Detection of fast Mg ions using the photon-burst method

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Single magnesium ions at superthermal velocities (200 eV energy) are detected for the first time by the photon-burst method. An optical isotopic resolution of $> 10^3$ is demonstrated.

Direct atom counting with lasers is becoming an attractive choice for radiochemical dating applications utilizing long-lived isotopes, particularly when small samples or short measurement times are required. Recent advances in laser techniques project the ability to measure atomic isotope ratios in the 10^{-11} to 10^{-15} range without preenrichment steps.[1–3] In this paper we report the results of initial experiments that demonstrate the fundamental principle of one of these methods, photon-burst mass spectrometry[1,4] (PBMS); that is, the selective identification, by a photon burst detector, of fast ions emerging from a mass spectrometer.

In the photon-burst method, n photons of laser-induced fluorescence are registered in a short time as a single atom or ion passes through the detection region of a resonant laser beam. High discrimination against random backgrounds and the tails of nearby lines is obtained due to the nonlinearity of n-event Poisson statistics.[1,4–7] The photon-burst method has been used previously for ultrasensitive spectroscopy on thermal atoms[5–7] and cooled ions in a trap.[8,9] We report here the first observation of superthermal ions by this technique.

Due to ever-present inefficiencies in optical collection and photon detection, 20 or more photons must be emitted by an atom for every photon that is detected. Thus a "two-level atom" configuration is desired, in order that optical pumping does not prematurely terminate the absorption-emission cycle. This is the case for the $^2S_{1/2} \rightarrow 2P_{3/2}{}^{\circ}$ transition at 279.5 nm in the $^{24}Mg^+$ and $^{26}Mg^+$ isotopes of magnesium. In $^{25}Mg^+$, only the $F''=3 \rightarrow F'=4$ hyperfine line fulfills this criterion, and can produce a photon burst of maximal size.

Our experimental apparatus is shown schematically in Fig. 1. Magnesium ions are generated in a Colutron ion source at 200 eV energy. A Wien ($\mathbf{E}\times\mathbf{B}$) mass filter allows either the simultaneous passage of all three Mg isotopes or the specific selection of an individual isotope. The Mg^+ ion beam is collimated to 5 mm diameter with an einzel lens and aligned by x-y deflection plates to be exactly antiparallel to the laser beam in the photon-burst detector.

The laser beam is generated in a frequency-stabilized (<2 MHz linewidth) ring-dye laser (CR 699), equipped with a high-reflectivity end mirror, intercavity potassium dihydrogen phosphate (KDP) doubling crystal, and dichroic beam splitter to couple out only the second harmonic light.[10] Two mirrors are used to align the laser beam to the central path of the photon-burst detector, where the stray light level is minimum. Since the available laser power in the photon-burst detector, 40 μW, is not sufficient to saturate the full area of the ion beam, the laser is focused to a 60-μm waist radius with a 1-m lens. This produces a maximum intensity of 1.4 times the saturation intensity at line center for the even isotopes. When the laser radius is much smaller than the ion-beam radius, only a fraction of the ions pass through the detection volume, and angular alignment of the laser and ion beams to within the required 1 mrad can be difficult. These problems will be eliminated when a laser power of 5–10 mW becomes available with an improved doubling system.[10]

The $^{25}Mg^+$-ion current in these experiments is 30 pA, corresponding to 10^5 $^{25}Mg^+$ ions/sec through the focal area of the laser. Due to Doppler broadening only one-eighth of these ions actually interact with the laser at line center. Taking into account the ~ 1 μsec transit time of ions through the detector, the average number of resonant ions in the detection volume is determined to be ~ 0.01.

