SUPERHEAVY ELEMENT RESEARCH AT THE VELOCITY FILTER SHIP

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The Separator for Heavy Ion Reaction Products (SHIP) is a velocity filter located at the UNILAC accelerator of GSI Darmstadt, Germany. For about 35 years a broad experimental program in the field of superheavy element research is running at SHIP. During the last years particularly investigations in the region of the heaviest known nuclei were performed. In fusion reactions of ^{48}Ca + ^{248}Cm → ^{296}116* a total of six decay chains was observed which could be attributed to the evaporation residues ^{292}116 and ^{293}116. In this experiment, data measured previously on the same isotopes in Dubna were well confirmed. Besides, two attempts were made to synthesize isotopes of the still unobserved element Z = 120 in reactions of ^{64}Ni + ^{238}U and ^{54}Cr + ^{248}Cm. No events were observed in these experiments leading to one-event cross-section limits of 90 fb and 560 fb, respectively. For future superheavy element research, a new superconducting continuous wave LINAC is planned at GSI which shall deliver beam intensities of up to 10^{14} particles per second. In this context we are developing a next generation separator and new detection techniques.

1. Introduction

Until the year 2005, so-called cold fusion reactions using doubly magic Pb or Bi targets were applied at SHIP for the synthesis of superheavy elements and lead to the discovery of the elements Z = 107 - 112 [1 - 6]. Starting in 2005, hot fusion reactions with actinide targets (^{238}U, ^{248}Cm) were introduced at SHIP. This was motivated by experimental results from the Dubna gas-filled separator (DGFRS) [7] at the Flerov Laboratory of Nuclear Reactions where relatively large cross-sections on the order of 1 pb were observed for nuclei with Z = 114 - 118 in hot fusion [8]. In contrast, the cross-sections in cold fusion reactions drop by about one order of magnitude for every two protons more in the compound nucleus and become already as low as 30 fb for Z = 113 [9]. The application of hot fusion reactions at SHIP is an important step not only for the synthesis of new elements with Z > 118 but equally for the confirmation of existing data from hot fusion on $112 \leq Z \leq 118$. The latter have been synthesized until recently only at gas-filled separators, mainly in Dubna. SHIP is presently the only velocity filter applied for the synthesis of the heaviest elements with exception of the energy filter VASSILISSA [10] in Dubna. Therefore, the cross-sections and decay properties of superheavy nuclei measured at SHIP represent an important cross-check of the existing data with respect to possible systematic errors. Additionally, SHIP allows the identification of the reaction channel (xn, αxn, transfer etc.) with good resolution by measuring the velocity spectra of the reaction products [11]. The knowledge of the reaction channel is crucial for assigning an observed decay chain to the correct mother isotope since the mass and nuclear charge of the superheavy nuclei are not measured directly.

The production cross-sections of superheavy nuclei are small and reach the sub-picobarn region for the heaviest known isotopes. This presently limits the synthesis of new elements with the available beam intensities of $5 \cdot 10^{12}$ ions per second. Reaction cross-sections on the order of 100 fb require several months of beam time to reach the one-event cross-section limit for the desired isotope. Therefore, the availability of higher beam intensities and the related necessary upgrades of the separation and detection techniques are decisive steps for the future research in this field. At GSI, a new superconducting continuous wave LINAC is planned. The beam intensities are expected to be 10 to 50 times larger than the presently available currents. Within this frame, we are developing a concept for a new separator which allows a strong suppression of primary beams with intensities up to several 10^{14} particles per second. In parallel to the separator design, we are testing and developing new detection techniques for nuclei which are long-lived and/or undergo beta-decay or spontaneous fission and are therefore not accessible with the available techniques.

2. SHIP: Experimental Setup

A scheme of the experimental setup is shown in Fig. 1. SHIP is a Wien filter which separates the ions according to their velocities [12]. Reaction products which leave the target at forward angles of (0 ± 2) degree with respect to the beam direction are accepted by the entrance aperture of SHIP. The electric and magnetic fields are chosen such that the relatively light and fast projectiles and projectile-like reaction products are deflected to the beam stop while the much slower evaporation residues pass SHIP and reach the focal plane detector. The accepted velocity window at a given setting is $\Delta v/v = 0.1$ (FWHM). All reaction products which pass the velocity filter are implanted in a position sensitive 16 strip silicon detector (”stop detector”) where their time of implantation, position, kinetic energy and radioactive decays are registered [13]. Such, especially the alpha decay properties allow for an unambiguous identification of single isotopes. Six further Si detectors are installed in a box-like arrangement (“box detector”) in front of the stop detector and cover 85 % of the backward hemisphere in order to register alpha particles and fission fragments escaping from the stop detector.

