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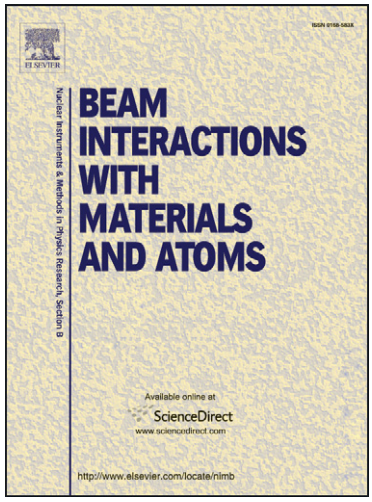

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Isobar suppression in AMS using Laser Photodetachment

O. Forstner [a,*] P. Andersson [b] C. Diehl [c] R. Golser [a] D. Hanstorp [b] W. Kutschera [a] A. Lindahl [b]
A. Priller [a] P. Steier [a] A. Wallner [a]

[a] *Faculty of Physics, University of Vienna, A-1090 Wien, Austria*
[b] *Department of Physics, Göteborg University, SE-412 96 Göteborg, Sweden*
[c] *Institut für Physik, Johannes Gutenberg Universität Mainz, D-55099 Mainz, Germany*

Abstract

We are investigating the possibility of using laser photodetachment of negative atomic and molecular ions as an additional isobaric selection filter in accelerator mass spectrometry. The aim of this study is to find a possibility to further improve the detection limit for long-lived heavy radionuclides at AMS facilities. We will focus on the astrophysical relevant radionuclide ^{182}Hf, which is one of the isotopes measured with the 3 MV tandem AMS facility VERA (Vienna Environmental Research Accelerator) at the University of Vienna. Laser–induced isobar suppression is also of importance for radioactive–beam facilities.

The present detection limit for measuring the isotope ratio ^{182}Hf/Hf at VERA is 1×10^{-11}. The limiting factor is the strong background of the stable isobar ^{182}W. Currently this background is suppressed using suitable molecular ions in the injection stage. Selective laser photodetachment of the negative ions at the injector can lead to an additional suppression of the interfering isobar. Test experiments have been carried out at the negative ion laser spectroscopy setup at Göteborg University. In a small ion beam apparatus pulsed tunable laser radiation is used to measure the photodetachment cross–section of different atomic and molecular negative ions. We will present studies of the photodetachment process for various Tungsten and Hafnium molecules with the aim to find a selective isobaric suppression scheme using laser photodetachment spectroscopy in combination with AMS.

Key words: Photodetachment, Negative ion, Mass spectrometry
PACS: 32.10.Hq, 32.80.Gc, 33.15.Ry, 07.75.+h

1. Introduction

Accelerator Mass Spectrometry (AMS) has proven to be a powerful tool not only for radiocarbon dating but also for research in many other fields. Detection of long–lived radioisotopes at typical abundances in the range of $10^{-12} - 10^{-15}$ has various applications as tracers or chronometers in geology, archaeology and biomedicine. Notably in the field of nuclear astrophysics AMS has proven to be a valuable tool in the determination of reaction cross–sections of long–lived isotopes and the search for live supernova remnants on earth. Tuniz et al. give a detailed overview of the technique as well as a discussion of the numerous applications [1].

One of the main tasks in AMS measurements is the separation of the rare radioisotope from stable isobaric background. There are various ways to achieve this separation after acceleration using different methods like energy loss in matter, measuring time of flights, using the mean charge state in a gas–filled magnet or full stripping in a subsequent stripper stage. Alternatively, it could be advantageous to suppress the background isobars already before entering the accelerator. In some cases this task can be efficiently fulfilled using negative ions. Examples for this cases are ^{14}C, ^{26}Al and ^{129}I where the stable isobars ^{14}N, ^{26}Mg and ^{129}Xe do not form negative ions. In other cases negative molecular ions can be used. One example is the measurement of ^{41}Ca where the interference of ^{41}K is strongly suppressed by using $^{41}CaH_3^-$ ions through the instability of $^{41}KH_3^-$. In the measurement of ^{182}Hf the negative HfF_5^- molecule is used in the injection stage. This strongly suppresses the isobaric background of ^{182}W, since WF_5^- ions are rarely produced. In this way a detection limit of the isotope ratio ^{182}Hf/Hf of 1×10^{-11} at the 3 MV tandem AMS facility VERA (Vienna Environmental Research Accelerator) can be achieved [2].

