A negative-ion TPC with ultra-high energy resolution for 0-ν double beta decay search in $^{136}$Xe

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Abstract. Future searches for the neutrino-less double beta decay mode in candidate nuclei must confront the need for sensitivities at the level of $10^{-5}$ to $10^{-50}$ meV effective neutrino mass. Current techniques may not be able to scale simultaneously to the needed mass of active isotope with both improved energy resolution and much higher levels of background rejection. To address these severe challenges, a novel approach based on a negative-ion time projection chamber filled primarily with high-pressure $^{136}$Xe gas is developed. In addition to the provision of a nearly ideal active fiducial boundary surface, an energy resolution of $\sim 5 \times 10^{-3}$ FWHM appears possible at the Q-value of 2.48 MeV. The background rejection advantages of detailed track information and a single monolithic detector are addressed. A barium daughter-tagging scheme based on mobility differences and a new approach addressing the goal of efficient electron capture and release in high-density gas exploiting chemical reactions are introduced. Many R&D issues are suggested by this new concept.

1. Introduction

The discovery of neutrino oscillation in atmospheric, reactor, and solar neutrino experiments provides solid evidence that at least two neutrinos have non-zero mass. These results have greatly increased interest in the search for neutrino-less double beta decay since one of the requirements for this decay to occur is that the neutrino have non-zero mass. A positive result, at any level, would establish a Majorana nature of the neutrino and demonstrate the violation of lepton number. Measurements of the squared mass differences by the oscillation experiments and the recent global picture including cosmological constraints have established an upper limit for the sum of neutrino masses around 0.17 eV. For reviews of the current experimental and theoretical situation, see e.g., [1 - 4]. The sole claim for a positive 0-ν $\beta\beta$ result remains controversial [5 - 6].

It seems clear that a robust discovery of this decay mode, should it exist, may require detectors with active mass M of hundreds to thousands of kilograms of isotope in the detector. The realization of such increased sensitivity with corresponding reduction in backgrounds and improvements in energy resolution presents major technical challenges. Rejection of the various backgrounds in previous experimental approaches at the ~10 kg level has been arguably marginal to inadequate. For any candidate nucleus, the allowed 2-ν $\beta\beta$ decay mode occurs with a spectrum extending to the Q-value of the decay. This introduces an “intrinsic background” for which the only remedy is energy resolution sufficient to prevent overlap of the 2-ν $\beta\beta$ and 0-ν $\beta\beta$ populations. Beyond that, $\gamma$ rays and $\beta$ particles from radionuclides with high Q$^\beta$ such as the radon daughters $^{214}$Bi and $^{208}$Tl, can induce processes that may mimic $\beta\beta$ decay events in both topology and energy release.
Many attractive neutrino-less double beta decay candidate isotopes do not exist with useful natural abundances, and affordable centrifuge enrichment processes exist only for those elements that are naturally gaseous or can form gaseous chemical compounds. $^{136}\text{Xe}$ is one of the few candidate isotopes for which a large quantity of enriched material has been obtained [7].

Because of the extreme technical challenges for future $0\nu\beta\beta$ searches, many approaches have been considered, including various applications of the Time Projection Chamber (TPC) idea as the basis for experimental design [8 - 10]. High pressure xenon gas (HPXe) at a density of $\rho = 0.11 \text{ g/cm}^3$ (~20 bars) contains ~1000 kg in a cylindrical volume of 2.25 m diameter and 2.25 m length, suggesting that one single-ended HPXe TPC could contain a metric ton mass in a volume feasible for deep laboratory conditions. As an overall optimization has not yet been attempted, other considerations may require a different operating density or geometry.

2. Intrinsic Energy Resolution

The number of free electron/ion pairs liberated by an ionizing event with energy $E$ is

$$n = \frac{E}{W},$$  

(1)

where $W$ is the average energy to produce an electron/ion pair in the medium. $W$ decreases with applied electric field because more electrons are swept away from the attractive influence of parent ions, diminishing recombination. For pure xenon, the zero-field value is $W = 21.9 \text{ eV}$ [11].

The intrinsic fluctuation $\sigma_n$ in the number of electron/ion pairs at energy $E$ is

$$\sigma_n = \left(\frac{F E}{W}\right)^{1/2},$$  

(2)

where $F$ is the Fano factor for the detection medium [12]. The Fano factor reflects a constraint in the balance between energy losses to ionization and other processes, predominantly excitations; as the excitations are relatively much more numerous, their statistical uncertainty is relatively small. Hence, for a given energy, the intrinsic energy resolution is typically considerably better than the naive value neglecting the Fano effect (i.e., $F = 1$). In gases, $F$ ranges from low values such as 0.05 for gas mixtures with a Penning effect, to values from 0.13 to 0.17 for pure xenon [14 - 17]. At the $^{136}\text{Xe}$ Q value energy, 2.48 MeV, the zero-field number of electron/ion pairs $n = Q/W = \sim 1.13 \times 10^5$. Using the measured Fano factor $F = 0.13$ for pure xenon [14], the intrinsic fluctuation is not worse than

$$\sigma_n = \left(\frac{F Q}{W}\right)^{1/2} = \sim 121 \text{ electrons rms},$$  

(3)

leading directly to

$$\frac{\delta E}{E} = 2.5 \times 10^{-3} \text{ FWHM}$$  

(4)

This value is in accord, by extrapolation, with measured energy resolution studies at 667 keV [11].

3. Negative-ion TPC

In a conventional TPC, image transport is by free electrons, with high drift velocities up to ~10 cm/µs. Transverse diffusion of electrons during drift can be undesirably large, but may be highly suppressed by a strong magnetic field parallel to the drift field [18]. In the negative-ion time projection chamber (NITPC) concept developed by Martoff et al [19], electrons are captured by the electronegative

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1 On the other hand, due to the onset at high density of complex molecular physics associated with ionization-density dependent fluctuations in the competition between net ionization and recombination scintillation, Fano factors as large as ~20 have been observed in liquid xenon [13]. A strong anti-correlation of scintillation and ionization is observed, and both must be measured to determine energy.
molecule CS$_2$, with subsequent image transport by CS$^-$ ions. CS$_2$ was originally discovered in the 1940’s as a superior quenching agent for Geiger counters [20].

