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Detection of the barium daughter in $^{136}Xe \rightarrow {}^{136}Ba + 2e^-$ by *in situ* single-molecule fluorescence imaging

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ABSTRACT

A robust technique for rejection of all γ-ray induced backgrounds in the search for the decay $^{136}Xe \rightarrow {}^{136}Ba + 2e^-$ is needed to proceed to ton-scale detection systems. Efficient detection of the barium daughter would provide a long-sought pathway toward this goal. Single-molecule fluorescent imaging may offer a new way to detect the barium daughter atom in a naturally ionized state. A doubly charged barium ion can initiate a chelation process with a non-fluorescent precursor molecule, leading to a highly fluorescent complex. Repeated photo-excitation of the complex can reveal both presence and location of a single ionized atom with high precision and selectivity. Detection would be automatic, and is accomplished within the active volume of a xenon gas Time Projection Chamber operating at high pressure.

1. Context

Observation of neutrino-less double beta-decay in any of several candidate nuclei establishes that lepton number is not conserved and that the neutrino is its own anti-particle, a unique property among spin 1/2 particles [1]. The experimental quest, now more than half of a century old since the first insights of Racah, Goeppert-Mayer and Furry in the 1930s, has been stimulated by the discovery of non-zero neutrino mass [2]. A clear observation of this decay mode would strengthen, for example, the idea of leptogenesis, which predicts accurately the baryon-antibaryon asymmetry of the universe and the very small but non-zero neutrino mass scale [3]. As observation of this decay mode would constitute a major advance, the quality of evidence for a discovery claim must accordingly be definitive. Recent results for ^{136}Xe have established lower limits of $\sim 2 \times 10^{25}$ years, corresponding to effective neutrino mass $\langle m\beta\beta \rangle$ sensitivities of $150 < \langle m\beta\beta \rangle < 280$ meV [4]. These results are sufficient to rule out an earlier observational claim based on ^{76}Ge [5]. The known neutrino mass parameters [6] establish for the inverted mass ordering an effective neutrino mass $\langle m\beta\beta \rangle$ range of 20–50 meV, corresponding to decay lifetimes T on the order of 10^{27} years.

To reach this unprecedented sensitivity goal, backgrounds from the allowed two-neutrino double beta-decay and natural radioactivity must be reduced to insignificance. An energy resolution requirement, prudently not worse than 1% FWHM [3], is sufficient for the former. However, natural radioactivity presents a daunting experimental challenge as current results suggest that further reductions by a factor of ~100 are needed to reduce the latter to insignificance. If the probability of even *one* background event is significant, sensitivity to running time evolves toward $t^{1/4}$ [7]. Further run time yields incremental sensitivity and scientific reach is effectively blocked. Semi-quantitatively, if background is zero, an exposure of about 1 ton-year is sufficient for most candidate isotopes, but if backgrounds in the *true event class* (after cuts, usually the energy region-of-interest or ROI) reach, for example, 10 events/ton-year, then an exposure of about 100 ton-years is needed for a 90% confidence result. This example illustrates the catastrophic impact of experimental backgrounds. 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 3%, near the two sigma level. 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 to $P \leq 1 \times 10^{-6}$ counts/keV kg-year. Germanium ionization detectors and bolometric crystals have energy resolutions of ~0.1% FWHM, but heroic radio-purity regimens for detector materials and shielding have been insufficient for background-free results at the tens- to hundred- kg scale in these techniques.

To probe deeply into the inverted mass ordering range in a reasonable time, active isotopic masses approaching ton-scale are necessary. It may be that no contemporary technique can demonstrate the necessary level of background rejection prior to commitment to and realization of a major project. A new method to relieve the daunting radio-purity challenges seems urgent. A

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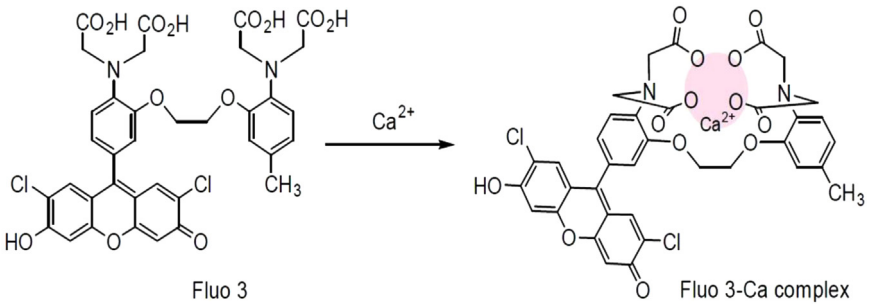

Fig. 1. The skeletal formula for Fluo-3 is shown before and after complex formation. The conformal change that Fluo-3 undergoes in chelation with Ca^{++} creates a fluorescent complex. In that state, the fluorescent response increases by a factor of 60 - > 100 in the cellular environment. *Image source*: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

new era has arrived, in which future experiments will be very costly in time-scale, careers, and resources, but which may lack a robust, *a priori* demonstrable, background-free capability. It seems clear that the current bazaar of technically interesting but scientifically inadequate approaches must yield to a small number of'discovery class' experiments: ton-scale exposures with negligible backgrounds.

