RECENT UPGRADES OF THE SHIPTRAP SETUP: ON THE FINISH LINE TOWARDS DIRECT MASS SPECTROSCOPY OF SUPERHEAVY ELEMENTS*

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With the Penning-trap mass spectrometer SHIPTRAP at GSI, Darmstadt, it is possible to investigate exotic nuclei in the region of the heaviest elements. Few years ago, challenging experiments led to the direct measurements of the masses of neutron-deficient isotopes with $Z = 102, 103$ around $N = 152$. Thanks to recent advances in cooling and ion-manipulation techniques, a major technical upgrade of the setup has been recently accomplished to boost its efficiency. At present, the gap to reach more rare and shorter-lived species at the limits of the nuclear landscape has been narrowed.

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1. Introduction

One of the most exciting challenges in nuclear physics in the last eighty years has been the unceasing quest for new elements. In this time span, at the upper end of the chart of nuclides the search for the next closed proton and neutron shell beyond ^{208}Pb has yielded exciting discoveries, extending the Z of the known elements from 92 to 118 (see, for instance, Refs. [1, 2]). In this region, exotic superheavy elements (SHE) exhibit unexpected and extraordinary atomic and nuclear properties. They exist as bound systems only because shell effects counterbalance the strong Coulomb repulsion that would cause their spontaneous fission. All the same, their atomic structure and chemical behaviour are significantly influenced by relativistic effects. In order to understand the existence of the SHEs, the evolution of the strength of shell effects should be experimentally explored. This was difficult until recently since information on the mass of SHEs was deduced only indirectly and with uncertainties of hundreds of keV.

Direct precision measurements of SHE atomic masses have become possible exploiting Penning-trap mass spectrometry (PTMS) [3–5]. Applied to rare and unstable nuclides, this technique enables us to investigate their nuclear structure since the absolute binding energies and derived quantities such as the nucleon separation energies can be readily measured even for radionuclides whose detailed nuclear structure is unknown.

The first direct mass measurements in the region above fermium ($Z = 100$) have been performed at the GSI Darmstadt, Germany with the Penning-trap mass spectrometer SHIPTRAP [6–8]. The binding energies of six nobelium and lawrencium isotopes ($Z = 102, 103$, respectively) have been determined with a precision of the order of $\delta m/m \approx 10^{-8}$ and allowed mapping the strength of shell effects in the vicinity of $N = 152$, confirming the existence of a region of enhanced shell stabilization for nuclides above the doubly magic ^{208}Pb. Furthermore, these data have been combined with results from the decay spectroscopy where superheavy elements are identified based on their α-decay chain. The masses of short-lived heavier nuclides up to ^{270}Ds, previously known from spectroscopic studies, have been improved using the masses of nobelium and lawrencium isotopes as anchor points [7].

In order to investigate the evolution of shell effects at $N = 152$ and further at $N = 162$ for different proton numbers, new accurate mass measurements of heavier nuclides, as ^{254}Lr and ^{257}Rf, are foreseen [9]. However, the corresponding cross sections decrease steeply to the level of a few nb or even pb. Only with the implementation of new technical and methodical developments, that will be described in the next sections along with an overview of SHIPTRAP, the sensitivity and efficiency of the whole setup is improved to match such challenging needs.

2. The SHIPTRAP mass spectrometer

The Penning-trap mass spectrometer SHIPTRAP takes advantage of its unique position, being operated at the SHIP recoil separator [10]. The latter provides, at one time, a high transmission for the reaction products of interest and an enhanced suppression of the primary beam and unwanted secondary products. The primary beam (typically ^{40}Ar, ^{48}Ca, ^{50}Ti) delivered by the UNILAC accelerator is sent to a rotating target wheel consisting of isotopically enriched Pb or Bi compounds with a typical thickness of $\approx 500\ \mu g/cm^2$. The products of fusion-evaporation reactions that recoil out of the target are then focused and selected depending on their velocity by SHIP that is configured as a two-stage velocity filter (Wien filter).

