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TITLE

PROSPECTS FOR LARGE DYNAMIC RANGE ISOTOPE ANALYSIS USING PHOTON BURST MASS SPECTROMETRY

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Prospects for large dynamic range isotope analysis using photon burst mass spectrometry

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ABSTRACT: Prospects and progress on the use of photon burst detectors in conjunction with a mass spectrometer are described. It is projected that isotope ratios in the 10^{-11} to 10^{-15} range could be measured on a photon burst mass spectrometer without any preenrichment steps.

1. INTRODUCTION

In the last decade advances in Accelerator Mass Spectrometry (AMS) have made possible, for the first time, direct atom counting of isotope ratios in the 10^{-11} to 10^{-15} range. As a consequence, new applications in dating have developed based on ^{14}C, ^{10}Be, and a few other elements with especially favorable properties. Laser methods of single atom detection, including Resonance Ionization Spectroscopy (RIS) and the Photon Burst method (Greenlees 1977), offer avenues to extend the new dating technology to many other elements in the periodic table. The noble gases, for example, have a wide range of dating applications (Lehmann 1984) and cannot be done by AMS.

Krypton isotope ratios at the 10^{-11} level have been measured recently using RIS (Thonnard 1988). However, the process required extensive preenrichment steps which attenuate the stable isotopes by 10^8, due to the limited isotopic selectivity of pulsed RIS coupled with a quadrupole mass spectrometer. The higher isotopic and isobaric selectivity available with the photon burst method should make possible measurements to the 10-15 level without any preenrichment steps. In this paper, the prospects and progress to date in Photon Burst Mass Spectrometry (PBMS) are reviewed. Sample calculations for argon and krypton are presented.

2. PHOTON BURST MASS SPECTROMETRY

Photon Burst Mass Spectrometry has been described in some detail by Keller (1981) and Fairbank (1987). It involves the coupling of one or more photon burst detectors with a high quality mass spectrometer. Each device has moderate selectivity, so that a large dynamic range is achieved by multiplication of discrimination factors.

Two types of PBMS instruments are illustrated in Figure 1. In (a) photon burst detectors provide additional isotopic and isobaric selectivity to enhance the capabilities of a conventional magnetic sector instrument. These photon burst detectors are operated in high vacuum and detect fast ions from the magnetic sector or fast metastable neutral atoms created by charge exchange in an alkali vapor cell. Since the ions arrive continuously, a CW dye laser is used. The collinear laser and ion beam geometry is used for two reasons. First it naturally accomodates multiple photon burst detectors, with efficient usage of dye laser power. Second, the large Doppler shift in the collinear geometry results in a compression of the residual Doppler width and introduces an additional frequency shift (an artificial isotope shift) between isotopes (Anton 1974). Thus it allows more complete coverage of the velocity distribution while enhancing the isotopic selectivity.

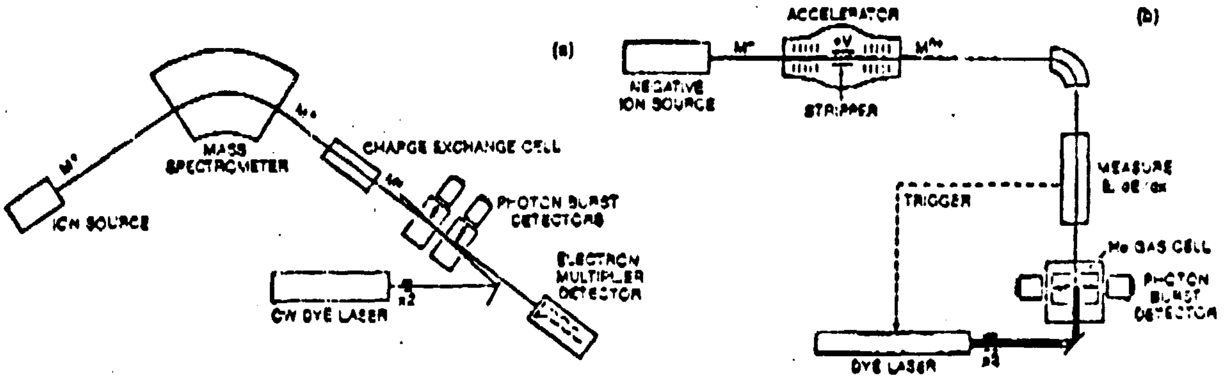

Figure 1. Photon Burst Mass Spectrometer systems: (a) with a conventional magnetic sector instrument and (b) with an acclerator mass spectrometer.

In Photon Burst Accelerator Mass Spectrometry (b), the photon burst detector serves as a means of elemental confirmation, and is used mainly to eliminate interferences from atomic isobars. In this case the ions are too fast for direct photon burst detection and must be stopped in helium gas. Large bursts can be generated in this case because the atoms diffuse away slowly. Either CW or flashlamp-pumped dye lasers, triggered by the AMS particle detectors, can be used.

The basic principle of a photon burst detector is that a single two-level atom can be excited and spontaneously emit many photons per second, when excited on resonance by a dye laser beam of moderate intensity (~mW/mm^2). Typically hundreds of photons are emitted by a single atom during a 1-10 microsecond transit time through a photon burst detector. For fast atoms in vacuum an elliptical cylinder light collector is preferred. With >50% light collection efficiency and 10-20% photomultiplier quantum efficiency, the detection of five or more photons from a single atom or ion is certainly feasible. Stray light counts can be kept to a manageable level, 10^{-12} to 10^{-14} of the incident flux, if reflections and beam quality are adequately controlled.

