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# Barium tagging from nEXO using Resonance Ionization Spectroscopy

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**Abstract**

nEXO is a 5-ton liquid enriched-xenon time projection chamber (TPC) to search for neutrinoless double-beta decay, designed to have the sensitivity to completely probe the inverted mass hierarchy of Majorana neutrinos. The detector will accommodate—as a background reduction technique—a system to recover and identify the barium decay product. This upgrade will allow a background-free measurement of neutrinoless double-beta decay and increase the half-life sensitivity of the experiment by at least one order of magnitude. Ongoing research and development includes a system to test barium extraction from liquid xenon using surface adsorption and Resonance Ionization Spectroscopy (RIS).

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**1. Introduction**

Neutrinos are potentially Majorana fermions, a property that can be investigated through neutrinoless double-beta decay ( $0\nu\beta\beta$ ), a nuclear decay postulated for several neutron-rich isotopes [1]. Double-beta decay,



is allowed by the Standard Model for  $n = 2$  ( $2\nu\beta\beta$ ). If the neutrino is a Majorana particle, then double-beta decay could proceed through the neutrinoless channel, where  $n = 0$  [1]. The EXO-200 experiment successfully measured the  $2\nu\beta\beta$  decay of  $^{136}\text{Xe}$  [2], and set some of the most competitive limits on the neutrinoless mode [3]; however, a larger source mass is necessary to probe lighter neutrino masses.

The nEXO experiment seeks to build on that success with a twenty-five times larger source mass and the potential upgrade of a barium-tagging system, allowing the experiment to fully probe the inverted mass hierarchy of Majorana neutrinos. nEXO is a 5-ton liquid enriched-xenon time projection chamber (TPC), designed specifically to limit backgrounds in the region of interest around the  $^{136}\text{Xe}$   $0\nu\beta\beta$  decay  $Q$ -value.  $^{136}\text{Xe}$  was chosen because it offers an acceptably high natural abundance and a relatively easy enrichment process. Furthermore, it can be used as the detection medium in a TPC [4].

Similar to EXO-200, nEXO is a thin-walled copper TPC with position resolution to distinguish  $\beta$ -like events from  $\gamma$ -like events [5]. The goal sensitivity to  $0\nu\beta\beta$  nEXO for five years of exposure (without barium tagging) is  $T_{1/2}^{0\nu\beta\beta} < 6.0 \times 10^{27}$  years.

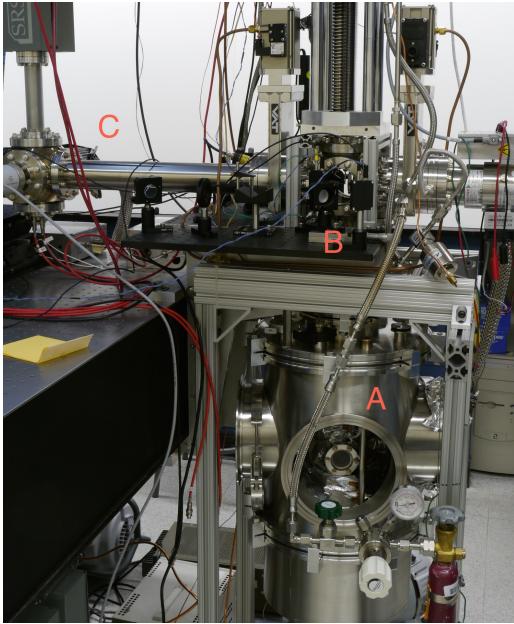


Fig. 1. (Color Online) The liquid xenon barium tagging device. The insulation vacuum jacket with one of the access ports open (**A**). A breadboard mounted off the optical table supports the desorption and RIS optics, focused into the load-lock (**B**). The probe is actuated by the vertical lead screw above the optical access. The time-of-flight mass spectrometer extends to the left of the image (**C**).

Detection of the decay product— $^{136}\text{Ba}$ —offers complete elimination of backgrounds in the region of interest [6] [4]. Liquid or gas xenon detectors are the only  $0\nu\beta\beta$  experiments that may allow this background reduction. With such a barium tagging scheme, the sensitivity to the neutrino mass grows as  $\sqrt{Nt}$ , where  $N$  is the number of source atoms and  $t$  is the exposure time—faster than the case where some background has to be subtracted statistically ( $(Nt)^{1/4}$  scaling). nEXO with barium tagging will achieve a half-life sensitivity of  $T_{1/2}^{0\nu\beta\beta} < 3.2 \times 10^{28}$  years.

