Barium Tagging in Xenon Gas for Neutrinoless Double Beta Decay

The NEXT Collaboration *

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Snowmass LOI submitted to the attention of Working Groups:

NF05: Neutrino properties; IF9: Cross cutting instrumentation; RF04: LNV processes

1 Barium Tagging

It is well understood that the only known practically sensitive way to establish the Majorana nature of the neutrino is via direct and robust observation of neutrinoless double beta decay ($0\nu\beta\beta$). This question is central to understanding the physics of neutrino mass, likely the only observed manifestation of physics at energy scales above the electroweak, and compelling as a potential window into the mystery of the dominance of matter over antimatter in the Universe.

All existing techniques to search for $0\nu\beta\beta$ have been limited by backgrounds from radiogenic activity in detector materials. It is widely agreed that to achieve sensitivity sufficient to cross the inverted mass ordering band of $m_{\beta\beta}$, detectors with O(1 Ton) of isotope and background indices of order $b \sim 0.1ct[ton\,ROI\,yr]^{-1}$ are required. Existing technologies have demonstrated background rates between 2 and 200 in these units at the 100 kg scale. Factors of between 20 and 2000 are thus required to meet the goals for the coming generation of ton-scale experiments, presenting difficult technological challenges. The prospects for extending sensitivities into the normal hierarchy band of parameter space, on the other hand, appear truly formidable. The relevant experiments would employ many tons of active isotope and require background indices still lower than 0.1. A distinct but related question is how to confirm as a true signature of $0\nu\beta\beta$ a suggestive but inconclusive signal that may be observed by the next generation of background-limited ton-scale experiments. In either scenario, new and possibly radically new ultra-low background technologies will be required to follow the coming generation of experiments. The timescale to bring such technologies to maturity is sufficiently long that the time to invest in the relevant R&D is now.

One especially promising technical approach is "barium tagging"- identification of the daughter ion produced in the double beta decay of ^{136}Xe. It has long been recognized that an efficient barium ion tag with low background could reduce contamination from radiogenic backgrounds to near zero. Demonstrating a method for capture from one to several tons of xenon, transport to a sensor, and imaging at single ion level, requires many technological advances. In the last 2-3 years, there has been an upsurge in barium tagging activity, enabled by two key advances. First, demonstration of single barium ion sensitivity with single molecule fluorescence imaging (SMFI) for xenon gas [1]; and second, demonstration of single barium atom sensitivity in xenon ice layers [2] for use in liquid xenon. We describe R&D aimed at realization of efficient barium daughter ion tagging in a large-scale high-pressure xenon gas TPC. This effort is one of two distinct, coordinated approaches toward this goal within the NEXT collaboration.

2 Present State of SMFI-based Barium Tagging

The proposal to use SMFI to tag the daughter ion in $0\nu\beta\beta$ was first presented in Ref. [3] and expanded upon in Ref. [4]. Commercial dyes developed for Ca^{++} sensing in biological applications were demonstrated as sensitive barium tagging agents in solution phase. Soon thereafter, single ion sensitivity was achieved [1]. The model system there, liquid droplets within a polymer matrix, is not well representative of conditions within a xenon gas experiment. Several aspects of the chemistry of binding and fluorescence of the commercial dyes were found to be inadequate for dry barium sensing. This prompted a program of novel organic fluorophore development [5–9], which has culminated in multiple molecular candidates for single barium ion sensing based on crown-ether derivatives.

The family of molecules proposed in Ref. [7, 9] use turn-on fluorescence to identify the presence of new barium ions, whereas the related family developed in Ref. [8] change color upon binding, as well as increasing in intensity. Selectivity is strong in both cases, with only Sr and Hg identified as possible confounding species. While such competing ions are not expected in pure xenon gas, selectivity is required to avoid signal obfuscation at the single ion level by trace metals in re-agents and solvents used to make the sensor, typically present at ppb level. Ref. [9] explored tuning selectivity of the dye based on modifications to the crown ether group, ultimately culminating in

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demonstration of solventless single-ion sensitivity. Ref. [8] further demonstrated in-vacuo reaction with sublimated barium perchlorate. Both Refs. [8, 9] provide evidence that binding and fluorescence properties of crown-ether fluorophores are computationally tractable with density functional theory, allowing for future optimization in silico. The NEXT collaboration is now working to develop sensor surfaces based on self assembled monolayers of these active barium chemosensors for deployment in xenon gas test environments.

