Prospects for Barium Tagging in Gaseous Xenon

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Abstract. Tagging events with the coincident detection of a barium ion would greatly reduce the background for a neutrino-less double beta decay search in xenon. This paper describes progress towards realizing this goal. It outlines a source that can produce large quantities of Ba++ in gas, shows that this can be extracted to vacuum, and demonstrates a mechanism by which the Ba++ can be efficiently converted to Ba+ as required for laser identification.

1. Introduction
The concept of rejecting the backgrounds in the search for neutrino-less double beta decay by tagging events with the coincident detection of the barium daughter ion was first suggested by M. Moe [1]. This suggestion was developed into a conceptual design for a gaseous xenon detector.
by M. Danilov et al. [2] based on extending the design of the Gotthard detector [3]. At the time, single ions of Ba+ had been successfully detected using laser fluorescence for ions contained in RF quadrupole traps [4]. The concept presented by Danilov et al. introduced laser beams into a gas TPC and guided the beams by means of movable mirrors to the location of the possible barium ion. The fluorescence would be detected by an array of photomultiplier tubes placed around the field cage.

There are a number of potential difficulties with this scheme.

- The barium ion is created as Ba++ (it may be in a higher charge state but will rapidly find electrons to reach the doubly charged state) but the species that can be fluoresced is only the Ba+ ion. Ba++ cannot pick up an electron from a single Xe atom and it is not known if it will convert to Ba+ due to the formation of xenon clusters.
- If the barium ion is in high pressure gas, the transitions will be pressure broadened. This requires increasing the laser power by a factor of about 1000 if the fluorescence rate is to be maintained at the level used in traps. With this laser power in high pressure gas there will be a significant scattered background which will make the positive identification of a single barium ion challenging.
- It is highly desirable to detect the scintillation light from the ionizing event and if one uses normal gas gain detection devices for the energy measurement, it is likely that the quench gas used will quench the scintillation by de-exciting the Xe dimmers.

We have therefore been exploring an alternate concept for the detector. In this concept we operate a TPC in pure xenon. The scintillation light and ionization electrons are measured using an electroluminescence process as discussed by Graham [5]. When a candidate event is observed in the ionization signal, the barium ion will be guided towards a nozzle leading to a vacuum region, by shaping the electric drift field. The ion would be swept out of the chamber by the gas flow and then separated from the gas using an ion guide system. This type of system is commonly used for the production of secondary (possibly radioactive) beams for nuclear physics research, and by the bio-molecular community in their mass spectrometers. The ion would then be converted to Ba+ (if it does not naturally happen in xenon) and detected using the proven techniques of laser fluorescence.

In order to establish the feasibility of this concept several measurements must be carried out.

- We need to understand the ability of the electroluminescence detection to determine the decay energy and the event vertex.
- We need to measure the mobility of Ba++ in Xe.
- We need to understand any loss mechanisms of Ba++ either to Xe or to contaminants.
- We need to demonstrate that Ba++ ions can be extracted from the high pressure gas and determine the efficiency.
- We need to establish a feasible way to effectively convert Ba++ to Ba+.
- We need to develop and prove a concept for fluorescence of the Ba+ ions either by introducing them to a trap or by looking at the fluorescence in flight.

2. An Ion Source
The first step in addressing these issues has been to find a mechanism for conveniently producing Ba++ ions in high-pressure gas. Several sources were investigated.

Production of barium ions using a spark discharge with barium on the electrodes is a technique that can produce barium ions. Figure 1 shows such a discharge with a laser tuned to the valance S1/2 to P1/2 transition passing through the arc. The fluorescence of the ions is obvious. This source can be used over a small pressure range to study the energy shift and broadening of the
transition but it is not clear how to identify any Ba++ production. The environment about the ions is very different from the environment in which our Ba will be created.

Figure 1. Production of barium ions in an arc. A laser beam passing through the gas fluoresces the Ba+ ions as they diffuse away from the arc.

