Nuclear Structure of the Heaviest Elements – Investigated at SHIP-GSI

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Abstract. The quest for the heaviest nuclei that can exist is a basic topic in natural science as their stability is characterized by a delicate interplay of short range nuclear forces acting between the nucleons (protons and neutrons) and long-range Coulomb forces acting solely between charged particles, i.e. the protons. As the stability of a nucleus is strongly correlated to its structure, understanding the nuclear structure of heaviest nuclei is presently a main challenge of experimental and theoretical investigations concerning the field of Superheavy Elements. At the velocity filter SHIP at GSI Darmstadt an extensive program on nuclear structure investigations has been started about a decade ago. The project covered both as well systematic investigations of single particle levels in odd-mass isotopes populated by α-decay as investigation of two- or four-quasi-particle states forming K isomers and was supplemented by direct mass measurements at SHIPTRAP and investigation of spontaneous fission properties. Recent experimental studies allowed to extend the systematics of low lying levels in N = 151 and N = 153 up to ^{255}Rf and ^{259}Sg, investigation of possible relations between nuclear structure and fission properties of odd-mass nuclei and investigation of shell strengths at N = 152 and towards N = 162.

1 Introduction

On the basis of the nuclear drop model, first suggested by C.F. von Weizsäcker in 1935 [1] instability of the atomic nucleus against prompt disruption was expected at $Z \approx 106$, the exact value depending on the parametrization of the contributions, like volume energy, Coulomb energy etc. Extrapolations of the nuclear shell model performed in the midths of the sixties of the last century lead to the prediction of spherical proton and neutron shells at Z = 114 and N = 184 [2,3]. Nuclei around these shell closures, soon denoted as 'superheavy', were expected to be stabilized against prompt fission by large ground state correction energies ('shell effects'); thus long half-lives were expected.

For about three decades predictions were based on macroscopic-microscopic models which agreed in the originally predicted spherical shell closures and, in advanced versions [4,5], predicted another island of enhanced stability in a region of strong prolate deformation around Z = 108 and N = 162. By the end of the 20th century first results from self-consistent Hartree-Fock-Bogoliubov (HFB) calculations using Skyrme- or Gogny- type forces and Relativistic Mean Field (RMF) calculations using different parametrizations were published. According to these, the location of the shell closures

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became under discussion, Z = 120 or even Z = 126 appeared as possible proton shell closures, while besides N = 184, some parametrizations predicted N = 172 as neutron shell (see e.g. [6]). Therefore besides the quests for the strength of the shell effects and the extension of the region of highly stabilized nuclei also that for its location arose.

Principally, there are two approaches to these questions: a) synthesis of isotopes in the region of the predicted shells and measuring their basic decay properties, which has been successful up to Z = 118, and b) detailed nuclear structure investigations in the transfermium region, typically around N = 152. While in case a) already properties derived from a few observed decays (α-energy, half-life, spontaneous fission branch) allow for some conclusion on a possible stabilization, in case b) a larger number (typically >>100) is necessary and allows only for indirect conclusions. Some of the single particle levels relevant for existance and strengths of the proton shell gaps at Z = 114 or Z = 120 and the neutron shell gap at N = 172 or N = 184 come close to the Fermi level at Z ≈ 100 – 106 and N ≈ 152. So their positions and orderings there will be relevant for their positions in the 'SHE – region', i.e. the data measured there will have drawback on the predictions for the spherical shells.

Under these considerations an extensive program on nuclear structure investigations of nuclei in the range Z =100 – 108 and N ≈ 145 – 162 was started a decade ago. The interest was focussed on extending systematics in excitation energies and ordering of low lying Nilsson levels, typically at E^* < 0.5 MeV, in odd-mass isotopes. The studies were supplemented by investigation of spontaneous fission properties and direct mass measurements allowing to deduce experimental masses of α-decay precursors; in nuclei with odd protons and/or neutrons where no intense ground-state to ground-state transitions are observed this was achieved in combination with γ-spectroscopy. The results can be used to test theoretical models on mass and shell correction energies predictions.

In the present paper we will discuss some aspects related to the properties of superheavy elements, specifically systematics of low lying Nilsson levels in N = 151 and N = 153 even-Z isotones, possible relations between nuclear structure and fission hindrance in odd mass nuclei, and the trend of 2n – binding energies in N – Z = 50 even-even nuclei towards the predicted neutron shell at N = 162.

2 Experiments

The experiments were performed at the Universal Linear Accelerator (UNILAC) at GSI – Helmholtzzentrum für Schwerionenforschung GmbH, Darmstadt, Germany.