Our photon-burst detector consists of a 4.6-cm-long elliptical cylinder with flat end plates, both polished and coated to 70% reflectivity at 280 nm. The laser and ion beams pass through one line focus of the ellipse, and a 5-mm slit is placed at the other focus. A uv bandpass filter (84% transmission at 280 nm) and a photomultiplier tube (EMI 9328QA) are located directly above the slit. The combined collection efficiency of the ellipse and filter is about 35% over the central 3.4 cm of beam path. With an estimated photomultiplier quantum efficiency of 15% (counts per incident photon), the total detection efficiency for emitted photons is approximately 5%. The ellipse, photomultiplier tube, and dynode divider chain are mounted inside the vacuum chamber.

The detection electronics for these initial experiments consist of an amplifier-discriminator unit (manufactured by Princeton Applied Research), a resettable counter (the clip count extender unit for a Malvern digital correlator) to look for $\geq n$ counts (a "burst") during a sample period,

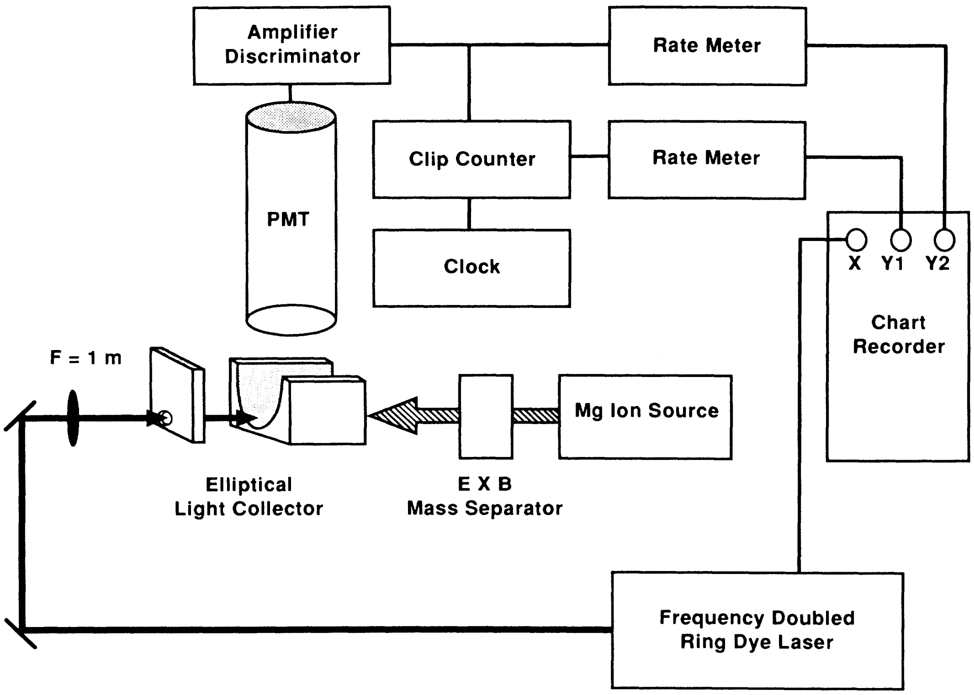

FIG. 1. Experimental apparatus.

and a clock which marks off 1.5-μsec sample periods. Two rate meters, monitoring the total count rate and the rate of bursts, supply analog signals to the y inputs of a dual-channel x-y recorder. The abscissa signal is obtained from the dye-laser scan drive.

Reduction in stray laser photons reaching the photomultiplier through various scattering paths is achieved by using a Brewster-angle input window and collimating apertures before and after the photon-burst detector. However, in these experiments, aperture position, laser-beam quality, and laser-beam termination were not optimal, and the residual stray light level of $\sim 10^{-9}$ counts per incident photon constituted a major background. Also, since there was no bend in the ion beam, stray light from the ion source contributed comparably to the background. In later experiments with the photon-burst detector alone, a stray light level of 5×10^{-13} was measured with a TEM_{00} He-Ne laser beam. Our uv laser beam, even without spatial filtering, yielded a stray light level of 1.5×10^{-11}. Thus a great reduction in background is anticipated with the installation of a bend in the ion source in the near future.

