

## **Extracts of the 2010 Annual Report** **of the (former) JGU “Institute of Nuclear Chemistry”**

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# Progress of the on-line coupling of TRIGA-SPEC

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**Introduction:** All interactions inside a nucleus are reflected by ground state-properties like mass, magnetic moment, spin and charge radius. Thus, high-precision measurements of these properties are of fundamental importance for nucleosynthesis and nuclear structure studies. The TRIGA-SPEC experiment, which is located at the research reactor TRIGA Mainz, aims for high-precision investigations on neutron-rich radionuclides to obtain the ground-state properties model-independently by means of Penning trap mass spectrometry and collinear laser spectroscopy [1].

The radionuclides are produced by thermal neutron-induced fission of an actinoid target like <sup>235</sup>U or <sup>249</sup>Cf mounted in a target chamber near the reactor core. The fission products are thermalized in an argon atmosphere at a pressure of about 1.4 bars pressure. After thermalization the non-volatile fission products attach to carbon aerosol particles seeded in the argon gas. The aerosol is subsequently flushed out through a capillary to the experimental setup, where carrier gas and aerosol particles are separated by a skimmer. The fission products break off from the aerosol particles in an electron plasma inside an ECR or in a high-temperature surface ion source on a high-voltage platform. After ionization the fission products are mass-separated in a dipole magnet, cooled and bunched in a radiofrequency quadrupole, and finally transferred either towards the collinear laser spectroscopy beam line or the Penning trap mass spectrometer setup.

**Experimental setup and results:** The experimental setup of the ECR ion source with the skimmer chamber mounted on an high-voltage platform is shown in Fig. 1. When the gas leaves the capillary, it expands in a Mach cone. The hole of the skimmer is placed inside the cone, the so-called zone of silence, where the supersonic expansion takes place without turbulences.

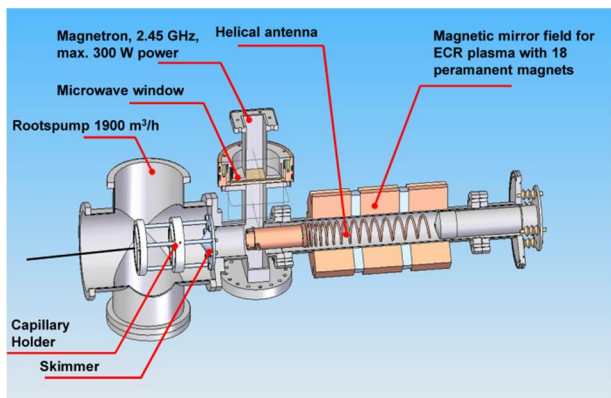


Fig. 1: Sketch of the ECR ion source with the skimmer region. The gas-jet enters the skimmer region through the capillary on the left, passes the skimmer, and is ionized in the plasma.

While most of the light carrier gas is pumped away by a roots pump, the heavy aerosol particles in the center of the Mach cone pass the skimmer and enter the ECR ion source. Inside the ion source the electrons are heated by a 2.45-GHz microwave on a closed surface of constant magnetic field strength, where the excitation frequency matches the cyclotron frequency of the electrons. Thereby, a hot electron plasma is created and confined due to the magnetic mirror effect.

Pure argon was used as carrier gas for the first tests of the ECR ion source and mass separator. The ion source was operated at a high-voltage of 20 kV and after ionization argon and residual ions were extracted from the plasma to ground. The beam was then mass-separated in the dipole magnet and detected on a Faraday cup. Figure 2 shows the beam current as a function of the magnetic field. In the data evaluation the mass-to-charge ratio was assigned to the detected ions and a present mass resolving power of about 100 was achieved.

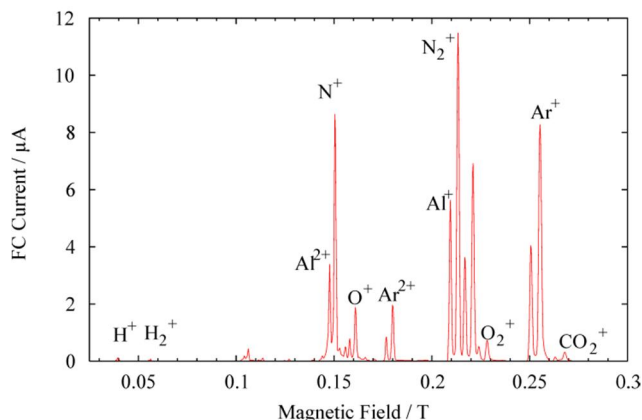


Fig. 2: Mass spectrum of the ions produced in the ECR ion source in off-line operation.

**Conclusions and outlook:** The ECR ion source as well as the mass separator are being commissioned. Next, the ionization of different elements and the separation of fission products and aerosol particles will be investigated. In addition, a radiofrequency quadrupole cooler buncher will be set up and the resolution of the magnet will be increased by implementing a slit system.

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## Acknowledgements

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# Production and characterization of gadolinium nitrate targets on superhydrophobic surfaces for high-precision mass spectrometry

D. Renisch<sup>1,2</sup>, T. Beyer<sup>2,3</sup>, K. Blaum<sup>2,3</sup>, Ch. E. Düllmann<sup>1,4,5</sup>, K. Eberhardt<sup>1</sup>, M. Eibach<sup>1,2</sup>, A. Gonschior<sup>1,4</sup>, J. Ketelaer<sup>2</sup>, Sz. Nagy<sup>2,4</sup>, D. Neidherr<sup>2,5</sup>, W. Nörtershäuser<sup>1,4</sup> and C. Smorra<sup>1,3</sup>

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**Introduction:** The TRIGA-TRAP project carries out high-precision mass measurements of neutron-rich fission products and actinide nuclides using a Penning trap [1]. In case of off-line measurements, the investigated elements are ionized and injected into the apparatus using a laser ablation ion source [2]. For this purpose a target, which consists of the element of interest or an appropriate compound deposited on a backing material, is used. Ions are produced by laser irradiation and extracted towards the Penning traps. Up to now, the usual target preparation technique was to evaporate a drop of a solution on the backing, leading to an inhomogeneous deposition of the dissolved material. To obtain improved targets, several different backing materials (zinc, copper, silver, titanium, glassy carbon) were chemically and physically modified to create superhydrophobic surfaces with contact angles  $\geq 150^\circ$ . Droplets of aqueous solutions retain their spherical shape on such surfaces, because wetting of the surface is energetically unfavourable. Evaporation of the drop yields a circular, homogeneous spot of the precipitate [3].