The vacuum photon burst detector is especially attractive for large dynamic rang. isotope analysis because for detected bursts of n photons the signal spectrum is proportional to a Lorentzian to the nth power. Thus the tails from stable isotopes go down as the isotope shift to the 2nth power. Furthermore, discrimination against molecular isobars is very high because molecules cannot generate a burst. After the first emitted photon, a molecule has a high probability of being in a different vibrational or rotational state and therefore transparent to the laser. Finally, with coincidence requirements in multiple detectors, dark current and statistical fluctuations from multiple atoms of stable isotopes can be eliminated completely.

3. PROSPECTS FOR NOBLE GAS MEASUREMENTS

Photon Burst Mass Spectrometry is potentially applicable to up to half of the elements of the periodic table using laser wavelengths in the 200-900 nm range (Fairbank 1987). To illustrate the considerations which go into determining the prospects for doing PBMS on a given element, the noble gases, especially ^{85}Kr, ^{81}Kr and ^{39}Ar, will be considered in more detail in the following discussion. Note that the modern isotopic abundances of these isotopes, 1×10^{-11}, 5×10^{-13} and 8×10^{-16}, respectively, span the range of our discussion. While ^{39}Ar may be the most difficult to measure, it is also the case where the need is greatest since the current RIS method (Thonnard 1988) is limited to Kr and Xe.

In Table 1 the parameters and results for several sample problems are summarized. We have considered photon burst detectors on two types of magnetic sector mass spectrometers which are in use at Los Alamos National Laboratory. The static mass spectrometer can have high efficiency (>50%) since the atoms have multiple chances to be ionized. However, low throughput (0.4nA maximum) should limit its applicability to the 10^{-11} level. Thus a first level of enrichment, e.g., like that being done for Kr by Lehmann (1988), will be required before ^{81}Kr and ^{39}Ar can be counted in this instrument. The second type of mass spectrometer is a tandem magnetic sector. It operates in the dynamic mode with a steady flow of sample through the source. New high current ion sources (4 μA, >10% efficiency) developed for nuclear physics (Kirschner 1981) will have to be adapted to the present instrument in order to reach the 10^{-15} level.

Table 1. Calculated PBMS signals for modern air samples of rare noble gas isotopes. The results quoted are for the dynamic mass spectrometer. The predicted signals for the static mass spectrometer are identical to those for the dynamic mass spectrometer, and the backgrounds are even lower.

Isotope	^{85}Kr	^{81}Kr	^{39}Ar
Isotope ratio	1×10^{-11}	5×10^{-13}	8×10^{-16}
Static Mass Spectrometer:			
Kr/Ar sample size (cm^3 STP)	2×10^{-5}	2×10^{-5}	2×10^{-5}
Measurement time (hours)	30	30	30
Current (nA)	0.4	0.4	0.4
Preenrichment required	none	20	10^4
Dynamic mass spectrometer:			
Kr/Ar sample size (cm^3 STP)	10^{-4}	2×10^{-3}	1.25
Measurement time (hours)	0.3	0.6	30
Current (nA)	40	400	4000
Preenrichment required	none	none	none
Two detectors:			
Energy (eV)	44	44	20
Background bursts	10-11	10-11	10-7
Signal bursts	20	20	18
Ten detectors:			
Energy (eV)	1100	1100	500
Background bursts	10-4	10 4	10^{-2}
Signal bursts	100	100	90

The details which lead to the results presented in Table 1 are discussed at a greater length in a previous paper (Fairbank 1987). For the case of two detectors a positive signal is defined as recorded bursts of 5 or more counts in both detectors. For the ten detector case, the registration of single counts in seven or more detectors is required. In the latter case the required burst size is lower because the atoms are travelling five times faster. However, five times as many rare atoms are detected due to Doppler width reduction. In all cases the dominant background was stray light. A modest stray light reduction factor of 10^{-12} was assumed. Note that 10-100 rare atoms are counted in these examples with essentially zero background. For comparison, the initial samples contained 5400 and 27,000 rare atoms in the static and dynamic cases, respectively.

The one assumption in these calculations which is not well understood and documented concerns the charge exchange process. If the wings of the Doppler distribution are substantially altered by charge exchange collisions, a few stable isotopes might be Doppler shifted into resonance, resulting in a background of significance. The handwaving argument against this scenario says that most charge exchange collisions occur at large impact parameter due to the large cross section ($>10^{-15}$ cm^2 or higher) for the process (Anton 1974). Such collisions have small angular deviations and minimal spread in kinetic energy transfer. The few atoms which have more direct collisions will be deflected through larger angles and will tend to miss the photon burst detectors. Charge exchange has been widely used in high resolution spectroscopy of rare nuclei. However, to our knowledge, the wings of the lines have never been investigated in detail. Further studies of this type are clearly needed.

4. CURRENT WORK

We have recently demonstarted the fundamentals of Photon Burst Mass Spectrometry using a Colutron ion source with a Wien filter mass spectrometer. Photon bursts of six counts or more at 279.5 nm have been recorded from single ^{24}Mg, ^{25}Mg and ^{26}Mg ions exiting the spectrometer at 200 eV energy. Tuning the mass filter successively eliminated adjacent mass peaks as expected. To our knowledge this is the first time that single fast ions have been seen by the Photon Burst method. Improvements are being made so that detailed studies of the isotopic discrimination obtained by this method can be performed.

5. CONCLUSIONS

Photon Burst Mass Spectrometry is a relatively new and untried method which may complement and extend the impressive achievements of Resonance Ionization Mass Spectrometry and Accelerator Mass Spectrometry in the field of large dynamic range isotope analysis. Theoretical predictions indicate that measurements in the 10^{-11} to 10^{-15} range are possible in a reasonable period of time with zero background. Experimentally only the very first demonstrations of PBMS with stable isotopes have been completed.

6. ACKNOWLEDGEMENTS

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