Initial results obtained with the mass filter turned off are shown in Fig. 2. The tail of the abundant $^{24}Mg^+$ isotope and peaks from $^{25}Mg^+$ and $^{26}Mg^+$ can be seen in the total count rate spectrum, with relatively high noise content due to the large stray light background. When the rate of bursts with ≥ 5 counts is monitored, large signals are seen on the even isotopes and only one of the hyperfine components for $^{25}Mg^+$. Note that the background level is greatly diminished compared to the peak values. The burst rate expected from random statistics on the total count rate, Fig. 2(a), taking into account the measured rate (0.3%) of double pulsing in the photomultiplier, is indicated in Fig. 2(c). In the wings of the lines, the observed background of 1–4 bursts per second, Fig. 2(b), is in excellent agreement with the predictions of Fig. 2(c). In addition, the off-scale burst signals recorded at the peaks are more than an order of magnitude larger than random statistics predict and therefore are a clear indication of the detection of photon bursts from single ions.

Simple theoretical estimates,[4,11] based on atomic properties of Mg^+ and the experimental parameters listed above, yield peak values of 2000 and 6500 bursts per second for $^{25}Mg^+$ ($F''=3\rightarrow F'=4$) and $^{26}Mg^+$. Uncertainties in the parameters and small angular differences in the paths of these two isotopes, arising from the residual magnetic field in the Wien filter, make the uncertainty in these predictions at least a factor of 2. Nevertheless, reasonable extrapolations of the observed peak values (2000–6000 bursts/sec) are in acceptable agreement with theory. The limited burst signals observed on the $^{25}Mg^+$ peak on the left are the result of absorption-emission cycles terminated by optical pumping into noninteracting F and m_F states.

The isotopic selectivity demonstrated in Fig. 2, considering the background of 1–4 bursts/sec and the peak heights of 2000–6000 bursts/sec, is approximately 10^3. This selectivity is clearly limited solely by the stray light background and not by fluorescence from the tails of neighboring isotopes, which are seen to be very steeply declining in the burst signal. We expect that with the anticipated reduction in stray light we will be able to improve the isotopic selectivity of our photon-burst detector by many orders of magnitude.

Figure 2 also illustrates the enhanced isotopic resolution obtained in collinear laser spectroscopy.[4,12] For example, 77% of the observed isotope shift between the two largest peaks comes from the different Doppler shifts of isotopes accelerated to the same energy. For elements with smaller natural isotope shifts, this artificial isotope shift will be an important factor in achieving selective excitation of rare isotopes.

A scan taken with the Wien filter turned to pass an individual isotope $^{25}Mg^+$ is presented in Fig. 3. In the total count rate spectrum, the largest signal arises from the tail of the mass peak for the most abundant (80%) isotope $^{24}Mg^+$. In the burst spectrum, however, the second $^{25}Mg^+$ peak dominates. This illustrates an additional way the photon burst detector can provide discrimination against the tails of neighboring mass spectrometer peaks. In this case, these $^{24}Mg^+$ ions pass at an angle through the laser beam and do not stay in the detection volume long enough to generate a full burst. Notice that since the relative heights of the two largest peaks in Figs. 3(a) and 3(b) are opposite, random statistics alone cannot explain the observations. Since stray light still dominates the background level, the Wien filter provides, at present, no additional enhancement in the observed isotopic selectivity. This illustrates the importance of stray light reduction to the overall success of PBMS.

