Transfer reaction studies in the region of heavy and superheavy nuclei at SHIP

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Abstract. We studied multi-nucleon transfer reactions in the region of heavy and superheavy nuclei. The goal was to investigate these reactions as possibility to create new superheavy neutron-rich isotopes, which cannot be produced in fusion reactions. The experiments have been performed at the velocity filter SHIP at GSI. At SHIP we can detect and identify the heavy, target-like, transfer products. Due to the low background at the focal plane detector and the isotope identification via radioactive decays, the setup allows to reach an upper cross-section limit of 10 pb/sr within one day of beamtime. We investigated the systems ⁵⁸,⁶⁴Ni + ²⁰⁷Pb and ⁴⁸Ca + ²⁴⁸Cm at beam energies below and up to 20% above the Coulomb barrier. At all energies we observed a massive transfer of protons and neutrons, where transfer products with up to eight neutrons more than the target nucleus could be identified.

1. Introduction
Isotopes in the region of superheavy nuclei with Z > 100 are usually produced in fusion-evaporation reactions applying Pb and Bi targets (cold fusion) or actinide targets (hot fusion) [1, 2]. Several isotopes with Z ≤ 101 have also been made in nucleon transfer reactions using heavy and intermediate heavy beams on actinide targets like ²³⁸U + ²⁴⁸Cm or ⁴⁸Ca + ²⁴⁸Cm [3, 4]. By comparing the same reaction product, fusion reactions lead usually to (10 – 50) times higher yields than transfer reactions. However, the isotopes resulting from fusion-evaporation reactions are located in the rather neutron-deficient region of the nuclear chart. This is due to the limited number of stable isotopes which can be applied as beams and target materials. Therefore, other reactions than fusion have to be considered in order to extent the nuclear chart towards more neutron-rich nuclei. In this context, multi-nucleon transfer reactions, which have been already extensively studied in the late 1970s up to the 1990s are brought into discussion again. Especially from theoretical side many contributions have been published in recent years concerning the creation of superheavy nuclei in transfer reactions with medium-heavy and very heavy beams [5, 6], while new experimental data in the region around Z = 100 and beyond are not available. Concerning the experiments performed some decades ago only those which applied chemical methods for particle identification were able to distinguish isotopes up to Z = 101 [3, 4]. These isotopes were still located within the region of nuclei which can also be
reached in fusion reactions. In collisions of $^{238}\text{U} + ^{248}\text{Cm}$ numerous isotopes have been observed, where the heaviest one was $Z = 101, N = 157$ [3]. These data are compared with model calculations for the same system at a centre-of-mass energy of $E_{cm} = 800$ MeV ($6.6\times A$ MeV) [6] in figure 1. The calculations are represented by the solid lines, while experimental data are represented by the different symbols. The calculations reveal a pronounced shoulder in the yield distributions at $Z = 106$ and a steep drop in the yield for isotopes with $Z > 106$. The shoulder appears due to shell effects which govern the break-up of the di-nuclear system (DNS) $\text{U} + \text{Cm}$ after nucleon exchange. The strong shell closures at $Z = 82$ and $N = 126$ in $^{208}\text{Pb}$ lead to a pronounced minimum in the potential energy surface and to a preferred production of isotopes close to $^{208}\text{Pb}$ in the transfer reactions. In this case the heavier transfer product is located close to $^{256}\text{Sg}$ ($Z = 106, N = 172$). For proton numbers of $Z < 82$ and $Z > 106$, respectively, the potential increases steeply, which leads to the steep drop in the yields for isotopes with $Z < 82$ and $Z > 106$. For comparison, the dashed line indicates the cross-sections, which are expected according to the diffusion model, without assuming the influence of the $^{208}\text{Pb}$ shells. In this case, the cross-sections drop exponentially with increasing number of transferred protons. The data in figure 1 reveal that the maxima of the experimental distributions rather follow the yields expected from the diffusion model. This is especially visible for the distributions for $Z = 98, 99$.

Experimental data from medium heavy beams on heavy targets are available from collisions of $^{40,44,48}\text{Ca}$ with $^{248}\text{Cm}$ targets [4]. Here, the heaviest measured isotope, $Z = 100, N = 156$, was observed with the most neutron rich beam, $^{48}\text{Ca}$, with a total cross-section of $\approx 1$ µb. By comparing the experimental data for the isotopes up to $Z = 101$ for medium heavy and very heavy projectiles, similar cross-sections were measured with $^{40}\text{Ca}$ and $^{238}\text{U}$ beams. From the technical point of view intermediate heavy beams are more favourable, because they are available at about 100 times higher intensities than the heavy beams like uranium. This is an important point regarding the expected low cross-sections of nanobarn and below for nuclei with $Z > 100$. 

