An On-Line Laser Ion Source Based on Resonance Photoionization in a Gas Cell

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(Received 9 March 1994)

First results obtained with a new type of on-line laser ion source based on resonant photoionization in a buffer gas cell are presented. The main advantages of this new target-ion-source system are its high efficiency, its selectivity, its short delay time, and its universality. This is demonstrated with the production of an isotopically and isobarically pure beam of ^{55}Ni ($T_{1/2} \approx 200$ ms).

PACS numbers: 29.25.Ni

Already during the first conference on nuclei far from stability dealing with the question "Why and how should we investigate nuclides far off the stability line" (Lysekil, 1966 [1]), it was realized that on-line mass separators are powerful tools for the exploration of the nuclear chart up to its boundaries. Three decades of experimental work at on-line mass separators have resulted in a vast improvement of the quality of the mass-separated beams, mainly because of developments in the target-ion-source systems [2]. Nowadays such beams are also injected in postaccelerators [3] and a wealth of new results in many different fields like nuclear physics, astrophysics (synthesis of unstable nuclei), and nuclear medicine are expected [4]. For the study of very unstable nuclei, the target-ion-source system should fulfill three criteria: the ionization efficiency should be high ($> 10\%$), the delay time short (< 100 ms), and, since in most cases the nuclear reactions are only partly selective, the element selectivity should be high (suppression of the isobaric contamination > 100). With the presently available techniques only about 30% of the elements can be produced according to these three criteria: alkali and alkaline-earth elements, noble gases, halogens, and the elements Ga, In, Tl, Zn, Cd, and Hg [2]. For all these elements, different target-ion-source combinations have to be used. In this paper we present a new and general concept for a laser ion source that will allow the production of about 80% of the elements meeting all the above-mentioned criteria. We will discuss the underlying principles and present the results of a feasibility study performed at the LISOL facility [5], whereby the production of a pure beam of radioactive ^{55}Ni ($T_{1/2} \sim 200$ ms [6,7]) ions was achieved and the ^{55}Ni half-life was remeasured.

An elegant way to obtain an extremely high degree of selectivity is using resonance photoionization with lasers. Stepwise excitation of the atoms followed by photoionization in the last step delivers isotopically pure ions. With today's commercially available laser systems it becomes, in principle, possible to obtain an ionization efficiency near unity for about 80% of all elements. So far only pulsed lasers can deliver the high power needed to achieve a high efficiency. These lasers have one major drawback, namely their low-duty cycle. To solve this duty cycle problem, Kluge *et al.* [8] and Andreev *et al.* [9] proposed to store the atoms of interest in a moderately heated cavity connected to a target or an external oven. This technique has now been used at several on-line mass separators [10–12] showing its high potentiality. But the target-storage system has its limitations: only elements diffusing rapidly out of the target and remaining volatile in the cavity are efficiently ionized; and second, unwanted thermoionization sometimes seriously contaminates the ion beam.

The laser ion source we propose combines the fast and universal thermalization of reaction products in a gas, used in He jet and ion-guide techniques [13], the high selectivity of resonance photoionization, and, to overcome the low-duty cycle problem, the ion-storage capacity of noble gases. A layout of the target-ion-source system is given in Fig. 1; the numbers mentioned in the figure are also used in the text. The cyclotron beam (11) enters the source through a 5 μm Havar window (3) and hits the target (4). The reaction products recoil out of the target and thermalize to a 1^+ charge state in the helium gas (typically 500 mbar pressure). After a few milliseconds all the ions are neutralized [14]. This short ion-survival time, which forms a major limitation for the efficiency of the ion-guide technique, is believed to be due to the plasma induced by the cyclotron beam [14]. As will be shown below, the ion-survival time in a gas cell without cyclotron beam is much longer. Therefore the cyclotron beam is bunched in "50 ms on" and "50 ms off" periods. About 5 ms after the end of the cyclotron beam pulse, the source is free of charged particles and a laser pulse (10) is fired axially into the laser ion source. Essentially all atoms in the laser beam path ($\phi = 2$ mm) are ionized and are transported by the helium flow (0.12 l/s) as ions through the exit hole (6). The ions are accelerated towards the skimmer (7) (negatively polarized by a few hundred volts relative to the gas cell) where most of the helium is

