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# Can the “intrinsic” energy resolution in xenon be surpassed?

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**Abstract.** Energy resolution is determined by fluctuations in the transformation of deposited energy to ionization and excitation. For noble gases, adding a molecular impurity that introduces a strong Penning effect reduces the Fano factor, improving energy resolution. I show that in xenon, adding trimethylamine (TMA) may provide a strong Penning effect and also permit electroluminescence (EL) at wavelengths characteristic of TMA. This combination may permit a large TPC to be operated with better than “intrinsic” energy resolution in the search for neutrino-less double beta decay in  $^{136}\text{Xe}$ . This path is being explored within the context of the NEXT collaboration.

## 1. Introduction

A large EL TPC filled with high-pressure xenon gas appears to be an attractive approach for a neutrino-less double-beta decay ( $0\nu\beta\beta$ ) search in  $^{136}\text{Xe}$  [1-6]. This expected rarity of this decay requires best possible energy resolution and knowledge of event topology to suppress backgrounds [7]. A good tracking capability, such as provided by the TPC, has been shown to permit suppression of single electron backgrounds by a large factor [8,9].

With regard to energy resolution, the performance champions are based on bolometric or germanium diode systems. These detectors generally offer one or two parts per thousand resolution FWHM at MeV energies [10,11]. In contrast, large detectors based on liquid or gaseous xenon have typically shown substantially worse energy resolution [4,12,13]; the exception with competitive resolution uses signal detection based on electroluminescence (EL) [14]. I show here that an avenue may exist for a large tracking TPC to approach energy resolution comparable to the performance champions, perhaps within a factor of two or three. How can this be? To begin, the conventional “intrinsic” resolution needs to be defined.

## 2. The intrinsic energy resolution

For a fixed energy input  $E$  to some medium, the variance  $\sigma_I$  in the observed ionization signal is customarily defined as

$$\sigma_I^2 = FN_I \quad (1)$$

where  $F$  is the Fano factor [15] and  $N_I$  is the expectation for primary electrons for the energy  $E$ ,  $N_I$  is the ionization signal to be detected, and is given by the ratio of  $E/w$ , where  $w$  is the average energy to make an electron-ion pair. Generally, in addition to  $E$ ,  $w$  depends on density and other properties of

the medium, and the strength of ambient electric field, if any. From this, the “intrinsic” energy resolution is

$$\delta E/E \equiv 2.35 \sigma_I/N_I \text{ FWHM} \quad (2)$$

$$= 2.35 (Fw/E)^{1/2} \quad (3)$$

The Fano factor for the ionization signal for gaseous xenon ( $\rho < 0.5 \text{ g/cm}^3$ ) and MeV energies has been found experimentally to lie in the range  $F_{\text{gxe}} = 0.15 \pm 0.02$  [16-19].<sup>1</sup> For energetic electrons, the parameter  $w$  falls in the range of 24 – 25 eV, depending on ambient electric field and density [20]. For  $^{136}\text{Xe}$ , a Q-value of 2457 keV is released in a decay to  $^{136}\text{Ba}$  [22,23]. From these numbers, the “intrinsic” energy resolution in gaseous xenon for the ionization signal is

$$\delta E/E = 2.7 \times 10^{-3} \text{ FWHM} \quad (4)$$

This expression ignores all degrading effects of detection and measurement; the essential challenge is to realize something close to this in a large-scale system.

For  $E = 2457 \text{ keV}$ , the ionization yield of electron/ion pairs is

$$N_I \sim 1 \times 10^5 \quad (5)$$

depending slightly on imposed electric field. At this energy, the fluctuation expected, from (1), is

$$\sigma_I \sim 122 \text{ electrons rms} \quad (6)$$

From an experimental perspective, this is a soberingly small number.  $N_I$  electrons must be detected while adding noise or fluctuations that are small on the scale of (6).