**Experimental:** To produce superhydrophobic surfaces it is necessary to have both, a rough surface and a water-repellent layer [4]. Thus all used metal foils were etched in an appropriate acid, and the glassy carbon backings were sandblasted before further treatment. The next step was the deposition of a thin, hydrophobic layer. For the metal foils the most promising technique was the adsorption of polyfluorinated alkyl chains with an anchor group that binds to the metal surface, so that self-assembled monolayers (SAM) were formed. Heptadecafluoro-1-decanthiol (HDFT) was used for Zn, Cu and Ag, (Heptadecafluoro-decylsulfanyl)-ethylphosphonic acid (HDFP) was applied for Ti. Because there are no appropriate anchor groups for pure carbon, it was not possible to create SAMs on glassy carbon surfaces. Therefore a commercially available cyclosiloxane (Tegotop<sup>®</sup>105) solution was sprayed onto the glassy carbon backings. In a last step a drop of the solution of the element under investigation was put on the hydrophobic surface and evaporated. Gadolinium nitrate was chosen as target material for all test series.

**Results:** Radiographic imaging is used to check the homogeneity of the deposited material, using Gd-153 as radioactive tracer. As shown in Fig. 1, pre-treatment of the backing material results in a much more homogeneous deposition of the target material. To investigate the improvement in ionization efficiency with the new targets, a MALDI-TOF mass spectrometer (BRUKER Reflex III) was equipped with a laser system similar to that of the TRIGA-TRAP setup, so that a laser

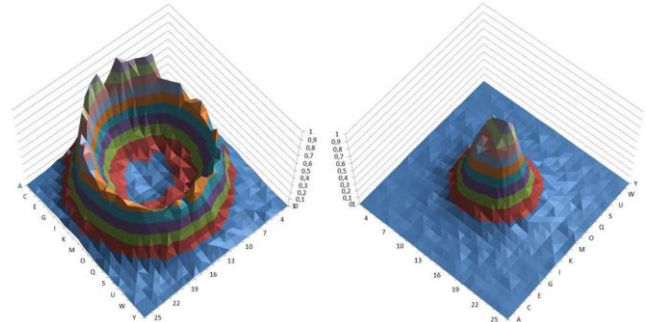


Figure 1. Radiographic imaging of radioactive Gd targets. The target on the left side was prepared on an untreated Ti foil, the one on the right side was prepared on a superhydrophobic Ti foil.

ablation ion source was available for test purposes. The results of the tests with Ti backings are shown in Fig. 2. The yield of ions per laser pulse is significantly increased by using the new targets compared to the two references (non-modified Ti backings). The Tegotop<sup>®</sup>105 layer is not stable under laser irradiation conditions, and accordingly, no satisfactory results were obtained with the glassy carbon backings [5].

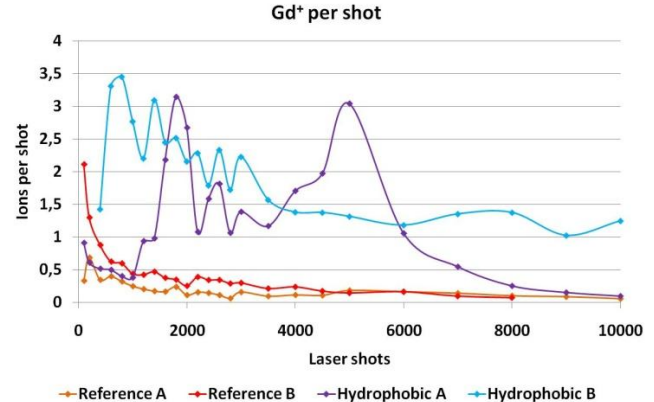


Figure 2. Gd<sup>+</sup> ions per laser shot detected with the modified MALDI-TOF mass spectrometer. The two reference targets were prepared on an untreated Ti foil, the two hydrophobic targets were prepared on a Ti foil which was pre-treated with HDFP.

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# Toward large-area targets for “TRAKULA”

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**Introduction:** TRAKULA (Transmutationsrelevante kernphysikalische Untersuchungen langlebiger Aktinide, i.e., nuclear physical investigations of long-lived actinides with relevance to transmutation) is a joint research project on nuclear physics investigations with modern scientific, technological and numerical methods, in which experiments concerning the transmutation of radioactive waste are a central topic. For this, large-area samples ( $\geq 40 \text{ cm}^2$ ) of  $^{235,238}\text{U}$  and  $^{239,242}\text{Pu}$  are required for the calibration of fission chambers and for neutron-induced fission yield measurements. Another topic requires large-area targets for precise measurements of the half-life,  $t_{1/2}$ , of very long-lived  $\alpha$ -particle emitters like  $^{144}\text{Nd}$  ( $t_{1/2} \approx 2 \cdot 10^{15} \text{ y}$ ).

Electrodeposition tests with Gd and Nd (used as chemical homologues of the actinides), were performed to find optimal deposition conditions for small-area targets that should be applicable to large-area targets.

**Experimental:** The layers were produced by Molecular Plating (MP) on  $5 \mu\text{m}$  backings. The backing foils were pre-cleaned with 6 M HCl, water, and isopropanol. The lanthanide compound in the nitrate form was dissolved in 0.1 M  $\text{HNO}_3$  to an elemental concentration of  $\approx 25 \text{ mg/ml}$ . Two different procedures were followed to produce targets: (i) an aliquot of  $100 \mu\text{l}$  of the stock-solution was mixed with 1 ml isopropanol, and transferred into the electrochemical cell, which was then filled up with 24 ml isobutanol (hereafter referred to as inactive MP); (ii) an aliquot of  $100 \mu\text{l}$  of the stock solution was irradiated in the TRIGA Mainz research reactor, in order to produce  $^{153}\text{Gd}$  ( $t_{1/2} = 239.47 \text{ d}$ ) and  $^{147}\text{Nd}$  ( $t_{1/2} = 10.98 \text{ d}$ ), respectively, as radioactive tracers. The irradiated solution was then used for the MP (hereafter referred to as active MP). The plating solution was stirred either with a magnetic stirrer (Variomag Compact) operated at 1000 rotations per minute (rpm) or a quartz tip ultrasonic stirrer (Bandelin Sonopuls HD 2070) operated at 30% power pulse. MP was carried out at  $14^\circ\text{C}$  by applying two different constant currents: 2 mA and 4.2 mA, yielding current densities, respectively, of  $0.7 \text{ mA/cm}^2$  and  $1.4 \text{ mA/cm}^2$ .

