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# Detection of trace levels of radioactive decay produets: Is it possible to determine beta-beta decay half-lives?

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## ABSTRACT

Multiphoton resonance ionization (MPRI) spectroscopy coupled with energetic ion bombardment has been used to detect sub-part-per-billion quantities of material desorbed from a surface. We report a sensitivity for the detection of In, uniformily doped into Si, of 9 parts-per-trillion. Using this result, the feasibility of applying MPRI to the detection of trace levels of radiodecay products is discussed.

## Introduction

The phenomenon of double beta decay is one of the rarest events in nature. It can be defined as a process in which the charge (Z) of a nucleus X changes by two units while the number of nucleons remains the same. One mechanism describing the double beta decay event involves the emission of two beta particles ($e^-$) and two antineutrinos ($\bar{\nu}$) (designated as the two neutrino (2ν) mode): ${}_{z}{}^{A}X \rightarrow {}_{z+2}{}^{A}X^{++} + 2e^- + 2\bar{\nu}$. In general, the lifetimes for this process are in excess of $10^{18}$ years. The indirect observation of two double beta decay events has been reported for $^{130}$Te and $^{82}$Se parent nuclei in geological formations using noble gas mass spectroscopic measurements of the daughter atoms (Commins 1983). Recently Elliott et al. have claimed the first direct observation of the $^{82}$Se 2ν double beta decay event using a time projection chamber (Elliott 1987).

A more exotic mechanism for double beta decay is the neutrinoless mode (designated as the 0ν mode): ${}_{z}{}^{A}X \rightarrow {}_{z+2}{}^{A}X^{++} + 2e^-$. This mode is predicted to have an even longer lifetime than the 2ν radiodecay. The significance of a mere observation of a neutrinoless event would indicate that the neutrino has a non-zero rest mass (Caldwell 1988), contrary to standard theories.

Our approach for measuring the lifetime of a double beta transmutation consists of determining the number of daughter atoms produced in a given time period under carefully controlled laboratory conditions. Specifically, we have begun a series of experiments aimed toward measuring the double beta decay lifetime of $^{136}$Xe by carefully collecting and then counting the $^{136}$Ba daughter nuclei. To illustrate the difficulty of this measurement, consider that 1 mole of $^{136}$Xe should produce ≈1100 $^{136}$Ba atoms per year if the theoretical lifetime of $3.8 \times 10^{20}$ years (Mitchell 1987) is correct. This extrapolates to a surface concentration of 1 ppt. Clearly, the measurement of such a small quantity of decay product requires an ultra-sensitive surface analytical method.

It has been previously shown that energetic ion bombardment coupled with multiphoton resonance ionization (MPRI) spectroscopy is an effective tool for the analysis of solids (Winograd 1982 and Kimock 1984). Indeed, the inherent sensitivity of this method has been demonstrated by analytical measurements down to the few parts-per-billion (Parks 1984) and several hundred parts-per-trillion level (Young 1987).

We propose to count the collected daughter atoms using MPRI detection of sputtered neutrals. This particular experiment is sensitive primarily to the 2v decay mode of $^{136}Xe$ since it is ≈1000 times more probable than the 0v mode. From the results, the lifetime can be calculated which will provide some insight into the correct nuclear structure theory. A complementary experiment is presently underway using a time projection chamber that is capable of measuring the 0v decay lifetime. Thus, combining the results of the two experiments, the rest mass of the neutrino may be calculated (Mitchell 1987).

In order to demonstrate the feasibility of the $^{136}Xe$ double beta decay experiment, ultra-trace analyses of uniformly distributed materials have been pursued to determine the detection limit of the MPRI apparatus (Figure 1). In addition, work currently underway exploits the surface specificity of the method in the quantitative trace analysis of impurities deposited upon the topmost layer of solids.

In this work we report the current detection limit of the MPRI technique. This value will be used as a means of determining the feasibility of measuring double beta decay half-lives through daughter atom counting. Toward this end, a new ultra-trace analysis apparatus utilizing a high current ion source and a reflection time-of-flight mass spectrometer has been constructed (Pappas 1988).

## Experimental

All experiments were performed in an ion pumped Perkin-Elmer TNB-X UHV chamber (Figure 1).