A novel scheme to remove stable isobars in a low energy negative ion beam was proposed by Berkovits et al. [3,4]. The basic idea is to neutralize the isobars by selective pho-

* Corresponding author. Address: Währinger Str. 17, A-1090 Wien, Tel: +43-1-4277-51763, Fax: +43-1-4277-9517
Email address: Oliver.Forstner@univie.ac.at (O. Forstner).

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todetachment with laser light whereas the radioisotope ions remain unaffected. To adopt this method the radioisotope has to have a larger electron affinity (EA) than the stable isobar. Tuning the laser to a frequency lower then the EA of the radioisotope but higher than the one of the stable isobar, only the latter will be photodetached and hence neutralized.

A proof of principle of the selective photodetachement was shown in demonstration experiments for the cases ^{36}Cl–^{36}S and ^{59}Ni–^{59}Co [3,4]. However, the reported overall degree of isobar suppression was far from being practically useful for actual AMS measurements without further improvements. Liu et al. [5,6] incorporated a linear gas–filled rf quadrupole ion guide into the system to decelerate the ions during their interaction with the laser light. In this way the interaction time between the ions and the photons could be increased from a few microseconds to the millisecond range.

The aim of this work is to investigate the possibility to decrease the detection limit for ^{182}Hf by incorporating a laser photodetachment step in an AMS facility. As a first step, electron affinities of various tungsten and hafnium fluorides have to be determined, and such investigations are currently carried out using the Göteborg negative ion beam apparatus described in section 3.

2. Laser photodetachment principles

In laser photodetachment a negative ion interacts with a photon of an energy greater than the binding energy of the electron thus neutralizing the ion:

$$X^- + h\nu \rightarrow X + e^- \quad (1)$$

The electron affinity (EA) is defined as the energy difference between the ground states of the ion and parent atom or molecule. A laser photodetachment threshold (LPT) measurement is performed by tuning the energy of the photons around the EA and monitoring either the produced neutrals or photoelectrons. The opening of the detachment channel is seen as a threshold when the photon energy equals the EA. The cross section behavior just above threshold is described by the Wigner law [7]:

$$\sigma(E) \propto (E - E_{Th})^{l+1/2}, \quad (2)$$

with $\sigma(E) = 0$ for $(E \leqslant E_{Th})$. Here, E represents the photon energy, E_{Th} is the electron affinity and l the angular momentum of the outgoing electron. The threshold position and therefore the electron affinity can be determined from the frequency dependence of the photodetachment process. A critical aspect of an accurate determination of the electron affinity is the precise measurement of the photon frequency. One has to take into account that the frequency in the laboratory frame, ν_{Lab}, is connected with the frequency ν, as seen by the ion moving with the velocity v relative to the laboratory in its rest frame, by [8]

