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# Search for efficient laser resonance ionization schemes of tantalum using a newly developed time-of-flight mass-spectrometer in KISS



BEAM INTERACTIONS WITH MATERIALS AND ATOMS

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#### ABSTRACT

The technique of laser resonance ionization is employed for an element-selective ionization of multinucleon transfer reaction products which are stopped and neutralized in a gas cell filled with argon gas at 50 kPa. We have been searching for efficient laser ionization schemes for refractory elements of Z = 73–78 using a time-of-flight mass-spectrometer (TOF-MS) chamber. To evaluate the isotope shift and ionization efficiency for each candidate of the ionization scheme, isotope separation using the TOF-MS was devised. The TOF-MS was designed to separate the isotopes using two-stage linear acceleration with a mass resolving power  $M/\Delta M$  of >350. A mass resolving power of 250 was experimentally confirmed by measuring the TOF of laser-ionized tantalum (Z = 73) ions with mass number 181. We searched for a laser resonance ionization scheme of tantalum using the TOF-MS.

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## 1. Introduction

An element-selective isotope separator KISS (KEK Isotope Separation System) [1] has been developed at the RIKEN Nishina Center (RNC) to investigate beta-decay properties of r-process waiting point nuclei with  $A \sim 200$  and N = 126 [2]. Initially, we will investigate the nuclei with N = 126 such as <sup>203</sup>Ir, <sup>202</sup>Os, <sup>201</sup>Re and <sup>200</sup>W, which are located in the vicinity of the waiting point region. To access these unexplored neutron-rich nuclei, multi-nucleon transfer (MNT) reactions of <sup>136</sup>Xe (projectile) + <sup>198</sup>Pt (target) will be used. At KISS, an argon gas cell system and laser resonance ionization technique have been employed for collecting all the reaction products and for the element-selective separation, respectively.

Reaction products from MNT reactions are stopped and neutralized in the gas cell which is filled with argon gas at a pressure of 50 kPa. Neutral atoms are transported towards the exit of the gas cell by laminar gas flow. Before the exit of the gas cell, atoms are ionized element-selectively by irradiation with two-color lasers. The two-color two-step laser ionization allows element-selective

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excitation of atoms by laser irradiation of wavelength  $\lambda_1$  and efficient ionization via an excited state to an auto-ionizing state (AIS) by laser irradiation of wavelength  $\lambda_2$  [3]. AIS is a resonance state located above the ionization potential. The selectivity of laser ionization (the ratio of the ion count rate with lasers on vs lasers off) is typically 1000. The ions are transported to a low pressure region of  $10^{-4}$  Pa by using a sextupole ion guide (SPIG) [4] and accelerated by an electric potential of 20 kV. The mass number of the ions is selected by using a dipole magnet. The ions are transported to a detector station to study the beta-decay properties.

The extraction efficiency of KISS consists of a stopping efficiency of reaction products (0.87), transport efficiencies in the gas cell (0.6) and the SPIG (1), an acceptance of the mass separator (0.9), decay losses against beta-decays of unstable nuclei (during a transport time ~200 ms from the stopping position to the detector position), and a laser ionization efficiency in the gas cell. The required extraction efficiency is 5% for the beta-decay half-life measurement of <sup>200</sup>W ( $T_{1/2} = 801 \text{ ms}$  [5]) within a few days. In this estimation, we assumed the <sup>198</sup>Pt beam intensity is 40 pnA and 1000 events are detected as beta-rays decaying from <sup>200</sup>W. A laser ionization efficiency of more than 15% is therefore required. Therefore, it is necessary to search for laser ionization schemes ( $\lambda_1$ ,  $\lambda_2$ ) by which such ionization efficiency in the gas cell for the refractory elements of *Z* = 73–78 can be achieved.