As ions remain thermal in the low electric field of the drift region, diffusion is reduced to the limit

$$\sigma = 0.07 \text{mm}(L/E)^{1/2}$$

(5)

(L in cm, E in kV/cm). For a typical drift length of 100 cm and an electric field of 4 kv/cm, $\sigma$ is ~0.35 mm rms. At pressures of ~1 bar, ion drift velocity is on the order of 100 m/s under these conditions. Very high spatial resolution is obtained, but at the price of relatively slow information capture. The low diffusion in a NITPC is of crucial importance in some applications such as nuclear recoil detection in WIMP searches. Two crucial aspects of the NITPC operation are the efficient capture of the ionization electrons in the drift region by interactions with electronegative molecules, and efficient stripping of electrons from the negative ions in a high-field electron multiplication region.

3.1 Adaptation of NITPC for ultra-high energy resolution

Here, instead of low diffusion, the slow image transport by ions in the NITPC is the fundamental advantage. In fact, the slowest ion transport consistent with other operational constraints is desired. This new method blends the NITPC concept with that of the Time Expansion Chamber, in which the electron clusters of primary ionization are detected by use of a very low drift field [21]. More precisely, in this new scenario the negative ion arrival times at the readout plane are sufficiently separated in time that each pulse generated as an individual electron is stripped from an ion is recognizably distinct from all other pulses. Each pulse can then be detected and counted in this ideal realization. An NITPC capable of counting each primary free electron liberated in the xenon gas by an ionizing event will approach the intrinsic energy resolution set by the intrinsic fluctuations in the conversion of energy to ionization. The NITPC exploiting single-ion counting is hereinafter referred to as the Ultra-High Resolution NITPC or UHR NITPC.

However, it appears that, even if the counting efficiency is significantly less than 100%, the energy resolution can still remain close to the intrinsic resolution. The reason for this is that if the fraction of counts lost is not large, the level of fluctuations in the lost counts is small relative to the intrinsic fluctuations in the number of electron/ion pairs in the total track. This assertion implicitly assumes that losses in counting efficiency are statistically constant and do not, for example, depend on event topology. While the important condition of statistical constancy of counting loss cannot strictly be true, the impact in practice may be negligible if steps are taken to spread out the ionization along the drift direction, and measurement of event topology has sufficient detail to permit adequate corrections.

An example is illustrative. Taking a density of 3 bar ($\rho = 0.11 \text{ g/cm}^3$), a typical reduced ion mobility of 2 cm$^2$/V-s, and an electrical field of 400 V/cm, an ionic drift velocity of ~ 40 cm/s is expected; the drift velocity of any candidate capture agent will likely differ by less than a factor of two from this value. Minimum ionizing tracks create ~6300 electron/ion pairs/cm in xenon of this density, leading to a average ion arrival rate of 2.5 $\times$ 10$^7$/s for track segments arriving perpendicular to the readout plane. If the local pulse pair resolution of the readout plane is assumed to be 50 ns and arrival times are assumed to be random at this rate, a pileup rate of ~6.3 kHz results. This leads to a predicted 2.5% counting inefficiency due to pileup. As double beta decay in xenon liberates ~1 $\times$ 10$^5$ electron/ion pairs, a 2.5% average pileup loss adds a fluctuation of only ~50 counts. Relative to the intrinsic fluctuation (3), a ~2.5% pileup loss has a surprisingly small additional impact. In this example, the energy resolution changes from 2.5 to ~2.8 $\times$ 10$^{-3}$ FWHM. In other words, the 2.5% loss worsens the energy resolution by only 0.3%. A lower HV may be closer to optimum, further reducing this loss mechanism.

Of course, track segments can arrive with any angle relative to the readout plane. The impact of variable track topology can be made less important, perhaps insignificant, by allowing electron capture to be prolonged, so that the ionic density spreads out along the field direction and is hence
approximately independent of track orientation to the field. This operational mode transforms the primary electron tracks into ionic curtains (see also section 4.1). Considerable flexibility may be available to optimize.

Taking $\varepsilon$ as the overall efficiency for counting each electron, then $\eta = (1 - \varepsilon)$ is the probability of not counting an electron. All loss mechanisms such as columnar recombination, unreleased electrons, finite electronic threshold, pulse pile-up, gain fluctuations, failure of ions to enter the amplification channels, charge sharing between pixels\(^2\), etc, can then be included in one loss parameter $\eta$. For simplicity, I make the simplifying and reasonable assumption (in gases) that any correlations between intrinsic fluctuations and all loss-inducing effects may be neglected. It is then elementary to show that an inefficiency, $\eta$, in counting individual electrons is equivalent to another, Fano-like, factor:

$$\sigma_n = ((F + \eta)E/W)^{1/2}$$  \hspace{3cm} (6)

The energy resolution is now

$$\delta E/E = ((F + \eta)W/E)^{1/2}/(1 - \eta)$$  \hspace{3cm} (7)

From this, it is clear that the UHR NITPC with high efficiency for counting primary electrons can approach the intrinsic resolution. If, for example, letting $\eta = F = 0.13$, \textit{i.e.}, a 13% counting inefficiency, a still excellent energy resolution

$$\delta E/E = 5 \times 10^{-3} \text{ FWHM}$$  \hspace{3cm} (8)

is predicted. This resolution, if realized, is a factor of 2 worse than (4), but only a factor of $\sim 4$ worse than that obtainable with germanium diodes at 2.5 MeV. The very mild dependence on $\eta$ offers strong motivation to exploit the UHR NITPC concept for any low-counting rate experiment where excellent energy resolution is important. Such a capability is directly relevant to future $0\nu\beta\beta$ searches, and could also offer benefits to nuclear spectroscopy, low background counting, or Compton $\gamma$ imaging.

