Detecting the barium daughter in ^{136}Xe 0-$\nu\beta\beta$ decay using single-molecule fluorescence imaging techniques

David R. Nygren

Department of Physics, University of Texas at Arlington
Box 19059, Arlington, TX 76015

nygren@uta.edu

Abstract. Single-molecule fluorescent imaging may provide an avenue to efficiently detect the Ba^{++} daughter atom in the decay ^{136}Xe $\rightarrow$ Ba + 2e$^-$, and, unambiguously associate the birth point in space within the electron trajectories of the decay event. Chelation of doubly-charged alkaline earth elements such as calcium and barium by certain precursor molecules converts the resulting complex from a non-fluorescent to a fluorescent state. Repeated photo-excitation of a single fluorescent complex reveals both presence and location with high precision. This technique, widespread now in biochemistry, biophysics and biology, may permit a similar discriminating response in a large high-pressure xenon gas TPC for the Ba^{++} ion from xenon double-beta decay. The TPC measures the event time and energy of the two nascent electrons, as well as topology and position in 3-D from their trajectories in the gas. Measurement of the 2-D location of the molecular ion after arrival at the cathode plane permits an association of ion with the event. Demonstration of an efficient, highly specific detection of the barium daughter would provide a long-sought pathway to a background-free result in the search for this decay mode, of central importance for determining the nature of the neutrino.

1. Introduction

Observation of neutrino-less double beta-decay in any of several candidate nuclei will establish that the neutrino is its own anti-particle, a unique property among spin 1/2 particles, and that lepton number is not conserved [1]. Any observation of this decay mode implies a natural and attractive connection to very high masses and a plausible mechanism for the baryon-antibaryon asymmetry of the universe, physics well beyond the Standard Model. The known neutrino mass parameters establish for the inverted mass ordering hierarchy an effective neutrino mass $\langle m_{\beta\beta} \rangle$ range of 20 - 50 meV, corresponding to possible decay lifetimes T in excess of $\sim 10^{27}$ years. Recent results [2] for ^{136}Xe have established lower limits of $\sim 2 \times 10^{25}$ years, corresponding to sensitivities of approximately $150 < \langle m_{\beta\beta} \rangle < 280$ meV. These results are sufficient to rule out a controversial earlier result for ^{76}Ge, but include contaminating backgrounds and make no discovery claims. Such long lifetimes present daunting experimental challenges. To probe deeply into the range of the inverted mass ordering hierarchy, active masses approaching or exceeding ton-scale are necessary.

In addition, substantial technical advances to reduce backgrounds are essential. In a background-free experiment, sensitivity to the neutrino effective mass increases slowly, as (run-time)$^{1/2}$. However, if the probability of even *one* background event is significant, sensitivity evolves toward (run-time)$^{1/4}$. As soon as background is present in an experiment, further running adds marginal sensitivity; further

scientific reach is effectively blocked. The natural sensitivity goal is to span the inverted mass ordering range, *i.e.*, to reach ~20 meV. If background is demonstrably zero, only about 1 ton-year is needed. But if backgrounds in the true event class reach 10 events/ton-year, then an exposure of about 100 ton-years is needed for 90% confidence, illustrating the catastrophic impact of background. Quantitatively, one might arguably take *background-free* to mean that the probability P for any background event satisfying all true event criteria in one year of running must be not greater than, say, 6%, or possibly as low as 1%; I'll choose 3%. At the ton scale, P = 3% for background contamination of the energy ROI corresponds to $P \leq 3 \times 10^{-5}$ counts/kg-year. With 1% FWHM energy resolution this corresponds approximately to $P < 1 \times 10^{-6}$ counts/keV-kg-year. No contemporary or planned experiment is able to claim or approach this level of discrimination.

As backgrounds may arise from both external radioactivity and from the allowed two-neutrino double beta-decay, various strategies are needed for mitigation. Heroic radio-purity regimens for materials are essential but have not yet been sufficient. Rejection of background events from two-neutrino double beta-decay will require energy resolution generally not worse than 1% FWHM. Germanium ionization detectors and tellurium oxide bolometric crystals have energy resolutions ten times better this, but have not yet been able to reject adequately all ambient radioactivity.[1] A new method to relieve the daunting future challenges of radio-purity will be most welcome.

The experimental quest, now more than half of a century old since the first insights of Racah, Goeppert-Mayer and Furry in the 1930s, thus remains an exciting but increasingly exacting scientific arena [3]. As a definitive observation of this decay mode would constitute a major discovery, the quality of evidence for a discovery claim must accordingly be very high. A new era has arrived– apparently without much notice–in which future experiments will be very costly in time-scale, careers, and resources and cannot be justified without very robust *a priori* background estimates supporting a credible, nearly background-free capability. The bazaar of technically interesting but scientifically marginal technical approaches explored earlier must yield to a small number of demonstrably robust 'discovery class' experiments.