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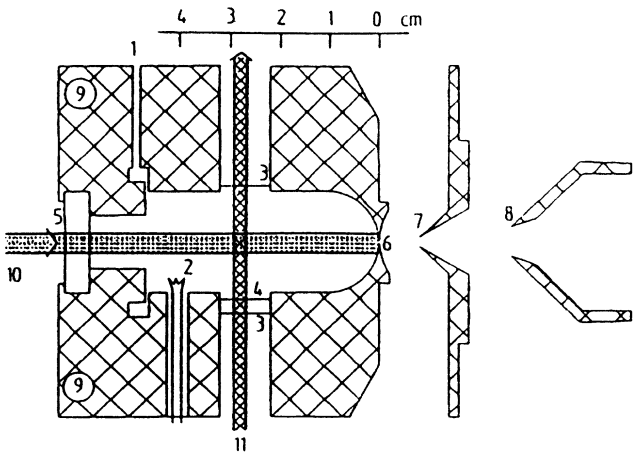

FIG. 1. Schematic layout of the laser ion source. The chamber was made out of stainless steel and was sealed with indium fittings. The numbers in the figure represent the following: (1) helium inlet; (2) nickel filament; (3) 5 μm Havar window; (4) target; (5) quartz window; (6) exit hole (⌀ 0.5 mm); (7) skimmer (⌀ 1.5 mm); (8) extraction electrode (⌀ 5.5 mm); (9) water cooling; (10) laser beam; (11) cyclotron beam.

removed by differential pumping. The ion beam is further accelerated towards the extraction electrode (8) and the analyzing magnet of the mass separator. The timing of the laser and the cyclotron beam is shown in Fig. 2. By choosing the volume of the ionization region and the exit-hole diameter in such a way that the evacuation time (t_{ev}), the time between subsequent laser pulses (t_{rep}), and the ion survival time (t_{ion}) satisfy the condition

$$t_{ion} > t_{ev} > t_{rep},$$

the low-duty cycle problem is overcome. In this case every atom in the laser-interaction region is irradiated at least once by a laser pulse and every photoion can survive its transportation towards the exit hole.

First, the resonance ionization process was studied. The possibility of laser excitation and ionization of reaction products, thermalized in a buffer gas cell, has recently been demonstrated [15–18]. Here we used a two-step, one-color photoionization technique, whereby the same laser wavelength is used for the excitation and the subsequent ionization of nickel atoms. About 4.5 mJ per pulse of UV laser light (10 ns pulse duration, 2 mm diameter) was obtained after frequency doubling of the output of a pulsed dye laser operated with DCM, which was pumped by the 532 nm output of a Q-switched 10 Hz Nd:YAG laser. For the strongest transitions observed (λ = 322.698 nm, λ = 324.846 nm, ...) the ionization step was readily saturated with the available laser power density (for more details, see [19,20]).

The performance of the laser ion source was tested with stable nickel atoms—produced in the cell by heating a Ni filament (2 in Fig. 1)—as well as with ^{55}Ni produced on-line using a 27 MeV ^{3}He beam on an enriched ^{54}Fe (3 mg/cm^2) target. Figure 3 shows the signal of stable nickel ions (mass 58) produced in the gas cell by resonant laser light and detected in the collection chamber of the mass separator [5]. The amplitude of the ion signal (proportional to the number of detected ions per time unit) is shown versus the time elapsed since the impact of the laser beam pulse in the gas cell. Note that the time scale can be related to a position inside the gas cell: for example, the ions created at the crossing point of the laser and cyclotron beam (2.5 cm from the exit hole) are detected 20 ms after the laser has fired. This figure demonstrates the long ion-survival time in the absence of the cyclotron beam. The recoil energy of the ^{55}Ni atoms was 1.4 MeV leading to an effective target thickness of 0.43 mg/cm^2 and a primary production rate of 4.2 $\times$ 10^4 atoms/μC of ^{55}Ni recoiling out of the target. The cross section of this reaction (1.4 mb) has been calculated with the HIVAP code [21]. The uncertainty on this number could be rather high. After mass separation, the radioactive isotopes were implanted in a tape behind which a silicon surface barrier

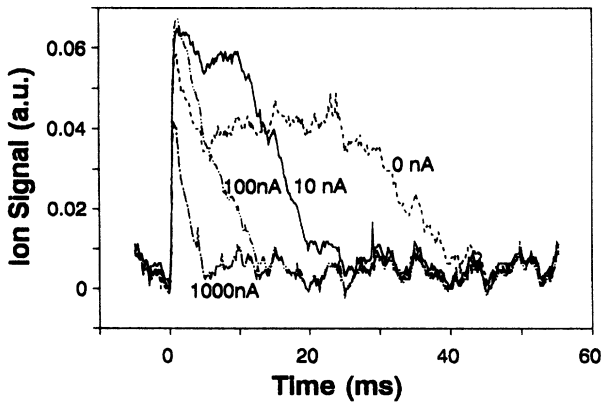

FIG. 2. Timing of the laser beam and the cyclotron beam used for the production measurements of ^{55}Ni.