The isotopes were produced in complete fusion reactions using beams of ^{48}Ca, ^{50}Ti and ^{54}Cr and targets of $^{206-208}$Pb or ^{209}Bi. The evaporation residues were separated in-flight from the projectile beam and unwanted reaction products by the velocity filter SHIP [7] and implanted into a position sensitive 16-strip Si-detector (stop detector) allowing for measuring the evaporation residues, α-particles, fission products and conversion electrons. Gamma radiation emitted in prompt or delayed coincidence with particles was measured with a Ge-Clover detector placed behind the stop detector. Further details on the experimental set-up and methods can be found in [8].

3 Nilsson levels systematics in N = 151 and N = 153 isotones

In the region between the established neutron shell at N = 126 and the predicted one at N = 184 the orbitals $2g_{9/2}$, $1i_{11/2}$, $1j_{15/2}$, $3d_{5/2}$, $2g_{7/2}$, $4s_{1/2}$ and $3d_{3/2}$ are expected, with a relatively large energy gap of ≈1.5 MeV between the $1j_{15/2}$ and $3d_{5/2}$ orbitals [9]. At deformation these orbitals split into the Nilsson levels, some of them increasing in energy at deformation, some decreasing in energy. Part of these resulting Nilsson levels are close to the Fermi surface at N ≈ 152 or responsible for the strength of the deformed neutron shell at N = 152. So performing nuclear structure investigations there will have feedback to the energies and ordering of the neutron orbitals and thus on the strength and probably also on the location of the next spherical neutron shell. Vice versa this also holds for the protons.

It is well established that in the transuranium region similar nuclear structure is observed within the isotone line in even-Z nuclei and along the isotope lines in odd-Z nuclei thus allowing to study the development (excitation energy change and possible change of ordering) of single particle levels adding pairs of neutrons or protons, resp. Recent experiments concentrated on decay studies of the $N = 153$ isotones ^{257}Rf [10] and ^{259}Sg [11, 12]. Results are presented in Fig. 1, where simplified level schemes are shown. Striking for the $N = 153$ isotones is the steep decrease of the 11/2$^-$[725] – level with increasing proton number. In ^{253}Fm it forms an isomer of $T_{1/2} = 0.5$ µs located at $E^* \approx 350$ keV, similar to the case of ^{251}Cf (not shown in fig. 1), where it is located at $E^* = 370$ keV and has a half-life of $T_{1/2} = 1.4$ µs. In ^{257}Rf it was identified as an isomeric state of $T_{1/2} = 4.9$ s at $E^* = 70$ keV decaying by α-emission, while in ^{259}Sg it is assigned as the ground-state. The 11/2$^-$[725] – level originates from the $1j_{15/2}$ – orbital and is expected to increase with increasing quadrupole deformation, while the 1/2$^+$[620] – level arises from the $2g_{7/2}$ – orbital and is expected to decrease with increasing deformation [9]. A change in the ground-state configuration thus could hint to a decrease of the quadrupole deformation already from $Z = 100$ to $Z = 106$ for the $N = 153$ isotones, while theory predicts such a decrease starting from $Z = 106$ to $Z = 108$ [5].
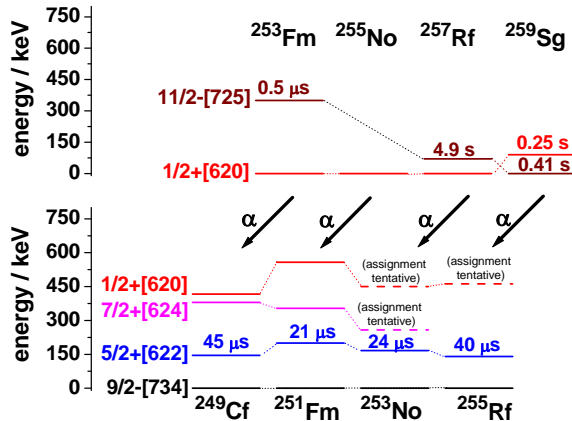
Figure 1. Simplified level schemes for $N = 151$ (^{249}Cf, ^{251}Fm, ^{253}No, ^{255}Rf) and $N = 153$ (^{253}Fm, ^{255}No, ^{257}Rf, ^{259}Sg) isotones. Only those levels are shown, which have been identified in at least three nuclides. Data are taken from [13,14] (^{253}Fm-^{249}Cf), [15] (^{255}No - ^{251}Fm), [10] (^{257}Rf - ^{253}Fm], [11,12] (^{259}Sg - ^{255}Rf).