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The laser ionization efficiency is governed by the hyperfine structure, power broadening and pressure broadening (the details were reported in Ref. [6]). To estimate the laser ionization efficiency for the established ionization scheme, we need to study the elements off-line. Meanwhile, in order to apply the efficient ionization scheme to unknown neutron-rich isotopes far away from the stability line, it is necessary to take into account isotope shifts of  $\lambda_1$  and  $\lambda_2$ . For this purpose, it is essential to study the size of the isotope shift of the established ionization scheme by using stable isotopes, and extrapolate the amount of isotope shift for unstable nuclei. For the estimation and extrapolation, it is important to select one isotope with high mass resolving power. Therefore, we developed TOF-MS for isotope separation, and studied the ionization scheme and efficiency. In this report, we explain the newly developed TOF-MS and the result for the ionization scheme of tantalum.

### 2. TOF-MS

The TOF-MS consists of two-stage linear acceleration electrodes, a drift tube, a channel electron multiplier (CEM) and a filament as shown in Fig. 1. The lasers were focused to a spot of a few mm<sup>2</sup> at the center between the electrodes of the first acceleration stage, and were overlapped temporally and spatially. Neutral atoms were evaporated by heating the filament and ionized by laser irradiation. The ions were accelerated by the two-stage constant electric fields and detected by the CEM. The mass resolving power  $M/\Delta M$  of the TOF-MS is defined as  $t/2\Delta t_{FWHM}$ . The TOF-MS was designed to separate isotopes of A = 180-200 with a mass resolving power of ~400.

It is considered that three parameters affect the mass resolving power of the TOF-MS, an initial position distribution of atoms in the ionization region (or size of the laser spot), an initial velocity distribution of atoms along the acceleration direction in the ionization region, the time duration of the laser pulse, and the resolution of detector and electronic modules. The present design of the acceleration electrodes enables a reduction of the time-deviation, which was caused by the initial position distribution of atoms, as much as possible. The distance between electrodes, length of the drift region and electric potentials were optimized under the conditions that the first and second degree of differential equations in relation to the initial position of the TOF equal zero [7].

Fig. 1 shows the optimized setup of the two-stage linear acceleration electrodes. Distances between the electrodes where voltage are applied are 52.5 mm and 28 mm, and the length of the drift region is 560.5 mm. Optimized acceleration potentials are  $U_{a1} = 850$  V and  $U_{a2} = 1550$  V, respectively. Correction electrodes are installed to suppress distortion of the electric field by 0 V (GND) potential generated by the TOF-MS vacuum chamber.

Fig. 2 shows the calculated ion trajectory by SIMION. In this simulation, an initial position distribution of atoms with a 3dimensional Gaussian distribution of  $\sigma = 1$  mm, an initial velocity distribution taking into account the geometry and the Maxwell distribution at the filament temperature, a laser pulse width with a Gaussian distribution of  $\sigma$  = 5.1 ns and an additional time resolution with a Gaussian distribution of  $\sigma = 2.3$  ns were considered. The additional time resolution consists of the time fluctuation of an overlap time profile between two excimer lasers (0.85 ns) and time resolutions of detector and electronic circuits (assumed to be 2.1 ns in total). The ion trajectory is biased towards the upper direction in Fig. 2. This bias originates from the vertical component of initial energy of the ions along the acceleration direction. 70-80% of ions are lost by the ion trajectory broadening. A mass resolution of about 380-450 was obtained in the simulation for an ion of A = 180-200 with acceleration potentials of  $U_{a1} = 850$  V and  $U_{a2} = 1550$  V.

The measured mass resolving power using a tantalum filament (natural abundance of <sup>181</sup>Ta is 99.988%.) was 252 as shown in Fig. 3. It is therefore possible to separate the mass of nuclei with A = 180-200. This measured resolving power can be almost reproduced by simulation considered the small variations of the TOF-MS geometry and the fluctuation of laser irradiation position etc. to the ideal



**Fig. 1.** Arrangement of the acceleration region in the present TOF-MS. Red lines represent equipotential lines calculated by SIMION [8] with acceleration potentials of  $U_{a1} = 850$  V and  $U_{a2} = 1550$  V. Voltages are applied to three electrodes from external power supplies. Voltage for the left-side electrode of the ionization region is  $\frac{475}{52}U_{a1} + U_{a2}$  considering the position of the electrode. Other linear acceleration fields are made by voltage division. The lower plot shows the axial potential as a function of position on the center axis. (For interpretation of the references to colour in this figure caption, the reader is referred to the web version of this article.)

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