Barium extraction and identification from the nEXO detector requires single-ion detection and high transport efficiency, issues which require significant research and development. Several groups within the nEXO collaboration are developing barium tagging methods, including single-barium fluorescence detection in frozen xenon, probe systems to collect Ba from the site of the decay, and gas ion transport to allow drifting ions out of future gas xenon detectors [7, 8]. This publication focuses on a technique using surface adsorption and Resonance Ionization Spectroscopy (RIS).

## 2. Ba-tagging prototype

A prototype setup has been built (Figure 1) to transport barium ions out of the liquid xenon (LXe) into a vacuum chamber where they can be identified using RIS [9]. In this setup, a source produces barium ions that are electrostatically drawn to and adsorbed onto the surface of a carrier substrate. The probe tip is then mechanically removed from the LXe to a chamber which can be valved out and rapidly pumped to vacuum for identification of the barium while LXe is maintained below (Figure 2). In order to recover the bulk of the xenon, the gas is cryopumped into a recovery bottle, while a small amount of it is pumped out to reach a base pressure suitable for time-of-flight mass spectrometry. The base pressure of the vacuum system is  $5.0 \times 10^{-9}$  mBar.

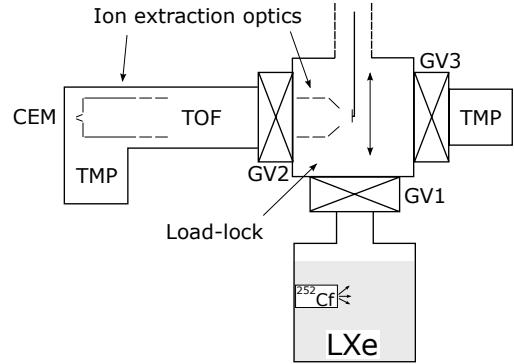


Fig. 2. Schematic of the apparatus in Fig. 1. The probe and substrate move between the liquid xenon (LXe) at the bottom and the time-of-flight spectrometer (TOF) at the top. Three gate valves (GV) isolate the LXe at atmospheric pressure from the TOF and channel electron multiplier (CEM) at high vacuum pumped out by turbomolecular pumps (TMP) at each end. The diagram also depicts the  $^{252}\text{Cf}$  source in the LXe.

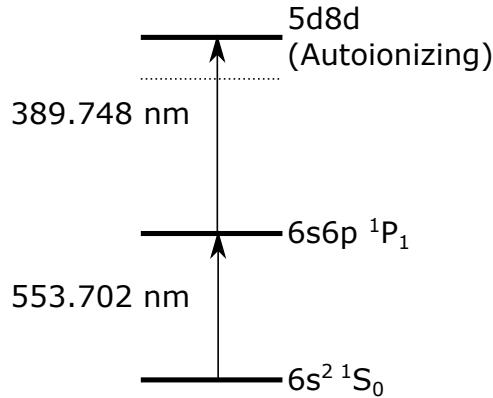


Fig. 3. The Resonance Ionization Spectroscopy scheme. Dye lasers are used to saturate both of these transitions for efficient and selective ionization.

Neutral barium atoms are thermally desorbed from the surface by a 1064 nm pulsed Nd:YAG laser. The desorption beam is focused onto the surface to a size of  $275 \mu\text{m} \times 750 \mu\text{m}$  (Gaussian  $\sigma$ ) resulting from a 70-degree angle of incidence with respect to the surface normal (constrained by optical access). Pulsed dye lasers pumped by a second Nd:YAG resonantly photoionize previously-desorbed barium atoms using 553.702 nm and 389.748 nm light (Figure 3). These RIS lasers are delayed by 1  $\mu\text{s}$  with respect to the desorption laser to optimize overlap between the RIS lasers and the desorbed plume moving away from the surface at thermal velocity. The RIS lasers drive both transitions at well above their saturation energies and are aligned parallel to the substrate, as close as possible to it without ablating ions from the surface.

The ions produced in this fashion are accelerated into a time-of-flight mass spectrometer.  $\text{Ba}^+$  ions take roughly 37.2  $\mu\text{s}$  after the ionizing laser pulse to travel through the 67.2 cm-long time-of-flight.

Existing alkali surface contamination is used to cross check the calibration and to establish the mass resolution of the time of flight. The time of flight system has a resolution of  $m/\Delta m \approx 80$  (FWHM) in the region of interest at 136 amu/e, as measured by the width of a  $^{133}\text{Cs}$  peak (ionized by the IR desorption laser).