It is quite reasonable to expect that fluctuations in the energy deposition process are uncorrelated with additional fluctuations or noise added in the subsequent detection process. Therefore, (3) can be expanded to include a term  $G$ , encompassing all degrading effects of detection and measurement:

$$\delta E/E = 2.35 ((F + G)E/w)^{1/2} \quad (6)$$

$G$ , ideally, should be small compared to  $F$ , or at the very least, not larger. This challenge cannot be met with any avalanche gain device, since early gain fluctuations are amplified exponentially;  $G$  values around 0.7 are expected [24]. Nor can a unity gain approach suffice, since electronic noise and microphonic effects cannot be made sufficiently small in a large system.

### 3. Electroluminescence

The only ultra-low-noise gain process available appears to be electroluminescence (EL), in which an ionization signal is converted first to an optical signal, then reconverted to an electronic signal typically using photomultipliers (PMT) or other photon sensors [25-29]. While this approach may seem awkward, perhaps even irrational, excellent performance is realizable. An effective noise level much less than one electron rms can be achieved, offering single electron sensitivity, a remarkable circumstance in a large system. Space charge effects have no impact on gain or linearity, and microphonic effects are highly suppressed.

<sup>1</sup> As xenon density is increased above  $\rho \sim 0.5 \text{ g/cm}^3$ , increasingly large fluctuations appear in the partitioning of energy between scintillation and ionization, seriously complicating the measurement of energy [14]. At densities near liquid, the Fano factor approaches 20 [21], resulting in a factor of  $\sim 11$  worse resolution than low-density gas.

In EL, the primary ionization electrons drift into a high-field region where they rapidly gain energy. Collisions are entirely elastic up to the first excited state of xenon at 8.32 eV. If the EL field is high enough, very little energy is lost in collisions with xenon atoms, and the electron energy increases rapidly in the high field. When an electron energy exceeds 8.32 eV, an inelastic collision then transfers most of the electron's kinetic energy to a xenon atom. The electron begins again to acquire energy, producing another excitation, but under proper conditions never acquires energy sufficient to ionize xenon atoms.

The excited xenon gas atom rapidly forms an excimer. An infrared transition produces a relaxed excimer, for which the characteristic excitation energy is  $\sim 7.3$  eV, (175 nm) somewhat less than the first excited state of xenon at 8.32 eV. The excimers emit VUV radiation in a band around 175 nm. This band is not absorbed by xenon and the light can be detected at large distances.

Each primary electron falls through the same gap potential  $V$ , producing  $M$  photons in a linear gain process. The variance of EL gain  $M$  can be described by a factor  $J_{CP}$ , for which  $J_{CP} = 1$  would be characteristic of a Poisson distribution.<sup>2</sup> Due to the linearity of EL gain, the variance of  $M$  can reach values as low as  $J_{CP} = 0.01$ , remarkably sub-Poisson [30]. Well above the threshold field for producing EL (but below the threshold for additional ionization), the EL efficiency is high: about 60 – 80% of acquired energy is converted to light. In other words, for broadly optimal  $E/p$  conditions, only about  $\sim 10 - 12$  V is expended per photon. For example, a gain  $M = 1000$  is possible with 10 – 12 kV across the gap supporting the EL field. In practice, due to limited geometrical coverage and PMT quantum efficiency, the detection efficiency of the EL light is less than 10%, and the number of detected photons  $n_{pe}$  is the quantum bottleneck dominating the statistical precision.

Nevertheless, in a real sense, EL offers a method to count individual primary electrons with high statistical precision. As photodetectors vary in their response to single photons, here denoted by  $\sigma_{pd}$ , the narrower that response, the better the electron counting precision. Taking these factors in quadrature, it is easy to show that [3]

$$G = J_{CP} / M + (1 + \sigma_{pd}^2) / n_{pe} \quad (7)$$

A practical goal is that  $G \leq F$ . Perhaps,  $G \ll F$  can be achieved someday. In this paper, I focus on the possibility that it may be possible to reduce  $F$  itself, substantially, while maintaining  $G \leq F$  by a proportional increase in optical gain.

