm^{2}g^{-1}) increases as the TMA fraction increases. That can be interpreted as an effect of increased recombination, since recombination is larger at lower electric field and becomes negligible at high electric field. Therefore, the larger slope indicates larger amount of recombination at a low electric field with more TMA. Furthermore, we observed that the charge yield at a high electric field of $\sim 10^5$ Vcm^{2}g^{-1} increases as the TMA fraction increases, which we interpreted as the effect of Penning transfer. We also found that charge amplification starts at lower electric field with the addition of TMA. This could be due to lower ionization potential of TMA and also due to Penning transfer.

Figure 7 shows the relative size of charge yield for $10^5 < E/\rho < 2 \times 10^5$ (Vcm^{2}g^{-1}), as a function of TMA concentration. We found a clear correlation between charge increase and TMA concentration. The charge yield is increased by $\sim 5\%$ with 1% TMA concentration. In gaseous Xe, the relative size of excitation and ionization is measured to be $1/W_{sc} : 1/W_i \sim 1 : 2.5$ [12], where $W_{sc(i)}$ is the amount of energy loss of electrons needed to excite (ionize) one Xe atom. Penning efficiency, $\epsilon(Penning)$, can be calculated using the relative charge yield between Xe+TMA mixture to pure Xe, I(Xe+TMA)/I(pure Xe), as

$$\epsilon(Penning) = \frac{W_{sc}}{W_i}\left(\frac{I(\text{Xe+TMA})}{I(\text{pure Xe})} - 1\right). \quad (1)$$

Hence, a 5% increase in the charge yield translates into 10-15% of Penning transfer efficiency. We observed a clear signature of Penning transfer. However, the amount of Penning transfer is found to be relatively small to make any significant impact on the ionization signal.

5.2. Light yield

Figure 8 shows results of light yield measurements with various TMA concentrations with an unfiltered PMT. We found a large reduction of primary scintillation light once a very small TMA fraction, as low as $\sim 0.01\%$, is introduced. On the other hand, we observed electroluminescence light at higher electric field, as shown in Fig. 9. This EL light is observed for both unfiltered and filtered PMTs, which indicates that it is scintillation light from TMA and the light from Xe is practically lost.

Although the amount of primary scintillation light is largely suppressed with Xe+TMA mixture, we observed non-zero amount of the scintillation light. Figure 10 shows the observed

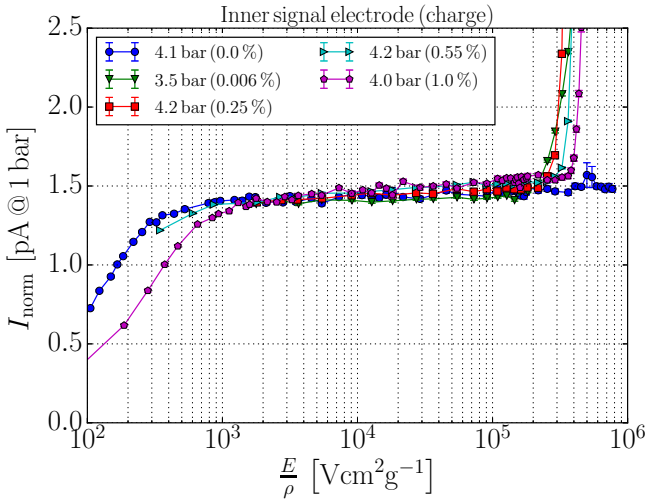

Figure 6. Charge yield of gas mixture of TMA and Xe as a function of external electric field for a total pressure of approximately 4 bar and various TMA concentrations.

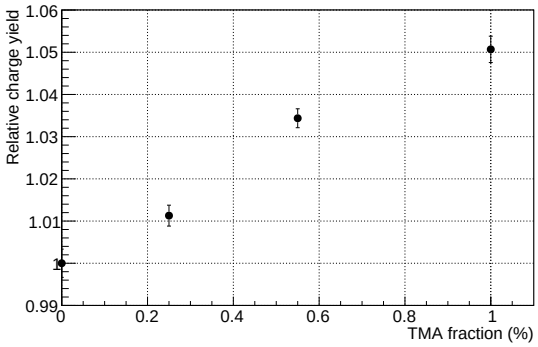

Figure 7. Relative size of averaged charge as a function of TMA concentration for a total pressure of approximately 4 bar. The data for $10^5 < E/\rho < 2 \times 10^5$ (Vcm^{2}g^{-1}) are used to avoid the effect of the recombination process.

