THE DISCOVERIES OF BOHRIUM, HASSIUM, MEITNERIUM, AND THE NEW REGION OF DEFORMED SHELL NUCLEI

Gottfried Münzenberg
Gesellschaft für Schwerionenforschung (GSI) mbH, Planckstrasse 1,
D-64220 Darmstadt, Germany
and Johannes Gutenberg-Universität Mainz, Germany
e-mail: g.muenzenberg@gsi.de

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Dedicated to Adam Sobiczewski in honour of his 70th birthday

The investigation of the light trans-actinide elements was not only exciting as it included the discovery of a number of new chemical elements. It led also to the discovery of a new region of shell nuclei existing beyond the macroscopic stability limit. Theory explained this in terms of a new shell region of deformed nuclei which bridge the trans-uranium nuclei and the predicted superheavy elements. This contribution will give a brief historic overview over these discoveries, experimental developments, and the impact on ongoing and future superheavy-element research.

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1. Introduction

One of the fundamental questions is, how many chemical elements can exist, and what is the limitation of their number. Already with the discovery of fission it became clear that the number of chemical elements is limited by this decay mode. In the macroscopic description this limit is located near rutherfordium ($Z = 104$), the heaviest actinide element. With the development of the nuclear shell model the question arose, where the next double shell closure above ^{208}Pb is located and whether it could be strong enough to allow the existence of superheavy nuclei beyond the liquid drop limit. Since those times the ultimate goal of heavy-element research is the synthesis and investigation of the superheavy elements, a new kind of nuclear species which exist only by shell stabilisation.

The early investigations of the trans-actinide nuclei were carried out with the gas-jet technique and led to the discovery of dubnium and seaborgium [1,2]. The heavy nuclei produced by complete fusion of heavy ions recoil from

the target, are stopped in a gas cell, and transported through a thin capillary to surface barrier detectors where they are identified by their decay characteristics with the alpha–alpha parent–daughter-correlation method [3]. This technique is limited to half-lives of seconds. A method, applicable to fissioning species, was the rotating drum [4]. The target material, deposited at the surface of a rotating cylinder, serves simultaneously as stopper for the fusion products. The fission fragments emerging from the decay of the heavy nuclei sticking at the cylinder surface are detected with mica track detectors. The rotation velocity of the cylinder determines the accessible half-live region which ranges down to milliseconds. Consequently only alpha-emitters with half-lives of seconds and fission with half-lives down to milliseconds were known. The heaviest element for which α-decay had been observed so far was seaborgium. All elements beyond were reported to undergo spontaneous fission. This was in good agreement with theory and systematics. Systematics as well as theory [5] predicted fission half-lives to decrease below milliseconds for the trans actinide elements. According to these predictions the superheavy nuclei form an island, well separated from the trans uranium elements.

The situation changed with the application of the in-flight separation technique. Separation times became as short as micro seconds. Alpha decay as well as fission could be detected at the same time in the same detector down to this limit. This new technique opened the way to the full exploration of the trans-actinide region [1,2].

2. Separation in-flight and the parent–daughter method for heavy-element identification

The key to the discovery and investigation of short lived nuclides at the very limits of nuclear stability is the separation in-flight. It is fast, sensitive, and independent from the chemical properties of the species to separate. The separation time, determined by the flight time through the separator, is of the order of micro seconds. The break-through with in-flight separation for heavy-element research was the investigation of the trans-actinide region with SHIP. Up to then the odd-mass isotopes of rutherfordium, which are α-emitters with half-lives of seconds were detected with the helium gas-jet whereas the even–even isotopes undergoing spontaneous fission with decay times of milliseconds were observed with the rotating drum. With SHIP for the first time the simultaneous measurement of fission and α-decay became possible.

The experiments at SHIP gave the first unambiguous proof for the cold heavy fusion, the production of weakly excited heavy compound nuclei by complete fusion using lead or bismuth targets, as proposed by Oganessian

[6], by the observation of the $1n$-channel in irradiations of ^{208}Pb with ^{50}Ti leading to ^{257}Rf. This indicates the production of cold compound nuclei with excitation energies of close to 10 MeV [7]. Cooling down the compound nucleus by the evaporation of only one neutron assures the largest survival probability for the heaviest, extremely fissile elements. It opened the way to the production of the new elements of Bohrium to $Z = 112$ [8], see contribution of S. Hofmann to this volume.