#### 4. The Fano factor and the Penning effect

The Fano factor reflects the impact of a fixed energy input on the energy partition among major pathways of energy loss. The fluctuations in number populations of ionization, excitation, and heat are constrained by the fixed energy input. Providentially, the Fano factor for not-too-dense xenon, as foot-noted above, is much smaller than 1. The only apparent way to reduce  $F$  further is to eliminate or severely reduce the population of excitations through the Penning effect. Because one of two populations is severely reduced, the fluctuations in both populations is reduced, producing the desired result for ionization.

The Penning effect occurs in noble gases, to which an impurity atom/molecule with an ionization potential (IP) slightly smaller than the excitation energy of the long-lived noble gas atoms or excimers has been added. Collisional de-excitation of the majority noble gas by the impurity leads to

<sup>2</sup> The factor  $J_{CP}$  honors the pioneering contributions of C. A. N. Conde and A. J. P. Policarpo, University of Coimbra, Portugal to the development of EL as a technique.

ionization of the impurity. With xenon, no noble gas Penning effect candidates are possible, leaving only molecular species. For molecular impurities, the efficiency of the Penning process appears to depend not only on the IP, but on chemical or structural characteristics as well, as breakup reactions may also occur, instead of ionization. The choices appear to be few.

After the energy deposition process, an EL TPC with a strong Penning effect present will, by design, deplete the primary excitation population, reducing  $F$  and increasing  $N_i$ , as desired. Subsequently, after drift to the high-field EL region, the primary electrons will gain energy and collide with the molecular impurities as well as with xenon. It is essential that the energetic electrons never ionize the impurity molecules, since that would lead to a large  $G$ . This imposes a requirement that the Penning molecule also will provide efficient fluorescence, fluorescence that would be otherwise be provided by the xenon atoms. Paradoxically, the molecule ionizes freely when exposed to collisions with excited xenon atoms/excimers, but never when exposed to a population of electrons gaining energy in an increased electric field. Instead, the molecule must convert the kinetic energy of the electron population to light.

In short, I seek a molecule displaying a strong Penning effect with xenon, strong fluorescence, and low electronegativity at all relevant electron energies. Do such molecules exist? A starting point is the family of aliphatic amines, which are known to display strong fluorescence. The only candidates I have found that may meet all criteria are trimethylamine (TMA) and dimethylamine (DMA); but there may be others. The IPs of TMA and DMA are  $7.85 \pm 0.05$  eV and  $8.32 \pm 0.03$  eV, respectively [31, 32]. Both are claimed to display a strong Penning effect [31], inferred from a large reduction in operating voltage in xenon-TMA or xenon-DMA mixtures, relative to other similar xenon + molecular gas mixtures at one bar. The evidence for the Penning effect presence thus is indirect, but the effect is quite large. Interestingly, the response in xenon-triethylamine (TEA) mixtures did not suggest the presence of a significant Penning effect [31], even though the IP of TEA is  $\sim 7.5$  eV.

TMA and TEA fluoresce strongly in 280 – 310 nm range [33]. Probably DMA does as well, but no studies appear to exist, but I have not found any studies of EL response in xenon-TMA or xenon-DMA mixtures. However, early work with a related mixture, argon – TEA showed a strong EL response, and a fairly wide range of optical gain before charge gain begins [29]. As the  $\sim 300$  nm range corresponds to  $\sim 4$  eV, less energy is needed per photon in a Penning – EL mixture; detection is easier as well.

