LASER ION SOURCES

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ABSTRACT

Stepwise excitation by resonant laser radiation and photo ionization in the last transition can be used to ionize atoms selectively and efficiently in order to obtain a high performance laser ion source. For on-line isotope separators like ISOLDE at CERN, the main advantages are the pulsed structure of the extracted radioactive beam and the reduction of isobaric impurities. Preliminary experiments which make use of the resonant ionization process and time-of-flight spectroscopy are described and the possible layout of an on-line version of a laser source is discussed. Ionization efficiencies of the order of 20 to 40% are expected for elements with one atomic ground state populated thermally.

INTRODUCTION

The main task and art in constructing ion sources for on-line isotope separation (ISOL) of radioactive isotopes is to achieve an isotopically pure beam with high efficiency. The extraordinary progress obtained in the last 10 years is documented, e.g., for the ISOLDE on-line mass separator in /RA84a, RA84b, IS85/. As shown in Fig.1, over 60 elements are obtainable as radioactive beams, some of them with more than 30 isotopes and production rates of up to $10^{11}$ atoms per sec and mass number. About 20 elements are produced

<table>
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<tr>
<th>PERIODIC TABLE OF THE ELEMENTS</th>
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Fig.1. Elements available at ISOLDE as radioactive beams.
with high yields and without major isobaric impurities. In these latter cases, the existing technique is most appropriate and straightforward and no major improvement can be expected. For the on-line use of the other elements, however, continuous effort has to be put into the further development of high-efficient ion sources, the investigation of suitable target-projectile combinations or the application of alternative ionizing schemes.

A new and promising scheme is resonant ionization spectroscopy (RIS), where the atom under investigation is stepwise excited by absorption of resonant laser light and finally ionized. The high selectivity of this process enables an efficient discrimination against isobars and, if desired, also against isotopes. In order to demonstrate the advantages of a RIS ion source, the production rates of some rare earth elements are shown in Fig. 2 as obtained at ISOLDE by bombarding a Ta-foil target with 600 MeV protons. Due to the chemical similarities of the rare earth elements, there is no physico-chemical way to obtain an ion source-target combination which delivers radioactive beams of only one element. In fact the yields of Fig. 2 were measured by laser spectroscopy /NE82/. A target such as the Ta-foil one is only of limited value for most application (with the exception of laser spectroscopy which is selective itself) because of the strong contamination by isobars. Clearly, the RIS ion source added to the Ta-foil target would give

![Graph showing production rates of rare earth elements](image-url)
10 beams which are isobarically pure and which could be added to the list of 20 favourable cases mentioned above.

There is another advantage of RIS-based ion sources: Since pulsed lasers have to be used for RIS, the radioactive beam will also be pulsed. A bunched ion beam with a pulse length of about 10 ns is useful for many applications, like injection into accelerators, storage rings or ion traps. It can also lead to an increase of the signal-to-background ratio as in collinear spectroscopy (for references see /KL84/) by many orders of magnitude. With present technology a gain of $10^4$ is expected which would bring down the detection limit of collinear spectroscopy to 100 atom/sec in favourable cases.

Finally, it should be mentioned that the demand for isotopically pure beams results in a large variety of different target-ion source combinations e.g., cold or hot transfer lines from the target to the ion source, cold (Ta) or hot (W) surface ionizer or a plasma ion source. For example, the La target of ISOLDE is combined with 3 different ion sources at ISOLDE to optimize the production of Xe, Cs and Ba. The need of several target ion source combinations (each at a cost of 4000 $) and the loss of beam time due to the need of changing and outgassing the target (which takes about 24h) would be reduced by use of the laser ionization technique. Of course, there would be the added cost of a highly sophisticated and expensive laser system.

THE RESONANT IONIZATION SCHEME

Resonance ionization spectroscopy (RIS) /AM71, HU79, LE83/ has been developed in parallel with the improved performance of powerful tunable dye lasers. The main areas of application of that technique are today trace analysis and isotope separation. By means of several laser beams the atoms are stepwise excited to (i) a Rydberg level where they are field-ionized, or (ii) photo-ionized by an excitation into the continuum, or (iii) photo-ionized by a transition to an auto-ionizing state.