The deposition yield was determined by an indirect and a direct method, respectively: Neutron Activation Analysis (NAA) and  $\gamma$ -spectroscopy. NAA was used after inactive MPs to determine the residual concentration of the lanthanide element in the supernatant solution via  $\gamma$ -spectroscopy of a standard sample and an aliquot of the supernatant solution irradiated simultaneously (indirect yield measurement). Gamma-spectroscopy was used after active MPs. This technique employed a high-purity germanium detector (HPGe) (GEM 23158 P-Plus, ORTEC Company) to measure the active samples (direct yield measurement). To obtain quantitative data, reference sources with

known amounts of the tracer were prepared. They consisted of filter papers with the same geometry as the targets, soaked with the tracer-containing solution. To avoid any kind of contamination inside the  $\gamma$ -spectrometer, the active samples were sealed before being inserted into the sample holder.

The homogeneity of the radioactive targets was inspected by using Radiographic Imaging (RI), using a FUJIFILM FLA 7000 equipped with reusable imaging plates and a 650 nm laser for the reading process.

The morphology of the targets was studied by using a Scanning Electron Microscope (SEM) (Philips XL30), operated at 20 kV.

The surface roughness of the targets was investigated by using an Atomic Force Microscope (AFM) (MFP 3D, Asylum Research). Fig.1 shows a surface whose mean roughness is  $\sim 100 \text{ nm}$ . The AFM used cantilevers with a resonance frequency of 283 kHz for imaging in the AC mode (tapping mode).

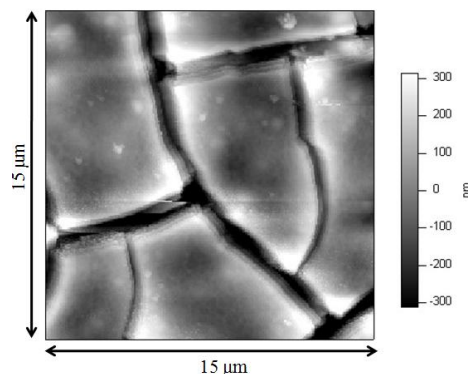


Figure 1. AFM picture of the surface of a Nd-target obtained at high current density ( $1.4 \text{ mA/cm}^2$ ), applying ultrasonic stirring.

**Results:** The characterization of the layers with different analytical techniques (i.e. NAA,  $\gamma$ -spectroscopy, RI, SEM, AFM) proved that ultrasonic stirring is superior to mechanical stirring, especially for the production of large-area targets, and that the two different current densities applied, i.e.  $0.7 \text{ mA/cm}^2$  and  $1.4 \text{ mA/cm}^2$ , are both suitable for target preparation.

According to the results obtained from these tests, a new electrochemical cell for the production of large-area targets was designed, built and recently tested with success.

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# Liquid – liquid – extraction with the MicroSISAK – system

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For studies of the chemical properties of the heaviest elements ( $Z > 103$ ), it is indispensable to use fast and efficient systems, because of their short half-lives and low production rates [1]. An established possibility for liquid chemistry in this field is the liquid – liquid – extraction, like it is used in the SISAK – system [2]. The *Institut für Mikrotechnik Mainz* (IMM) developed a device that agitates two liquids via a static digital mixer and separates them again via a hydrophobic Teflon membrane by using micro reaction technology [3]. It is called **MicroSISAK** and consists of micromechanical discs made of titanium with an inner volume in the  $\mu\text{l}$  – range. Previous experiments have shown that the principal idea could be realised with this apparatus.

Complete phase separation is achieved by setting a backpressure at the outlet of the aqueous phase of the device [4].

In a series of experiments we studied the influence of the temperature on the extraction yield in the MicroSISAK – apparatus. For this we chose the extraction system Hf in  $\text{H}_2\text{SO}_4$  vs. TOA in toluene. Temperature control is achieved via an electric heating element in a copper strip around the microreactor. As expected, the extraction yield rises with increasing temperature for different flow rates or concentrations of TOA and  $\text{H}_2\text{SO}_4$  respectively (figure 1). The extraction yield was determined using Hf-181 as a radioactive tracer.

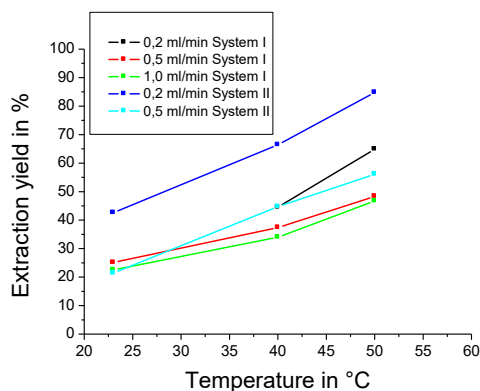


Figure 1. Extraction yield in the MicroSISAK – apparatus depending on the temperature for different flow rates and concentrations. System I: 1,0 mol/l TOA in toluene vs. 0,5 mol/l  $\text{H}_2\text{SO}_4$ . system II: 0,1 mol/l TOA in toluene vs. 0,1 mol/l  $\text{H}_2\text{SO}_4$ .

Another possibility to get higher extraction yields is to extend the contact time of the two phases. If the flow rates shall not be changed, one has to increase the volume between the mixer and the separation unit. Therefore the IMM designed a new disc with meandering flow scheme, to integrate into the MicroSISAK stack system (figure 2). Now extraction experiments were made for different flow rates of the system  $\text{H}_2\text{SO}_4$  (0,1 mol/l) with Hf-181 ( $10^{-6}$  mol/l) vs. TOA in toluene (0,1 mol/l).

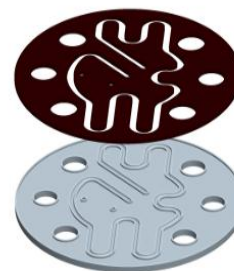


Figure 2. Scheme of the new disc (grey) for the MicroSISAK to increase the contact volume, with a seal (black)

Here also the extraction yield rises as it was expected from 25-40 % without the new disc (see [5]) to now 60-80 % with it (see figure 3).