Figure 1. Calculated (solid lines) [6] and measured (symbols) [3] cross-sections for transfer products from $^{238}\text{U} + ^{248}\text{Cm}$ collisions. The dashed line represents the expected yields assuming a diffusion process.
At the velocity filter SHIP we started the investigation of transfer reactions in heavy systems, applying very sensitive detection techniques, which are also used for the identification of superheavy nuclei produced in fusion evaporation reactions. The identification of the isotopes is performed via their radioactive alpha-, beta- or spontaneous fission (SF) decays. This method together with the low background on the detection system allows to reach the cross-section limit of 10 pb/sr for a certain isotope, within one day of beam time. In a first experiment we investigated transfer reactions with $^{207}$Pb targets applying $^{58,64}$Ni beams. The application of lead targets leads to the population of a broad region of alpha emitters with proton numbers between $Z = 82$ and $Z = 92$. This allowed us to investigate the transfer from the projectile to the target nucleus in the region of heavy nuclei and its dependence on the projectile neutron number. Further, we could investigate the applied detection techniques and their limitations. In a follow-up experiment we studied transfer reactions using $^{248}$Cm targets ($Z = 96$) and $^{48}$Ca beams, where we explored the population of isotopes in the region around $Z = 100$. All experiments were performed at rather low beam energies, starting from about 10% below the Coulomb barrier up to about 20% above the barrier. At these low energies the (primary) transfer products are created at low excitation energies. This reduces the losses due to fission of the primary nuclei, which is an important point for the heavy nuclei around $Z = 100$, which have low fission barriers. Moreover, for the primary isotopes surviving sequential fission low excitation energies reduce the number of evaporated neutrons and lead to more neutron rich secondary nuclei.

2. Experimental setup and method

A sketch of the experimental setup is shown in figure 2. The beams of $^{48}$Ca and $^{58,64}$Ni were delivered by the UNILAC accelerator of GSI. The UNILAC accelerator delivers beams with a pulsed structure. The 5 ms long beam-on periods are followed by 15 ms long beam-off periods. Eight targets were mounted on a rotating target wheel, which is installed at a distance of $\approx 500$ mm in front of the first quadrupole triplet of SHIP. Reaction products which leave the target at forward angles of $(0 \pm 2)$ degree with respect to the beam direction are accepted by the entrance aperture of SHIP. The respective fraction of the solid angle is $\Delta \Omega = 10$ msr. The electric and magnetic fields are chosen such that the relatively light projectiles and projectile-like reaction products, which have about ten times larger velocities than the transfer products, are deflected to the beam stop. Only the slow ($v \approx 0.01c$) target-like nuclei can pass SHIP and reach the focal plane detector. All reaction products which pass the velocity filter are implanted in a position sensitive silicon detector in the focal plane of SHIP where their time of implantation, position, kinetic energy and radioactive decays are registered. Especially the alpha decay properties allow for an unambiguous identification of the respective isotope. In order to reach the focal plane the nuclei must have half-lives of at least 1 $\mu$s which
corresponds to the flight time of the reaction products through the 11 m long setup. For the identification via radioactive decay chains their half-life has to be \( \geq 20 \mu s \) which corresponds to the conversion time plus dead time of the data acquisition system. For nuclei with shorter half-lives pile-up of the signals from the implanted reaction product and the particle emitted during its successive decay occurs.

For the identification of the reaction channel (transfer, fusion-evaporation etc.) we measured the velocity spectra of the produced isotopes. To obtain the spectra we varied the electric and magnetic field values, E and B, of the velocity filter accordingly. The ratio E/B determines at which velocity a particle can pass SHIP. The accepted velocity width at a given setting is \( \Delta v/v = 0.1 \) (FWHM).

The SHIP setup allows the identification of single nuclei. This results from the combination of strong background suppression and the isotope identification via the radioactive decays of the nuclei. This is valid for isotopes produced in fusion-evaporation reactions as well as for isotopes produced in nucleon transfer reactions. The suppression of the primary beam depends on the velocity difference between the projectile- and the target-like nucleus. This difference increases with increasing mass asymmetry between projectile and target. In the here-discussed reactions of \(^{58,64}\text{Ni} + ^{207}\text{Pb}\) and \(^{48}\text{Ca} + ^{248}\text{Cm}\) the factor of beam suppression is on the order of \(10^{11} – 10^{12}\). The remaining projectile-like particles which still reach the focal plane can be distinguished from target-like particles by a time-of-flight (TOF) and energy measurement. Such, projectile- and target-like particles are located on different branches in the energy-versus-TOF spectrum. The obtained mass resolution is \(A/\Delta A \approx 10\). The decisive point for identification of a certain isotope is its radioactive decay or decay chain, respectively, which is registered in the focal plane detector as described above. However, this method is only applicable for nuclei with sufficiently short half-lives. The upper limits for the correlation times are given by the number of background events which have identical signatures like true events [7]. The here described method of identification allows to reach cross-section limits of 10 pb within one day of beam time by applying usual beam intensities of several \(10^{12}\) particles per second. For more details concerning the experimental setup and identification see ref [1, 8].