A second source technique that was investigated is laser ablation. Barium metal and barium compounds were ablated using a focused, pulsed UV laser beam. In vacuum the products can be identified by measuring the time of flight of the ions to a detector. A typical spectrum is shown in Figure 2. In can be see that this is a convenient source for production of Ba+ but there is no evidence for production of Ba++. 
Figure 2. Ions produced by a laser ablation source using BaAl as the target and identification of ions by time of flight. The prediction is based on the use of the SIMION code with a source temperature taken to be 30,000K.

A third source was based on production of barium ions by alpha decay recoil. An alpha emitting material is deposited on a silicon detector and a thin barium salt is deposited over the source. When an alpha decay takes place, the alpha can be detected in the silicon detector and the recoil may knock barium out of the covering layer. Some of these atoms may emerge as ions. The ion spectrum can be measured by time of flight using the silicon detector as the start signal. A spectrum quite similar to Figure 2 above is found. No evidence is seen for the production of Ba++. This source has been described in more detail in [6].

The final source which we investigated, and found to work well, is electrospray. In an electrospray source of barium, a salt of barium is dissolved in water and mixed with methanol. The liquid is sprayed through a very fine nozzle which is which is maintained at a high negative potential. The flow of heated gas at atmospheric pressure causes the droplets to evaporate and as they reach the Rayleigh limit they produce a copious stream of ions. As the barium in the water tends to be in the form Ba++, it is expected that this will be the dominant ion produced.

A mass spectrometer (a Fisons Quattro II) with an electrospray source was used to test this mechanism. Ions from the source pass through a small orifice through a roughing pumped region into a high vacuum maintained by a turbo pump. The ions are trapped in a sextupole ion guide and travel to a quadrupole spectrometer. When the source conditions are optimized for Ba++ production the spectrum shown in Figure 3 is obtained. The dominant peak is Ba++. Figure 4 shows a portion of the spectrum expanded about m/z=69. We can confirm that this group is Ba++
by noting that all of the isotopes of Ba are seen in the proper intensity ratio and the group must be a $z=2$ group because the peaks are spaced by $\frac{1}{2}$ unit in m/z.

Figure 3. The spectrum obtained using a quadrupole mass spectrometer with an electrospray source. The source solution was 1 mM barium acetate in an equal mix of water and methanol.
3. A Charge Changing Technique

The Quattro II contains two quadrupole spectrometers separated by a gas collision cell where the ions are contained using an RF sextupole. We have used this system to look at conversion of Ba++ to Ba+. Our first attempts used triethylthene (TEA) as a low ionization potential gas in the cell. For this measurement the first quadrupole was set to pass only m/z = 69. Ions were accelerated into the cell and the products were measured with the second quadrupole. Measurements were taken for a range of pressures and acceleration voltages. An example spectrum at high pressure and voltage is shown in Figure 5. We see that all of the Ba++ has been converted to Ba+ with the production of TEA+. The Ba peak is broad because the ion enters the spectrometer with a high velocity while the TEA picks up very little momentum in the transfer and is sharp. There is no evidence for the formation of any other species or molecules.
Figure 5. The spectrum of particles produced when Ba\textsuperscript{++} passes through TEA. The Ba\textsuperscript{++} ions are accelerated through 100 volts and the TEA pressure is 10\textsuperscript{-3} mbar. The length of the cell is about 20 cm.

4. Implications of the Measurements
It is clear from the above discussion that electrospray is a convenient mechanism for producing Ba\textsuperscript{++} is gas at atmospheric pressure. It is likely that the source will perform just as effectively at higher pressures. Even though the source region has water vapour and methanol vapour at the 0.3\% level, there is no evidence for molecular formation. The use of TEA offers an effective method to achieve the charge state conversion. The overall design of the ion extraction from high pressure to vacuum is very similar to the scheme proposed for the final detector and this appears to work well although the efficiency is not yet determined.

5. Future Program
The next step in our investigation will be to operate an electrospray source in xenon gas. To do this a special source is being fabricated which will allow efficient re-circulation of the gas. A feature of the source will be a drift region where the mobility of the ions can be investigated by countering the motion due to the gas flow with motion due to an electric field. A sketch of the source is provided in Figure 6 below.
Figure 6. Exploded view of the nano-spray and mobility channel now under construction for further studies.

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