Alternatively the energy difference of both levels could also be strongly influenced by the hexadecapole deformation which changes gradually from $\beta_4 = 0.024$ (^{251}Cf) to $\beta_4 = -0.023$ (^{259}Sg) [5]. The low lying Nilsson levels in the $N = 151$ isotones 9/2$^-$[734] (from $1j_{15/2}$), 5/2$^+$[622] (from $1i_{11/2}$), 7/2$^+$[624] (from $2g_{9/2}$) and 1/2$^+$[620] play a crucial role in the strength of the shell gap at $N = 152$. Calculations predict them as the lowest lying ones in the $N = 151$ isotones [16,17], but at a different ordering, such as the location of the 5/2$^+$ above the 7/2$^+$. The occurance of the 5/2$^+$[622] as the first excited Nilsson-state, as observed experimentally (see fig. 1), results in the existence of shortlived isomeric states, located along the isotone line in quite narrow energy and halflife intervals $E^* \approx$ (140-230) keV and $T_{1/2} \approx$ (15-45) µs. This trend is also continued for ^{255}Rf, where the isomer has been identified recently [12].

4 Spontaneous fission and nuclear structure

Spontaneous fission properties are a sensitive probe for shell stabilization of superheavy nuclei. As the liquid drop fission barrier decreases below the ground-state motion energy (≈ 0.5 MeV) at around $Z = 104$, fission barriers of heavier nuclei are determined by the shell structure. For fission

properties, among others as e.g. the effective inertia, not only the size of the shell effects play a role, but also their dependence from nuclear deformation or, in other words, their development along the fission path. For nuclei with odd proton and /or odd neutron numbers the change of the energy of the fissioning state along the fission path plays a dececive role, as due to spin and parity conservation at crossing points of Nilsson levels the unpaired nucleon cannot change the level as it is the case for nucleon pairs in even-even nuclei. This leads to an effective increase of the fission barrier, in literature denoted as 'specalization energy' [18]. Quantitatively this influence can be expressed by a 'hindrance factor' HF, defined as

$$HF = T_{sf}(Z,N) / T_{ee}(Z,N) \qquad (1)$$

where T_{sf} is the experimental fission half-life and T_{ee} the unhindered half-life defined as the geometric mean of the neighbouring even-even nuclei [19], e.g. for odd neutron number.

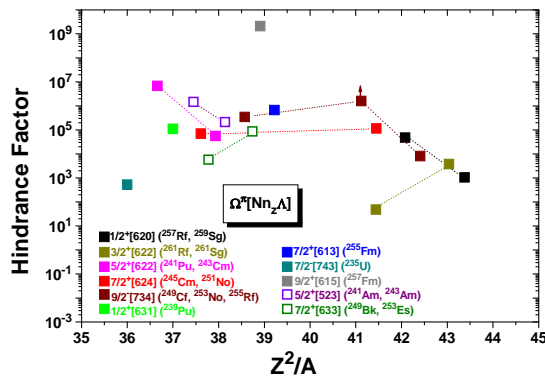$$T_{ee}(Z,N) = (T_{sf}(N-1,Z) \; x \; T_{sf}(N+1,Z))^{1/2} \qquad (2)$$

Fig. 2: Spontaneous fission hindrance factors of odd mass nuclei as function of Z^2/A. Nuclei with the same Nilsson levels are connected by dotted lines to guide the eye.

The situation is displayed in fig. 2. for the cases where spin and parity of the fissioning Nilsson levels ($\Omega^{\pi}[Nn_z\Lambda]$) are experimentally established and where fission halflives of the neighbouring even-even nuclei are experimentally known. Evidently the hindrance factors vary in a range of about nine orders of magnitude, but despite the strong variations a tendency to decrease with increasing fissility, expressed by Z^2/A, is evident. Striking differences to this general trend are only observed for three cases: ^{235}U, ^{257}Fm and ^{261}Rf. The unusual high value for ^{257}Fm was explained in [18] by a strong increase of the fission barrier due to the $9/2^+$[615] level, seemingly connected with the strong upsloping in energy of this level with increasing deformation [9]; but already in [19] the very low half-life of the neighbouring ^{258}Fm (0.3 ms), which indeed means a decrease of T_{sf} by more than seven order of magnitude from ^{256}Fm to ^{258}Fm, was considered as a possible reason, as in this specific case relation (2) either may not be not a good measure for $T_{ee}(^{257}$Fm) or the decrease of the outer fission barrier, which should be responsible for the short fission half-life of ^{258}Fm may not yet be effective for ^{257}Fm; the extremely low hindrance factor found for ^{235}U, vice versa, may possibly be connected with the strong downsloping of the $7/2^-$[743] level at increasing deformation [9]. However, one should also keep in mind that in the case of ^{235}U the reported fission branchings [13] of the envolved nuclei, $^{234-236}$U, are extremely small ($b_{sf} < 10^{-9}$), so the hindrance factor is very sensitive to possible systematic errors. Seemingly unclear is the low hindrance factor of ^{261}Rf; its decay properties and these of the neighboring ^{262}Rf had been under debate for a long time; the analysis here is based on recently published decay data and level assignments, which are in agreement for ^{261}Rf and thus seem quite

