A CHEMICALLY SELECTIVE LASER ION SOURCE FOR ON-LINE MASS SEPARATION

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ABSTRACT

Laser resonance ionization has been applied to ionize radioactive atoms released from thick ISOLDE/CERN targets. Laser light from pulsed dye lasers with a high repetition rate (10 kHz) was used for stepwise excitation and ionization. By this method the efficiency as well as the selectivity of the target/ion source system could be improved. In a series of off-line and on-line studies the resonance ionization of elements with different ionization potentials such as Yb ($E_i = 6.2$ eV), Tm ($E_i = 5.8$ eV) and Sn ($E_i = 7.3$ eV) was investigated. For Yb, photoionization efficiencies of up to 15\% have been measured. An efficiency of 0.2\% was obtained for Sn at the on-line-separator ISOLDE-3. However, this value can be improved by about two orders of magnitude using a transition to an autoionizing state at 59375.9 cm\textsuperscript{-1}.

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1. INTRODUCTION

A general problem of conventional ion sources at on-line isotope separators is their limited chemical selectivity. Very often the isotopes of several elements are produced simultaneously in nuclear reactions [1]. As a result the mass separated ion beams are contaminated by isobars. For detailed investigation of single isotopes a chemically selective ion source is desirable. The selectivity but also the efficiency of conventional ion sources as applied at on-line mass separators can be improved by using resonance ionization spectroscopy (RIS). In a laser ion source (LIS) a pulsed ion beam is produced via stepwise resonant excitation and finally ionization by light of a pulsed laser system. The pulse structure of the photo ions can be utilized to discriminate against the continuous background of surface-ionized atoms.

Such a laser ion source has been installed at the on-line isotope separator ISOLDE–3 (Fig.1) at CERN/Geneva [2]. The laser beams are incidented into the hot tube (insert of Fig.1) through the hole of the extraction electrode. The photo ions are extracted and mass analysed by the ISOLDE mass separator.

At GSI/Darmstadt we plan to set up a similar LIS for $\gamma$-spectroscopy of $^{102}$Sn. This experiment requires a high efficiency to reach a resonable yield of $^{102}$Sn as well as a high selectivity. Until now such an experiment could not be performed due to the contamination by $^{102}$In as obtained with a FEBIAD ion source [3].
2. PRINCIPLE OF THE LASER ION SOURCE

The atoms of the element under investigation are excited by two resonant excitation steps. The ionization can be achieved in a third step non-resonantly into the continuum with cross-sections of typically $\sigma = 10^{-17}$ cm$^2$ - $10^{-18}$ cm$^2$ or resonantly via an autoionizing state with cross-sections of the order of $\sigma = 10^{-14}$ cm$^2$ - $10^{-16}$ cm$^2$. The RIS schemes for Yb, Tm and Sn used for the measurements at CERN are shown in Fig.2. Pulse energies of a few mJ/cm$^2$ are required for saturation of resonance transitions. Such energies are available with dye lasers pumped by copper vapour lasers and a laser beam focus of 1 mm diameter. The laser pulse energy, however, is insufficient for saturation of the non-resonant ionization step. Therefore it is essential to use transitions to autoionizing states in the last step. In the case of tin, no suitable path for resonance ionization leading to an autoionizing state was known [4].

Generally three different wavelengths and hence three dye lasers are required as shown in Fig.2b for Tm. In the case of Yb an excitation scheme can be used, where the wavelengths for the second and third step are identical within the laser bandwidth ($\Delta \nu_L = 24$ GHz), so that only two dye lasers are necessary (Fig.2a). For the ionization of Sn laser light for the second transition is also used for excitation into the continuum (Fig.2c).

The photoionization efficiency $\epsilon$ of the ion source can be estimated by comparing the rate of photoionization with the rate of diffusion as neutral atom out of the orifice of the hot tube. Assuming complete extraction of the produced photo ions one obtains for a tube of length $L$

$$\epsilon = \frac{\nu_{rep} \epsilon_{ion}}{\nu_{rep} \epsilon_{ion} + \frac{v}{4L}},$$

(1)
where $\nu_{rep}$ is the pulse repetition rate of the lasers, $\epsilon_{ion}$ is the probability for resonant ionization of the atoms interacting with the laser beams and $v$ the thermal velocity of the atoms. As can be seen from (1) the efficiency of the LIS is independent of the diameter of the tube. High efficiency requires a high pulse repetition rate of the lasers and a long tube length. Furthermore saturation in all transitions is prerequisite to get an $\epsilon_{ion}$ value close to one. Today, only dye lasers pumped by copper vapour lasers provide a very high repetition rate ($\nu_{rep} \approx 10$ kHz) at sufficient output power. One calculates an efficiency of $\epsilon = 62\%$ for $\nu_{rep} = 10$ kHz, $\epsilon_{ion} = 1.0$, $L = 4$ cm and $v = 10^5$ cm/s (for atoms with a mass number $A = 60$ and a temperature of the tube of $T = 2000^\circ$ C).