3.2 \textit{Impact of Avalanche Statistics}

It is noteworthy that, for energy measurements based on analog sums of electron avalanche multiplication, a factor $B$ reflecting an additional variance due to gain fluctuations must be introduced in a similar way [16]:

$$\sigma_n = ((F + B)E/W)^{1/2}$$  \hspace{3cm} (9)

For cylindrical proportional counters, values of $B$ in the range $0.6 < B < 0.8$ were calculated for gains $10^2 < G < 10^5$ [22]. Gain structures with sharper transitions from drift to gain regions may be expected to exhibit somewhat lower values of $B$, but this remains to be demonstrated. Since $B$ is much larger than $F$, energy measurements with any analog system based on wire plane multiplication will be dominated by the gain fluctuations, and the potential benefit of a small $F$ value is lost. From (9), using $F = 0.13$ and $B = 0.7$,

$$\sigma_n = ((F + B)E/W)^{1/2} = 307$$  \hspace{3cm} (10)

\(^2\) Charge sharing between pixels can also lead to double counting; this, as well as any other over-counting effect can probably be included in the same $\eta$ parameter as a contributor to resolution. However, some gain structures can be geometrically matched to avoid depositing charge on pixel boundaries, eliminating charge sharing.
very close to \((Q/W)^{1/2} = 337\); most of the benefit of the Fano factor is lost. From (10), the implied energy resolution is

\[
\delta E/E = 6.4 \times 10^{-3} \text{ FWHM} \quad (11)
\]

However, for energy deposits in the 2.5 MeV range, the observed energy resolution of wire plane systems has been significantly worse than the prediction of (11)

\[
\delta E/E = 40 \times 10^{-3} \text{ FWHM} \quad (12)
\]

\[
\delta E/E = 66 \times 10^{-3} \text{ FWHM} \quad (13)
\]

A number of factors may also enter here, such as:

- **electronics**: noise, ballistic deficit, non-linearities and other calibration subtleties;
- **gain**: variations due to density and gas composition instabilities, gain map uncertainties, and head-tail space charge distortions;
- **other**: microphonics, loss of signal to electronegative contaminants, etc.

The more than one order of magnitude differences between the estimate (4) and experiment, such as (12) and (13), indicate the value of the opportunity that the UHR NITPC appears to offer for high resolution in the MeV \(\beta-\gamma\) energy range.

### 4 Factors affecting UHR NITPC energy resolution

#### 4.1. Impact of event topologies

Because thermal diffusion in any NITPC is small, and because portions of event tracks may arrive parallel to the readout plane, a strategy is needed to ensure that pile-up in arrival times is reduced to an acceptably low level. This can be realized by maintaining the capture agent at a sufficiently dilute level that capture is not instantaneous. Before capture, the electrons will drift at cm/\(\mu\)s speeds, and diffuse both longitudinally and transversely. If the effective capture length is arranged to be roughly equal to the readout plane pixel size, then the arrival time distribution of the negative ions will be roughly independent of track orientation. In this scenario, the electron tracks are converted into “curtains” drifting toward the anode plane. The use of a dilute capture agent will also reduce geminate recombination losses since the electrons are able to drift rapidly away from their ionic parents and neighbors. The positive ion tracks are not affected (see section 6.3). The impact of the curtain effect on track reconstruction should be minimal. It also could turn out the impact of cluster formation could be turned to advantage, leading to such very low mobilities that pile-up is negligible.

#### 4.1.2 Recombination effects

Consistently, improved energy resolution and increased net ionization are observed as density is reduced and electric field is increased [11]. These effects are attributed to recombination phenomena, particularly strong in locally dense clusters of ionization in \(\delta\)-rays. There exists a sharp threshold density at \(\rho \sim 0.55 \text{ g/cm}^3\), beyond which non-Poissonian fluctuations in net ionization become increasingly prominent. At the density of LXe, \(\rho \sim 3.06 \text{ g/cm}^3\), recombination effects introduce a very strong anti-correlation between net ionization and scintillation at any practical electric field [13]. Subsequent columnar ionic recombination could be an issue, particularly for track segments parallel to the applied field, and needs more study; NITPC studies with \(\alpha\) particle emitters would make these effects much more prominent. The detailed track reconstruction will make corrections possible. On balance, the use of a dilute capture agent is motivated by this concern as well.

#### 4.1.3 Margin and resolution shape

While it could be argued that such ultra-high energy resolution is not needed to reach somewhat beyond the present neutrino effective mass sensitivity, a counter-argument maintains that a margin of safety here is not only prudent but perhaps essential since the energy resolution in xenon will display, at the minimum, non-gaussian characteristics on the low energy side due to Bremsstrahlung if those energy losses are not recovered. Unanticipated resolution effects on the high energy side may become evident only after a large-scale experiment is operated for
a substantial time. As noted above, there appear to be very few, if any, examples in the literature where extended $\beta$ tracks (in the MeV range) in either an ionization chamber or gas detector with proportional gain have been measured with energy resolution approaching intrinsic values. In HPXe ionization chambers, resolution is good at sub-MeV energies, where event size (track length) is not too large. But at low sub-MeV energies (<100 keV), electronic noise and microphonics typically dominate the observed resolution. By counting individual electrons, one eliminates entirely these conventional resolution-limiting noise sources.

4.1.4 Capture of event energy. With the assumption that the UHR NITPC provides detailed 3-D event topological reconstruction, it should be possible to detect and associate small energy deposits carried away from the $\beta$ tracks by bremsstrahlung photons but converted in the xenon nearby. From this perspective, the larger the detector, the better will be the capture of all satellite energy deposits, except for those very large bremsstrahlung losses that escape to the periphery. If so, the energy resolution function will be narrow around the Q value, but will also possess a small but long tail to low energies. Except for a small loss of efficiency for larger bremsstrahlung, this non-gaussian resolution effect should be rather benign.