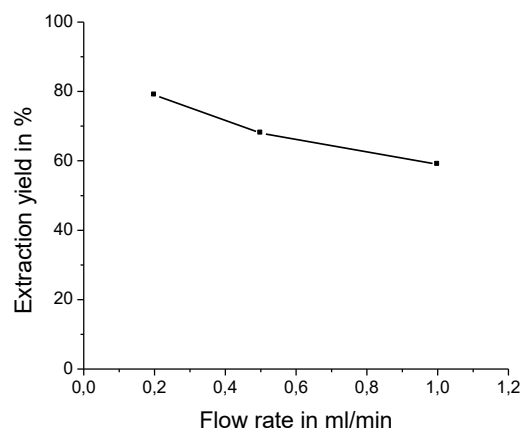


Figure 3. Extraction yield depending on the flow rate for the modified MicroSISAK - apparatus

Similar experiments - larger contact volume and higher temperature - were also made with the system Tc-99m (carrier-free) in nitric acid (0,01 mol/l) vs. TPAC in chloroform ( $10^{-4}$  mol/l) and evidenced the same results: higher extraction yield than in earlier experiments [5].

In the near future, on-line experiments with fission products are planned at the research reactor TRIGA Mainz.

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# Towards fully-relativistic simulations of the adsorption of super-heavy elements on $\alpha$ -SiO<sub>2</sub> surfaces \*

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While in the last two decades super-heavy elements with  $Z \leq 112$  have been studied, the focus of the present work is on the chemical properties of element 114. Our theoretical calculations have been motivated by two conflicting gas-chromatography experiments, which aimed on studying the interaction strength of element 114 with a gold surface. The experiment by Eichler *et al.* [1] reported adsorption in the chromatography column at only very low temperatures of approximately  $-90^\circ\text{C}$ , from which they concluded a weak interaction between element 114 and the gold surface. In contrast, experiments performed at GSI [2] observed adsorption at room temperature, indicating a much stronger bond between element 114 and gold. To resolve this conflict further experiments will be performed at GSI within the next two years, where besides gold SiO<sub>2</sub> will be used as detector material.

Besides the previously mentioned experiments, extensive theoretical studies on the adsorption of element 114 on gold surfaces were performed using fully-relativistic DFT methods [3], while the adsorption on inert surfaces such as SiO<sub>2</sub> were estimated using semi-empirical methods in conjunction with computed properties of atoms, dimers, or small molecules [3, 4]. Unfortunately a fully-relativistic treatment of the entire adsorption process is beyond the capacities of nowadays computing resources. However, this work can be divided into two steps: (i) extensive studies on SiO<sub>2</sub> bulk and surface properties (e.g. stable bulk-phases or possible surface structures and terminations) using a non-relativistic approach; (ii) fully-relativistic calculations on the adsorption process of element 114 on SiO<sub>2</sub>, where the most stable and interesting surface structures obtained in the first step serve as basis.

So far we have focused on the first task, understanding the surface structure of SiO<sub>2</sub> under realistic experimental conditions. For these calculations, the CASTEP code [6] with Vanderbilt-type ultrasoft pseudopotentials [7] and the PBE exchange–correlation functional has been used. The obtained DFT-energies were then used in conjunction with the *ab initio* atomistic thermodynamics approach [5] to evaluate the surface phase diagrams, providing information of the surface stability as function of surrounding temperature and pressure. Starting with bulk systems, our calculations show that at experimental conditions ( $p_{\text{O}_2} = 10^{-13}$  atm,  $100\text{ K} < T < 320\text{ K}$ ) the most stable bulk structure is the so called  $\alpha$ -quartz. Using this crystal structure as basis, various surface orientations and morpholo-

gies were studied. We find that the thermodynamically preferred structure is the  $\alpha$ -SiO<sub>2</sub>(001) surface, which could assume three different morphologies: one Si-terminated and two different O-terminated structures.

Figure 1 shows the surface free energy,  $\gamma(p, T)$ , for the most stable surface structures as function of the oxygen chemical potential. Structure **a**, which is Si-terminated, is favored at  $\Delta\mu_{\text{O}} < -6.24\text{ eV}$  ( $T \gg 1000\text{ K}$  under UHV conditions). In the temperature range until  $\sim 100\text{ K}$  the surface is O-terminated (structure **b**). The phase diagram shows the existence of a fourth, but less stable structure (structure **d**), which is terminated by a single O-layer.

After understanding the surface morphology of SiO<sub>2</sub> under experimental conditions, the next step will be to perform fully-relativistic DFT calculations on the actual adsorption process of elements 112 and 114 and their homologs on a  $\alpha$ -SiO<sub>2</sub>(001) surface (structure **b**).

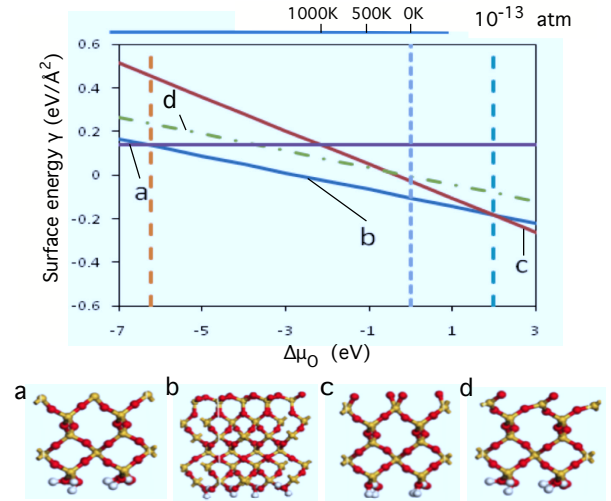


Figure 1: Surface free energy of the  $\alpha$ -SiO<sub>2</sub>(001) as function of  $\Delta\mu_{\text{O}}$  or temperature at fixed pressure of  $10^{-13}$  atm.

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# Transport of fission products using a gas-jet with dry ice aerosol particles

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**Introduction:** The TRIGA-SPEC experiment [1], which is installed at the TRIGA Mainz reactor aims to determine nuclear ground-state properties of neutron-rich fission products like masses, magnetic moments, spin and charge radii in a model independent way by means of Penning trap mass spectrometry and laser spectroscopy. Neutron-rich fission products are produced at the TRIGA Mainz reactor in a target chamber containing a fissionable target, e.g. <sup>235</sup>U or <sup>249</sup>Cf close to the reactor core. To transfer these products to the TRIGA-SPEC setup, a gas-jet system using N<sub>2</sub>/CO<sub>2</sub> or CO<sub>2</sub> gas and dry ice aerosol particles has been tested.

**Experiment:** Dry ice aerosol particles were produced by allowing liquid CO<sub>2</sub> (kept at a pressure of about 60 bars) to expand to atmospheric pressure through a nozzle. In the expansion zone behind the nozzle solid dry ice particles (clusters) form and flow together with the carrier gas through a polyethylene capillary to the target chamber.