The velocity filter SHIP (Separator for Heavy Ion reaction Products), Fig. 1, is a kinematic separator, using the specific properties of complete fusion products to recoil with center of mass velocity from the reaction target [9]. Lead or bismuth targets with a thickness of about 0.5 mg/cm^2 are used. The reaction products recoil almost unretarded from the thin target. To improve the separation quality, SHIP is a combination of two velocity filters with separated electrostatic and magnetic deflection fields. Magnetic quadrupole triplets at the entrance collect the recoils with almost the full solid angle. At the symmetry plane the projectile beam is separated from the recoils. A quadrupole triplet at the exit focuses the recoils onto the detector system.

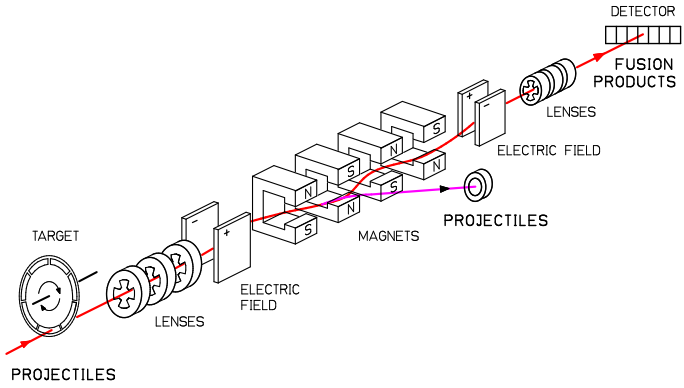

Fig. 1. Schematic view of the velocity filter SHIP as used for the early experiments.

All in-flight separated nuclei are identified atom-by-atom by their nuclear decay characteristics with the use of the α–α parent–daughter-correlation [3]. At SHIP the parent–daughter correlation technique has been developed to the highest possible sensitivity, the identification of new isotopes or even elements by individual atoms [10]. The implantation of the in-flight separated recoils into a position sensitive silicon detector allows to follow their individual history in situ over hours or even days. Thin carbon foil secondary-electron detectors allow for time-of flight measurement to give a rough information on the recoil mass in combination with the energy signal from the implantation.

3. The discoveries of elements bohrium (107), hassium (108), and meitnerium (109)

Element 107, meitnerium, has been produced in irradiations of ^{209}Bi with ^{54}Cr [11]. In the first experiment the isotope ^{262}107, formed by the evaporation of one neutron from the compound nucleus, was identified on the basis of six decay chains, one of them originating from an isomeric state. The cross section was 0.2 nb. The chains were assigned using the parent–daughter-correlation method. For this purpose the daughter isotopes of element 105 had to be investigated before this experiment as they were unknown at that time (Fig. 2). They were produced in a companion experiment irradiating ^{209}Bi with ^{50}Ti. This procedure ensured the best performance in terms of experimental uncertainties including the calibration procedure and stability of the set-up. In a later experiment the isotope with mass 261 was observed. These results were confirmed later.