Concerning the transmission of SHIP there is a significant difference between fusion-evaporation residues and nuclei produced in transfer reactions (the term transmission comprises the angular acceptance and the transport efficiency through SHIP). The full momentum transfer in fusion reactions leads to an emission of the compound nuclei in the direction of the primary beam. Scattering angles, which differ from zero degree result predominantly from small angle scattering in the target. The effect of small angle scattering increases with increasing mass asymmetry between projectile and target nucleus. This causes the drop in transmission for very asymmetric combinations of projectile and target mass [9]. Usual transmissions obtained for fusion-evaporation reactions in the region of superheavy nuclei are \((30 – 50)\%\). In transfer reactions, only a fraction of the incident momentum is transferred to the heavier transfer product, while the remaining fraction is carried by the lighter one. This leads to relatively broad angular distributions in the centre-of-mass as well as in the laboratory frame. For quasi-elastic transfer like pickup or stripping reactions, the angular distributions peak at the grazing angle. But for deep inelastic multi-nucleon transfer with long lifetimes of the underlying DNS the distributions broaden significantly and can be isotropic in the utmost case. Therefore, the transmission of SHIP for transfer products depends strongly on the angular distribution of the transfer products from a certain reaction, which is usually not fully known. In order to estimate the lower limit of the SHIP transmission for transfer products with \(Z > 100\) we assumed the extreme case of an isotropic angular distribution in the centre-of-mass frame. In the laboratory, the heavy target-like product is still emitted within a rather forward directed cone with opening angles up to about 40°. In this case we obtain a transmission of \(\approx 5\%\). Even with this relatively small transmission one can detect one event per day for reactions with total cross-sections of \(200\) pb. The small angle scattering is not dominant for transfer products since they have three times higher kinetic energies than fusion products created at the same beam energy.
Figure 3. Alpha spectrum measured in $^{64}\text{Ni} + ^{207}\text{Pb}$ collisions during beam-off periods. All observed alpha lines can be attributed to isotopes, which have been created in transfer reactions.

Figure 4. Velocity spectrum for $^{214}\text{Ra}$ isotopes created in transfer reactions of $^{64}\text{Ni} + ^{207}\text{Pb}$ at 5.9×A MeV (for description see text).

3. Results and discussion

1.1. Transfer reactions in the systems $^{58}\text{Ni} + ^{207}\text{Pb}$ and $^{64}\text{Ni} + ^{207}\text{Pb}$

Transfer reactions with Pb targets lead to the population of a broad region of alpha emitters between proton numbers $Z = 82$ and $Z = 92$. This allowed us to investigate nucleon transfer from the projectile to the target nucleus in the region of heavy nuclei as well as the available identification and detection methods. We made experiments with the relatively neutron-rich $^{64}\text{Ni}$ and the more neutron-deficient $^{58}\text{Ni}$ beams. The energies of the $^{64}\text{Ni}$ beam were in the range $(4.80 – 5.92)\times A$ MeV, corresponding to $(0.95 – 1.17)$ times the Bass barrier ($V_{\text{Bass}} = 5.05\times A$ MeV). The respective grazing angles are $(180 – 96)^\circ$ in the centre-of-mass system, which correspond to $(0 – 42)^\circ$ in the laboratory system for the target-like transfer product. For the $^{58}\text{Ni}$ beam we applied energies of $(4.85 – 6.50)\times A$ MeV, corresponding to $(0.90 – 1.20)$ times the Bass barrier ($5.4\times A$ MeV). The grazing angles were $(180 – 91)^\circ$ in the centre-of-mass and $(0 – 45)^\circ$ in the laboratory system. The targets were prepared from enriched $^{207}\text{Pb}$. Layers of $380 \mu g/cm^2$ thickness of the chemical compound $^{207}\text{PbS}$ have been evaporated on carbon foils of $40 \mu g/cm^2$ thickness and covered by $10 \mu g/cm^2$ carbon. Eight targets were mounted on a rotating wheel. At each beam energy we scanned the velocity setting of SHIP in the interval $(0.2 – 2.0)$ times the compound nucleus velocity $v_{CN}$ in steps of $0.05\times v_{CN}$.