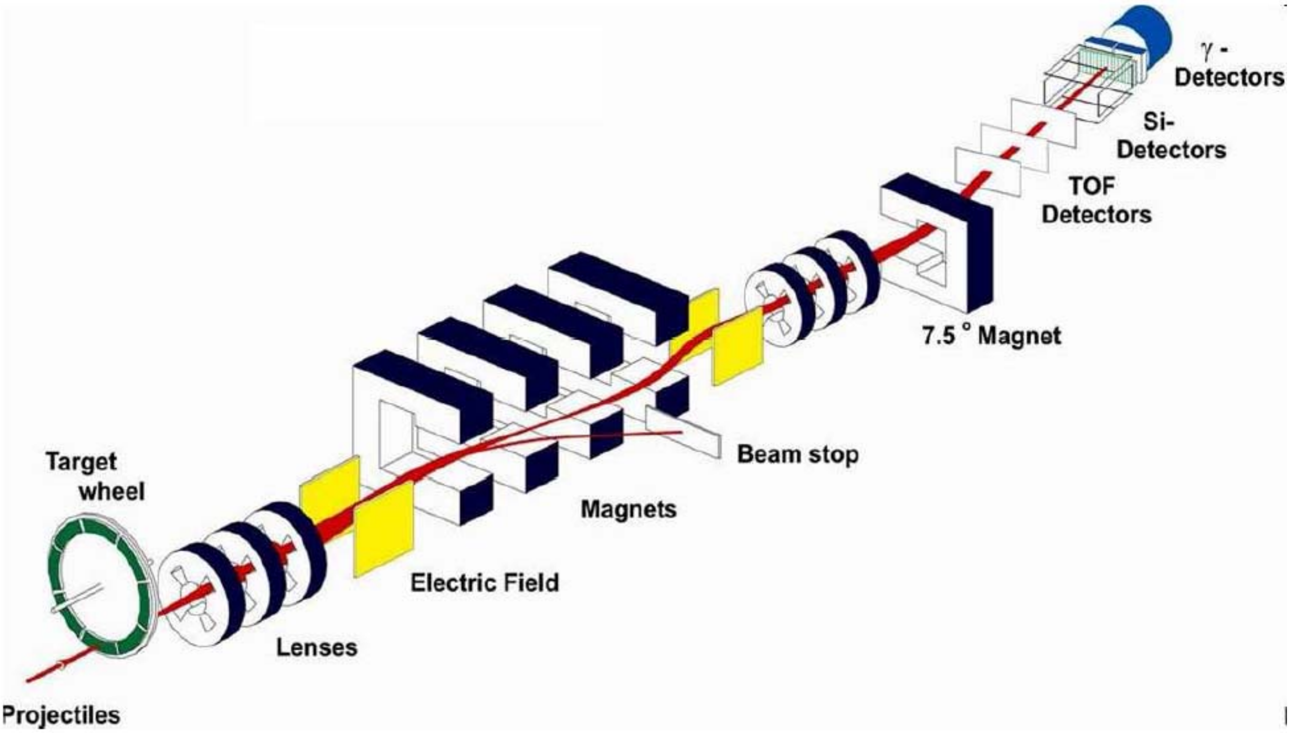

Fig. 1 The velocity filter SHIP and detection system. For details see text.

Finally, a germanium clover detector is mounted behind the stop detector. It consists of four germanium crystals which register gamma rays from excited reaction products implanted in the stop detector. Three time-of-flight (TOF) detectors [14] are installed in front of the silicon detectors. They fulfill two tasks: On one hand the measured TOF together with the energy deposited in the silicon detector allows to distinguish between projectile-like nuclei, target-like nuclei and fusion evaporation residues. Secondly, they allow to distinguish between ions which have been produced in the target and radioactive decay products (alpha particles or fission fragments) which have been created in the silicon detector by decays of implanted mother isotopes. In the first case the particles have to pass the TOF detectors and create a TOF signal while in the latter case they don't. Further details concerning the experimental setup can be found in [15].

The combination of isotope identification via radioactive decays and the strong background suppression by the separator allows the identification of single nuclei where cross-section limits of 10 pb can be reached within one day of beam time by applying usual beam intensities of several 10^{12} particles per second. For additional suppression of alpha-like and fission-like background events the beam-off periods are used which are provided by the pulsed structure of the beam consisting of 5 ms long beam-on periods followed by 15 ms long beam-off periods.