3. EXPERIMENTAL SET-UP AND MEASUREMENTS

The laser system consists of three dye lasers pumped by two copper vapour lasers ($\nu_{rep} = 10$ kHz, pulse length 20 ns). One copper vapour laser is used as an oscillator, the other one as an amplifier. The LIS has first been tested at the off-line mass separator at CERN with Yb and Tm. In these studies efficiencies of $\epsilon = 15\%$ have been achieved for Yb. The lower efficiency than expected from eq. 1 can be explained by incomplete saturation in the last step, transmission losses of the mass separator, the tube length of only 3 cm and diffusion into the bulk material of the ionizer tube. The selectivity (defined as the ratio of the number of photo ions relative to the number of surface ions) can be increased at lower temperatures and reaches a value of up to $10^4$ at a temperature of $T = 1600$ K with TaC as construction material for the tube. Since the release
time is quite long at low temperatures, such temperatures are not applicable to on-line experiments on Yb isotopes.

At the on-line separator ISOLDE–3/CERN the LIS has also been applied to the radioactive nuclei $^{157-167}$Yb released from a thick ISOLDE target (122 g/cm$^2$ Ta-foil). In this experiment the laser beams were focused over a distance of about 20 m through a hole in the first magnet of the on-line mass separator (Fig.1) into the tube where the atoms under investigation have been photo-ionized. The diameter of the laser beams was 1 mm at the entrance of the tube. The laser spot as well as its alignment in respect to the tube were controlled via a theodolite. Different materials for the tube (W or Ta) were tested with inner diameters ranging from 1 mm to 3 mm and lengths between 30 mm and 40 mm. The photo ions were detected at the focal point of the second magnet of the ISOLDE–3 mass separator (Fig.1).

The tube was heated by a DC current of about 100 A to a temperature of $T \approx 2000^\circ$C. This current causes a voltage drop of a few volts along the tube which is used to extract the photo ions. Although the photo ions are produced within the laser pulse length of only 20 ns, they arrive at the ion detector placed at the focal point F2 of the mass separator with a time spread of 10–50 $\mu$s. This is caused by the slow extraction of the ions out of the ionizer tube. By a gated detection of the photo ions the continuous background stemming from surface ionization is partially suppressed and the selectivity is improved from 10 without gated detection to about 90 with gated detection.

The shape of the photo ion pulse was determined by measuring the current of the mass separated ions in a Faraday cup at F2 for different delays between the laser pulse and a short time
window ($\Delta T \approx 5 \mu s$) for detection (Fig.3). The pulse shape was shown to depend on the voltage drop along the tube, the laser beam alignment and the element under investigation. The pulse width is reduced if only a fraction of the ions is extracted. With a higher voltage of about 100 V along an insulator tube it should be possible to reduce the pulse width, so that the selectivity can be further improved.

For Sn an efficiency of only 0.2% has been obtained at the on-line separator with the excitation scheme shown in Fig.2c. This is due to the low efficiency of a transition into the continuum for the ionization step being only $\epsilon_{\text{ion}} \approx 0.01$. In a recent experiment with a time-of-flight spectrometer [4] we have found a new excitation scheme for Sn leading to an autoionizing state (Fig.4) at 59375.9 cm$^{-1}$ [5]. With this scheme it should be possible to improve the efficiency by about two orders of magnitude and to perform $\gamma$-spectroscopy on $^{102}$Sn. This scheme enables the saturation of the third transition with a pulse energy of 1.5 mJ/cm$^2$ as delivered by a dye laser system pumped by copper vapour lasers.

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Figure captions:

Figure 1:  Laser ion source (LIS) at the on-line separator ISOLDE-3/CERN. The ion optics (lenses and magnets) are shown for the horizontal plane. M: mirror, LB: laser beams, M1: first magnet, F1: focal point of the first magnet, M2: second magnet, PD: pulsed deflector, F2: focal point of the second magnet, ID: ion detector. Insert: Schematics of the target container, transfer tube, ionizer tube and extraction electrode of the laser ion source.

Figure 2:  Atomic level schemes for resonance ionization of (a): Yb \((E_i = 6.2 \text{ eV})\), (b): Tm \((E_i = 5.8 \text{ eV})\) and (c): Sn \((E_i = 7.3 \text{ eV})\) as used in the experiments at ISOLDE/CERN.

Figure 3:  Pulse shapes as obtained at the focal point of the second magnet of the on-line mass separator ISOLDE-3 for stable \(^{174}\text{Yb}\). The Ta–tube (length \(L = 4 \text{ cm}\), inner diameter \(d = 1 \text{ mm}\) was heated by a DC current of \(I = 95 \text{ A (open square)}\) and of \(I = 137 \text{ A (asterix)}\). The lines are fits to the experimental points by Gaussian line profiles.

Figure 4:  Atomic level scheme for resonance ionization of Sn leading to an autoionizing state.
(a) Yb

52240 \text{ cm}^{-1}

581.0 \text{ nm}

35197 \text{ cm}^{-1}

581.0 \text{ nm}

17992 \text{ cm}^{-1}

555.6 \text{ nm}

0

(b) Tm

51829 \text{ cm}^{-1}

575.5 \text{ nm}

34457.9 \text{ cm}^{-1}

571.2 \text{ nm}

16967 \text{ cm}^{-1}

589.6 \text{ nm}

0

(c) Sn

606.9 \text{ nm}

51171 \text{ cm}^{-1}

606.9 \text{ nm}

34641 \text{ cm}^{-1}

303.4 \text{ nm}

1692 \text{ cm}^{-1}

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Fig. 2