4.1.5 Loss mechanisms. To obtain a very low ionic drift velocity, the drift electric field has to be small. Thus, the ratio of the amplification field to the drift electric field will be sufficiently high that the loss due to ions failing to enter the high-field channels will be small compared to $F$, for any likely gain structure. However, a more subtle mechanism is the possibility that the higher ionic temperatures in the amplification region excite dissociation reactions of the capture agent; the reaction products may exhibit deeply bound states of the electron, such that release does not occur.

5 Reconstruction of event candidates

The capabilities to reconstruct complex, dense events with sub-mm tracking resolution and to place events in space are hallmarks of the TPC idea, and these capabilities, along with excellent energy resolution, should provide the means to distinguish most false events from true events. However, the 3-D placement of events is only an automatic feature of the TPC concept as long as some robust mechanism, such as a scintillation signal or some other equivalent information, exists to provide an event time origin.

5.1 Event time origin, $t_0$. In pure xenon, gas or liquid, copious scintillation occurs. In HPXe, the scintillation phenomenon yields a pressure-dependent spectrum centered about 7 – 8 eV. The time dependence is complex, reflecting competition in the underlying molecular processes [24]. In any case, most of the scintillation light typically occurs within 100 ns, fast enough to provide a natural $t_0$ signal. Use of this signal is surely preferred to other possible schemes. However, the presence of a small amount of a complex molecule such as methane is needed to stabilize avalanche gain. Methane is attractive because it does not absorb xenon scintillation, but even at a low 0.23% concentration, methane appears to quench primary scintillation by 25% [25]. In the UHR NITPC, the presence of the capture agent could also compromise the scintillation yield, either through quenching of the primary scintillation process, or by secondary absorption of the UV photons. However, the needed capture agent density is likely to be far below 0.01%. Overall, it seems possible that an operational regime can be realized such that most of the potential primary UV scintillation will survive and reach the detector periphery, where scintillator bars with appropriate wave-shifter components can provide efficient UV detection, as well as capture of higher energy bremsstrahlung. $N_2$ is known to provide efficient wave-shifting characteristics in argon gas, and may do so also in xenon.

5.2 Spatial resolution. Although the primary data is digital, the integrated count per pixel as a function of time leads to a very high “analog charge” information quality for the purpose of spatial reconstruction. The sharing of the “charge” on adjacent pixels by direct projection and by diffusion provides superb intrinsic spatial resolution. The event tracks typically provide $\sim10^7$ electrons/mm, and with average diffusion less than 2 mm, the intrinsic spatial resolution is on the order of 50 $\mu$m rms, per
mm of track. Since multiple scattering in the xenon is large, however, there is little motivation to exploit tracking to the intrinsic limit. Pixel size can be chosen to match the number of points needed for topology reconstruction, likely on the order of 50 for an event with nearly full energy. This suggests a pixel size on the order of 4 – 6 mm. Despite the coarseness, track spatial resolution will typically be sub-mm except for the more convoluted segments near the endpoints.

6. Background rejection
6.1 A seamless, continuously sensitive, virtual fiducial volume surface
With 3-D placement of events, this closed surface can be defined to exist within the sensitive TPC volume and enclose nearly all the active mass. Tracks passing through this surface would be identified as either compromised true events leaving the active volume or false events originating outside the active xenon gas, likely from a structural element or electrode surface. While first-pass data analysis may be done blind, with set boundary conditions, the fiducial volume surface may be adjusted at will ex post facto to understand sensitivities. Electronic dead-time should be negligible in this low rate environment. Previous experiments based on solid-state detectors have suffered due to the presence of partially dead or insensitive surfaces, or more accurately, the complete absence of a genuine fiducial surface. This is not the case for the UHR NITPC, as the fiducial surface is closed, precise, dead-timeless, and completely active.

6.2 Other stratagems
6.2.1 Energy resolution. If the high resolution asserted above can be realized, then the narrowness of the region of interest will reduce directly any residual continuum background.
6.2.2 Magnetic field. The presence of a magnetic field, in the absence of multiple scattering, would induce an S-shaped curvature onto the topology of a genuine double beta decay event (or any other process that creates two energetic electrons); pair production by γ-rays would take on a C-shaped curvature. As multiple scattering in xenon is quite pronounced, the large random deflections can only be overcome by fields above ~1 kG. Very large magnetic fields curl events to ribbons with cm-scale transverse dimension, complicating track reconstruction but improving containment within the fiducial volume. Algorithmic development to discriminate against large, clearly non-magnetic deflections may show that moderate fields provide useful additional constraints against confusion with pair production by energetic γ-rays, beyond that provided automatically by conversion of positron annihilation γ-rays. There would be no requirement that the magnetic field uniformity match that of the electric drift field.
6.2.3 Charged particles. No β or other minimum-ionizing particle can pass through the fiducial surface undetected. Protons, α particles, and other heavily ionizing tracks are easily distinguished from β’s by their very different signatures.
6.2.4 Radon daughters. The heavily ionizing radon α decay will never be confused as a β. The heavy element daughters will be plated out at the cathode within seconds, since they are almost certain to be ionized at birth. Subsequent decays into β particles will be detected as false events.
6.2.5 Neutral particles. γ-rays originating from radio-nuclides in the detector components and the decays of the plated-out radon daughters into γ-rays will be problematic, and the techniques to purify materials and aggregate experience of the field will be essential. Neutrons can in principle lead to short-lived radioactive species within the fiducial volume, and will also need study. Since true ββ decays typically have strong scattering at both endpoints, their topologies will differ from most single β decays, and the detailed imaging can be expected to help. Photoelectric conversion in xenon of background γ rays will lead to K-shell fluorescence with 85% probability; these false events can be distinguished by a characteristic satellite energy deposit of ~30 keV.