In order to distinguish barium from various backgrounds, one or more of the three laser pulses can be omitted (the desorption IR pulse and the subsequent simultaneous RIS pulses). The standard data-taking cycle alternates between shots where all three lasers are fired with shots omitting the RIS lasers. This cyclic operation is important because the measurement is intrinsically destructive—a small number of Ba atoms is consumed by the process of measuring the recovery efficiency.

While future work in LXe will be performed dispensing Ba (along with other species) from a  $^{252}\text{Cf}$  fission source, in the initial work presented here the substrate is loaded in vacuum using a nuclear recoil-driven Ba source [10]. This source provides lower energy ( $< 100 \text{ eV}$ ) Ba that, unlike in the case of the fission source, is not implanted deeply into the substrate. The higher energy products from the fission source are expected to be more appropriate for the use in LXe, where it is desirable to deliver ions at an appreciable distance from the source. In both cases—and in a large LXe double-beta decay detector—the RIS selectivity is essential to tag the Ba atoms from other species.

Selectivity of the RIS process coupled with the mass spectrometer eliminates most of the background, leaving efficient transport of Ba from the LXe to detection as the main challenge. This is primarily a problem of surface science, as different surfaces offer different thermal properties and adsorption energies for barium. The surface must have a high melting point and a low desorption energy to allow efficient laser-induced thermal desorption and also be clean of Ba impurities. This applies also, to some extent, to the bulk of the substrate, as diffusion easily brings to the surface impurities at a single atom level. Finally, notwithstanding the selectivity of RIS, it is desirable that the desorption of all other species is generally low, in order not to clutter the TOF spectrum.

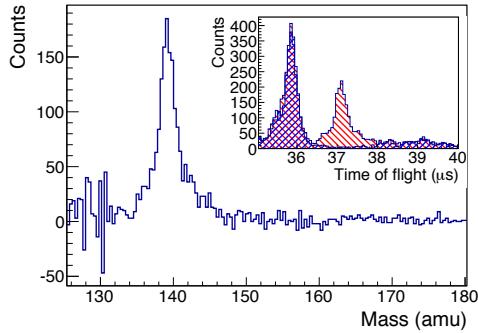


Fig. 4. (Color online) An RIS Ba signal with minimal background, from surface contamination on a Si surface. The background subtracted spectrum shows a clear, resonantly ionized barium signal at 138 amu. Inset: The blue histogram shows the signal without the RIS lasers, representing background from ions ablated using only the IR desorption laser. The red histogram shows barium that has been desorbed as a neutral atom and photoionized through the RIS scheme. Note that the resonantly photoionized barium arrives delayed relative to other peaks by the time delay of the RIS lasers, about 1  $\mu$ s.

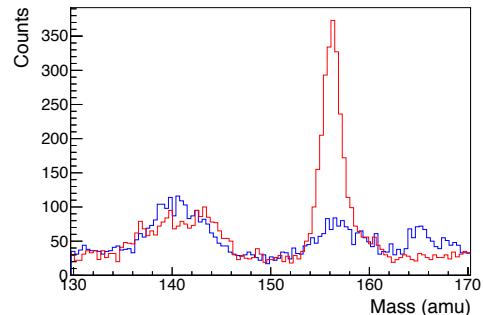


Fig. 5. (Color online) Ablated ion time of flight spectrum before (blue) and after (red) of deposition of  $\text{Ba}^+$  and  $\text{BaF}^+$  on the clean Si (111) surface. The peak at 140 amu corresponds to  $\text{Si}_5^+$ . Note the appearance of the peak centered at 157 amu, the appropriate mass for  $\text{BaF}^+$ . This  $\text{BaF}^+$  peak is unaffected by the RIS lasers.

Silicon wafers offer many of these desired surface properties. Studies of Ba on Si demonstrate that Ba can be thermally desorbed from the surface, highlighting the potential of this surface for highly efficient recovery of barium atoms [11]. Additionally, commercially available silicon wafers have very low levels of impurities, and further cleaning *in situ* is possible.

This cleaning is accomplished through outgassing the Si for hours at 700 C, then removing the oxide layer by flashing the Si to 1250 C for a few minutes. This system is constructed to allow resistive heating of the silicon surface.