3 Near-term R&D

With promising molecules for barium tagging now available, near term R&D focuses on implementation of microscopy schemes applicable within high pressure gas environments. These programs require both single ion sensitive optics, reliable single ion sources, and mechanisms to bring the ion to the sensor, or vice versa.

The **GodXilla** program in the United States has demonstrated single-ion imaging using epi-fluorescence microscopy within a polymer layer held inside a 10 bar argon or xenon environment with a 30$\times$ 30 micron field of view (FOV). An upgrade is in progress to sensitize a 1mm^2 surface, sufficient to enable barium tagging with methods that employ ion concentration before imaging, such as RF carpets. A program of RF carpet R&D in high pressure xenon is underway using the **CARIBU** facility, where selected ^{144} Ba from ^{252}Cf decay will be injected into xenon gas and concentrated by a 10 cm diameter RF carpet. SIMION simulations and RF breakdown studies of xenon gas [10] suggest efficient transport at 10 bar will be achievable. Experimental verification is crucial, especially given non-trivial effects of molecular ion formation in high density gases which have been explored theoretically [11]. This program targets a full system demonstration of concentration and efficient fluorescent detection of barium ions within high pressure xenon gas on a 1-2 year timescale. The complementary **SABAT** program has also made important advances, and is the subject of a separate LOI.

4 Demonstrators and Future Experiments

An invaluable test-beam for a barium tagging system is the two neutrino double beta decay ($2\nu\beta\beta$) of ^{136}Xe. Following demonstration of one or more methods of single ion barium collection and tagging, realization of a demonstrator utilizing $2\nu\beta\beta$ to characterize performance of the system is a crucial step. While the ultimate role of topological information for future multi-ton scale experiments is unclear, topology remains a mandatory feature of a barium tagging demonstrator using $2\nu\beta\beta$, as the essential ingredient that allows for separation of signal events from the Compton continua of various high energy gamma rays on a per-event level.

At least one demonstrator experiment using $2\nu\beta\beta$ at the 10 kg scale will be a key technological step during this Snowmass period. The **CRAB** system, under construction at Argonne National Laboratory, will synthesize camera-based TPC readout with RF-carpet technology to produce a fully active barium tagging $0\nu\beta\beta$ TPC. This detector will initially run in an above-ground phase to exercise topological identification of $\beta\beta$ events using a SiPM-less tracking plane, followed by an underground deployment to demonstrate barium tagging. Injection of tagged barium ions from CARIBU is also under consideration. The CRAB system allow for quantification of both efficiency and background index of a barium tagging sensor in realistic operating conditions, while side-stepping engineering challenges associated with large-scale tracking detectors, potentially offering a truly scalable concept for barium tagging in xenon gas at the ton-scale and beyond.

Given the relative conceptual simplicity of a barium tagging-based $0\nu\beta\beta$ experiment in high pressure gas, following demonstration at this scale, the development of credible designs for a ton- or multi-ton scale system is plausible. Given suitable investment in technological development now, such systems could achieve readiness on the appropriate timescale to follow imminent ton-scale phases $0\nu\beta\beta$ experiments, including but not limited to NEXT-HD (the subject of an independent Snowmass LOI).

5 Conclusion

Barium tagging is a potentially paradigm-shifting approach to searches for $0\nu\beta\beta$ that may be critical for surpassing present-generation background indices of $b \sim 1ct[ton\,ROI\,yr]^{-1}$. Recent dramatic advances in single-ion tagging technology make the realization of detectors based on this method plausible on the Snowmass timescale. We advocate here for 1) continued support of R&D in chemistry and ion manipulation needed to bring the approach to full maturity, and 2) realization of demonstrator-scale experiments that can prove the technique using $2\nu\beta\beta$ events into the US / world neutrino physics program on a 3-5 year timescale. This will allow for preparation of the robust and physics-ready technology needed to follow the present generation of ton-scale $0\nu\beta\beta$ experiments.

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