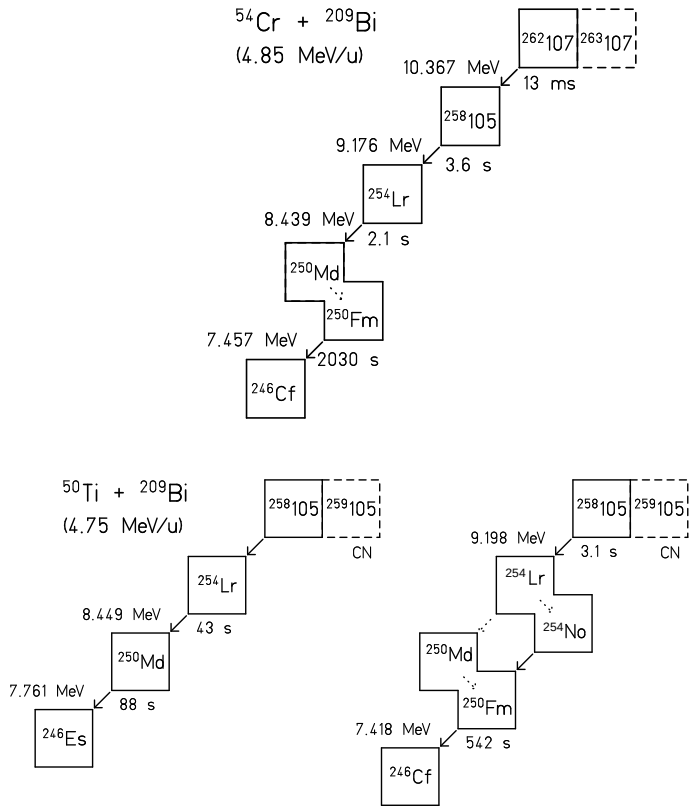

Fig. 2. The first decay chain of element 107 together with two decay chains from the corresponding daughter nuclides of element 105.

The question was to which element to proceed next. According to systematics, the cross section for the production of element 108 in an irradiation of ^{208}Pb with ^{58}Fe should be only by a factor of three smaller than the one for element 107. On the other hand, to observe α-chains, essential for the assignment of the new element, it would be more safe to proceed to element 109, bohrium. From theory as well as from the systematics of fission isomers it was known, that the hindrance factor for fission would be of the order of 10^3 for each odd nucleon, highest for an odd-Z element and its odd-N isotope. So we decided to proceed to element 109, in spite of the fact that we expected a cross section to be smaller by a factor of ten as compared to 107.

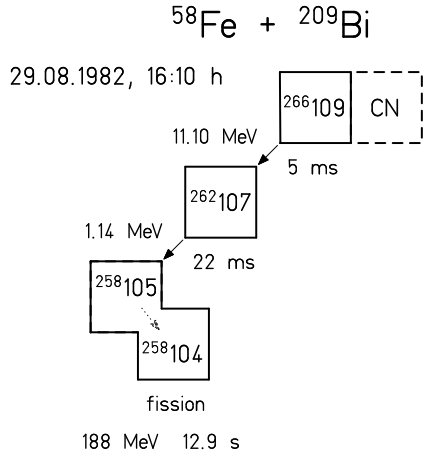

Fig. 3. The first decay chain of element 109.

The experiment was successful: Element 109 was identified by the observation of one single atom (Fig. 3) in the irradiation of ^{209}Bi with ^{58}Fe of 250 h duration with an integral ion dose of 7×10^{17} [12]. The assignment based on a careful statistical analysis to exclude that the observed chain is a random sequence of events and to find the most probable assignment by comparing the observed daughter decays to all possible decay paths along known isotopes. The assignment was difficult as the daughter α-decay was not detected with its full energy. It had escaped the implantation detector so that only the observed time interval could be used. The chain was terminated by fission. The α-decay energy of ^{266}109 was measured to 11.10 MeV, the half-live about 3.5 milliseconds. The production cross section is of the order of 15 pb. This result was confirmed later in a number of experiments [8]. An independent and very interesting confirmation came from a series of chemical experiments at Dubna [13] in which the long-lived end-member of this chain, ^{246}Cf was identified by its α-decay after off-line chemical separation, and in two succeeding GSI experiments [8].

After this positive and unexpected result, the observation of α-decay for element 109, we were optimistic to start the synthesis of element 108, hassium. This was produced in irradiations of ^{208}Pb with ^{58}Fe. For the same reason as in the synthesis of element 107, in a preceding companion experiment the synthesis of element 106 isotopes was carried out in irradiations with ^{54}Cr. Three α-decay chains from the element 108-isotope with mass 265 were identified. Their α-decay energy is 10.36 MeV, the half-life close to 2 ms [14]. The example of the analysis and assignment procedure is given in Fig. 4. The first step is the analysis of the α-spectrum taken between the accelerator beam-bursts, without any further condition. It shows pre-