### 3. Results

Most impurity atoms are cleaned from the Si surface by resistively heating to 1000 C for several minutes. Additional cleaning by rastering the IR desorption laser across the surface has been found to provide a very clean time of flight with only K and  $\text{Si}_n^+$  clusters, up to  $n = 5$  in some datasets (Fig. 5). The combination of these two cleaning methods may be particularly effective because although resistive heating mobilizes contaminants (including Ba) it cannot fully exhaust the relatively large bulk reservoir, and the laser then removes the surface contamination without disturbing what is left in the bulk.

Although a small amount of K remains after this cleaning process, Li, Na, and Rb are entirely eliminated from the surface. RIS of Ba allows separation of these backgrounds from the signal, shown in Figure 4. The inset shows the raw data, with desorption-only signal shown in the blue histogram and desorption-RIS data shown in the red histogram. The peak appearing in the red histogram only is the RIS Ba signal, while the peak at 36  $\mu$ s is Ba desorbed and ionized by the IR laser. These data demonstrate detection of barium through the desorption-RIS process. The background (blue) can be subtracted from the signal (red) to give the background-subtracted RIS Ba signal shown in the main panel of Figure 4.

After cleaning using the above process, exposing the surface to the Gd-driven barium source for 14 hours, with the substrate charged to  $-230$  V to attract  $\text{Ba}^+$  and  $\text{BaF}^+$  ions results in a peak at 157 amu, as shown in Figure 5. This is consistent with  $\text{BaF}^+$  ions, a known byproduct of the source owing to the chemical form of the Ba in the coating ( $\text{BaF}_2$ ) [10], showing effective transfer and detection using this system. Based on the source known yield and the loading ion-optics, we estimate that  $10^7$  barium ions are collected on the

substrate. The peak at 157 amu is depleted over 200 laser shots, indicating that the small amount of BaF<sup>+</sup> deposited on the surface has been removed by laser thermal desorption.

#### 4. Outlook

Ongoing work is focused on reducing backgrounds due to surface contaminations of barium and silicon clusters. Other ultra pure materials, such as silicon carbide, are under investigation as possible alternatives to silicon. Potential reactants such as oxygen may exist on the surface and should be eliminated if they are present. This progress must demonstrate that barium can be recovered efficiently and spectroscopically identified from vacuum before moving to liquid xenon recovery.

#### 5. Acknowledgements

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#### References

- [1] F. Avignone, S. R. Elliott, J. Engel, Double beta decay, Majorana neutrinos, and neutrino mass, *Rev. Mod. Phys.* 80 (2008) 481–516.
- [2] J. B. Albert, M. Auger, D. J. Auty, et al., Improved measurement of the  $2\nu\beta\beta$  half-life of  $^{136}\text{Xe}$  with the EXO-200 detector, *Phys. Rev. C* 89 (2014) 015502.
- [3] J. B. Albert, D. J. Auty, P. S. Barbeau, et al., Search for Majorana neutrinos with the first two years of EXO-200 data, arXiv:1402.6956v1.
- [4] M. K. Moe, Detection of neutrinoless double-beta decay, *Phys. Rev. C* 44 (1991) R931–R934.
- [5] M. Auger, D. J. Auty, P. S. Barbeau, et al., The EXO-200 detector, part I: detector design and construction, *J. Inst.* 7 (2012) P05010.
- [6] M. Danilov, R. DeVoe, A. Dolgolenko, et al., Detection of very small neutrino masses in double-beta decay using laser tagging, *Phys. Lett. B* 480 (2000) 12–18.
- [7] T. Brunner, D. Fudenberg, G. Gratta, et al., A setup for Ba-ion extraction from high pressure Xe gas for double-beta decay studies with EXO, *NIM B* 317, Part B (2013) 473–475.
- [8] D. Sinclair, M. Breidenbach, R. DeVoe, et al., Prospects for Barium Tagging in Gaseous Xenon, *J. Phys.: Conf. Ser.* 309 (2011) 012005.
- [9] G. S. Hurst, M. G. Payne, S. D. Kramer, et al., Resonance ionization spectroscopy and one-atom detection, *Rev. Mod. Phys.* 51 (1979) 767.
- [10] M. Montero Díez, K. Twelker, W. Fairbank, Jr, et al., A simple radionuclide-driven single-ion source, *Rev. Sci. Inst.* 81 (2010) 113301.
- [11] S. Hongo, K. Ojima, S. Taniguchi, et al., Observation of the interface of Ba/Si (100) by MDS and TDS, *Appl. Surf. Sci.* 82 (1994) 537–542.