$$\nu = \nu_{Lab}(1 - (v/c)\cos\alpha)/[1 - (v/c)^2]^{1/2}. \quad (3)$$

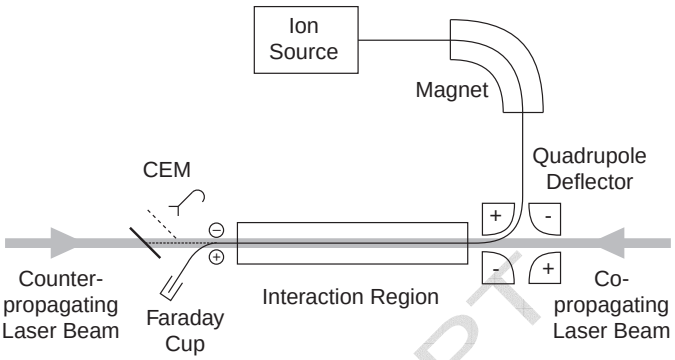

Fig. 1. Schematic of the experimental apparatus

Depending on the experimental geometry and ion velocity, the lab threshold frequency will therefore differ from the electron affinity frequency $\nu_{EA} = E_{EA}/h$, needed to promote electron detachment in the ion rest frame. By measuring the photodetachment thresholds both with parallel and anti–parallel laser and ion beams it is possible to extract the rest frame threshold frequency without explicitly measure the ion beam velocity. Otherwise the velocity of the ions has to been accurate enough to calculate the shift.

3. Experimental procedures

The experiments were performed using a negative ion beam apparatus at Göteborg University. A schematic of the setup is shown in Fig. 1. Negative ions are produced in a single–cathode cesium–beam sputter ion source Model PSX-120 from Peabody Scientific [9]. For the photodetachment measurements of W^- a mixture of tungsten and silver powder was used as cathode material. To produce beams of the various WF_x^- ions a tungsten powder was mixed with silver–fluoride. A beam with an energy of 6 keV was extracted from the source. The ions were mass separated using a 90° sector magnet with a mass resolution of $m/\Delta m \approx 200$. Ions of the selected mass were deflected into the interaction regions by the means of an electrostatic quadrupole deflector. In the interaction region the ion beam was merged with a laser beam over a length of 0.7 m. The interaction region was defined by two 3 mm diameter apertures. Downstream of the second aperture, the ions were deflected out of the laser beam into a Faraday cup by applying an electric field produced by two cylindrical electrodes. The measured ion current at this point was typically 10−30 pA. The vacuum system consisted of three differentially pumped sections which resulted in a pressure of the order of 10^{-9} mbar in the detection chamber.

The negative ions that were neutralized within the interaction region were not affected by the electric field and continued to move in the forward direction. The fast atoms impinged on a transparent tin-doped indium oxide (In_2O_3:SN) glass plate where secondary electrons were emitted. These electrons were detected with a channel electron multiplier (CEM). The glass plate and the CEM were housed in a box which was biased at −50 V. In this

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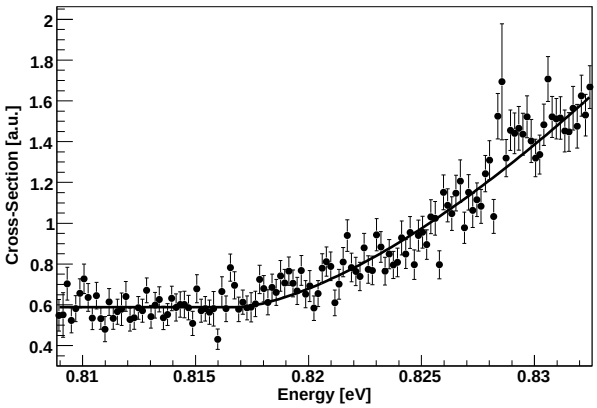

Fig. 2. The threshold region of the cross–section for photodetachment of W^-. The data points are obtained by scanning the laser in the vicinity of the onset of W atom production. The solid curve is a fit of the data points to a threshold curve described by a outgoing p–wave electron.

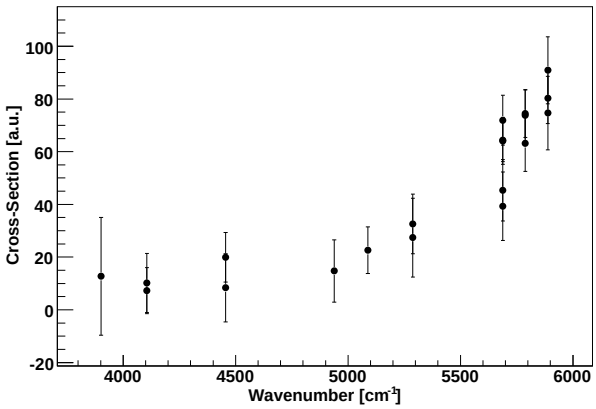

Fig. 3. The threshold region of the cross–section for photodetachment of WF^-. The data points are obtained by scanning the laser in the vicinity of the onset of WF atom production.