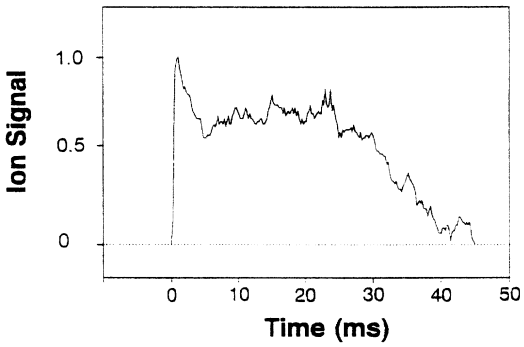

FIG. 3. Amplitude of the ion signal produced by mass-separated stable nickel photoions (proportional to the number of ions impinging on the detector) as a function of the time elapsed since the impact of the laser pulse. Here the pressure of helium is 100 mbar. The ion signal is given in arbitrary units.

detector (400 μm thickness, 150 mm^2 active surface) was positioned for the detection of β particles. A production rate of 54 atoms/μC was measured when the laser was tuned at the 322.698 nm line and with a target chamber pressure of 500 mbar. This corresponds to a total efficiency of 0.13%. A lower limit of 64 for the selectivity, defined as the ratio of the production rates with the laser on and off resonance, was measured, a limit which was almost exclusively determined by the ambient radioactivity seen by the β detector. When tuning on the 324.846 nm line—ionization from the lowest metastable state [19]—a production rate of 11 atoms/μC was observed. To clarify the 0.13% total efficiency, we summarize in Table I the different processes that determine the efficiency in our laser ion source.

In order to identify firmly the radioactivity at mass 55, we performed half-life measurements. During these measurements the separator beam was pulsed (1 s beam on, 2 s beam off) and for every β event the energy and the time elapsed since the start of the beam on period were recorded. Figure 4 shows the time spectrum obtained at mass 55. Using a Poisson optimization procedure, we obtained a half-life of 155 $\pm$ 10 ms which is in disagreement with the values of Hornshøj *et al.* (189 $\pm$ 5 ms) [6] and of Äystö *et al.* (208 $\pm$ 5 ms) [7]. We checked our setup in a subsequent experiment by measuring the half-life of ^{54}Co using a conventional ion guide, yielding a half-life of 192 $\pm$ 2 ms in agreement with the literature value: 193.23(14) ms [22]. The reason for the discrepancy must be sought in unwanted and longer-living activity present in the previous experiments [6,7]. In all three experiments, the reaction used is ^{3}He on ^{54}Fe. The experiment of Hornshøj *et al.* is an in-beam experiment without mass separation; the half-life

TABLE I. The estimated efficiency (%) of the relevant processes that contribute to the total efficiency of the laser ion source.

Stopping efficiency	85
Survival against diffusion	44
Laser ionization efficiency	90
Metastable-state production	< 80
Spatial overlap with the laser beam	< 10
Time overlap (duty cycle)[a]	31
Transmission through the separator[b]	20
Molecular sidebands	< 100
Total estimated efficiency	< 0.18
Experimental efficiency[c]	0.13

[a]Only ^{55}Ni atoms produced in the last 15 ms of the cyclotron beam on period are still in the ion source at the moment of the laser pulse.

[b]The low transmission through the separator is not related to the laser ion source but is a purely technical problem.

[c]Since the nuclear reaction cross section enters directly in the experimental efficiency, the quoted value for this efficiency has a large uncertainty.