At Mainz, we have tested the technique for different elements. The main goals are the determination of hyperfine structure (HFS) and isotope shift (IS) of neutron-deficient Au isotopes in the D, optical transition at ISOLDE /ST85, BO84/ and the trace analysis of Pu atoms in the environment /KR85, PE85/. Fig.3 shows the optical transitions used for a number of elements. The first two resonant transitions have a high effective cross section, of the order of $10^{-10}$ to $10^{-12}$ cm$^2$. High laser power is needed in the case of a two-photon transition, a transition to a high Rydberg level, or one into the continuum ($< 10^{-17}$ cm$^2$).
Fig. 3. Resonance ionization schemes used for the investigation of In, Xe, Hg, Au, U, Pu and Gd. The broken line indicates a virtual intermediate state used in the two-photon experiment, solid lines represent real states. The wavelengths for the different experiments are indicated.

Fig. 4. Experimental setup for resonance ionization spectroscopy in combination with a measurement of the mass of the ion by time of flight.

The experimental setup is shown in Fig. 4. An atomic beam is formed by heating an oven (in which the ISOLDE beam will be sent in case of Au) or a filament (on which the sample has been electrolytically deposited as in the case of Pu). This beam is intersected by the pulsed laser beams used for stepwise photo ionization. The ions created in resonance are accelerated in an electric field and allowed to drift to a channel plate where they are detected. Since the short laser pulse (pulse length = 10^-9 s) represents an ideal start signal, and a time focus at the detector can be obtained with appropriate voltages in the region of acceleration, the mass resolution obtainable is about 2000.
A time-of-flight spectrum obtained for Pu is shown in Fig. 5. No background event is observed in this mass region. Gating on mass A=239 with a time window and scanning the frequency of the two tunable dye lasers, a two-dimensional plot of the optical resonance is obtained (Fig. 6). Again, no background is observed off resonance, indicating the inherent high selectivity of the RIS technique.

LASER ION SOURCE

The setup shown in Fig. 4 represents in principle a laser ion source. Since a typical optical transition has a transition frequency of about $5 \cdot 10^{14}$ Hz and a line width $< 10^9$ Hz, and the atomic level density is extremely low, the chance to hit accidentally a resonance of another element is less than $10^{-5}$ per excitation step. Hence, isobaric selectivity is assured for essentially all ISOL applications. Isotope selectivity might be achieved by taking advantage of the HFS and IS in
the first excitation step differing from one isotope to another. In most cases, however, a mass separator will already do this job and therefore it is advantageous to use a laser for the first step with a band width which is large enough ($\Delta v_{\text{laser}} < 30 \text{ GHz}$) in order to cover the HFS and IS of all isotopes produced.

**Sensitivity**

The ionization efficiency is the most important figure of merit of an ion source. The overall efficiency of the set-up shown in Fig.4 is given in Table I and is defined as the ratio of the registered electronic pulses of the channel plate detector in the appropriate time window to the number of atoms put into the oven or on the filament. In case of the Au and Gd experiments the saturation of all optical transitions could be experimentally proved. This is shown in Fig.7 where the Gd ion yield is plotted as a function of the wavelength of the third laser and the dependance of this step on the laser power is given in the insert. The resonances in Fig.7 are transitions to autoionizing states. The pedestal of the resonances indicates the normal cross section of the third step into the continuum. A gain of 3 orders of magnitude is obtained by tuning the third laser to an autoionizing transition. The main factors reducing the efficiencies as given in Table I are (i) the long distance between the oven/filament to the zone of

<table>
<thead>
<tr>
<th>INVESTIGATED ELEMENT</th>
<th>SOURCE</th>
<th>LASER/REPETITION RATE</th>
<th>DIAMETER OF LASER BEAM</th>
<th>EFFICIENCY</th>
</tr>
</thead>
<tbody>
<tr>
<td>Au</td>
<td>oven</td>
<td>Nd:Yag laser pumped Dye lasers/ 10Hz</td>
<td>0.3cm</td>
<td>$10^{-8}$</td>
</tr>
<tr>
<td>Pu</td>
<td>filament</td>
<td>Cu-vapour laser pumped Dye lasers/ 6KHz</td>
<td>0.1cm</td>
<td>$10^{-7}$</td>
</tr>
<tr>
<td>Gd</td>
<td>oven</td>
<td>Cu-vapour laser pumped Dye lasers/ 6KHz</td>
<td>1cm</td>
<td>$10^{-5}$</td>
</tr>
</tbody>
</table>
Fig. 7. Yield of Gd ions as a function of the wavelength of the third laser. The dependence of the yield on the laser power is given in the inset at $\lambda_3=613.3\text{nm}$. The laser beams have a diameter of 1cm.

interaction with the laser (all cases), (ii) the low repetition rate of the laser used (Au) and (iii) the short traversing time of the atom through the laser beam in comparison to the laser repetition rate (Pu). Hence, an efficient ISOL ion source should have an almost vanishing distance between oven and interaction zone and only those atoms should leave the interaction zone which are ionized.