way stray electrons from other parts of the chamber kept from being detected. The output signal from the CEM was fed into a gated photon counter. The signal of the neutral atoms was delayed relative to the laser pulse by the finite time of flight between the interaction region and the detector. A narrow time window was set up in the photon counter to coincide with the arrival of the neutrals at the detector. The signals from the photon counter, the laser power meter and the Faraday cup were recorded in a PC using a LabView–based data acquisition system.

The laser beam was produced by an OPO/OPA system. As pump laser a Nd:YAG laser seeded by a solid state laser providing pulses with 6 ns length at a rate of 10 Hz was used. The fundamental output of 1064 nm from the Nd:YAG laser was frequency doubled and sent to the Optical Parameter Oscillator (OPO). This stage was tunable producing an idler beam in the range from 1350 to 2100 nm. The signal output was sent to a wavelength meter for measuring the tuned wavelength. The idler output was sent to an Optical Parametric Amplifier (OPA), which was pumped by the 1064 nm fundamental of the Nd:YAG pump laser. By using either the signal or the idler output of the OPA an infrared laser beam in the range from 1350 to 5000 nm is produced.

4. Results

In a first step the already known electron affinity of W^- was measured. The adopted value measured by Feigerle et al. [10] obtained by laser photodetachment electron spectroscopy is 0.815(8) eV. Our measurement was performed to show the ability of measuring laser photodetachment thresholds of heavy negative ions with masses around $M \approx 200$ amu with the current setup. A compilation of various scans around the onset of tungsten atom production is shown in Fig. 2. The detached electron is emitted as a p–wave which is incorporated into the fit of the onset according to the Wigner law. The measured photodetachment threshold agrees well with the value for the electron affinity published by Feigerle et al. The statistical uncertainty of our measurement is in the order of 0.3 meV, i.e. 25 times smaller than the uncertainty in the work by Feigerle et al. However, an accurate determination of the electron affinity of W^- will require an careful calibration of the energy scale. Such work is currently under progress and we expect to publish an improved value of the electron affinity of tungsten in the near future.

After successfully showing the ability of measuring photodetachment thresholds of heavy ions using the Göteborg negative ion beam apparatus we will proceed to measure the electron affinities of the various hafnium– and tungsten–fluorides. After succesfully identifying WF^- ions in the mass spectra photodetachment covering large energy regions was conducted for this molecular ion. In molecules various rotational and vibrational states can be excited, and complicated threshold behaviors than in the case of atomic negative ions can be expected. Fig. 3 shows a compilation of measured data points at different laser energies. These measurements suggest a threshold for the detachment of the bound electron around 4700 cm^{-1} (0.58 eV).

Threshold measurements of the higher tungsten fluorides as well as of the hafnium fluorides were limited by the transmission and the mass resolution of the current setup.

5. Discussion

The first results of the measurement were promising and proved that the determination of the electron affinity of atomic and molecular ions at masses around $M \approx 200$ amu is possible with the setup at Göteborg University. Two major problems have, however, prevented us from measuring heavier tungsten and hafnium fluorides with the present setup. The mass resolution of the system is not high enough to distinctively separate neighboring masses. This is crucial for the molecules as the various combinations of the differ-

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ent isotopes of tungsten and hafnium together with fluor, oxygen and hydrogen results in complicated mass spectra. As the current setup has a mass resolution of the order of $m/\Delta m \approx 200$ a considerable improvement is necessary. Another problem is the low transmission of the system which results in low ion currents around 10 to 30 pA. A redesign of the Göteborg apparatus is currently underway and the required modifications are foreseen for the end of 2007. After this modifications an accurate determination of the various fluorides up to the tungsten and hafnium pentafluorides will be possible.

Acknowledgments

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