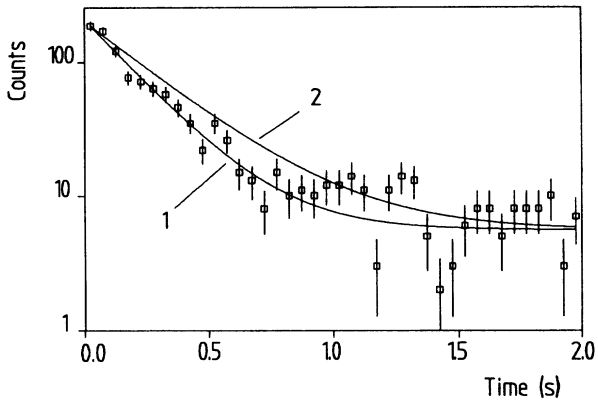

FIG. 4. Number of beta counts versus time. The curve labeled "1" shows the fitted lifetime (155 ms); curve "2" is drawn using the lifetime value of [7] (208 ms).

is deduced from the high-energy positrons and Hornshøj *et al.* claim that by properly choosing the beam energy (15.3 MeV) channels leading to high-energy positron emitters other than ^{55}Ni are closed. However, they do not take into account that ^{54}Fe(^{3}He, t)^{54}Co channel (threshold energy 8.7 MeV) which leads to the two β^+ emitting states of ^{54}Co (with $T_{1/2}$ = 193 ms and 1.46 min). The beam energy in the Äystö *et al.* experiment is higher (27 MeV) and more reaction channels are open. Although there is mass separation, contamination of longer-living Co nuclei cannot be excluded. Only our experiment has full element and mass selectivity.

The prototype laser ion source presented here has already the same production rate for ^{55}Ni as the conventional ion guide system, but with element selectivity. As can be seen in Table I, further optimization is possible. Losses due to the presence of metastable-state atoms and molecules can be reduced by introducing quenching gas mixtures. The weakest point in the present setup is the spatial and time overlap (cumulative efficiency < 3%). These losses can completely be removed by using a high pulse-repetition-rate laser system, two-color-two-step ionization schemes and an optical multipass system. The transmission problem through the separator has meanwhile been solved. With all these improvements an ion source efficiency of the order of 30% can be obtained. Another advantage of the laser ion source is the separation of the thermalization and the ionization processes. It allows the use of other gases at different pressures to increase the stopping efficiency and the survival against diffusion. It also allows multitarget stacks to increase the effective target thickness. This makes the laser ion source also very efficient for heavy ion induced fusion, for spallation, and for fission. Furthermore, the separation of the thermalization and ionization region makes it also possible to screen the ionization region from the beam-induced plasma. This makes the pulsing of the primary beam unnecessary.

In conclusion, we have shown the feasibility of a laser ion source based on resonance photoionization in a gas cell. The use of a higher repetition rate laser system (repetition rate > 100 Hz) will allow the production of radioactive beams of short-lived exotic nuclei independent of the chemical properties of the isotope provided that their ionization potential < 10 eV (80% of the elements). As the major part of the elements are covered by the existing selective target-ion-source systems, unstable nuclei of virtually all elements can be produced in a fast (< 100 ms), efficient ($> 10\%$), and element selective (suppression factor > 100) way. The discrepancy between the published ^{55}Ni half-life and our value indicates that element purity is often a key issue for spectroscopic studies near the proton or neutron drip line. Also in other regions of the nuclear chart the element selectivity combined with a mass separator is of prime importance for the study of exotic nuclei. For example, studying the nuclei around the double magic ^{78}Ni nucleus produced in the very unselective fission reaction needs a clear identification of the mass and the charge of the nucleus. With this laser ion source these requirements can be fulfilled and new studies of short-lived nuclei far from stability will become possible offering us new vistas in the understanding of the nuclear properties of highly unstable nuclei. Moreover, by using the laser ion source in conjunction with a multitarget system, intense and pure radioactive beams can be produced for injection in postaccelerators. This would relax three severe constraints on the presently proposed radioactive ion beam facilities [4]. The multitarget system, effectively cooled by the helium flow, can withstand intense beams. Second, the ionization by laser light brings all critical parts of the ion source far from the irradiation zone, enhancing the reliability of the whole system. Third, the element selectivity rules out the need for isobaric mass separation prior to acceleration.

We would like to thank J. Gentens and P. Van Den Bergh for running and maintaining the LISOL separator. We also like to thank Professor Ir. J. Berghmans and Ir. J. Hennissen (Mechanical Engineering Department, Leuven) for performing calculations on the flow properties of helium in the ion source. This work is supported by the Belgian Interuniversity Institute for Nuclear Sciences (I.I.K.W.) and by the Concerted Action (G.O.A.) and Inter-University Attraction Poles (I.U.A.P.) Research Programs. One of us (L.V.) is Postdoctoraal Onderzoeker of the Belgian National Fund for Scientific Research (N.F.W.O.).

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