2. Detection of the daughter atom

Detection of the daughter atom has been long recognized as a possible avenue to realize background rejection, since conventional processes do not introduce a new atom with Z ± 2. For barium, xenon's double-beta decay daughter, attention has been given to spectroscopic features of the singly ionized state, Ba^+, as this atomic configuration permits a sequence of repetitive excitation/de-excitation cycles with red and blue light involving a long-lived triplet D state [4]. Detection of an extended sequence of photons of two colors from a single atom is generally accepted as a robust determination of the presence of Ba^+.

Ba^+ two-color excitation and scintillation is, however, only possible in near-vacuum conditions. D-state quenching prevents measuring <u>both</u> spectroscopic features of single Ba^+ in high pressure or liquid xenon *in situ*. Conversely, the extraction of a single ion from a large mass of liquid or high-pressure gaseous xenon with rapid transport into a low-pressure trap where spectral interrogation becomes effective also appears to be extremely difficult, perhaps intractable, at least to some observers.

In the decay $^{136}Xe \rightarrow Ba + 2e^-$, with or without neutrinos, it is very likely that the daughter barium atom will be highly ionized by the disruptive departure of the nascent electrons from the nucleus. A highly ionized barium atom will strip electrons from nearby neutral xenon atoms until it is unable to de-ionize further. The process halts when barium reaches the doubly ionized state Ba^{++}. At this point,

---

[1] This is perhaps due to the need to embed a large number of small detectors with relatively high surface-to-volume ratio within an inter-penetrating structural harness, unlike the case of the monolithic xenon detectors.

the second ionization potential of barium, 10.04 eV, is less than the first ionization potential of xenon, 12.14 eV, [5]. Further extraction of electrons from neutral xenon atoms is energetically forbidden. So in pure xenon, the natural state of the equilibrium barium ion is expected to be, instead of singly ionized, doubly charged Ba^{++}. This may turn out to be a great advantage.

3. Single-molecule fluorescent imaging

But there may be another way for sensing the creation of barium, one that may be compatible with high-pressure xenon gas. Single-molecule fluorescent imaging (SMFI) is among the most powerful techniques in contemporary biological sciences. SMFI, developed by physicists, has been adopted and perfected by biologists and chemists.[2] The basic SMFI idea is to interrogate repeatedly a small, optically thin region with blue or near-UV photons that can excite a molecule of interest. Repeated interrogations can provide statistically precise localization down to the few nm^2 level, surpassing the Abbe diffraction limit. Detection of a fluorescent response is made possible by a chromatic shift or a delay in response time relative to a pulsed excitation. Fluorescent response quantum yields approach unity in many cases. Two-photon excitation by IR offers a way to obtain, by filtering, near-perfect rejection of the excitation wavelength. Many texts and references are available [6,7].

A wide variety of chemical and biological molecules has been found or constructed for SMFI. Of particular interest, for two reasons, are calcium-chelating fluorophores. First, doubly-charged calcium ions are fundamental in biology and hence prominent in biochemistry research. Second, since barium and calcium are congeners, what works for Ca^{++} may be relevant for barium. For example, Fluo-3, Fluo-4, and PET-1 are commonly used for calcium chelation. Calcium chelation by Fluo-3 is shown schematically in figure 1. Typical excitation of Fluo-3 is at 488 nm, with response peaking at 550 nm. Quoted response ratios between unchelated and chelated states vary from 60 to more than100. In a dry environment, the response ratio is unknown and may be quite different.
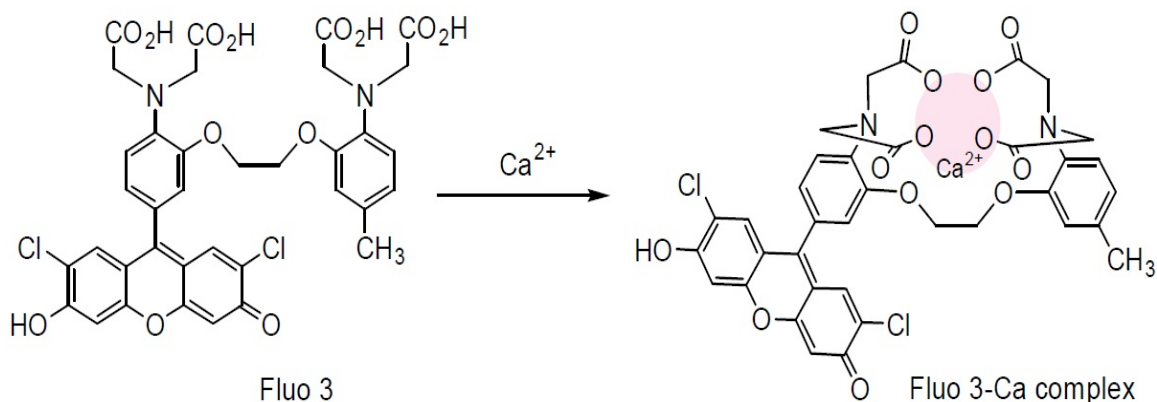