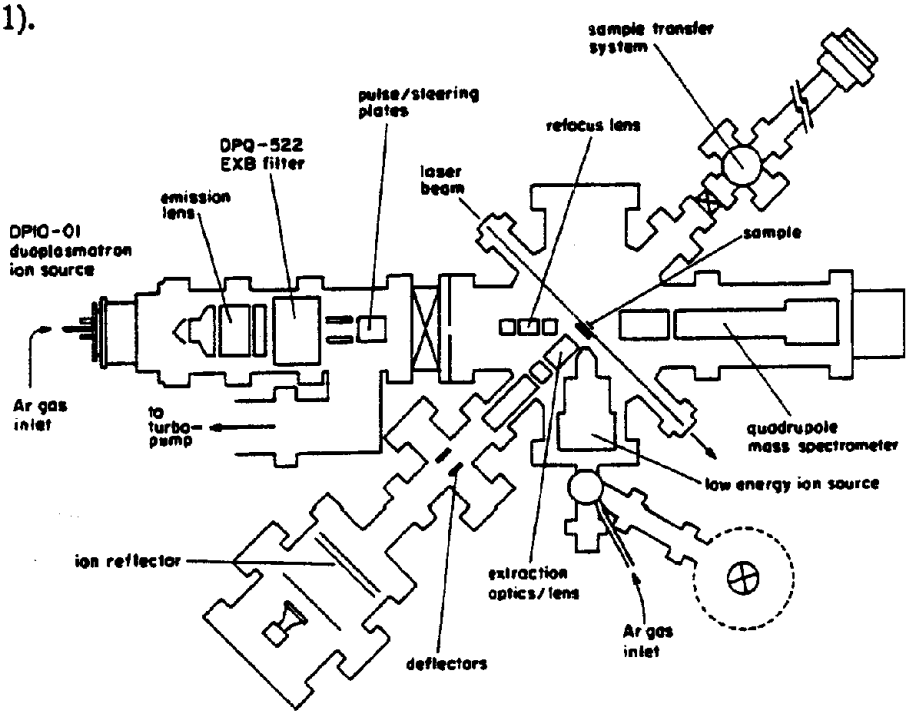


Figure 1. Schematic diagram of the MPRI apparatus.

The primary beam of $Ar^+$ ions (60 μA, 10 keV) was generated by a Physicon DP10-01 ion source with a typical pulse duration of 5.6 μs. The laser system used to provide tunable photon radiation consisted of a Quanta-Ray Model PDL-2 dye laser pumped by a Model DCR-2A Nd:YAG laser. Frequency doubling and mixing of the dye laser output was accomplished using a Model WEX-1 wavelength extension unit.

Following resonant post-ionization, the photoions are extracted into a reflecting time-of-flight mass spectrometer and detected by a Galileo Electro-Optics Corp. Model FTD 2002 dual microchannel plate (MCP) assembly. The output signal from the MCP is processed digitally using a Princeton Applied Research Model 1121A amplifier/discriminator unit and a gated Princeton Applied Research Model 1112 photon counter/processor.

In an effort to demonstrate the current sensitivity and linearity of the MPRI method, a set of Si samples (Hughes Research Laboratories and Atom Sciences, Inc.), uniformly doped with In, were analyzed. The targets were characterized by resistivity measurements, yielding In concentrations of 2000, 36.5, and 3.85 ppb. In addition, the $^{113}In$ isotope from the 3.85 ppb target provided a 0.165 ppb sample for analysis.

Prior to the measurement, each sample was sputter-etched to establish a steady-state, reproducible response indicative of the bulk concentration. The analysis time was 5 minutes (9000 laser pulses at a repetition rate of 30 Hz) for both the signal and background determinations. From consideration of the duty cycle of the experiment, the amount of material removed during the 5 minute analysis time corresponded to less than a monolayer.

## Results

The linearity of the MPRI method is demonstrated in Figure 2, where the In background subtracted count rates are plotted against the reference concentrations (determined by Hughes). The MPRI results appear to correlate well with the expected values down to the few ppt level.

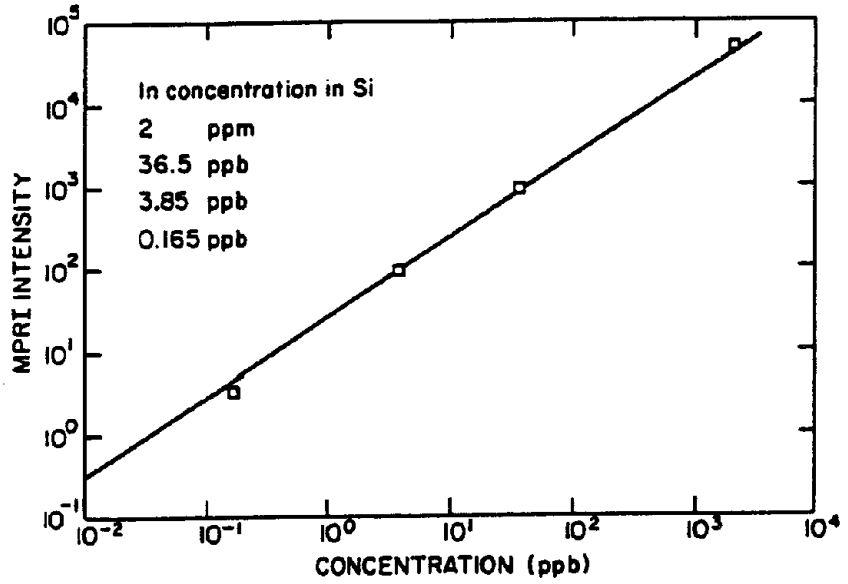


Figure 2. Comparison of MPRI intensities with Hughes reference values for In in Si.