Figure 3 shows the alpha spectrum measured in reactions of $^{64}\text{Ni} + ^{207}\text{Pb}$. It contains the sum over all beam energy and velocity settings. We detected isotopes of elements with $84 \leq Z \leq 88$, which were directly produced in the reaction, i.e. they were not populated by the decay of mother nuclei. Nuclei with $Z \leq 83$ could not be identified because they are not alpha emitters or have too long half-lives. With $^{58}\text{Ni}$ beams the same elements were populated, but the isotopic distributions peak at the more neutron-deficient side. All observed alpha lines can be attributed to nuclei produced in transfer reactions.

The velocity spectrum of $^{214}\text{Ra}$ ($Z = 88$) is shown in figure 4 representatively for the isotopes created in transfer reactions. The velocity of the detected reaction product was normalized on the compound nucleus velocity. For the different velocity settings limits for the differential cross-sections of $(d\sigma/d\Omega) = (10 – 50) \text{ pb/sr}$ were reached, depending on the respective beam dose and measurement time.
Figure 5. Excitation functions for the isotopes $^{211}$Po, $^{213}$Rn, $^{213}$Fr and $^{214}$Ra measured in $^{64}$Ni + $^{207}$Pb transfer reactions. The arrow marks the energy corresponding to the Bass barrier.

Figure 6. TKE values as a function of the proton number $Z$ of the detected heavy reaction products for different beam energies (the respective beam energies in the centre of mass are given in the inset). For details see text.

For all isotopes we found pronounced maxima around velocities of $v/v_{CN} = 1.6$ and further weak maxima around $v/v_{CN} = 0.4$. The maxima are located symmetrically with respect to the compound nucleus velocity. This structure is caused by the narrow angular acceptance of SHIP of $(0 \pm 2)\degree$. Only transfer products which are created in central collisions can enter the separator. In this case the lighter, projectile-like nucleus is scattered in backward direction and the heavier, target-like nucleus in beam direction with a velocity of $1.6v_{CN}$. This peak position cannot be explained by quasi-elastic transfer, which would lead to velocities of $v/v_{CN} = 1.9$. Rather, the DNS which is formed after capture re-separates with the Coulomb-barrier energy of the exit channel, which means that the isotopes originate from deep inelastic transfer processes (see discussion below). The low-velocity component is consistent with this scenario if we assume that the DNS can rotate around its centre of gravity after a non-central collision. The rotation brings the system again to an orientation parallel to the beam axis where, however, projectile and target have changed their positions with respect to the above described case of a central collision. Therefore, if the re-separation occurs in this orientation, the lighter, projectile-like fragment will be emitted in forward direction and the heavier, target-like in backward direction in the centre-of-mass frame. In the laboratory frame the target-like nucleus is still emitted in beam direction but with reduced velocity [10].

As an example, the excitation functions for the isotopes $^{211}$Po ($Z = 84$), $^{213}$Rn ($Z = 86$), $^{213}$Fr ($Z = 87$) and $^{214}$Ra ($Z = 88$) measured in $^{64}$Ni + $^{207}$Pb reactions are shown in figure 5. One has to note, that the detected nuclei are secondary reaction products which result from the excited primary products after evaporation of neutrons and, at sufficiently high excitation energies, also protons. The excitation functions reveal a systematic broadening and shift of the maximum with increasing number of transferred protons. For $^{211}$Po and $^{213}$Rn the maxima are located at $E \approx 0.95\ V_{\text{Bass}}$ and at $E \approx V_{\text{Bass}}$, respectively. At these beam energies the grazing angle is $180\degree$ in the centre-of-mass. This corresponds to a scattering angle of the target-like nucleus of $0\degree$ in the laboratory frame, which is also the detection angle of SHIP. This explanation holds in the case of direct reactions. However, the excitation functions for $^{213}$Fr and $^{214}$Ra do no longer fit into this scheme. For a possible explanation we took into account the $Q_{\text{gg}}$ values of the $^{64}$Ni + $^{207}$Pb reactions leading to the different isotopes. In the case of polonium and radon the least negative $Q_{\text{gg}}$ values are obtained when $^{211}$Po and $^{213}$Rn are created. This means that the detected $^{211}$Po and $^{213}$Rn nuclei are very close to the primary fragments and must have been created mainly at low excitation energies or beam energies, respectively. For the creation of
Figure 7. Isotopic yields of transfer products from $^{64}$Ni + $^{207}$Pb (top) and $^{58}$Ni + $^{207}$Pb reactions (bottom) as a function of the neutron number N of the reaction products. The neutron number of N = 125 of the target nucleus $^{207}$Pb is indicated by the green dot.