Figure 1. The skeletal formula for Fluo-3 is shown before and after complex formation. The conformal change that Fluo-3 undergoes in chelation with Ca^{++} creates a fluorescent complex. In that state, the fluorescent response increases by a factor of 60 - >100 in the cellular environment. *Image source*: Dojindo Molecular Industries, Inc.

Especially noteworthy is pyrene-stabilized monoazacryptand **1** [8]. This fluorophore is responsive with extremely high specificity to Ba^{++}, ignoring almost completely other alkaline earth doubly charged ions, even Ca^{++}, as well as all alkaline metal ions. Excitation at 342 nm leads to fluorescence in a 330-420 nm band. At this very early stage, with so many unanswered questions, the existence of numerous possible molecular avenues is comforting.

---

[2] The 2014 Nobel Prize in Chemistry was awarded to three scientists, trained as physicists.

In the presence of the TPC electric field, the barium ion will drift slowly to the cathode plane. The cathode is presents a thin dielectric layer to avoid neutralizing the arriving ion. The search problem is thus reduced from 3-D to a 2-D problem. If the cathode dielectric surface is coated with the incipient fluorophore, and if it is further assumed that the chelation process works in the high-pressure xenon gas, then, exactly one molecular complex becomes responsive to excitation whereas prior to that occurrence there is none. The unsolved challenge is to ascertain by interrogation that this is the case.

The interrogation/response rate in SMFI can exceed 10^5 per second. With the assumptions of Gaussian statistics, negligible photon background, and that calibrations establish an average single ion response to be, say, 100 detected photons, one might naïvely expect that the detection of 100 fluorescence photons from a single site establishes the presence of a single ion with variance $\sigma = 10$. The probability that two or more ions (or zero!) are responding to yield 100 $\pm 3\sigma$ detected photons is, in this idealized case, extremely small. An isolated complex can be interrogated in less than 100 μs. Only those event candidates with energy near the parent-daughter Q-value need detailed scrutiny. However, for an event to be considered as a double beta-decay candidate, it will be necessary to ascertain that exactly one new detected barium ion is now present within the general locus of the event candidate as projected on the cathode plane. Diffusion of the ion during drift, on the order of 1 mm rms, requires a relatively large search area within the projected event locus.
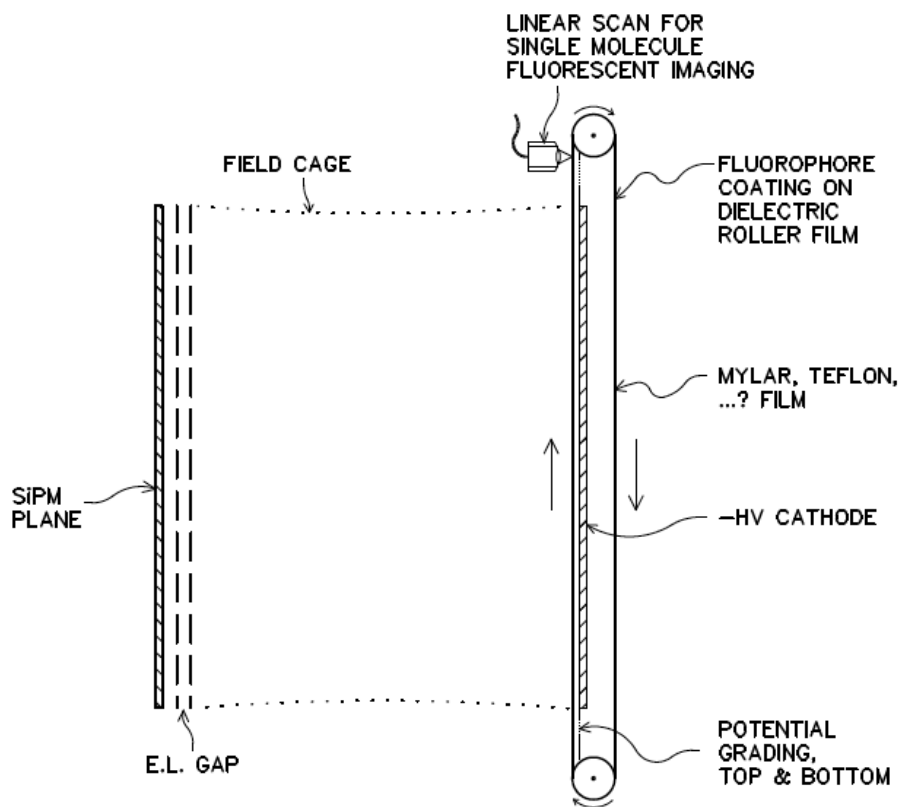
An immediate problem in this scenario is that the un-complexed molecules have to be unresponsive to a very high level if the search area is 'large'. A monolayer presents approximately 10^{10} molecules/ mm^2. A way out of this dilemma, with the addition of some magical thinking, is to imagine that the dielectric surface is a moving belt that transports the ion to a region where only charged molecules are selected. This could be achieved by having the moving belt surface pass by a reversed field region in a convenient location. At this point, all charged complexes would transfer to a co-moving roller where the scan occurs, leaving all the un-complexed ones on the belt. Thus the precursors do not contribute in the actual scan. This scenario is reminiscent of the familiar xerographic copy process, for which nano-xerography has been explored with some success. Some aspects of this are illustrated in figure 2.