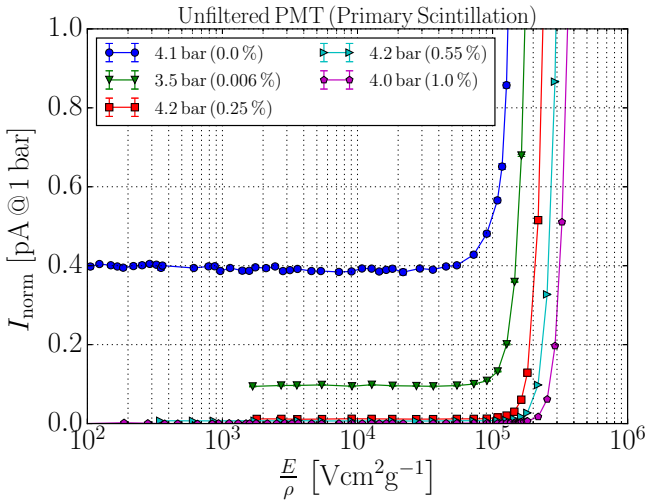

Figure 8. Primary scintillation light yield with Xe+TMA gas mixture, measured at approximately 4 bar total pressure and various TMA concentration.

current from the unfiltered PMT with $\sim 0.5\%$ TMA concentration for different total pressures. The size of the signal is approximately 1-4% of that of pure Xe. This is roughly consistent with the expectation from direct excitation of TMA, after taking into account the difference of PMT quantum efficiency between 170 nm and 300 nm and the difference in excitation cross sections between Xe and TMA. A consistent amount of scintillation light was observed with the filtered

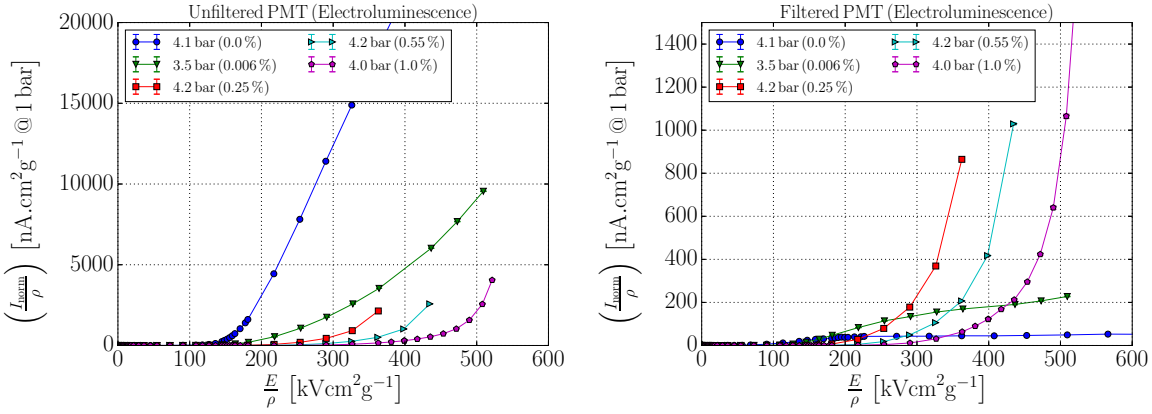

Figure 9. Electroluminescence light yield with Xe+TMA gas mixture, measured at approximately 4 bar total pressure and for various TMA concentrations. The left(right) panel shows the results from unfiltered(filtered) PMT.

PMT as well, which also supports the conclusion that the observed light is only emitted from TMA. The suppression of the light yield at higher pressure is consistent with the self-quenching effect of the TMA [13]. A more careful investigation of this self-quenching effect is underway. If the TMA recombination produces fluorescence light, it would be anti-correlated to the charge yield and would appear as a negative slope of light yield as a function of electric field. However, we observed a constant light yield as a function of electric field for $10^3 < E/\rho < 10^5$ (Vcm^{2}g^{-1}). Therefore, we conclude that no significant additional scintillation light, such as light from TMA recombination, energy transfer from Xe, is observed.

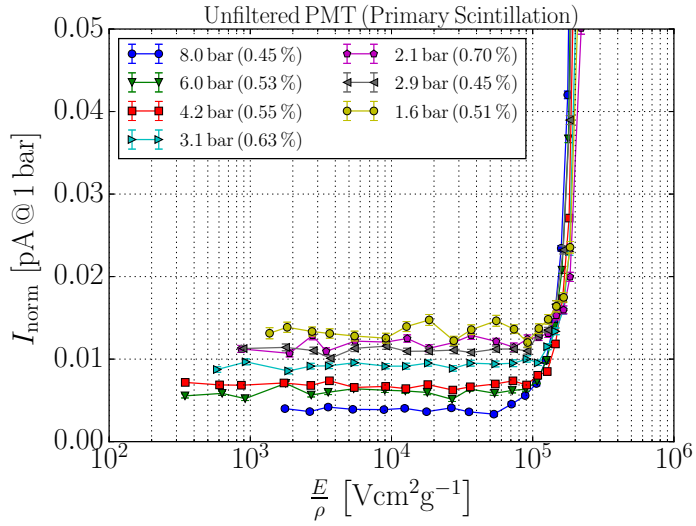

Figure 10. Primary scintillation light yield with Xe+TMA gas mixture. Results of unfiltered PMT at approximately 0.5% TMA concentration and for different total pressures are shown.