Francium and radium isotopes the optimum $Q_{gg}$ values are obtained for mass numbers $A = 216$ and $A = 218$, respectively. Therefore, $^{213}$Fr and $^{214}$Ra result after the evaporation of three to four neutrons. Larger beam energies are needed to obtain the required excitation energies of the primary fragments for the evaporation of several neutrons. The broadening of the curves we interpret as a hint that the correlation between incident energy and grazing angle becomes weak like it is observed in deep-inelastic transfer reactions with relatively long lifetimes of the underlying DNS.

Indeed, the total kinetic energies (TKE) in the exit channel reveal deep inelastic kinematics for all detected isotopes (figure 6). The TKE values were reconstructed from the measured kinetic energy of the target-like transfer product by assuming two-body breakup. The data points in figure 6 which belong to the same value of Z but have been created at different beam energies have been plotted with an offset for better discrimination. The values expected from the Viola systematic (crosses) are also shown. The TKE values of the reaction products are, within error bars, independent of the beam energy, which points to a deep inelastic transfer process. In comparison, the open circles represent the TKE values, which are expected if the detected isotopes would originate from elastic kinematics at 4.80×$A$ MeV (the $Q_{gg}$ values have been taken into account). The maximum available excitation energy of the DNS is the difference between the centre-of-mass energies in the entrance and exit channels. This is (50 – 60) MeV for the lowest beam energy of 4.80×$A$ MeV (see figure 6). For more details on the $^{64}$Ni + $^{207}$Pb reactions see ref [11].

The observations described for the $^{64}$Ni beams concerning excitation functions and TKE values are also true for the reactions with $^{58}$Ni beams. The only exception is $^{211}$Po, which we will not discuss here.
The isotopic yield distributions obtained with $^{64}$Ni and $^{58}$Ni beams are compared in figure 7 for the Rn, Fr and Ra isotopes ($Z = 86, 87, 88$). For these graphs all beam energies have been added. A common feature for both projectile-target combinations is the drop of the cross-sections by roughly a factor of five to ten with every transferred proton. This is true for all detected isotopes. Further, two characteristic differences between $^{58}$Ni and $^{64}$Ni reactions can be observed. With $^{58}$Ni beams rather neutron-deficient isotopes are populated and the maximum of the distributions shift to the more neutron-rich side with increasing proton number of the transfer product (see figure 7, bottom). A steep drop in the yields can be observed for $N > 126$. For the more neutron-rich $^{64}$Ni beams the yield distributions extend noticeably to the neutron-rich side. The isotope with the largest neutron number is $^{221}$Ra ($N = 133$), which has eight neutrons more than the target nucleus. The corresponding differential cross-section is 500 pb/sr, which is 100 times lower than the differential cross-section of $^{213}$Ra ($N = 125$) which is located in the maximum of the yield curve. Contrary to the $^{58}$Ni reactions, the maxima of the distributions shift to the neutron deficient side with increasing proton transfer. Concerning the yields, we found about two times larger cross-sections for $^{58}$Ni reactions. These results are in agreement with observations from previous experiments described in ref [4] with $^{40,44,48}$Ca beams on $^{248}$Cm targets. The largest neutron transfer observed in the experiments of ref [4] was $\Delta N = 4$. The reason is probably, that the half-lives of more neutron-rich isotopes are on the scale of milliseconds or seconds and are therefore not accessible with chemical methods, which require several minutes. In our experiment the observation of the most neutron-rich isotopes with $N > 128$ was only possible by searching for alpha decay chains correlated in time with a preceding implanted recoil nucleus. This method, which is in the same way applied at SHIP for the identification of superheavy nuclei produced in fusion reactions, allows the identification of single events and gives access to the short-living isotopes with half-lives down to 20 µs. Further isotopes with still larger neutron numbers could not be identified, because they have decay chains with short-living members with half-lives in the region of microseconds and below which cannot be distinguished with the present data acquisition system of SHIP. For the same reason also no Rn isotopes with neutron numbers larger than $N = 127$ could be identified.