Using an electrostatic classifier we measured the particle size distribution [2]. At a gas flow rate of 1.0 l/min particle diameters were from 10 nm to 500 nm lognormally distributed around a maximum at about 80 nm, however, the mean size decreased to 30 nm within 30 min. The gas-jet containing these particles was fed into the target chamber, which was equipped with a <sup>249</sup>Cf target. The reactor was operated at a thermal power of 100 kW. The fission products were thermalized in the gas at a pressure of about 2.5 bars, attached to dry ice aerosol particles, and were flushed out of the target chamber in a laminar flow through a 8-m long capillary of 0.5 mm inner diameter to a glass-fiber filter, where they were collected. The transport efficiency from the target chamber to the filter was measured. Sample collection lasted for 4 min. Measurement of the filter with a Ge(Li)  $\gamma$ -detector was started 1 min after the end of collection. A typical spectrum is shown in Fig.1.

**Results:** The extraction of fission products using dry ice aerosol particles was demonstrated for different gas flows of CO<sub>2</sub> and N<sub>2</sub> (see Fig.2). The transport efficiency will be investigated in detail. Values up to 60% (relative to a N<sub>2</sub>/KCl gas-jet) have been reached in first tests.

**Outlook:** The gas-jet will be connected to a skimmer system where the gas will be separated from the aerosol particles. To release the fission products from the aerosol particles and to ionize them, an ECR ion source is being installed on a high-voltage platform [3]. After ionization the fission products will be mass separated in a 90° dipole magnet and finally transferred either towards the laser spectroscopy experiment TRIGA-

LASER or the Penning trap mass spectrometer TRIGA-TRAP.

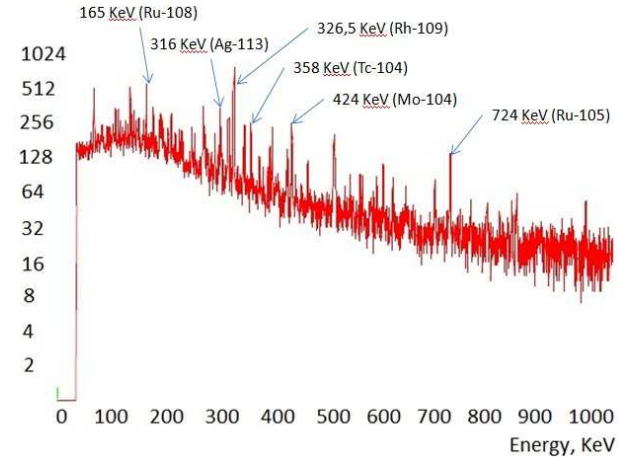


Figure 1.  $\gamma$ -spectrum of fission products obtained at a gas flow of 250 ml/min of CO<sub>2</sub> and 250 ml/min of N<sub>2</sub>. Some prominent  $\gamma$ -rays are labeled in the spectrum.

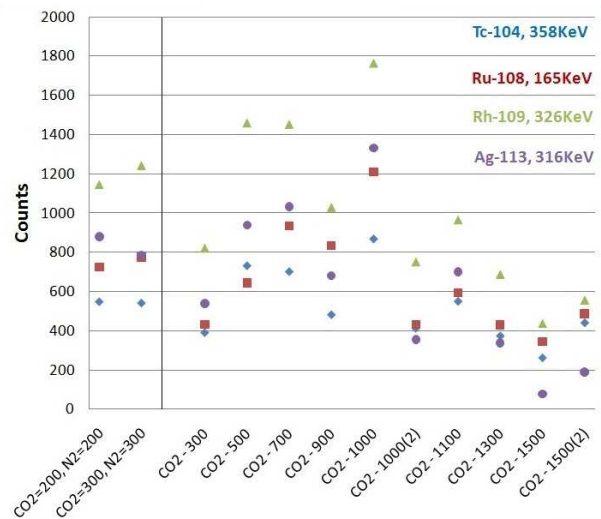


Figure 2. Relative yield of different fission products (Tc-104, Ru-108, Rh-109 and Ag-113) as a function of the gas flow rate.

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## Acknowledgement

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# Gas pressure influence on average charges of heavy recoils in TASCA

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Several experimental works at gas-filled separators have been reported that lay the basis for a correct prediction of the average charges of heavy and superheavy ions [1-3]. These works resulted in differing semi-empirical parameterizations.

An interesting feature of heavy ion "charge-exchange" collisions is the so-called "density effect", which has been observed at the Dubna gas-filled recoil separator DGFRS [2]. However, such an influence of the gas pressure has never been included in any of the above-mentioned semi-empirical expressions. The effect was also observed at the gas-filled separator TASCA [4]. Here we report results for <sup>252,254</sup>No ions which were produced in the fusion-evaporation reactions <sup>48</sup>Ca+<sup>206,208</sup>Pb.

The experimental setup and the reactions were the same as in [4]. Average charges and magnetic rigidities of No ions were deduced using the experimental distribution of evaporation residues in a focal plane detector [4]. The analysis method was the same as in [3].

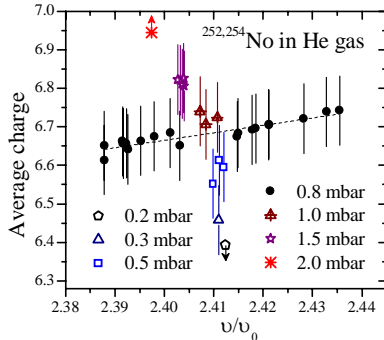


Figure 1: Measured average charges of the <sup>252,254</sup>No depending on their velocity (in units of the Bohr velocity). The line shows a linear fit. Arrows are marking limits for average charges.

Deduced average charges as a function of the velocity (expressed in units of the Bohr velocity,  $v_0$ ) are shown in Fig. 1. Average charges measured at 0.8 mbar gas pressure for both No ions (black solid dots) exhibit the linear dependence of the average charges on velocity. However, measured average charges at other pressures deviate significantly from this line. Deduced magnetic rigidities are shown in Fig. 2 for <sup>252,254</sup>No ions in helium (He) as well

\* These studies were financially supported the GSI F&E program and by the Research Center "Elementary Forces and Mathematical Foundations" (EMG).

as for <sup>254</sup>No in hydrogen (H<sub>2</sub>). These data show an increase of magnetic rigidities with decreasing pressure for both filling gases.