6.3 Barium daughter tagging
The barium daughter, whether singly or doubly charged, will display a mobility different from that of xenon ions, due to the phenomenon of resonant charge exchange (RCE) [26]. RCE occurs efficiently
for ions of the majority component, even for large impact parameters, since there is no energy penalty for charge exchange and collisions are frequent. The effective size of xenon ions is increased, inducing a higher viscosity for the xenon ions. Low-pressure measurements suggest that the difference between xenon and barium mobilities would be ~40%, a very large effect [27]. If the cathode plane surface is prepared such that the arrival of the barium ion leads to prompt, efficient emission of at least one electron, the ionic return of this signal to the readout plane, and subsequent detection there, will comprise an “echo” signal. Detection of this echo pulse should provide a very robust tag for \( \beta \beta \) decay because of the very precise timing and spatial characteristics of the echo pulse. The arrival time of the signal caused by the barium daughter will, in general, be very well separated from echo pulses of xenon ion track. This idea remains unexplored at present. Clustering of xenon atoms around ions may completely alter this scenario, but such effects remain undetermined at the densities of interest [28].

The barium daughter is likely to be created in a highly ionized state [29]. Electrons will be captured rapidly from xenon neighbors to yield a doubly ionized state at 10.004 eV. If impurities with ionization potentials lower than 10 eV are kept sufficiently low, the doubly charged barium daughter will survive the journey to the cathode. The cathode plane surface would likely need to be prepared with a high emissivity surface. Such a cathode may also act as a photocathode for primary scintillation; a wave of “photo-anions” would arrive at the readout plane after the event is collected.

7. Good scaling behavior

With the assumption that performance goals at the active mass levels characteristic of a small prototype are met, the performance expected for large-scale implementations can be extrapolated with confidence. A schematic illustration of a vertically oriented ton-scale system is shown in figure 1.

7.1 Density, size, and surface-to-volume ratio. Contrary to those experiments made of numerous small modules, the ratio of fiducial surface to active volume for the UHR NITPC improves automatically with increasing scale. The contamination of the active volume by penetrating \( \gamma \) backgrounds is reduced with increasing scale. For a single cylindrical geometry of length \( L \) and radius \( r \), the minimum surface/volume ratio occurs when \( L = 2r \), a natural TPC geometry. A single-ended TPC design with a vertical axis of symmetry may provide a convenient construction/maintenance scenario. Furthermore, from simple dimensional considerations, for a given active mass and taking the detector as a cube of dimension \( L \) and event track length as \( \lambda \), the ratio \( \lambda L \propto \rho^{-2/3} \). This shows that (as long as \( \rho < 0.55 \text{ g/cm}^3 \)) higher pressures lead to a smaller region where true events may be compromised by passing through the fiducial surface. Thus, for a constant mass thickness of xenon outside the fiducial surface, the fraction of isotope within the fiducial volume increases with higher density.

Finally, neon can be added to reach an optimum density if the available mass of active isotope is insufficient. A neon-xenon mixture will display a beneficial Penning effect, reducing the Fano factor.

7.2 Maximum drift length and high voltage. Diffusion, both transverse and longitudinal, is at the thermal level, and is small for present purposes and scales slowly, as the square root of drift distance \( L \); rather long drift distances can be envisaged without significant degradation of information. For the same reason, the number of electronic channels triggered at the readout plane for a given event will be nearly constant with scale. Drift region high voltage (HV) scales linearly with \( L \), and for a ton-scale example in section 1, the HV for the 2.25 m drift length would be 90 kV.\footnote{While linear HV scaling is obvious in the gas TPC, for semiconductors such as the \(^{68}\text{Ge} \) diodes for 0-\( \nu \beta \beta \) searches, the depletion voltage needed depends quadratically with drift distance, limiting the size of the modules in these systems.}

7.3 Readout plane. The need to maximize counting efficiency translates to an optimization process for pulse-pair resolution (high electronic bandwidth and fast detector pulse characteristics), high detection efficiency and the ability to distinguish events from background noise.

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\footnote{The electron emission probability is greatly enhanced if the ionization potential of the incident ion is at least twice the work function of the surface [30], and if the incident ion is multiply charged. From these perspectives, the doubly charged Ba\(^{++} \) daughter will likely be much more efficient than the singly charged ion Ba\(^+ \).}
efficiency (good pulse height for single electron pulses and minimum electronic noise to achieve a low threshold for an acceptable power budget), etc. This optimization has not been done, but conservatively, these goals suggest that the readout plane will need to be pixelized, with one channel per pixel. However, the pixel size can be rather large, perhaps 4 – 6 mm diameter. Such a low channel density should facilitate implementation. The number $N$ of readout plane electronic channels is modest, $\sim 4 \times 10^4 \text{m}^2$; $N$ scales linearly with the readout plane area and hence as $M^{2/3}$.

**Figure 1.** A schematic illustration of a ton scale system. The cylindrical active volume is 2.25 m diameter and 2.25 m height. The major components are:

- a. Sensitive volume filled at ~20 bars with xenon + capture agent mix;
- b. Field cage, comprised of rings to establish uniform equipotential surfaces;
- c. Cathode plane, at negative HV;
- d. Neutron absorber, HV insulator, and filler to force xenon into active volume - possibly polyethylene;
- e. HV module, or feedthrough;
- f. Plastic scintillator or wave-shifter bars to convert UV scintillation for event start time signal;
- g. HV insulator, neutron absorber, and filler, as in d;
- h. Readout plane, with gain structures and front-end electronics;
- i. Annular ring supporting service feedthroughs and data flow;
- j. Neutron absorber, and filler.

### 8. Implementation: R&D Issues

The R&D issues are numerous and interconnected, but are in many respects rather straightforward.

