SUPERHEAVY ELEMENTS AT GSI — INVESTIGATING EXOTIC NUCLEAR MATTER*

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The search for the next closed proton and neutron shells beyond 208 Pb has yielded a number of exciting results in terms of the synthesis of new elements at the upper end of the charts of nuclides, in a region of exotic high-Z nuclear matter. In particular, the results obtained at the Flerov Laboratory of Nuclear Reactions (FLNR) for a rich number of decay patterns for 48 Ca induced reactions on actinide targets have by now been confirmed for reactions on 238 U, 244 Pu and 248 Cm at GSI, and on 242 Pu at LBNL. These superheavy elements (SHE), however, are a nuclear structure phenomenon. They owe their existence to shell effects, an energy contribution of quantum mechanical origin to the nuclear potential, without which they would not be bound. Experimental activities in this field, apart from attempts to directly synthesize new elements, have to pursue reaction mechanism studies and, in particular, nuclear structure investigations to study the development of single particle levels towards the expected gap for the proton and neutron shell closure in the region of the spherical SHE.

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1. Introduction

More then four decades after the first prediction of the "island of stability" of SHE in the late sixties of the last century [1] its localization seems almost in reach with experimental indications up to a Z of 118 [2,3]. The two approaches cold and hot fusion, have yielded the synthesis of a large number of isotopes in the region of highest Z and A. At GSI cold fusion reactions have been employed to produce elements up to Z = 112 [3] whereas the heaviest nucleus for this approach has been synthesized at RIKEN in the reaction 70 Zn + 209 Bi [4]. In ⁴⁸Ca induced reactions on actinide target

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nuclei decay patterns have been observed at Dubna which were assigned to the production of nuclei spanning the more neutron rich area from ²⁶⁶Rf to ²⁹⁴118. Some of these results for decay chains assigned to isotopes of the elements Z = 112, 114 and 116 have been confirmed independently at the velocity filter SHIP at GSI [5], at the Berkeley gas-filled separator BGS at LBNL [6], and most recently for the reaction ⁴⁸Ca on ²⁴⁴Pu at the gas-filled separator TASCA at GSI [7]. The results of the investigation of the reaction ${}^{48}\text{Ca} + {}^{248}\text{Cm} \rightarrow {}^{296}116^*$ at SHIP are presently under analysis. Beyond the successful synthesis of heavy nuclei, the high beam intensities nowadays available, together with advanced particle and γ detector set-ups allow for detailed nuclear structure investigations of heavy and superheavy nuclei like 252,254 No [8,15] and 270 Ds [9], which are discussed in this paper. In addition fast SHE chemistry, most recently performed after pre-separation with the gas-filled separator TASCA, and precision mass measurements in the penning trap system SHIPTRAP complete the experimental means available at GSI. As one of the highlights in the field of heavy and superheavy nuclei. the masses of the nobelium isotopes ^{252,253,254}No could be measured with high precision at SHIPTRAP recently [10]. Chemistry studies and possibly X-ray detection of SHE have the potential to provide the, for the hot fusion reactions still missing, unambiguous Z assignment, mandatory for element identification. Precision mass measurements yield an important input in terms of nuclear binding energies for theoretical models. In recent years the development of efficient experimental set-ups, including separators and advanced particle and photon detection arrangements, allowed for more detailed nuclear structure studies for nuclei at and beyond Z = 100. Among the most interesting features is the observation of K-isomeric states. We could recently establish and/or confirm such states in the even-even isotopes 252,254 No. The heaviest nucleus where such a state was found is 270 Ds with Z = 110 as we reported in 2001. Those nuclear structure studies lay out the grounds for a detailed understanding of these heavy and high-Z nuclear systems, and contribute at the same time valuable information to the preparation of strategies to successfully continue the hunt for the localisation of the next spherical proton and neutron shells beyond ²⁰⁸Pb. All these investigations call for a constant development of higher beam intensities due to the ever lower production cross-sections.