According to the reaction kinematics, the transmitted products leave the separator with a kinetic energy of about 40–45 MeV. They are slowed down in the following stage of the setup comprising a buffer-gas stopping cell [11]. The energy of such ions is degraded by a metallic entrance window that gives access to the active volume of the buffer-gas cell where 50 mbar ultrahigh-purity helium is used to stop the reaction products. They are guided by electric DC gradients and an RF field towards an extraction aperture (a de Laval nozzle) at the far end side. They are extracted after few ms as singly or doubly charged ions in a supersonic gas jet and efficiently guided by a radio frequency quadrupole (RFQ) at a buffer gas pressure of 10^{-2} mbar. A pumping barrier marks off the injection to a second RFQ, where ions are guided and accumulated into a potential well before being extracted in short bunches (≈ 100 ns long), with an overall efficiency of about 60% [12]. Electrostatic ion optics guides the ion bunches inside the 7 T superconducting solenoid magnet, where the double Penning-trap system is located. The trap electrodes have a cylindrical geometry, with the trap centres located in the homogenous magnetic field regions of the superconducting magnet and additional correction electrodes that ensure the harmonicity of the electrostatic trap potential. In the first trap, the incoming ion bunch is prepared for the measurement with a further buffer-gas cooling stage and isobaric purification with a mass resolving power up to 10^5 [13]. In the second trap, the actual mass measurement is performed in ultrahigh vacuum. A sketch of the setup is given in Fig. 1.

The mass of ions confined in a homogenous magnetic field is determined via the measurement of the cyclotron frequency $\nu_c = qB/(2\pi m)$ of their ionic form (where q is the ion charge and m the ion mass). In a Penning trap, the confinement is achieved by the combination of electrostatic and magnetic fields. The ion motion can be described as the superposition of three independent harmonic eigenmotions, one along the axial direction (z-axis) and two radial components, a fast *cyclotron motion* ν_+ and a slow

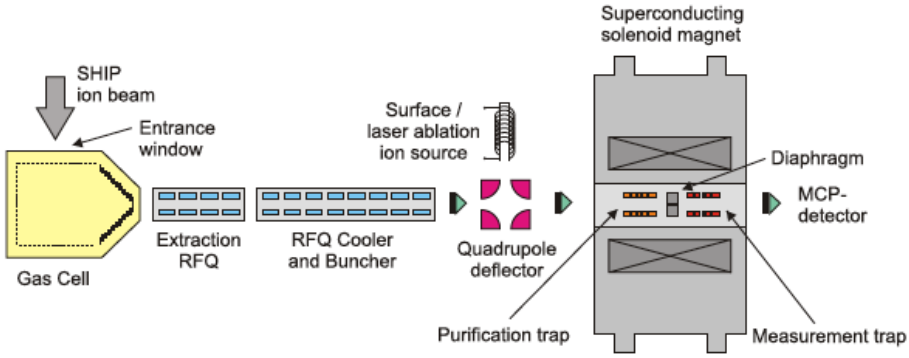

Fig. 1. Schematic overview of the Penning-trap mass spectrometer SHIPTRAP at GSI, in the configuration used for the No and Lr mass measurements.

magnetron motion ν_- along the xy-plane. In an ideal Penning trap, the condition $\nu_c = \nu_- + \nu_+$ is fulfilled [14]. Applying external RF dipolar or quadrupolar fields, the amplitude of the eigenmotions can be manipulated as well as a periodic interconversion between the two radial modes is achieved. Usually, the measurement of the cyclotron frequency for the ion of interest is compared to the value of a reference ion to perform a calibration of the magnetic field. For the determination of the mass of nobelium and lawrencium isotopes, the adopted reference ion with the well-known mass and similar m/q ratio as doubly charged No and Lr ions was $^{133}Cs^+$. The cyclotron frequency was measured following the standard ToF-ICR (Time-of-Flight Ion-Cyclotron-Resonance) technique [15].

3. The cryogenic buffer-gas cell

A new cryogenic buffer-gas cell (CryoCell) has been developed and characterised in the last years [16, 17]. It will go beyond some of the constraints of the cell operated at room temperature, featuring a larger stopping volume and a more favourable on-axis injection, and will boost the overall efficiency which allows extending the reach towards nuclides with lower production cross section than ^{256}Lr.