#### 8.1 Preservation of the scintillation signal

In addition to the capture agent, signal amplification by electron avalanche multiplication requires a molecular additive to avoid Geiger-like unstable behavior in the readout plane gain stage. The need to realize unattenuated 170 nm UV propagation for a $t_0$ signal is antagonistic to the purpose of stable gain. However, it may be possible to operate in a regime where both can be realized. Methane has very little UV absorption below 9 eV because it has only C – H bonds, which display a characteristic absorption band at 125 nm. So the measured losses seen in [25] are attributed to quenching of primary scintillation. At 0.23% methane, however, the loss is only ~25%, dependent to some extent on applied field. As the absolute density of 0.23% methane at 26 bars is equal to that of a ~6% mixture at 1 bar, and since good gain behavior can be obtained with such a mixture at 1 bar, it may be that a 0.23% fraction of methane may be more than sufficient to permit both UV detection for $t_0$ signals and stable readout plane gain in a large scale device.
However, any hydrocarbon may interfere with the electron regeneration scheme outlined in section 8.2.3, and any molecule with C–C bonds will absorb the primary scintillation. Candidate capture agents will pose a dilemma if their UV absorption is high above 6 eV; CS$_2$ is in this category.

8.2 Capture agent

Electron capture, ion transport, electron release in practical electric fields, and needed gas gain all must be obtained simultaneously in HPXe with a capture agent of optimal electron affinity (EA), acceptable toxicity and flammability, good chemical stability and limited aggressiveness, and minimal UV quenching and absorption. While electron capture has been studied theoretically and experimentally in a wide range of substances and conditions, detachment processes in the NITPC context are less well understood. The conventional view of release is that the negative ion must gain kinetic energy in the high E/P region until a collision sufficiently exceeds the binding energy.

The processes governing capture and release in CS$_2$ are complex. The EA of CS$_2$ is double-valued, 0.6 eV and 0.9 eV, depending on a bent or linear ionic molecular structure; both can coexist, with relative populations depending on temperature [31]. With such high EA value, it seems evident that the CS$_2^-$ ion must somehow reach very high temperatures, as the Arrhenius factor for the CS$_2^-$ ion, $\exp(-0.9/0.025)$, is roughly $2 \times 10^{16}$. NITPC operation at higher gas density, 700 Torr (CS$_2$ 200 Torr plus He 500 Torr), has been explored; results indicate a large decrease in ionic mobility between total pressures of 200 to 700 Torr [32]. This unexplained effect could arise from the onset of clustering. From all these perspectives, CS$_2$ presents some serious limitations. For the high densities of interest here, it seems likely that a new capture agent with much smaller electron affinity (EA) may be essential. An EA of perhaps $\sim 6 – 18$ kT may be ideal.

Other effects may enter. The presence of molecular levels may permit excitations in the ion to become populated, acting as a staircase to release. The ionic polarization energy of the ion in the electric field of the amplification region may also increase the chance of release through the creation of energy levels accessible by collision (however, the field needed for rapid tunneling in vacuum exceeds 2 MV/cm [33]). The release may be complicated by, or conceivably even assisted by, the formation of clusters in HPXe around ions [28].

There appear to be four roughly basic categories, depending on the EA:

a. EA $\sim 1 – 4$ kT “Equilibrium”

b. EA $\sim 6 – 18$ kT “Ideal”

c. EA $\sim 20 – 60$ kT “Inhibited”

d. EA $> \sim 60$ kT “Unsuitable”

8.2.1 Category a: “Equilibrium”. For capture agents in category a, a cascade of capture/release/capture/release cycles occurs at a high rate during thermal agitation, and the ratio of time spent as free or ionic will depend sensitively on the EA and molecular and ionic energy levels. The capture rate is tunable by capture agent density and gas temperature. If circumstances can be arranged such that the ionic lifetime is, e.g., 10,000 times longer than the free electron lifetime, then the apparent drift velocity would be very slow, resembling that of ionic drift, but actually be almost entirely due to drift as a free electron. Good operation in the amplification region would obtain only if the mean interval between release/capture is a small fraction of the thickness of the amplification region, or if a slight increase in thermal agitation due to the high field is sufficient to initiate release. Although operating with a relatively low electric field would offer a practical advantage, geminate and columnar recombination would increase.

As two possible examples in category a, nitric oxide, NO, has an EA of $\sim 0.026$ eV, and nitrous oxide, N$_2$O, has a positive energy state with an activation energy of $\sim 0.03$ eV [34]. A strong dependence with gas temperature of the apparent electron capture rate in N$_2$O has been observed [35,36], but this phenomenon is actually reflecting a different process: N$_2$O + e$^-$ $\rightarrow$ 0.21 eV $\rightarrow$ N$_2$ + O$^-$. The conversion of a free electron to a negative atomic oxygen ion places the process in category e
because the EA of $O^-$ is 1.465 eV. Unfortunately, $N_2O$ has strong VUV absorption in the 160 – 180 nm band [37].

8.2.2 Category b: “Ideal” Here, capture consistent with the curtain effect is obtained by optimizing capture agent density. Once an electron is captured, the EA of the ion is sufficient, just barely, to maintain ionic integrity during drift; some re-ionization/recapture would not be harmful, and may be helpful. When the high field amplification region is reached, ion kinetic energy increases somewhat, and release is immediate; an avalanche commences without loss to further capture. It is not yet clear whether this scenario will hold at the high densities appropriate to the double beta decay search, nor is it clear at which EA all this would obtain.

Oxygen may come close to falling in this category as the EA of $O^-$ is 0.46 eV or 18.4 kT. However, if the reaction $O_2 \rightarrow O^- + O$ occurs, the oxygen ion has an EA of 1.465 eV and now falls in category c. Acetylene has an EA of 0.49 eV and may represent another interesting candidate.

