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TITLE DETECTION OF PHOTON BURSTS FROM SINGLE 200 eV Mg IONS,
PROGRESS IN PHOTON BURST MASS SPECTROMETRY

AUTHOR(S) W. M. Fairbank, Jr., R. D. LaBelle, R. A. Keller,
and E. P. Chamberlin

SUBMITTED TO Proceedings of the 9th International Conference on
Laser Spectroscopy, June 19-23, 1989, New Hampshire

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Los Alamos National Laboratory
Los Alamos, New Mexico 87545

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Detection of Photon Bursts from Single 200eV Mg Ions, Progress in Photon Burst Mass Spectrometry

W. M. Fairbank, Jr. and R. D. LaBelle

Department of Physics, Colorado State University
Fort Collins, CO 80523, USA

Richard. A. Keller and E. Philip Chamberlin

Los Alamos National Laboratory,
Los Alamos, NM 87545, USA

Laser Spectroscopy IX, Springer-Verlag, 1989, in press.

Modern atom counting methods, based on advances in laser and accelerator technology, provide a valuable complement to traditional decay counting methods for radioisotope dating and tracer work. Tandem Accelerator Mass Spectrometry (TAMS) has already had a large impact on ^{14}C dating and is beginning to provide new opportunities with ^{10}Be and several other isotopes. We report here on progress in the development of a laser-based technique, Photon Burst Mass Spectrometry,^{1,2} which is potentially capable of analyzing many of the elements which are forbidden in TAMS because they do not form negative ions. We are especially interested in the noble gases, which have a variety of potential scientific and environmental applications.³

Photon Burst Mass Spectrometry (PBMS) is based on the coupling of a highly selective laser single atom or ion counter, the photon burst detector, with a mass spectrometer, which serves as an on-line prefilter to reduce the abundant isotopes by 10^6 (Fig. 1). In two previous papers, calculations have been presented for several sample isotopes, which indicate that isotope ratios as low as 10^{-15} are measurable in 30 hours or less by the PBMS method.^{2,4} In the last year or so, our primary effort has been in demonstrating experimentally the parameters upon which these calculations were based and in designing and testing the important components of the proposed PBMS instrument. Progress on these goals is the major focus of this paper.

The photon burst detector is being developed and tested with a simple Colutron ion source, equipped with an ExB (Wien) filter. It provides mass-selected Mg^+ ions at 200 eV at a low current on the order of picoamps. The ions pass

Fig. 1. Photon Burst Mass Spectrometer.

collinearly through a photon burst detector with a focussed laser beam tuned to the $^2S_{1/2} - ^2P_{3/2}$ Mg^+ resonance at 279.5 nm. Focussing was required because the laser power, 40 μW , was insufficient to saturate the full diameter of the ion beam.

Results obtained with the mass filter off are shown in Fig. 2. In the total fluorescence signal, peaks are seen from all three isotopes, but a large stray light background (4×10^{-9} of the incident light) limits the signal-to-background ratio to about 2. Much greater contrast is seen in the burst signal, with nearly zero background. The steepness of the sides of the peaks indicates that the contributions from neighboring isotopes at line center are well below the stray light background. With measured experimental parameters and an estimated 7% collection/quantum efficiency, predicted maximum values for the two $^{25}Mg^+$ and $^{26}Mg^+$ peaks from left to right are 83, 1500 and 6450 bursts/sec, respectively. The predicted stray light background is 2.2 bursts/sec, including measured effects of double pulsing in the photomultiplier. All these predictions agree well (within a factor of two or so) with the observations. The derived average burst size for resonant $^{25}Mg^+$ and $^{26}Mg^+$ ions is, respectively 3.1 and 4.5 counts.

This is the first time that photon bursts from single ions, other than in a trap, have been observed. It is also the first time that superthermal particles have been detected by the photon burst method. It is worth noting that the high optical efficiency and burst sizes demonstrated in these experiments, as well as the good agreement of the experimental data with theoretical predictions, represent important milestones in the development and testing of the PBMS method. The relatively high stray light level in these experiments, due partly to light from the ion source, prevented a better demonstration of the isotopic selectivity of the photon burst method. Nevertheless, an abundance sensitivity of at least 10^3 is shown in Fig. 2. The calculated average background from neighboring isotopes is 10^4 lower than the stray light level, and contributed negligibly to background bursts.

Fig. 2. Fluorescence and photon burst spectra of Mg ions obtained with the Wien filter off: (a) total count rate, and (b) rate of bursts with 25 counts. Curve (c) represents the calculated burst rate if the photon arrival rate in (a) were uncorrelated, except for a 0.3% afterpulsing correction. From the left the peaks are: $^{24}Mg^+$, $^{25}Mg^+$ ($F''=2 \rightarrow F'=1, 2, 3$), $^{26}Mg^+$ ($F''=3 \rightarrow F'=2, 3, 4$) and

Recently we have installed an electrostatic bend between the ion source and the photon burst detector, as illustrated in the PBMS diagram (Fig. 1). This effectively eliminates background photons from ion source. A stray light level of 10^{-10} for the whole system has been achieved at 280 nm. In more careful work with the photon burst detector alone, 10^{-11} was measured at 280 nm and 10^{-12} with a clean TEM₀₀ beam at 633 nm. It is anticipated that proper spatial filtering of the beam with a pinhole or single-mode optical fiber will allow the latter level to be reached at all wavelengths. Thus we have demonstrated essentially all of the required features of the proposed photon burst detector.

The planned mass spectrometer for our full PBMS demonstration is a 126° magnetic isotope separator at Los Alamos National Laboratory.⁵ A microwave source for this instrument⁶ has been installed and tested. It produces a 10 μ A current in a 1-2 mm spot at the exit of the spectrometer with 50% overall ionization and transmission efficiency and reasonable divergence. An abundance sensitivity of 10^6 at the krypton masses has been measured. The background isobars observed at the 10^{-6} level pose no problem due to the high isobaric selectivity of the photon burst detector. Thus most of our assumed mass spectrometer specification are exceeded by this instrument.

Many of the important features of the Photon Burst Mass Spectrometry method, including selective counting of single fast ions, have now been confirmed experimentally. Progress is being made toward adapting an isotope separator at Los Alamos National Laboratory for the final demonstration of the full capabilities of the PBMS method. Further studies are still required on the most important remaining uncertainty in the method, the magnitude of longitudinal velocity changes during charge exchange to metastable atomic states. This is not thought to be a severe problem because most charge exchange collisions occur mainly at large impact parameters.

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