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PHOTON BURST MASS
SPECTROMETRY--ULTRASENSITIVE DETECTION OF
RARE ISOTOPES

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Photon burst mass spectrometry--ultrasensitive detection of rare isotopes

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ABSTRACT

Progress is reported on the development of a new technique for measurement of trace levels of radioisotopes which is based on fluorescence detection of output from a mass spectrometer. Significant achievements include the observation of fluorescence and burst signals from Kr isotopes, including enriched samples of ^{85}Kr with a 4-collector system. An abundance sensitivity is demonstrated with ^{83}Kr and ^{85}Kr .

Keywords: krypton, mass spectrometry, photon burst, rare isotope detection

1. INTRODUCTION

Many long lived radioisotopes can be used in dating, tracer and monitoring applications. Traditional radioactive decay methods are not well suited for such isotopes because of the prohibitive sample sizes and/or measurement times required. Many of these radioisotopes have abundances of 10^{-11} to 10^{-16} , ruling out direct atom counting by conventional mass spectrometers, which have abundance sensitivities in the 10^{-6} range, limited by molecular isobars and tails of adjacent isotopes. Tandem accelerator mass spectrometry (TAMS) has been used very effectively for direct counting of such rare isotopes as ^{14}C , ^{10}Be , and many others. However, TAMS is not able to measure noble gas rare isotopes such as ^{39}Ar , ^{81}Kr and ^{85}Kr which are important for oceanography, groundwater dating and environmental monitoring applications.¹

Photon burst mass spectrometry (PBMS) combines a conventional mass spectrometer with photon burst detection, a highly isobaric and isotopically selective process based on the principle that a single atom or ion in an optically isolated electronic state can absorb and emit a large number, or burst, of photons as it passes through a resonant laser beam.^{2,3} The basic principles of PBMS have been examined in detail,⁴⁻⁶ and it is expected that when fully developed it will be able to measure isotopes of about half the periodic table at abundances in the 10^{-11} to 10^{-16} range. Presently the research has focused on ^{85}Kr , which has a half life of 10.76 years and a natural abundance of $\sim 10^{-11}$.

Figure 1. Photon Burst Mass Spectrometry apparatus for noble gas measurements.

2. PHOTON BURST DETECTION OF KRYPTON

The PBMS instrument is illustrated in Fig. 1. It consists of a microwave ion source operating at 2.45 GHz,⁷ a 126° magnetic sector with a radius of 1.5m,⁸ a decelerator to slow the exiting ions from 50 keV to 400-2000 eV, a charge exchange cell to convert Kr or Ar ions to metastable atoms, and a photon burst detector assembly consisting of 4-10 light collectors and photomultiplier tubes. The atomic excitation is provided by a tunable diode laser at 811 nm. Performance of the mass spectrometer and initial studies of the charge exchange process are discussed elsewhere.⁹ We use Rb in the charge exchange cell and typically achieve about 90% neutralization of the Kr ions. For the data in this paper, 4 light collectors were used in the detection region. A gas feed manifold allows us to ionize Kr gas in a N₂ or Ar buffer gas and leak in enriched samples of ⁸⁵Kr at abundance levels ranging from 10⁻³ down to background levels (~10⁻¹¹).

With this system we have seen fluorescence and burst signals from all the major and minor isotopes of krypton as well as from enriched samples of ⁸⁵Kr. The spectra of the ⁸⁴Kr resonance in Fig. 2 illustrate the significant reduction in background achieved by requiring time correlated bursts on multiple collectors as compared to the fluorescence on a single collector. Although the signal level drops due to decreased detection efficiency, the signal-to-background ratio increases dramatically. The burst peak is also somewhat narrower than the fluorescence peak. Note that the various possible time correlated burst conditions are identified by a "clip level" (required number of photons detected on a single collector) and a "coincidence level" (required number of detectors producing a time correlated signal.) For example a "clip 2, coincidence 3" condition corresponds to bursts of at least 2 photons being detected on at least 3 collectors in the correct time sequence. This scan was obtained by ramping the final atomic beam energy through ~20 V in 100 seconds with 0.1 seconds integration per point.

After adjusting system parameters to minimize the energy spread of the krypton beam, we typically have a FWHM of about 3 eV. The ⁸³Kr fluorescence spectrum shown in Fig. 3 has a FWHM of 2.7 eV which is equivalent to a frequency FWHM of about 115 MHz. A large contribution to this energy spread can probably be attributed to a >2 volt ripple between the source and the deceleration electrodes due to ground loops.

Figure 2. Fluorescence and burst spectra of ⁸⁴Kr.