8.2.3 Category c: “Inhibited” Here, the highly non-thermal agitation needed for re-ionization is sufficiently large that the needed electric fields at high densities become difficult to sustain. For the present purpose CS$_2$ may fall in this category, since it is only known to work at 40 Torr. However, there appears to be another way to deal with this, at least in some cases, by the provision of chemical energy. This new notion for UHR NITPC operation is based on gas-phase chemical reactions, exploiting chemical energy to regenerate free electrons (see Appendix). The electron freed by a chemical reaction is subsequently captured again, producing a new anion. A cascade of release/capture cycles occurs as the electron/anion drifts toward the amplification region. If the mean drift length as a free electron is sufficiently small, this process would ensure that electrons are freed in the high field avalanche region early enough, so that well-behaved amplification will occur. The total fraction of time spent as free electrons must be sufficiently small that the effective drift velocity will resemble ionic mobility.

8.2.4 Category d: “Unsuitable” Here, the EA is very large, and no chemical reactions are available to provide the efficient conversions needed to maintain availability of free electrons. SF$_6$ appears to fall in this category. Although it has an attractive positive-energy two-body capture process at thermal conditions, subsequent collisions can lead to trapping in a deep state at 1 eV. Dissociation reactions such as $SF_6 \rightarrow SF_5 + F$ lead to even more deeply bound states at several eV.

8.3 Ion mobility and drift velocity
There appears to be little data for ion mobility in xenon at the densities of interest. The onset of clustering in xenon at some density is expected [38]. R&D for this topic would include:
1. Measurements of positive and negative ion mobilities as a function of xenon density, for all candidate capture agents;
2. Determination of uniqueness or non-uniqueness of mobility as a function of xenon density;
3. Exploration of the impact on clustering by addition of neon;
4. Exploration of the lowest possible drift velocity consistent with track integrity.

8.4 Electron multiplication
Advances in gas amplification structures such as the Well [39], Micromegas [40], gaseous electron multiplier (GEM) [41], and large electron multiplier (LEM) [42] devices offer the prospect of fast signals and adequately high gain. The Well and Micromegas will likely offer the fastest signals, but the very high spatial resolution possible with GEM and micromegas devices is not needed. The LEM may also offer operational advantages, such as high gain in one stage and a possibility to match pixel boundaries to the hole pattern such that charge-sharing is avoided. It may be possible to combine the LEM and Well geometries directly to realize a robust high-gain structure. As noted, it seems reasonable to expect that 0.2% methane, or even a wide range around this value, might work adequately. The combination of agents needed for good capture and release behaviors might be sufficient without methane. R&D for this topic could include:
1. Multiplication by avalanche in GEM, LEM, micromegas;
2. pulse height spectrum optimization
3. maximum gain limits
4. intrinsic pulse-pair time resolution limits
5. range of quencher concentration providing stable gain
6. multiplication by photo-conversion of field-induced scintillation, without quencher.

8.5 Electronic channel
Fast shaping, together with lowest practical electronic noise, small threshold margins, and power dissipation drive the design. Each electron pulse leads to an increment of +1 in an internal counter. If the ion drift velocity is 0.5 m/s, time-stamping each sum at ~2 kHz provides track sampling at sub-mm resolution. At a density of \( \rho = 0.1 \text{ g/cm}^3 \) (~20 bars), a minimum ionizing particle will liberate about 6000 electron/ion pairs/mm; their arrival rate is roughly \( 2.5 \times 10^5 \text{ /s} \). If the pulse-pair resolution is, e.g., 50 ns, pile-up losses occur at the ~2.5% level. The counter will typically hold about 125 counts per time slice, at most ~20 kbytes/s per active channel. Corrections to the pile-up counting loss can be made, including the possibility to measure time-over-threshold, or even waveform capture. Only those channels with counts above zero would be read out, and with such a low data rate the data acquisition system is not a challenge. Even slower ionic drift velocities and/or lower ionization density (less pressure) may turn out to be optimal.

9. Summary and perspective
The idea of using a NITPC designed to count individual negative ions, admitting less than 100% efficiency but still with the goals of ultra-high energy resolution and excellent track reconstruction, appears to be both attractive and novel. Possible applications include the search for \( 0^- \beta \beta \) decays, Compton \( \gamma \) imagery, and low-rate \( \gamma \) spectroscopy. Many technical issues remain to be explored, such as the scintillation yield possible within the constraints of electron capture, detachment, and electron avalanche multiplication in HPXe in the presence of a capture agent. Methane is likely to be the only hydrocarbon transparent to the primary scintillation, but would be problematic if atomic oxygen ions are produced. New capture agents are likely essential. Gas-phase chemistry may offer new opportunities for capture, transport, and release through regeneration. Electronic issues appear manageable, and the lack of an ideal ASIC is not seen as an impediment for most R&D goals. Although speculative at the present level of knowledge, barium daughter-tagging through detection of mobility differences may be possible.

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Appendix: Gas-phase chemistry
Gas-phase chemistry may open new possibilities for both NITPC and UHR NITPC operation. Ionic mobility studies have been frequently confounded by undetected impurities that, even at extremely low levels, initiate reactions that dominate final reaction products. Mass spectrometry of the migrating species has often been essential to the resolution of conflicting results. In the UHR NITPC context, \( \text{O}_2 \) and \( \text{N}_2\text{O} \) are two illustrative but still incompletely understood examples.

A.1 Oxygen. Molecular oxygen is electronegative, non-toxic, and safe at the presumably very low levels needed. The basic attachment processes have been the subjects of many experimental and theoretical studies [43-46]. In dense gases, the negative ion \( \text{O}_2^- \)

\[
\text{O}_2 + e^- + \text{M} \rightarrow \text{O}_2^- + \text{M}
\]  

(A.1)

is created in a multi-step process with the participation of another molecule to transfer energy and momentum. \( \text{O}_2^- \) has an EA of 0.46 eV - about 18 kT [34], and this value appears to place oxygen in the category b, the ideal case. Another attachment process, the dissociative reaction
\[ O_2 + e^- \rightarrow O^- + O \]  \hspace{1cm} (A.2)

requires 3.6 - 4 eV of electron energy [47,48], and is improbable during drift, as free electrons would be near thermal. Once \( O_2^- \) is formed, the dissociation reaction

\[ O_2^- \rightarrow O^- + O \]  \hspace{1cm} (A.3)

can also lead to \( O^- \) production. As (A.3) is endothermic with an activation energy of 1.005 eV, this reaction is also unlikely during drift. However, (A.3) could become prominent in the high field amplification region. As the EA of the nascent \( O^- \) is very high, 1.465 eV, \( O^- \), once formed, is likely to stay permanently bound. Therefore, to the extent that (A.2) or (A.3) do occur anywhere, these reactions would represent a direct loss of primary electrons.