2. Synthesis of superheavy elements

The Z- and A-identification of a new isotope is the main task of successful SHE synthesis. Evaporation residue- α correlations providing this assignment for the cold fusion approach, have to be complemented by other means like chemistry or X-ray detection for the nuclei produced in ⁴⁸Ca induced reactions on actinide targets, as their decay chains end in spontaneous fission of unknown nuclides. Their production cross-section values remain surprisingly high around and above the 1-pb-level for Z up to 118 [2]. In order to confirm these tempting results with the promising perspectives of relatively high production yields, we investigated the reactions ${}^{48}Ca + {}^{238}U$ [5] and ${}^{48}\text{Ca} + {}^{248}\text{Cm}$. The decay properties observed for the first reaction are in good agreement with the data obtained at the FLNR [11,12]. In addition to our confirmation, there was a chemical study of the reaction ${}^{48}\text{Ca} + {}^{244}\text{Pu}$ which observed also consistent decay patterns [12]. Most recently the same reaction was used at the gas-filled separator TASCA at GSI to successfully reproduce the same decay patterns assigned to isotopes of element Z = 114 [7] by Oganessian *et al.* [2]. Those results produced consistently in different laboratories and at different types of separators are a qualitative step forward towards a final A and Z assignment which is despite the impressive body of data still not final. To reach nuclei beyond Z = 118synthesis experiments for Z = 119 and 120 are presently discussed at GSI.

3. Nuclear structure of superheavy elements

A comprehensive review of nuclear structure investigations for heavy nuclei has been published by Herzberg and Greenlees [13]. Inbeam studies using a γ -spectroscopy set-up at the target position yield access to the high spin region of nuclear excitation in heavy ion collisions. They are, however, usually hampered by the high background rate from the close by and unshielded target what limits the beam current to moderate intensities. Decay spectroscopy after beam separation yields access to low lying states populated by α -decay or below isomeric states only. They are, however, almost background free due to separation and decay coincidences, and the full intensities of high current accelerators can be used. The combination of the UNILAC accelerator with SHIP and the decay spectroscopy detector in its focal plane is among the most efficient setups for these studies worldwide.

3.1. Decay spectroscopy after separation

The detailed understanding of nuclear structure and its development in the vicinity of closed shells, in regions of deformation and towards heavier masses and higher Z is a necessary ingredient for a successful progress in the synthesis of new heavy elements. The possible trends in single particle levels are the most sensitive probe for the formation of low level density, and eventually the appearance of shell gaps and regions of (shell-) stabilized nuclei. Decay spectroscopy of α -emitters stopped after separation is a powerful tool to study their daughter products or isomeric states via α fine structure or α - γ spectroscopy by ER- α or ER- α - γ coincidence measurements. Here

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the fusion reaction products are after separation implanted into a solid state ("stop") detector for the residue and α detection, which is combined with a high resolving γ -ray (Ge-) detector array. This method is very clean as compared to in-beam studies because of the effective shielding from target background due to its spatial separation and the effective cleaning by the ER- α coincidence technique. It is highly efficient due to the close geometry of the particle and γ detectors, and for the nuclei implanted in the solid state detector no Doppler shift correction is needed. As illustrated in Fig. 1 with this method various features can be studied like γ -rays emitted from an isomeric state surviving the flight time through the separator, γ -rays originating from an excited state populated by α -decay in the daughter nucleus as well as conversion electrons and X-rays.



Fig. 1. Observable decay modes for evaporation residues after separation and implantation in a decay spectroscopy setup.

3.2. (K) isomers

The K-quantum number is the projection of the total angular momentum J on the symmetry axis of an axially deformed nucleus. In prolate deformed nuclei isomeric states are formed when their decay requires a large change in K value. Transitions are called "K-forbidden" when the change in K value is larger than the transition multipole order λ with the degree of forbiddenness defined as $\nu = \Delta K - \lambda$ (see *e.g.* [15, 16]).

Xu *et al.*, employing configuration constrained potential energy surface calculations (PES), predict high K isomers to be a general feature of prolate deformed nuclei in the region for Z>100 in terms of nucleon pair breaking into multi-quasiparticle excitations at low excitation energies ($\approx 1-2$ MeV) for even–even isotopes [14]. In fact, experimentally about 13 cases have

been identified in the region of Z>96 as shown in Fig. 2. A table of K isomers known in even-even isotopes in that region is given in Ref. [13]. K isomers or indications of their existence have been found for almost all even-Z elements in the region Z = 100 to 110. We could recently establish and/or confirm such states in the even-even isotopes 252,254 No [15] (see also [16]). The heaviest nucleus where such a state was found is 270 Ds as reported by our group in 2001 [9]. In the same work we noticed the possibility of an isomeric state in the ²⁷⁰Ds daughter nucleus ²⁶⁶Hs (see below). For Z = 106 the search was up to now not successful. However, K isomers were found also in even-odd and odd-even isotopes in this region like *e.q.* ²⁵¹No [15], ²⁵³No [17], ²⁵⁵No [18], and ²⁵⁵Lr [8,19]. Xu *et al.* pointed out that high-spin K isomerism has consequences for the fission barrier and α -decay. which could lead to higher stability and longer lifetime of the isomeric state as compared to the ground state (g.s.) for a certain class of superheavy nuclei. They state that via the population of high-K states in experiments, one may be able to extend the nuclear chart further into the island of superheavy *nuclei*. Examples for such an isomer-g.s. lifetime inversion are 250 No [19] and 270 Ds [9], the heaviest nucleus for which a K isomer has been observed.