Figure 3. Fluorescence spectrum of ⁸³Kr. The FWHM energy spread is 2.7 eV.

3. MINIMUM ISOTOPIC ABUNDANCE SENSITIVITY MEASUREMENTS

Krypton-83 was used in an initial attempt to experimentally determine a limit on the isotopic abundance sensitivity of the PBMS instrument. To obtain the data in Fig. 4, Kr was ionized in an Ar buffer gas yielding 0.89 nA of ^{83}Kr in the detection system. A time correlated on-resonance burst signal (clip 2, coincidence 4) was integrated for 1000 seconds. Then the beam energy was raised 17 V, corresponding to a detuning of -724 MHz, and the background signal was integrated for 1000 seconds. The 1.44×10^5 counts on resonance compared to the 11 counts off resonance gives a signal-to-background ratio of 1.31×10^4 . The measured minimum isotopic abundance sensitivity obtained by scaling this number by the isotopic abundance of ^{83}Kr (.115), is 8.8×10^{-6} . If we scale, in addition, by the total Kr current when running pure krypton in the source ($\sim 13 \mu\text{A}$), the equivalent abundance sensitivity is 5.3×10^{-9} . The latter step is justified because we have seen no evidence of increased background signal off resonance with higher Kr current. The 11 counts of background were due mainly to the 70,000 background counts per second on each collector arising from dark counts in the photomultiplier tubes ($\sim 7,000$) and stray laser light.

This experiment was later repeated using an enriched sample of ^{85}Kr . This time N_2 was used as the buffer gas, since the ^{85}Kr sample had an N_2 buffer. The beam currents were 3.5 pA and $1.84 \mu\text{A}$ for ^{85}Kr and total Kr respectively. The signal-to-noise for one measurement consisting of 100 seconds on resonance and 100 seconds off resonance was 4.4, and was about the same for total fluorescence and burst measurements. This corresponds to an abundance sensitivity of 4.3×10^{-7} . Once again if pure Kr were used in the source ($17 \mu\text{A}$), this is equivalent to an abundance sensitivity of 4.2×10^{-8} .

In a final experiment the burst signal from 55 pA of ^{85}Kr was compared to the background signal level with $17 \mu\text{A}$ of Kr and the mass separator and laser tuned to look at ^{85}Kr . This yielded a signal-to-noise ratio of 200 corresponding to an abundance sensitivity of 1.6×10^{-8} for a 100 second measurement period.

Figure 4. The photon burst signal from 0.89 nA of ^{83}Kr for 1000 sec. on resonance and 1000 sec. off resonance. Burst condition is clip 2, coincidence 4.

Figure 5. A comparison of model predictions and experimental results for ^{84}Kr at several burst conditions and 2 different laser powers.

4. COMPARISON WITH MODEL

Two models have been developed to help analyze the photon burst detection process. The first is a spreadsheet model based on average quantities, and the second is a detailed, and much more time consuming, Monte Carlo simulation. Since there is good agreement between the two models, we feel confident in using the spreadsheet model for real time analysis and feedback. The diameter of the ion beam is the only adjusted parameter. Experimental agreement with the model for a series of measurements on ^{84}Kr at two different laser powers is shown in Fig. 5. Agreement is better for the lower power, but is reasonable in both cases, with the exception of the high clip/coincidence conditions where the signal was small.

5. RECENT EFFORT

Recent work has focused on increasing the detection efficiency and reducing the background levels, thereby increasing the sensitivity. Specifically, six additional detector assemblies have been installed along with their associated electronics. This will allow us to reduce the clip level for individual detectors while maintaining low background by requiring a higher coincidence level. Secondly, a new deceleration lens has been designed that will allow us to run a 2 keV beam improving the spatial and frequency overlap of the atomic and laser beams. Both changes are expected to lead to significantly larger overall efficiency.

The background count rate has been decreased in two ways. More efficient cooling of the photomultipliers has dropped the dark count rate from ~7,000 counts per second on each tube, to about 1,000 counts per second. Spatial filtering, higher quality optics and better light baffling design have all contributed to a drop from >60,000 background counts per second (at 100 μW of laser power), due to scattered laser light, down to ~2,000 counts per second. With future changes we plan on reducing this number another order of magnitude.

6. SUMMARY

Significant progress has been made in characterizing and testing the capabilities of the PBMS instrument. In the near future the 10-detector system will be on line and ready for further testing. Based on model predictions, this system will be able to perform isotopic analysis at the 10^{-11} level.

6. ACKNOWLEDGMENTS

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