An expression to estimate the "density effect" has been given in [5] and we use it as a fit function for our experimental data. Average charges of heavy ions in a gas with pressure  $P$  can be presented as a sum of the equilibrated average charge and a correction term due to the "density effect",  $q_{ion} = \langle q \rangle + \Delta q$  [5]. Charge correction can be determined as  $\Delta q = a/(b+y)$  with  $y = [(v/v_0)P]^f$  in according to [5]. The function  $(B\rho)_{ion} = (c+dy)/(f+y)$  can be used to fit measured magnetic rigidities, with  $c, d$  and  $f$  being parameters.

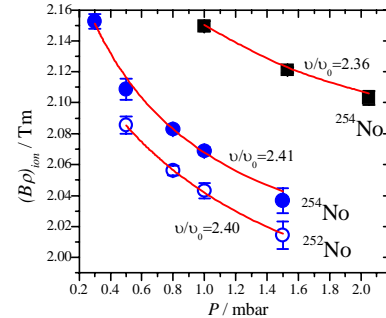


Figure 2: Magnetic rigidities of the <sup>252,254</sup>No ions as a function of the pressure. Circles and squares are denoting magnetic rigidities in He and H<sub>2</sub>, respectively. Lines show fits to the data according to the relations given in the text.

Results of fits are shown in Fig. 2. The behavior of the fitted curves for both <sup>252,254</sup>No ions seems to be similar, as can be expected for ions with an identical atomic shell structure. The linear fit function  $(B\rho)_{ion} = c+dy$  was used in the case of H<sub>2</sub> due to the limited number of experimental data points. These semi-empirical expressions which describe the "density effect" can be used as a correction for predicted average charges when using expressions from [1-3]. More detailed information will be provided in [6].

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## Average charges of heavy ions in a gas mixture

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At present, gas-filled separators are typically filled with either pure helium (He) or pure hydrogen (H<sub>2</sub>). Hydrogen seems to provide better suppression of background related to target-like ions [1]. However, as the average charges are lower than in pure He, a stronger dipole magnet is needed to separate evaporation residues. Therefore, at the gas-filled separator TASCA, experiments were performed with a fill-gas mixture of these two gases to investigate whether this allows for a combination of the advantages of the two gases, i.e., which allows a good suppression of the background while still keeping rather high average charges of evaporation residues. However, no data exists how to predict the average charges of heavy ions in gas mixtures. Thus, we aimed to study systematically average charges of heavy ions.

Earlier studies at TASCA clearly showed that the average charge is a function of the gas pressure [1]. This so called “density effect” was investigated on <sup>252,254</sup>No ions and a corresponding semi-empirical expression for the determination of the average charges was given in [1]. These expressions were used for the prediction of pressure dependent average charges of No ions in pure He and H<sub>2</sub>. The experimental setup and the nuclear reactions used in the studies reported here are the same as in [1]. Magnetic rigidities were deduced as described in [2].

Measured magnetic rigidities of <sup>254</sup>No ions in the mixtures at certain relative amounts of the gases  $v$  (He/H<sub>2</sub>), are shown in Fig. 1 as a function of the pressure. Clearly, the “density effect” is observed also in the mixtures.

Charge equilibration of heavy ions moving in the gas is determined by a system of coupled homogeneous linear equations for fractions of each charge state and cross-sections of “charge-exchange” collisions (see [3] for details).

Let us assume that the charge equilibration is also occurring in gas mixtures, and that heavy ions (with initial charge state fractions of  $F_i$ ) after the “charge-exchange” collisions have fractions  $F_i^{mix}$  ( $\sum F_i^{mix} = 1$ ) for each  $i$ -th charge state. Each heavy ion with an  $i$ -th charge state has a probability  $P_{He} = v/(v+1)$  and  $P_{H_2} = 1/(v+1)$  to collide with either a He or a H<sub>2</sub> atom, respectively, with  $P_{He} + P_{H_2} = 1$ . Then, the fraction of each  $i$ -th charge state in gas mixtures can be written as:  $F_i^{mix} = F_i \cdot P_{He} + F_i \cdot P_{H_2}$ . Average equilibrated charges in the gas mixtures can be derived as:

\* These studies were financially supported the GSI F&E program and by the Research Center “Elementary Forces and Mathematical Foundations” (EMG).

$$\bar{q}^{mix} = \sum q_i \cdot F_i^{mix} = \frac{\bar{q}^{He} \cdot v + \bar{q}^{H_2}}{v + 1}$$

where,  $\bar{q}^{He}$  and  $\bar{q}^{H_2}$  are average charges of <sup>254</sup>No ions in pure He and H<sub>2</sub> at the same pressure as the gas mixture, respectively. Corresponding magnetic rigidities can be put into the following expression:

$$(B\rho)^{mix} = \frac{(B\rho)^{He} \cdot (B\rho)^{H_2} \cdot (1+v)}{(B\rho)^{He} \cdot (v/v_0)_{H_2} + (B\rho)^{H_2} \cdot (v/v_0)_{He} \cdot v} \cdot (v/v_0)_{mix}$$

where  $(v/v_0)$  with different indices are the velocities of the <sup>254</sup>No ions in different gases (He and H<sub>2</sub> cases are given in [2]). In our experiments:  $(v/v_0)_{mix} = 2.39 \pm 0.03$ . Predicted magnetic rigidities from the above expression with  $(v/v_0)_{mix} = 2.39$  are shown in Fig. 1 by open dots. Experimental magnetic rigidities are well reproduced for  $v=1$  and 2, except for some underestimations at 0.8 mbar. Slightly underestimated magnetic rigidities were also observed in the case of  $v=3$  and 4. Again, in these cases gas pressures were 0.8 mbar. This discrepancy could be due to the linear function which was used to fit the observed “density effect” within the region (1-2) mbar in the case of pure H<sub>2</sub> [2]. More detailed information will be given in [4].

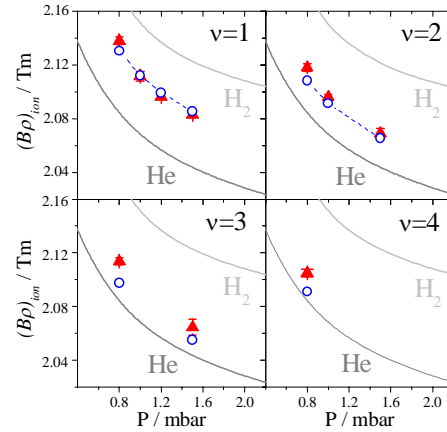


Figure 1: Magnetic rigidities of <sup>254</sup>No ions in gas mixtures  $v$  depending on gas pressures  $P$ . Triangles and open circles are showing the measured and predicted magnetic rigidities, respectively. Lines are showing the fitted results for pure He and H<sub>2</sub> gases [1].