Fig. 2. Excerpt of the chart of nuclides indicating the K isomers observed for heavy nuclei in the region $Z \leq 96$ and the decay chain for the proposed reaction $^{64}\text{Ni} + ^{207}\text{Pb}$. Half-life, decay energy, spin and parity values are given for K isomers only.

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For the latter we observed in the reaction ${}^{64}\text{Ni} + {}^{207}\text{Pb}$ a total of 8 decay chains, consisting of evaporation residue(ER)- α - α -sf correlations which we all attributed to the production of ${}^{270}\text{Ds}$ followed by the sequential emission of two α -particles leading to the daughter ${}^{266}\text{Hs}$ and the granddaughter ${}^{262}\text{Sg}$ which eventually decayed by spontaneous fission [9]. During a period of 7.3 days we collected a beam dose of 1.3×10^{18} projectiles at a beam energy of 317 MeV. At the resulting excitation energy of 14 MeV we obtained a production cross-section of 13 ± 5 pb which was close to the value obtained for the synthesis of ${}^{271}\text{Ds}$ in the reaction ${}^{64}\text{Ni} + {}^{207}\text{Pb}$.

For two of the decay sequences the first α escaped from the detector in backward direction without generating an electronic signal in the Si detector. For the six remaining chains we observed the 270 Ds α decay which we assigned to two groups of decay times with half-lives of $6.0^{+8.2}_{-2.2}$ ms and 100^{+140}_{-40} µs, respectively. The α decay energy of the shorter lived g.s. was 11.03 ± 0.05 MeV, whereas for the longer lived isomer we observed α particles with 10.95 ± 0.02 MeV, 11.15 ± 0.02 MeV and 12.15 ± 0.05 MeV. We could assign those six α -decays to various decay paths which are consistent with the calculated level scheme [20]. We interpreted one 270 Ds α decay (chain number 7), that belonged to the slower decay group and that was followed by a γ -ray, as the isomer decaying into an excited state of the daughter ²⁶⁶Hs. The first α of chain number 8, being also part of the longer lived group, with the highest decay energy was assigned to the isomer-to-g.s. decay. From the energy difference of this decay and the g.s.-to-g.s. transition energy, we derived the excitation energy of the isomer of 1.13 MeV. The last slow decay, having the same α energy as the three faster decays is consistent with the isomer decaying by γ -emission (not observed) into the ^{270g.s.}Ds which then decays directly into the ²⁶⁶Hs ground state. The remaining three observed α particles from the shorter lived group we view as g.s.-g.s. transitions. Note: this interpretation is consistent with theoretical expectations [21] which were also confirmed by Xu et al. [14] who produced a similar decay scenario. However, the derived ^{270m}Ds excitation energy is based on just one measured α decay. The probability is non-zero that the transition assigned as isomer-g.s. transition populates a state above the ground state instead. In that case this excitation energy value would be a lower limit. Other properties like e.q. spin and parity of the ²⁷⁰Ds K isomer or the excitation energy of the states populated in ²⁶⁶Hs are still missing. For the daughter ²⁶⁶Hs the observed eight decay times were consistent with a single half-life of $2.3^{+1.3}_{-0.6}$ ms with an α energy of 10.18 ± 0.02 MeV. The time distribution of the ²⁶⁶Hs α decay is relatively wide spread and we pointed out the possibility of the presence of two decay components with half-lives of $0.35_{-0.11}^{+0.28}$ and $6.3_{-2.3}^{+8.6}$ ms, respectively. This would be in line with the expectations from the theoretical predictions of the existence of K isomers

in the whole region of deformed heavy and superheavy elements. Following our decay assignment above, one out of six 270 Ds α decays (chain number 7) populates an excited state in 266 Hs.

The last member of the decay chain decays by fission with a half-life of $6.9^{+3.8}_{-1.8}$ ms and a total kinetic energy of the fission fragments of 222 ± 10 MeV. The time distributions are shown in Fig. 3. The 8⁺ to 6⁺ transition in the g.s. rotational band of ²⁶⁶Hs is predicted to have a transition energy of 229 keV, a value which is close to the 218 keV we measured for a γ -ray in coincidence with the 11.15 MeV α particle of the longer lived ²⁷⁰Ds decay (decay chain number 7) in our first measurement.