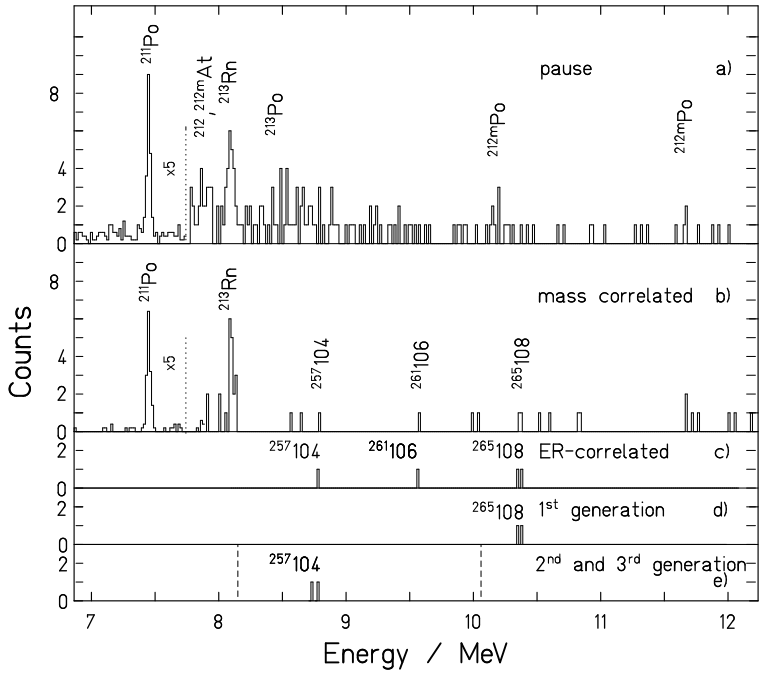

Fig. 4. The α-spectra analysed for the identification of element 108 by its isotope with mass 265 with various analysing conditions. Please note that only those alphas from element 108 which appear in the beam pause are analysed in the spectra [14].

dominantly contaminants from the transfer products polonium, astatine and radon. Most of those are suppressed when the mass correlation is introduced, *e.g.* the condition that the α-decay is measured after the implantation of a recoil. The background disappears after setting a window on the masses of the implanted recoils. The next spectra show the daughter and granddaughter decays. The heaviest doubly even isotope ^{264}108 was also observed as an α-emitter, but unfortunately only as an escape, *e.g.* not with its full energy,

so the Q_α-value could only be estimated from the measured time interval using a Geiger–Nuttal systematics adapted to the heaviest elements [14].These result were confirmed later [8]. The striking result was the observation of α-decay for all three chains, a clear indication for an enhancement of stability with respect to fission, not expected from the earlier predictions.

4. The new shell region around hassium

The observation that α-decay dominates in the decay of elements 107, 108, and 109 points towards a stabilisation against fission in the explored region. In fact, already the analysis of the element 106-isotopes with masses 259, 260, and 261, and the earlier observed one with mass 263, are α-emitters [15]. The doubly even $^{260}106$ has a fission branch of 50%. This was the direct proof of a result which was deduced earlier form a series of cross bombardments with the rotating drum [16].

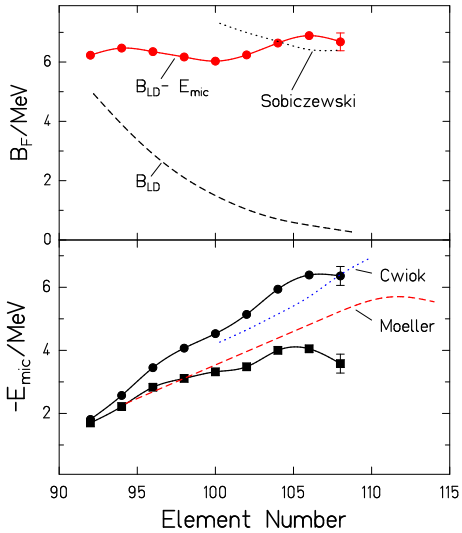

Fig. 5. Experimental shell correction energies (lower panel) and fission barrier heights (upper panel) along the N–$Z = 48$ α-chain [14].