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# Pulse Shape Analysis for the TASISpec Implantation Detector\*

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In TASISpec – TASCA in Small Image Mode Spectroscopy setup, which aims at decay spectroscopy of super-heavy elements [1] – heavy ions are implanted into a double-sided silicon strip detector (DSSSD). Their subsequent decays are recorded in this, as well as in the surrounding silicon and germanium detectors. The use of sampling ADCs in the experimental setup open up for new possibilities, such as particle identification, as they allow for investigations of the actual pulse shapes from the detectors. Pulses from the DSSSD were integrated in charge-sensitive preamplifiers [2] and studied by splitting the signals between the standard electronics read-out chain and sampling ADCs (CAEN V1724) as shown in Fig. 1. The sampling ADCs digitise the pulses at a rate of 100 MHz. For each event, a time span of  $2.56 \mu\text{s}$  around the arrival of the pulse was recorded and analysed offline. Triggers from the standard electronics were used. The accumulated data originates from a 3-line  $\alpha$  source and an in-beam experiment in which the DSSSD was irradiated with heavy ions using the reaction  $^{207}\text{Pb}(^{48}\text{Ca}, 2n)^{253}\text{No}$ .

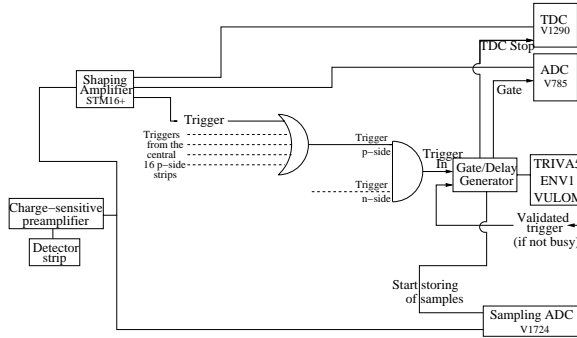


Figure 1: Electronics scheme for the DSSSD of TASISpec.

Any particle information present in the pulse shape resides within the rise of the pulse. The pulses were differentiated in order to emphasize this region of interest. Figure 2 shows the summed derivatives of pulses originating from the p-side (implantation side) of one pixel of the DSSSD; from  $\alpha$  particles (red) and from implanted heavy ions (black). A difference between the pulse shapes appears in the “tail” of the derivative, which corresponds to the top of the original signal. This discrepancy was characterised by calculating the ratio of the integral over the particle-dependent area, and the integral of the main peak in the derivative. The regions used are marked in the figure.

\* This work was supported by the Research Center “Elementary Forces and Mathematical Foundations”.

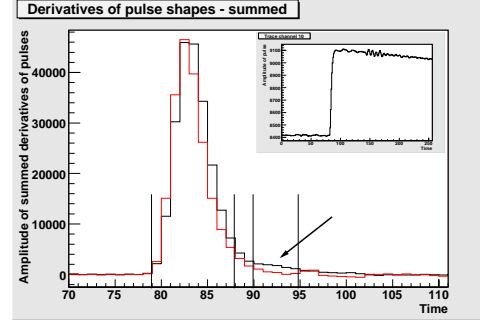


Figure 2: Summed derivatives of pulses from  $\alpha$  particles (red) and implanted heavy ions (black). Insert: Typical pulse shape before software treatment.

The ratio between the two integrals was calculated for all pulses from the pixel, and the resulting distributions are shown in Fig. 3. The distribution from heavy ions is clearly shifted to the right compared to the one from  $\alpha$  particles. Other pixels that were investigated show the same tendencies. This testifies that particle information is indeed present in the pulse shapes.

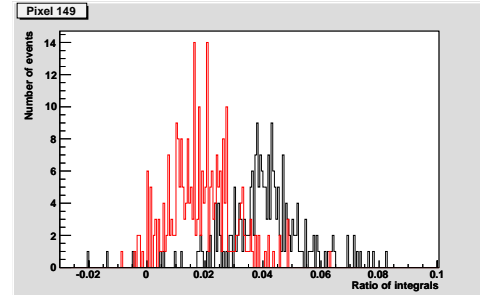


Figure 3: The ratio between the integrals for  $\alpha$  particles (red) and implanted heavy ions (black).

In order to use the existing differences for a separation into  $\alpha$  particles and implanted heavy ions, each strip, and possibly also every pixel, must be analysed individually in order to optimise the parameters used for the characterisation of the differences. This issue will be addressed in future analyses of pulse shapes from the TASISpec implantation detector.

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# Ion-optical simulations for the Inelastic Reaction Isotope Separator IRiS

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## Introduction

An impressive advancement in investigation of superheavy elements (SHE) was achieved in the past 25 years. The heaviest currently reported superheavy element contains 118 protons and novel challenging experiments pursuing the synthesis of elements 119 and 120 are under preparation. Yet all of these heaviest currently claimed elements were synthesized in nuclear fusion reactions, which can yield only neutron deficient products. Neutron-rich isotopes of the heaviest elements, which are of special interest e.g. in the context of nuclear chemistry and nuclear astrophysics, cannot be produced this way. The only viable production mechanism for neutron-rich nuclides is through multi-nucleon transfer reactions (MNTR), the application of which will give access to tens of new neutron-rich isotopes of the heaviest elements. Currently available separators are optimized for fusion products emitted under 0° and are poorly suited for MNTR studies due to their limited angular acceptance.

A new Inelastic Reaction Isotope Separator (IRiS) [1], dedicated to the investigation of neutron-rich isotopes of the heaviest elements produced in MNTR, will be constructed and set-up at the GSI in a joint effort of an international collaboration, headed by the Johannes Gutenberg University Mainz, the Helmholtz Institute Mainz, and the GSI Helmholtzzentrum für Schwerionenforschung Darmstadt. The main design goal of the IRiS is the ability to separate the heavy products formed in MNTR and deliver them to a focal plane. Here ion implantation and decay is detected in focal plane detector. To perform (i) chemical studies, (ii) mass measurements, and (iii) nuclear and atomic spectroscopy of the heavy ions separated in the IRiS, the detector setup will be retracted and allow the separated products to enter a gas cell, where they will be stopped and extracted for further investigation. A conceptual design scheme is shown in Fig. 1.