Fig. 3. (a), (b) time distribution of the subsequent α -decays and (c) fission events — measured for $^{64}\text{Ni} + ^{207}\text{Pb}$. The curves represent the density distribution (arbitrary ordinate scale) of counts in a radioactive decay on a logarithmic time scale with logarithmically increasing channel width [22]. The position of the curve, adjusted to the data by a least-squares analysis, determines the lifetime. The dashed curve (b) indicates that the data are also compatible with two lifetimes of 0.5 and 9 ms. The numbers from 1 to 8 assign the events to the measured decay chains (Table 1 in Ref. [9]). (Figure taken from Ref. [9].)

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The expected α -sf branching ratios for ²⁷⁰Ds and ²⁶⁶Hs are relatively low. In Ref. [9] we estimated upper limits with 0.2 and 1.4%, respectively. A search for those sf-branches is very challenging. For ²⁶²Sg our measured limit for an α -branch of 22% was recently improved be Gregorich *et al.* [23] who obtained a limit value of 16%. This is very close to the prediction of 15% [9]. The observation of this α decay would complete the chain from



Fig. 4. Proposed decay and level scheme for ²⁷⁰Ds and its decay products on the basis of our observations [9] and the predictions of Hartree–Fock–Bogoliubov calculations [21], taken from Ref. [9] (center). The insert (upper left) illustrates the decay paths for ²⁷⁰Ds corresponding to the six observed decay chains identified by the chain numbers from Ref. [9]. On the lower right the now newly established decay pattern for ²⁷⁰Ds is shown, including the new ²⁶⁶Hs spontaneous fission branch, and the searched for and now found ²⁶²Sg α branch providing the Q_{α} connection to ²⁵⁴No [24].

²⁷⁰Ds to ²⁵⁴No for which we recently performed a precision mass measurement [10] and for the first time yield an experimental mass value for an even–even nucleus in this superheavy element region. To settle these questions more decay data were collected in an experiment at SHIP, which was performed while this conference took place. During this run a total of 25 decay chains of ²⁷⁰Ds had been collected in more than 40 days of beam on target. Although the data is presently under analysis, some of the achievements can be reported here, like the spontaneous fission branch which was observed for the first time for ²⁶⁶Hs. More importantly, however, also the searched for α decay of ²⁶²Sg mentioned above was found and with this an experimental mass for ²⁷⁰Ds has been established. The final results of this analysis will be published soon [24]. Fig. 4 shows the previously proposed decay pattern together with the now newly established decay properties of ²⁷⁰Ds and its decay products.

4. Outlook: towards the island of stability

Low cross-sections, the advances in nuclear structure investigations, reaction mechanism studies, chemistry and SHE synthesis experiments with a steadily increasing demand for higher beam intensities, more sensitive and more sophisticated detection set-ups and new methods determine the road map for future SHE investigations. Intensity increase is one of the major issues in this context. The presently pursued upgrade of the UNILAC accelerator at GSI, consisting of a new 28 GHZ ECR source and a new RFQ injector providing an order of magnitude higher beam current, is only a first step towards a dedicated continuous wave (CW) accelerator which would increase the beam intensity by a factor of four already by extending the 25% UNILAC duty cycle to 100%. Additional increases due to the advanced accelerator technology can be expected. Improvement of the detection system in terms of higher efficiencies is as mandatory as the employment of additional measurement parameters. Mass determination by an adequate spectrometer, complementary to precision mass measurements with a trap, would be extremely helpful for a final confirmation of the unconnected chains obtained in the ⁴⁸Ca induced reactions on actinide targets. The unambiguous experimental establishment of the Z of the nuclides produced in 48 Ca reactions on actinide targets is the most important task in the field of SHE, presently even more important then the synthesis of nuclei with a presumably higher Z.

In conclusion, the roadmap towards spherical shell stabilized nuclei is laid out. The challenges are obvious. It is up to us to take them on. The recent experiments were performed together with M. Block, H.-G. Burkhard, Ch. Dröse, S. Heinz, F.P. Hessberger, S. Hofmann, J. Khuyabaatar, I. Kojouharov, R. Mann, J. Maurer, E. Minaya, B. Sulignano (IRFU Saclay), A.G. Popeko, A.V. Yeremin (FLNR-JINR Dubna), S. Antalic, Š. Šaro, M. Venhart (University of Bratislava), P. Greenlees, M. Leino, J. Sorri, and J. Uusitalo (University of Jyväskylä). The author would like to thank the GSI target laboratory for their skillful and minute support. He acknowledges, in particular, the great and professional afford of W. Barth, L. Dahl, P. Gerhard and the entire accelerator team to provide the ⁶⁴Ni beam for the ²⁷⁰Ds experiment.

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