1.2. Transfer reactions in the system $^{48}$Ca + $^{248}$Cm

The cross-sections of transfer products drop by up to one order of magnitude with every transferred proton. Therefore, targets with the largest available proton numbers must be applied for the population of superheavy nuclei. Moreover, in order to reach out to the neutron-rich side, the neutron numbers of the projectile as well as of the target nucleus should also be as large as possible. One of the heaviest available target materials is the radioactive $^{248}$Cm with a half-life of $3.4 \times 10^5$ y ($Z = 96, N = 152$). With these targets we investigated transfer reactions using the neutron-rich $^{48}$Ca beams. $^{48}$Ca is a doubly magic nucleus and $^{248}$Cm has a subshell closure at $N = 152$, which leads in both cases to high binding energies. In this case we expect, in analogy to fusion reactions, relatively low excitation energies of the primary transfer products. $^{248}$Cm targets are applied at SHIP since June 2010. The first application was from June 25 to July 26, 2010, for the production of different isotopes of element 116 in the fusion reactions of $^{48}$Ca + $^{248}$Cm $\rightarrow ^{296}116^*$. This experiment, where a total of six decay chains of isotopes $^{296}116$ and $^{297}116$ were observed as well as the applied target technique are discussed in detail in ref [12]. After the setting for fusion-evaporation residues we applied two days of beam time for the investigation of transfer reactions at the beam energy of 5.63×$\Delta$ MeV. This energy is 15% larger than the Coulomb barrier, which is obtained by assuming spherical Cm nuclei. Data were taken at five different velocity settings, 1.70, 1.80, 1.85, 1.90 and 1.95 times the compound nucleus velocity $v_{CN}$. The analysis of the transfer reactions from $^{48}$Ca + $^{248}$Cm is still in progress. Therefore, more detailed or improved information than discussed below might be available after finishing the analysis.
We identified numerous isotopes in the region of $Z < 96$ via alpha decays. The respective alpha energy spectrum is shown in figure 8. Like in the case of $^{58,64}$Ni + $^{207}$Pb reactions the detected nuclei are secondary reaction products which result after evaporation of neutrons from the excited primary products. Figure 8 reveals alpha lines which belong to isotopes with proton numbers $Z = 84$ (polonium) to $Z = 88$ (radium). We observed the maximum production yield for isotopes with $Z = 88$, which result after the transfer of 8 protons from the target to the projectile nucleus. The respective differential cross-sections are $d\sigma/d\Omega \approx (100 - 500) \text{ nb/sr}$ related to the angular range of $(0 \pm 2)^{\circ}$ accepted by SHIP. According to the present status of analysis we can identify isotopes down to $Z = 86$ as directly produced, i.e. not populated by the decay of mother isotopes. The main contribution to the alpha line intensities for $Z \leq 86$ nuclei comes from the population due to mother decays of $Z = 88$ and $Z = 87$ isotopes. If also the $Z = 84$ lines contain a contribution from direct production is subject to the ongoing analysis. It is noteworthy, that we observed a sudden steep drop in the yield by more than a factor of ten for nuclei with proton numbers $Z \geq 89$. We identified these isotopes by their alpha decay chains. In figure 8 they give only a small contribution to the much more dominant alpha lines of isotopes with lower proton numbers. The only exception is some isotopes around the $^{248}$Cm target nucleus, which were populated in quasi-elastic nucleon stripping or pickup reactions. They were produced with differential cross-sections of up to $\approx 10 \text{ µb/sr}$. We identified these nuclei due to their beta-decays, which lead to excited states of the respective daughter nuclei. The gamma rays following the de-excitation were detected in coincidence with the electron from the preceding beta-decay. The gamma-ray energies allowed for the assignment of the respective mother isotope. We are not yet sure about the cross-section maximum observed for $Z = 88$. Actually, we would expect that the isotopes closer to the target are populated stronger and that the cross-sections decrease with every transferred proton, like observed in the reactions of $^{58,64}$Ni + $^{207}$Pb. A possible reason for the large cross-section for radium isotopes might be connected with the fission barriers, which decrease gradually with increasing proton number in the region of actinide nuclei. Since the measured nuclei are secondary reaction products the observed alpha spectra reflect also the loss of primary fragments due to sequential fission or nucleon evaporation, where especially the probability for sequential fission increases for nuclei closer to $^{248}$Cm, while the nuclei closer to lead are more resistive. This might lead to the yield maximum at $Z = 88$ for the secondary isotopes, while the yield of the primary isotopes still decreases gradually with increasing number of transferred protons. Theoretical studies are presently undertaken to investigate this scenario [13].