However, gas-phase chemical reactions may be introduced to supply energy, releasing the captive electron. If molecular hydrogen or carbon monoxide is also added, the exothermic reactions

\[ O^- + H_2 \rightarrow H_2O + e^- + 3.5 \text{ eV} \]  \hspace{1cm} (A.4)

\[ O^- + CO \rightarrow CO_2 + e^- + 4 \text{ eV} \]  \hspace{1cm} (A.5)

will regenerate the electron again [48-50]. As oxygen is a minor component, the kinetic energy of the regenerated electrons will likely dissipate through collisions, rather than lead to further reactions of process (A.2). CO appears to be about an order of magnitude more efficient than \( H_2 \) in promoting detachment, while NO appears to be about an order of magnitude less efficient [49]. The \( CO_2 \) and \( H_2O \) products should be benign for the UHR NITPC. Other reactions, such as

\[ H_2 + O_2^- \rightarrow H_2O + e^- + O \]  \hspace{1cm} (A.6)

\[ \rightarrow H_2O + O \]  \hspace{1cm} (A.7)

may also compete, but would not represent a loss mechanism if (A.4) and (A.5) are effective. However, reactions leading to hydroxyl groups can also occur, such as:

\[ H_2 + O_2^- \rightarrow OH^- + H + 0.16 \text{ eV}. \]  \hspace{1cm} (A.8)

Although (A.8) is slightly exothermic, the production of hydroxyl radicals appears to be nearly absent in pure \( H_2 \) [48]. The EA of the \( OH^- \) is very high, 1.828 eV. If \( OH^- \) blocks electron regeneration in \( Xe + \text{dilute} \,(H_2 + O_2) \), even if hydroxyl production is very low, a gradual decay of signal would occur as the drift cascade continues. In that event, perhaps only \( CO \) or \( NO \) would be suitable agents for electron regeneration. The possible interactions of \( O_2^- \) with \( CO \) and \( NO \) need further understanding, as \( CO_3 \) and \( NO_3 \) have EAs of 2.69 eV and 3.937 eV, respectively, and formation of these species would be fatal to further regeneration.

While novel, the possible exploitation of gas-phase chemistry at this point leaves the basic question unanswered: is there an optimum mixture of \( O + (H_2 + CO + NO) + Xe \), such that electron regeneration is efficiently maintained while providing a drift velocity characteristic of ions?

**A.2 NO.** In the case of nitrous oxide, electron attachment/detachment processes relevant to UHR NITPC functionality are even more complex than those for \( O_2 \) [35,50,51]. A state \( NO^- \) with an activation energy of \( ~0.03 \text{ eV} \) has been recently predicted [52], resolving some experimental inconsistencies. This state is produced in a three-body process:

\[ N_2O + e^- + 0.03 \text{ eV} + M \rightarrow N_2O^- + M \]  \hspace{1cm} (A.9)

\( N_2O^- \) is unstable against detachment at thermal energies. It is conceivable that (A.9) provides a natural attach/detach process for UHR NITPC operation at some accessible temperature. The mobility of thermal electrons in \( N_2O \) has apparently not yet been measured.

A second important channel is the endothermic dissociative attachment reaction

\[ N_2O + e^- + 0.21 \text{ eV} \rightarrow N_2 + O^- \]  \hspace{1cm} (A.10)

As expected, (A.10) occurs more readily if the gas is hot or if electrons are heated in the gas [35]; the attachment rate increases by several orders of magnitude as the mean electron energy rises from thermal to \( ~2 \text{ eV} \) [50]. With the electron regeneration implied by (4) and (5), the loss mechanism implied by reaction (10) may be preventable. Again, the basic question remains unanswered: is there an optimum mixture of \( N_2O + (H_2 + CO + NO) + Xe \), such that electron regeneration is efficiently maintained while providing a drift velocity characteristic of ions?
A.3 Hydrocarbons. In gases with a hydrocarbon component, represented as RH, the reaction
\[ \text{O}^{-} + \text{RH} \rightarrow \text{OH}^{-} + \text{R} \]  
blocks further electron regeneration \[51\]. As noted, the EA of the OH\(^{-}\) is high, 1.828 eV, implying a permanently bound state in the UHR NITPC if no electron regeneration reactions involving a hydroxyl radical are possible. This implies that the use of methane as a quencher for avalanche gain may be infeasible for \(\text{N}_2\text{O}\) or \(\text{O}_2\). More complex hydrocarbons have numerous other charge-transfer dissociation reactions that would likely proscribe their use as well. However, adequate gain may be possible without the use of methane or other hydrocarbon.

A.4 Barium daughter tagging and charge exchange. \(\text{O}_2\), \(\text{N}_2\), \(\text{CO}\), and \(\text{N}_2\text{O}\) have ionization potentials (IP) of 12.0697, 15.58, 14.014, and 12.886 eV, respectively, and would not reduce \(\text{Ba}^{2+}\) ions (IP = 10.004 eV) to the singly ionized state, whereas NO, with an ionization potential of 9.26 eV, would do so \[34\]. The presence of dilute \(\text{O}_2\) in xenon (IP = 12.1298 eV) or NO will likely lead to charge exchange reactions with \(\text{Xe}^+\) ions. As minority ions, oxygen and nitric oxide will likely frustrate the barium daughter-tagging scheme based on mobility differences. From these considerations, only the \(\text{N}_2\text{O} - \text{CO}\) combination would maintain the desired mobility differences. There may, of course, be other suitable molecular candidates not yet discerned.

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