To investigate the microscopic corrections, an analysis of the ground-state masses obtained from the measured Q_α-values of the doubly even nuclides with the N–$Z = 48$ isotopes along the α-chain starting from $^{264}108$ has been made [14, 15]. From the ground state mass the shell correction energy was obtained using a macroscopic-microscopic reference. The result, displayed in Fig. 5, clearly shows the increase of the microscopic energy towards element 108, where it apparently saturates at a value close to 6 MeV, roughly half of the shell strength of the doubly magic ^{208}Pb, the strongest

shell we know in nature. Even more striking is the result that the fission barriers from uranium to element 108 are almost constant, and close to 6 MeV. They were obtained to a rough approximation by adding the experimental microscopic correction to a macroscopic fission barrier taken from a systematics [18]. Both results agree well with theory including the fission barrier heights [17].

The origin of this enhanced stability can only be revealed by a theoretical analysis. A thorough theoretical investigation analysis of the properties of the trans-actinide elements [19] showed that the enhanced stability of the trans-actinide nuclei is due to a shell gap at $Z = 108$ and $N = 162$, associated with a hexadecapole deformation in the ground state. The map of calculated shell correction energies for the trans-actinide nuclei, Fig. 6, shows the new shell region, interconnecting the trans-uranium elements and the predicted region of superheavy nuclei around $Z = 114$. It shows a maximum at $^{270}108$ with shell correction energies of up to 7 MeV, in excellent agreement with the experimental data, taking into account that these end at $^{264}108$, still six neutrons remote from the maximum. This finding had dramatic consequences for the investigation of the heavy and superheavy elements. It opened a completely new region of the nuclear chart for heavy-element research and made a bridge to the superheavy elements, which allows to approach them stepwise.

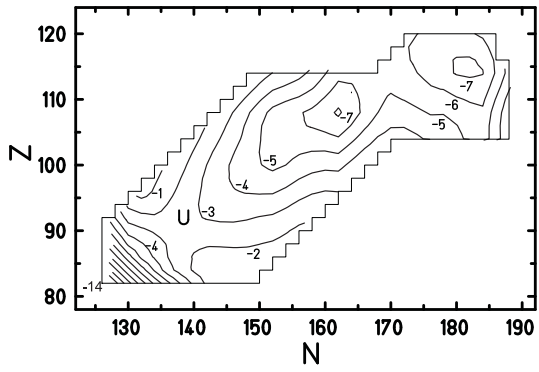

Fig. 6. Groundstate shell corrections for the region from lead to the spherical superheavy elements [21].

The theoretical analysis of the fission barriers explained, on the first glance inconclusive observation, that the fission barriers are constant from uranium to element 108 while the partial fission half-lives drop from 8×10^{15} yr for ^{238}U to 1 ms for ^{264}Hs, by more than 23 orders of magnitude. Fig. 7 gives the explanation [20]: The fission barriers of the trans-actinides, created only by the microscopic effects, are extremely narrow, whereas the fission barriers of the actinides, dominated by macroscopic effects, are thick.

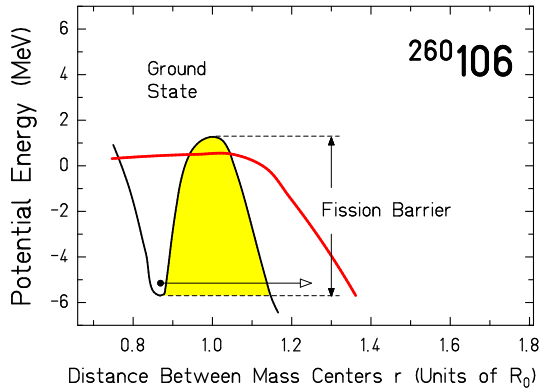

Fig. 7. Calculated fission barrier for $^{260}106$, as a typical trans-actinide shell barrier [17].

As the fission half live is related to the barrier penetrability p and the number n of barrier assaults per unit time, the half-life is

$$T_{1/2} = \frac{\ln 2}{pn}$$

with

$$p = \left[1 + \frac{\exp(2\pi B_f)}{\hbar\omega_f}\right]^{-1/2} ,$$

where B_f denotes the height of the fission barrier and $\hbar\omega_f$ the barrier curvature. The barrier curvature, respectively thickness, varies significantly from the liquid drop character to the typical shell barrier, and appears in the exponential, which explains the dramatic changes in half-life. Fig. 7 also shows the shallow liquid drop barrier of less than 0.5 MeV, which is not able to stabilise the nucleus against spontaneous disintegration.