In a commissioning beam time in September 2015, ^{254}No ions produced in the fusion-evaporation reaction $^{208}Pb(^{48}Ca,2n)^{254}No$ and separated by SHIP were injected into the CryoCell with a rate of about 3 ions/s. After extraction, the ions were implanted in a 0.8 μm Al foil and their α decay was recorded in an Si detector. Although the nominal operation temperature of 40 K was not reached due to lack of time, an overall efficiency of 33(5)% at about 90 K was obtained. It corresponds to an increase in the overall efficiency by more than a factor of 3 in comparison with the previous

buffer-gas cell. Further improvements are expected when the CryoCell is operated with an optimum entrance window thickness and gas pressure, at a temperature of 40 K.

In order to integrate the CryoCell into the SHIPTRAP setup, the entire beam line has been relocated at an angle of 3.5 degrees with respect to the recoil separator SHIP in the first semester of 2016 (see Fig. 2). The combined operation of the buffer-gas cell and buncher has been thoroughly optimized and the whole setup is fully operational again.

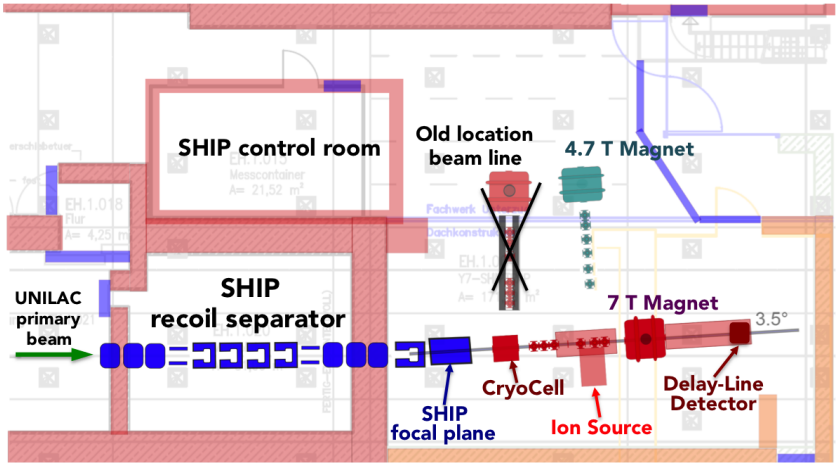

Fig. 2. Layout of the SHIPTRAP experimental hall after the relocation of the setup. The old beam line position is marked with a black cross.

4. New mass measurement techniques

Recently, a novel technique to measure the cyclotron frequency has been developed and applied at SHIPTRAP [18, 19]. The Phase-Imaging Ion-Cyclotron Resonance (PI-ICR) method overcomes the limitations of the ToF-ICR technique whose resolving power is limited by the excitation time and, therefore, by the half-life of the ion of interest. This new method is about 25 times faster and provides a 40-fold increased resolving power that enables the separation of low-lying isomeric states few tens of keV above the ground state. These features are particularly favourable when applied to the heaviest nuclides, in order to investigate their low-lying isomeric states, and to access shorter-lived isotopes. The same level of precision can be now reached in shorter measurement times, as well as with a smaller number of ions per measurement.

The ion cyclotron frequency ν_c is determined projecting its radial motion in the trap onto a high-resolution position-sensitive microchannel plate detector after ejection from the trap. Applying a dipolar RF field with an initial phase ϕ_i, the ions are exited to a certain orbit with radius R from the trap center and perform a number n of full revolutions in a certain accumulation time t. From the phase difference ϕ for two different revolution times, the frequency is obtained according to $\nu = (\phi + 2\pi n)/2\pi t$, where t is the phase accumulation time, and n is the number of full revolutions an ion performs in a time t. The position-sensitve detector has been installed at the far end of the SHIPTRAP beam line, at about a meter distance from the measurement trap center, with a manipulator system to optimize its position under vacuum. An ion optical system guarantees the transport of the ions to the detector in a homogenous constant electric field, resulting in a magnification factor of the ion spot on the detector surface of about 20. The PI-ICR method has been first applied at SHIPTRAP to measure the mass difference of long-lived and stable nuclides reaching a relative precision of about 10^{-10} [20–22]. First on-line mass measurements with the new technique are planned for the coming beam time.