reliable [20,21,22,23]. For the cases with the same Nilsson levels (connected by dotted lines in fig. 2) hindrance factors are found to be significantly different. For the even-Z isotopes the values (except for the pair [245]Cm – [251]No) vary within a factor of 40 to 120, for the odd-Z nuclei the differences are somewhat smaller with a factor of 6.6 for [241]Am – [243]Am and 15 for [249]Bk – [253]Es. Neither a distinctive correlation between the spin and the fission hindrance or the fissility and the fission hindrance for the nuclei with the same Nilsson configuration is visible for the available data.

5 Investigation of shell strengths

An important feature regarding nuclear stabilization by the shell structure is the shell strength; measures for it are usually the 2n- (or 2p) – separation energies ($S_{2n}(N,Z)$) or the 'shell gap parameter' $\delta_{2n}(N,Z)$ defined as the difference of the 2n – separation energies ($\delta_{2n}(N,Z) = S_{2n}(N,Z) - S_{2n}(N+2,Z)$).

The masses of nobelium – and lawrencium isotopes measured directly with SHIPTRAP have been recently used to determine the strength of the N = 152 shell [24]; similarly, the directly measured masses for [252]No and [254]No can be used to determine the ground-state masses of the α-decay precursors. For [252]No this chain has been known up to [264]Hs already for a quite long time [25], for [254]No, the recently discovered α-decay branches of [262]Sg [26] and [258]Rf [27] allow for a mass determination up to [270]Ds and hence for an experimental estimation of the 2n – binding energies for the even-even N – Z = 50 nuclei up to [266]Hs. The results are shown and compared with the mass predictions of [4] in fig. 3.
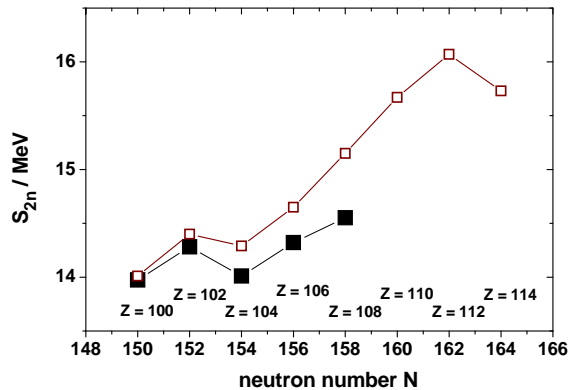

Fig. 3: Two – neutron separation energies ΔS_{2n} of N-Z = 50 nuclei; full squares – experimental values; open squares – predictions from theory [4].

The known shell gap at N = 152 is visible by a local maximum, beyond N = 154 the 2n- separation energies are increasing again due to the influence of the expected neutron shell at N = 162; obviously, the increase of the experimental values is smaller than predicted; this might be accidental, maybe a 'local' effect, but it could be a hint that the N = 162 shell is weaker than predicted. Some indication for this assumption can be found also in the fission half-lives; the agreement between the predicted values [28] and experimental values for [254,256,258,260]Rf and [258,260]Sg is better than a factor of three, while for [262]Sg the predicted value is already a factor of about six higher than the experimental one [26,28]. In a recent experiment at SHIP a fission branch of $b_{sf} = 0.24 \pm 0.09$ has been established in [266]Hs [26], resulting in a partial fission half-life of $T_{sf} \approx 9$ ms, which is almost a factor of 200 lower than the predicted value of $T_{sf} = 1.6$ s [28].

6 Conclusions

Superheavy elements represent a specific class of exotic nuclei. As liquid drop fission barriers are vanished their existence, stability and decay properties are solely defined by their shell structure and thus they are a unique laboratory to study fundamentel interactions. Thanks to technical improvements nuclear structure investigations have become a powerful instrument within the last decade to study nuclear properties at least in the range up to $Z \approx 110$ in more detail. Still, the research work is rather at the beginning. New set-ups increasing the sensitivity significantly are required to perform nuclear structure investigations of similar quality in the region of the expected spherical proton and neutron shells and to investigate properties of nuclei there. This will lead to new insights in understanding the existence and stability of our world.

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