In conclusion, the theoretical calculations, as illustrated in Fig. 7, prove two important new facts:

- the trans-actinide nuclei are shell nuclei, they exist only by microscopic stabilisation;

- the new shell region interconnects the actinides, at the upper end of the chart of nuclides, to the predicted region of spherical superheavy nuclei. This has dramatic consequences for experiment: it creates the possibility to approach the superheavy nuclei stepwise along this new bridge.

5. New opportunities for SHE research

The implication of this new region of deformed shell nuclei on half-lives has also been investigated [17]. Fig. 8 shows the half-lives of the nuclides around the new shell region. The nuclides above element 106 are predicted to be α-emitters with half-lives of above milliseconds, ranging up to hundred

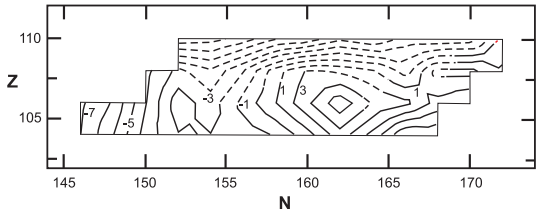

Fig. 8. Predicted half-lives for the trans-actinide nuclei.

seconds. This opens the trans-actinide region not only for investigations with in-flight methods, but also for chemical investigations, and atomic physics. In fact the discovery of this new shell region is the basis of the ongoing heavy-element research. It opened the region for three more new elements [8], 110, 111, and 112. Only recently Dubna reports experimental results on elements 114, and 116, entering the region of the spherical SHE [22].

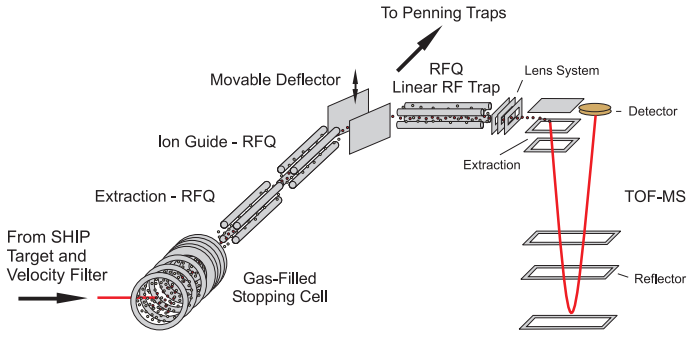

Fig. 9. A schematic view of the SHIPTRAP setup with the gas stopper cell, the ion guide, and a time-of-flight mass spectrometer.

The comparatively long half-lives of the order of seconds allowed to chemically investigate the elements seaborgium, meitnerium, and recently even hassium [23]. New opportunities will be opened with SHIPTRAP, a trap positioned behind SHIP to investigate nuclear, atomic, and chemical properties of the heaviest elements [24]. Fig. 9 shows a schematic sketch of the setup. The atoms separated in-flight with SHIP are stopped in a gas cell and then gas cooled and transported to a trap system. First experiments

will be carried out with a multi-reflection time-of flight spectrometer which will not only help for determination of the mass number, but also allow for direct mass measurement. Having passed the time-of flight mass spectrometer they can be implanted into silicon surface-barrier detectors for decay studies.

6. Conclusion

The exploration of the trans-actinide elements with the in-flight method led to the discovery of a new shell region, interconnecting the trans-uranium elements and the predicted region of superheavy elements. It created the basis of present heavy-elements research. The comparatively long half-lives allowed also chemical investigations and will open this region for investigations in ion traps.

The close collaboration with theoreticians, and in particular with Adam Sobiczewski and his school helped to understand these results, to reveal the origin of the experimentally enhanced stability. The predictions based on these theoretical models will help to explore the heaviest elements SHE physics and lead us on the way to the spherical superheavy nuclei.

We always highly appreciate the close connection with Adam Sobiczewski, the inspiring and always fruitful discussions, and his eminent contribution to the understanding and the physical interpretation of our experimental results which largely contributed to the discovery of the deformed shell nuclei around hassium.

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