3. Hot fusion reactions at SHIP

3.1 The reaction ^{48}Ca + ^{248}Cm → ^{296}116*

The nuclei with the largest proton numbers synthesized so far at SHIP, and also at GSI, are isotopes of element Z = 116 in reactions of ^{48}Ca + ^{248}Cm → ^{296}116* [16] (June 25 - July 26, 2010). The same reaction was already studied earlier at the Dubna gas-filled separator in several experiments during the years 2000 to 2004 at compound nucleus excitation energies of 33 and 39 MeV [17, 18]. In the Dubna experiments five decay chains were observed which were attributed to the decay of the isotope ^{293}116 (3n evaporation channel) and six decay chains which were attributed to ^{292}116 (4n channel). At SHIP, we continued the excitation function to higher energies of 41 and 45 MeV. At 41 MeV we observed six alpha decay chains of different lengths, all of them terminated by a spontaneously fissioning nucleus (Fig. 2). Four of the chains consist of an implanted recoil nucleus followed by two alpha decays and a fission event. The length of these chains as well as the half-lives and alpha energies of the chain members are well in agreement with the data measured in Dubna for the isotope ^{292}116. Also the corresponding cross-section of 3.4 pb well continues the excitation function measured in Dubna as shown in Fig. 3. Fig. 3 shows also the expected excitation functions from model calculations [19]. The good agreement between experimental data and theoretical expectations further supports the attribution of the four decay chains to the mother isotope ^{292}116.

Besides, two further chains were observed. One of them consists of an implanted recoil nucleus followed by three α-decays and fission decay. This decay sequence, the α-energies and half-lives are consistent with the data measured in Dubna for the isotope ^{293}116 (3n channel). Also the related cross-section of 0.9 pb agrees well with these data and with theoretical predictions (Fig. 3). In the other decay chain the recoil nucleus was followed by four α-decays and a fission event. In this chain only the energy of the first α-particle was in agreement with the values measured in Dubna for the nucleus $^{293}116$. Therefore, we attributed this chain preliminary also to the mother isotope $^{293}116$. The last α-decay in the chain, with the energy of 9.315 MeV would then originate from an alpha branch of the isotope ^{281}Ds. An α-decay of ^{281}Ds was so far only observed in an experiment at the gas-filled separator TASCA at GSI [20], however with an energy of 8.727 MeV which is 588 keV less than the energy observed at SHIP. The half-lives of the four α-decays observed at SHIP and attributed to $^{293}116$, $^{289}114$, ^{285}Cn and ^{281}Ds are in agreement with the literature values within statistical fluctuations. But the α-decay energies of the daughter nuclei $^{289}114$ and ^{285}Cn are 211 keV and 523 keV larger than the literature values.