The ultimate sensitivity for these measurements was determined using the results of the analysis for the 0.165 ppb $^{113}$In in Si sample. This target yielded 37 signal counts and 4 background counts during the 5 minute counting period for a primary ion current of 46 μA and an ionization efficiency of 0.55. Thus, for a signal-to-noise (S/N) level of 2, the concentration of the 0.165 ppb sample may be extrapolated to 0.036 ppb or 36 ppt. This implies that for an obtainable incident ion current of 100 μA and an ionization efficiency of 1, the detection limit is 9 ppt. Given a primary ion beam diameter of 3 mm (0.07 cm$^2$), the number of atoms in the sampling area is 7 x 10$^{13}$. Thus, for the removal of 1 monolayer of material during an analysis and using 9 ppt as the detection limit, this experiment is sensitive to 630 atoms.

Present Work

For the collection of radiodecay products, a chamber has been constructed which is filled with a known pressure of parent gas (Figure 3).

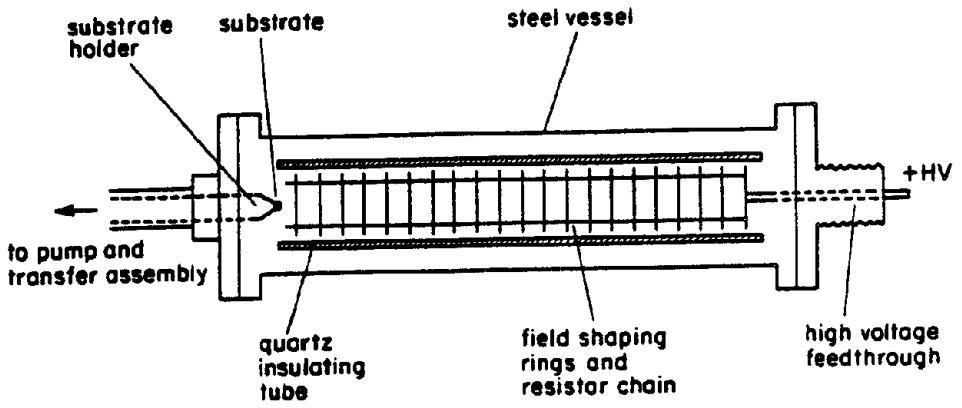


Figure 3. Schematic diagram of the chamber used to collect radiodecay products.

The daughter ions drift and deposit onto a pure substrate which is held at a small negative bias. Ideally, all or most of the daughter ions produced by the disintegration will be collected on the surface layer of the substrate.

In order to investigate and optimize the proposed collection method, a test system is required. The system we have chosen is the detection of $^{133}$Cs produced by the single beta decay of $^{133}$Xe. Briefly, the chamber is filled with $^{133}$Xe which is known to undergo ordinary beta decay to $^{133}$Cs with a half-life of 5.25 days. The $^{133}$Cs daughters are then collected on the tip of Au ingots which initially contain no measurable amounts of $^{133}$Cs. The Au substrates are mounted on a transfer rod assembly which is evacuated and interfaced to the MPRI chamber for future analysis.

For the collection of $^{136}$Ba produced by the double beta decay of $^{136}$Xe, a 10 L drift chamber will be attached to a 355 L time projection chamber located in the Gotthard tunnel in Switzerland. The parent gas will circulate throughout the time projection chamber and drift chamber for several months, depositing $^{136}$Ba as previously described onto a Au substrate.

## Summary

We have demonstrated a sensitivity for the detection of In in Si of 9 ppt. At present, the major limitation of this value is the background level. Improvements in the apparatus are presently underway which will isolate and reduce noise sources. Our results indicate that the MPRI experiment is sensitive to 630 surface atoms. In order to perform the double beta decay experiment, it is necessary that the analysis technique be sensitive to at least 1100 surface atoms. Thus, at the present time, it still appears possible to determine double beta decay half-lives through daughter atom counting via MPRI. Coupling these results with the quantitative trace analysis of $^{133}$Cs (the beta decay product of $^{133}$Xe) that is in progress will complete the assessment of the feasibility of double beta decay detection.

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