In October 2016, a second superconducting magnet with a 4.7 T field has been installed at SHIPTRAP. In the near future, it will be connected to the existing beam line perpendicularly to the surface and laser ablation ion sources (see Fig. 2). The new beam line branch is devoted to the development of single-ion mass spectrometry based on the Fourier-Transform Ion-Cyclotron-Resonance (FT-ICR) technique where the cyclotron frequency is obtained by the measurement of the induced image charge by the ions into the trap electrodes [23]. A major advantage of the non-destructive FT-ICR technique is the possibility to perform mass measurements with a single ion. For long-lived nuclides, multiple measurements can be performed on the same ion until it decays. However, cryogenic operation of the trap system is mandatory to reach the single-ion sensitivity. A bath cryostat for the trap cooling has been designed and will be soon installed at SHIPTRAP [24, 25].

5. Summary

With the pioneering high-accuracy direct mass measurements of several nobelium and lawrencium isotopes at SHIPTRAP, the limit of the nuclides with the smallest production rate ever investigated at an on-line Penning-trap mass spectrometer (^{256}Lr) has been reached. After several technical and methodical developments, the sensitivity and efficiency of the SHIPTRAP setup have been further improved such that mass measurements can be performed in a shorter time and with a smaller amount of ions. With these upgrades, the investigation of the heavier elements is now within reach.

REFERENCES

[1] G. Münzenberg, *Nucl. Phys. A* **944**, 5 (2015).
[2] Y. Oganessian, *J. Phys. G: Nucl. Part. Phys.* **34**, R165 (2007).
[3] K. Blaum, *Phys. Rep.* **425**, 1 (2006).
[4] H.J. Kluge, *Int. J. Mass Spectrom.* **349**, 26 (2013).
[5] D. Lunney, J. Pearson, C. Thibault, *Rev. Mod. Phys.* **75**, 1021 (2003).
[6] M. Block *et al.*, *Nature* **463**, 785 (2010).
[7] M. Dworschak *et al.*, *Phys. Rev. C* **81**, 064312 (2010).
[8] E. Minaya Ramirez *et al.*, *Science* **337**, 1207 (2012).
[9] M. Block, *Nucl. Phys. A* **944**, 471 (2015).
[10] G. Münzenberg *et al.*, *Nucl. Instrum. Methods* **161**, 65 (1979).
[11] J. Neumayr *et al.*, *Nucl. Instrum. Methods Phys. Res. B* **244**, 489 (2006).
[12] D. Rodriguez, Ph.D. Thesis, Universidad de Valencia, 2003.
[13] G. Savard *et al.*, *Phys. Lett. A* **158**, 247 (1991).
[14] G. Gabrielse, *Int. J. Mass Spectrom.* **279**, 107 (2009).
[15] M. König *et al.*, *Int. J. Mass Spectrom. Ion Process.* **142**, 95 (1995).
[16] S. Eliseev *et al.*, *Nucl. Instrum. Methods Phys. Res. B* **266**, 4475 (2008).
[17] C. Droese *et al.*, *Nucl. Instrum. Methods Phys. Res. B* **338**, 126 (2014).
[18] S. Eliseev *et al.*, *Phys. Rev. Lett.* **110**, 082501 (2013).
[19] S. Eliseev *et al.*, *Appl. Phys. B* **114**, 107 (2014).
[20] D. Nesterenko *et al.*, *Phys. Rev. C* **90**, 042501 (2014).
[21] S. Eliseev *et al.*, *Phys. Rev. Lett.* **115**, 062501 (2015).
[22] P. Filianin *et al.*, *Phys. Lett. B* **758**, 407 (2016).
[23] A.G. Marshall *et al.*, *Mass Spectrom. Rev.* **17**, 1 (1998).
[24] C. Weber *et al.*, in: The 4$^{\text{th}}$ Int. Conf. on Exotic Nuclei and Atomic Masses, (Eds.) C.J. Gross, W. Nazarewicz, K.P. Rykaczewski Springer, 2005, pp. 65–66, DOI: 10.1007/3-540-37642-9_19.
[25] R. Ferrer *et al.*, *Eur. Phys. J. Spec. Top.* **150**, 347 (2007).