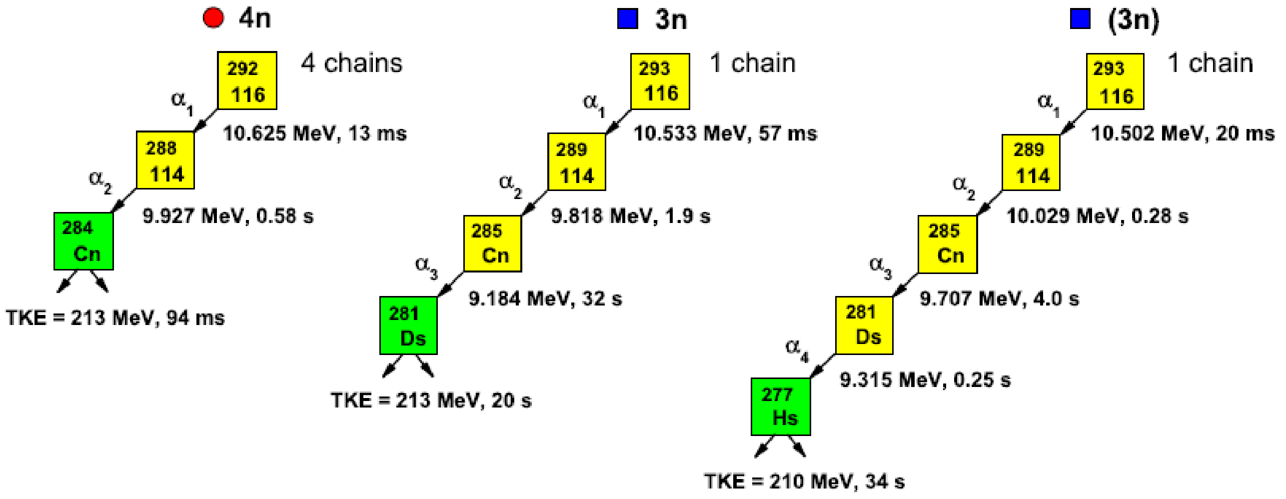

Fig. 2 Decay chains observed at SHIP in fusion reactions of ^{48}Ca + ^{248}Cm leading to the compound nucleus $^{296}116^*$ at an excitation energy of 41 MeV. The given half-lives and alpha decay energies include all available data from different experiments. The decay sequence (α–α–α–α–fission) was so far only observed at SHIP and preliminary attributed to the 3n evaporation channel. For this case the half-lives and decay energies represent the values measured at SHIP.

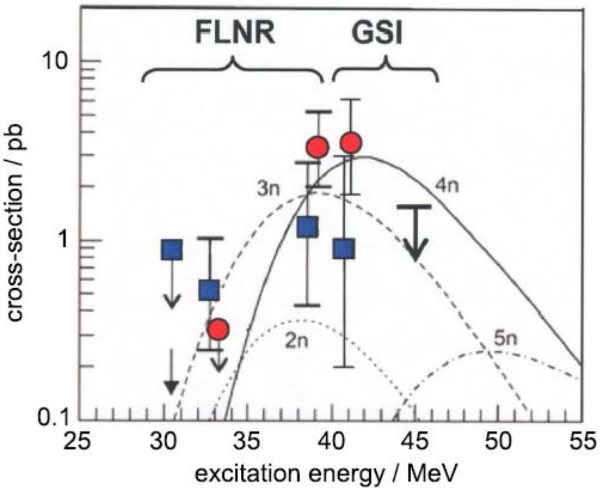

Fig. 3. Measured cross-sections for the 3n (squares) and 4n (circles) evaporation channels of the reaction ^{48}Ca + ^{248}Cm → $^{296}116^*$. The Figure shows both, the data measured in Dubna [17, 18] and at SHIP. Also shown are the calculated excitation functions for the 2n, 3n, 4n and 5n evaporation channels [19].

A possible explanation for the deviating α-energies observed at SHIP might be the population of isomeric states which are located rather close to the ground state. Theoretical calculations for the decay chain of $^{293}116$ [21] result in the existence of high and low spin states close to the ground state which would enable the existence of isomeric states. The observation of very similar half-lives for the decay chains from the hypothetical ground and isomeric states is not contradicting according to the calculations in [21] since they allow for the existence of decay chains from the ground as well as from isomeric states with similar angular momentum leading to similar half-lives. However, an assignment of the observed transitions by comparing experimental and theoretical data was not possible.

3.2 Search for Z = 120

The reaction ^{64}Ni + ^{238}U was studied at SHIP to search for evaporation residues of the compound nucleus $^{302}120^*$. This proton number is particularly interesting because in some theoretical models a spherical shell closure is expected there. The compound nucleus $^{302}120^*$ formed in fusion reactions of ^{64}Ni and ^{238}U has 182 neutrons. This would be the closest approach to the predicted closed neutron shell N = 184 reached so far in superheavy element experiments. Calculations within different models have been performed for this reaction which resulted in strongly different production cross-sections. The calculated values for the 4n channel are compared in the Table, column 1 for several models.

Expected cross-sections for the isotope $^{298}120$ according to different model calculations. The nuclei are produced in 4n evaporation channels from excited $^{302}120^*$ compound nuclei created in fusion reactions with different projectile-target combinations. References are given in the Table.

^{64}Ni + ^{238}U	^{58}Fe + ^{244}Pu	^{54}Cr + ^{248}Cm	ref.
3.2 fb	5.3 fb	25 fb	[22]
0.022 fb	1 fb	800 fb	[23]
0.02 fb	0.015 fb	0.07 fb	[24]
5 fb	32 fb	54 fb	[24]
< 90 fb [25]	< 400 fb [26]	< 560 fb [27]	experiment