Besides the alpha activity described above, we observed also numerous SF decays during the settings for transfer products. After the beam time the registration of SF events was continued and will run for
about half a year more in order to see also possible long-living activities (see below). Since the energies of spontaneous fission events are no clear fingerprint of the decaying nucleus, especially not at small numbers of events, an assignment can only be made via recoil-fission correlations. This method is, however, restricted to sufficiently short-living isotopes. The maximum possible correlation time is given by the counting rate of possible recoils. In our experiment it allowed non-ambiguous correlations within time windows of 100 ms. The observed correlations between SF events and preceding recoil nuclei within a time of 100 ms are shown in figure 9. Two groups of SF events with different half-lives can be distinguished. For the group of events with correlation times below 10 ms we obtain a mean correlation time of $t(\text{Re-SF}) = (1.63 \pm 1.14)$ ms and for the group with correlation times larger than 10 ms we obtain $t(\text{Re-SF}) = (25.26 \pm 14.88)$ ms. We attributed these events to fission isomers in $^{242}\text{Am}$ ($T_{1/2} = 14$ ms) and $^{244}\text{Am}$ ($T_{1/2} = 0.9$ ms) with mean lifetimes of 20.2 ms and 1.30 ms, respectively. The americium isotopes (Z = 95) can be reached by the exchange of one proton and the exchange or/and evaporation of several neutrons. The calculated envelopes of the time distributions, which we expect for events from $^{242}\text{Am}$ and $^{244}\text{Am}$ are also drawn in figure 9. The respective differential cross-sections are $\approx 700$ pb/sr for $^{244m}\text{Am}$ and $\approx 100$ pb/sr for $^{244m}\text{Am}$. The activity with the shorter half-live would also fit to an isomeric state in $^{244}\text{Am}$ with a lifetime of 1.36 ms ($T_{1/2} = 0.94$ ms). However, within error bars, we can not decide, if the SF events with $\tau = 1.63$ ms originate from $^{240m}\text{Am}$ or from $^{244m}\text{Am}$ or if there are contributions from both. One has still to note that for Am nuclei we can only identify the isomeric but not the ground states, because they are too long-living.

The time distribution of the long-living SF events is shown in figure 10 for the first 15 days after the beam time. The respective measurement was started just after the end of the beam time. Each bar contains the events which were observed within one day. The activity measured within the first day after beam time is significantly higher than on the following days. Within statistical fluctuations, the activity decreases from the second day on. The missing events for days nine and ten are due to a stop of the measurement during these two days, while on days 11, 13 and 15 no decays occurred. The shape of the distribution suggests, that there are contributions from several activities with different half-lives. A more detailed analysis is in progress.

For the long-living fission events a clear assignment to a certain mother nucleus via time correlations is no longer possible. Different time components can only be roughly distinguished. Figure 11 shows the isotopes, which are the most probable candidates for the observed SF decays. The location of the target nucleus, $^{248}\text{Cm}$, is also shown for orientation. The chart contains only isotopes which have half-lives shorter than one year and sufficiently large SF branches of $> 0.5\%$. Nuclei, with longer half-lives
and smaller fission branches are not detectable at SHIP taking into account the expected small production cross-sections. The half-lives of several isotopes are too similar and cannot be unambiguously attributed to a certain nucleus. However, one can deduce from these results that the region of nuclei with \( Z \approx 100 \) was populated during this experiment with high probability.

In order to reach the neutron-rich nuclei in the region of \( Z \geq 100 \), low beam energies should be favourable. The nuclei in this region have relatively low fission barriers. As a consequence, the probability that the excited primary transfer product undergoes sequential fission is larger than for lighter nuclei and is increasing with increasing excitation energy or beam energy, respectively.

Secondly, the nuclei which survive sequential fission de-excite by evaporating neutrons. The lower the excitation energy, the lower is the number of evaporated neutrons and the more neutron-rich are the secondary nuclei. Similar to fusion-evaporation reactions, the cross-sections for the production of a certain transfer product can be written as the product of three factors:

\[
\sigma = \sigma_{\text{capture}} \times P_{\text{transfer}} \times P_{\text{survival}}.
\]

The factor \( \sigma_{\text{capture}} \) is the capture cross-section and \( P_{\text{transfer}} \) is the probability to create a certain primary transfer product. These two values increase when the beam energy exceeds the Coulomb barrier. The factor \( P_{\text{survival}} \) is the probability that the primary transfer product survives sequential fission and that it evaporates just the number of neutrons which lead to the desired secondary product. \( P_{\text{survival}} \) decreases with increasing excitation energy. Especially the influence of sequential fission on \( P_{\text{survival}} \) becomes more critical for very heavy nuclei with decreasing fission barriers. The extreme case is superheavy nuclei, where the fission barriers are only determined by the shell correction energies. These qualitative considerations are also found by theoretical model calculations [13]. Figure 12 shows the excitation functions for the isotopes \(^{259}\text{No} (Z = 102), ^{264}\text{Lr} (Z = 103)\) and \(^{271}\text{Sg} (Z = 106)\) created in transfer reactions in the collision system \(^{48}\text{Ca} + ^{248}\text{Cm}\). These three isotopes are already located towards the neutron-rich side of the nuclear chart. \(^{264}\text{Lr}\) and \(^{271}\text{Sg}\) are also new isotopes in the sense, that they have not been produced directly, so far. According to the model calculations, the maximum cross-sections for \(^{264}\text{Lr}\) and \(^{271}\text{Sg}\) are located at energies, which are only 2% above the Coulomb barrier.