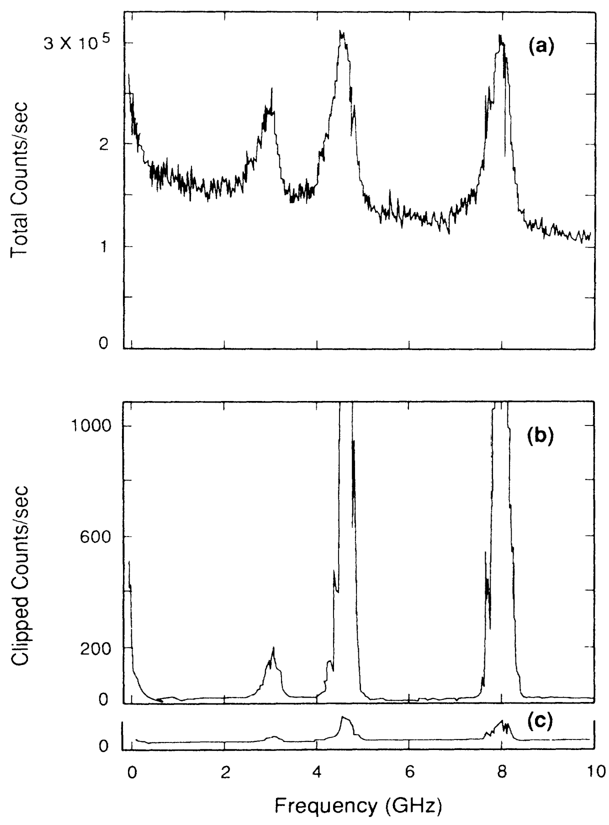

FIG. 2. Fluorescence and photon-burst spectra of Mg ions obtained with the Wien filter off: (a) total count rate and (b) rate of bursts with ≥ 5 counts. Curve (c) represents the calculated burst rate under the assumption that the photon arrival rate in (a) is uncorrelated, except for a 0.3% afterpulsing correction. The vertical scale of (c) is the same as for (b). From the left the peaks are $^{24}Mg^+$, $^{25}Mg^+$ ($F''=2 \rightarrow F'=1,2,3$), $^{25}Mg^+$ ($F''=3 \rightarrow F'=2,3,4$), and $^{26}Mg^+$.

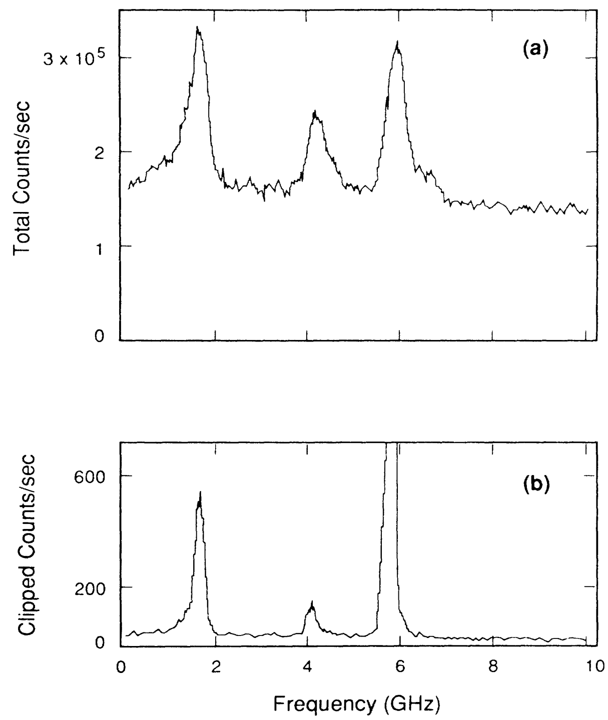

FIG. 3. Mg-ion spectra obtained with the Wien filter tuned to select $^{25}Mg^+$: (a) total count rate and (b) rate of bursts with ≥ 6 counts. The small $^{24}Mg^+$ peak on the left remains because conditions of low mass resolution (small **B** and **E**) were used. The $^{26}Mg^+$ peak expected at the 9.2-GHz position is absent.

In summary, the passage of single fast ions has been recorded for the first time by the photon-burst method. The general features and signal levels of the observed photon-burst spectra are in reasonable agreement with expectations. The demonstrated optical isotopic selectivity of $> 10^3$ is a first step toward the 10^6-10^9 level required for successful isotope ratio measurements in the 10^{-11} to 10^{-15} range by photon-burst mass spectrometry.[4]

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