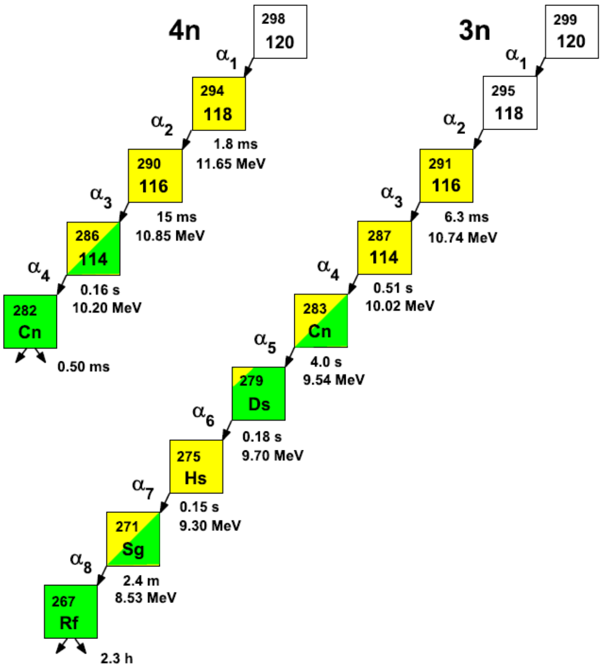

Fig. 4 Expected decay chains of the isotopes $^{298}120$ and $^{299}120$. The white squares represent still unobserved isotopes.

They differ by more than three orders of magnitude which reflect the high sensitivity of the cross-sections to the entrance and exit channel parameters like fusion barriers and shell correction energies which vary in the different models or have different impact, respectively. One has to note that all models named in the Table were able to reasonably reproduce the cross-sections for lighter superheavy nuclei measured in Dubna. The models underlying the values in the Table assumed fission barriers of about 7 MeV and a proton shell closure at Z = 114. However, if the fission barriers are actually higher and/or the shell closure is located at Z = 120, much higher cross-sections can be expected. Concerning the strength of the shell corrections the model predictions vary on a large scale resulting in values for the shell correction energies from -6 MeV to -12 MeV which corresponds to fission barriers of (6 - 12) MeV [28]. By trend, one can assume that the cross-section increases by one order of magnitude if the fission barrier increases by 1 MeV. This was the motivation to investigate this reaction. A total of 120 days of beam time was applied in 2007 and 2008. The beam energy was chosen such that the compound nuclei would be created with excitation energy of 36 MeV which should predominantly lead to the evaporation residues $^{298}120$ and $^{299}120$. From both isotopes long α-decay chains can be expected which join the already known decay chains of $^{294}118$ and $^{291}116$, respectively (Fig. 4). Finally, no event was observed which showed the expected signatures of the decay of a Z = 120 isotope. The upper cross-section limit reached in this experiment is 90 fb at a total number of $2.64 \cdot 10^{19}$ projectiles on target [25]. This indicates that the fission barriers are not higher than 8.3 MeV for the isotopes $^{298,299}120$.

Besides in fusion reactions of ^{64}Ni + ^{238}U, the same compound nucleus, $^{302}120^*$, can also be created in collisions of ^{58}Fe + ^{244}Pu and ^{54}Cr + ^{248}Cm. The respective calculated cross-sections for the 3n channel are listed in the Table, column 2 and 3. The reaction $^{58}\text{Fe} + ^{244}\text{Pu} \rightarrow ^{302}120^*$ was investigated in Dubna [26]. No event was observed which could be attributed to the decay of a Z = 120 isotope. The corresponding upper cross-section limit for the observation of one event was 400 fb.

At SHIP, the reaction ^{54}Cr + ^{248}Cm was investigated from April 23 to June 1st, 2011, with a total of $7.9 \cdot 10^{18}$ projectiles on target [27]. Also here no event was observed leading to an upper cross-section limit of 560 fb. The limit cross-sections reached so far in the experiments are still not low enough to meet the cross-sections predicted by most of the models. However, the experimental data allow the exclusion of extraordinary high fission barriers as predicted by some models. From the experimental cross-section limits one can deduce that the barriers do not significantly exceed the value of 8 MeV.

The expected half-lives for isotopes with Z = 120 deserve special attention. The Q-values for α-decay are increasing with the proton number which leads to a decrease in the half-lives. According to the macroscopic-microscopic model which assumes the shell closure at Z = 114 the Q-values are about 13 MeV leading to half-lives on the order of (1 – 10) μs for the isotopes $^{298,299}120$. This is already on the same range as the flight time of the fusion products through the separator. Therefore one has to keep in mind that the evaporation residues could already decay before the detector is reached. If, however, the shell closure is assumed at Z = 120 one can expect significantly longer half-lives on the order of 1 s. The related Q-values are around 11 MeV. Therefore, the observation of relatively long half-lives and α-decay energies around 11 MeV would be a strong hint for a shell closure at Z = 120.