4. Summary and outlook

We studied multi-nucleon transfer reactions in the collision systems \(^{58,64}\text{Ni} + ^{207}\text{Pb}\) and \(^{48}\text{Ca} + ^{248}\text{Cm}\). In both cases we observed a massive transfer of protons and neutrons already at beam energies below the Coulomb barrier. The transfer from the projectile to the target nucleus was of special interest, since we investigated the possibility to create new neutron-rich nuclei in the region of superheavy elements.
with $Z \geq 100$, which cannot be reached in fusion-evaporation reactions. We performed the studies at the velocity filter SHIP. The setup allows the detection of the heavy, target-like transfer products and their identification via radioactive decays. Such, even single nuclei can be unambiguously identified. With this detection method, which is also applied for superheavy nuclei produced in fusion reactions, in combination with the low background on the focal plane detector a cross-section limit of 10 pb can be reached within one day of beam time applying usual beam intensities of $5 \times 10^{12}$ particles per second. Therefore SHIP is presently the most sensitive apparatus for the study of such transfer reactions for which cross-sections of nanobarn and below are expected. A further advantage is that SHIP allows the detection of the transfer products, which are emitted in forward direction. This is essential if the reactions are performed at beam energies below and close to the Coulomb barrier. Such low beam energies seem favourable for the production of very heavy nuclei with large neutron numbers, since excitation energies are low in this case, which prevents the excited primary transfer products from sequential fission or from the evaporation of too many neutrons.

In collisions of $^{58}$Ni and the more neutron-rich $^{64}$Ni with $^{207}$Pb nuclei we observed the transfer of up to six protons from the projectile to the target nucleus. With $^{68}$Ni beams we reached out to the neutron-rich side with respect to the target neutron number $N = 125$. We detected isotopes resulting from the transfer of up to eight neutrons ($^{221}$Ra). Still more neutron-rich nuclei were not observed. They would have been accessible concerning the cross-sections, but their radioactive decay chains comprised short-living members (<20 µs), which were not accessible with the data acquisition system. Also in the $^{48}$Ca + $^{248}$Cm collisions we observed numerous transfer products with proton numbers below as well as above the target proton number. The transfer products with $Z < Z_{\text{cm}}$ can be identified via their alpha decays. But for the transfer products with $Z > Z_{\text{cm}}$ the limitations of the present detection technique become visible. The nuclei in this region are very long-living and can no longer be identified via their decay chains. Further, the nuclei on the neutron-rich side undergo SF or beta-decays, which allow, in contrary to alpha decay chains, no unambiguous identification of the mother nucleus. In our experiment we observed several SF activities which can be roughly attributed to isotopes with different half-lives. However, an unambiguous attribution to certain mother nuclei was not possible. Therefore, for long-living isotopes and those which undergo SF- or beta-decays other detection methods have to be applied. Presently we are investigating the possibility to identify them via precision mass measurements. Penning traps, like SHIPTRAP at GSI [14], and time-of-flight mass spectrometers, like the MR-TOF-MS developed at the University of Gießen [15], have sufficiently large resolution of $m/\Delta m > 10^5$ to perform also an isobaric separation for most of the isotopes. Part of

![Calculated excitation functions for the isotopes $^{259}$No, $^{271}$Sg and $^{264}$Lr produced in nucleon transfer reactions in the collision system $^{48}$Ca + $^{248}$Cm [13]. The arrow denotes the Coulomb barrier energy.](image)
our program at GSI is to make feasibility studies of this method for the application to cross-sections below nanobarn. Especially the efficiency of the buffer gas cell has to be improved, which is presently on the level of 1%. The buffer gas cell is needed for stopping the reaction products, which have initial energies of ≈100 MeV, for the injection to a Penning trap or time-of-flight mass spectrometer, which requires ion energies on the order of 100 eV. The application of alternative detection methods is also essential to study the development of production cross-sections with increasing proton and neutron transfer. With intermediate heavy beams on heavy targets, like $^{48}$Ca + $^{248}$Cm, neutron-rich isotopes with proton numbers up to $Z \approx 105$ are predicted by model calculations with total cross-sections of several ten up to several 100 pb. This would make transfer reactions a feasible means to create these nuclei if the predicted cross-sections can be verified experimentally.

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