Layout of an ISOL Ion Source

Laser ion sources for ISOL applications have been proposed by Letokhov /LE84/ and Lee /LEE84/. The geometry proposed here is different and based on the experience gained at Mainz. It will be tested soon in the existing setup with Gd. A scheme of the source is shown in Fig. 8.

Fig. 8. Possible layout of laser ion sources.

The laser beams enter the ionization chamber (which is connected by a transfer tube with the target) through the extraction hole in opposite direction to the extracted ion.
The rate of ionization $R_I$ by RIS is given by

$$R_I = \frac{V_{\text{INTER}} \times v_{\text{LASER}} \times \epsilon_{\text{ION}}}{V_{\text{TOTAL}}}$$

where $V_{\text{INTER}}$ is the volume where the atoms in the ionization chamber of total volume $V_{\text{TOTAL}}$ can interact with the laser beams, $v_{\text{LASER}}$ is the repetition rate of the laser pulses and $\epsilon_{\text{ION}}$ is the total ionization probability.

There are two possible modes of escape out of the ionization chamber: Diffusion through the extraction hole as neutral atom and escape after surface ionization.

The rate of escape by diffusion $R_E$ is given by the total surface area of the ionization chamber $A_{\text{TOTAL}}$ divided by the surface area of the extraction hole $A_{\text{HOLE}}$ times the collision rate of the atom on the surface, which is given by the mean velocity $\bar{u}$ and the mean free path $\lambda$ in the chamber:

$$R_E = \left(\frac{A_{\text{HOLE}}}{A_{\text{TOTAL}}}\right) \times (\bar{u}/\lambda)$$

The loss by surface ionization $R_S$ is determined by the probability of surface ionization $\epsilon_{\text{SUR}}$ times the collision rate times the escape probability. If a magnetic field of sufficient strength is applied, only those ions can escape which are surface ionized on the area $A_{\text{HOLE}}$ just opposite to the extraction hole. Hence, this escape rate is given by

$$R_S = \left(\frac{A_{\text{HOLE}}}{A_{\text{TOTAL}}}\right) \times \epsilon_{\text{SUR}} \times (\bar{u}/\lambda)$$

The efficiency of the laser ion source is given by the ratio

$$\epsilon = \frac{R_I}{R_{\text{TOTAL}}} = \frac{R_I}{(R_I + R_E + R_S)}$$

which simplifies to

$$\epsilon = \frac{v_{\text{LASER}} \times \epsilon_{\text{ION}}}{\bar{u}/6\lambda \times (1 + \epsilon_{\text{SUR}}) + v_{\text{LASER}} \times \epsilon_{\text{ION}}}$$

if a cubic ionization chamber is taken for ease of calculation. Hence, the efficiency of the laser ion source does not depend on the size of the extraction hole (which of course determines the delay time in the ionization chamber). For
an extraction hole of 1 to 3 mm$^2$ one can therefore put $\varepsilon_{\text{ION}} = 1$ (see the example of Gd discussed above) if there is only one fine structure level thermally populated. The essential factor of (5) is then the ratio between laser repetition and collision rate. This dictates the use of pulsed lasers with high repetition rate which are today only realized by Cu vapour lasers. Taking for example $\nu_{\text{LASER}} = 10^4$ Hz, $\nu = 10^5$ cm/sec, $\lambda = 1$ cm one obtains

$$
\varepsilon = \begin{cases} 
0.37 & \text{for } \varepsilon_{\text{surf}} = 0 \\
0.22 & \text{for } \varepsilon_{\text{surf}} = 1
\end{cases} 
$$

(6)

which are quite good figures even for conventional ion sources without isobaric selectivity.

CONCLUSION

Ion sources for ISOL applications can be constructed with today's technology. The problem to be solved is the construction of the ionization chamber where an insulator has to be incorporated which works at high temperature and is chemically inert in respect to the radioactive atoms (short sticking time). The advantages of a laser ion source are

- high efficiency
- bunched ion beams
- small phase space
- rejection of isocbars.

There are many applications of such sources. Examples at ISOL facilities are

- isotopically pure beams
- improvement of signal-to-background
- ease of injection into storage rings, ion traps or accelerators

In addition, of course, there is the general interest in isotope separation, the use of the source as analytical instrument in trace analysis, the determination of isotope ratios for cosmological investigations and all those many other fields where mass spectrometers are essential tools.
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