The basic idea presented here is an augmentation of the high-pressure xenon gas (HPXe) electroluminescent (EL) TPC concept [9,10]. The HPXe EL TPC provides the desired energy resolution $\delta E/E < 1\%$ FWHM for the two nascent electrons through electroluminescence (EL), a nearly noiseless linear gain method [11]. Two-electron event topology (or single electron, as from γ-rays) is revealed in excellent detail in xenon gas at densities corresponding to ~10 bars. The increasing multiple scattering as the two electrons range out reveals the two-electron topology signature.

4. Perspective
The scale of interest is to achieve sensitivity that spans the inverted mass ordering. This implies a exposure goal of at least 1 ton-year with demonstrably negligible backgrounds. In the author's view, following a path that leads in a ton-scale experiment to a background-dominated result is failure.

The possibility to exploit SMFI is both appealing and admittedly bizarre at first read. A large number of questions arise for this concept, and the exploratory path forward will likely present numerous cul-de-sacs along the way. Nevertheless, the quest to find a technical approach that provides adequate and robust background rejection is compelling. A radically novel approach, if plausibly credible, is more rational to pursue and develop than following familiar paths to certain failure. Perhaps the biochemistry of $^{136}Xe \rightarrow {}^{136}Ba^{++}$ can show us the path to discovery.

LINEAR SCAN FOR
SINGLE MOLECULE
FLUORESCENT IMAGING

FIELD CAGE

FLUOROPHORE
COATING ON
DIELECTRIC
ROLLER FILM

MYLAR, TEFLON,
...? FILM

SiPM
PLANE

—HV CATHODE

E.L. GAP

POTENTIAL
GRADING,
TOP & BOTTOM

HIGH PRESSURE XENON GAS ELECTROLUMINESCENT TPC
WITH SINGLE MOLECULE FLUORESCENT IMAGING OF BARIUM DAUGHTER

Figure 2. The 3-D search problem for a single barium ion is reduced to 2-D by collection of the ion on a dielectric cathode surface. The cathode surface is a slowly moving belt that transports the complex to a 1-D scanner that senses the latent image with resolution of ~0.1 mm. The primary decay electrons, imaged in 3-D at the anode plane, predict the locus on the belt where, for true events, a barium ion complex should appear.

References
[1]   Valle J W F, arXiv:hep-ph/060810v1
[2]   Auger M *et al.* (EXO collaboration) arXiv:1205.5608v2 [hep-ex]; Asakura K *et al.* (KamLAND-ZEN collaboration) arXiv:1409.0077v1
[3]   Elliott S R and Vogel P, "Double Beta Decay" *Annual Reviews of Nuclear and Particle Science 2002* vol 52 p115-151
[4]   Moe M Physical Review C vol 44 (1991) p931
[5]   IAU Symposium 210 Uppsala; see IAU Symposium 210
[6]   Valeur B and Berberan-Santos M N, *Molecular Fluorescence* 2nd ed Wiley-VCH 2013
[7]   Lakowicz J R *Principles of Fluorescence Spectroscopy* 3rd ed Springer 2010
[8]   Nakahara Y, Kida T, Naktsuji Y, and Akashi M *A novel fluorescent indicatior for Ba^{++}in aqueous micellar solutions* The Royal Society of Chemistry Communications 2004 p224
[9]   Nygren D, Nuclear Instruments and Methods in Physics Research vol A 603 (2009) p337-348
[10] Gomez-Cadenas J J (NEXT collaboration) arXiv:1411.2433v1
[11] Alvarez V *et al.* arXiv:1211.4474v1 [physics.ins-det]