4. Perspectives for the future of superheavy element research

The small production cross-sections of the heaviest nuclei presently limit the synthesis of new isotopes or elements, respectively. Presently available typical beam intensities are on the scale of $5 \cdot 10^{12}$ ions / s which leads to the situation that cross-sections on the order of 100 fb require already several months of beam time to reach the one-event cross-section limit for the desired isotope. Therefore, the availability of more intense ion beams and the related necessary upgrades of the separation and detection techniques are decisive steps for the future research in this field. This concerns equally, beside the synthesis of new isotopes, all experiments in the field of superheavy nuclei like spectroscopic and chemical investigations, precision mass measurements and nuclear reaction studies.

At GSI, a new superconducting continuous wave (cw) LINAC is planned which shall provide ion beams with intensities up to 10^{14} particles / s. Presently, a so-called demonstrator, consisting of a superconducting CH-cavity and two superconducting solenoids is built and shall be tested with beam in 2013 / 2014 at GSI [29]. In parallel, we are developing a new separator for the future superheavy element experimental program which will be adjusted to the requirements arising with the higher beam intensities, namely, it must be capable of strong background suppression to handle the 10 to 50 times higher beam intensities. The new separator will also be based on the concept of a velocity filter like the present SHIP following from the positive long-term experience with SHIP. Velocity filters provide several advantages in comparison to gas-filled separators: (i) the separation according to velocities is, for the typical reactions applied here, about five times stronger than the separation according to magnetic rigidity in gas. Therefore, velocity filters provide a stronger suppression especially of target-like quasi-elastic and deep inelastic background events; (ii) velocity filters allow the determination of the reaction channel in which a certain isotope was created by measuring the velocity spectra of the reaction products; (iii) due to the relatively strong separation of different reaction channels, velocity filters are also suitable for the study of transfer reactions with low cross-sections, especially at beam energies below and close to the Coulomb barrier. The study of transfer reactions is very interesting since in recent times new theoretical calculations suggest to produce new neutron-rich heavy and superheavy nuclei in multi-nucleon transfer reactions which are not accessible in other reactions (see e. g. [22, 30]).

For the new separator we plan a more compact design in comparison to SHIP. It will also be a two-stage separator but the condenser field shall be placed inside the magnetic dipole field while SHIP has separated electric and magnetic fields. To enhance especially the angular efficiency for transfer products, the acceptance angle will be increased by a factor of 2 to 3 with respect to the present acceptance of SHIP which is 10 msr.

In parallel to the separator design, we are testing and developing detection techniques for heavy long-lived nuclei and for nuclei which do not undergo α-decays and are therefore not accessible with the present technique of α-decay tagging. One possibility is the application of high-precision mass measurements. Penning traps or multiple reflection time-of-flight mass spectrometers have mass resolving powers up to $m/\Delta m \approx 10^7$ where, however, a resolving power of 10^5 is already sufficient for an isobaric separation of most of the isotopes. The mass measurement is performed after the separation stage. Penning traps and TOF spectrometers can only trap ions with low energies on the scale of (100 - 1000) eV therefore a buffer gas cell and an ion guide system has to be used after the separator for stopping, extracting and transporting the ions to the mass measurement device. The presently running gas cells which are applied for ions in the required mass and energy range have overall (i. e. stopping and extraction) efficiencies of ≈ 1 %. This leads to a rather strong loss of ions which presently requires production cross-sections of at least 10 nb. However, the new generation of cryogenic gas cells [31, 32] shows at least a factor of 10 more efficiency which has been demonstrated in the commissioning of such a cell at the Fragment Separator at GSI. Further, a newly developed multiple reflection time-of-flight mass spectrometer (MR-TOF-MS [33]) at the University of Gießen has promising features for the application as detection system since it allows for a broadband detection. This is especially for transfer products very efficient since a large number of them with rather different mass numbers can pass the separator at the same setting.

Besides, we are investigating the applicability of calorimetric low temperature detectors (bolometers) [34] for energy measurement of very heavy nuclei. These detectors register the increase of temperature when an ion deposits its energy in the detector. In this case, the usually occurring plasma effects which lead to a pulse height deficit can be circumvented. As a consequence, a considerably better energy resolution can be obtained than with the usually applied semiconductor detectors.

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