2. The barium daughter atom

Detection of the daughter atom in double-beta decay has been long recognized as a very strong positive criterion for discovery, since no conventional process can introduce a new atom with $Z \pm 2$. 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 [8]. A highly ionized barium atom strips electrons from nearby neutral xenon atoms until further extraction of electrons from neutral xenon atoms is highly disfavored energetically. The process stops at the doubly ionized state, Ba^{++}, as the second ionization potential of barium, 10.04 eV, is substantially less than the first ionization potential of xenon, 12.14 eV, [9]. Free electrons liberated nearby by the emergent beta particles could also partially or completely neutralize the Ba^{++} ion. However, simulations indicate that recombination is very unlikely in a high-pressure pure xenon gas TPC. A further danger is the presence of impurities with ionization potentials lower than 10 eV, which can induce charge exchange and convert Ba^{++} to the singly ionized Ba^{+}.

For xenon's double-beta decay daughter atom barium, 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 alternating red and blue light involving a long-lived triplet D state [10]. Repeated detection of an alternating sequence of photons of two colors from a single atom is a robust determination of the presence of Ba^{+} in a trap. Ba^{+} two-color excitation and fluorescence is, however, only possible in near-vacuum conditions. Pressure broadening and D-state quenching prevent measuring both spectroscopic features of single Ba^{+} in high pressure or liquid xenon *in situ*. In practice, however, the sequence of extraction of a doubly charged ion from a large mass of high-purity cryogenic liquid or high-pressure gaseous xenon, conversion to a singly charged state, efficient transport into a low-pressure trap, extended two-color spectral interrogation, with credible spatial and temporal correlation to the double-beta decay event candidate, appears to be difficult [11].

In sufficiently pure xenon gas, the Ba^{++} state seems likely to survive at least until it reaches the cathode under the influence of the TPC electric field. Its arrival presents an opportunity for detection.

3. Single-molecule fluorescent imaging

There may be a hitherto unrecognized way for sensing the creation of a barium daughter atom: *single-molecule fluorescent imaging* (SMFI). SMFI is among the most powerful techniques in contemporary biological sciences. Developed originally by physicists, SMFI has been adopted and advanced by biologists and chemists.[1] Remarkably, the molecules serving SMFI purposes survive and perform in the wet and chemically active cellular environment. SMFI may plausibly be compatible with *in situ* imaging in high-pressure xenon gas. In SMFI, a small, optically thin region is interrogated repeatedly with blue or near-UV photons that excite a molecule of interest. The molecule is complexed with a doubly charged ion, typically Ca^{++}. The molecular complex fluoresces strongly, whereas un-complexed molecules respond very weakly. Image-intensified CCD cameras are used to detect single photons with pixel-scale spatial resolution. Repeated interrogations provide statistically precise identification and localization of a single molecule—even inside living cells. Spatial resolutions down to the few nm^2 level, far surpassing the Abbé diffraction limit, are routine. The interrogation rate in SMFI can exceed 10^5 per second and fluorescence quantum yields approach unity in many cases. Detection of a fluorescent response is facilitated by a chromatic stokes shift or a delay in response time relative to pulsed excitation. Two-photon excitation by IR offers a way to filter away the excitation wavelength. Many texts and references are available, such as [12,13].

A wide variety of chemical and biological molecules now exists for SMFI purposes. Of particular interest here are fluorophores such as Fluo-3, Fluo-4, and PET-1, which chelate Ca^{++}. Calcium chelation by Fluo-3 is shown schematically in Fig. 1 [14]. 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 than 100 in biological milieu. Although barium is uncommon in biochemistry research, the fact that barium and calcium are congeners suggests that techniques for Ca^{++} may have relevance for Ba^{++}.

In this regard, another fluorophore, pyrene-stabilized monoazacryptand (PSMA) is especially noteworthy [15]. This molecule, a crown ether with an attached pyrene group, is shown schematically in Fig. 2. PSMA displays extremely high specificity to Ba^{++}, ignoring almost completely other alkaline earth doubly charged ions, even Ca^{++}, as well as all alkaline metal ions. While a background arising from doubly charged calcium ions in high-pressure xenon gas seems very unlikely, it is remarkable that such specificity for barium is readily available. Excitation of PSMA at 342 nm leads to fluorescence in a wide band, 360–430 nm. According to [15], PSMA fluorescent functionality in a wet environment is protected from quenching by encapsulation in a micelle. The micelle would presumably not be needed in a dry environment

[1] The 2014 Nobel Prize in Chemistry was awarded to three physicists for their seminal contributions to SMFI.