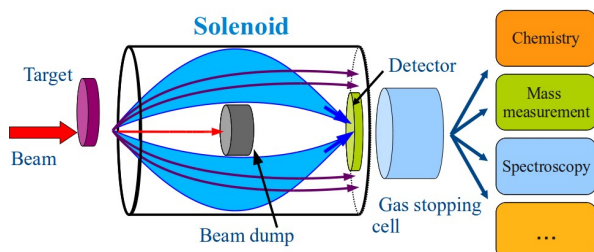


Figure 1: Schematic drawing of the IRiS design concept. Products are separated in a gas-filled solenoid magnet according to their magnetic rigidity. A beam dump stops the beam particles as well as 0° products.

## Simulations

An essential part of the IRiS design process is the development of a computer simulation for the IRiS ion-optics. Due to the availability of theoretical predictions, the following two nuclear reactions were chosen as input for the simulation:  $^{48}\text{Ca}+^{248}\text{Cm}$  at  $E_{\text{CM}}=209$  MeV [2], and  $^{238}\text{U}+^{248}\text{Cm}$  at  $E_{\text{CM}}=750$  MeV [3]. The simulation code using the ROOT [4] framework is already highly advanced and includes: (i) Simulation of unreacted projectiles and products of elastic and inelastic scattering including MNTR; (ii) energy loss and straggling in the target; (iii) ion interaction with gas molecules and (iv) ion tracking in a realistic solenoidal magnetic field.

## Simulation results

Simulations showed that relatively strong magnetic fields are necessary to achieve acceptable separation in a gas-filled solenoid. While several solenoid dimensions and magnetic field strengths were successfully tested, a stored energy of about 10 MJ was necessary in all cases. In symmetric ion-optical geometries, as shown in Fig. 2, efficiencies of typically about 20% for collecting the heaviest products ( $Z \geq 102$ ) in the detector area were reached, while the detector count-rate due to background was estimated to about few kHz. Although not optimal for the identification of products in a focal plane detector, these conditions are well suited for use of a gas cell.

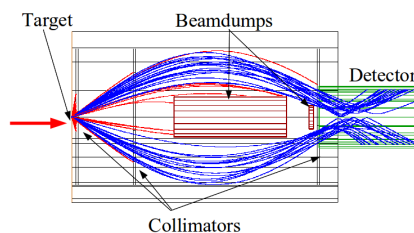


Figure 2: Trajectories of the heaviest products ( $Z \geq 102$ ) produced in the reaction  $^{48}\text{Ca} + ^{248}\text{Cm}$ .

## Acknowledgements

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# Mass Measurements of No and Lr isotopes with SHIPTRAP\*

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High-precision mass measurements of radionuclides provide information about their binding energy and are thus a powerful method to study their nuclear structure and to benchmark nuclear models. The region of superheavy elements that owe their existence to nuclear shell effects is of particular interest. However, these nuclides can only be produced in fusion-evaporation reactions at rather low rates of a few particles per second or less. Therefore, information about their masses was so far exclusively available from the observation of their decay. Recently, the first direct measurements on transuranium nuclides, three nobelium ( $Z = 102$ ) isotopes, have been performed with SHIPTRAP [1]. The obtained accurate mass values provide anchor points to fix  $\alpha$ -decay chains as demonstrated for the nobelium isotopes  $^{252-254}\text{No}$  [2]. This is especially important for odd-odd and odd- $A$  nuclides where the  $\alpha$  decay typically populates excited states complicating an unambiguous mass determination.

SHIPTRAP receives radionuclides after separation from the primary beam in the velocity filter SHIP. It is presently the only Penning trap for high-precision mass measurements of elements above fermium. Recently, the masses of the nuclides  $^{255}\text{No}$  and  $^{255,256}\text{Lr}$  ( $Z = 103$ ) produced in the reactions  $^{208}\text{Pb}(^{48}\text{Ca},n)^{255}\text{No}$  and  $^{209}\text{Bi}(^{48}\text{Ca},xn)^{257-x}\text{Lr}$  have been measured. A primary beam energy of 4.55 AMeV was chosen resulting in production cross sections of about 50–200 nb. The corresponding production rate was as low as about one particle per minute entering the SHIPTRAP gas cell in the case of the nuclide  $^{256}\text{Lr}$ , the lowest yield for which a Penning trap mass measurement was performed to date. The reaction products were decelerated in degrader foils and stopped in a gas cell in 50 mbar helium. The nuclides were extracted from the gas cell mainly as doubly charged ions. After cooling and accumulation in a radiofrequency quadrupole ion-beam cooler short bunches were injected into a double-Penning trap system inside a  $B = 7$  T magnet. The mass was determined by measuring the cyclotron frequency  $\nu_c = qB/(2\pi m)$  of the ions using a time-of-flight ion-cyclotron-resonance detection technique.

An example of a resonance for  $^{255}\text{No}$  is shown in Fig. 1. An important development enabling these measurements

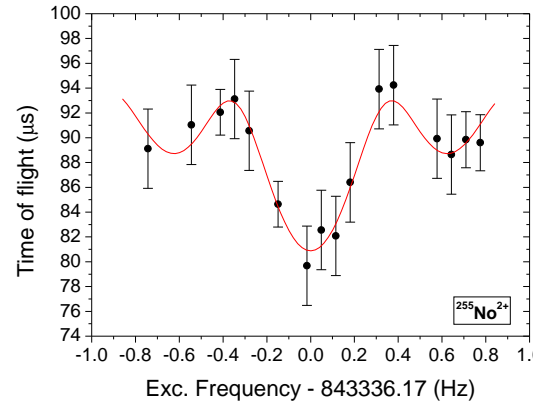


Figure 1: Cyclotron resonance of  $^{255}\text{No}^{2+}$ . The solid line is a fit of the theoretical line shape to the data points.

was the implementation of active regulation systems controlling the pressure in the helium cryostat and the temperature in the bore of the superconducting solenoid [3]. In this way fluctuations of the magnetic field are reduced substantially and the time interval between successive calibration measurements can be increased. This is crucial for measurements of rare isotopes with low yield such as  $^{256}\text{Lr}$  where a single resonance was recorded over a period of about 48 hours. Prior to our measurements the masses of both Lr isotopes listed in the Atomic-Mass Evaluation 2003 were only estimated from systematic trends. Now these isotopes have been established as new anchor points above uranium. In addition, the chain of neighboring nobelium isotopes whose masses have been measured directly has been extended across neutron number  $N = 152$ . This allows, for example, studying the neutron shell gap via the three-point difference of binding energies  $\Delta^{(3)}(N) = (-1)^N/2[B(N+1) + B(N-1) - 2B(N)]$ .

The SHIPTRAP measurements on nobelium and lawrencium are important steps towards direct mass measurements of superheavy nuclides in the near future.

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