### 5. EL TPC with Penning effect molecules?

A central element of this concept is that, as the electrons gain energy in the EL region, they transfer their excess energy efficiently to the TMA or DMA by excitation. The efficient transfer of energy is needed to ensure that no electrons reach energies corresponding to the IPs of TMA/DMA. The molecular density must be sufficient to scavenge energy from all electrons with energies above excitation,  $\sim 4$  eV. So, in this scenario, TMA or DMA completely “replaces” xenon, even though the TMA or DMA is present only at the  $\sim 1\%$  level, (perhaps much less). In other words, the role of xenon in EL is now non-existent, extracting negligible energy in collisions with energetic electrons as they gain energy up to the 4 eV range. Above 4 eV, collisional energy transfer efficiently extinguishes the electron population. Since the first excitation level of xenon is 8.32 eV, far above the EL range of TMA/DMA, the noble gas with TMA or DMA plays no role in EL [34]. There should be a range of electric field where strong TMA/DMA EL exists with no excess ionization. The range of electric field satisfying this requirement needs to be explored experimentally, varying the relative concentration of TMA or DMA in xenon, and total density  $\rho$ . Thus it should be possible to map out optimum conditions for EL in a small TPC. It could turn out that while DMA mixtures are also worth exploring, concern arises as to possibility of higher electronegativity in this gas since DMA is less symmetric.

What about the Fano factor  $F$ ? Will the optimum conditions for EL production be sufficiently broad that an overlap exists for minimizing  $F$  as well? This question remains unanswered. Experimental work, hopefully coincident with EL exploration outlined above, will be required. A direct manifestation of the Penning effect should appear as increased charge yield for the same energy input, with improved energy resolution relative to the pure gas. The energy resolution of x-ray lines below 100 keV should provide a direct measure of the improvement as long as EL production is high enough to provide good statistical precision in detected photons.

The reduction of the Fano factor remains speculative until measurements are made and optimal conditions are determined. In some mixtures, a Fano factor as low as 0.05 has been calculated [35]. Possibly then, a reduction factor of 2 is not unreasonable. For this value and the assumption that  $G = F = 0.075$ , the “sub-intrinsic energy resolution” is found:

$$\delta E/E = 1.9 \times 10^{-3} \text{ FWHM} \quad (8)$$

This value is of course quite idealistic, as other effects, such as Bremsstrahlung and energy gained/lost due to electrons traversing the drift field, present obstacles to realizing this goal. Nevertheless, any advance in energy resolution adds to the sensitivity of the search in the case that background is expected in the energy region of interest. In the case of  $^{136}\text{Xe}$ , the 2457 keV Q-value is quite close to a weak but dangerous  $^{214}\text{Bi}$   $\gamma$ -ray line just 10 keV below. It is at least conceivable that if resolution close to (8) is realized, then the discrimination against this background by energy resolution would be dramatically better.

Within the context of R&D for the NEXT-100 TPC, described at this Symposium, an experimental effort is being prepared to seek the optima for a Penning effect in xenon-TMA, and for EL in xenon-TMA. If the optima overlap well, a new pathway will emerge.

## 6. Discussion

Adding any molecule to xenon introduces profound changes for scintillation yields. With TMA present, the excitation population is extinguished by design. Hence the primary scintillation characteristic of pure xenon would be quenched. In this case, no fast signal will be available to provide the  $t_0$  signal needed by the TPC to place an event in space. Even if all other aspects turn out to be optimally beneficial, the complete absence of the  $t_0$  signal would make the goal of a Penning-enhanced EL TPC infeasible or unattractive. On the other hand, it is conceivable that not all of the excitation population is converted to ionization, and that some of the energy extracted by TMA is effectively wavelength-shifted to the more convenient 300 nm range. The understanding of remnant primary scintillation at either 175 or 300 nm is also an important part of the developing experimental effort within the context of the NEXT collaboration. That program will be described elsewhere.

Finally, the presence of complex molecules will reduce substantially diffusion of the primary ionization electrons during drift, even at the parts per thousand level. This is beneficial for event topology reconstruction. In the drift region, it is the lower-lying molecular levels that are important for cooling the electron population, not the excitation levels around 4 eV relevant to fluorescence. Some change in the drift velocity may also occur, but is not particularly important except for optimization of digitization rates for the tracking elements.

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