6. Discussion

One of the most significant effects that we observed with the Xe and TMA mixture is the high suppression of the primary scintillation light. It is likely that TMA is either absorbing Xe scintillation light, or taking out energy from Xe excited states but not releasing the energy in the

form of observable light. We observed enhancement of the recombination process through the reduction of charge yield at low electric field. However, we did not observe a corresponding increase of scintillation light. This can be due to either lack of charge-exchange process ($Xe^+ + TMA \rightarrow Xe + TMA^+$) and also due to lack of fluorescence after TMA recombination. An increase of charge yield due to Penning effect was clearly observed, and Penning efficiency was found to be 10-15% for 1% TMA concentration at 4 bar total pressure. This, however, corresponds to an increase in the number of ionization electrons at the level of only 5% and would not give significant improvement in the detector performance. All of those observations indicate that, unfortunately, TMA is not a suitable additive to enhance performance of gaseous Xe TPC for dark matter searches. While the suppression of scintillation light is also problematic for neutrinoless double beta decay searches, other features of TMA, such as reduction of diffusion and increase of charge yield, can still be beneficial for this application and are actively studied [8, 14].

On the other hand, our system is capable of measuring scintillation and ionization yield for any gas mixture at high precision. Other gas mixtures, such as Xe and Triethylamine(TEA), may be tested to explore ideal gas mixtures.

7. Summary

Using a test parallel-plate ionization chamber, we successfully made high-precision measurement of scintillation and ionization yield for Xe+TMA gas mixture at various electric fields. Although we found the Penning effect and enhancement of the recombination process with the addition of TMA, many undesired features, such as strong suppression of the scintillation light and no sign of recombination light, were also found. Therefore, it is likely difficult to utilize TMA to enhance the performance of gaseous Xe TPCs in dark matter searches. Other features of TMA, such as diffusion reduction and charge gain increase, can still benefit neutrinoless double beta decay searches.

Although TMA is found to be not an ideal additive, our system has proven to be capable of measuring scintillation and ionization yield at high precision. We are exploring the possibility of testing other gas mixtures, such as Xe+TEA.

8. Acknowledgments

We acknowledge the indispensable technical support from Tom Miller (LBNL) during the design, construction and running of the detector. This work was supported by the Director, Office of Science, Office of Basic Energy Sciences, of the US Department of Energy under contract no. DE-AC02-05CH11231. J. Renner (LBNL) acknowledges the support of a US DOE NNSA Stewardship Science Graduate Fellowship under contract no. DE-FC52-08NA28752.

References

[1] Alvarez V *et al.* (NEXT Collaboratoin) 2013 *Nucl.Instrum.Meth.* **A708** 101–114
[2] Alvarez V *et al.* (NEXT Collaboratoin) 2013 *JINST* **8** P09011
[3] Granena F *et al.* (NEXT Collaboratoin) 2009 *Preprint* arXiv:0907.4054
[4] Nygren D 2013 *J.Phys.Conf.Ser.* **460** 012006
[5] Nakajima Y *et al. In these proceedings*
[6] NIST Chemistry WebBook URL `http://webbook.nist.gov/cgi/inchi/InChI%3D1S/C3H9N/c1-4(2)3/h1-3H3`
[7] NIST Atomic Spectra Database URL `http://www.nist.gov/pml/data/asd.cfm`
[8] Gonzalez-Diaz D, Alvarez V, Borges F, Camargo M, Carcel S *et al.* 2015 *Preprint* arXiv:1504.03678
[9] Oliveira C, Gehman V, Goldschmidt A, Nygren D and Renner J 2015 *Physics Procedia* **61** 742 – 749
[10] Monteiro C, Fernandes L, Lopes J, Coelho L, Veloso J *et al.* 2007 *JINST* **2** P05001
[11] Freitas E, Monteiro C, Ball M, Gomez-Cadenas J, Lopes J *et al.* 2010 *Phys.Lett.* **B684** 205–210
[12] Renner J *et al.* (NEXT Collaboration) 2014 *Preprint* arXiv:1409.2853
[13] Cureton C G, Hara K, O'Connor D V and Phillips D 1981 *Chemical Physics* **63** 31–49

[14] Alvarez V *et al.* (NEXT Collaboration) 2014 *JINST* **9** C04015